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The National University of Ireland, Cork

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# Sustainable Food Packaging Engineering

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*The thesis is presented to the National University of Ireland in fulfilment of the requirements for the Degree of Doctor of Philosophy*

(PhD in Process & Chemical Engineering)

Under the supervision and co-supervision of

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# Declaration

I hereby declare that the present dissertation is my own work and has not been submitted to another degree neither at the University College Cork nor elsewhere. Where the work of others was necessary to build an argument, it was acknowledged and referenced properly.

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Ina Bremenkamp

*December 2023*

# Executive Summary

Food is a basic need and food packaging plays an important role in providing safe food to the world. Food packaging innovations in the past were important milestones for the development of the food sector, by ensuring safety, prolonging quality, and enabling the current supply chain. Nevertheless, the focus has drifted and innovations in the food sector are starting to be driven by ecological needs. The current food packaging options present challenges due to the dependency on fossil raw materials and the accumulation of durable single used packaging waste. Therefore, food packaging innovations need to be adjusted to the new demands, by not being an environmental burden, but still providing product protection, safety, and supporting various supply chains. Design strategies as eco-packaging design, circular economy design and design for recycling are important drivers. Furthermore, the development, improvement, and standardisation of biopolymers for replacing fossil sourced packaging materials are being widely studied. This opens up opportunities for relieving the current environmental burden of food packaging.

This thesis focuses on alternative food packaging for ready to eat (RtE) seafood products, a food category that ultimately provides healthy and convenient food to consumers. RtE is a food product group with increasing turnovers, supporting different life styles, e.g., eating on the go, fast and simplified product preparation as well as healthy eating. Seafood products provide essential nutrients and are a valuable food source, rich in nutrients that are particularly important for elderly people, a growing consumer group all over the world. The objective of this thesis was the investigation of biobased edible coatings as sustainable packaging development for RtE seafood products by using a quantitative methodology. A quantitative packaging development approach, in contrast to the often-used qualitative approach in industry, supports an optimal packaging material selection based on the product characteristics and its potential supply chain.

Currently a challenging task in the development of eco-food packaging systems is the assessment of the environmental burden. Every action or product has an environmental impact, but the environmental impact can be significantly influenced by the selected options. A systematic review of existing life cycle assessments (LCA) for investigating environmental challenges of novel food packaging systems in the field of RtE fish and meat products was undertaken and revealed a significant impact of the selected LCA approach on the evaluated environmental impact, highlighting the importance of transparent LCA studies, using fair comparisons and a holistic scope. A step by step approach by shifting the LCA focus to different objectives, such as the whole food-packaging system, comparing different packaging materials and systems, is recommended to allow a comprehensive understanding of the environmental impact of food-packaging system. This enables industry stakeholders to make informed decisions, taking an active role that balance necessity, wastefulness, and creating efficient and sustainable packaging solutions.

A critical review of edible coatings for chilled RtE food products was also performed to understand the state of the art and identify knowledge gaps for further developments. Edible coatings have been studied for RtE food products mainly to improve the microbiological stability; with other quality parameters receiving much less attention. Only few products within the RtE seafood category were tested in combination with edible coatings. Edible coatings can be sourced from natural resources, including land or marine based biomass. The direct use of food resources as packaging can create a

burden on the food supply chain. The conversion of food industry waste products to edible coatings allows an optimal use of the full harvested biomass, but has some challenges, e.g., volume availability, collection, and possible high energy demand during transformation. Also, during the selection of an edible coating material, the supply chain impact should be reviewed, and additionally industrial scale up feasibility should be assessed for a successful industrial application. Biobased materials often have a limited potential, but by combining multiple biobased materials, the coating properties can be optimised. Aspects considered included the methodology for coating application, assessment of coating performance, and edible coating challenges as part of the food system were also discussed. Edible coatings are a complex topic but provide an interesting approach for developing eco-food packaging systems.

Developing a tailored packaging system for a RtE seafood product requires the identification of critical quality parameters and understanding of the influence of environmental conditions on the product quality. Therefore, the degradation process of two RtE seafood products, a RtE baked fish product and a RtE minced fish patty, were investigated by studying the microbiological, chemical, and physical properties. The studied sorption isotherm behaviour showed a sigmodal shape, and the best model fitting was reached with the Peleg model, followed by the Guggenheim-Anderson-de Boer (GAB) model. The storage experiments revealed that water loss and microbial growth are important product quality parameters for both tested RtE seafood products, while fat oxidation was only a critical quality parameter for RtE seafood products with a higher fat content. By understanding the underlying reaction of degradation processes and the effect of environmental factors such as oxygen, light, and relative humidity, the required packaging properties were identified.

The application of chitosan and alginate coatings for RtE baked fish products were studied. A full factorial experimental design was used to investigate the effect of coating material composition on microbial growth, water loss and lipid oxidation under optimal (4°C) and abuse (14°C) storage conditions. A 3^2 full factorial design was used to study the effect of chitosan concentration (1, 2 and 3%), and glycerol concentration (0, 15 and 30% w/w chitosan). The effect of the composition of an alginate coating was studying with a 2^3 full factorial design. The studied factors were alginate concentration (1 or 2% (w/v)), glycerol concentration (0 or 1.5% (w/w) alginate) and crosslinking the alginate coating with $CaCl_2$ to form calcium alginate (yes/no). The results showed that a chitosan coating with 1% (w/v) chitosan in 1% (v/v) acetic acid, and 15% (w/w chitosan) glycerol, or 1% (w/v) alginate coating with no glycerol, and no crosslinking showed the best performance in controlling the tested safety and quality parameters. Additionally, a full experimental factorial design was performed to study the combined effect of an alginate and chitosan coating applied as a double coating, and single coating. The double coating tested did not provide a combined product protection. A desirability method was used to identify the shelf life of chitosan, alginate and double coated RtE products based on multi quality parameters. It was concluded that chitosan coated samples showed the best performance with a three- fold shelf life extension compared to uncoated products.

Improving product safety and quality by combining different protection methods, known as hurdle technology, is a common approach. The applied hurdles can differ. Well applied examples are sanitizing, packing and cold storage for fruits and vegetables. A novel sanitizing system consists of the use of cold plasma treatment, avoiding the use of chemicals to reduce microbiological contamination and increase safety. The combination of cold plasma treatment and an equilibrium modified atmosphere packaging (EMAP) for cherry tomatoes was investigated under controlled temperature (10 and 20°C) throughout storage. Statistical analysis of the results showed that plasma treatment did not negatively affect the quality of the product. A higher microbiological growth was detected within the packed cherry tomatoes, while a plasma treatment before packing reduced growth of microorganism during the early days of storage. Package was important to prevent weight loss and changes in total soluble solid content over 14 days of storage. The combination of cold plasma, temperature and EMAP design ensured the quality retention of cherry tomatoes, which suggests that they have potential to be used as hurdle technology.

A packaging based on an edible coating, requires a second packaging component which can be a packaging film. Combining the edible coating and an outside layer should provide an optimal product protection over the required shelf life. A mathematical modelling approach was taken to estimate the required water barrier properties for the packaging film material. The water loss was identified as the remaining most critical quality parameter after a chitosan coating was applied. Film materials were selected based on their properties and end of life options, and the water vapour barrier properties at lower storage temperatures were tested. The water vapour transmission rate (WVTR) allowing a water loss of 10% within 21 days was calculated using the GAB model, and using a linear approximation of the isotherm was estimated to be 94 and 81 g/m$^2$ day, respectively. The simplified linear model showed similar results to the GAB model approach for predicting moisture loss of a high moisture food product. When increasing the required shelf life only a small reduction of the WVTR was seen. Testing the WVTR at chilled storage conditions showed that depending on the material selected, the temperature can impact the water vapour barrier of the packaging film materials, especially for biobased materials. The recommended film material in combination with a chitosan edible coating for a RtE baked fish product was a cellulose based compostable film.

Food packaging is essential but only with efficient solutions, an open mind and with various approaches the negative effects of food packaging on the environment can be tackled. Special emphasis should be given to food packaging that allows to use naturally available protection systems. It was highlighted that including circular economy aspects and environmental considerations during all development stages of a food packaging improves the sustainability of a packed food product. An integrated precision packaging, based on an edible coating and a film layer, for RtE seafood products can be an eco-packaging solution, without compromises in food safety and quality.

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# General Introduction

## Background

Nowadays, food packaging is seen as a threat from environmental conscious consumers. Images of packaging accumulations in the sea, beaches or lands are circulating in the media. Studies revealing the omnipresence of plastic particles, often used for food packaging, in nearly every part of the world and even traces have been found in organisms. Environmental pollution due to packaging waste is only one challenge related to food packaging, another is the dependency of fossil resources. These aspects prevail the positive contribution of food packaging. Without food packaging shelf life of food products would be reduced, product safety could be at risk, current product ranges in the markets would not be possible, and handling would be more difficult, to only mentioning some advantages of food packaging.

The main challenges for food packaging are therefore, to find a solution to eliminate the environmental threats without compromising the current food safety and guaranteeing food supply. Diverse strategies are being explored to improve the sustainability of food packaging, whereby some focusing on the optimization within the available packaging supply chain infrastructure, and others exploring new possibilities. Keeping a broader view on the possibilities to improve the packaging sustainability is important to address the diversity of food products, markets, and distribution chains. Food packaging waste accumulation statistics show that there is still a long way to go to a sustainable food packaging system. The selection of a food packaging is an interdisciplinary task, combining requirements of many disciplines such as technology, quality & safety, supply chain, sales, marketing, and sustainability aspects; highlighting the complexibility of food packaging developments.

## Rational and Objectives

For high perishable products like vegetables, fruits, fish and meat, that provide important nutrients, packaging is essential for protecting the quality and safety of the products and extending the shelf life. Packaging addresses two goals of the sustainability strategy of the United Nations to provide enough food for the growing population (SDG2) by ensuring best usage of products grown and therefore, avoiding food waste (SDG12). Another important aspect to consider for the food sector is the shift of the population age distribution to an increasing elderly population. As well as the consumer behaviours and demands, for save, healthy, nutritious and convenient products. Therefore, special emphasis in this work have been given to sustainable packaging developments for RtE healthy and nutritious food products.

Hurdle technologies are a common way in the food industry to improve the product preservation, by combining various barriers to delay food degradation processes. Hurdles can be factors like product preparation, preservation methods, temperature control, or physical barriers. The most common hurdles for foods are cold storage and a package. Other hurdle technologies in food packaging are modified packaging, active packaging, intelligent packaging, antimicrobial packaging, edible packaging, or nanotechnology. By using hurdle technologies, food manufacturers can not only improve food safety, extend shelf life, and maintain product quality, but also improve the sustainability. The principle to reduce the environmental impact of a food packaging by using complementary packaging parts is still an under investigated topic and was further explored in this work.

In 2018 the EU Directive 94/62/EC on Packaging and Packaging Waste to reduce food packaging environmental impact published by the European Parliament and the Council was amended to set a recycling target to 65, and 70% by weight of all packaging waste to be achieved by 2025, and 2030, respectively. The focus is placed on improving the recyclability by using design for recycling, favouring few packaging materials, improving the recycling infrastructure, and creating a market for recyclate. Here, also the use of synthetic plastic materials from biobased resources as biobased PET is another aspect, but the use of biobased and biodegradable materials is less addressed. Biobased and biodegradable packaging systems have often been seen as contamination of the current packaging material streams in existing waste management systems. Nevertheless, biobased, and biodegradable food packaging materials have potential for reducing fossil dependency and reducing environmental pollution. A packaging concept with barriers based on biobased edible and biobased compostable materials was investigated within this thesis.

Edible packaging is produced from natural edible materials and already commercialised for fruits and vegetables. Another application area is for fast moving consumer goods as eating and drinking on the go, as introduced by start-ups companies, e.g., Notpla. Edible packaging is designed to be eaten with the product or being easily biodegradable, often having a similar biodegradation time as the food product itself. Ideally, they are produced from food residuals, applied to the product as coating or film, and provide protection functions by controlling product degradation processes. The concept of edible coatings to food product groups, as RtE foods, can be of interest to provide high quality, and safe products and at the same time to reduce packaging waste. Research studies often use edible coatings in combination with a synthetic food packaging with the aim to prolong the shelf life. Whereby, the aim for a maximum possible shelf life is a concept that should be reconsidered. A specific shelf life is prerequisite to support flexibility in production, transportation, retailing and handling at the consumer, but aiming for a maximum possible shelf life is not always desirable. Overprotection can lead to the use of packaging materials with unnecessary high barrier properties, often know to have a high environmental impact. Another strategy goal for a packaging concept based on an edible coating and a film layer can be to improve the sustainability of the packaging system by providing a maximal necessary shelf life. Both the edible coating and the film layer can be designed to provide a protection synergy and ensuring a low environmental impact. Hurdle packaging systems are a powerful tool to design packages that can prolong shelf life of food products, but it is necessary to transfer the concept to food as RtE foods. Avail of a better understanding of the concept of edible coatings for this food group, as well as related possibilities of edible coatings for developing more sustainable, eco-food packaging systems is necessary. Therefore, the development and selection of a tailored complementary food packaging consisting of an edible coating and film layer was intensively investigated within this thesis.

### Thesis structure

The research structure used within this thesis to address the defined objectives is visualized in Figure 0-1.

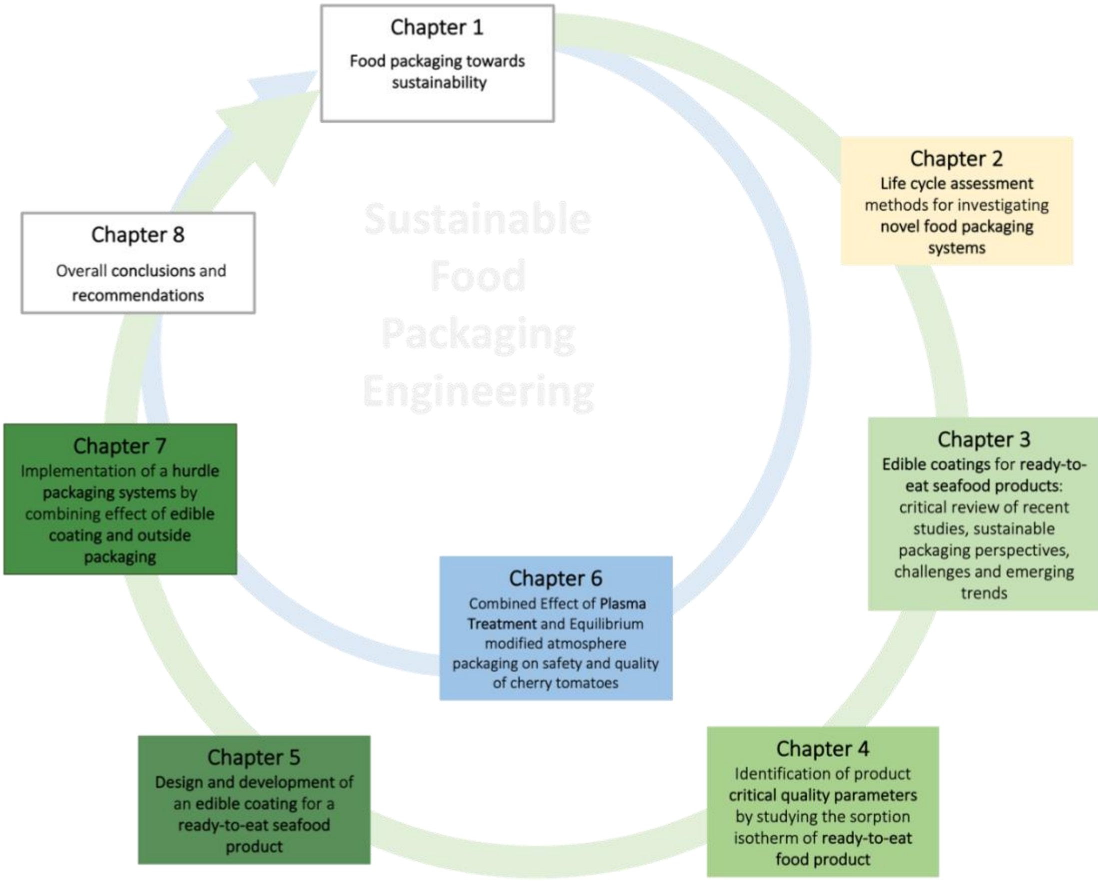


*Figure 0-1: Graphical representation of the thesis structure.*

**Chapter 1** covers challenges in the food industry related to food packaging, and its impacts. Additionally, trends in the food sector to address these challenges, as eco-design, circular economy, design for recycling, biobased materials, biodegradable materials, edible packaging, and the role of government regulations were discussed. An overview of the current approach for food packaging selection and development were presented. This chapter provides an overview and essential background knowledge about environmental developments within the food packaging area. Only with a great understanding of the whole concept, innovative, impactful research can be conducted.

Assessing environmental impact is crucial for a successful implementation of changes toward a sustainable food packaging. Identifying environmental hotspots within a supply chain and comparing different packaging concepts are of relevance. Different approaches are applied. Depending on the approach taken the focus of the LCA study can differ and the outcome can be significantly influenced. Therefore, in Chapter 2 LCA methodologies with the focus on food packaging were compared by reviewing LCA studies for RtE seafood and similar products. A special emphasize was given to LCA approaches for novel food packaging systems.

The gained knowledge from Chapter 2 was applied for the investigation of edible coatings as a sustainable food packaging concept for RtE seafood products. Chapter 3 focuses on a literature review by summarizing the state-of-art, sustainable packaging perspectives, challenges, and emerging trends of edible coatings for RtE seafood products.

When developing a tailored packaging, understanding of product behaviour, degradation processes and identifying critical quality parameters is of ultimate importance. In Chapter 4, the degradation reactions of RtE baked fish product were established by studying the sorption behaviour, water loss, microbiological degradation, and fat oxidation during chilled and abused storage conditions. The results support the development of an innovative tailored synergetic packaging for a RtE seafood product based on an edible coating and a film layer.

In Chapter 5 the experimental results of the design and development of an edible coating for a RtE seafood product are presented. Circular economy principles were considered by focusing on coatings originated from marine environment. Edible coatings based on alginate, a sustainable sourced biomass from the sea and chitosan, mainly produced from crustacean shells a waste product of the food industry, were optimized to provide protection against degradation processes of RtE seafood products, such as microbiological growth, water loss and fat oxidation.

A holistic approach was considered by adding a study about hurdle technology for food packaging systems. The effect of a packaging hurdle concept for cherry tomatoes was investigated in Chapter 6 to improve product quality and safety. Plasma treatment, a novel sanitizing method, in combination with equilibrium modified atmosphere packaging (EMAP) was studied. This hurdle packaging concept can provide sustainable improvements due to three reasons: An increased product stability, safety, and reduced food waste along the supply chain. A plasma disinfection treatment avoids standardly applied chemical treatment processes. An EMAP is a packaging concept reaching for a natural respiration control. The gained learning outcomes were transferred to a hurdle technology based on an edible coating and film layer.

An edible coating is part of an integrated packaging concept, as only combined with a second packaging component all packaging requirements can be fulfilled. The focus in Chapter 7 was the selection of a complementary film material to the previously developed edible coating tailored to a chilled RtE seafood product. Film materials were selected based on the mathematical estimated minimum required water barrier for the product and were compared with the measured barrier properties of the film materials at chilled storage conditions. Environmental considerations were investigated for film materials fulfilling the product requirements and were used for the final selection of a sustainable film material. The outcome is a hurdle packaging system for a RtE seafood product with a synergetic effect between an edible coating and a film material providing a full product protection.

**Chapter 8** provides the overall findings and conclusions of the presented thesis work. A critical viewpoint analysis was conducted to assess the potential of packaging concepts based on edible coatings. Recommendations are given for supporting the journey of food packaging toward sustainability.

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## Publications

A list of research results for publication in journals, and conferences are presented following the thesis structure:

Chapter 1:

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2019) Co-creation workshop and co-design of eco-innovative packaging solution. SeafoodAge meeting, 6th Nov 2019, Lisbon, Portugal, oral presentation.

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2020). Novel eco-friendly food plastic packaging. Linking the Food Chain. College of Science, Engineering and Food Science.

Chapter 2:

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2023) Life cycle assessment methods for investigating novel food packaging systems. Environmental Science Advances. (submitted)

Chapter 3:

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2021). Coatings as a natural invisible product protection. SeafoodAge Workshop, 25th May 2021, Braga, Portugal, oral presentation.

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2023) Edible coatings for ready to eat seafood products: critical review of recent studies, sustainable packaging perspectives, challenges, and emerging trends. Packaging Technology and Science. (submitted)

Chapter 4/5:

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2022) Design and development of an eco-packaging for the ready to eat seafood product. SeafoodAge Conference, 19th Oct 2022, Vigo, Spain, oral presentation.

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2022). Marine based coatings as an eco-food packaging for ready to eat seafood products. In 50th Annual Food Science and Technology Conference, date 6 & 7th Dec 2022, Cork, Ireland, oral presentation.

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2024) Design and development of an edible coating for ready to eat seafood product. Polymers, 16, 346. Doi:https://doi.org/10.3390/polym16030346

Chapter 6:

**Bremenkamp, I.,** Ramos, A. V., Lu, P., Patange, A., Bourke, P., & Sousa-Gallagher, M. J. (2021). Combined effect of plasma treatment and equilibrium modified atmosphere packaging on safety and quality of cherry tomatoes. Future Foods, 100011. doi:https://doi.org/10.1016/j.fufo.2021.100011

Chapter 7:

**Bremenkamp, I.,** and Sousa-Gallagher, M. J. (2022) Investigation of an edible coating in combination with an outside layer. SeafoodAge Meeting, 11th May 2022, Cork, Ireland, oral presentation.

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# Nomenclature

| Symbol | Unit | Description |
|---|---|---|
| $\dot{v}_{out}$ | m/s | Air velocity outside |
| $\overline{x_{exp,i}}$ | - | Mean of N experimental moisture content |
| A | $m^2$ | Surface area of packaging |
| A,B,C,D,E | - | Dimensionless parameters |
| $A_p$ | $m^2$ | Surface area of fish |
| $a_w$ | - | Water activity |
| $B_1$ | - | Parameter 1 |
| $B_2$ | - | Parameter 2 |
| $b_{lin}$ | g water/g d.m | Slope of linear isotherm |
| $C_0$ | - | Dimensionless parameter constant |
| $C_g$ | - | Guggenheim constant |
| $C_{GAB}$ | - | GAB model constant |
| $C_h$ | | Dimensionless Hailwood-Horrobin parameter constant |
| $C_{lin}$ | g water/g d.m. | Constant for linear isotherm |
| d | - | Desirability score |
| D | - | Desirability |
| E% | % | Mean relative deviations modulus |
| h | - | Interval between steps |
| $h_f$ | m | Height of film |
| $h_p$ | m | Height of the product |
| j or P/X | mol m/$m^3$ s Pa | Permeance |
| $k_{GAB}$ | - | GAB model constant |
| $l_f$ | m | Length of film |
| $l_p$ | m | Length of the product |
| m | Kg water/Kg solids | Moisture content |
| $M_{0,GAB}$ | Kg water/Kg solids | Moisture content of monolayer for GAB isotherm of food |
| $m_{cal,i}$ | db% | Calculated moisture content dry basis at specific water activity |
| $m_{db}$ | Kg water/Kg solids | Moisture content dry basis |
| $m_e$ | Kg water/Kg solids | Moisture content at equilibrium |
| $m_{exp,i}$ | db% | Experimental moisture content dry basis at specific water activity |
| $m_i$ | Kg water/Kg solids | Moisture content initial |
| $m_{m,GAB}$ | - | Moisture content constant GAB model |
| $M_{r,H20}$ | Kg/mol | Molecular mass of water |
| MSE | - | Means square error |
| $m_{wb}$ | Kg water/Kg solids | Moisture content wet basis |
| N | - | Number of experimental data |
| OTR | cc/$m^2$ day | Oxygen transmission rate |
| P | mol m/$m^2$ s Pa | Permeability |
| $p_0$ | Pa | Saturated water vapour pressure |
| $p_p$ | Pa | Vapour pressure of water exerted by the system |
| $p_{p,ins}$ | Pa | Partial vapour pressure of water vapour inside food package |
| $p_{p,out}$ | Pa | Partial vapour pressure of water vapour outside food package |
| $p_{sat}$ | Pa | Saturated vapour pressure of pure water at defined temperature |
| $R^2$ | - | Coefficient of determination |
| RH | - | Relative humidity |
| RH% | % | Relative humidity |
| $RH_{in}$ | - | Relative humidity inside the packaging |
| $RH_{out}$ | - | Relative humidity outside the packaging |
| RMSE | - | Root mean square error |
| s | - | Model parameters |
| SSR | - | Sum of square of residuals |
| t | s | Time |
| T | °C | Temperature |
| $T_K$ | K | Absolute temperature |
| TR | g/$m^2$ day | Water vapour transmission rate |
| $W_0$ | g | Weight initial |

| | | |
|---|---|---|
| $w_f$ | m | Width of film |
| $W_L$ | g | Weight loss |
| $w_p$ | m | Width of the product |
| $W_s$ | g | Weight of dry solids enclosed |
| WVT | $g/m^2$ day | Water vapour transmission rate |
| X | m | Film thickness |
| yi+x | - | Next estimated solution value |

# Abbreviations

| | |
|---|---|
| AL | Aluminium |
| ANOVA | Analysis of variance |
| aPET | Amorphous polyethylene terephthalate |
| BET | Brunauer-Emmett-Teller |
| bio-PE | Biological sourced Polyethylene |
| bio-PET | Biological sourced Polyethylene terephthalate |
| bio-PP | Biological sourced polypropylene |
| BOPP | biaxially-oriented polypropylene |
| BVOH | Butenediol vinyl alcohol |
| CDTR | Carbon dioxide transmission rate |
| CFU | Colony forming units |
| CL | Colour index |
| CoP | Cut of point |
| CQP | Critical quality parameters |
| CtGa | Cradle to Gate |
| CtGr | Cradle to Grave |
| D4R | Design for Recycling |
| db | Dry basis |
| Eco | Ecological |
| EMAP | Equilibrium modified atmosphere packaging |
| EO | Essential oils |
| EoL | End of life |
| equ. | Equivalent |
| EU | European union |
| EVOH | Ethylene vinylalcohol |
| EVOH | Ethylene-vinyl alcohol-copolymer |
| FFS | Film forming solution |
| FLW | Food loss and waste |
| FTP | Food to packaging ratio |
| GAB | Guggenheim-Anderson-de Boer |
| GHG | Greenhouse gas |
| GIZ | Deutsche Gesellschaft für internationale Zusammenarbeit |
| HMW | High molecular weight |
| LAB | lactic acid bacteria |
| LCA | Life cycle assessment |
| LCC | Life cycle costing |
| LCC-VA | Life cycle costing – value added |
| LCI | Life cycle inventory analysis |
| LCIA | Life cycle inventory assessment |
| LDPE | Low density polyethylene |
| MAP | Modified atmosphere packaging |
| MC | Moisture content |
| MDA | Malondialdehyde |
| MDA | Malondialdehyde |
| MMW | Medium molecular weight |
| MPP | Macro perforated packaging |
| Mt | Megatons |
| OEO | Oregano essential oils |
| OPP | Oriented polypropylene |

| | |
|---|---|
| PA | Polyamide |
| PBS | Polybutylene succinate |
| PE | Polyethylene |
| PET | Polyethylene terephthalate |
| PHA | Polyhydroxyalkanoates |
| PHA | Polyhydroxyalkanoates |
| PLA | Polylactic acid |
| PP | Polypropylene |
| PS | Polystyrene |
| PvdC | Polyvinylidene chloride |
| PVOH | polyvinyl alcohol |
| QR Code | Quick response code |
| RH | Relative humidity |
| RtC | Ready-to-cook |
| RtE | Ready to eat |
| SDG | Sustainable Development Goals of United Nations |
| SEM | Scanning electron microscope |
| SLCA | Social life cycle assessment |
| TAC | Total aerobic count |
| TBARS | Thiobarbituric acid reactive substance assay |
| TPS | Thermoplastic starch |
| TSS | Total soluble solids |
| USD | United states dollar |
| wb | Wet basis |
| WVTR | Water vapour transmission rate |
| XPS | Expanded polystyrene |
| ZnO | Zinc oxide |

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Chapter ONE

# Food packaging towards sustainability

## Abstract

*This chapter assesses the current food packaging situation as only with a great understanding of the whole concept, innovative and impactful research can be conducted. Therefore, challenges in food industry and packaging sector were addressed with the focus on food loss and waste, fossil resource dependency and packaging waste. High awareness of consumers, industry, researchers, and governments regarding these topics was and still is an important driving force for identifying issues, researching for alternatives, and implementing changes. Research and actions are undergone to move towards sustainable food packaging and are summarized under the section trends in food packaging. An interesting aspect for developing sustainable food packaging systems is to understand the current decision-making process for the selection of a packaging material. Providing insights on where and how to implement new developing strategies. It showed which packaging requirements are prioritized and drives the current selection process.*



*Figure 1-1: Graphical abstract - Food packaging towards sustainability.*


## Keywords

*Food packaging*
*Environmental challenges*
*Sustainable developments*

## 1.1 Food Packaging

Food packaging refers to the materials and containers used to store, protect, and transport food products. Often the term food packaging is associated with the primary food packaging, besides referring also to secondary and tertiary packaging. Primary food packaging is the packaging that comes into direct contact with the food product, also called sales packaging. Its primary functions are protection against food contamination and damage. Secondary packaging refers to overpacks, a packaging that is used to group or contain primary packages. Its primary function is to facilitate handling, transport, and storage of the primary packages. Tertiary packaging refers to the packaging that is used to transport and store large quantities of secondary packages. Its function is to protect the secondary packages during transport and to facilitate storage and handling in warehouse and distribution centres. As this work focused on primary food packaging for simplification reasons the term food packaging from now one refers to primary food packaging, when not otherwise mentioned. Food packaging has multitude essential tasks as protection and preserving during storage and delivery of food from the producer to the consumer. It can simplify the product usage and functions as information and communication platform between producers and consumers. Food packaging can be made from a wide variety of materials, such as plastic, paper, metal, glass, or combination of those, and can come in many different colours and forms, such as bags, boxes, bottles, or cans. The type of packaging used depends on the type of food being stored, and transportation system used, as well as other factors such as cost, convenience, safety, consumer preferences, industrial processing properties and environmental aspects. Whereby most products nowadays are packed in plastic packaging mainly due to technological and economic reasons. The complexity of food packaging is high and important elements are inter alia material type, material composition, colours, additives, labels, inks and adhesives (Robertson, 2012). Food packaging is an interdisciplinary topic, which is represented by the variety in products to pack, big range of packaging materials, different transportation systems, supply chains, or storage conditions. Food packaging is a core element of the food industry and without the advances in food packaging the current way of living and global food chain would not be possible.

## 1.2 Challenges in food industry & packaging sector

The food industry faces a number of global issues, including food safety, changing consumer behaviour, consumer environmental awareness, population growth, population distribution shift towards more elderly people, supply chain disruptions and government regulations (Han et al., 2018; United Nations Department of Economic and Social Affairs, Population Division, 2022; Yan et al., 2022). In regard to food packaging, especially the challenges of food waste, dependency of fossil raw materials and packaging waste are crucial (Figure 1-2).

*Figure 1-2: Challenges of the food industry.*

### 1.2.1 Food losses and waste

Food losses and waste is a major global problem that has social, environmental and economic consequences (Council of European Union, 2016). Food losses and waste refers to loss or disposal of food that is still edible or usable and can occur at all stages of the food system from production to consumer level. The sustainable development goal report of the United Nation states the world's food loss at around 13% after harvesting and before reaching retail markets, and the total food loss at around 17% at retail levels, household or food services (United Nations, 2022). Causes include inefficient agricultural practices, overproduction, supply chain inefficiencies, consumer behaviour, inadequate storage, and transportation infrastructures. By reducing food waste, simultaneously the pressure on feeding the growing world population is reduced and the use of the full potential of available resources is ensured. Regarding the environment, food waste has a significant environmental impact, as it contributes to greenhouse gas (GHG) emissions and implies a wasteful use of scares resources such as water, land, and energy. When food waste ends up in landfills, it produces methane, a potent GHG. GHG emissions related to food ending up in landfills was estimated as 8 to 10% of global GHG emissions (United Nations, 2022). Additionally, food waste leads to economic costs, estimated to be around USD 990 billion annually (Council of European Union, 2016). It represents a loss of resources and money for both consumer, businesses, and communities. There are many ways to address food waste, including reducing food waste at the consumer level by planning meals, shopping wisely, and using leftovers, improving supply chain efficiency, and supporting policies and programs that promote food recovery and redistribution (European Commission, 2023). Nevertheless, also packaging plays a valuable role in minimizing food loss and waste, by ensuring a safe and easier product distribution, increasing shelf life and providing a communication platform about product usage to the consumer (Brennan et al., 2021).

### 1.2.2 Dependency of fossil raw materials

The packaging sector has historically been dependent on fossil raw materials such as oil and gas, which are used to produce plastic packaging, but also metals for cans or tubes. Another indirect fossil dependency of the food industry is the amount of energy required for producing food, producing packaging or recycling packaging materials. Especially, the production as well as recycling of metals is an energy intensive process (Riedewald et al., 2022). The use of fossil-based materials for packaging has several drawbacks, including environmental impact, finite resources, linear supply chains, releasing of captured $CO_2$, prize volatility and dependency on fossil markets (Karin Molenveld et al., 2015). Especially, increasing uncertainties such as supply chain disruptions or prize changes due to political developments are challenges for packaging companies and the whole food sector. Consumers are increasingly concerned about the environmental impact of packaging, and continuing business as usual with fossil-based materials, can provide an image of disinterest, and in the worst case can harm a company's reputation or brand image. Ways to address the fossil dependency of food packaging materials are the use of renewable materials which can be seen as carbon neutral, or reducing the need for virgin materials by ensuring that the materials stay in the material cycle by reusing or recycling practices.

### 1.2.3 Packaging waste

Every food packaging is disposed after they serve their indented purpose, resulting in a waste generation of around 80 Mt in 2020, which are in average 178 Kg/a per EU inhabitant (Eurostats, 2023). Not only the food packaging littering is an issue, as it can harm wildlife and ecosystems (United Nations Environmental Programme, 2021), but also the tremendous amounts of food packaging waste that needs to be collected, sorted and handled in landfills, incineration or recycling facilities. Packaging accounted for one-third of all waste collected at kerbside in Ireland in 2019, accounting for 1,1 Mt (EPA, 2021a). Depending on the end of life (EoL) option the environmental impact can vary significantly. The EoL cycle of a packaging depends on a combination of various factors, including materials used, packaging design, consumer disposal behaviour, local waste management infrastructure, as well as local, national, and international regulations. The food packaging waste hierarchy gives an overview of the preferable order of waste treatment options for EU, whereby other sequences can be more suitable for other regions: reduce, re-use, recycle (mechanical, chemical, biological), energy recovery and disposal in landfilling (Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008). Whereby, the uncontrolled release of food packaging waste into the environmental should be avoided at every cause. In general, the most recyclable materials at present are paper and cardboard packaging (82%), metallic packaging (77%), glass packaging (75%), plastic packaging (41%) and wooden packaging (31%), resulting in an overall recycling rate of municipal packaging waste in the European Union (EU-27) in 2019 of nearly 65% (Statista, 2023). Whereby, the recycling rates differ significantly between countries. Packaging waste is an important challenge to overcome. The awareness of the consumers regarding packaging waste issues rises and imposes pressure onto the market and governments to reduce environmental issues related to food packaging (Brennan et al., 2021).

## 1.3 Trends in food packaging

Food packaging trends are pursuing the aim to reduce food waste, decrease fossil dependency and reduce food packaging waste by investigating the potential of renewable, biodegradable, or recyclable materials. Consumer trends towards convenience and eating on-the-go is also driving the development of new packaging formats that are portable, reusable, edible, and easy to use or provide portion control. Other research efforts are placed on investigating new packaging systems, as active or intelligent packaging. Additionally, a growing interest was reported in packaging that offers personalisation, as tailored to the need of a specific consumer group, transparency, and interactive experience for consumers, such as interactive QR codes that provide information about the product, packaging, and company environmental commitments. The best packaging for reaching zero environmental impact is no packaging, known as ‘nude food’. However, no packaging is only an option for few food products, food services or supply chains. Often food packaging is essential to provide high quality and safe food to consumers. Overall, development trends in food packaging are innovative packaging solutions that can improve safety, convenience, customization, and sustainability of food packaging. Design for circular economy, eco-design, biobased materials, edible packaging, reusable food containers, active and intelligent packaging, and recyclability, are relevant packaging trends and are briefly discussed further. Additionally, food packaging regulations are discussed as they play an important role in driving innovations.

### 1.3.1 Eco-packaging design

Ecological (Eco) stands for an interaction of an organisms or object with the environment. Eco-design is approached with the intention to decrease the environmental impact of a food packaging (Lewis, 2008). Eco-design not only focuses on environmental impact, but also covers social aspects and business viability, taking the entire life cycle into consideration. Thus, covering the three pillars of sustainability - people, planet and profit - as defined in the Brundtland report (United Nations, 1987). Food packaging designed with the aim to be safe for the environment or reducing negative environmental effects compared to a traditional packaging falls into the category of eco-packaging. It is a continuously evolving process and can be achieved by different developments. In terms of food packaging, the term “eco” could refer to reusable, biodegradable, recyclable, compostable or edible packaging considering the packaging end of life phase. Improvements in early life cycle steps, as the use of biobased materials or improving production technologies by the reduction of hazardous chemicals or energy demand, have also been reported. Many eco-design strategies exists, whereby the advantages are often measured on theoretical assumptions or are focused on a specific part of the life cycle. A challenge is the evaluation of the effect on the whole life cycle, and by considering existing supply chain scenarios. For example, biodegradable products can reduce packaging waste, but can potentially increase methane production during decomposition or even could contaminate current recycling streams. The term eco covers an elastic term, where no guideline or regulations are yet defined (GIZ, 2020).

### 1.3.2 Circular economy design

Circular economy aims at reducing primary resource extraction by using all materials at their EoL phase (Ellen McArthur Foundation, 2022). Ensuring that materials either stay in the same material cycle or enter a new cycle, with the priority to keeping products, components and materials at their highest value at all times. In an ideal circular economy no primary resources shall enter production processes, only secondary (recycled) materials. Therefore, a precondition is that products must be dismountable and further treated in an appropriate EoL option. More specific for food packaging it involves, reuse, collection, sorting, recycling and the use of recyclate as raw materials in the same or another material cycle. The transition towards circular economy in the food sector already starts at the packaging design stage, as here considerations for EoL treatments and recyclability can be taken into account (GIZ, 2020). EoL treatments includes mechanical, chemical or biological recycling of packaging materials, but available sorting, and recycling options at the place of the packaging market should be favoured. An often stated gap in the system of circular economy is the uncertainty of saving environmental pollution due to recycling. In some cases the recycling process is more cost intensive and the use of raw materials is more economically, and/or environmentally. Regulations for food contact materials can limited the use of low quality recyclate in the same life cycle due to hygiene requirements. Developments towards a circular economy are bioplastics, packaging designed for recycling, optimisation of recycling infrastructure, or promotion of reusable packaging. With the overall aim to transform the mainly linear economy to a circular model characterized by make, use, reuse, recycle and repeat.

### 1.3.3 Design for Recycling

Design for recycling (D4R) is a design principle to improve large-scale recyclability, in order to achieve a significant impact. By extending packaging requirements focusing on product quality and safety, to ensuring recyclability of the packaging in existing waste management structures. D4R aims to prevent waste generation due to recycling and the creation of new high quality material streams that could be used as a raw material for the same or another product. Benefits can be achieved at different levels, the society as a whole by increasing recycling rates and reducing waste accumulation, the recycler by achieving more high quality feedstock, or the producers by reputational benefits or extended producer responsibility savings (Triolett and Alvarez Martinez, 2021). The prerequisites for a successfully implementation of D4R is first a broad knowledge about the technical recyclability of the used materials, and secondly understanding the actual recyclability by conveniently available collection, sorting and recycling systems in the areas of sales to a reasonable proportion of consumers. Recyclability of a packaging is defined by the ISO 14021, Sec. 7.71 as 'A characteristic of a product, packaging or associated component that can be diverted from the waste stream through available processes and programs and can be collected, processed and returned to use in the form of raw materials or products'. Important information's are inter alia large-scale collection, sorting and recycling processes and technology established, shares of material in the overall waste volume, value of waste streams or recyclate, and packaging design choices that can impact the quality of recyclate. For a successful implementation a cross-sectoral cooperation and transparency between packaging manufactory, food sector, recycler and recyclate users is essential.

Many D4R guidelines are available (Table 1-1), especially for the food sector with different prioritizations as material groups, food product groups or packaging systems (Watkins et al., 2020). These guidelines evolve with developments on packaging product design, recycling technologies, sorting systems or recyclate market developments and can significantly differ based on domestic waste streams and local recycling sectors. D4R is an instrument for manufacturers in the food industry to improve the theoretical recyclability of packaging place on to the market by referring to the actual waste management process but cannot ensure that the sorting and recycling is actually done.

Table 1-1: Design for recycling guidelines with the focus on material group, food products or packaging systems.

| Guideline | Source |
|---|---|
| EPR toolbox | (Brinkmann et al., 2022) |
| 4Evergreen campaign for optimization fibre based packaging circularity | (4evergreen, 2022) |
| Better recyclability of paper and board packaging | (WRAP, 2020) |
| Design for Recycling – State of play | (GIZ, 2020) |
| Packaging & Design for the circular economy | (REPAK, 2020) |

### 1.3.4 Biopolymers

Biopolymers is a general term, and relates to biobased, biodegradable and/or compostable packaging materials (Han et al., 2018). These are different concepts as biobased reverse to the origin, whereby biodegradable and compostable to the EoL of the material. Whereby, one is not a condition for the other, or *vice* versa. Bioeconomy is one trend to tackle the current challenges related to food packaging.

#### *1.3.4.1 Biobased materials*

Biobased polymers are made in whole or partially from renewable biological resources (Ashter, 2016). The use of biobased materials as raw material source for food packaging follows the aim to reduce the dependency of fossil resources. It is considered as carbon neutral, based on the principle that the carbon dioxide released during biodegradation or incineration of the materials can be considered to be reabsorbed by the next crop that is farmed to produce more biopolymers (Parker, 2008). The term biobased in relation with food packaging was studied to have an increasing consumer acceptance and resulting in a more positive packaging image (Karin Molenveld et al., 2015). The bioplastic market showed a steadily increasing demand in recent years. Biopolymers can be produced by chemical synthesis starting from renewable biobased monomers (e.g., sugar cane), or natural polymers sourced from vegetal or animal sources such as polysaccharides (e.g., starch, alginate, cellulose, or chitosan) and proteins (e.g., whey, soy, gluten, gelatine). Bioplastic as bio-PET, bio-PE, bio-PP, PLA, PBS, and PHAs or material based on biopolymers as starch or cellulose are available at industrial scales and find applications for food packaging. Whereby, others are at development level due to necessary performance improvements (Ashter, 2016).

Points of criticism for biobased materials are potential increasing pressure on agriculture crops for food and packaging, pressure on sustainable agriculture practices, high costs and reduced compatibility with the current waste management system. Biobased materials are theoretical recyclable, but the actual recycling is low. If the sorting facilities are not able to separate biobased materials from current recycling streams, it can come to contaminations of existing high value recycling streams. Only few recycling schemes are available for this kind of materials and the driving force for investment is low due to small amounts of biobased materials in the recycling materials (Ashter, 2016; Parker, 2008). Excluding biobased materials with the same chemical composition as synthetic materials (bio-PET or bio-PE) which can be recycled in the same way as the material originated from fossil sources. An additional, important consideration for biobased materials is that the crops themselves are renewable, but the production framework is reliant on fossil fuels for fertiliser production, power farm machinery, transport, or electricity. Therefore, not only a switch from fossil to renewable in the material is necessary but also developments in other sectors to have a real carbon neutral material. The principle of using biobased materials will play an important role in the development toward environmentally friendly packaging systems. The development needs to be further supported by incorporating biobased materials in standard waste management systems and increasing the market share (Parker, 2008). Also, regulations favouring this strategy will play a significant role in promoting biobased packaging materials.

#### *1.3.4.2 Biodegradable and compostable materials*

Materials are considered biodegradable when materials are rapidly broken down under the influence of microbes under defined conditions (European Environmental Agency, 2023). When a complete microbial assimilation of the material happens within 180 days in an industrial compost environment, it is considered as industrial compostable (Ashter, 2016). Biodegradable polymers used in food packaging are PLA, PHA, zein, soy protein isolate, starches, cellulose, gluten, whey protein or chitosan and demonstrated a growing market interest (Peelman et al., 2013). Biodegradable or compostable materials are interesting for reducing packaging waste accumulation. They are particularly seen as a solution for packaging products that are difficult to recycle as film laminates, strongly contaminated with product residuals or difficult to separate from the product (Karin Molenveld et al., 2015). Packaging materials with a biodegradation period similar to the food product shelf life avoids the use of materials with a degradation time of several decades. The concept of biodegradation and composting requires rethinking of a durable packaging, to a packaging with an own shelf life. Due to the required behaviour change it is more difficult to implement biodegradable or compostable materials in the current food and packaging life cycle.

#### *1.3.4.3 Edible packaging*

Edible packaging concepts following the aim to either apply an edible layer to the product surface (edible coating) or surrounding the product with an edible film that proves product protection, and increased the shelf life (Dehghani et al., 2018). The concept of edible packaging tackles food industry challenges by reducing the need for synthetic packaging materials by being biobased, avoiding the generation of packaging waste, and reducing food waste. Edible packaging is already commonly applied to fruits and vegetables, or for fast moving consumer goods and eating on the go products. Whereby, the use for other food groups is under research. Edible coatings can be safely consumed with the product, and edible films wrapped around the product can be either consumed, or can be disposed in the organic or normal waste bin (Dehghani et al., 2018). Although edible packaging is not meant to entirely replace traditional packaging, the adequacy of food protection can be improved as well as new packaging material combination can be explored by combining an edible coating and non-edible biobased or synthetic packaging. The second packaging part is required for handling, hygienic reasons and product distribution (Dehghani et al., 2018). An important aspect for the development of edible packaging is consumer communication to show increasing importance of new packaging systems and improving consumer acceptance and trust.

### 1.3.5 Regulations and commitments

Regulations and government strategies are an important driver for lowering food packaging challenges. Setting a driving force for industry to use eco-friendly-sustainable packages. There are several local, national, and international food packaging regulations in place promoting safer, more sustainable, and more responsible packaging practices. Regulations can help to incentivize producers to contribute to reducing food packaging, packaging waste and invest in sustainable packaging solutions. The regulations may include requirements for the use of recyclable materials, minimum recycling rates and implementation of waste reduction strategies. Regulations can favour developments into a certain direction or can set ultimatums. A resent example is the ban of certain single-used-plastic packaging components in the EU (EU, 2019). Another example is extended producer responsibility schemes, which require producers to take responsibility for the EoL management of their packaging products to increase food packaging recycling. The EU strategy for reducing packaging waste and reducing the raw material dependency is stated in the EU packaging waste directive. Hereby, the main focus is on preventing packaging waste and on implementing a circular economy system by ensuring open or close life cycle loops to promote reduce, recycling or other forms of recovering of packaging waste (Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008). The directive defines recycling targets for EU Member States for all packaging of 65% by 2025 and 70% by 2030, additionally specific targets are set for each packaging material section within the overall targets. The EU follows the strategy of recycling with the advantages of reducing the dependency of virgin material needed, using, and developing existing waste management facilities, generating markets for recyclate by focusing on specific material groups to ensure high material streams for a high recyclability. Beside government regulations, also commitments of industrial alliances can impact the road map for reducing packaging environmental impacts, often outperformance regulations target settings. In the new plastic economic global commitment it was agreed to innovate all plastics needed to be designed to be safely for reuse, recycling or composting by 2025 (Ellen McArthur Foundation, 2022).

## 1.4 Food packaging development

The development of food packaging, including primary, secondary, and tertiary packaging, is a complex process where many disciplines, technologist, engineers, logistic, marketing, and sales professionals; are involved. The selection is often an interdisciplinary process finding a balance between different requirements from safety & quality, engineering & manufacturing, marketing & sales, and regulatory & economic point of view presented in Table 1-2 (Coles, 2003; Robertson, 2012). The prioritisation of environmental aspects is growing in recent years due to discussed environmental concerns related to food packaging. Packaging selection involves many considerations whereby the overall aim to protect the food product during the supply chain and ensure a safe and high quality product to guarantee sufficient shelf life and avoid food waste, should not be lost. Within this thesis the step of the primary packaging material selection based on technological and sustainable requirements were further reviewed. Different strategies exist, which can be broadly categorized into qualitative and quantitative approaches.

Table 1-2: Overview of potential technical, cost, marketing, legislation and environmental packaging requirements.

| **Product requirements** | **Technical requirements** | **Cost** | **Marketing aspects** | **Legislative requirements** | **Environmental aspects** |
|---|---|---|---|---|---|
| Oxygen barrier<br>Water barrier<br>Grease resistance<br>Light barrier<br>Chemical resistance<br>Odour barrier | Surface qualities<br>Thermal properties<br>Electrical properties<br>Mechanical properties<br>Available machinery | <u>Direct costs</u><br>Packaging material<br>Packaging process<br><u>Indirect costs</u><br>Storage<br>Transportation<br>Energy consumption<br>Labour<br>Line efficiency<br>Investments<br>Sales<br>Quality control<br>Waste | Aesthetics<br>Physical properties<br>Perceived quality<br>Convenience<br>Displaying properties<br>Communication<br>Design & branding | Food hygiene regulations<br>Labelling regulations<br>Food contact regulations<br>Migration regulations<br>Environmental regulations | Energy use<br>Raw material production<br>Ratio of package weight to product<br>Reusability<br>Recyclability<br>Biodegradability<br>Waste production |

### 1.4.1 Qualitative approach

In the qualitative approach the barrier properties are selected based on comparable products on the market, packaging manufacture or product manufacture experiences. The packaging systems with the product of interest is mostly tested with a “trial and error” approach. Which can result in sub-optimised packaging systems and significant need of capital expenditures (Coles, 2003; Robertson, 2010). Another difficulty is the identification of the used packaging material of a comparable product, due to lacking transparency of the material used. Within the EU, Italy recently obligated the resin identification code on food products. Nevertheless, the resin identification code only identifies the main component of packaging. Detailed information about the real packaging composition is still lacking e.g., when multi-layer films are used as barrier properties depend on the thickness of each layer. The qualitative approach comes with an error term as it is based on several assumptions in the selection process. When selecting barrier properties based on other food products on the market the identified packaging requirements are not based on the actual product properties of the product of interest or designed for the individual distribution chain. It also relays on the assumption that the comparable product was optimally designed. Overprotection is a common outcome of the qualitative approach, leading to less environmentally friendly packaging systems. This practice is a common strategy used in industry (Coles, 2003) often due to lack of required resources. This is signalling a huge improvement potential.

### 1.4.2 Quantitative approach

In a quantitative approach the barrier properties are selected based on experimentally obtained data. One quantitative strategy for the identification of barrier properties is by experimentally measuring the material properties of existing food product packaging. This approach is an extension of the qualitative approach and allows to narrow down the estimated required range of the barrier properties. Nevertheless, this approach only gives information of the barrier requirements of commercial marketed products and are not based on the requirements of the product to be packed. Which could lead to an overprotection of the product and could narrow down the range of possible suitable materials. Another quantitative approach is by investigating the required barrier properties based on the product to be packed and the individual supply chain, known under “shelf life approach”. In this approach the shelf life of the product is identified, degradation processes are investigated, and other relevant product and supply chain information are gathered. Information is then processed with the aid of a mathematical model to investigate the required barrier properties. The shelf life approach allows a specified material selection based on selected quantitative details about the required packaging properties based on product shelf life (Robertson, 2010). Advantages are inter alia reduction of resources, minimizing of physical testing's and less required capital expenses. Additionally, system properties can be adjusted, or new packaging systems can be investigated prior to physical testing.

Chapter TWO

# Life cycle assessment methods for investigating novel food packaging systems

## Abstract

*High volume of plastic waste generated and potential harm to wildlife and ecosystems, are negative consequences from poor food packaging design and poor end of life food packaging management. Nowadays, an essential part of designing a food packaging is to minimize its environmental impact, a significant challenge for industry. The aim was to examine existing life cycle assessment (LCA) approaches for investigating environmental impact of novel food packaging systems in the field of ready to eat (RtE) fish and meat products by using a systematic review approach. The results showed that different scopes were used, resulting in different conclusions. Some included food products, others focusing on comparing packaging materials or packaging concepts. Either considering direct impacts, or considering indirect impacts. LCA approaches reviewed, showed how different focuses could be combined and used as sequential steps in getting a comprehensive understanding of the environmental impact of a food-packaging system. By considering a holistic LCA approach and comparing environmental performance of different packages, industry stakeholders and consumers can make informed decisions, taking an active role that balance necessity, wastefulness, creating efficient and sustainable packaging solutions.*


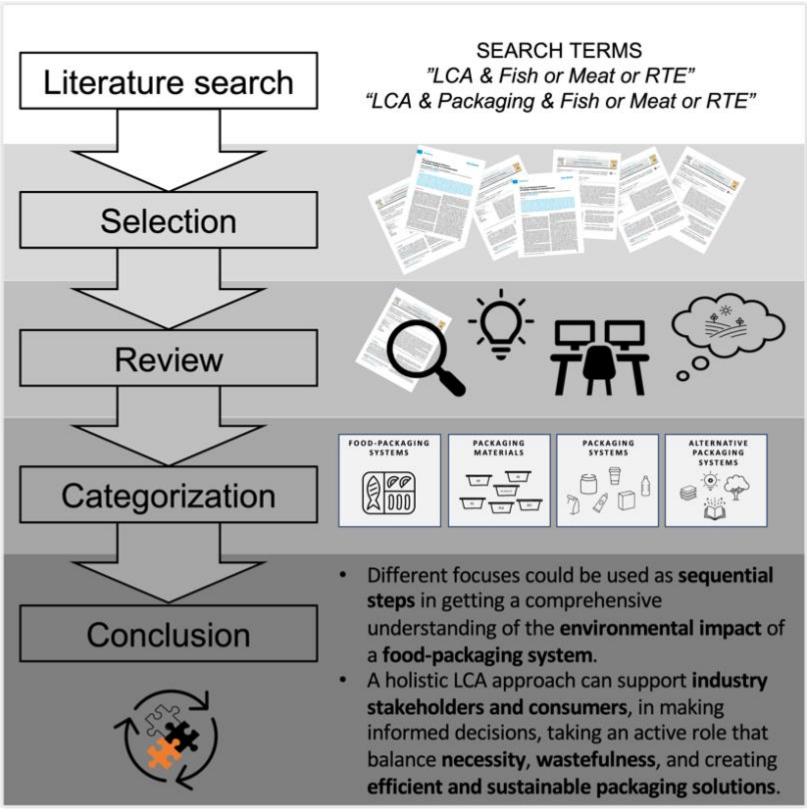


*Figure 2-1: Graphical Abstract - Life cycle assessment methods for investigating novel food packaging systems.*


## Keywords

*Life cycle assessment methodologies*
*Food packaging*
*Novel food packaging developments*
*RtE seafood products*

## 2.1 Introduction

Improving the packaging environmental impact is a challenging task and life cycle assessment (LCA) is a tool, which can guide and support the decision-making process for an eco-packaging design. A design where the sustainable performance of the packaging is included in the core requirements of a packaging (Jepsen et al., 2019). Successful and sustainable innovations depend on a clear understanding of the impacts and benefits of packaging system throughout the whole life cycle. Attributes as biobased, recyclable or biodegradable, where proven as no direct indicators for reducing life cycle environmental impact of food packaging (Vendries et al., 2020). The environmental sustainability of a product or process can be quantified by using LCA. Additional sustainable categories which are not included in LCAs, which are the other two pillars of sustainability as economic (life cycle costing – LCC) and social (social life cycle assessment – SLCA), can be also included to provide a holistic sustainable performance investigation (Verghese et al., 2012). Nevertheless, studies combining all three aspects are difficult to perform and are less widely studied (Zira et al., 2021).

LCA are defined in the ISO standard 14040 (BS EN ISO 14040, 2016), focusing on the principles and framework, and 14044 (ISO 14044, 2006) giving more detailed requirements and guidelines. Further instructions on LCA based on the ISO 14044/44 are given in the International References Life Cycle Data System Handbook (European Commission - Joint Research Center, 2010) or the handbook on LCA Operational Guide to the ISO standards (Guinee et al., 2002). The LCAs are divided into four steps according to ISO 14044, which are interrelated throughout the entire assessment and each fulfilling an important role. 1) Goal and scope definition: defining the functional unit, system boundaries, impact categories and geographical scope. 2) Life cycle inventory analysis (LCI): collection of data to meet the objective of the LCA study. 3) Life cycle impact assessment (LCIA): converting the collected LCI into related environmental impacts. 4) Interpretation: summary of the LCI and LCIA, sensitive analysis, conclusions, and recommendations. LCA can be used with different focuses, concerning developing an eco-food packaging; it can be used to investigate environmental hotspots of a packed food product, understand the impact of a packaging material, or comparing the environmental effects of alternative packaging systems to a benchmark product. LCA has evolved in recent years and were further standardised, but limitations as complexity of the analysis, the comparison of results, and the required full transparency of the selected methods, data sources and results are hurdles (Lehtinen et al., 2011; Verghese et al., 2012).

When developing eco-packaging solutions it is important to investigate the environmental influences of the purposed eco-design option to minimize the environmental impact of a packaging material, packaging system or food-packaging system. Almeida et al. (2021) highlighted that food packaging systems that can improve product shelf life and at the same time can limit the negative environmental impact of the food packaging are of growing interest. LCAs can generated valuable outputs and can support the decision-making process about more sustainable packaging. However, there are many different approaches to investigate the environmental impact of a food packaging. The aim of this state-of-the-art review was to study the applied LCA approaches to support the development of novel eco-packaging solutions with a focus on solutions for ready to eat (RtE) seafood products. A strategical performance literature review was performance and based on the found research papers the focus points of this chapter were divided into four subsections: 1) LCA studies focusing on food-packaging systems, 2) LCA studies comparing different packaging materials, 3) LCA studies comparing different packaging systems, and 4) LCA studies with alternative innovative (novel) packaging systems.

## 2.2 Experimental procedures

A literature search was carried out by investigating publications in peer-reviewed indexed journals through electronic databases (Scopus, Google Scholar and Science Direct). Only publications in English and published between 2014 till 2023 were considered. The search terms used were combinations of LCA and fish, meat or RtE food products, and/or packaging. An overview of the found studies within the selected search words for each electronic database are shown in Figure 2-2. Studies in the range of three digits could be found with the search words 'meat and LCA' or 'fish and LCA', whereby only studies in the range of two digits could be found for the search terms, 'meat and LCA and packaging' or 'fish and LCA and packaging'. Limited studies were found when using the search terms 'LCA and RtE' or 'LCA and RtE and packaging'. Giving a broad overview of the consideration of packaging in LCA studies and indicating a lack of research on LCA studies with RtE products. The gathered articles were further investigated and only considered when fitting into the previous described focus areas of this review.

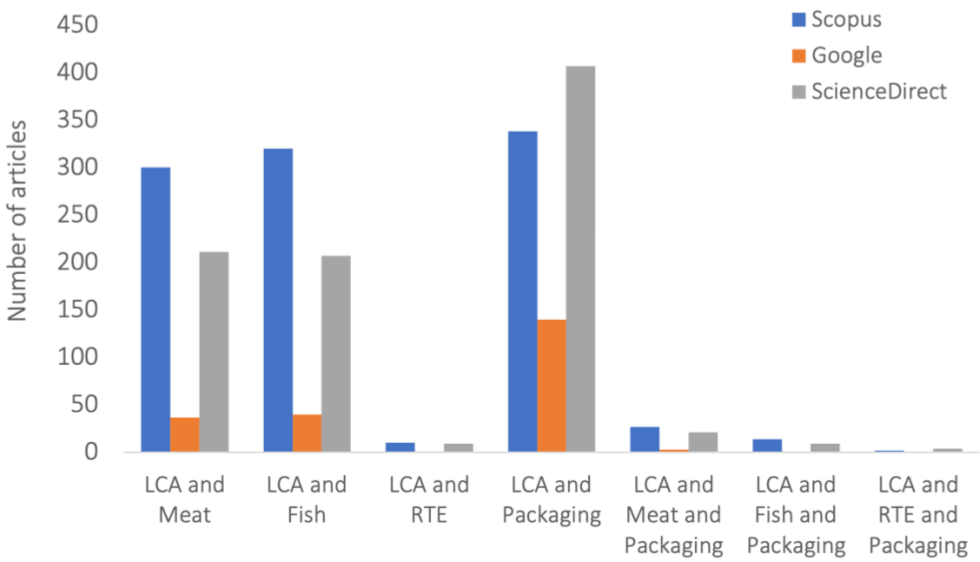


*Figure 2-2: Overview of search results per search platform* and selected search words**. Only articles from 2014 till 2023 and published in English were considered. (*Scopus: in title, keywords or abstract, all open access, Google Scholar: without citations, all in title, ScienceDirect: Title, abstract or keywords) (**"LCA" or "life cycle assessment" , "ready to eat" or "ready-to-eat" or "RtE").*

## 2.3 LCA studies focusing on food product

Only few studies were found focusing on the LCAs of the food supply chain, particularly in relation to RtE fish products and their packaging. A summary of the investigated LCA studies is presented in Table 2-2, highlighting functional units, impact categories, packaging details, system boundaries and main LCA conclusion regarding the packaging. The studies aim to understand the environmental impacts associated with these products and packaging throughout their entire life cycle and to identify environmental hotspots. The studies investigated the complete life cycle of a packed RtE food product with a system boundary from cradle to grave. The considerations encompassed various stages such as raw material sourcing, product production, transportation, retailing, use phase and end of life (EoL) phase. It was also noted that food loss and waste at different stages of the food life cycle should be included (Silvenius et al., 2014). Regarding the packaging, some studies included the direct impact of the primary packaging of the final product, other incorporated secondary, tertiary packaging, or intermediate packaging (Figure 2-3). Direct packaging environmental impact encompassed raw material sourcing, packaging material production, packaging production, and EoL material management. The packaging impact of RtE products packed in heavy materials, as glass and metal, was significant (Molina-Besch, 2016; Sieti et al., 2019; Vázquez-Rowe et al., 2014; Wiloso et al., 2022). When other packaging materials were used the share of the packaging on the overall impact of the packed product was low, and the contribution of product production was identified as the main hotspot (Molina-Besch, 2016).

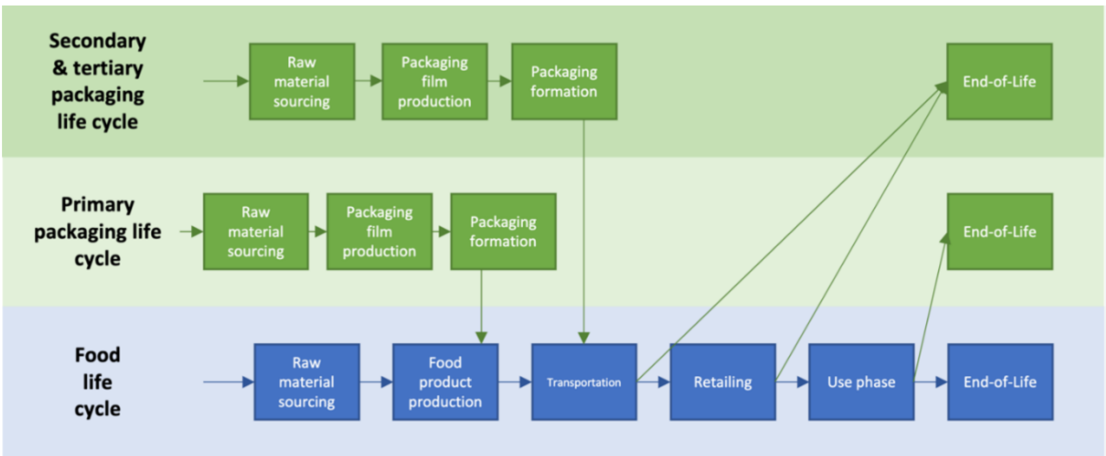


*Figure 2-3: Schematical overview of relation between direct environmental impacts steps of food and packaging life cycles.*

In general, it could be seen that the share of the direct environmental impact of a packaging of the total impact of the food-packaging system, can vary significantly within different LCA studies, mainly related to the considered product. The food to packaging ratio (FTP) in terms of greenhouse gas (GHG) emission of diverse products were compared by Heller et al. (2019) reporting large differences from 0.06 up to 700. Fish and seafood were identified beside dairy, cereals and meat as a food category with a higher FTP ratio or high environmental impact (Del Borghi et al., 2018). A high ratio is an indication that the impact of the packaging is minor compared to the impact of the product, and therefore changes in packaging configuration that led to food waste reduction would more likely result in net system decrease of the environmental impact, even when the packaging impact is increased (Heller et al., 2019). The investigated global warming potential for seafood products exhibited a wide range from 0.7 up to 31 $CO_2$-eq./Kg product (Table 2-1), attributed to factors as fish species, fishing methods, transportation, storage, or production. Evaluations of the environmental impact of packaging on seafood supply showed that the share of the packaging impact presents only a small portion of around 5%, which represents less than 1 Kg $CO_2$ eq./Kg, of the products climate change impact, and packaging represents in average 6% of the product weight (Molina-Besch, 2016).

Table 2-1: Overview of the environmental impact of RtE seafood and similar products.

| **Product** | **Environmental impact category** | **Environmental impact [Kg $CO_2$ eq./Kg product]** | **Source** |
|---|---|---|---|
| Fresh seafood products (chilled) | Climate change | 1.9 to 31 | (Almeida et al., 2021) |
| Fresh seafood products (herring to salmon) | Carbon footprint | 0.7 – 14 (average 3.2) | (Ziegler et al., 2013) |
| RtE baked tuna in tomato sauce | Greenhouse gas effect | 11.87* | (Zufia and Arana, 2008) |
| RtE surimi (minced fish paste) | Global warming potential | 1.3 – 7.1 | (Phungrassami and Usubharatana, 2020) |
| Fresh chicken | Global warming potential | 2.93* | (Kalhor et al., 2016) |
| Fresh fish: all species combined | Global warming potential | 4.41 | (Clune et al., 2017) |
| US beef (consumed, boneless) | Global warming potential | 48.4 | (Asem-Hiablie et al., 2019) |

*Adjusted to Kg $CO_2$ eq./Kg packed product

The investigated studies showed that the focus on food-packaging LCAs is to investigate the whole food-packaging life cycle, and to identify high environmental impact phases. Therefore, often only the direct packaging impacts were considered, whereby the consideration of indirect impacts were more comment when comparing packaging systems. The results highlighting the importance of the packaging to provide a sufficient product protection to avoid food waste. Nevertheless, considering the huge amount of product packed and the accumulated of used food packaging materials, also the reductions in the environmental impact of the packaging material can be significant.

Table 2-2: Overview of LCA studies of packed RtE seafood and similar products published in the last 10 years (2014-2023).

| Food product | Functional unit | Impact categories* | Packaging details | System boundary | Main conclusions on packaging | Source |
|---|---|---|---|---|---|---|
| RtE pork and bean stew | 1 Kg of prepared stew | 16 midpoint categories<br>1 end point category (carbon footprint) | Primary packaging of RtE product: steel cans or aluminium cans<br>Secondary packaging of RtE product<br>Primary packaging of intermediate products: cradle to gate, without EoL | Cradle to grave | - The tinplate employed to fabricate the metal cans is responsible for most of the impact associated with the packaging. Overall environmental savings can be achieved due to recycling of the tinplate.<br>- Substituting tinplate with aluminium is not recommended due primarily to lower environmental savings during aluminium recycling.<br>- recommendation to reduce packaging impacts are reducing weight, increasing the recycled content and/or increasing its recyclability. | (San Miguel and Ruiz, 2021) |
| RtE wet/dry baby porridge | Consumption of 1 porridge meal (125 g) | global warming potential, abiotic depletion potential of elements and fossil resources, acidification, and eutrophication potentials, freshwater aquatic ecotoxicity potential, Human toxicity potential, marine aquatic ecotoxicity potential, terrestrial ecotoxicity potential, Photochemical oxidants creation potential, ozone layer depletion potential | Primary packaging:<br>- dry porridge: plastic bag in cardboard box<br>- wet porridge: glass jars with metal cap with an aluminium and plastic lining | Cradle to grave | - packaging is only a hotspot for the wet porridge option<br>- Main hotspots for the wet product are manufacturing and packaging of raw materials<br>- using a plastic pouch instead of a glass jar would decrease most environmental impacts of wet porridge by 7-89%. | (Sieti et al., 2019) |
| Ready-made meals (e.g., Fisherman's pie) | Chilled ready-made meal for one person consumed at home in the UK | global warming potential, abiotic depletion potential of elements, fossil fuels, acidification potential, eutrophication potential, freshwater aquatic ecotoxicity potential, photo-chemical oxidation potential, ozone depletion potential, terrestrial ecotoxicity | primary, secondary, and tertiary packaging of RtE meal<br>Additional packaging stages: raw material packaging<br>plastic bag at consumption | Cradle to grave (Including food loss and waste) | - the impact of the packaging is below 10% in the tested impact categories beside for fossil with around 22%. | (Schmidt Rivera and Azapagic, 2019) |
| RtE dinner meals | One unit of RtE meal (507 g food) | Greenhouse gas emission, Energy use, waste generation | Primary, secondary, and tertiary packaging | Cradle to grave | - Packaging has a significant impact for the environmental impact of RtE products | (Hanssen et al., 2017) |

| | | | | | | |
|---|---|---|---|---|---|---|
| | | | | | - Consumer critics the overpackaging of RtE food products<br>- RtE meals had higher gross and net weight of packaging compared to the other meals.<br>- Packaging optimization recommendations for RtE meals: environmental improvements and fulfil consumers views | |
| RtE steamed Indonesian canned crab | 1 t of canned product at market | global warming, acidification, eutrophication, abiotic depletion | primary packaging: can, plastic cup, or pouch | Cradle to market | - The processing stage was found to be the highest contributor for most of the impact categories considered, mainly due to the use of tin cans for packaging.<br>- Environmental improvements can be achieved by substitution of packaging material from cans to plastic cups or pouches with an overall impact reduction of 70-85% per FU. | (Wiloso et al., 2022) |
| RtE cooked European pilchard (Sardina pilchardus) | Amount of protein supplied by one can of sardines in olive oil (equal. to 17.26 g protein) | climate change, ozone depletion; human toxicity; photochemical oxidant formation; particulate matter formation; ionizing radiation; terrestrial acidification; freshwater eutrophication; marine eutrophication; terrestrial ecotoxicity; freshwater ecotoxicity; marine ecotoxicity; agricultural land occupation; urban land occupation; water depletion; metal depletion; fossil depletion | Primary packaging: can<br>Secondary packaging: Cardboard boxes | Cradle to grave | - The packaging has a significant impact for canned products. | (Vázquez-Rowe et al., 2014) |
| RtE cooked Sardine | 1 Kg of edible product of canned sardine | abiotic depletion potential, acidification potential, cumulative energy demand, eutrophication potential, global warming potential in 100 years, ozone depletion potential, marine aquatic ecotoxicology potential, photochemical oxidation potential | Primary packaging: aluminium cans and boxboard<br>Secondary packaging: corrugated board boxes, pallets, and LDPE film | Cradle to factory gate | - Production of aluminium cans is the most significant process for all impact categories, except ozone depletion potential and eutrophication potential, resulting from the high energy demand and the extraction of raw materials.<br>- recommendation for optimising the environmental performance of packaging: replacing the packaging material | (Almeida et al., 2015) |

## 2.4 LCA studies comparing different packaging materials

In LCA studies of the whole food-packaging system it was demonstrated that the packaging only contributes to low percentage of the total environmental impact for RtE or fish products, whereby the main environmental impact is related to the food supply chain. Nevertheless, even when the environmental impact per mass of the product is low, huge amounts of products are produced and packed, accumulating food packaging waste (Foschi et al., 2020). Comparison of the packaging material alone can support decision-making processes regarding material selection and improvement of the environmental performance. Reviewed life cycle studies focusing on packaging material are summarized in Table 2-3. The most comment packaging materials used for food packaging are paper and paperboard, plastics, metals, and glass, in descending order of usage in weight in the EU. In the RtE market, plastic packaging is the most dominant one, as it provides diverse properties and can be tailored to the high product needs as oxygen barrier, water barrier or grease resistance. In this tailored approach, either mono materials or composite materials (blended or layered) were used. Common conclusions of the listed studies could be drawn, beside the differences in the materials investigated, impact categories, or system boundaries. Overall, the reviewed LCA studies focused on comparing the direct impacts of the packaging materials. The selected functional units were per square meter material, per tray or amount of material needed for a defined product volume. Only, two studies compared materials with similar properties (Hutchings et al., 2021; Toniolo et al., 2013), allowing the assumption of a similar product-packaging effect. Additionally, Hutchings et al. (2021) factored the indirect impact of packaging materials by selecting a functional unit as material thickness providing a similar product shelf life. This approach allowed to include direct and indirect packaging impacts and at the same time avoided the difficulties of finding a relation between shelf life extension and food waste reduction. The challenge was to find materials with equal barrier properties and comparable other functions to have a fair comparison (Silvenius et al., 2014).

It was shown that LCAs are a useful tool to assess the environmental influences of packaging materials, revealing high impact steps of the individual supply chains and differences between packaging materials. The main environmental impact of the packaging could be allocated to the material production and waste management process (Almeida et al., 2021). The use of recyclate or product waste streams can improve the environmental performance of packaging materials (Hutchings et al., 2021; Maga et al., 2019; Toniolo et al., 2013). Mono-material solutions should be preferred upon multi-material solutions, mainly to the non-recyclability of the later (Maga et al., 2019; Pauer et al., 2020; Toniolo et al., 2013). Although, recyclable packaging does not directly mean the most environmentally friendly packaging, as technical recycling does not automatically lead to actual recycling, especially for plastic films (Pauer et al., 2020). It was identified that the energy source used during production can have a significant influence on the LCA results. The use of renewable energy sources was shown to improve the environmental performance of foamy tray materials (Ingrao et al., 2015; Suwanmanee et al., 2013). Additionally, the amount of material used, and weight reduction was identified as the most important factor to improve the environmental performance of food packaging. It often even outweighs possible recyclability benefits (Maga et al., 2019; Pauer et al., 2020).

Comparative LCAs can be a useful tool to assess differences in the environmental performance of packaging materials to support decision-making processes concerning a more sustainable solution or to identify improvement options. Nevertheless, due to the significant higher impact of the product compared to the packaging, a clear priority for material selection is the product protection (Pauer et al., 2020), avoidance of food waste, and then followed by the packaging environmental performance. A LCA of packaging material can give useful insides for developing an eco-packaging solution, but also has limitations. The material LCA should be a complementary element of a LCA study where the complete food-packaging system is investigated, for a holistic view for the decision-making process in regard to an eco-packaging solution.

Table 2-3: Overview of recent publications on LCAs comparing packaging materials for meat, fish or RtE products.

| **Packaging materials** | **Functional unit** | **Approach** | **Ref.** |
|---|---|---|---|
| XPS closed cells, XPS open cells, XPS-EVOH, PS-EVOH, aPET, rPET, rPET-PE, PP and PLA | Tray (with/without absorption pad) with a volume of 1 L preserving 500 g meat | Cradle to gate with end of life approach | (Maga et al., 2019) |
| PS, PLA, PLA/starch | 10000 units of trays with a fixed dimension<br>(Different materials have different weights) | Cradle to consumer gate | (Suwanmanee et al., 2013) |
| Composite lidding films for MAP: LDPE/EVOH/LDPE vs. PHA/BVOH/PHA | Amount (g) of film required for 1 Kg of produce<br>A) with the same carbon dioxide transmission rate<br>B) CDTR providing the same shelf life | Cradle to grave (without packaging, retail, and consumer stage) | (Hutchings et al., 2021) |
| PA/PE film, PE/EVOH film, PA/PE bag, PA/PE bag, PE/PVdC shrink bag, PA/EVOH/PE shrink bag | 550 $cm^2$ multilayer film for packaging 500 g bacon product | Cradle to grave | (Pauer et al., 2020) |
| Foamy PS tray | 1 Kg of packed trays | Cradle to grave (raw material extraction, tray production, transportation, EoL) | (Ingrao et al., 2015) |
| Multilayer multi-material tray (PE/PET)<br>Multilayer mono-material tray (PET) | 1 tray with a sealed lid for sliced meat (volume: 0.54 L, 30 g) with similar properties | Cradle to grave (manufacturing films, tray production, transport, assembly and EoL) | (Toniolo et al., 2013) |
| PP film (commercial)<br>Chitosan film (lab scale) | 1 $m^2$ packaging film | Cradle to grave (material extraction, film manufacturing, EoL) | (Leceta et al., 2013) |
| PHA-TPS layered material (biodegradable) and PP (commercial) | 1 Kg of packaged product at the house | Cradle to grave | (Dilkes-Hoffman et al., 2018) |

* aPET: amorphous polyethylene terephthalate, BVOH: butenediol vinyl alcohol, CDTR: carbon dioxide transmission rate, EVOH: ethylene vinylalcohol, LDPE: low density polyethylene, MAP: modified atmosphere packaging, PA: polyamid, PE: Polyethylene, PHA: polyhydroxyalkanoates, PLA: Polylactic acid, PP: Polypropylene, PS: polystyrene, PvdC: Polyvinylidene chloride, rPET: recycled polyethylene terephthalate, TPS: thermoplastic starch, XPS: expanded polystyrene

## 2.5 LCA studies comparing different packaging systems

Packages cannot only differ in the material, but also in other design aspects as shape or size, e.g., a tray with a lidding film, a bag, or a tube. An overview of reviewed LCA studies comparing different packaging systems with the focus on RtE seafood products, or similar products are summarized in Table 2-4. The reviewed LCA studies considered a wide variety of different packaging systems, system boundaries, functional units, and used diverse approaches with the aim to compare the environmental performance of different packaging systems. There were many LCA studies focusing on the environmental sustainability, where only few studies include economic, or social sustainability aspects as life cycle costing analysis, consumer behaviour scenario analysis, consumer preference analysis, or circular analysis (Gutierrez et al., 2017; Wohner et al., 2020). It was highlighted that there is a need for a balance decision-making approach including all aspects by e.g., using multi-component analysis. Nevertheless, the focus on this review was on LCA studies.

The functional units were chosen based on the specific research goals and the aspects of the packaging system that the studies aim to evaluate. Functional units selected were packaging material amount, one unit of packaging system containing a specific amount of product, amount of product eating by consumer or amount of packed product. Comparing packaging systems based on the direct environmental impacts, which include aspects like material production, packaging production and EoL options, is a common approach in LCA studies (Banar and Çokaygil, 2009). This approach allows to understand the immediate direct environmental burdens associated with the different packaging choices. Whereby, indirect impacts based on different packaging systems were neglected. Another LCA approach was focusing on direct packaging effects by comparing packaging systems with equal packaging properties, and therefore assuming the same indirect environmental effects as product shelf life (Humbert et al., 2009; Schenker et al., 2021; Zampori and Dotelli, 2014). Schenker et al. (2021) purposed the approach to first study the direct environmental impact of the packaging material itself with a functional unit of 1 Kg packaging material, which then can be transferred to compare the direct impact of different packaging systems with a functional unit of one packaging unit to pack a specific product amount. Another purposed approach was to include indirect environmental packaging effects; beside considering direct environmental impacts as packaging not only influenced the environment impact of a food-packaging system by direct impacts, but also indirectly by interacting with the food supply chain (Molina-Besch, 2016). It has been under discussion if LCA studies only considering the direct impact of packaging are potentially coming to misleading conclusions regarding the effect of the packaging (Molina-Besch et al., 2019; Silvenius et al., 2014). Indirect environmental impacts were investigated in various extends. Potential indirect environmental impacts of a packaging are summarised in Figure 2-4. Multiply indirect effects combining packaging system design and consumer behaviour, including easy to empty, easy to clean, easy to separate, easy to fold, mass, containing a desired quantity, information about how to sort and preserving effect (shelf life) were considered in the study by Wikström et al. (2016). Other studies focused on a specific indirect impact as food loss reduction due to shelf life extension (Casson et al., 2022; Matar et al., 2021; Yokokawa et al., 2018), emptiability (Wohner et al., 2019b; Wohner et al., 2020), content amount (Silvenius et al., 2014) or consumer behaviour (Yokokawa et al., 2018); effect on transportation and storage phase due to packaging weight, shape (Calderón et al., 2010); or effect on product preparation at factory or at the consumer phase (Zampori and Dotelli, 2014).

The challenge when considering indirect packaging effects was the quantification of the relation between food packaging and indirect effects. Relations between packaging and indirect effects were drawn by using experimental data, literature data, mathematical models, or consumer surveys. Another approach was the investigation of break-even point or trade-off situation. Heller et al. (2019) calculated the relative increase in packaging system impact in two impact categories that could be afforded by a hypothetical food waste reduction of 10%, based on the food waste rate estimated from the U.S. department of agriculture. Wikström et al. (2019) concluded that a 1% reduction of red meat waste, would allow a threefold increase in the packaging impact without increasing the climate impact of the entire product-packaging system. The packaging in this study only contributed to 0.3% of the GHG emission of the product. A model for the calculation of the trade-offs between product protection, packaging environmental footprint, packaging recycling, and FLW was presented by Williams and Wikström (2011). A consumer survey of Swedish households determined that 20 to 25% of household food waste was related to packaging design attributes, including the attributes easy to empty and containing the correct quantity. When such attributes were considered from the standpoint of reducing food waste, the potential of packaging to improve system environmental performance may be realized (Heller et al., 2019). In relation to seafood or RtE products further studies about the relation between the effect on food packaging and product waste are needed (Almeida et al., 2021).

Besides, the variations in the LCA studies comparing different packaging systems, it can be summarized that different conclusions about the sustainability of a packaging systems can be drawn when only the direct environmental impacts or both, the direct and indirect impacts were considered. When only direct packaging impacts were reviewed design options to reduce material amount, improving transportation, or a switch to light weight options can have a higher positive environmental effect. When additionally indirect effects were included other packaging systems were favoured. As often packaging systems with the highest preservation properties, permitted the lowest food loss and lead to the lowest environmental impacts, especially for high impact food groups. The use of highly functional packaging systems was often justified by the counterbalance between higher direct environmental impacts and the indirect impacts as food loss reduction or energy savings (break-even-rate). Additionally, besides the importance of the indirect effects, it was shown that when assessing the eco-design of packaging systems also consumer behaviour and economic aspects were important. Ignoring consumer behaviour and preferences during packaging selection and only focusing on environmental aspects can be difficult to be suitable in the real market. Overall, it was highlighted that there are different important aspects in the LCA of packaging systems, as packaging weight, packaging functionality (format options), energetic mix used in the supply chain, logistic, food waste reduction, household waste collection system, selection technology for waste treatment and EoL options as recycling and incineration with energy recovery. Therefore, to identify an eco-design packaging options it is important to study the present individual case scenario.

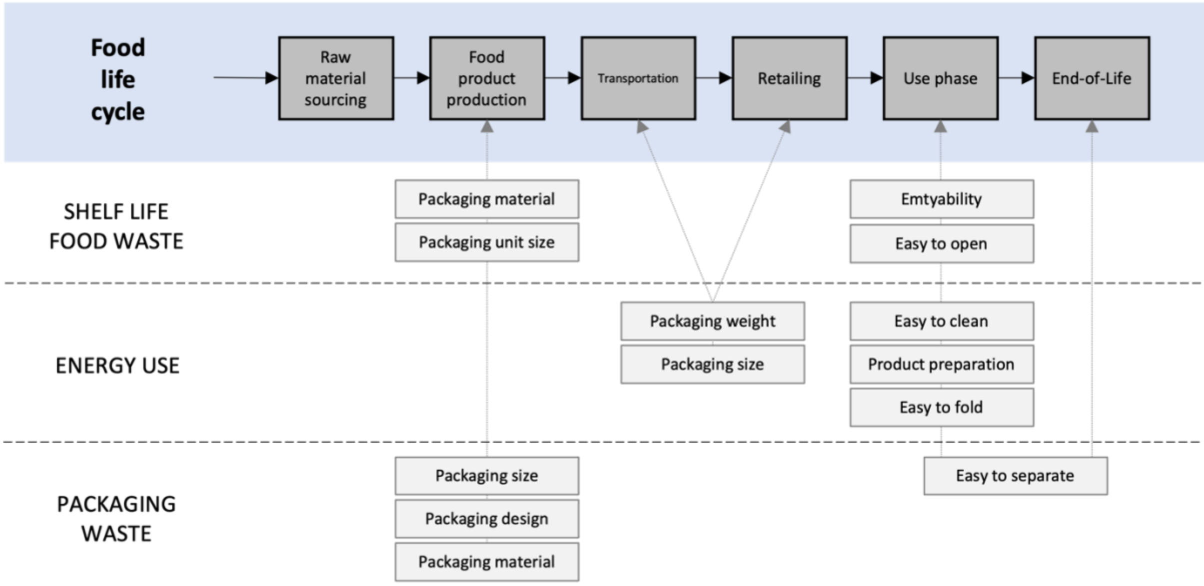


*Figure 2-4: Schematical overview of indirect environmental packaging impacts on food waste, energy use and packaging waste, and their site of action in the food life cycle.*

Table 2-4: Overview of recent publications on comparative LCAs studies for food-packaging systems with the focus on meat, fish and RtE products.

| Packaging systems | Functional unit | Approach | Impact categories | Source |
|---|---|---|---|---|
| Overwrap, high oxygen MAP, or vacuum skin packaging | 1 unit of packaging containing 500 g of sliced beef | LCA of packaging*1<br>LCA of food-packaging system (food waste reduction based on empirical model) | Abiotic depletion, global warming, ozone layer depletion, human toxicity, freshwater aquatic ecotoxicity, marine aquatic ecotoxicity, terrestrial ecotoxicity, photochemical oxidation, acidification, eutrophication | (Casson et al., 2022) |
| Cellulosic fibre-based stand up poaches, flexible flow wrap, food trays and moulded pulp vs. BOPP flexible flow wrap, OPP/PE stand up poach, PET thermoformed tray, PP thermoformed lid | 1 Kg of packaging material,<br>amount of packaging for a specific product amount | LCA of packaging material*1<br>LCA of packaging systems with equal protection functions for unspecified product | Climate change | (Schenker et al., 2021) |
| EMAP standard, EMAP optimized and MPP | 1 Kg of strawberries eaten by the consumer | LCA of food-packaging system (direct and indirect effects, mainly food waste and loss) | Acidification terrestrial and freshwater, cancer human health effects, climate change, ecotoxicity freshwater, eutrophication marine and freshwater, eutrophication terrestrial, ionizing radiation, land use, non-cancer human health effects, ozone depletion, photochemical ozone formation, resource use - energy carrier, resource use - mineral and metals, respiratory inorganics, water scarcity | (Matar et al., 2021) |
| 1) Coloured PP bottle with coloured PP cap, multilayer seal (PE/PET/adhesive/AL), PP label;<br>2) Clear transparent PP bottle, coloured PP cap, multilayer seal, and PP label;<br>3) Clear transparent PP bottle, coloured PP cap, multilayer seal, paper label;<br>4) Flint packaging glass, tinplate screw cap, paper labels | Per average consumption per capital in Austria (3.8 Kg consumed product) | A) Food loss quantification: determination of food waste due to poor emptability<br>B) LCA and LCC-VA<br>C) Combining the results of LCA and LCC-VA using multi-criteria decision analysis | Climate change, resource use, fossils, water use, eutrophication, freshwater, acidification, particulate matter | (Wohner et al., 2020) |
| Diverse dairy product packaging systems | 1 Kg of consumed product | Streamline LCA of food-packaging system (including food waste based on packaging emptability) | Acidification, respiratory effect, inorganics, climate change, eutrophication terrestrial and freshwater, resource use- fossils | (Wohner et al., 2019a) |
| XPS tray with film vs. high barrier vacuum skin pack | 1 Kg of food eaten by consumer | Food-packaging LCA (CtGr, direct and indirect effect: on food loss reduction at consumer phase)<br>Break-even rate calculation<br>Consumer behaviour scenario analysis | GHG emission | (Yokokawa et al., 2018) |
| PET tray with wrapping with air headspace vs. tray with wrapping with MAP headspace | Tray containing two cheesecakes (total 300 g) | A) Food-packaging screening LCA (CtGa + disposal of food and packaging, direct and indirect effects: shelf life extension)<br>B) economic and environmental based decision making | Climate change human health, ozone depletion, human toxicity, photochemical oxidant formation, particulate matter formation, ionising radiation, climate change ecosystems, terrestrial acidification, freshwater eutrophication, terrestrial ecotoxicity, freshwater ecotoxicity, marine ecotoxicity, agricultural land occupation, urban land occupation, natural land transformation, metal depletion, fossil depletion | (Gutierrez et al., 2017) |
| Tube vs tray | 1 Kg eaten minced meat | Simplified food packaging LCA (direct and indirect effects: consumer behaviour as easy to empty, clean, separate, and fold, | GHG emissions, acidification, ozone depletion | (Wikström et al., 2016) |

| | | | | |
|---|---|---|---|---|
| | | mass, sorting information, shelf life) | | |
| PS-based tray vs. Al- bowl | One tray (for one piece of poultry product) with the same function and same performance (shelf life) | Packaging LCA (CtGr, direct impact and indirect impact: energy use during cooking) | GHG emission, cumulative energy demand, climate change, ozone depletion, human toxicity, particulate matter, ionizing radiation, photochemical ozone formation, acidification, terrestrial-fresh water, and marine eutrophication, freshwater ecotoxicity, land use, water resource depletion, mineral, fossil & resource depletion | (Zampori and Dotelli, 2014) |
| Daypacks, glass jars, and steel cans | Packages for 1 t of olives for aperitif and cooking usage | Packaging LCA (direct impact) *1 | Climate change, human toxicity, particulate matter formation, fossil depletion and ionizing radiation | (Bertoluci et al., 2014) |
| Metal can vs. plastic retort pouches vs. plastic retort cup | Retail unit containing 80 - 85 g tuna | Food-packaging system (CtGr, direct impact) | Total carbon footprint, greenhouse gas emission | (Poovarodom et al., 2012) |
| Tin can, PP bag, PET bag, glass jar, PLA bag and TPS bag | 1 Kg of packaging material<br>1 Kg of packed product | A) LCA packaging material (CtGr)<br>B) Food-packaging LCA (CtGr, direct impacts and indirect impacts: packaging weight) | Land use, fossil fuels, respiratory inorganics, minerals, carcinogens, acidification/eutrophication, marine aquatic ecotoxicity, fresh water ecotoxicity, acidification, eutrophication, abiotic depletion, global warming, terrestrial ecotoxicity, climate change, ozone | (Calderón et al., 2010) |
| PP; tin - PE; and carton - PE | Packaging unit for 1 Kg cheese | Comparative packaging LCA (CtGr without consumer phase) | Cancer human health effect, respiratory effect, climate change, radiation, ozone layer, ecotoxicity, acidification potential, eutrophication potential, land use, mineral extraction, fossil fuels | (Banar and Çokaygil, 2009) |
| Glass jar, plastic pot | One baby food unit of 200 g, with equal properties (shelf life) | Packaging LCA (CtG) | Cancer human health effect, non-cancer human health effect, respiratory effect, ionizing radiation, ozone depletion potential, photochemical oxidation potential, ecotoxicity, terrestrial ecotoxicity potential, terrestrial acidification, land occupation, acidification potential, eutrophication potential, global warming potential, non-renewable cumulative energy, metal depletion | (Humbert et al., 2009) |
| PE/PP/PA/EVOH, Carton/PE/EVOH, APET/EVOH/PE, PE/EVOH/PET | 1000 Kg of each product consumed by the consumer | Food-packaging LCA (direct and indirect impacts) | Climate change, eutrophication potential, acidification potential | (Silvenius et al., 2014) |

*[1]LCA of packaging: material production, packaging production and EoL *[2] AL: aluminium, BOPP: biaxially-oriented polypropylene, CtGa: Cradle to Gate, CtGr: Cradle to Grave, EMAP: equilibrium modified atmosphere packaging, LCA: Life cycle assessment, LCC-VA: life cycle costing – value added, MAP: modified atmosphere packaging, MPP: macro perforated packaging, OPP: oriented polypropylene, PE: polyethylene, PET: polyethylene terephthalate, PS: polystyrene, PLA: Polylactic acid, TPS: thermoplastic starch, XPS: expanded polystyrene

## 2.6 LCA studies with alternative packaging systems

The identified studies with alternative packaging systems, which are innovative and novel packaging approaches, under the selected search terms are summarized in Table 2-5. The studies were reviewed with the focus on the investigated packaging system, used functional unit, and the applied approach. The studies investigated different types of alternative packaging systems, including active coatings applied to packaging films (Manfredi et al., 2015; Settier-Ramirez et al., 2022; Stramarkou et al., 2022; Venkatesh et al., 2018; Zhang et al., 2015), smart-active packaging (Stramarkou et al., 2022), and packaging with active nanoparticles (Vigil et al., 2020; Zhang et al., 2016; Zhang et al., 2019a). Similar functional units were selected, based on a defined packaging unit for a specific amount of product, but different approaches were used to study the environmental effects of the alternative packaging systems. Either only the direct impacts of packaging systems were compared (Venkatesh et al., 2018; Zhang et al., 2016), or a food-packaging system LCA approach including the indirect effects were applied (Manfredi et al., 2015; Zhang et al., 2015; Zhang et al., 2019a), or both approach were combined (Settier-Ramirez et al., 2022; Stramarkou et al., 2022; Vigil et al., 2020). Venkatesh et al. (2018) additionally, combined economic and environmental aspects for a broader approach. Different strategies were used to include indirect packaging impacts. Stramarkou et al. (2022) studied different waste reduction scenarios from 30% food waste generation with the conventional packaging, and for the alternative packaging system a food waste production of 5, 10 and 20% were assumed. Zhang et al. (2019a) used a survey approach to generate a function between shelf life extension and food waste. Zhang et al. (2015) calculated the break-even point to estimate the minimal required waste reduction of the tested alternative packaging systems for four impact categories, including global warming, fossil energy demand, acidification potential and eutrophication potential. Besides, the different approaches taken in the analysed studies, they agreed in their main conclusions. In nearly all studied cases, the additional packaging material increased the environmental burden of the packaging, whereby the amount depends on the additional material needed. Nevertheless, when the product-packaging system or the indirect impacts of the alternative packaging systems regarding shelf life extension and food waste reduction was considered, all alternative packaging systems showed an overall reduction in the studied environmental impact categories. The effect depended on the packed product, as shown by Zhang et al. (2019a) with an off set of the negative impact by 2.3 times for fresh fruits and up to 112 times for processed meat. It was highlighted that an important advantage of using alternative, novel packages was a higher product protection, which should also be valued when assessing the environmental impact. Including the shelf life extension is a decisive aspect when assessing the environmental impact of novel packages. Beside the challenges regarding waste estimation, it should be considered in all assessments of packaging solutions (Manfredi et al., 2015; Settier-Ramirez et al., 2022; Vigil et al., 2020).

Table 2-5: Overview of LCA studies with alternative packaging systems.

| Packaging system | Function unit | Approach | Reference |
|---|---|---|---|
| Bioactive bag (PE coated with active coatings: PVOH and nisin producing LAB) vs. conventional PE bag | Bag with a capacity of 200 mL (or 218 g pastry cream) | LCA of packaging<br>LCA of food-packaging system | (Settier-Ramirez et al., 2022) |
| Active packaging (OEO) and sensor vs. conventional packaging | Packaging container for about 10 Kg of sensitive food product | LCA of packaging<br>LCA of food-packaging system | (Stramarkou et al., 2022) |
| PLA coated film with NP (ZnO) vs. PP coated film with NP (ZnO) vs. PP film | Packaging unit for of 130 g of fresh cut lectures | LCA of packaging<br>LCA of food-packaging system | (Vigil et al., 2020) |
| Four nano-packaging systems for different food products | Amount of packaging to pack 1 Kg of food product | LCA of food packaging-system<br>with a trade-off calculation and a consumer study to investigate the relation between food waste and shelf life extension | (Zhang et al., 2019a) |
| Packaging film (PE) with alternative barrier coatings: Starch based, latex + kaolin, EVOH + kaolin PE | 1 Kg of films with the same functionalities | Partial LCA of packaging with focus of production and end of life | (Venkatesh et al., 2018) |
| PLA + silver NP, PLA + titanium dioxide NP, PLA + mixture of both | 1 Kg active packaging material that provides equivalent effectiveness to ensure food safety and quality | LCA of packaging | (Zhang et al., 2016) |
| Conventional MAP packaging (PP/EVOH) vs<br>PP/EVOH + active coating (thymol/carvacrol) | Packaging unit for 1 Kg fresh beef | LCA of food-packaging system (with focus on food waste) | (Zhang et al., 2015) |
| Tetra Top beverage container coated with active coating vs. Tetra Top beverage container | 1 L of consumed milk | Food-packaging system (with focus on food waste) | (Manfredi et al., 2015) |

*EVOH: Ethylene-vinyl alcohol-copolymer, LAB: lactic acid bacteria, NP: nanoparticles, OEO: oregano essential oils, PE: Polyethylene, PLA: polylactic acid, PP: polypropylene, PVOH: polyvinyl alcohol, ZnO: zinc oxide

## 2.7 Conclusion

LCA is a tool to support the decision-making process for improving environmental impact of a packaging. Only a packaging which reduces the environmental impact of a commercial or current used food-packaging system is a better solution for the environment, assuming that other core requirements as product protection, transport and communication are assured. This review emphasizes that approaches taken to investigate the environmental performance of a packaging can vary and support different developing stages.

A) The packaging can be investigated within a LCA study focusing on as supply chain of a packed food product. The fraction of the environmental impact of the packaging in the whole food-packaging impact can be identified. The focus was on direct packaging impacts such as raw material sourcing, packaging material production, packaging production, and EoL management. For products with a high environmental impact, the impact of the packaging represented only a small fraction of the overall environmental impact. This emphasised, the importance to protect the product and ensure that the products are used as nutritional source for consumption instead of ending as waste, and therefore accumulating all environmental impacts of the supply chain without any use. In other words, the worst-case scenario. Nevertheless, even when the portion of the environmental impact of one packaging unit of high impact food products was small, it should be taken in consideration that the impact can be huge when considering the accumulated impact of all used packaging unit. B) Consequential LCA of packaging materials can be a useful tool to select the most appropriate material, e.g., comparing a mono-material with a multilayer material. In this category different approaches were taken, either only the direct effects of the materials were compared, or studies were designed to compare materials with potential similar indirect effects as providing the same shelf life. The later come with more hurdles but provide a more realistic outcome. These approaches were focused on the packaging material and can be extend by additionally considering the food-packaging system, the third approach (C). Having the advantages, that a more holistic decision can be taken. LCA comparing different packaging systems often included indirect packaging impacts, beside direct impacts. Although, quantifying the relation between food packaging and the food supply chain can be challenging. Additionally, studies often focus on specific indirect impacts, whereby fewer studies with multiple indirect impacts were published.

By reviewing LCA studies of novel packaging systems it could be seen that often when only considering the direct environmental impact the novel packaging systems have a higher environmental impact, and therefore including indirect impacts is important. When including indirect impacts, mostly waste reduction due to shelf life extension, the environmental advantages are on the side of the novel packaging system. Overall, it was highlighted that beside the reduced environmental effect, also consumer preferences concerning the novel food packaging system should be included when selecting the packaging.

In conclusion, the different LCA approaches taken were diverse, and each one covers a specific goal. By seeing the selected categories as sequential steps, a holistic understanding of the environmental impact of novel food packaging can be achieved. A clear understanding of the impacts and benefits of innovative packaging systems is an important driver for successful and sustainable innovations.

Chapter THREE

# Edible coatings for ready to eat seafood products: critical review of recent studies, sustainable packaging perspectives, challenges, and emerging trends

## Abstract

*The potential of edible coatings as an innovative approach for eco-friendly packaging systems needs further investigation to realise the potential of edible coatings for the ready to eat (RTE) food sector. This chapter reviews the state-of-art developments of edible coatings for chilled RtE food products with a focus on sustainability, functionality, and industrial feasibility. It discusses associated challenges with the use of edible coatings as eco-friendly packaging system in RtE food sector, including compatibility with food products, processing conditions, shelf life conditions, storage conditions, and regulatory requirements. Additionally, emerging trends are discussed, including biodegradable and eco-friendly coatings, shelf life extension, active and intelligent coatings, customization and personalization opportunities. Overall, edible coatings can offer benefits in the RtE food sector, but there are several challenges that must be addressed to ensure their successful implementation. Research and development efforts are needed to optimize the performance and stability of coatings, while ensuring compliance with regulatory requirements and addressing cost concerns. The potential of edible coatings as eco-friendly packaging system should be further studied to highlight the full potential of edible coatings.*


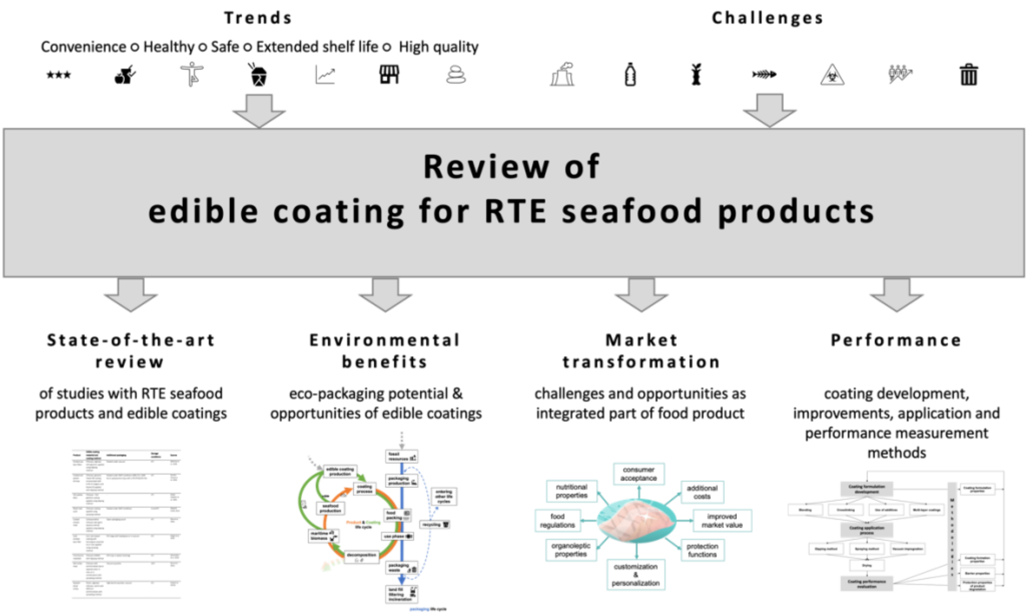


*Figure 3-1: Graphical Abstract - Edible coatings for ready to eat seafood products: critical review of recent studies, sustainable packaging perspectives, challenges and emerging trends.*


## Keywords

*edible coating*
*ready to eat seafood*
*eco-friendly innovation*
*novel food packaging system*
*shelf-life extension*

## 3.1 Introduction

More sustainable and novel food-packaging systems are in high demand like never before. In order to meet the world's growing need for sustainable and high-quality food supply chains, two major challenges need to be addressed: food loss and waste reduction, as well as packaging and packaging waste reduction, which are target issues of the sustainable development goals defined by the United Nations. Additionally, consumer demands for mild preserved, fresh, nutritious, safe, convenient food products with enhanced shelf life and longer retailing distances have driven the need for novel and innovative food-packaging systems (Han et al., 2018).

Edible coatings have already been explored as packaging alternatives for food product groups as vegetables, fruits and fresh meat products (Janes and Dai, 2012). However, can their potential be harnessed for RtE food products, which is a fast growing and emerging market, interesting for diverse consumer groups? Seafood products are particularly susceptible to spoilage and deterioration, and the use of edible coatings can help to extend the shelf life of RtE seafood products and ensure their overall quality. Several studies have investigated the use of edible coatings based on different materials, with a focus on improving safety, shelf life, and sensory properties of products. Challenges associated with the use of edible coatings in the RtE seafood sector include the development of coatings that are compatible with different types of seafood products, as well as ensuring the safety and stability of the coatings over time. Additionally, regulatory requirements related to food safety and packaging materials must be addressed.

This critical review focuses on RtE seafood products and reports on current advances, challenges, and knowledge gaps that need to be addressed to enhance further developments in this area. Additionally, the potential of edible coatings for developing environmentally friendly responsible packaging systems for the RtE food sector was explored.

## 3.2 Edible coatings

Edible coatings are thin protection layers that are directly applied to the product surface and can be consumed along with it (Bourlieu et al., 2008). They are made from natural film formers that adhere to the product surface, creating a protective layer that can help to prolong the shelf life by reducing or inhibiting natural chemical, biochemical or microbiological degradation processes caused by gas contact, gas diffusion or water transfer (Cerqueira et al., 2010). Essential properties of an edible coating include adhesiveness to the product, an invisible and uniform surface appearance, neutral or positive sensory attributes, a defect-free cover, and a composition that conforms to food regulations (Nešić et al., 2019).

## 3.3 Recent studies with edible coatings for RtE products

Edible coatings have great potential for use in RtE food products. Yousuf et al. (2021) have noted this potential, but limited research has been conducted on the use of such coatings in RtE muscle food products. The current state-of-the-art of studies investigating the use of edible coatings in RtE seafood were reviewed. The investigation was broadened by including studies with edible coatings for muscle RtE products. The properties of single material with and without additives as well as blends or multi-layer applications are explored. Some of the key findings are summarised in Table 3-1.

Smoked sea bass fillets coated with chitosan or alginate packed in a vacuum bag and stored at 4°C showed that a chitosan coating reduced the viable counts of mesophilic, psychrophilic, and anaerobic bacteria and additionally, retarded oxidation (Martínez et al., 2018). The alginate coated products improved the protection against oxidation but did not show an inhibition in bacteria growth. Cooked and peeled shrimps with a chitosan-thyme coating and packed under MAP conditions in a tray with a film stored at 4°C for up to 12 days, showed a positive effect in inhibiting the growth of lactic acid and psychotropic bacteria (Carrión-Granda et al., 2016). Retail maki sushi with salmon coated with chitosan, especially in combination with a low $CO_2$ – MAP packaging, was reported to increase the microbiological stability and preserve the colorimetric properties of the sample stored at abused temperature conditions (8°C) and was recommend as a suitable hurdle technology for the safety measure against temperature abuse in the food cold chain (Sørbø and Lerfall, 2022). Spiced cooked RtE chicken meat with an edible crude nano-coating solution based on carboxymethyl chitosan, a water-soluble derivative of chitosan, and garlic aqueous extract inhibited the growth of microorganisms and delayed the rate of protein break-down and lipid oxidation (Diao et al., 2020). Cold-smoked sunshine bass fillets coated with corn zein-based coating with lemongrass essential oil or nisin packed in bags with headspace air or vacuumed was studied with the focus on the inhibition of Listeria monocytogenes, showing that both coatings reduced the cell count in both studied packaging systems (Hager et al., 2019). The nisin treated samples were found to be the most effective additives against *L. monocytogenes*, over the tested storage time of 42 days. Additionally, it was concluded that only the samples treated with corn zein-lemongrass essential oil in bag with headspace air were found to inhibit the growth of spoilage organisms. Other studies focused on controlling *L. monocytogenes* in RtE meat products confirmed a positive effect of edible coatings with or without antimicrobials to reduce or even inhibition the growth (Antoniadou et al., 2019; Guo et al., 2014; Jiang et al., 2011b). Antoniadou et al. (2019) studied a chitosan coating on RtE meatballs, beside the inhibitor effect of *L. monocytogenes*, also other microbial populations were counted, leading to a shelf life extension of around 14 days. The conducted sensory test showed that the chitosan coating did not have a negative effect on the sensory characteristics of RtE meat balls. A packaging system consisting of an edible chitosan coating without additives and synthetic polymer bag was tested for RtE roast beef under cooled storage (Beverly et al., 2008). The focus was to study the effect of the chitosan molecular weight and the solvent solution for the coating preparation on the reduction effect against *L. monocytogenes*. Chitosan coating prepared with an acetic acid solution were more effective in reducing *L. monocytogenes* compared to lactic acid solvent. The study also confirmed that chitosan coatings can be used to control *L. monocytogenes* on the surface of RtE roast beef products. The anti-listeria activity of chitosan based edible coatings in combination with a vacuum packaging was also tested on cold-smoked salmon (Jiang et al., 2011a). It was confirmed that a chitosan coating without antimicrobials was able to inhibit the population of *L. monocytogenes* on cold-smoked salmon during vacuum storage under refrigerated conditions, but tested chitosan coatings with antimicrobials had the greatest anti-listeria activity. Juck et al. (2010) studied the effect of edible coatings for two RtE turkey products to control microbial growth with a focus on *L. monocytogenes*. Different polysaccharide coatings were tested as carrier materials as well as different antimicrobials in single and binary combinations. Alginate, kappa – carrageenan, pectin, xanthan gum and starch displayed a good adherence and stability on the turkey products. The applied coatings had no negative effect on the texture, colour, smell, and overall appearance of the product. The different antimicrobial treatments had comparable efficiencies across the tested carrier materials, although pectin was slightly less effective than the others. Overall, it was shown that alginate-based antimicrobial coatings can improve the safety of poached and roasted turkey by inhibiting the growth of *L. monocytogenes* and reducing background microbiota levels throughout an 8-week storage period at 4°C. Though, to a different extend depending on the selected antimicrobials. Another study focused on controlling the growth of *L. monocytogenes* on roasted turkey with a packaging system with edible antimicrobial coatings vacuumed packed into high barrier pouches (Jiang et al., 2011b). Starch, chitosan, alginate, and pectin coatings with different antimicrobials were studied on sliced roasted turkey during cold storage and frozen storage. The results showed that pectin-based antimicrobial edible coatings hold promise in enhancing the safety of RtE poultry products and frozen storage has the potential to enhance their effectiveness. The anti-listeria effectives of the tested polysaccharide-based carrier coatings with antimicrobials ranked in the order of pectin, followed by alginate, and followed by chitosan and starch.

The investigated studies explored edible coatings for RtE fish products as smoked fish bass, smoked salmon, cooked shrimps, and retail maki sushi. Studied RtE meat products are cooked chicken breast, fried meat balls, cooked turkey meat and roast beef. Mainly, chitosan based edible coatings with or without antimicrobial additives were examined. Additionally, corn-zein, pectin, alginate, starch, gelatine, kappa-carrageenan, and xanthan gum were examined as carrier coatings for antimicrobials. The focus of the investigated edible coating studies was the control of microbial growth with a special emphasize on *L. monocytogenes* a comment food born pathogen in RtE foods. It was shown that current packaging systems as high barrier synthetic films, pouches or trays were improved by the addition of a coating protection layer on the surface of the food product by extending the safety and quality of RtE products. Edible coating methods was purposed as potential fresh-keeping method for RtE products (Diao et al., 2020), as well as could be a suitable hurdle technology for the safety measure against temperature abuse in the food cold chain (Sørbø and Lerfall, 2022). Only few studies focused on other protection properties as fat oxidation or water loss, beside the microbial degradation (López-Caballero et al., 2005; Martínez et al., 2018). The fact that an edible coating can supply multiple packaging functions by also protecting physical, biological, or chemical quality changes of food products is under investigated in the case for RtE food products. Controlling multiply degradation processes can increase the potential use of a coating as in integrated part of a packaging system. It was noticed that the tested edible coatings were developed as an added protection layer to the existing primary package but not with the focus on overtaking functions of the current packaging system. The synergy between an edible coating and an outside layer can be of interest for an eco-packaging design option and needs further investigation. Additionally, it was noticed that only composite coatings with a polysaccharide carrier material and antimicrobials were studied. Other potential methods, e.g., multilayer coatings or blends, for improving the coating performance were not studied with RtE seafood products.

Table 3-1: Overview of edible coating studies with RtE seafood and meat products.

| **Product** | **Edible coating material and coating method** | **Additional packaging** | **Storage conditions** | **Source** |
|---|---|---|---|---|
| Smoked sea bass fillets | Chitosan, alginate with glycerol, applied using dipping method | Packed under vacuum | 4°C | (Martínez et al., 2018) |
| Cooked and peeled shrimps | Chitosan, glycerol, Tween 80 coating incorporated with 0.5% of oregano and thyme EO applied with dipping method | Packed under MAP conditions (40% $CO_2$ / 60% $N_2$) in polystyrene trays with a PET/PVdC/PE film | 4°C | (Carrión-Granda et al., 2016) |
| Fish patties (RtC) | Chitosan – fish gelatine coating applied using dipping method | - | 2°C | (López-Caballero et al., 2005) |
| Retail maki sushi | Chitosan coating applied using spraying method | Packed under MAP conditions | 4 and 8°C | (Sørbø and Lerfall, 2022) |
| Cooked chicken meat | Carboxymethyl chitosan with garlic aqueous extract applied using dipping method | Open packaging pouch | 4°C | (Diao et al., 2020) |
| Cold-smoked bass fillet | Corn zein-based coating with lemongrass essential oil or nisin applied using spraying method | PVC bags with headspace or in vacuum | 4°C | (Hager et al., 2019) |
| Fried bovine meatballs | Chitosan (HMW) with dipping method | EPS trays in plastic food bag | 5°C | (Antoniadou et al., 2019) |
| Deli turkey meat | Chitosan with antimicrobials (lauric arginate ester or nisin or in combination) with spreading method | Vacuum pouches | 10°C | (Guo et al., 2014) |
| Roasted sliced turkey | Pectin, alginate, chitosan, starch with different antimicrobials with spreading method | High barrier pouches, vacuum | 4°C | (Jiang et al., 2011b) |
| Cold smoked salmon | Chitosan coating with and without antimicrobials with spreading method | Vacuum packed | 4°C | (Jiang et al., 2011a) |
| Deli turkey and in pouched cooked sliced turkey | Polysaccharide carry materials (alginate, kappa – carrageenan, pectin, xanthan gum, starch) with different antimicrobials) with spreading method | Vacuum packed | 22°C, 4°C | (Juck et al., 2010) |
| Roast beef | Chitosan coating (different molecular weights and solvents) with dipping method | Bagged | 4°C | (Beverly et al., 2008) |
| Precooked beef patties | Wheat gluten, soy protein, carrageenan, and chitosan coatings with dipping method | - | 4°C | (Wu et al., 2000) |
| RtE squid rings | Oregano and thyme essential oils with dipping method | Packed under MAP conditions (70% $N_2$, 25% $CO_2$, 5% $O_2$) in film bag | 4°C | (Sanjuás-Rey et al., 2012) |
| Dry sausages | Alginate, polyglycerol esters of fatty acids, pea protein and collagen with extrusion method | - | 13°C | (Marcos et al., 2020) |

## 3.4 Challenges & emerging trends

To explore the full potential of edible coatings for RtE seafood products, the concept was reviewed under a holistic approach. Potential challenging aspects, emerging trends and opportunities were discussed (Figure 3-4). Sustainability aspects of edible coatings and coating integration requirements for being part of the food product were investigated. Additionally, strategies to improve coating performances, application methods and methodologies for testing the coating performance were reviewed.

### 3.4.1 Can an edible coating provide an eco-packaging solution?

Food packaging is complex, multidimensional and plays a crucial role in ensuring the quality, safety, and security of food products. Food packaging offers several functions and services to products as protection, containment, communication, and convenience (Singh et al., 2017). One important aspect of food packaging is its protective function, which helps to prevent degradation processes, such as mechanical, physical, chemical, or biological damages. Depending on the food and its characteristics, degradation processes vary, and the emphasis of packaging requirements is placed on different prioritized protective functions (FAO, 2019; Farris and Piergiovanni, 2012; Morris et al., 2017). However, in addition to providing protection, food packaging also has an important role to play in minimizing environmental impact. An environmental responsible food-packaging system involves a synergy between reducing food losses and waste from production to consumption, as well as using packaging materials that require minimal resources and generate minimal waste (Emblem, 2012). RtE seafood products are highly perishable and require specialised packaging to ensure their safety and quality. However, traditional used multilayer packaging systems, which combine different materials to achieve specific packaging properties, offers strengths and weaknesses (Hopewell et al., 2009; Kaiser et al., 2018; Morris, 2017). Multilayer food packaging is a lightweight solution, but can be difficult to recycle, and hinders achieving circularity (Dainelli, 2008; Hopewell et al., 2009; Kaiser et al., 2018). The dependency of fossil raw materials is an additional environmental burden. A good balance between eco-friendliness and the preservation of shelf life is crucial for food-packaging systems. While incorporating and edible coating into a packaging system can incur additional expenses, such as those related to coating development, application, and product-coating interactions, it is worthwhile to examine the potential benefits of using an edible coating as a primary packaging system. An edible coating is sourced from biobased materials, whereby the environmental impact of an edible coating depends on the selected coating material as further discussed in section 4.2. An edible coating is designed to be consumed with the product and therefore no additional packaging waste is generated. Often only a small amount of coating material is needed to cover the food product surface and supply a protection function.

Numerous studies have investigated the use of edible coatings in combination with a traditional synthetic primary packaging, with a focus on increasing product shelf life and reducing food waste. By reducing degradation processes as microbiological growth, but also increasing the product robustness against temperature changes during the supply chain (Sørbø and Lerfall, 2022). Studies focusing on the coating performance without an external layer have shown that the coating alone can protect and extend product shelf life (López-Caballero et al., 2005; Wu et al., 2000). This could indicate that an edible coating in combination with an external packaging provides protection even when the integrity of the external packaging is broken at the consumer stage. This approach can support the use of larger purchasing units, reducing packaging waste and providing convenience to the consumer by reducing shopping frequency, and gives more flexibility in the consumption of the product.

An under investigated concept of edible coatings is the use of edible coatings as synergetic part of the two-component primary packaging. Hereby, the focus is on developing environmentally friendly packaging systems by reducing the environmental impact of the external layer, instead of increasing the shelf life with a coating. The concept is based on the ability of a coating to overtake packaging functions of the packaging by controlling single or multiply degradation processes e.g., water loss, fat oxidation or microbiological growth. With the aim to reduce the requirements of the second packaging component, and potentially opens the possibility to select more environmentally friendly materials. Viable options could be mono-material application, more easily recycled materials, or biobased materials. Further investigation is needed into the concept of edible coatings as an integrated part of primary packaging systems, including applied packaging and theoretical modelling studies.

Overall, a synergetic packaging consisting of an edible coating and second component could lead to a more sustainable packaging system, by reducing food waste, using bio based (edible) packaging materials, providing more flexibility in the external packaging selection, providing convenience to the consumer, and increasing shelf life leading to more flexibility for producers, wholesalers and retailers in the supply chain (Figure 3-2). An edible coating cannot stand alone as primary packaging as it does not protect for contamination during storage, transportation, or does not provide containment or communication, but it should be seen as an added or integrated part of the packaging system. Edible coatings in combination with a second packaging part is described as a fast and cost-effective technology with a substantial industrial potential easy to implement (Sørbø and Lerfall, 2022). An edible coating could meet the consumer demand for high quality food products in combination with the environmental need to reduce packaging waste (Puttalingamma, 2014). Nevertheless, more studies are needed considering an edible coating as integrated part of the packaging systems to reduce the environmental impact of the packaging system with a similar shelf life, instead of an added part to extend the product shelf life. The full potential of edible coatings as drivers for sustainability is still not explored. Life cycle assessments (LCA) studies should be used to provide evidence of the developments and functions of an edible coating as an eco-packaging system.

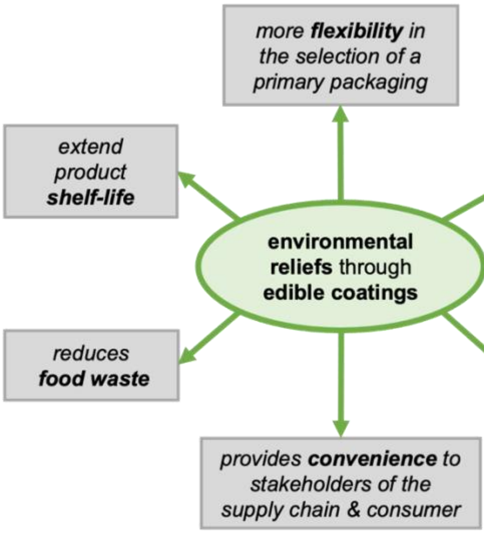


*Figure 3-2: Environmental advantages due to eco-food packaging systems with edible coatings.*

### 3.4.2 Developing strategies for edible coatings

RtE muscle foods are highly perishable products, intended for direct consumption. These products are often transported and stored in chilled supply chain and have a limited shelf life due to microbiological and biological reactions as well as chemical degradation. As a result, ensuring food safety is a critical factor. In addition to this, it is important to control common deterioration reactions in RtE muscle foods such as water loss and fat oxidation to prolong product quality and safety. When selecting coating materials, it is important to consider their ability to provide characteristic protection functions, as well as nutritional and sensory aspects as discussed in section 3.4.3. Moreover, it is essential to investigate the potential for environmental conservation and conscious handling of available resources in the selection of coating materials. A circular economy principle supports a design that includes options to make use of resources, materials, and products after the use phase (European Commission, 2014). Avoiding, a linear consumption pattern is essential, often used for many global resources where the focus lies on the consumption of resources without planning further use at the EoL. Prioritising circular aspects will lead to supporting sustainable aspects during the design phase (Ghisellini et al., 2016). A wide range of natural edible coating materials are available from land, or water resources. In the case of RtE seafood products, exploring coating materials from the maritime environment was favoured to support an integrated circular life cycle, in contrast through an often-linear life cycle of a synthetic packaging material (Figure 3-3). Therefore, it is of value to explore potential marine based coating materials by considering available biomass, materials from waste streams, their resource availability, or competition with food supply.

An overview of generated products from maritime biomass, including finfish-based, shellfish-based, and plant-based biomass can be found Olsen et al. (2014). Interesting products for edible coating materials include, among others, polysaccharides sources from seaweeds such as alginate, carrageenan and agar, sources from crustacean shell waste such as chitin and chitosan, or sources from algae as cellulose and starch (Table 3-2). Interesting protein sources are collagen and gelatine, and lipid sources are fish oil from fin-fish by-products. Additionally, minerals and other functional ingredients can be of interest to improve the coating properties and improve the nutritional value. By exploring marine waste products for potential coating applications new markets are explored and environmental pollutions can be avoided. Waste products that are not valorised can end up in landfills, or soil dumping, or be discarded in seawater, resulting in major surface pollution, unpleasant smell in coastal areas, and contributing to major environmental concerns (Kandra et al., 2012). Various marine by products have been researched and developed for many years for different applications. While some are successfully implemented and established on the market, such as chitosan production from crustacean shells, others have been limited due to several drawbacks, particularly for fin-fish by-products. These limitations may be due to overestimation of market possibilities, too small amounts of high-quality by-products available on a regular basis, excessive costs of isolating specific components often present in lesser amounts in the by-products, or cheaper and more stable production methods using genetically modified microbes or chemical synthesis. Furthermore, regulations and documentation costs for upcycling by-products for human consumption may also pose challenges (Olsen et al., 2014). Marine plants are a fast-growing sustainable recourse for biomass, which can be produced without significant environmental impacts (Mouritsen, 2013). They are considered as next generation bio-based materials (Fabra et al., 2017; Zhang et al., 2019b), and have advantages such as minimizing or avoiding competition with arable land and nutrients used for conventional agriculture, reducing competition for limited freshwater supply, utilizing wastewater, reclaimed water and saline water and recycling carbon dioxide during growth by carbon dioxide fixation and oxygen production (Merz and Main, 2017). Additionally, they have an extensive environmental tolerance (Mathiot et al., 2019; Zia et al., 2017).

Table 3-2: Overview of available maritime materials for edible coatings.

| **Product category** | **Product name** | **Maritime source** | **Market implementation** |
|---|---|---|---|
| Polysaccharide | Alginate | Seaweed (brown algae) | Established |
| Polysaccharide | Agar | Seaweed (red algae) | Established |
| Polysaccharide | Carrageenan | Seaweed (red algae) | Established |
| Polysaccharide | Chitin | Crustacean exoskeleton (waste product), algae | Established |
| Polysaccharide | Chitosan | Crustacean exoskeleton (waste product), algae | Established |
| Polysaccharide | Cellulose | Microalgae | Development stage |
| Polysaccharide | Starch | Microalgae | Development stage |
| Protein | Collagen | Fish waste (skin/bone) | Established |
| Protein | Gelatine | Fish waste (skin/bone) | Established |
| Protein | Glycoprotein | Fish (blood) | Established |
| Lipid | Fish oil | Fish waste | Established |
| Minerals | Calcium chloride | Crustacean exoskeleton (waste product) | Industrial side product |

The available biomass from aquatic resources includes high-value polysaccharides, proteins, lipids, and minerals that can be used for the development of edible coating materials. Some materials, such as chitosan, alginate, agar, carrageenan, fish oil, and fish gelatine, are already implemented into the current market, while others, such cellulose or starch from algae, are still at research level and need to be scaled up to reach a competitive market value.

### 3.4.3 Edible coatings: An integrated part of food products

An edible coating is an integrated part of the food product, intended for direct human consumption and ideally unnoticeable to the consumer. Thus, in addition to protection functions, organoleptic, nutritional aspects and food regulations play a significant role for edible coatings (Nešić et al., 2019). Specialized edible coatings can be designed to have specific protection functionalities, such as antimicrobial properties, or oxygen scavenging, so called active coatings. Additionally, coatings find use within intelligent packaging systems to monitor freshness and enable real-time monitoring of quality and safety of food product during whole supply chain. Active and intelligent coatings are of high research interest.

The mouthfeel, texture, taste, and aroma throughout the whole shelf life of the product should be taken into account when developing an edible coating to increase consumer acceptance and market transformation. Additional values of edible coatings are customization and personalization potential. Consumers are seeking more personalized and customized products, and edible coatings can provide a platform for product differentiation. For example, coatings can be customized with different flavours or colours to enhance the sensory experience of the consumer. Another emerging trend is the use of edible coatings as carriers for nutraceuticals such as vitamins or minerals. Allowing product tailoring for meeting specific demands of consumer groups as elderly people. Difficulties may arise due to interactions between coatings and product components, which could impact the organoleptic or nutritional aspects. These aspects are conditional for the use of an edible coating for industrial products.

The use of edible coatings in the RtE food sector is subject to regulatory requirements related to food safety, labelling, and packaging materials. Compliance with these requirements can be challenging and time-consuming (Yousuf et al., 2021). Additionally, further research is required to explore consumer view about edible packaging. By better understanding the acceptance and hurdles, relevant communication can be developed about for products with edible coatings. A drawback of the development and implementation of edible coatings into the RtE food sector are additional costs due to coating development, implementation, material costs or equipment investments. Nevertheless, additional direct costs can be justified by providing additional product and market value as product quality and safety or, product differentiation to competitive products. In the realm of studies with edible coating for RtE seafood products, standard methodologies considering both protection properties and sensory aspects should be further implemented to support the interdisciplinary fields of a coating development, and its implementation into industrial application.

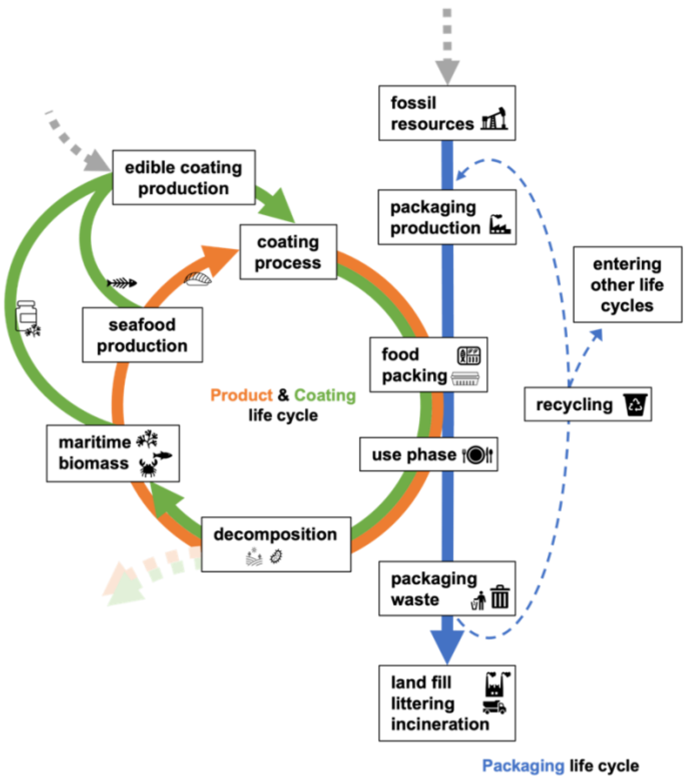


*Figure 3-3: Overview of the synergy between food product, edible coating and packaging life cycle.*

### 3.4.4 Strategies to improve coating performance: Active and intelligent coatings

The more effective an edible coating is at protecting the product, the less reliance there is on the external packaging. To optimise the protection function for a specific food product, it is important to consider different options for tailoring the edible coating. Using a single material may only provide limited protection, therefore, composite or multilayer coating can combine the unique properties of different materials and form a protective layer for different critical quality parameters simultaneously (Hosseini and Gómez-Guillén, 2018; Yousuf et al., 2021).

Coating properties can be tailored by blending of different materials (composite coating), either from the same material group as polysaccharides, proteins or lipids or different groups. However, the latter case is more comment, since the individual protection properties are more diverse (Yousuf et al., 2021). For instance, hydrocolloids know to supplying a good gas barrier, can be blended with a lipid component to improve water barrier properties and enhance mechanical and structural coating properties (Bourlieu et al., 2008; Wu et al., 2001). Nevertheless, blending materials has its limitations, such as difficulties in forming a homogeneous coating solution, turbidity due to small light breaking particle distributed in the solution, and the formation of aggregates or phase separation.

Crosslinking is an irreversible change of the polymer structure through a chemical reaction and is another interesting way to tailor coating formulations. It is often applied through a double coating process. For instance, sodium alginate can be cross-linked with calcium carbonate to form a water insoluble gel coating. But also, other materials are cross-linked to tailor their coating properties as chitosan coatings (Azaza et al., 2022).

Another strategy is the addition of synthetic or natural additives such as antioxidants, antimicrobials, gas scavengers, nutraceuticals, plasticizers, or nanoparticles. Active coatings are defined as coating that interact with the food to deliberately incorporate components that would release or absorb substances into or from the packaged food or the environmental surrounding the food to extend the shelf life of packaged food (European Commission, 2009). The research focus is mainly on using natural compounds to eliminate potential health concerns and increasing consumer acceptance. Antimicrobial agents such as, nisin, lysozyme, essential oils; antioxidant agents such as gelatine hydrolysates (Mirzapour-Kouhdasht and Moosavi-Nasab, 2020) and nanoparticles such as nanocellulose or clay (Souza et al., 2020) were studied. However, the use of additives has limitations, such as application costs, their intensive aroma or potential toxicity (Souza et al., 2020), and the potential migration of nanoparticles to the human body (Mahmud et al., 2021).

A different approach to improve the coating performance is the use of multi-layer coatings. Composite coatings involve blending materials to form a stable mixture, while bilayer coatings can be prepared through layer-by-layer or unstable mixture technique (Pérez-Gago and Rhim, 2014). Multi-layer coating systems form a layer-by-layer structure on the product surface, and have been studied for application of lipid-based coatings (Pérez-Gago and Rhim, 2014). Multi-layer coatings of chitosan and alginate coatings have been used on fresh cut watermelon to combine coatings with opposite charged polymers (Poverenov et al., 2014). Whereby, lipid-based bilayer coatings based on different lipid components or lipid and hydrocolloid layers have been reported to provide a much higher moisture barrier compared to composite coatings of the same materials (Bourlieu et al., 2009). While bilayer coatings can add up the properties of single material coatings (Morillon et al., 2002), they also have drawbacks, such as forming a thicker coating layer that may lower consumer acceptance and require an additional processing step, which can be a hurdle for the implementation in industrial applications.

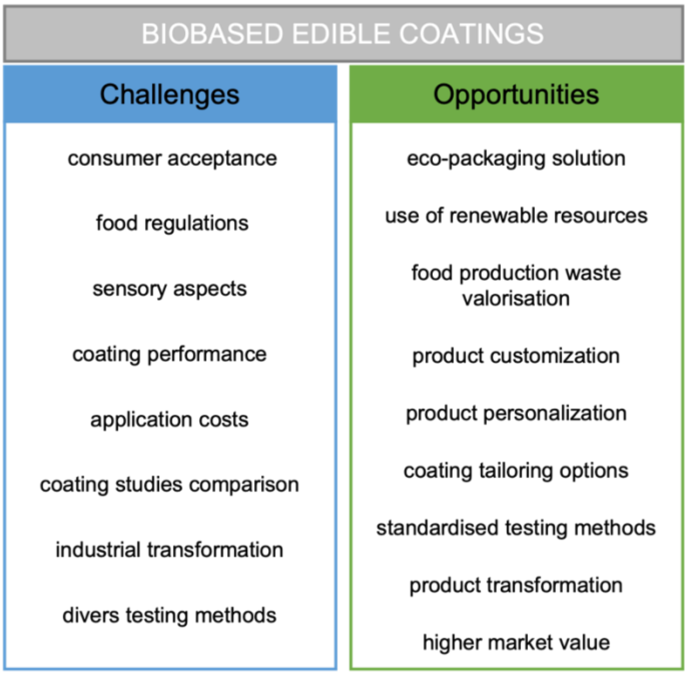


*Figure 3-4: Overview of challenges and opportunities of biobased edible coatings.*

### 3.4.5 Coating application methods

Biobased coatings can be applied to the product surface by dipping, spraying or vacuum impregnation. For a more detailed information, please refer to Senturk Parreidt et al. (2018). In all methods, first coating formulation must be prepared by dissolving the biopolymer in a solvent, either followed by addition of components, homogenization, and a degassing step (filtration, ultra-bath, centrifugation or resting). Depending on the coating materials, other steps as heating may be necessary to ensure structural breakdown or improve dissolving properties.

At the laboratory scale, dipping is the most commonly used coating method (Senturk Parreidt et al., 2018). The product is coated by being dipped into a coating formulation for a defined time, then drained to remove excess solution, and finally dried by evaporating the solvent. The coating process is typically repeated with the same or a different coating solution to ensure a continuous film formation on the surface of the product. Dipping creates an even coating of products with complex and rough surfaces. The coating thickness can be controlled to some extend with the viscosity of the coating formulation (Cisneros-Zevallos and Krochta, 2003). However, compared to the spraying methods, coatings applied with dipping are generally thicker (Senturk Parreidt et al., 2018). Difficulties may arise when coating hydrophilic product surfaces with a hydrophobic coating solution. A layer-by-layer coating method can solve this issue by promoting physical and chemical bonding between oppositely charged materials (Senturk Parreidt et al., 2018). One drawback of the dipping method is the potential cross-contamination when multiple products are dipped into the same coating solution, which can result in product components dissolving into the coating solution.

Spraying is a costing method where the coating solution is distributed as droplets over the targeted food surface area through a nozzle. This method has several advantages, such as the need for less coating solution due to applied high spraying pressure, creating a uniform coating with a controlled thickness, enabling temperature control of the solution, and being suitable for multilayer applications. Additionally, this method avoids contamination of the coating solution, making it possible to work with larger product surface area. The spraying system's coating efficiency is affected by various factors, such as the properties of the coating solution (density, viscosity, and surface tension), operational conditions (flow rate, air pressure etc.), and system conditions (nozzle design, spray angle etc.). It is important to consider the spray-flowing characteristics of the coating solution, where a low viscosity of the coating solution is preferred. However, many biobased polymers have a high viscosity at low polymer concentration, which can be an issue. The combination of dipping and spraying methods is possible, and it provides advantages as a thinner layer, better coverage and controlled thickness (Amanatidou et al., 2000).

Vacuum impregnation is a third coating method that involves the same dipping draining steps as the dipping method. However, in vacuum impregnation, the samples are coated in a vacuum chamber to ensure rapid and direct migration of the coating material into the porous tissues of the samples. The method works by removing gas and liquids from the pores of the product and replacing them with coating solutions under restored atmospheric pressure (Zhao et al., 2019).

After the coating application, the samples were often allowed to dry before being further packed or stored to ensure complete coating coverage around the product. Drying at room temperature, during cold storage or at higher temperatures under laminar or turbulent air circulation were reported.

Only few research papers of edible coatings for RtE seafood or related products using spraying or vacuum impregnation method were found, most studies applied the dipping method. Further investigation of the spraying methods is recommended to bridge the gap between research and industrial applications.

### 3.4.6 Coating performance measurement methods

Important coating properties include adhesiveness, water barrier, gas barrier, antimicrobial properties, antioxidation properties, appearance, taste, and texture. The purpose of this section is to provide an overview of methodologies used to validate and compare coating performances (Table 3-3).

Viscoelastic properties of the coating formulations, such as pH value, elastic modulus, viscosity modulus, solubility, phase angle as a function of temperature, and electrical charge are important in ensuring easy handling, application, and increasing industrial feasibility (Bourlieu et al., 2008; López-Caballero et al., 2005).

When the coating is applied to the product, the adhesiveness, coating thickness and coating integrity properties are of interest. The adhesiveness of a coating material on the product depends on the nature of the material, and on the number of interactions or bonding between coating and support (Comaposada et al., 2018). Some researchers investigated coating adhesiveness by measuring the contact angle of the product of interest before and after coating (Cerqueira et al., 2010). In addition, coating integrity on the surface and penetration of the coating can be analysed through imaging analysis. The coating thickness can be measured with the aid of a scanning electron microscope (SEM).

Coating barrier properties against water and gas can provide relevant information about the coating performance. To evaluate barrier efficacy of coating materials, either the barrier properties of casted-dry films (permeability) or the barrier measurements of the coating directly applied onto the product (resistance) are determined. However, measuring the barrier properties of casted coating films has the disadvantage that the results are often difficult to transfer to the barrier properties of the coating material applied to the product, as they can be highly influenced by the food contact (Guo et al., 2014; Jiang et al., 2011a; Nowzari et al., 2013; Wu et al., 2000). To evaluate the relative water loss of coated products, the rate of water loss (g/h) is measured, or the water vapour resistance of the coating (s/cm) is determined to compare the performance of different coatings, considering parameters as water activity, relative humidity, water loss rate, surface area, saturation pressure and storage temperature (Avena-Bustillos et al., 1997; Ben-Yehoshua et al., 1985). Although, studying the effect of barrier properties of edible coatings with the product is still limited, this method has been applied to diverse coatings and food products (Bravin et al., 2006; Chiumarelli and Hubinger, 2012; Morillon et al., 2002; Vargas et al., 2008). This method allows for comparing different coating materials and assessing their efficiency on various products. Another direct method to study the gas barrier of a coating is by measuring the gas exchange rates (e.g., $O_2$ or $CO_2$) of the coated product with the headspace volume in a close system (Cerqueira et al., 2010).

The effect of the coating on the degradation process of food samples can be assessed through microbial, chemical, and physical analyses. Antimicrobial properties can be evaluated by measuring the growth of bacterial, yeast and mould, with the specific strains selected based on the food product and storage conditions of interest. Physical-chemical deterioration can be monitored by measuring factors such as water loss, water activity, nitrogen degradation, and fat oxidation (peroxide value, malondialdehyde value, total volatile basic nitrogen, or trimethylamine-nitrogen), as well as pH changes and adenosine triphosphate breakdown during storage (Socaciu et al., 2018). Changes in the appearance of the product can be measured using a colorimeter, while the texture can be assessed with a texture analyser that measures the hardness, elasticity, cohesiveness, adhesiveness, chewiness, and gumminess of the coated and non-coated products (López-Caballero et al., 2005). Sensory evaluations can also be conducted to study the appearance, odour, texture, firmness, colour, taste, juiciness, and flavour of coated products.

Various methodologies have been used in the literature to measure the coating performance. However, standard methods for measuring the coating integrity, penetration and barrier properties are still lacking. Additionally, the comparison of coatings applied to different food products is limited as the coating properties are highly affected by the product or interactive properties. Direct methods for studying the coating properties are limited, and indirect measurements are still prevalent in coating studies.

Table 3-3: Overview of applied coating performance measurement methods.

| **Studied material** | **Property category** | **Property details** | **Methodology** |
|---|---|---|---|
| Coating formulation | Viscoelastic properties | Handling<br>Application<br>Industrial feasibility | Ph<br>Elastic modulus<br>Viscosity modulus<br>Solubility<br>Phase angle<br>Electrical charge |
| Casted films | Barrier properties | Molecule transfer | Water permeability<br>Gas permeability |
| Coated product | Barrier properties | Molecule transfer | Water resistance<br>Gas exchange range |
| Coated product | Coating formation properties | Adhesiveness<br>Thickness<br>Penetration/absorption<br>Integrity<br>Durability | Contact angle<br>Imaging analysis<br>Scanning electron microscope<br>Wettability<br>Cohesiveness |
| Coated product | Sensory properties | Appearance<br>Texture<br>Odour<br>Taste | Colour/opacity<br>Hardness<br>Elasticity<br>Chewiness/gumminess |
| Coated product | Physical-chemical properties | Fat oxidation<br>Water migration | Water loss,<br>Water activity<br>Nitrogen degradation<br>Peroxide value<br>Malondialdehyde value<br>Total volatile basic nitrogen<br>Trimethylamine-nitrogen<br>Ph<br>Adenosine triphosphate breakdown |
| Coated product | Anti-microbial properties | Safety & quality degradation | Bacterial growth<br>Yeast growth<br>Mould growth |

## 3.5 Conclusion & future trends

The consumer demand for healthy, safe and convenient food products, as well as sustainable food-packaging solutions, is driving the development and market implementation of RtE products packed with eco-innovative packaging solutions. This review investigated the state-of-the-art of using edible coatings for RtE seafood products as an alternative packaging concept.

Reviewed studies show promising results for the use of edible coatings in combination with a synthetic primary packaging for improving the product safety and shelf life. Edible coatings have been highlighted as a potential method for fresh-keeping RtE products and improving the sensitivity of products against temperature abuses in the food cold chain. The focus of the limited available studies with edible coatings for RtE seafood products has been on microbiological control, particularly on *L. monocytogenes*, where coatings were tested as an additional protective layer with a commonly used primary packaging. To further develop the concept of edible coatings for RtE seafood products, it is recommended to broaden coating development studies to include other critical degradation processes. Developing coatings that can control multiple degradation processes simultaneously would increase the protection effect of edible coatings.

An important aspect driving the use of edible coatings is their potential to reduce the environmental impact of packed food products, by no additional waste generation, being biobased and improving the product shelf life. Life cycle studies are recommended to further highlight the potential of edible coatings to improve the environmental impact of packed food products. A circular economy approach considered during the development of an edible coating can create new markets for waste products of the maritime industry and biomass. Hurdles identified for the use of edible coatings for industrial products are the consideration of food appearance, regulations, labelling and consumer acceptance. Market value opportunities related to edible coatings are product customization and personalization options.

Different methods are applied to tailor coating properties to develop coatings with higher protection functions. Challenges are related to counter-effects between protection properties and sensory properties. The investigation on the applied coating application methods showed that it could be of value to study not only the dipping method, but also to study the spraying method in lab scale studies, to increase the potential to bridge the gap between laboratory and industrial applications. Developing standardised methodologies for testing edible coatings could improve the current difficulty of comparing edible coating performances among different studies.

Overall, edible coatings are of growing interest for RtE seafood products, due to sustainable, functional, and customization aspects. While edible coatings can offer potential benefits in the RtE food sector, there are several challenges that must be addressed to ensure their successful implementation. Research and development efforts are needed to further demonstrate their compatibility with food products, optimize their performance and stability, while ensuring compliance with regulatory requirements and addressing cost concerns. The development of edible coatings for RtE seafood products is an interdisciplinary task that needs to take into consideration technology, engineering, environmental and consumer aspects. Application of edible coatings as an integrated part of a RtE seafood product-packaging concept to tackle current challenges has promising prospects and should be further researched to explore the full potential.

Chapter FOUR

# Identification of critical quality parameters and product degradation process

## Abstract

*An optimized packaging system is product specific and developed for a defined supply chain scenario. Investigating the product to be packed and understanding interactions with the product, packaging and environment is of significant relevance, and should be the starting point for a packaging development. The aim of this chapter was to summarize product properties, identify critical quality parameters of a ready to eat (RtE) baked white fish fillet and a RtE minced fish patty to understand the product degradation during storage by studying chemical, microbiological and physical product properties. The RtE baked fish product sorption isotherm behaviour showed a sigmodal shape, and the best model fitting was obtained with the Peleg model ($R^2$: 0.9995,SSE 0.008), followed by the GAB model ($R^2$: 0.995, SSE 0.01). The storage experiments revealed that water loss and microbial growth are important product quality parameters for both RtE seafood products tested. Fat oxidation is only a critical quality parameter for RtE seafood products with fat content higher than 6%. By understanding the underlying reactions and factors that influence the identified critical quality parameters, required packaging properties such as oxygen, light, and water vapour barrier could be identified. Additionally, antimicrobial packaging properties should be considered.*


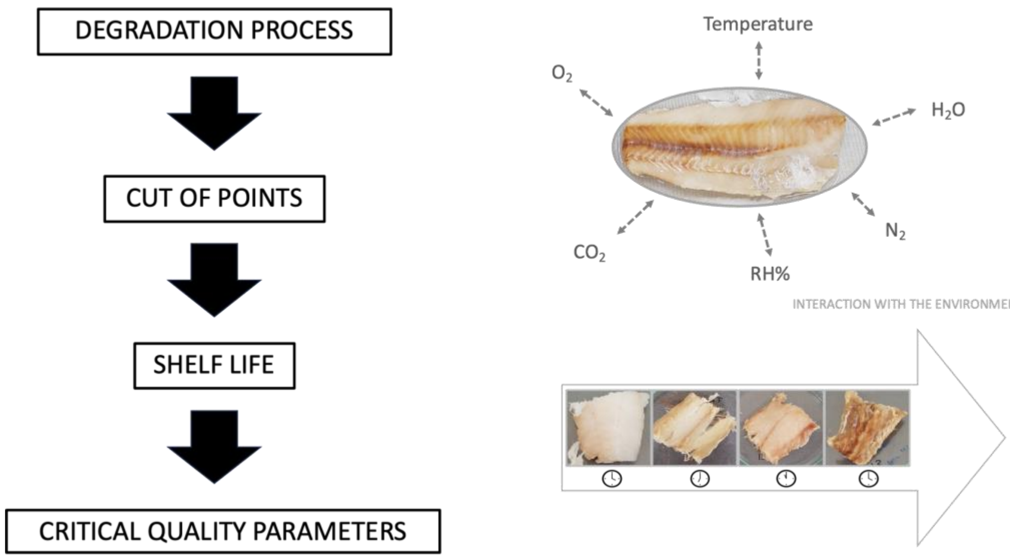


*Figure 4-1: Graphical Abstract - Identification of critical quality parameters and product degradation process.*


## Keywords

*Critical quality parameters*
*RtE fish product*
*Sorption isotherm*
*Degradation processes*

## 4.1 Introduction

Ready to eat (RtE) food products are a favoured product choice for different consumer groups, mainly as they provide convenience. Beside convenience RtE seafood products presents, a healthy food choice, another increasing consumer demand. RtE seafood products belong to the category of highly perishable food products and an appropriate packaging selection is important. For the development of a packaging design with a quantitative approach an important step is to understand the product degradation processes (Robertson, 2012) and identify critical quality parameters.

The sorption isotherm of a product is a useful measurement in food science and technology to get a better understanding of product stability, present product degradation reactions and influences of storage conditions (Ahmat et al., 2014; Robertson, 2012). In packaging development, sorption isotherm analysis provides useful information to determine appropriate packaging materials with properties to control degradation processes and established a more rational bases to improve packaging design (Comaposada et al., 2000).

A sorption isotherm describes the relationship between the water content and the water activity of a product at a defined temperature. Water activity is a critical parameter in the food industry, as it is closely related to product spoilage, texture, flavour, appearance and overall product stability, safety, and shelf life. The water activity of the product relates to the available water in a product and depends on the relative humidity of the surrounding air (Comaposada et al., 2000). The water content of a product is defined as all moisture present within the food product. Food products typically exhibit a range of sorption behaviours, depending on their composition, structure, and processing conditions and are categorized into type I to V (Andrade P. et al. 2011).

Sorption isotherms of diverse food products have been studied by many authors, although only few studies considered RtE products. There are several sorption isotherm models that are commonly studied to describe the moisture sorption behaviour of food products as Langmuir equation, Brunauer-Emmett-Teller model, Oswin model, Smith model, Halsey model, Henderson model, Iglesias-Chirife model, Guggenheim-Anderson-de Boer (GAB) model and Peleg model. These models are either theoretical based, empirical based or simplifications of more elaborated models (Andrade et al., 2011). Whereby, for food with a high-water activity the GAB model, Halsey model, Smith model, and Peleg model were identified as most suitable (Ahmat et al., 2014; Comaposada et al., 2000; Singh et al., 2006).

Understanding the degradation processes of a food product provides fundamental information for defining packaging requirements. Only few studies are available investigating the safety and quality deterioration of heat treated RtE seafood products during storage (González-Fandos et al., 2005). Critical quality parameters (CQP) are quality attributes that describe the product quality characteristics. By defined safety and quality limits for the CQP of a food product the overall product quality and safety can be assessed.

The aim of this study was to understand the product degradation processes and identify CQP for two RtE seafood products. The sorption isotherm characteristics at different temperatures were determined to understand the product behaviour during storage. The resulting data were used to generate a sorption isotherm curve that shows the relationship between water activity and moisture content of the food product. The model fitting was evaluated to find the most suitable model. A storage experiment was performed to study the product degradation process and identifying CQP. As a last steps, packaging requirements were defined based on the obtained results.

## 4.2 Materials and Methods

### 4.2.1 Chemicals

Lithium chloride, magnesium chloride hexahydrate, magnesium nitrate hexahydrate, potassium acetate, peptone buffer, plate count agar, potassium chloride, potassium sorbate, potassium sulphate, sodium bromide, sodium chloride, 1,1,3,3-tetraethoxypropane, thiobarbituric acid and trichloroacetic acid were purchased from Sigma-Aldrich.

### 4.2.2 Sample preparation

Two different products were used in this study, a baked white fish fillet and a RtE fish patty (Figure 4-2). For the baked fish fillet, frozen white fish fillet was purchased from a local supermarket. The frozen fillets were placed on a baking tray and prepared with a mild heating treatment at 80°C for 70 min. The baked fillets were allowed to cool down to room temperature in a sterile storage box to avoid any cross contamination, before the fillets were cut into pieces of 20 to 25 g. The samples were examined and those pieces with a similar thickness were used for the study. Fish fillet patties were provided by SeafoodAge project partner (WP5) at the CETMA in Lisbon. The patties are restructured fish product produced from fish mince of grey gurnard (*Eutrigla gurnardus*), horse mackerel (*Trachurus trachurus*), and pout (*Trisopterus luscus*) in the ration of 1:1:1. The fish patties were double heat treated, by first baking in an oven at 60°C for 40 min and then fried in a pan for 1 min on each side. Afterwards the samples were frozen to -18°C for transportation. Before use, the fish patties were slowly thawed at 4°C, cut into equal pieces with an average weight of 15 g.

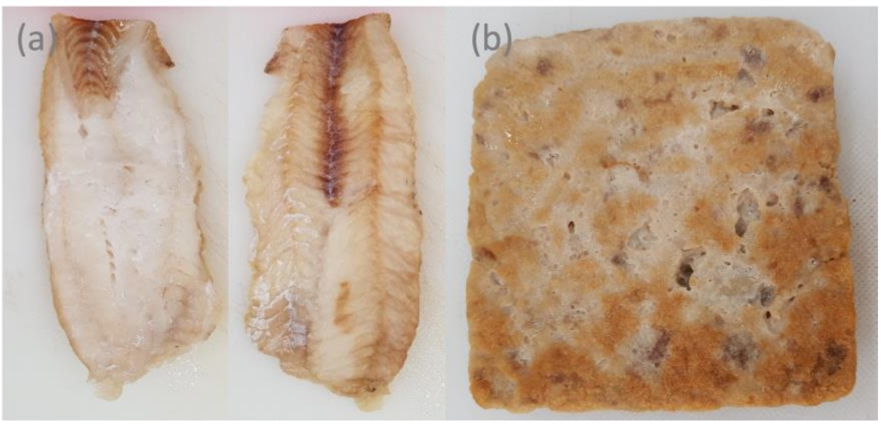


*Figure 4-2: RtE seafood samples. (a) baked white fish fillet (b) baked and fried minced fish patty.*

### 4.2.3 Determination of desorption isotherm

The moisture desorption isotherm of baked fish samples was obtained in duplicate at two temperatures (4 and 14°C). Desorption isotherm of baked fish samples was determined by using the gravimetric static method by placing around 4 g sample into hermetically sealed Kilnar jars containing saturated salt solutions in the humidity range of 0.12 to 0.99 (Table 4-1), stored at 4 and 14°C until equilibrium was reached. The weight changes of the samples were analysed in defined intervals by opening the jar, weighting the samples on a precision balance (Sartorius, Germany), and replacing the sample into the jar. The time when the equilibrium was reached was defined as less than 1 mg/g solid sample weight different to the previous measurement. To control deterioration processes of samples stored at relative humidity's greater than or equal to 50%RH, the sample surface was sprinkled with 0.05 mg potassium sorbate per g sample (Sigma-Aldrich, Germany). Potassium sorbate was reported to not influence the sorption isotherm of meat products (Ahmat et al., 2014; Singh et al., 2006). The water content of the equilibrated samples was measured by drying in an oven for 24 h at 104°C. Samples were crushed before drying to increase the surface area. Afterwards, samples were placed in a desiccator with silica for cooling to room temperature before weighting. The initial sample weight and weight after drying was measured with a precision balance. The water content was calculated based on the initial weight ($W_0$) and the drying weight ($W_s$) according to equation 4.2.-1. The relation between moisture content dry basis and wet basis is shown in equation 4.2-2. The water activity of the samples was measured with the aid of a water activity meter (AquaLab, USA) at 15°C.

$$m_{db,i} = \frac{W_0 - W_s}{W_s} \qquad \text{eq. 4.2-1}$$

$$m_{db} = \frac{m_{wb}}{1 - m_{wb}} \text{ or } m_{wb} = \frac{m_{db}}{1 + m_{db}} \qquad \text{eq. 4.2-2}$$

Table 4-1: $a_w$ value and solubility of saturated salt solutions (Bravin et al., 2006; KITIC et al., 1986; Wang et al., 2013).

| **Saturated salt solution** | **Chemical formular** | **5°C** | **15°C** | **15°C*** | **Water solubility [g/100g $H_2O$] @XX°C** |
|---|---|---|---|---|---|
| Lithium chloride | LiCl | 0.13 | 0.12 | 0.115 | 56.9 @20°C |
| Potassium acetate | $CH_3COOK$ | - | - | 0.238 | 9.82 @20°C |
| Magnesium chloride hexahydrate | $MgCl_2$ | 0.34 | 0.33 | 0.339 | 46.87 @20°C |
| Magnesium nitrate hexahydrate | $Mg(NO_3)_2$ | - | - | 0.573 | 42.0 @20°C |
| Sodium bromide | NaBr | - | - | 0.610 | 94.3 @25°C |
| Sodium chloride | NaCl | 0.76 | 0.753 | 0.772 | 31.7 @20°C |
| Potassium chloride | KCl | 0.87 | 0.859 | 0.876 | 35.5 @25°C |
| Potassium sulphate | $K_2SO_4$ | | 0.979 | 0.985 | 12.0 @25°C |

*measured with Aqualab

### 4.2.4 Modelling of desorption isotherm

Different sorption isotherm models have been developed to describe the different relationships between the water activity and moisture content for food products. Six sorption isotherm models were tested to model the sorption isotherm data of a baked fish product at two temperatures (Table 4-3). These models were selected based a good model fit for products with a similar sorption isotherm behaviour (Mbarek and Mihoubi, 2019). Statistica software was used to fit the sorption models to the experimental data and determining the model parameters. A non-linear least square regression analysis was used to predict the model parameters. Only models with a good acceptance represented by a random distribution of values in the residual plots were further investigated. The closeness of the fit of tested models was evaluated by the following statistical parameters: sum of square of residuals (SSR), mean square error (MSE), coefficient of determination ($R^2$), mean relative deviation modulus (E), and root mean square error (RMSE) (Table 4-2).

Table 4-2: Statistical parameters for evaluation of the closeness of fit.

| | |
|---|---|
| $SSR = \sum_{i=1}^{N}\left(m_{exp,i} - m_{cal,i}\right)^2$ | eq. 4.2-3 |
| $MSE = \frac{SSR}{(N-s)}$ | eq. 4.2-4 |
| $R^2 = 1 - \frac{SSR}{\sum_{i=1}^{N}\left(\overline{m_{exp,i}} - m_{exp,i}\right)^2}$ | eq. 4.2-5 |
| $E\% = \frac{100}{N}\sum\frac{\lvert m_{exp,i} - m_{cal,i}\rvert}{m_{exp,i}}$ | eq. 4.2-6 |
| $RMSE = \sqrt{\frac{1}{N-s}\sum_{i=1}^{N}\left(m_{exp,i} - m_{cal,i}\right)^2}$ | eq. 4.2-7 |

Abbreviations: N: number of experimental data, $m_{exp,i}$ : experimental moisture content dry basis at specific water activity i [db%], $m_{cal,i}$: calculated moisture content dry basis at specific water activity i [db%], s: model parameter, $\overline{x_{exp,i}}$ : mean of N experimental moisture content

Table 4-3: Sorption isotherm models.

| | | |
|---|---|---|
| Guggenheim-Anderson-de Boer (GAB) model | $\frac{m_{db,i}}{X_m} = \frac{C_g \cdot k_{GAB} \cdot a_w}{(1 - k_{GAB}a_w)(1 + C_g k_{GAB} a_w - k_{GAB} a_w)}$ | eq. 4.2-8 |
| Hailwood-Horrobin model | $m_{db,i} = \left(\frac{E}{a_w} + B - C_h a_w\right)^{-1}$ | eq. 4.2-9 |
| modified Hailwood-Horrobin model | $m_{db,i} = \left(T_K\left(\frac{E}{a_w} + B\right) - \frac{C_h}{T_K}a_w\right)^{-1}$ | eq. 4.2-10 |
| modified Oswin model | $m_{db,i} = (\mathrm{E} + BT_K)\left[\frac{a_w}{1 - a_w}\right]^{1/C_o}$ | eq. 4.2-11 |
| Halsey model | $m_{db,i} = \left[-\frac{E}{\ln(a_w)}\right]^{\frac{1}{B}}$ | eq. 4.2-12 |
| Oswin model | $m_{db,i} = \mathrm{E}\left(\frac{a_w}{1 - a_w}\right)^B$ | eq. 4.2-13 |
| Lewicki (A) | $m_{db,i} = \frac{E}{(1 - a_w)^B} - \frac{E}{(1 + a_w^C)}$ | eq. 4.2-14 |
| Lewicki (B) | $m_{db,i} = \mathrm{E}\left(\frac{1}{a_w} - 1\right)^{B-1}$ | eq. 4.2-15 |
| Peleg | $m_{db,i} = \mathrm{E}\, a_w^B + C a_w^D$ | eq. 4.2-16 |

Abbreviations: $m_i$ : moisture content dry basis at specific water activity i [db], $m_{m,GAB}$: moisture content corresponding to saturation of all primary adsorption sites by one water molecule (equivalent to the BET monolayer), $a_w$ : water activity, $k_{GAB}$ : Dimensionless GAB parameter (related to the heat of sorption of the multilayer region), $C_g$: Dimensionless GAB parameter or Guggenheim constant (related to the heat of sorption of the monolayer region), $C_O$: Dimensionless parameter constant, $C_h$: Dimensionless Hailwood-Horrobin parameter constant, B;C,D,E: Dimensionless parameter, $T_K$: absolute Temperature [K]

### 4.2.5 Storage test

The storage test was performed under defined temperature and controlled air circulation. The samples were placed into a storage box with a lid before placing the box into an incubator at 4°C or 14°C. The storage conditions were selected to assemble a realistic scenario. The effect of the air circulation was eliminated to decrease the rate of water loss to some extent and were reconsidered in the development of the outside layer. The selected storage conditions provided a closer scenario to the final packaging system with an outside layer.

### 4.2.6 Characterisation of product quality

Thiobarbituric acid reactive substance assay (TBARS) was used to determine the lipid oxidation of samples based on Rosmini et al. (1996). Briefly, 4 g sample was mixed with 10 mL of 10% trichloroacetic acid (w/v). Then, 5 mL of control solution, containing an aliquot of 30 μL of a $10^{-3}$ M 1,1,3,3-tetraethoxypropane or 5 mL of distilled water was added and vortex for 5 min. Freshly prepared 0.001 M thiobarbituric acid solution was added (5 mL) and mixed for 1 min. The reaction mixture was centrifugated at 3500 rpm for 5 min (Universal 16R, Hettich, Tuttlingen, Germany) and the supernatant was filter through a Whatman No. 1 filter. Afterwards the mixture was heated in a water bath (WB14, Memmert, Schwabach, Germany) at 95°C for 35 min and placed for 10 min into a cooled water bath for cooling. The supernatant absorbance was recorded against a blank, prepared as described above without the addition of sample, at 532 nm using a spectrophotometer (Libra S22, Biochrom, Cambridge, England). Standard solutions were prepared using 1,1,3,3-tetraethoxypropane to quantify the malondialdehyde (MDA) content, and results were expressed as mg MDA equ./Kg sample.

The weight of RtE fish samples was measured using a precision balance (Sartorius, Germany). The weight loss ($W_L$) in % was calculated based on the initial ($W_0$) and weight at time i ($W_i$) of the product, as follows:

$$W_L = [(W_0 - W_i)/W_0] \cdot 100 \qquad \text{eq. 4.2-17}$$

The total aerobic count (TAC) of the product was analysed by aseptically transferring around 10 - 15 g of sample to a sterile stomacher bag (Sewards LTD, UK) and adding peptone buffer to achieve a 10-fold dilution. The sample was mashed, and hand shacked for 2 min. Appropriated dilutions were prepared of the resulting suspension with serially diluting in peptone buffer. For the total microbial count, the pour plate method with a plate count agar (Sigma Aldrich) was applied. The agar plates were incubated at 30°C for 72 h (Zarandona et al., 2021). Results were expressed as $\log_{10}$ colony forming units per gram sample (log CFU/g).

# 4.3 Results & Discussion

## 4.3.1 Desorption moisture isotherm

The sorption isotherm of RtE baked fish product was constructed by plotting the equilibrium moisture content at steady-state wet basis and dry basis against the water activity (Figure 4-3). The sorption isotherm of baked fish fillet showed a sigmodal shape for both tested temperatures, a typical type II isotherm according to the classification by Brunauer et al. (1940). With a slowly rising water content from 7 to 20 wb%, with a simultaneously fast increasing of the water activity from 0.1 to 0.88. In the water activity range from 0.88 to nearly 1, the water content increases from 30 up to 75 wb%. Many food products follow a sigmodal sorption isotherm shape (Timmermann et al., 2001). A sorption isotherm in a similar maximum water content range as measured for the baked fish product around 3 db% was reported for fresh fish and apples (Garcia-Perez et al., 2014; Mbarek and Mihoubi, 2019; Toujani et al., 2011). Other authors reported a higher maximum moisture content around 120 db% for fish and meat products (Delgado and Sun, 2002; Ponwiboon and Rojanakorn, 2017). Differences can be explained due to the applied baking process. Due to the heat treatment, proteins denaturated and water bound to the proteins was released (Skipnes et al., 2011). Most of the remaining water is present as free water as weak water bounding sides are remaining. Resulting in an initial water content after baking of around 3 db% (75 wb%) and a high water activity.

Figure 4-3 also shows the temperature effect of the desorption isotherm of RtE baked fish product. As the water activity is temperature dependent the sorption isotherm at 4 and 14°C was studied. Only small differences between the sorption isotherms at the tested temperatures could be seen. Statistically no significant difference was observed. At a given water content, the water activity value is expected to be higher at higher storage temperature (Comaposada et al., 2000; Robertson, 2012). In the obtained results, this can be only confirmed with water activity value above 0.6.

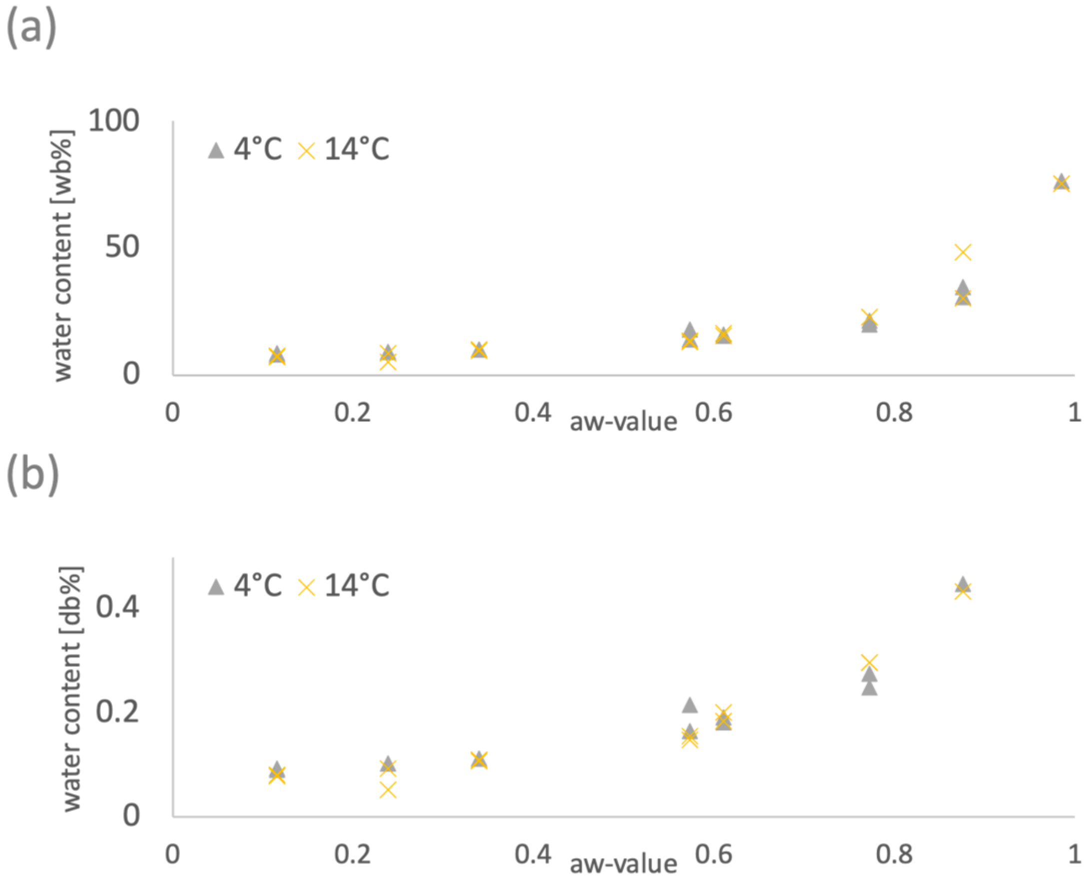


*Figure 4-3: Sorption isotherm of baked fish fillet determined with a gravimetrical static method with (a) water content wet basis [wb%] and (b) water content dry basis [db%].*

### 4.3.2 Sorption isotherm modelling

Different mathematical models were developed to describe sorption isotherms of food products. Therefore, different models were tested to find the best correlation of the experimental moisture adsorption data of the RtE baked fish product with sorption isotherm model. Based on literature search the GAB, Halsey, Oswin, Lewicki, and Peleg sorption isotherm models were most suitable for similar products (Garcia-Perez et al., 2014; Mbarek and Mihoubi, 2019; Toujani et al., 2011). The estimated specific model parameters and the corresponding model fitting parameters are shown in Table 4-4. The results showed that the best model fit at both tested temperatures were obtained with the Peleg model, followed by the GAB model. The $R^2$ was close to 1, with 0.9995 and 0.9911, and a MSE close to 0, with 0.0038 and 0.0026. The residual plots and the other tested statistical parameters were in an acceptable range. The relation between the experimental and predicted equilibrium moisture content values (db%) is shown in Figure 4-4a. The diagnosis plot between experimental and predicted values of equilibrium moisture content (Figure 4-4b), and the relation between experimental and predicted equilibrium moisture content values (Figure 4-4c) at 4°C and 14°C showed a good fit.

Table 4-4: Estimated model constants and model parameters.

| Model | Parameters and criteria's | Temperature | |
|---|---|---|---|
| | | **4°C** | **14°C** |
| Halsey | A | 0.0264 | 0.0290 |
| | B | 1.1839 | 1.3111 |
| | $R^2$ | 0.9989 | 0.9893 |
| | SSE | 0.0187 | 0.1617 |
| | MSE | 0.0013 | 0.0116 |
| | E% | 18.4243 | 20.7987 |
| | RMSE | 0.0365 | 0.1075 |
| Oswin | A | 0.1099 | 0.1513 |
| | B | 0.8083 | 0.7213 |
| | $R^2$ | 0.9976 | 0.9897 |
| | SSE | 0.0390 | 0.1557 |
| | MSE | 0.0028 | 0.0111 |
| | E% | 30.0936 | 24.1155 |
| | RMSE | 0.0528 | 0.1055 |
| Lewicki (A) | A | 0.0818 | 0.1518 |
| | B | 0.8788 | 0.7237 |
| | C | -5.1061 | 0.5518 |
| | $R^2$ | 0.9996 | 0.9896 |
| | SSE | 0.0072 | 0.1567 |
| | MSE | 0.0006 | 0.0121 |
| | E% | 5.1274 | 21.7317 |
| | RMSE | 0.0235 | 0.1098 |
| Lewicki (B) | A | 0.1099 | 0.1513 |
| | B | 0.1917 | 0.2787 |
| | $R^2$ | 0.9976 | 0.9897 |
| | SSE | 0.0390 | 0.1557 |
| | MSE | 0.0028 | 0.0111 |
| | E% | 30.09 | 24.12 |
| | RMSE | 0.0528 | 0.1055 |
| GAB | Xm | 0.0637 | 0.0613 |
| | C | 11.04 | 4871 |
| | K | 0.9954 | 0.9951 |
| | $R^2$ | 0.9988 | 0.9993 |
| | SSE | 0.0103 | 0.0049 |
| | MSE | 0.0021 | 0.0010 |
| | E% | 21.45 | 11.06 |
| | RMSE | 0.0453 | 0.0313 |
| Peleg | A | 4.1021 | 3.5841 |
| | B | 21.3479 | 15.6037 |
| | C | 0.2713 | 0.2635 |
| | D | 0.6584 | 0.7623 |
| | $R^2$ | 0.9995 | 0.9911 |
| | SSE | 0.0080 | 0.1337 |
| | MSE | 0.0038 | 0.0026 |
| | E% | 10.50 | 18.55 |
| | RMSE | 0.0258 | 0.1056 |

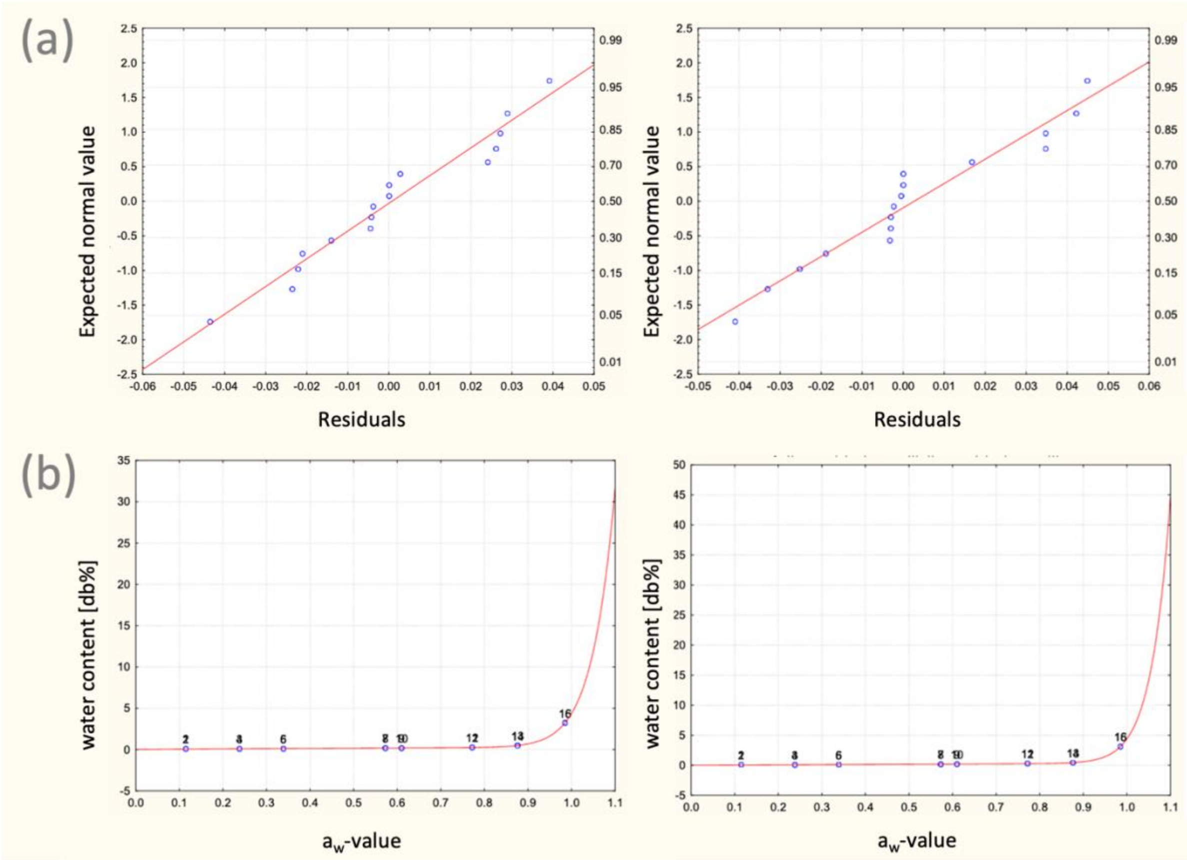


*Figure 4-4: Normal plot of the model residuals (a) and relation between experimental and predicted equilibrium moisture content values (b) for 4°C (left) and 14°C (right).*

### 4.3.3 Product degradation during cold storage

First, the initial state of the two RtE seafood products were investigated. Both products have an initial water content of around 75 wb% and showed a high water activity. The protein content is in a similar range for both products. The two products differ in the fat content, of 0.8% for the baked fish fillet and 6% for the minced fish patty as well as in the salt content. In the production process of the minced fish patty seasonings were added, including salt.

Table 4-5: Product composition of investigated RtE fish products.

| Parameter | Baked fish fillet | Minced fish patty |
|---|---|---|
| Water content [%] | 75 | 75 |
| Water activity [%] | 0.95 | 0.93 |
| Fat content [%] | 0.8 | 6* |
| Salt content [%] | 0 | 0.5 |
| Weight per piece [g] | 120 | 90 |
| Dimensions [mm] | 160 x 70 x 15 | 80 x 80 x 10 |

*estimated based on the patty composition

With the aid of the products initial water contents and the obtained sorption isotherm potential moisture content relative reactions rates can be detected based on Labuza and Dugan, Jr. (1971). The expected moisture content relative reaction rates in products with a high water activity in descending order is lipid oxidation, enzyme activity, mould growth, yeast growth and bacteria growth. For investigating the safety & quality of the products microbiological, chemical, and physical changes were analysed. These expected main degradation reactions were studied during storage to understand the underlying degradation processes of the products by measuring the fat oxidation, water loss and microbiological growth. A constant cold chain of 4°C is recommended for this food category as products which received only a mild heat treatment are likely to spoil due to microbial action (Gram and Huss, 1996). As already small changes can influence the microbiological growth significantly (González-Fandos et al., 2005), in this study additionally the shelf life was determined under abused storage conditions of 14°C. Understanding the underlying degradation process are valuable information for selecting the right packaging system to ensure appropriate handling during transportation and storage.

The obtained results for the baked fish product are summarized in Table 4-6. Lipid oxidation occurred during storage, first at a slow rate and with longer storage times with a higher reaction rate presented by the increasing values. Samples stored at 4°C for 21 days showed the highest value of 4.4 mg MDA eq./Kg sample. The measured value after 28 days showed a lower value as after 21 days, these results were unexpected as the MDA value in the product is accumulating during storage. Therefore, this value is mostly likely a result of a measuring error. Additionally, it could be seen that the fat oxidation is slightly enhanced at higher storage temperatures. Represented by the measured values of 1.1 mg MDA eq./Kg sample at 4°C and 1.8 mg MDA eq./Kg sample at 14°C. The water loss during storage is significant with a water loss of around 16% after 28 days of storage at 4°C. When the sample is stored at 14°C the water loss rate is doubled compared to samples stored at 4°C, with a water loss of 3.7% and 1.5% after 7 days respectively. The food safety of the product was observed by measuring the total aerobic count. The product had an initial microbial load of below 1 log CFU/g sample, representing a safe product. During storage the microbial load was increasing. The results showed that the growth rate is significantly influenced by the storage temperature. After 7 days of storage the measured total aerobic count for the baked fish samples was around 3.3 log CFU/g sample at 4°C and around 9.1 log CFU/g sample at 14°C.

The degradation process of the RtE fish patty were observed and the results are represented in Table 4-7. Main differences to the RtE baked fish fillet could be seen in the fat oxidation. The initial MDA value of the tested samples already showed an increased value of around 7.7mg MDA/Kg sample, during storage at 4°C this value increased to up to 17 mg MDA/Kg sample at 4°C. The higher fat oxidation can be explained due to the higher fat content of the product (Table 4-5). Interesting is the temperature effect on the fat oxidation, as the results after 7 days of storage at 4°C with 16.5 mg MDA/Kg samples are higher than samples stored at 14°C with 11.6 mg MDA/Kg sample. Additionally, a higher water loss rate was observed for the RtE fish patty during storage at both tested storage temperatures. The microbiological growth was in a similar range for both tested RtE samples.

Table 4-6: Lipid oxidation, water loss and total aerobic count of RtE baked fish product stored at 4°C (a) and 14°C (b).

(a)

| Time [days] | 0 | 7 | 14 | 21 | 28 |
|---|---|---|---|---|---|
| Lipid oxidation [mg MDA eq./kg sample] | 0.16 ±0.03 A | 1.10 ±0.62 A | 1.17 ±0.39 A | 4.38 ±1.20 B | 1.85 ±0.76 A |
| Water loss [%] | | 1.50 ±0.49 A | 3.65 ±0.98 A | 9.96 ±4.02 A | 14.52 ±5.04 B |
| Total aerobic count [log CFU/g sample] | < 1.06* ±0.10 A | < 2.86* ±0.84 A | 7.63 ±0.75 B | 9.10 ±1.04 B | - |

(b)

| Time [days] | 0 | 2 | 4 | 7 |
|---|---|---|---|---|
| Lipid oxidation [mg MDA eq./kg sample] | 0.16 ±0.03 A | 0.54 ±0.53 A | 0.66 ±0.27 A,B | 1.90 ±0.82 B |
| Water loss [%] | - | 1.16 ±0.35 A | 2.70 ±0.44 B | 3.70 ±0.32 B |
| Total aerobic count [log CFU/g sample] | < 1.06* ±0.10 A | 4.86 ±0.74 B | 7.19 ±0.58 C | 9.11 ±0.45 D |

*indicates an estimated value due at least one colonies count of the triplicates below 30 or above 300. >/< indicates the direction to the potential real TAC value

Table 4-7: Lipid oxidation, water loss and total aerobic count of RtE fish patty stored at 4°C (a) and 14°C (b).

(a)

| Time [days] | 0 | 7 | 14 | 21 | 28 |
|---|---|---|---|---|---|
| Lipid oxidation [mg MDA eq./kg sample] | 7.69 ±0.46 A | 16.51 ±0.57 B | 18.43 ±0.12 C | 16.15 ±0.58 B | 17.04 ±0.53 B,C |
| Water loss [%] | - | 1.59 ±0.31 A | 6.55 ±0.06 A | 16.70 ±2.13 B | 24.54 ±0.46 C |
| Total aerobic count [log CFU/g sample] | < 2.16* ±0.89 A | < 3.31* ±0.71 A,B | < 5.32* ±0.88 B | 8.73 ±0.68 C | < 9.26* ±0.56 C |

(b)

| Time [days] | 0 | 2 | 4 | 7 |
|---|---|---|---|---|
| Lipid oxidation [mg MDA eq./kg sample] | 7.69 ±0.46 A | 8.28 ±1.23 A,B | 11.65 ±0.79 B | 11.58 ±1.71 B |
| Water loss [%] | | 3.09 ±0.52 A | 4.41 ±0.34 A,B | 6.76 ±0.88 B |
| Total aerobic count [log CFU/g sample] | < 2.16* ±0.89 A | 4.22 ±0.45 A | 8.53 ±0.41 B | 9.12 ±0.03 B |

*indicates an estimated value due at least one colonies count of the triplicates below 30 or above 300. >/< indicates the direction to the potential real TAC value

To be able to define a shelf life based on the CQP the allowed maximum deterioration values were defined based on regulations and literature values. The cut off point for the fat oxidation was set to an increase of 8 mg MDA/Kg sample, for microbiological colony count below 7 log CFU/g sample and the accepted water loss before rejection was set to 10% weight loss. Based on the set cut off points, the shelf life of the RtE fish patty without packaging stored under chilled conditions at 4°C, with no air circulation was found to be below 7 days. The most critical quality parameter was the fat oxidation, followed by total aerobic count and water loss. The shelf life of the RtE baked fish product was determined between 7 and 14 days. The most critical quality parameter was the total aerobic count, followed by the moisture loss. The fat oxidation of the baked fish product was below the cut off point for the measured time of 28 days and is therefore higher than the desired shelf life value. For the baked fish product, the fat oxidation was not a critical quality parameter.

The critical quality parameters for RtE seafood products were defined as total aerobic count to ensure a safe product, as well as the fat oxidation to avoid off flavour formation in the product and as the third CQP the water loss was defined as the appearance of the product during storage is highly influenced by the water loss. Nevertheless, the results showed that the order of significant of the quality parameters can depends on the product composition and processing steps.

### 4.3.4 Investigation of supply chain

RtE seafood products can be categorized as perishable food and needs refrigeration since it is susceptible to post-cooking contamination and/or to the multiplication of microorganism that can be present after the mild heat treatment. A desired product shelf life of 14-21 days was estimated based on a potential distribution chain as presented in Table 4-8. A certain shelf life is inevitable to be suitable for the industrial food sector and consumer e.g., to avoid food waste and provide convenience. The aim of packaging design should be to achieve a minimum shelf life of 14 up to 21 days. A balance between a reasonable shelf life goal is important for the packaging development process to avoid overprotection, potential leading to a higher packaging environmental impact.

Table 4-8: Typical distribution supply chain of RtE seafood product.

| **Links in the supply chain** | **Day** | **Average temperature** |
|---|---|---|
| Processing | 1 | 4°C |
| Packing | 1 | 4°C |
| Cold storage at processing facilities | 1-2 | 4°C |
| Transportation to wholesaler | 2 | 4°C |
| Storage at wholesaler | 2-3 | 4°C |
| Transportation to retailer | 3 | 4°C |
| Storage at retailer | 3-7 | 4°C |
| Transport to display cabinet in retailer store | 3-14 | 4°C |
| Transport to consumer | 3-14 | 4°C |
| Storage at consumer | 3-14 | 4°C |
| Total | 14-21 | |

### 4.3.5 Selecting of packaging requirements

The deterioration reactions responsible for the chances of the critical quality parameters are chemical, biochemical reaction for the fat oxidation, physical chances for the water loss and biological chances for the total aerobic count. The related quality changes, consequences, nature of the reaction and factors influencing the reaction are presented in Table 4-9. Moreover, it was ascertained which of the CQP might be influenced by the packaging system and which packaging material properties needs to be controlled. The packaging should be able to control the microbiological growth, either providing antimicrobial properties, or by controlling the gas composition in contact with the product. Additionally, a water barrier is necessary to control the water loss during storage. For RtE seafood products with a high fat content, the fat oxidation should be controlled by limiting oxygen and light permeability. The fat oxidation is only relevant for products with a higher fat content.

Table 4-9: Identified critical quality parameter and there CoP, deterioration reaction, quality changes, consequences, nature of reactions, factors that control the reaction and the potential effect of a secondary package, cold storage and chitosan coating *(Anacleto et al., 2011; Robertson, 2012)*.

| **CQP** | **Fat oxidation** | **Water loss** | **Total aerobic count** |
|---|---|---|---|
| Cut of points | > 8 mg equ. MDA/Kg sample | 10% | < 7 log CFU/g |
| Deterioration reaction origin | Chemical or biochemical | Physical | Biological |
| Quality changes | Sensory and nutritional | Sensory and appearance | Safety and nutritional |
| Consequences | Loss of essential fatty acids,<br>off - flavour,<br>formation of toxicologically compounds,<br>loss of nutritive value | Loss of water<br>Texture changes as shrinking, toughening and colour chances.<br>Loss of water holding capacity | Food spoilage – Food safety<br>Degradation of proteins & minerals<br>Generation of toxic substances |
| Nature of this reactions | Auto-oxidation | Partial water pressure, Water gradient | Extensive multiplication of the MO's in the food |

| | | | |
|---|---|---|---|
| Factors that control/influence the reaction rates | Light<br>Local $O_2$ concentration<br>High temperature<br>Presence of catalysts<br>$a_w$-value | Temperature<br>Light<br>RH<br>Air flow | Initial microbiological load<br>pH<br>$a_w$<br>$E_h$<br>Antimicrobial constituent's biological structures<br>Temperature<br>RH surrounding air |
| Required packaging properties | Oxygen barrier<br>Headspace volume<br>Light barrier | Water vapour barrier | Oxygen barrier<br>Light barrier<br>Antimicrobial properties |

## 4.4 Conclusion

The sorption isotherm was studied to understand the sorption characteristics of a RtE baked fish product. A static gravimetrical method was applied in the $a_w$ range from 0.12 to 0.98. Additionally, the effect of the storage temperature on the sorption isotherm behaviour was studied at 4 and 14°C. Different models were tested to describe the relationship between the equilibrium moisture content and water activity. The sorption isotherm of a baked fish product showed a typical type II sorption isotherm shape as most food products. No significant differences could be observed between the sorption behaviour at 4°C and 14°C. All tested statistical models, GAB, Halsey, Oswin, Lewicki, and Peleg, showed a good model fitness. The highest goodness of fit was achieved with the Peleg model, closely followed by GAB model over the whole range of water activity and investigated temperature range. The sorption behaviour of the baked fish product gives reliable insights about present degradation processes. Sorption isotherm behaviour of a food product provides important information for the design of an optimal food packaging system.

The water loss and microbial growth could be identified as critical quality parameters for both investigated RtE seafood products, whereby the fat oxidation is only a critical factor when the fat content of the product is similar or higher as 6%. The cut of points for each critical quality parameter was defined and used to predict the product shelf life. The RtE baked fish product has a shelf life between 7 to 14 days, mainly restricted by the microbial growth. Predicted shelf life of the RtE fish patty was only below 7 days, mainly due to rapid fat oxidation. The potential supply chain for a RtE seafood product was investigated and the required shelf life was estimated to be around 14 to 21 days. Based on the results the following required packaging properties could be identified: oxygen barrier, light barrier, and water vapour barrier. Additionally, antimicrobial properties of the packaging can provide safety to the product. The gained information provides a fundamental knowledge for the development of an integrated precision food packaging, providing sufficient product safety and quality.

Chapter FIVE

# Design and development of an edible coating for a ready to eat seafood product

## Abstract

*The application of chitosan and alginate coatings for a ready to eat (RtE) baked fish product were studied. A full factorial experimental design was used to investigate the effect of coating material composition on microbial growth, water loss and lipid oxidation under optimal (4°C) and abuse (14°C) storage conditions. A 3^2 full factorial design was used to study the effect of chitosan concentration (1, 2 and 3%), and glycerol concentration (0, 15 and 30% w/w chitosan). The effect of the composition of an alginate coating was studying with a 2^3 full factorial design. The studied factors were alginate concentration (1 or 2% (w/v)), glycerol concentration (0 or 1.5% (w/w) alginate) and crosslinking the alginate coating with $CaCl_2$ to form calcium alginate (yes/no). The results showed that a chitosan coating with 1% (w/v) chitosan in 1% (v/v) acetic acid, and 15% (w/w chitosan) glycerol, or 1% (w/v) alginate coating with no glycerol, and no crosslinking showed the best performance in controlling the tested safety and quality parameters. Additionally, a desirability method was used to identify the shelf life of chitosan, alginate and double coated RtE products based on the measured critical quality parameters. Chitosan coated samples showed the best performance with a 3-fold shelf life extension compared to uncoated products.*


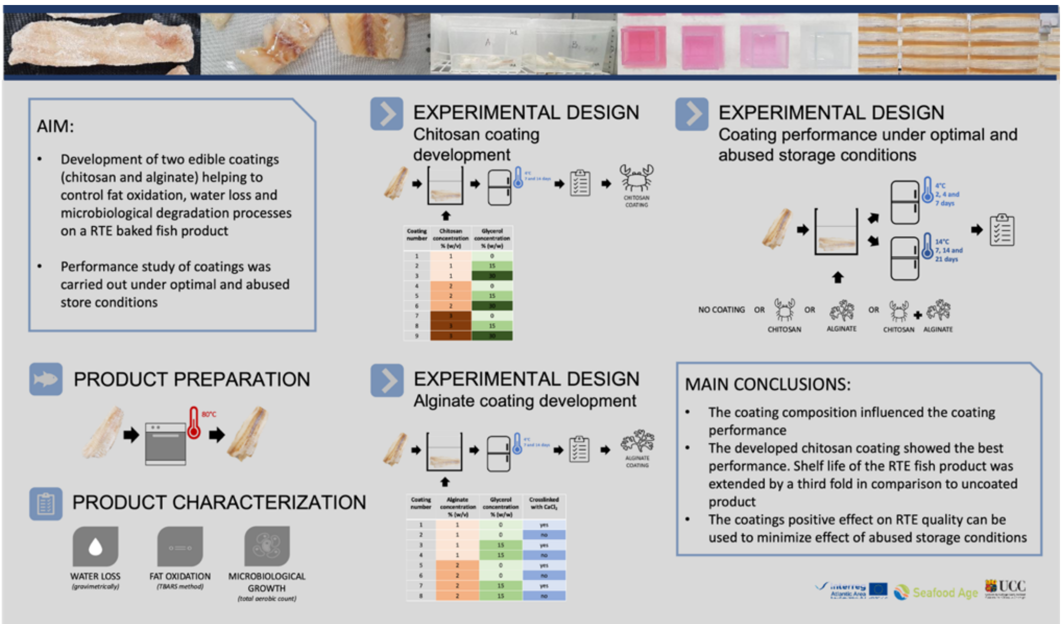


*Figure 5-1: Graphical abstract: Design and development of an edible coating for ready to eat seafood product.*


## Keywords

*Edible coating*
*Chitosan*
*Sodium alginate*
*Food quality and shelf life*
*RtE fish product*

## 5.1 Introduction

Rethinking packaging, for recyclability, circularity and sustainability is required to overcome the current packaging challenges as fossil raw material dependency or waste management. The use of an edible coating is one of the strategies, to improve the sustainability of food packages, which combines the need to use renewable natural biomaterials and reduces packaging waste by being edible or biodegradable. A coating is a thin homogeneous layer formed from a natural film forming material, as carbohydrates, proteins or lipids, on the product surface (Falguera et al., 2011) and acts as a packaging by preventing or controlling deteriorative reactions of the product. Potential renewable natural film forming polysaccharides, sourced from maritime environment are for example, chitosan and alginate. Advantages of using maritime polymeric materials as edible coatings for seafood products are their availability in the production area of the seafood products, and generation of a new market for processing waste products.

Chitosan is a derivative of chitin the second most abundant natural polymer in nature and is mainly produced from shrimp and crab shells, a waste product of the seafood industry with a global waste accumulation of 6 to 8 Mt (Muñoz et al., 2018; Vicente et al., 2022). Chitosan is a suitable material for edible coatings, as it is nontoxic, biodegradable, biocompatible and general recognized as safe by US Food and Drug Administration (Kanatt et al., 2013). Chitosan coatings showed an excellent coating behaviour by forming a transparent, invisible and adhesive layer around food products, and was shown to control microbial growth (Sørbø and Lerfall, 2022; Zarandona et al., 2021) and reduce fat oxidation (Martínez et al., 2018). Variations in chitosan coatings composition and processing factors were reported, e.g., chitosan with different degrees of acetylation from 70 to 95%, chitosan concentrations from 0.5 to 3%, different solvents with concentrations from 1 to 2%, stirring temperature from 23 to 40°C, or addition of glycerol from 0.5 to 75% of w/w chitosan (Arancibia et al., 2015; Bonilla et al., 2019; Carrión-Granda et al., 2016; Ebadi et al., 2019; Jeon et al., 2002; Li et al., 2012; Martínez et al., 2018; Mohan et al., 2012; Sørbø and Lerfall, 2022; Yu et al., 2018; Zarandona et al., 2021). The potential of chitosan coatings is well studied for different fresh fish products (Azaza et al., 2022; Bonilla et al., 2019; Li et al., 2012; Zarandona et al., 2021), but limited studies for ready to eat (RtE) food products, and particularly RtE seafood products (Carrión-Granda et al., 2016; Martínez et al., 2018; Sørbø and Lerfall, 2022) could be found.

Alginate can be found in cell-walls and intercellular regions of most brown algae species which are abundant in many coastlines. Alginates are seen as valuable and sustainable resource, avoiding competition for land use with food production and are a low cost product (Beata Łabowska et al., 2019). Alginate is biodegradable, biocompatibility, nontoxic, has excellent film forming properties and is widely studied as carrier coating material for natural antimicrobial or antioxidant agents for fresh fish products (Bazargani-Gilani, 2017; Bazargani-Gilani and Pajohi-Alamoti, 2020; Heydari et al., 2015; Nie et al., 2018; Sáez et al., 2020; Song et al., 2011; Vital et al., 2018). Alginate coating was reported to control food product degradations processes, as lipid oxidation (Bazargani-Gilani, 2017; Heydari et al., 2015; Nie et al., 2018; Sáez et al., 2020; Song et al., 2011; Vital et al., 2018) and microbiological growth (Bazargani-Gilani, 2017; Heydari et al., 2015; Nie et al., 2018). Studies with alginate coatings applied to RtE seafood products are rare (Martínez et al., 2018) and the effect can depend on the coating composition and development process.

RtE seafood products are in high demand considering the current consumer trends of convenience, healthy, nutritious, mildly preserved foods and products with an enhanced shelf life and controlled product quality (Han et al., 2018). Additionally, considering an upward trend on the growth of elderly population in every country in the world (United Nations, 2019), seafood products can provide an important food source with high nutritional value (Liu and Ralston, 2021). RtE food is defined as food intended for direct human consumption without the need for cooking or other processing preparation steps to eliminate or reduce microorganisms of concern (The Commission of the European Communities, 2005). The shelf life of chilled RtE products is limited, and suitable handling until consumption is crucial to maintain the quality and provide a safe product. The use of hurdle technology, as for example pre-preservation steps, cooled storage and packaging is a common strategy applied to provide a product with an extended shelf life. The application of coatings for RtE seafood products is still limited beside several advantages as enhancing the product safety and quality and being an alternative edible packaging option.

The objective of this study was to develop an edible coating for controlling degradation processes of a RtE baked fish fillet. As the protection functions of a coating are affected by material properties, coating composition and development process, for this study two maritime based coatings, based on chitosan and alginate were developed and optimised. Additionally, the performance of the selected coatings as well as a bi-layer coating, to combine the advantages of both coatings, were compared under optimal and abused storage conditions. The shelf life of the baked fish product with and without coatings were estimated using a desirability analysis to compare the overall performance of the studied coatings.

## 5.2 Materials & methods

### 5.2.1 Chemicals

Acetic acid, calcium chloride anhydrous, chitosan (medium molecular weight), glycerol, peptone buffer, plate count agar, sodium alginate, 1,1,3,3-tetraethoxypropane, thiobarbituric acid and trichloroacetic acid were purchased from Sigma-Aldrich.

### 5.2.2 Experimental design & set up

An overview of the performed experimental designs are illustrated in Table 5-1. A 3^2 full factorial design was used to study the effect of chitosan concentration (1, 2 and 3%), and glycerol concentration (0, 15 and 30% w/w chitosan) on the coating performance to control degradation processes of a baked fish sample stored at 4°C for 14 days. The effect of the composition of an alginate coating was studying with a 2^3 full factorial design. The studied factors were alginate concentration (1 or 2% (w/v)), glycerol concentration (0 or 1.5% (w/w) alginate) and crosslinking the alginate coating with $CaCl_2$ to form calcium alginate (yes/no).

Additionally, the performance of the selected chitosan coating, selected alginate coating, and samples coated with a bilayer coating consisting of a chitosan and alginate layer at optimal storage conditions (4°C) for 21 days and abused storage conditions (14°C) for 7 days were investigated. All samples were placed into a storage container (KIS C, XS) with a lid to control airflow and stored in an incubator (Sanyo) at defined temperature. The functionality of the coatings was assessed by measuring the lipid oxidation, water loss and the microbial total aerobic growth at day 0 and defined storage times (2, 4, 7 days or 7, 14, 21 days for samples stored at 14 and 4°C, respectively). An uncoated product was used as reference. All measurements were at least conducted in triplicates and values presented in this study are average values.

Table 5-1: Overview of experimental designs for a) development of a chitosan coating, b) development of an alginate coating and c) comparing the coating performances of alginate and chitosan coating at optimal and abused storage temperature.

**a)**

| **Factors** | **Levels** | | |
|---|---|---|---|
| Chitosan concentration [% w/v] | 1 | 2 | 3 |
| Glycerol concentration [% w/w chitosan] | 0 | 15 | 30 |

**b)**

| **Factors** | **Levels** | |
|---|---|---|
| Alginate concentration [% w/v] | 1 | 2 |
| Glycerol concentration [% w/w alginate] | 0 | 15 |
| Crosslinking with calcium chloride | yes | no |

**c)**

| **Factor** | **Levels** | |
|---|---|---|
| Storage temperature | 4 | 14 |
| Alginate coating | no | yes |
| Chitosan coating | no | yes |

### 5.2.3 Product preparation

Frozen white fish fillet was purchased from local supermarket. The nutritional composition of the product was 0.8% fat, 17% protein, 0.25% salt, and 0.5% fibre. The frozen fillets were placed on a baking tray and prepared with a mild heating treatment in an oven at 80°C for 70 min. The baked fillets were allowed to cool down to room temperature in a sterile storage box to avoid any cross contamination, before the fillets were cut into pieces of 20 to 25 g. The samples were examined and pieces with a similar thickness was selected for this study. The average water content of the RtE fish product after baking was 75%. Quality characteristics and microbiological contamination of the products were analysed at day 0, which was taken as reference point to analyse the deterioration process during storage.

### 5.2.4 Coating preparation & application

Chitosan film forming solutions (FFS) were prepared by either dissolving 1, 2 or 3% (w/v) chitosan in a 1% (v/v) acidic acid solution. The mixture was stirred with a head stirrer (IKA, Staufen) for 2 h at room temperature. Afterwards, when required 15 or 30% glycerol (w/w chitosan powder) was added and the mixture was stirred for additional 10 min. Alginate FFSs were prepared by dissolving sodium alginate (1 or 2% w/v) in distilled water by stirring for

30 min at 80°C. Afterwards, when required glycerol was added and the mixture was stirred for additional 10 min. The solutions were cooled down to room temperature. Both FFSs were allowed to rest for at least 12 h at 4°C before use.

Samples were coated by immersing the samples into FFS for 30s, followed by a draining step for 5 min. The process was repeated to ensure a complete coating formation on the sample surface. For the calcium-alginate coating the sodium alginate coated samples were immersed into a 0.2 M $CaCl_2$ solution. A double coating was applied by first immersing the sample into chitosan FFS, drained for 5 min and afterwards immersed into the alginate FFS. Samples were allowed to air dry during storage.

### 5.2.5 Characterisation of coating

Apparent viscosity of the FFS was measured using a Haake Rotor viscometer equipped with a coaxial cylinder geometry at 30°C ± 0.1. The shear rate was increased from 0.1 to 1000 $s^{-1}$ over 4 min, held at 1000 $s^{-1}$ for 3 min, after which it was decreased from 1000 to 0.1 $s^{-1}$ over 4 min.

The wet coating load (mg liquid coating/$cm^2$ sample) defined as the coating adhered to the sample after the coating process was calculated by weighting the samples before and after the coating process. Additionally, the surface area of the samples was manually measured with the aid of a ruler and calculated by assuming a rectangular shape of the fish samples.

### 5.2.6 Characterisation of product quality

Thiobarbituric acid reactive substance assay (TBARS) was used to determine the lipid oxidation of samples based on Rosmini et al. (1996). Briefly, 4 g sample was mixed with 10 mL of 10% trichloroacetic acid (w/v). Then, 5 mL of control solution, containing an aliquot of 30 μL of a $10^{-3}$ M 1,1,3,3-tetraethoxypropane or 5 mL of distilled water was added and vortex for 5 min. Freshly prepared 0.001 M thiobarbituric acid solution was added (5 mL) and mixed for 1 min. The reaction mixture was centrifugated at 3500 rpm for 5 min (Universal 16R, Hettich, Tuttlingen, Germany) and the supernatant was filter through a Whatman No. 1 filter. Afterwards the mixture was heated in a water bath (WB14, Memmert, Schwabach, Germany) at 95°C for 35 min and placed for 10 min into a cooled water bath for cooling. The supernatant absorbance was recorded against a blank, prepared as described above without the addition of sample, at 532 nm using a spectrophotometer (Libra S22, Biochrom, Cambridge, England). Standard solutions were prepared using 1,1,3,3-tetraethoxypropane to quantify the malondialdehyde (MDA) content, and results were expressed as mg MDA equ./Kg sample.

The weight of RtE fish samples was measured using a precision balance (Sartorius, Germany). The weight loss ($W_L$) in % was calculated based on the initial ($W_0$) and weight after time i ($W_i$) of the product (eq. 5.2-1). For coated samples, the initial weight was set to the product weight after the coating was applied.

$$W_L = [(W_0 - W_i)/W_0] \cdot 100 \qquad \text{eq. 5.2-1}$$

The total aerobic count (TAC) of the product was analysed by aseptically transferring around 10 - 15 g of sample to a sterile stomacher bag (Sewards LTD, UK) and adding peptone buffer to achieve a 10-fold dilution. The sample was mashed, and hand shacked for 2 min. Appropriated dilutions were prepared of the resulting suspension with serially diluting in peptone buffer. For the total microbial count, the pour plate method with a plate count agar (Sigma Aldrich) was applied. The agar plates were incubated at 30°C for 72 h (Zarandona et al., 2021). Results were expressed as $\log_{10}$ colony forming units per gram sample (log CFU/g).

### 5.2.7 Statistical analysis

Analysis of Variance (ANOVA) and post-hoc analysis using Tukey's test at 95% confidence level were used to assess significant differences between the tested coating formulations. Full factorial designs were analysed with a significance level of 5%. Additionally, the desirability was calculated as presented by Myers et al. (2016), as follows:

$$D = {d_1}^{w_1} \cdot {d_2}^{w_2} \cdot \ldots \cdot {d_n}^{w_n} \qquad \text{eq. 5.2-2}$$

Where, D is the total desirability, d is the desirability score of each property studied and w is the weight conferred to each property. Each property was weighted equally. The desirability score is determined using a linear transformation to the values from the Pareto Front, scaling the objectives between 0 and 1 from worst to best quality. The optimal combination of the tested coatings was validated qualitatively using characteristic quality and safety properties describing a lower level of product degradation, namely low lipid oxidation, water loss and TAC during storage. In order to avoid the elimination of some results that could be important to explain some observed phenomena, results with $0.05 < p < 0.1$ were considered marginally significant, and also considered to the analysis (Montgomery, 2013). The statistical analysis was performed by using Statistica software for Windows v. 7.1 (Tulsa, USA).

## 5.3 Results & Discussion

The coating material, coating formulation, and addition of additives as plasticisers can influence physical and protection properties of edible coatings. Therefore, the first step was to identify a suitable coating formulation for a RtE seafood product based on chitosan and alginate. The coating performance was measured by studying critical quality parameters of RtE baked fish fillet stored over 14 days at 4°C. Lipid oxidation is one of the main factors limiting the shelf life of muscle foods due to present of higher amounts unsaturated fatty acids, leading to off-flavour, colour and odour development and contribute to texture deterioration (Kanatt et al., 2013; Mendes et al., 2008). Therefore, lipid oxidation measured by the malondialdehyde content developed during storage indicating the degree of secondary lipid oxidation (Anacleto et al., 2011), was used as the first critical quality parameter. Controlling weight loss during storage was an additional important factor for quantifying the shelf life. Weight loss during storage can alter the product appearance and reduces consumer acceptance. The third critical quality parameter was the determination of the total aerobic count to assess the overall safety and quality of the RtE fish product. RtE food products which received only a mild heat treatment and are distributed at cold temperatures are likely to spoil due to microbial action

(Gram and Huss, 1996). As they are intended for direct consumption without any heat treatment prior to consumption, the safety of the product needs to be assured.

Table 5-2: Measured quality parameters (lipid oxidation, water loss and total aerobic count) of samples coated with single alginate, single chitosan and bilayer chitosan-alginate coating stored at a) 14°C over 7 days and b) 4°C over 21 days.

| **Lipid oxidation [mg MDA eq./Kg sample]** | | | | |
|---|---|---|---|---|
| **a)** | **day 0** | **day 2** | **day 4** | **day 7** |
| Control | 0.16 ±0.03 a ,A | 0.54 ±0.53 a ,A | 0.66 ±0.27 a ,A,B | 1.90 ±0.82 a ,B |
| Alginate | 0.16 ±0.03 a ,A | 0.79 ±0.22 a ,A | 1.56 ±0.91 a ,A | 1.10 ±0.10 a ,B |
| Chitosan | 0.16 ±0.03 a ,A | 0.28 ±0.17 a ,A,B | 0.50 ±0.12 a ,B | 1.81 ±0.38 a ,A,B |
| Chitosan + Alginate | 0.16 ±0.03 a ,A | 0.54 ±0.07 a ,B | 1.15 ±0.11 a ,C | 1.50 ±0.03 a ,D |
| **b)** | **day 0** | **day 7** | **day 14** | **day 21** |
| Control | 0.16 ±0.03 a ,A | 1.10 ±0.62 a ,A | 1.17 ±0.39 a ,A | 4.38 ±1.20 a ,B |
| Alginate | 0.16 ±0.03 a ,A | 1.55 ±0.17 a ,A,B | 2.21 ±0.50 b ,A,B,C | 4.36 ±0.96 a ,C |
| Chitosan | 0.16 ±0.03 a ,A | 1.05 ±0.38 a ,B | 2.40 ±0.42 a,b ,B | 4.22 ±1.09 a ,C |
| Chitosan + Alginate | 0.16 ±0.03 a ,A | 1.48 ±0.66 a ,A,B | 1.75 ±0.42 a,b ,A,B | 2.07 ±0.89 a ,A,B |
| **Water loss [%]** | | | | |
| **a)** | | **day 2** | **day 4** | **day 7** |
| Control | | 1.16 ±0.35 a,A | 2.70 ±0.44 a,B | 3.70 ±0.32 a,B |
| Alginate | | 0.96 ±0.17 a,A | 1.78 ±0.36 a,A,B | 2.87 ±0.54 a,B |
| Chitosan | | 1.02 ±0.31 a,A | 2.32 ±0.66 a,A | 3.13 ±1.00 a,A |
| Chitosan + Alginate | | 0.86 ±0.06 a,A | 1.59 ±0.11 a,A | 3.26 ±0.52 a,B |
| **b)** | | **day 7** | **day 14** | **day 21** |
| Control | | 1.50 ±0.49 a,A | 3.65 ±0.98 a,A | 9.96 ±4.02 a,A |
| Alginate | | 1.77 ±0.25 a,A | 2.63 ±0.30 a,A,B | 4.88 ±0.76 a,b,B |
| Chitosan | | 1.46 ±0.30 a,A | 1.99 ±0.28 a,A | 3.31 ±0.50 a,b,B |
| Chitosan + Alginate | | 1.52 ±0.68 a,A | 2.28 ±0.51 a,A | 2.94 ±0.71 b,A |
| **Total aerobic count [log CFU/g sample]** | | | | |
| **a)** | **day 0** | **day 2** | **day 4** | **day 7** |
| Control | < 1.06* ±0.10 a,A | 4.86 ±0.74 a,B | 7.19 ±0.58 a,b,C | 9.11 ±0.45 a,D |
| Alginate | < 1.06* ±0.10 a,A | 5.85 ±0.05 a,B | 7.70 ±0.58 b,C | 10.06 ±0.06 a,D |
| Chitosan | < 1.06* ±0.10 a,A | < 1.39* ±0.67 b,A | < 0.94* ±0.34 c,A | 5.07 ±2.56 b,A |
| Chitosan + Alginate | < 1.06* ±0.10 a,A | 2.79* ±0.74 b,A | 6.00 ±0.90 a ,B | 7.21* ±1.05 a,b,B |
| **b)** | **day 0** | **day 7** | **day 14** | **day 21** |
| Control | < 1.06* ±0.10 a,A | < 2.86* ±0.84 a,A | 7.63 ±0.75 a,B | 9.10 ±1.04 a,B |
| Alginate | < 1.06* ±0.10 a,A | < 2.13* ±0.23 a,b,B | > 8.48* ±0.00 a,C | 10.35 ±0.41 a,D |
| Chitosan | < 1.06* ±0.10 a,A | < 1.00* ±0.00 b,A | < 1.00* ±0.00 b,A | < 1.79* ±2.29 b,A |
| Chitosan + Alginate | < 1.06* ±0.10 a,A | < 1.40* ±0.70 a,b,A | 4.85* ±2.81 a,A | 10.06 ±0.00 a,B |

*Statistical analyses were conducted for samples at 14°C (a) and 4°C (b) separately. For each storage temperature, results on the same column followed by the same small letter and results in the same row followed by the same capital letter were not statistically different ($p < 0.05$) according to a Tukey's - HSD test. *Indicates an estimated value due at least one colony count of the triplicates below 30 or above 300. >/< indicates the direction to the potential real TAC value.

### 5.3.1 Development of a chitosan coating

The overall appearance of the coating formulations and coating properties were investigated visually. The chitosan FFS had a clear, slightly yellowish appearance. The viscosity was highly depended on the used chitosan concentration with higher viscosity with higher concentrations. When applied to the RtE fish samples, using the coating dipping method, all tested chitosan coatings adhered to the sample surface and coated samples had a wet, shiny appearance. After air drying during storage, no differences between coated and uncoated samples could be identified, indicating that all tested coating formulations are invisible.

Samples coated with 1% chitosan solution with a viscosity of 86 mPas, had a wet coating load of around 18.6 mg liquid coating/cm$^2$. It could be observed that a higher viscosity increased the amount of coating adhering to the sample. A relation between the coating thickness and FFS properties as viscosity, density and draining time was also reported in literature (Cisneros-Zevallos and Krochta, 2003). The used acidic acid solvent for dissolving the chitosan had a pH value of around 3.1, and the pH of the final chitosan FFSs were around 4.4. Previous studies showed that the low pH of the solvent alone did not affect the product quality (Kanatt et al., 2013).

Overall, lipid oxidation increased during storage for all samples, but even results after 14 days of storage with an average MDA value of 1.5 mg MDA eq./Kg sample were low. They can be classified into the category of 'perfect quality material' based on threshold criteria suggested by Socaciu et al. (2018), which were values < 3 mg MDA eq./Kg for perfect quality material, 3 ≤ 5 mg MDA eq./Kg for good quality material, and 5 < 8 mg MDA eq./Kg suitable for human consumption. The statistical analysis showed that there were no significant differences between the tested coating formulations, as well as no significant differences between 7 and 14 days of storage in regard to lipid oxidation (Figure 5-2ai). Variations in the selected design variables were in a similar range of magnitude as an identified noise factor, possible trace back to natural product variations, and therefore it was not possible to distinguish between these two. In conclusion, no effect of the tested chitosan coating composition on controlling lipid oxidation of RtE baked fish product could be observed within this experimental set up. These findings are in disagreement with results from Wu et al. (2000) and (Martínez et al., 2018). Both observed an effect of the tested chitosan coating on retarding oxidation when applied to a precooked beef patty or smoked sea bass fillet, respectively. A possible explanation for the different results, could be that the sample in this study are less prolonged to lipid oxidation or the effect was not visible under the selected experimental conditions.

All samples lost water during storage, as the driving force for the water gradient was in the direction of the surrounding air with a relative humidity of around 75%. Significant differences in the water loss between the tested coatings could be observed. The results of the full factorial design after 14 days of storage showed that the variation in the data set can be explained by the factors chitosan concentration, glycerol concentration and an interactive effect between these two factors (Figure 5-2aii). The lowest water loss could be seen when a coating with a low chitosan concentration in combination with a medium glycerol concentration was applied. When the chitosan concentration is increasing the water loss is increased, whereby this effect can be reduced with an increase of glycerol up to 15% (w/w chitosan) in the coating. With higher glycerol concentrations (30% w/w chitosan), the combination with a higher chitosan concentration reduces the water loss the most. Nevertheless, the lowest obtained value with 30% glycerol concentration was still higher than values with lower glycerol and chitosan concentrations. A similar effect of a chitosan coating, with a 2% (w/v) chitosan coating with 25% (w/w chitosan) glycerol, on the water loss of a precooked beef patty was reported by Wu et al. (2000).

All tested chitosan coatings showed a significant effect on the microbiological growth. The TAC of the coated samples was below the detection limit after 14 days of storage at 4°C, with one exception of samples coated with 1% chitosan solution with 30% (w/w chitosan) glycerol addition with a TAC below 2 log CFU/g after 14 days (Figure 5-2aiii). The control samples showed a TAC of around 4 log CFU/g after 14 days stored at 4°C. All tested chitosan coating formulations showed a similar effect on controlling the TAC of the RtE fish samples. Therefore, it can be concluded that the presence of glycerol in the solutions does not influence the antimicrobial effect of the chitosan and a 1% chitosan concentration showed the same effect as the highest tested chitosan concentration of 3%. The reported antimicrobial effect in literature of chitosan coatings can be confirmed with the observed results. The effect of a chitosan coating on ready-to-cook meat products packed in LDPE pouch showed that a chitosan coating can lead to retention of the good quality characteristics, improvement of microbiological safety and extension of shelf life during storage (Kanatt et al., 2013). The application of a chitosan coating with and without the incorporation of essential oils applied to RtE peeled shrimp tails at refrigerated storage packed under MAP conditions was studied by Carrión-Granda et al. (2016), showing that chitosan coatings could be used as a promising emerging technology in order to maintain the microbiological quality. Martínez et al. (2018) studied the effect of a chitosan coating in combination with a vacuum package, inter alia, as a conservative method for smoked sea bass fillets and found that the coating inhibited microbiological growth. An increasing microbial stability and improve colour stability in RtE maki sushi when coated with chitosan and packed under low-$CO_2$ MAP conditions, was reported by Sørbø and Lerfall (2022).

Summarizing, based on the findings of all three quality parameters, an FFS with 1% chitosan with 15% (w/w chitosan) glycerol showed the best performance in controlling the tested degradation processes during cold storage, mainly based on the effect of the coating to reduce water loss and control microbiological growth.

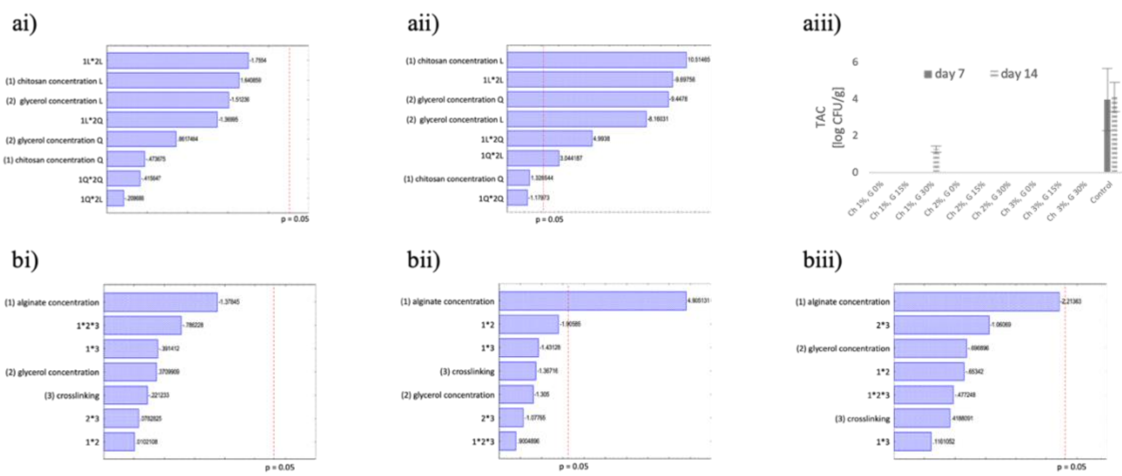


*Figure 5-2: Results of the screening experiment of (a) chitosan and (b) alginate coatings on the quality parameters (i) lipid oxidation, (ii) water loss and (iii) total aerobic microbiological count stored at 4°C under controlled air circulation over 14 days.*

### 5.3.2 Development of an alginate coating

The overall appearance of the coating formulations and coating properties were investigated. The alginate FFSs had a clear appearance and a similar viscosity of around 9.43 mPas for all tested compositions. When applied to the RtE fish samples, using the coating dipping method, FFSs adhered to the sample surface. When the alginate coating was cross-linked with calcium chloride a gel was formed at the product surface. Coated samples had a wet, shiny appearance. After drying during storage, no differences between coated and uncoated samples could be identified, indicating that the coatings were invisible. The wet coating load of the alginate coatings was determined at around 10 mg liquid coating/$cm^2$. Confirming that a lower viscosity decreases the coating adhering to the product, when compared to the higher viscosity of the FFS of the chitosan coatings (Kanatt et al., 2013). The results of the statistical analysis of the effect of the alginate concentration, glycerol concentration and crosslinking the alginate with $CaCl_2$ on the lipid oxidation, water loss and TAC of the baked fish sample are shown in Figure 5-2b.

An increase in MDA value was observed, indicating fat oxidation of all tested alginate coated samples, with the highest value of around 2 mg MDA eq./Kg for samples stored for 14 days. Under the assessed conditions, no tested factor on the alginate coating showed that the lipid oxidation was significant (Figure 5-2bi) and no significant differences to control samples were found. A higher standard deviation was found within each coating formulation, which could be related to natural variations within the tested baked fish samples. The tested alginate concentrations, glycerol addition or crosslinking with $CaCl_2$ did not influence the performance of the alginate coating, in providing an oxygen barrier to reduce lipid oxidation. In several studies, alginate coated fish products showed a reduction in lipid oxidation during cold storage. Martínez et al. (2018) studied the effect of an alginate coating in combination with a vacuum package, inter alia, as a conservative method for smoked sea bass fillets, and concluded that an alginate coating protected the fillets against oxidation. Other studies with fresh fish fillet showed that with longer storage time the use of an alginate coating can reduce lipid oxidation (Bazargani-Gilani, 2017; Sáez et al., 2020). Nevertheless, in these studies the lipid oxidation of the tested fish products was of a higher level, than in the present study. Therefore, the minimal detected effect can be potentially related to the low fat content of the RtE product, overall low lipid oxidation of the product during the tested storage conditions or were not yet visible under the selected storage conditions of 4°C up to 14 days. It could potentially be recommended to investigate the effect of the alginate coating on the lipid oxidation at higher storage temperature to increase the lipid oxidation reaction rate or for longer storage times. In general, the product degradation regarding the lipid oxidation could be of higher importance for high fat content RtE food products.

The statistical results of the effect of the tested alginate coatings on the moisture loss are presented in Figure 5-2bii. The moisture loss increased with longer storage times, whereby the moisture loss rate depends on the coating formulation. The results of the full factorial design showed that the factor alginate concentration was significant after 14 days of storage, emphasizing the importance of a lower alginate concentration to decrease the water loss. For shorter storage times also the interactive effect between alginate concentration and glycerol concentration was significant. For storage times up to 14 days the interactive effect was only marginally significant. When the alginate concentration was increased, the glycerol concentration needed to be increased for a better control of the water loss during cold storage. The third tested factor crosslinking of the alginate coating with $CaCl_2$ was not significant. Based on the effect of the coatings on the water loss a 1% alginate concentration, with no glycerol and not crosslinking was favoured for a RtE seafood product.

The statistical results of the TAC are shown in Figure 5-2biii. The microbiological growth increased during storage, after 7 days the average TAC was around 4 log CFU/g sample and after 14 days around 7 log CFU/g sample. The results are within the same range as the control sample without any coating. Statistical analysis of the results showed no significant differences between the tested alginate coating formulations after 7 and 14 days stored at 4°C. After 14 days of storage only a marginal effect of the alginate concentration was observed. Indicating that an alginate concentration of 2% was marginally significant for reducing the TAC. Different findings of the effect of alginate coating on microbiological growth were presented in literature. Some studies reported a reduction of microbiological growth for samples coated with sodium alginate due to the ability of the coating to act as a barrier against oxygen transfer (Nie et al., 2018), and the effect could be increased by the used of additives (Bazargani-Gilani, 2017; Heydari et al., 2015). Nevertheless, other researchers reported no reduction in microbial growth (Martínez et al., 2018; Sáez et al., 2020; Shankar et al., 2019).

In summary, considering the findings for all the tested quality parameters, a coating formulation with 1% alginate, without glycerol and no crosslinking was recommended to enhance the product quality. Nevertheless, the alginate coating showed good film forming properties but the effect of the alginate coating on the lipid oxidation, water loss and TAC on RtE baked fish product is limited and could be possible enhanced by the inclusion of natural additives as antioxidants or antimicrobials.

### 5.3.3 Comparing the performance of a chitosan, alginate, and bilayer coating under optimal and abused storage conditions

A follow up experiment was conducted to further investigate and compare the properties of the selected chitosan and alginate coatings. A 1% chitosan solution with 15% glycerol (w/w chitosan) and 1% alginate coating were previously identified to be the most suitable for the RtE fish product under study. Additionally, the potential of combining the individual coating properties of the chitosan and alginate coating was studied by applying a bilayer coating to the RtE baked fish fillet. A full factorial experimental design was used to study the effect of the coatings at optimal storage conditions at 4°C and abusive conditions of 14°C, over 21 days and 7 days, respectively (Table 5-2). The effect of the storage temperature was investigated after 7 days of storage.

All tested samples, including the chitosan coated, alginate coated, bi-layer coated samples and an untreated sample as control, showed a significant increase in MDA value over the tested storage time, indicating lipid oxidation. Though, the highest value measured over the tested storage conditions, with 4.4 mg MDA eq./Kg for uncoated and alginate coated samples stored at 4°C for 21 days, was still in the range of a good quality material based on the threshold criteria suggested by Socaciu et al. (2018). Results showed that even for longer storage times, up to 21 days for samples stored at 4°C and up to 7 days for samples stored at 14°C, the lipid oxidation was not a critical quality parameter for the product under study. The statistical analysis showed that there were no significant differences between the control, chitosan, alginate or a double coating at each sampling day regarding lipid oxidation (Table 5-2). When studying the effect of the storage temperature, a significant interactive effect between the storage temperature and chitosan coating could be observed (Figure 5-3). In general, as expected a slightly higher increase of lipid oxidation for samples stored at 14°C compared to samples stored at 4°C were seen. At higher storage temperatures, samples coated with chitosan showed a lower lipid oxidation. These findings are in accordance with other studies reporting no effect for short storage times and a slight effect for longer storage times of chitosan coatings on the lipid oxidation of RtE meat products. Wu et al. (2000) reported that chitosan coating on precooked beef patties stored for 3 days at 4°C, showed no significant effect of the chitosan coating reducing TBARS, when compared to an uncoated, unpacked sample. Kanatt et al. (2013) reported that chitosan coated ready-to-cook meat products displayed a longer shelf life compared to uncoated samples as the tested chitosan coating slightly retarded lipid oxidation in all the meat products during storage. No effect of the alginate coating regarding the storage temperature could be identified. Nevertheless, a bi-layer coating showed a reduced lipid oxidation at higher storage temperature. Most likely, due to the positive effect of the chitosan layer of the bi-layer coating.

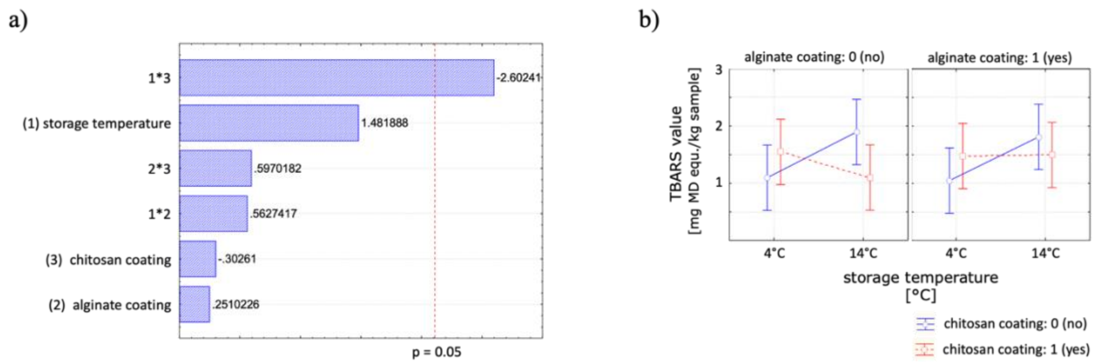


*Figure 5-3: Statistical analysis of the effect of the chitosan coated, alginate coating and storage temperature on the lipid oxidation. a) pareto chart and b) marginal plot with a confident limit of 95% at day 7.*

The effect of chitosan, alginate, and a bilayer coating, compared to an uncoated control sample, on the product water loss under optimal storage temperature of 4°C and abusive storage temperature of 14°C were studied and the results are presented in Table 5-2. Overall, for all the studied scenarios a decrease in water loss was observed. The effect of the coating treatments in regard to the weight loss was studied at 14°C (Figure 5-4i) and 4°C (Figure 5-4ii). For samples stored at 14°C only the alginate coating was significant with a slight reduction in water loss over time. The chitosan coating, the interactive effect between factors 'storage time and chitosan coating' and the interactive effect between the factors 'alginate coating and storage time' was significant for samples stored under cooled storage. For longer storage times, the applied coating reduced the water loss. When studying the storage temperature effect after 7 days, the storage temperature was the only significant parameter with a high effect on weight loss. The average moisture loss after 7 days stored at 14°C was 3.2% and at 4°C was 1.6%. In conclusion, the tested coating formulations were effective on reducing weight loss of the samples during storage. Samples, either coated with a single chitosan coating or a double coating had a weight loss of around 3.3 and 3% after 21 days of storage, compared to the control sample with a 10% weight loss. With the double coating it was not possible to combine the effect of the chitosan and the single alginate coating.

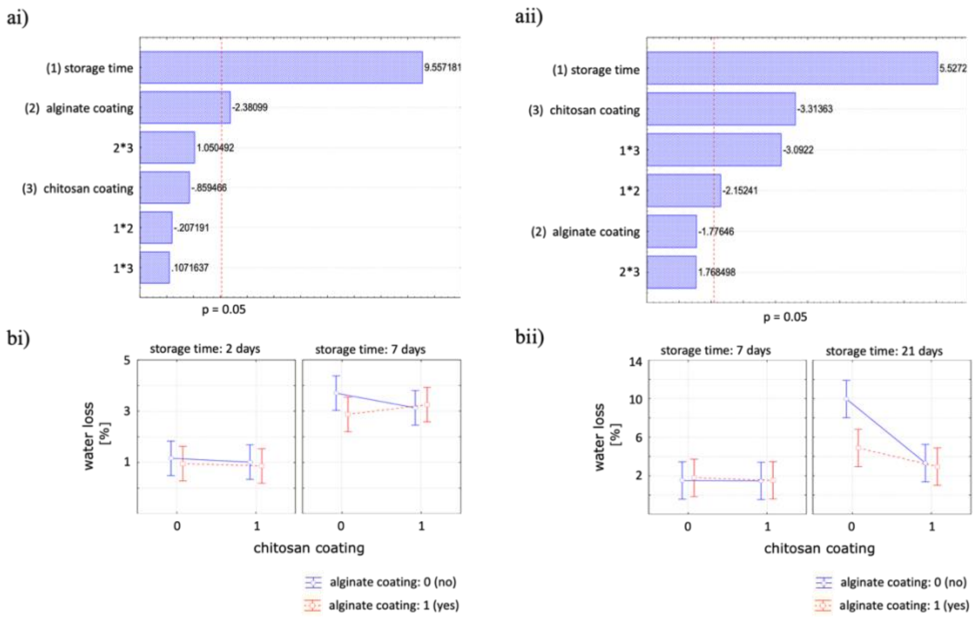


*Figure 5-4: Statistical analysis of the effect of chitosan coating, alginate coating and storage time on the moisture loss at (i) 14°C and (ii) 4°C. a) pareto charts, b) plot of marginal means with a confident limit of 95%.*

In this study the TAC was used to assess the overall safety and quality of the RtE fish product. The observed results of the TAC of the RtE fish samples are presented in Table 5-2. The initial TAC of freshly baked uncoated RtE fish product was in the range of 1 log CFU/g sample. In general, the TAC increases with storage time, though the growth rate can be significantly influenced by the storage temperature and by applying a coating to the product. The microbiological growth rate was reduced with decreasing temperature, shown by reaching the maximal allowed microbiological load of 7 log CFU/g sample at 14°C after 4 days and at 4°C after 14 days for uncoated control samples. After the storage temperature, the chitosan coating was the second significant parameter (Figure 5-5). The results showed that the microbial growth during storage can be reduced when a chitosan coating was applied. The chitosan coated samples were below the critical value of 7 log CFU/g during the tested storage times of 7 days at 14°C and 21 days at 4°C. The alginate coating showed a negative effect on the microbial growth. With longer storage times, samples coated with alginate resulted in a higher CFU values compared to the control samples. The samples coated with chitosan and alginate showed a reduced microbial growth when compared to the uncoated samples, nevertheless the former showed a higher CFU count than using a single chitosan coating. These findings are in accordance with other chitosan coating studies, reporting an antimicrobial effect of chitosan (Bonilla et al., 2019; Ebadi et al., 2019; Martínez et al., 2018; Zarandona et al., 2021). The effect of chitosan on microbial stability in RtE maki sushi under abused storage conditions was also reported by Sørbø and Lerfall (2022). Emphasizing, an increased safety of RtE products during optimal cooled chain, but also when exposed to abused conditions. Therefore, it is recommended to further test the ability of a chitosan coating under simulated real shelf life conditions.

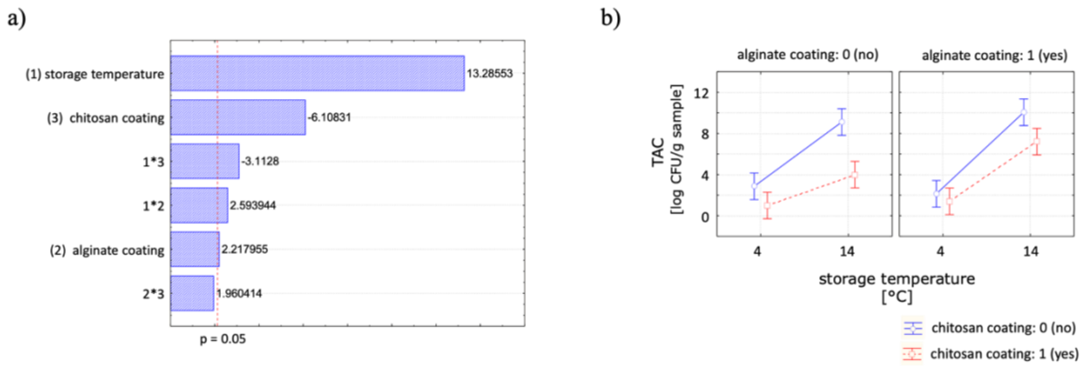


*Figure 5-5: Statistical analysis of the effect of the chitosan coating, alginate coating and storage temperature on total aerobic count. a) pareto chart and b) marginal means plot with a confident limit of 95% at day 7.*

### 5.3.4 Effect of the edible coatings on the product shelf life

A multi-response analysis was used to determine the shelf life of the uncoated and coated products. The cut of points for the shelf life estimation was 7 log CFU/g for TAC (Food Safety Authority of Ireland, 2020), 10% water loss and lipid oxidation value of below 8 mg MDA eq./Kg sample (Socaciu et al., 2018). The results of the total desirability score of the RtE fish products, weighting each quality parameter equally without the ability to comprise for another quality parameter, are presented in Table 5-3. Overall, the highest score at each storage time and temperature was reached with a chitosan coating providing an acceptable quality score until 21 and 7 days stored at 4 and 14°C, respectively. The second best coating was a bi-layer coating, though with a reduced quality and consequently a lower maximal shelf life. The used alginate coating only provides marginal advantage for prolonging the quality. The most critical drawback of the alginate coating under the tested three quality factors was the negative effect of the coating on the aerobic microbial growth.

Table 5-3: Overall desirability score for the quality of tested RtE baked fish samples coated and uncoated.

| Storage temp. | Coating | day 0 | day 2 | day 4 | day 7 | day 14 | day 21 | day 28 |
|---|---|---|---|---|---|---|---|---|
| 4°C | Alginate | 1 | | | 0.84 | 0 | 0 | 0 |
| | Chitosan | 1 | | | 0.89 | 0.84 | 0.65 | 0 |
| | Chitosan-Alginate | 1 | | | 0.87 | 0.61 | 0 | 0 |
| | Control | 1 | | | 0.80 | 0 | 0 | 0 |
| 14°C | Alginate | 1 | 0.56 | 0 | 0 | 0 | 0 | 0 |
| | Chitosan | 1 | 0.92 | 0.86 | 0.67 | 0 | 0 | 0 |
| | Chitosan-Alginate | 1 | 0.85 | 0.50 | 0 | 0 | 0 | 0 |
| | Control | 1 | 0.67 | 0 | 0 | 0 | 0 | 0 |

| 1 | 0.8 | 0.6 | 0.4 | 0.2 | 0 |
|---|---|---|---|---|---|

colour scale from 1 to 0

## 5.4 Conclusion

Edible coatings are alternative eco-packaging systems based on biobased materials and can provide packaging functions by controlling degradation processes of food products, resulting in advantages in terms of food safety and quality. In this study coatings based on chitosan and alginate were tested for controlling fat oxidation, water loss and microbial growth of a RtE baked fish product. First, the effect of changes in the coating composition was studied and secondly the effect of the developed coatings on the product shelf life was analysed under optimal and abused storage conditions. The results showed that the selected coating composition can influence the coating performance. A chitosan coating composition with 1% (w/v) chitosan dissolved in 1% (v/v) acetic acid solution with 15% (w/w chitosan) glycerol, and a 1% (w/v) sodium alginate without glycerol coating were the most effective coating solutions for the RtE seafood product of this study. Both chitosan and alginate coating applied as a single or bi-layer coating formed invisible adherent layers around the product. The tested alginate coatings only showed a limited effect on the tested quality parameters of the RtE seafood product. Whereas the chitosan coating presented a significant reduction in microbial growth during storage at optimal and abused temperatures, as well as reduction in weight loss. A shelf life extension of chitosan coated samples compared to uncoated control sample stored at optimal temperature of 4°C could be achieved from 7 to 21 days, and under abused conditions from 2 to 7 days. An effect of the tested coatings both at optimal and abused storage conditions could be seen, and therefore it is recommended using an edible coating as a safety measure against temperature abuse during the food supply chain. These results suggest that a chitosan coating is an effective packaging strategy to improve the safety and quality of a RtE seafood product during cold storage as well as under abused storage conditions.

## 5.5 Acknowledgments

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Chapter SIX

# Combined effect of plasma treatment and equilibrium modified atmosphere packaging on safety and quality of cherry tomatoes

## Abstract

*Combination of novel technologies for post-harvest processing can have advantages in terms of food safety and quality to reduce food waste. In this work the combination of cold plasma treatment and an equilibrium modified atmosphere packaging (EMAP) for cherry tomatoes was investigated under controlled temperature (10 and 20°C) throughout storage. Statistical analysis of the results showed that plasma treatment did not negatively affect the quality of the product. A higher microbiological growth was detected within the packed cherry tomatoes, while a plasma treatment before packing reduced growth of microorganism during the early days of storage. Package was important to prevent weight loss and changes in total soluble solid content over 14 days of storage. The combination of cold plasma, temperature and EMAP design ensured the quality retention of cherry tomatoes, which suggests that they have potential to be used as hurdle technology.*


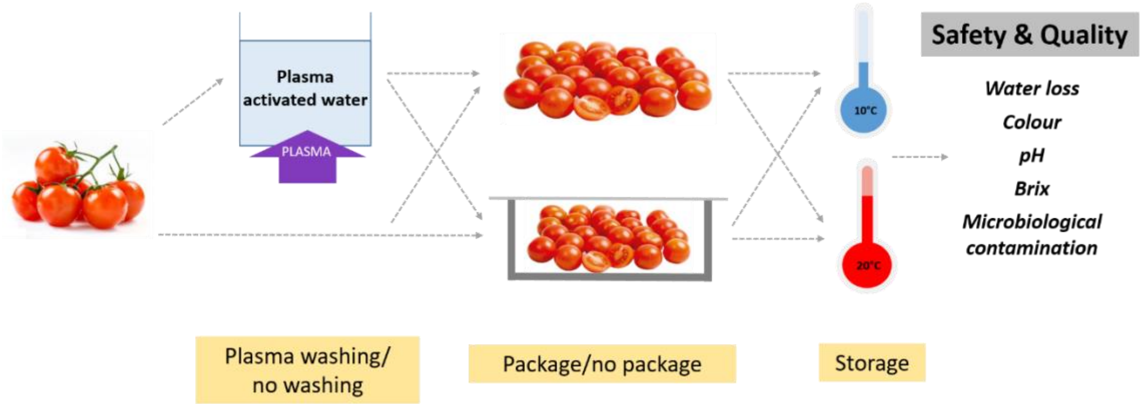


*Figure 6-1: Graphical Abstract – Combined effect of plasma treatment and equilibrium modified atmosphere packaging on safety and quality of cherry tomatoes.*


## Keywords

*Equilibrium modified atmosphere packaging*
*Cold plasma washing treatment*
*Food quality and safety*

## 6.1 Introduction

One-third of the food intended for human consumption was estimated to be lost or wasted globally over the total supply chain from harvest up to the household level (FAO, 2011). A recent estimate presents a food loss index of 14% including the food loss of the total food produced in the world from farm to retailer level, excluding the retail (FAO, 2019). Food waste is a significant problem for the environment in terms of inefficient use of resources, economical as well as social and moral implications relating to food security (Mattsson et al., 2018; Stuart, 2009). Addressing the food waste and losses over the food supply chain is an important step to reach the aim of feeding the world population at a sustainable level (Eriksson et al., 2017). Ensuring that the produce reaches the final consumer in full quality, safe and appealing, is a crucial step to reduce food waste within the supply chain and simultaneously increasing sustainability (Kaipia et al., 2013).

Vegetables and fruits are highly perishable commodities and have a short life span. The right handling of fruits and vegetables during harvesting, transportation, storage and in retailer stores are crucial to provide the consumer with a safe and high quality product. An average supply chain of fruits can be estimated as around two weeks, or several weeks depending on the country of origin (Barrett and Lloyd, 2012). This indicates the need for a full understanding of how to provide a maximum shelf life to different fruits and vegetables. Appropriate handling, pre-processing, and packaging can contribute to the waste reduction by guaranteeing a high quality, safe produce and prolonging the shelf life over the supply chain.

Tomatoes were identified as one of seven hotspot categories which contributes to the majority of the in-store waste produce at retailer level based on wasted mass, economic costs and climate impact (Mattsson et al., 2018). Tomatoes are climacteric fruits and therefore the shelf life is directly related to the respiration rate (Sousa et al., 2017). Main factors to decrease the natural metabolism are storage temperature and gas composition during storage. For a high quality product, the microbiological contamination levels of raw produce before packaging have to be as low as possible. Optimal storage temperature for tomatoes is between 8 and 12˚C and the ideal headspace oxygen concentration for cherry tomatoes is between 3 and 5% (Tumwesigye et al., 2017). Cherry tomatoes are frequently consumed raw; therefore, the safety and quality are extremely important during storage.

An appropriate packaging is an important tool to provide a shelf life extension of fruit and vegetables (Mahajan et al., 2017). Food packaging is designed to keep produce’s quality and safety, to provide protection against light, oxygen, temperature, moisture, and mechanical damage to the product. It works also as a communication device between producer and consumer (Subramaniam, 2016). Equilibrium modified atmosphere packaging (EMAP) is designed to generate naturally an equilibrium gas atmosphere within the package by controlling the respiration rate of the produce. EMAP design is reached by the mathematical integration of physiological characteristics of the produce, packaging material properties and optimum equilibrium atmospheric conditions for the given produce (Belay et al., 2016).

A sanitizing step with chemicals as a pre-treatment before applying a package to fruits and vegetables is common to reduce the microbiological contamination and increase safety (Misra et al., 2014). The use of chemicals such as chlorine is a safety concern and is strictly regulated or even prohibited in a number of European countries (Misra et al., 2014). Different alternative treatment methods for the inactivation of microorganisms without affecting the quality of fruits and vegetables are necessary. Cold plasma, is a novel technology under investigation for replacing chemical washing steps through the generation of plasma activated water for sanitizing product surfaces as well as for decontamination of used wash water for reuse (Chizoba Ekezie et al., 2017; Misra and Roopesh, 2019; Pan et al., 2019; Patange et al., 2019). It is a non-thermal processing method and can be produced at low levels of pressure and power. Cold plasma has potential in the food area for the decontamination of fruits and vegetables without significantly influencing the appearance of the product and competitive with existing industrial sanitization processes (Andrasch et al., 2017). The activated species generated by electron collision play a key role in the microbial plasma inactivation process (Mai-Prochnow et al., 2014)

In previous studies, hurdle technology to prevent microbiological growth, preserve quality and prolong the shelf life has been investigated by combining a washing pre-treatment and modified atmospheric packaging technology in fruits and vegetables (e.g. cherry tomatoes) during controlled storage conditions (Choi et al., 2015). Cold storage and package are the most common preservation methods used for preventing quality loss and inhibit microbiological growth; however, these methodologies are not sufficient to decrease microbiological contamination. Therefore, it is of high interest to combine an optimal designed package with a novel washing treatment to secure a safe product and prevent physical, chemical and microbiological deterioration or contamination in fruits and vegetables (e.g., cherry tomatoes) throughout the distribution chain.

Hence, the aim of the study was to investigate the combined effects of a plasma-washing treatment and an equilibrium modified atmosphere packaging design on the cherry tomatoes quality characteristics during storage at 10°C and 20°C for improving microbiological quality and maintaining quality characteristics during storage. The quality of cherry tomatoes was monitored by measuring the weight loss, soluble solid content, colour, and pH of the tomatoes, as well as detecting the aerobic bacteria, mould, and yeasts load.

## 6.2 Materials & Methods

### 6.2.1 Product Preparation

Fresh packed cherry tomatoes (red comet, origin Morocco) were purchased from local supermarket on the day of the experiments. Purchased in 250 g portions packed in a plastic film with three holes (⌀ 0.5 cm) on a central line on the upper side. Tomatoes were examined and those with smooth, shiny and a firm intact skin, with no visible moulds were used for the study. Quality characteristics and microbiological contamination of the product were analysed on the day of purchasing, which was taken as the reference point to analyse the deterioration process.

### 6.2.2 Nonthermal Plasma Treatment

The liquid plasma treatment system is mainly composed of a custom-built dielectric barrier discharge plasma source and a modular treatment acrylic chamber of 400 mL volume (Figure 6-2), connected to a high voltage AC power source PVM500 (Information Unlimited). The dielectric barrier plasma source was a perforated stainless steel flange which served as the ground electrode to enhance the local electric field as well as serving as the primary gas diffusion pore structure. Additionally, a water-proof breathable membrane was used as the main gas diffusor, which isolated the water treatment chamber from the plasma source to avoid any interference with the plasma discharge. Plasma discharge in air was generated at 25 kV peak-to-peak voltage. The amplitude of the current pulse was 500 mA and was measured from the ground electrode by the current probe. The frequency of the applied voltage was 25.3 kHz. The applied voltage waveform was sinusoidal. To diffuse the plasma treated air into the water phase, atmospheric air was fed into the reaction chamber from the bottom at a flow rate of 2 L/min. The system was allowed to stabilize until plasma was clearly visible (purple flashing) before the reactor chamber was loaded with 70 g of product and immediately flushed with distilled water (250 mL). The product was treated for 5 min in the plasma-activated water based on previously reported antimicrobial efficacy comparable to chloride treatment achieved within 5 min (Patange et al., 2019). The temperature was continuously monitored, and the pH of plasma-activated water was measured using a digital pH meter (FE20, Mettler Toledo, Schwerzenbach, Switzerland). After the required treatment time, the system was turned off, the tomatoes were removed with the aid of a sterile forceps and were placed on a clean, absorbance tissue to drain remaining water from the product. Plasma-activated water was removed from the chamber with a pump after each treatment.

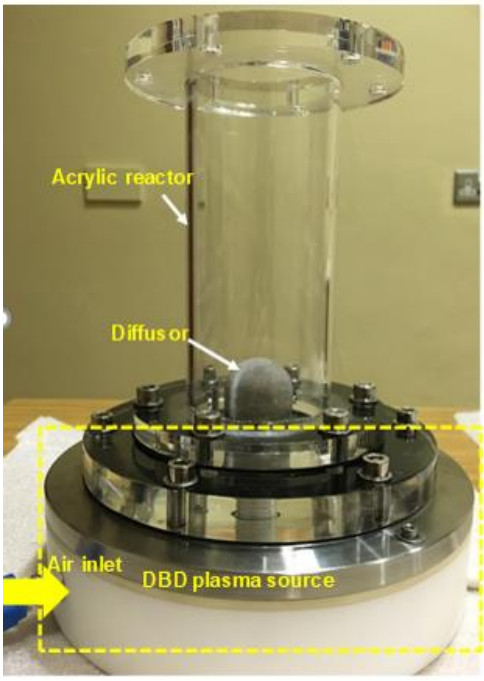


*Figure 6-2: 400ml Plasma water washing module.*

### 6.2.3 Equilibrium Modified Atmosphere Package design

Each 148 x 115 x 50 mm polypropylene tray (micro-pack 2000, Barquettes, France) was loaded with 145 ± 5 g of cherry tomatoes and packages were sealed with oriented polypropylene (OPP) film. The film was perforated with a needle (270 μm diameter) to ensure that the atmosphere conditions inside the package would reach the optimal amount of oxygen and carbon dioxide. The optimum perforation was determined using the respiration model for cherry tomatoes presented by Sousa et al. (2017).

### 6.2.4 Experimental design

A full factorial design was used to study the effect of a plasma washing step and the use of an EMAP under controlled recommended (10°C) and abusive (20°C) temperature conditions on the quality of cherry tomatoes throughout storage. The cherry tomatoes were stored unpacked and packed with and without previous plasma treatment; cherry tomatoes packed by the supermarket was used as a reference point for the shelf life extension. The quality of the cherry tomatoes during storage was assessed by measuring the weight loss, skin colour, total soluble solid (TSS) content and the pH over time. Microbiological contamination was monitored by determining the total aerobic microbes as well as yeast and mould present on the produce. The values presented in this report are average values of at least duplicates.

### 6.2.5 Chemical analyses of plasma-activated water

The acidity of the plasma-activated water was determined with a pH meter (FE20, Mettler Toledo, Schwerzenbach, Switzerland). Reactive oxygen and nitrogen species in the plasma-activated water in form of nitrite, nitrate, hydroxyl, and hydrogen peroxide were determined.

The concentration of nitrite in the solution was determined based on its reaction with Griess reagent by mixing 2.6 mL deionized water, 100 µL of Griess reagent (Sigma-Aldrich, 03553, Germany) and 300 µL of sample, blank or standard solution. Mixed solutions were allowed to develop at room temperature in the dark for 30 min, and nitrite concentration was calculated from the absorbance at 548 nm (spectrophotometer Libra S22, Biochrom, Cambridge, UK), based on a sodium nitrite standard curve (Patange et al., 2019).

The nitrate content was determined photometrically with Spectroquant®Nitrate Test (Merck, Darmstadt, Germany). Sample and standard solutions were measured at absorbance of 340 nm (spectrophotometer Libra S22, Biochrom, Cambridge, UK), and the concentration was calculated based on a calibration curve (Patange et al., 2019).

The peroxide content was determined based on the methodology proposed by Eisenberg (1943) with modifications. 2.5 mL of the sample was mixed to 250 µL of $TiOSO_4$ (Sigma-Aldrich, France). The mixtures were kept for 10 min in the dark at room temperature and the absorbance was measured at 405 nm. Hydrogen peroxide reacts with titanium sulfate reagent to form peritantic acid which has typical yellow colour. The peroxide concentration was calculated based on a calibration curve generated with hydrogen peroxide, where a linear regression was applied to the absorbance.

### 6.2.6 Quality characterisation

The weight of cherry tomatoes in each package was measured using a precision balance (Satorius, Germany). The weight loss (WL) was calculated based on the initial ($W_0$) and weight after time i ($W_i$) of the fruit, according to eq. 6.2-1.

$$W_L = [(W_0 - W_i)/W_0\,] \cdot 100 \qquad \text{eq. 6.2-1}$$

Visual colour degradation of the product was assessed using a Minolta ChromMeter (Model CR-300, Minolta Camera Co., Osaka, Japan), considering the saturation colour index (CI), which is dependent on Hunter colour parameters (a = red/green and b = yellow/blue) as shown in eq. 6.2-2. Three tomatoes per package were analysed and, on each tomato, two different parts were measured.

$$CI = a/b \qquad \text{eq. 6.2-2}$$

To measure the pH of the product, four tomatoes per package were previously homogenised with a blender and the pH of the solution was measured using a digital pH meter (FE20, Mettler Toledo, Schwerzenbach, Switzerland).

The TSS content of the product was measured using a digital refractometer (Refracto 30P, Mettler Toledo, Schwerzenbach, Switzerland). The values of TSS content were expressed as ˚Brix. The product was halved and gently squeezed to release the juice (~6 drops); three tomatoes per package were analysed.

For the microbiological analyses cherry tomatoes (around 30-40 g) were aseptically transferred to a sterile stomacher bag (Sewards LTD, UK) with 40 mL of sterile maximum recovery diluent (Sigma-Aldrich, India) and hand shacked (rubbed) for 2 min. Appropriate dilutions were made of the resulting suspension with serially diluting. Sample (0.1 mL) was spread plated onto a non-selective media tryptic soy agar (Sigma-Aldrich, India) to evaluate the total aerobic count. For the estimation of the yeasts and moulds, potato dextrose agar (Sigma-Aldrich, India) was used. The agar plates were incubated at 37°C and 25°C, for 48 h and 5 days, respectively. Results were expressed as $Log_{10}$ colony forming units per gram (CFU/g).

### 6.2.7 Statistical analysis

Analysis of Variance (ANOVA) and Tukey's test at 95% confidence level were used to assessing significant differences between different treatments. A full factorial design, with 3 factors at 2 levels, was used to evaluate the statistical significance of package, plasma treatment and temperature on the quality of the cherry tomatoes during storage, at a significance level of 5%.

Desirability was calculated as presented by Myers et al. (2016), as follows:

$$D = {d_1}^{w_1} \cdot {d_2}^{w_2} \cdot \ldots \cdot {d_n}^{w_n} \qquad \text{eq. 6.2-3}$$

where D is the total desirability, d is the desirability score of each property studied and w is the weight conferred to each property. Each property was weight equally. The desirability score is determined using a linear transformation to the values from the Pareto Front, scaling the objectives between 0 and 1 from worst to best quality. The optimal combination of the tested preservation technologies was validated qualitatively using characteristic quality and safety properties describing a lower level of product degradation, namely low pH, weight loss, colour index, aerobic count and TSS after 14 days of storage (Becker and Fricke, 2002; Romero, 1987; Tigist et al., 2013).

In order to avoid the elimination of some results that could be important to explain some observed phenomena, results with $0.05 < p < 0.1$ were considered marginally significant, and also considered to the analysis (Montgomery, 2013). The statistical analysis was performed by using Statistica software for Windows v. 7.1 (Tulsa, USA).

## 6.3 Results and discussion

The overall appearance of the cherry tomatoes was investigated visually. Packed tomatoes stored at 10°C could be stored for 35 days without comprising optical appearance, whereby packed cherry tomatoes stored at 20°C could be stored for 14 days (Figure 6-3). The main cut off criteria for unpacked cherry tomatoes was the shrinkage over time. The skin started to wrinkle, and the tomatoes dried out. Whereby, with packed tomatoes visual mould growth on isolated tomatoes could be observed and was used as cut off point.


*Figure 6-3: Plasma-treated cherry tomatoes stored for 35 days at 10°C unpacked (a) and packed (b), and stored for 14 days at 20°C unpacked (c) and packed (d).*

### 6.3.1 Microbiological growth

Alongside price, consumers consider freshness and food safety as the most important food purchase criteria (Röhr et al., 2005). Microorganism's metabolic activity renders the produce undesirable or unacceptable and therefore the aim throughout the food chain has to be to minimize the presence of microorganism as well as to prevent contamination and inhibit their preferable growth conditions (Lianou et al., 2016).

Plasma treated tomatoes show a decrease in aerobic microorganisms after a 5 min plasma washing step (Table 6-1). This findings are in accordance with Lee et al. (2018) who described an aerobic bacteria growth reduction on cherry tomatoes with a corona discharge plasma jet treatment. Whereby, a direct plasma treatment system with higher current and a shorter treatment time was used. Statistical results to evaluate the combined effect of plasma treatment, package and storage temperature show that after 3 days of storage, plasma treatment was the most important factor with effect on the aerobic total count, followed by the interaction effect temperature x treatment and the effect of the temperature (Figure 6-4a). Plasma treatment was responsible for decreasing the presence of aerobic microorganisms, and at 20°C their growth was favoured. However, after 7 and 14 days of storage, package became the most important factor with effect on the aerobic count. Packed products were more susceptible to the growth of aerobic microorganisms and a higher increase in microorganism in the packed tomatoes was observed. This is most likely due to the high humidity observed in the packages, which favours the growth of microorganism. OPP has low permeability to water vapour, and the perforation was not enough to prevent condensation inside the packages. Treatment and the interaction effect package x temperature had a marginally significant effect on the aerobic growth, which suggests that the treatment influenced the aerobic count, although less significant (Figure 6-4a). Notwithstanding, the average aerobic count until 14 days of storage was under $10^6$ CFU/g (Figure 6-5a), except for cherry tomatoes stored packed at 20°C, which presented an average count slightly above $10^6$ CFU/g. An aerobic count below $10^6$ CFU/g is the acceptable range for MAP produce based on Ireland's Guidelines for aerobic colony count microbiological quality for vegetables (FSAI, 2019).

A reduction in mould and yeast count by applying a 5 min plasma washing treatment to cherry tomatoes was observed (Figure 6-5a). For the mould growth on cherry tomatoes stored at 10°C the most important factor was the plasma treatment, followed by the package (Figure 6-4b). For short periods, plasma-washing treatment was able to control the mould growth satisfactorily, and unpacked products were also related to less mould count. However, at 14 and 21 days, none of the parameters presented any effect on the mould growth, and after 28 days of storage packaging had the only significant effect on the mould growth.

These results suggest that for longer periods, the water condensation inside the package might favour the mould growth. As this work indicated that packaging is essential to avoid weight loss of the produce, a film more permeable to water could be considered for the storage. Tumwesigye et al. (2017) proposed the use of cassava films as an alternative to the traditional OPP films with promising results; other biobased materials could be also evaluated.

Table 6-1: Quality determination (total solid content, colour index, pH and weight loss) of cherry tomatoes plasma-treated (T) or untreated (UT), packed (P) or unpacked (UP) stored at 10°C or 20°C throughout storage. Values represent mean (± S.D.) of measurements of at least triplicates.

| | **10°C** | | | | **20°C** | | | |
|---|---|---|---|---|---|---|---|---|
| | **day 3** | **day 7** | **day 14** | **day 35** | **day 3** | **day 7** | **day 14** | **day 28** |
| **Total soluble solid content [°Brix]** | | | | | | | | |
| NT,NP | 7.18 ± 0.59 b,A | 6.96 ± 0.47 a,b,A | 6.60 ± 0.74 a,b,A | 6.37 ± 1.10 a,b,A | 7.56 ± 0.34 a,A | 7.63 ± 0.40 a,A | 7.92 ± 0.78 a,A | - |
| T,NP | 7.25 ± 0.51 a,A | 7.02 ± 0.77 a,A | 6.90 ± 0.77 a,A | 6.67 ± 1.52 a,A | 7.43 ± 0.56 a,A | 7.09 ± 0.79 a,A,B | 7.85 ± 0.42 a,A | - |
| NT,P | 6.58 ± 0.40 a,A,B | 5.94 ± 0.38 b,c,A | 6.26 ± 0.31 a,b,A | 5.46 ± 0.48 c,A | 6.43 ± 0.70 a,b,A | 6.37 ± 0.64 a,b,B | 6.31 ± 0.63 a,b,B | 5.75 ± 0.49 b,A |
| T,P | 6.28 ± 0.46 a,b,B | 6.30 ± 0.82 a,b,A | 6.11 ± 0.45 a,b,A | 5.60 ± 0.52 b,A | 6.37 ± 0.66 a,A | 6.27 ± 0.43 a,B | 6.27 ± 0.59 a,B | 5.37 ± 0.48 b,A |
| **Colour index [%]** | | | | | | | | |
| NT,NP | 0.68 ± 0.09 c,A | 0.79 ± 0.09 a,c,A | 0.86 ± 0.09 a,b,A | 0.93 ± 0.11 b,A,B | 0.71 ± 0.18 a,A | 0.86 ± 0.05 a,b,A | 0.91 ± 0.05 b,A | - |
| T,NP | 0.71 ± 0.14 a,A | 0.75 ± 0.09 a,b,A | 0.88 ± 0.09 b,c,d,A | 0.97 ± 0.06 d,A | 0.71 ± 0.18 a,A | 0.86 ± 0.04 a,b,A | 0.89 ± 0.05 b,A | - |
| NT,P | 0.62 ± 0.16 a,A | 0.66 ± 0.14 a,A | 0.75 ± 0.12 a,b,A | 0.86 ± 0.07 b,B,C | 0.77 ± 0.12 a,A | 0.80 ± 0.14 a,A | 0.79 ± 0.14 a,A | 0.82 ± 0.15 a,A |
| T,P | 0.70 ± 0.12 a,A | 0.70 ± 0.20 a,A | 0.77 ± 0.14 a,A | 0.81 ± 0.08 a,C | 0.77 ± 0.12 a,A | 0.77 ± 0.10 a,A | 0.81 ± 0.11 a,A | 0.75 ± 0.17 a,A |
| pH | | | | | | | | |
| NT,NP | 4.48 ± 0.01 a,A | 4.57 ± 0.04 a,A | 4.51 ± 0.02 a,A | 4.60 ± 0.07 a,A | 4.51 ± 0.07 a,A | 4.52 ± 0.04 a,A | 4.62 ± 0.04 a,A | - |
| T,NP | 4.51 ± 0.03 a,A | 4.58 ± 0.04 a,A | 4.51 ± 0.05 a,A | 4.70 ± 0.11 b,A | 4.57 ± 0.05 a,b,A | 4.65 ± 0.09 a,b,A,B | 4.78 ± 0.00 b,A | - |
| NT,P | 4.48 ± 0.01 a,A | 4.61 ± 0.05 a,A | 4.63 ± 0.13 a,A | 4.66 ± 0.01 a,A | 4.52 ± 0.05 a,A | 4.60 ± 0.01 a,A | 4.53 ± 0.02 a,A | 4.50 ± 0.04 a,A |
| T,P | 4.47 ± 0.01 c,A | 4.59 ± 0.02 a,b,c,A | 4.54 ± 0.03 a,c,A | 4.70 ± 0.04 b,A | 4.55 ± 0.02 a,b,A | 4.67 ± 0.03 b,B | 4.57 ± 0.04 a,b,A | 4.52 ± 0.03 a,A |
| **Weight loss [%]** | | | | | | | | |
| NT,NP | 2.05 ± 0.19 a,A | 4.04 ± 0.46 a,b,A | 7.20 ± 0.56 b,c,A | 16.90 ± 2.00 e,A | 3.83 ± 0.61 a,A | 7.96 ± 0.82 b,A | 15.81 ± 2.64 c,A | - |
| T,NP | 1.71 ± 0.22 a,A | 4.06 ± 0.12 b,A | 7.99 ± 0.32 c,A | 15.42 ± 0.60 e,A | 3.38 ± 0.26 a,A | 7.74 ± 0.47 b,A | 11.75 ± 1.18 c,A | - |
| NT,P | 0.13 ± 0.02 a,B | 0.28 ± 0.01 a,B | 0.49 ± 0.08 a,b,B | 0.79 ± 0.01 a,b,B | 0.20 ± 0.07 a,B | 0.25 ± 0.04 a,B | 0.45 ± 0.01 b,B | 0.62 ± 0.01 c,B |
| T,P | 0.08 ± 0.05 a,B | 0.28 ± 0.07 a,b,B | 0.42 ± 0.02 a,b,B | 1.26 ± 0.21 d,C | 0.11 ± 0.01 a,B | 0.22 ± 0.03 b,B | 0.33 ± 0.01 c,C | 0.66 ± 0.04 e,B |

**Statistical analyses were conducted for samples at 10°C and 20°C separately. For each temperature, results on the same row followed by the same small letter and results in the same column followed by the same capital letter were not statistically different ($p < 0.05$) according to a Tukey-HSB test.*

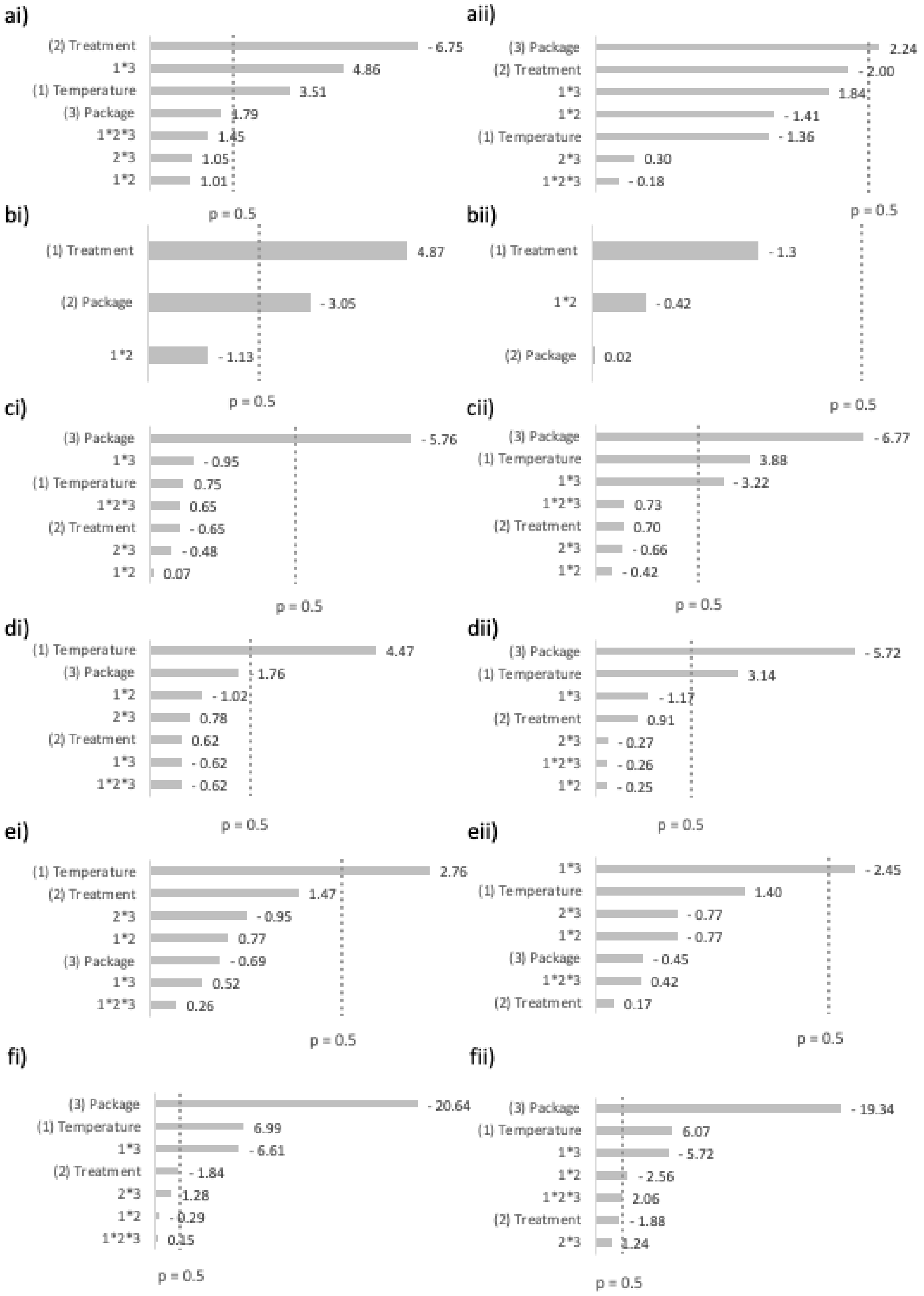


*Figure 6-4: Statistical analysis (pareto charts) of the effect of the plasma treatment, package and storage temperature on total aerobic mesophilic count (a), mould growth (b), total soluble solids content (c), colour index (d), pH (e) and weight loss (f) after 3 days of storage (i) and 14 days of storage (ii).*

### 6.3.2 Total Soluble Solid content

Changes in soluble solid contents over storage are expected as the metabolic activities result in the breakage of structural polymers. Glucose and other compounds are broken into soluble sugars and therefore an increase in TSS can be observed (Sharma et al., 2018).

The results obtained at 10°C and 20°C have shown that package has a huge influence on the TSS of the cherry tomatoes. Although all the products presented oscillation on their results over time and no significant differences could be seen (Table 6-1), unpacked products showed greater TSS values over storage compared to their initial content while packed products presented lower TSS than their initial values at 20°C. Lower TSS after few days of storage was observed by Tumwesigye et al. (2017) on cherry tomatoes packed with cassava and OPP films at 10°C and associated with higher levels of $CO_2$ and lower levels of $O_2$ surrounding the commodities due to respiration. High $CO_2$ concentration leads to lower respiration rates and therefore a decrease on the degradation of metabolites responsible for the increase on TSS (Wills and Golding, 2018). The statistical analysis showed that packaging was the most important factor influencing the TSS (Figure 6-4c) and emphasized the advantage of a package for the storage of cherry tomatoes. At 14 days of storage temperature was also a significant effect, followed by the combination of package x storage temperature. These results indicated that packed products exhibited lower TSS during the whole storage, as can be seen in Figure 6-6a. Plasma treatment did not present a statistically significant effect on the TSS as well no significant differences could be observed during treated and untreated stored product.

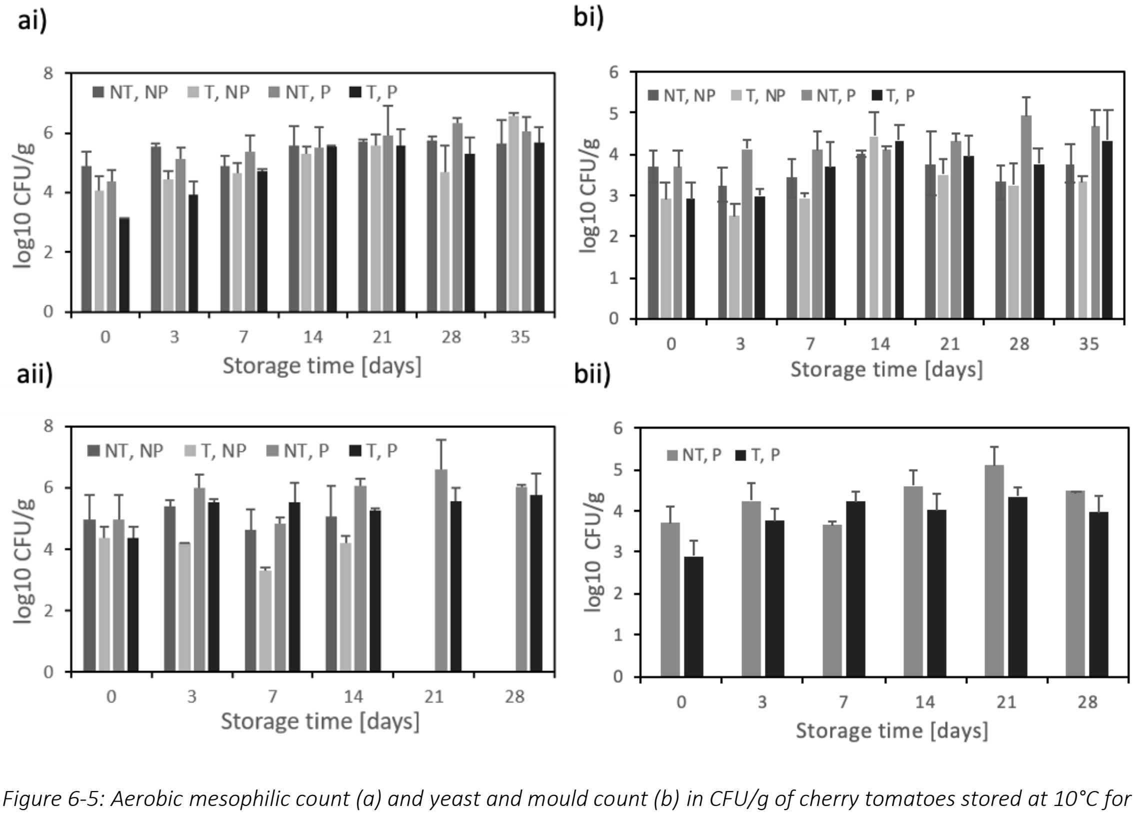


*Figure 6-5: Aerobic mesophilic count (a) and yeast and mould count (b) in CFU/g of cherry tomatoes stored at 10°C for 35 days (i) and at 20°C for 28 days (ii). NT are untreated cherry tomatoes, NP are unpacked products, T are treated products and P are packed cherry tomatoes.*

### 6.3.3 Colour changes

Colour is the major appearance aspect of most foods; it is the first sensory attribute that costumers can appraise. It often degrades during storage due to enzymatic and non-enzymatic reactions, oxidation, and physical reactions. Good correlation between colour, food quality and maturity has been reported (Subramaniam, 2016). The colour index is used to characterize the external colour of fruits and vegetables, and has been associated with maturation of the produce (Pathare et al., 2013). Plasma treatment did not affect the colour index as no significant differences could be observed on treated and untreated samples (Table 6-1). This findings are in agreement with cold plasma treated cherry tomatoes and other vegetable goods (Misra et al., 2014), were changes in colour amount of treated and untreated samples were found insignificant. A slight increase on the colour index over time for all samples stored at 10°C and a higher increase for all samples stored at 20°C was observed (Table 6-1). The increase in CI describes an increase in red colour throughout the storage period. Higher colour indices for unpacked samples compared to packed samples were observed at 10°C and 20°C, on the last day of storage (35 and 14 days, respectively). The statistical analysis show that after 14 days of storage, package was the most important factor with influence on the CI; the effect of temperature is also significant (Figure 6-4d). In conclusion, the colour index is greater at higher temperatures, and this effect is more pronounced in unpacked cherry tomatoes (Figure 6-6b). This finding is in accordance with packaged cherry tomatoes in a MAP with cassava film describing a slight increase CI during storage at 10°C (Tumwesigye et al., 2017) as well as the influence of the storage temperature on the colour changing rate (Cantwell et al., 2009)

### 6.3.4 pH

pH change during storage is an indicator of quality, where a lower pH value is related to slower respiration rate and better quality maintenance (Tigist et al., 2013). During ripening, organic acids are used as substrate in respiration processes, wherefore the pH of the tomatoes during storage increases (Duma et al., 2017; Tilahun et al., 2017). In this study, very little variation on pH over time was observed (Table 6-1). A tendency to a pH-increase at the end of storage time could be seen. However, there was no significant difference between pH measured at different storage times as well as no significant influence of the plasma treatment or packaging was observed It was also mentioned that a slightly higher pH was measured for plasma treated tomatoes, with no statistically significant. A same tendency can be found in the obtained results. A stable pH over the tested storage time and conditions implies a slow respiration and maturation. The statistical results show that after 3 days of storage, temperature was the only significant factor influencing the pH where higher pH values were observed at a storage temperature of 20°C. After 14 days of storage the interaction effect between temperature x package was most important (Figure 6-4e). These findings supports that a combination of a MAP and low temperature may reduce the degradation of ascorbic acid in fruits (Lee and Kader, 2000). Fagundes et al. (2015) studied the organic acid content of cherry tomatoes packaged under MAP conditions and observed no significant decrease of organic acid, except for tartaric acid (reduction of 0.16 g/L), over storage time of 25 days.

### 6.3.5 Weight loss

Weight loss is a result of nutrient depletion due to respiration and water exchange due to difference in gradient of water vapour pressure between the tissue and the surrounding air (Becker and Fricke, 2002; Romero, 1987). Both are highly dependent on the environmental conditions, namely temperature and relative humidity. Hence, it did not come as a surprise that package was the most significant parameter with effect on weight loss. Results presented in Table 6-1 show that in all studied cases a significant increase in water loss throughout storage was observed. However, cherry tomatoes exposed to the ambient conditions inside the incubators without the protection of a plastic film lost much more water than those packed. The packaging film clearly created a protection against air movement inside the incubator and created its own environment inside the tray, preventing the weight loss due to difference of water vapour pressure. Products stored for 3 days in open trays lost more water (WL 2-4%) than packed cherry tomatoes after 35 days of storage (WL 0.8-1.26%). This result emphasises the need for a good packaging design to prevent losses. On the other hand, the effect of plasma treatment used in this study was marginally significant to the weight loss ($0.05 < p < 0.1$). As it can be seen in Figure 6-6c, the weight loss of cherry tomatoes that were packed was very low, and did not change much from 10°C to 20°C or between treated and untreated samples; however, the weight loss was much greater when the products were not packed, and the loss was more pronounced at products that did not receive plasma treatment and were stored at 20°C. It is important to notice that cherry tomatoes packed and stored at 20°C presented good aspects until 21 days of storage (WL 0.52-0.56%); however, products that were not packed had a water loss of 11.7-15.8% after 14 days. At 10°C, the cherry tomatoes could be stored packed for 35 days. This indicates that the storage temperature is an important factor to prevent weight loss of the produce. Temperature and the interaction effect package x temperature were also statistically significant, which emphasises the relationship among relative humidity, temperature, and weight loss (Figure 6-4f). Research in EMAP for cherry tomatoes have suggested either 10°C (Tumwesigye et al., 2017) or 20°C (Briassoulis et al., 2013) as ideal storage temperatures for cherry tomatoes, which could be due to the effect of other parameters such as different atmosphere conditions and package materials used in the studies. The important of a package regarding the weight loss is in accordance with other storage experiments with tomatoes products e.g., cherry tomatoes packed in MAP had a WL of 0.18-0.6% after 21-25 days of storage at 5-7°C (Fagundes et al., 2015; Paulsen et al., 2019). Furthermore, the results of the unpacked cherry tomatoes are in alignment with this study reporting a weight loss of 10-19% after 23-25 days stored between 5 to 20°C (Fagundes et al., 2015; Paulsen et al., 2019).

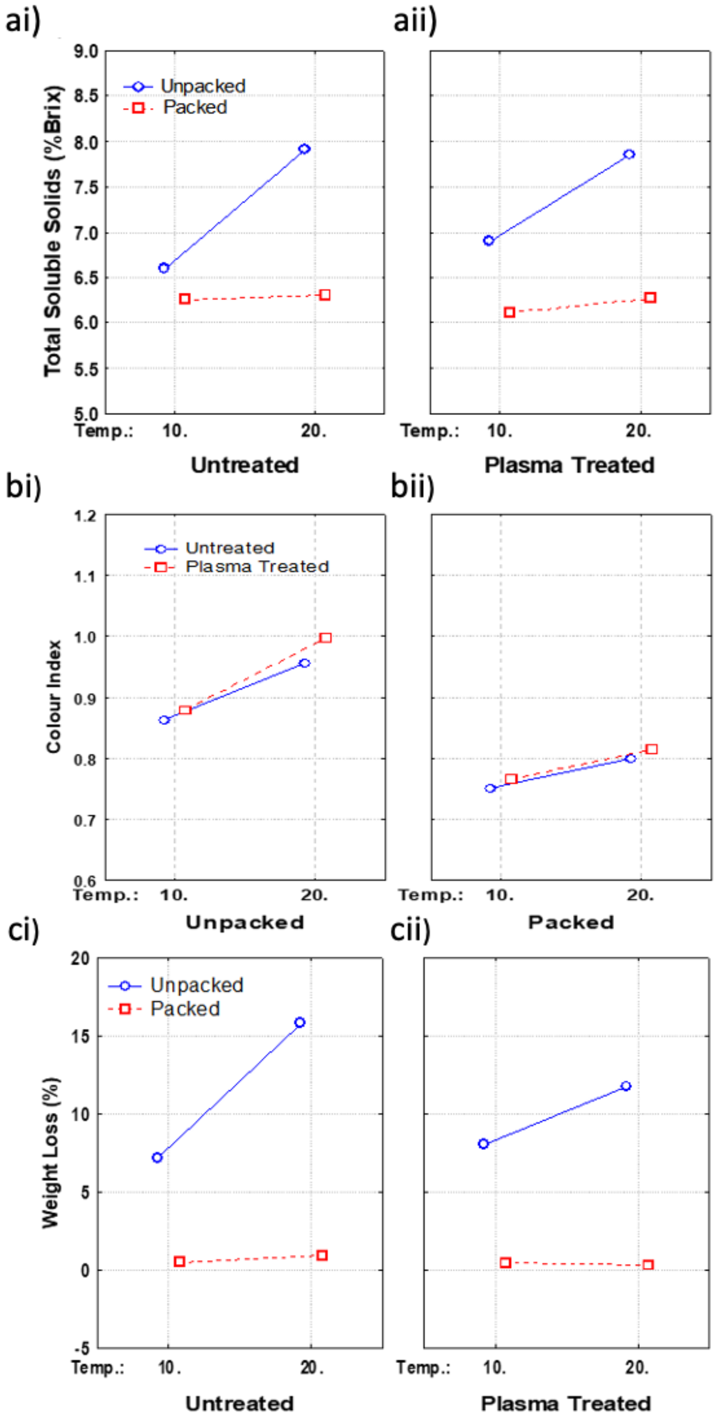


*Figure 6-6: Marginal means plot of total soluble solids (a) of cherry tomatoes stored at 10°C and 20°C after 14 days of storage, packed (-□-) and unpacked (—◇—) tomatoes, untreated (i) and plasma-treated (ii). Marginal means of colour index (b) of cherry tomatoes stored at 10°C and 20°C after 14 days of storage, plasma-treated (-□-) and untreated (—◇—) tomatoes, unpacked (i) and packed (ii). Marginal means plot of weight loss (c) of cherry tomatoes stored at 10°C and 20°C for 14 days of storage packed (-□-) and unpacked (—◇—) tomatoes, untreated (i) and plasma-treated (ii).*

### 6.3.6 Integration of a plasma washing, EMAP and storage temperature

The effect of temperature, plasma treatment and packaging on the different properties were studied separately to understand how they affect each aspect of the produce quality. However, as they relate differently to each of the parameters, it is important to establish which conditions maximise the maintenance of the shelf life. The simultaneous optimisation technique of Derringer and Suich (1980) was used for targeting the best storage conditions for cherry tomatoes. The desirable values were low pH, weight loss, colour index, aerobic count and TSS after 14 days of storage, as it characterises lower levels of product degradation and therefore quality maintenance over time.

The results in Figure 6-7 indicate that cherry tomatoes plasma-treated and packed presented the lowest quality degradation during storage; under these conditions, storage temperature had little effect on the quality of the produce. Untreated products presented better results when packed and stored at 20°C, which is probably due to the fact that at 10°C condensation was observed in the package. It also might explain the fact that unpacked products presented higher desirability (lower degradation) at lower temperatures, as there is no condensation when the products are not packed. However, when longer periods of storage are required (more than 14 days), temperature of 10°C is crucial to keep the products suitable for consumption. Nevertheless, the cherry tomatoes used in this research are commercialised with a window of 3 days to the best before date, and therefore the results of this research represent great improvement to the current best before date.

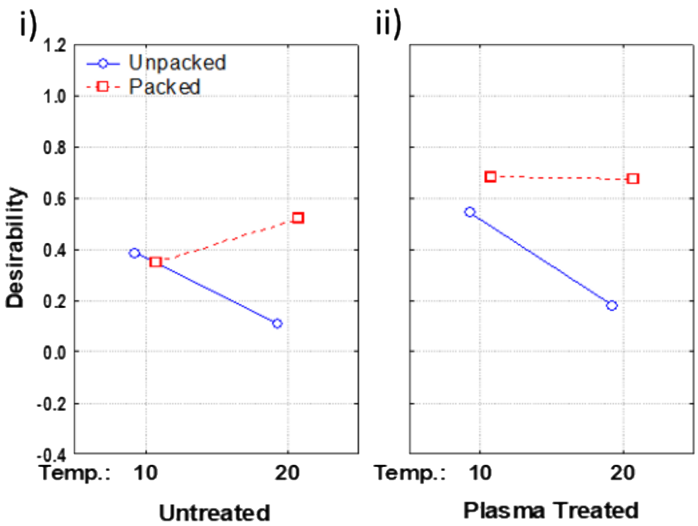


*Figure 6-7: Predicted optimal parameters based on desired low brix, colour index, aerobic count, pH and weight loss after 14 days of storage, packed (-□-) and unpacked (-◇-) tomatoes and untreated (i) and untreated (ii).*

## 6.4 Conclusion

Statistical analysis of the results showed the importance of a package to prevent weight loss and changes in total soluble solid content of cherry tomatoes over 14 days of storage. Temperature had a great impact on the shelf life of the product, as cherry tomatoes kept at 10°C could be stored for 35 days while at 20°C the produce lost their integrity after 14 days.

This represents a shelf life extension of 32 days and 11 days respectively, based on the best before date of the supplier. The combination of cold plasma and EMAP design was important to ensure the quality of cherry tomatoes, which suggests that they have potential to be used as hurdle technology and can be explored to reduced food waste by extending the shelf life of fresh produce. Cherry tomatoes stored to be consumed until 14 days of storage could be stored at 10°C or 20°C as long as they were packed. Untreated products suffered quality losses when stored unpacked at 20°C, but when packed they could be stored at 20°C. Although temperature and plasma treatment affected the properties of the products, EMAP was the parameter with higher effect on the shelf life of the cherry tomatoes, with great influence on preventing weight loss. Hence, the results showed that cherry tomatoes should be stored under EMAP conditions in order to prevent degradation.

## 6.5 Acknowledgments

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Chapter SEVEN

# Completion of a sustainable integrated precision packaging

## Abstract

*A packaging based on an edible coating, requires a complementary packaging component e.g., a packaging film. Combining the edible coating and a film layer provides an optimal product protection over the required shelf life. Therefore, a mathematical model approach was taken to estimate the required water barrier properties for the packaging film material as the water loss was identified as the remaining most critical quality parameter after a chitosan coating was applied. A water loss of 10% within 21 days was the selected cut of point. The maximal WVTR was calculated, using the GAB model and a linear approximation of the isotherm, to be 94 and 81 g/m$^2$ day, respectively. Indicating that the simplified linear model showed similar results to the GAB model approach for predicting the moisture loss of a high moisture food product. When increasing the required shelf life only a small reduction of the maximal allowed WVTR was seen. Film materials were selected based on the water barrier properties and environmental performance. Testing the WVTR at chilled storage conditions showed that depending on the material selected, the temperature can impact the water vapour barrier of the packaging film materials, especially for biobased film materials. The recommended film material in combination with a chitosan edible coating for a RtE baked fish product was a cellulose based compostable film material.*


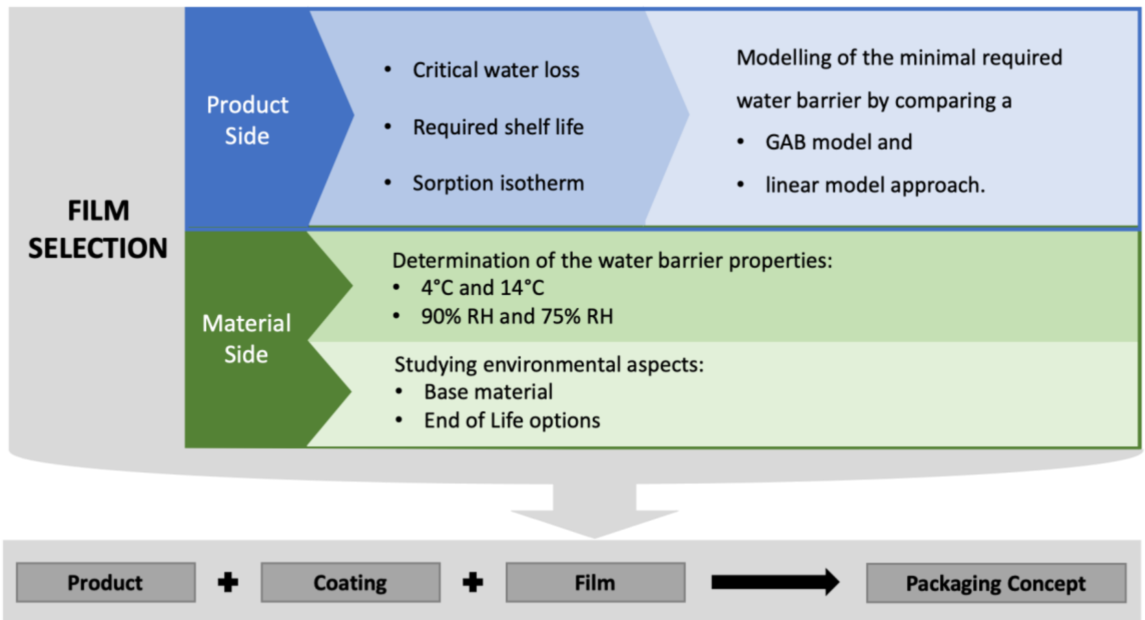


*Figure 7-1: Graphical Abstract – Completion of a sustainable integrated precision packaging.*


Keywords

*Selection of film material*
*Modelling water barrier requirements*
*WVTR of film materials*
*Environmental considerations*

## 7.1 Introduction

Currently used packaging concepts for chilled RtE seafood products often use a multilayer plastic film, sometimes packed under modified atmosphere to ensure their safety and quality. Considering the product protection these films have an excellent oxygen and water vapour barrier, controlling different degradation processes, which is critical for minimising food waste as per the circular economy approach (Hopewell et al., 2009; Kaiser et al., 2018; Morris, 2017). Multilayer plastic films are hardly recycled, ending up in incineration facilities or landfills (Dainelli, 2008; Hopewell et al., 2009; Kaiser et al., 2018). An alternative packaging concept is a hurdle packaging system consisting of an edible coating and an outside layer. The first hurdle is the edible coating directly applied to the product surface, and the second hurdle is a plastic film layer surrounding the product. Hurdle technology is a common way in the food supply chain to protect food products, with the aim to reduce degradation processes. An additional important hurdle is chilled storage as a lower temperature decreases the reaction rate of microbial growth, or chemical reactions.

The currently used food packaging concepts for high perishable food, as RtE seafood products, are mainly selected based on the limiting critical quality parameters, often the microbial growth. Nevertheless, it was shown that the microbial degradation can be well controlled with chitosan based edible coating applied to the product surface (as shown in chapter 5). When the food safety is protected by an edible coating, the water loss is the limiting shelf life factor. Moisture loss is a result of water exchange due to difference in gradient of water vapour pressure between the product and the surrounding air (Becker and Fricke, 2002). The loss of moisture influences the product appearance, an important sensory attribute for freshness and a significant driver for the consumer buying behaviour. The outside layer should therefore provide a sufficient water vapour barrier, beside other essential functions as a protection against cross-contamination, or a platform for product and supplier information.

The moisture transfer rate is affected by the water activity of the food, outside temperature, humidity, and air circulation, and it can be significantly controlled by the barrier properties of the plastic film layer. A common way for estimating the required water vapour barrier and selecting film materials for low and medium moisture foods is the assumption of steady state conditions of temperature and humidity in the surrounding environment. The relation between the water vapour permeation and relative humidity gradient can be mathematical described with a developed model based on Fick’s law (Robertson and Lee, 2021). Combining the mathematical models describing the relation between the water activity and water loss of the product, these models were used to predict the shelf life as the time to reach a critical moisture content value. The simplest function, assuming a linear relationship between the moisture content and water activity in the case for low to medium food products is only valid for water activity range of up 0.6, and is commonly applied in industry due to its easy application (Robertson, 2012; Robertson and Lee, 2021). The sorption isotherm of an RtE baked fish product was detected to follow a type II isotherm behaviour (chapter 2). The sorption isotherm could be well described by the mathematical non-linear function of GAB and Peleg over a broad range of water activity.

Beside the technical requirements, also environmental aspects are important to consider. Every product comes with an environmental burden, nevertheless the impacts can differ depending on the raw materials used, the packaging structure, and the end of life treatments. Circular economy aims at reducing primary resource extraction by using all materials at their EoL phase (Ellen McArthur Foundation, 2022). Ensuring that materials either stay in the same material cycle or enter a new cycle, with the priority to keeping products, components and materials at their highest value at all times. Therefore, a precondition is an EoL treatment of food packaging that materials can stay in the same or another material cycle.

The purpose of this chapter was to select a sustainable and suitable film packaging material based on technical and environmental requirements, for an RtE coated baked seafood product. Therefore, the required water vapour permeability of the outside film layer for a RtE baked fish product was determined by comparing two model approaches. The steady-state permeation equation describing the relation of the water pressure with a linear relation (A), and a GAB model relation (B). The predicted maximal WVTR was compared to detected WVTR of four film materials at cooled storage conditions. Additionally, the film material was investigated in regard to sustainable aspects for the final film material selection.

## 7.2 Materials & Methods

### 7.2.1 Mathematical relations

In the following all relevant mathematical relations are presented used for the prediction of the film material water vapour barrier properties:

Relation between the moisture content wet basis and dry basis:

$$m_{db} = \frac{m_{wb}}{1 - m_{wb}} \text{ or } m_{wb} = \frac{m_{db}}{1 + m_{db}} \qquad \text{eq. 7.2-1}$$

Relation between water activity and partial vapour pressure of the product:

$$RH = a_w = \frac{p_p}{p_{sat}} \qquad \text{eq. 7.2-2}$$

Mass transfer model based on Fick's law: Water vapour pressure in steady-state permeation for single packaging layer with water transport from the product to outside air:

$$\frac{dW}{dt} = \varphi \cdot A \cdot \left(p_{p,ins} - p_{p,out}\right) \qquad \text{eq. 7.2-3}$$

Relation between water loss and moisture content:

$$dW = dmW_s \qquad \text{eq. 7.2-4}$$

Relation between water vapour transmission rate, permeance and pressure:

$$\frac{P}{X} p_0 M_{r,H2O} = TR \qquad \text{eq. 7.2-5}$$

Euler's method:

$$y_{i+1} = x_i + h\, f(x_i, y_i) \qquad \text{eq. 7.2-6}$$

### 7.2.1.1 *Linear model approach*

Linear relation between moisture content and water activity:

$$m = b_{lin} a_w + C_{lin} \qquad \text{eq. 7.2-7}$$

Substitute aw for the expression in eq. 7.2-2. and solving the equation by $p_p$:

$$m = b_{lin} \frac{p_p}{p_{sat}} + C_{lin}$$

$$p_p = \frac{(m - C_{lin}) \cdot p_{sat}}{b_{lin}}$$

Substitution $p_p$ in eq. 7.2-3:

$$\frac{dm}{dt} = \frac{P}{X} \cdot \frac{A}{W_s} \cdot \left( \frac{(m_{\square} - C_{lin}) \cdot p_{sat}}{b_{lin}} - \frac{(m_e - C_{lin}) \cdot p_{sat}}{b_{lin}} \right)$$

Rearranging gives:

$$\frac{dm}{m - m_x} = \frac{P}{X} \cdot \frac{A}{W_s} \cdot \frac{p_{sat}}{b_{lin}} dt$$

with

$$m_x = m_e - 2 \cdot C_{lin}$$

On integrating gives:

$$\ln \frac{m_i - m_x}{m - m_x} = \frac{P}{X} \cdot \frac{A}{W_s} \cdot \frac{p_{sat}}{b_{lin}} t$$

Considering the relation 7.2-5 and rearranging the equation to TR gives:

$$TR = ln\left(\frac{m_i - m_x}{m_{\square} - m_x}\right) \cdot W_s \cdot \frac{b_{lin}}{t_{min} \cdot A} \qquad \text{eq. 7.2-8}$$

with

$$m_x = m_e - (2 \cdot C_{lin})$$

### 7.2.1.2 *GAB model approach*

GAB model relation between moisture content and water activity:

$$\frac{m_i}{m_{0,GAB}} = \frac{C_{GAB} \cdot k_{GAB} \cdot a_w}{(1 - k_{GAB} a_w)(1 + C_{GAB} k_{GAB} a_w - k_{GAB} a_w)} \qquad \text{eq. 7.2-9}$$

Rearranged GAB model to make the vapour pressure of water vapour the subject gives:

$$p_{\square} = p_0 \left[ \frac{-B_2 - \sqrt{B_2^2 - 4B_1}}{2B_1} \right] \qquad \text{eq. 7.2-10}$$

with

$$B_1 = k_{GAB}^2 (1 - C_{GAB})$$
$$B_2 = k_{GAB} \left( C_{GAB} - C_{GAB} \frac{m_{0,GAB}}{m} - 2 \right)$$

Substitution $p_p$ in eq. 7.2-3:

$$\frac{dm}{dt} = \frac{P}{X} \cdot M_{r,H20} \cdot A \cdot \frac{p_0}{M_s} \left[ \left( \frac{-B_2(m_e) - \sqrt{B_2(m_e)^2 - 4B_1}}{2B_1} \right) - \left( \frac{-B_2(m) - \sqrt{B_2(m)^2 - 4B_1}}{2B_1} \right) \right] \qquad \text{eq. 7.2-11}$$

with

$$B_1 = k_{GAB}^2 (1 - C_{GAB})$$
$$B_2(m_e) = k_{GAB} \left( C_{GAB} - C_{GAB} \frac{m_{0,GAB}}{m_e} - 2 \right)$$
$$B_2(m) = k_{GAB} \left( C_{GAB} - C_{GAB} \frac{m_{0,GAB}}{m} - 2 \right)$$

### 7.2.2 Studied film materials

Four plastic film materials were selected for this study, based on the composition and barrier properties at standard temperatures (Table 7-1).

Table 7-1: Film plastic materials and their properties.

| Films | Materials | Thickness [mm] | Properties |
|---|---|---|---|
| NatureFlex™ NK | Cellulose film covered with a heat-sealable moisture barrier coating, compostable | 30 | heat-sealable, moisture barrier, compostable |
| NatureFlex™ NVS | Cellulose with a coating on both side | 30 | Transparent, heat-sealable, home and industrial compostable |
| SOPP | Oriented polypropylene | 30 | Synthetic monolayer material |
| PropaFresh RK+ | Biaxially oriented PP film coated with two aqueous dispersions of polyvinylidene chloride (PVdC) copolymer | 35 | Synthetic, multilayer material |
| Aluminium foil | aluminium | 40 | Recyclable, when collected and sorted |

### 7.2.3 Determination of water vapour transmission rate

The WVTR of the films at specific temperature and relative humidity were studied. The temperatures were chosen based on the recommended storage temperature of 4°C, and abusiv conditions of 14°C (Table 7-2) and compared to the values provides from the film suppliers based on standard test conditions (38°C, 90% or 25°C, 75%RH).

Table 7-2: Studied WVTR parameters.

| **Levels** | 1 | 2 |
|---|---|---|
| **RH%** | 90 | 75 |
| **Temperature** | 4°C | 14°C |

The WVTR was determined by using the gravimetric “cup” method described in ASTM E96-95, 1995. Cylindrical PP test cups (6.4 cm bottom diameter, 8.4 cm depth and 7.4 cm top inner diameter) with a lid was used. The lid had an opening of 6.6 cm diameter to expose the film to the inside atmosphere of a dry anhydrous calcium chloride (0%) and outside atmosphere of selected RH. The exposed film surface area was 43 cm$^2$. The cup was filled with saturated salt solution to a maximum half capacity, the film was placed onto the opening and closed with the lid. To ensure an airtight sealing silicon grease was applied around the edge of the cups. The sealing was tested by covering a test cup with aluminium foil. The cups (triplicates) were kept under constant relative humidity and temperatures, by placing the cups in an airtight plastic container with different salt solutions in an incubator (Table 7-3). The mass of each cup was periodically monitored (daily basis for two weeks), by removing, weighting on an analytical balance, and replacing the cups in the air sealed boxes. The weight loss over time (g/day) was calculated from the measured data in steady state, which was confirmed by the linearity of weight loss with time (Macedo et al., 2013), and the water vapour transmission rate was calculated with the film surface area and was further transposed to water vapour permeability by the driving force (RH gradient) and the thickness of the tested films as described by Macedo (2011).

Table 7-3: Salt solution and their equilibrium relative humidity at defined temperature.

| **Salt** | **Factors** | **Levels** | | |
|---|---|---|---|---|
| | **Temperature** | **4°C** | **14°C** | **38°C** |
| Potassium Chloride | RH% | 87 | 88 | - |
| Sodium Chloride | | 76 | 76 | 75 |

## 7.3 Results & Discussion

### 7.3.1 Model prediction

To be able to control the water loss until a desired shelf life, a minimum allowed permeability of the film material can be estimated. Therefore, the permeability model was used assuming a steady state permeation of water vapour through the material with a constant driving force and varying internal vapour pressure due to moisture content changes over time. Based on the assumption that the moisture gradient inside the package is negligible and the film material is the major resistance to water vapour transport (Robertson, 2012). The moisture content changes were model with a linear and GAB model. Additional, set parameters to predict the minimum required water vapour permeability of an outside layer are summarized in Table 7-4. The results showed that the minimum required water vapour permeability allowing a water loss of less than 10% over 21 days are 81 g/m$^2$ day based on the linear model approach or 95 g/m$^2$ day based on GAB model approach for products stored at 4°C and a relative humidity of 75%. It could be seen that for the required water activity range above 0.9 the linear model provides similar results as the GAB model, therefore the simpler linear approach can be recommended as a standard method for predicting the required water vapour transmission rate under defined storage conditions. The models were used to estimate the minimal WVTR for different shelf life with a cut-off point of 10% water loss (Figure 7-2). An exponential behaviour could be seen, showing that with increasing shelf life a lower WVTR of the film material is required. A minimum required shelf life of the product was 21 days with a WVTR of around 81-95 g/m$^2$ day, but also increasing the shelf life to 28 or even 35 days would be possible with only small changes in the required WVTR (e.g., for 35 days the WVTR was estimated to be between 48-56 g/m$^2$ day). Every mathematical model underlay the assumption taken, therefore it is recommended to test the real product shelf life under realistic supply chain conditions. Nevertheless, the model approach was shown to be a good estimation of the required barrier properties (Kruger, 2011).

Table 7-4: Data used to for the prediction of the required water vapour permeability of baked fish fillet.

| **Parameter description** | **Symbol** | **Value** | **Unit** |
|---|---|---|---|
| GAB parameter at 4C | $k_{GAB}$ | 0.9954 | - |
| GAB parameter at 4C | $C_{GAB}$ | 11.04 | - |
| GAB parameter at 4C | $M_{0,GAB}$ | 0.0637 | Kg water/kg solid |
| Mass of product | W | 0.12 | Kg |
| Mass of solids | $W_s$ | 0.09 | Kg |
| Moisture content at equilibrium (dry basis) | $m_e$ | 0.27 | Kg water/Kg solid |
| Moisture content at equilibrium (wet basis) | $m_{e,wb}$ | 0.27 | Kg water/Kg product |
| Moisture content critical (dry basis) | $m_c$ | 1.86 | Kg water/Kg solid |
| Moisture content critical (wet basis) | $m_{c,wb}$ | 0.65 | Kg water/Kg product |
| Moisture content initial (dry basis) | $m_i$ | 3 | Kg water/Kg solid |
| Moisture content initial (wet basis) | $m_{i,db}$ | 0.75 | Kg water/Kg product |
| Molecular mass of water | $M_{r,H20}$ | 0.01802 | Kg/mol |
| Saturated vapour pressure of water vapour | $p_{0,\ 4°C,\ 75\%\ RH}$ | 813 | Pa |
| Shelf life | t | 21 | days |
| Surface area of packaging film* | A | 0.0293 | m$^2$ |
| Thickness of film | X | 0.00004 | m |
| Transfer rate | TR | $1.10x10^{-6}$ | Kg/m$^2$ s |
| Transfer rate | TR | 0.095 | Kg/m$^2$ day |
| Transfer rate | TR | 95.29 | g/m$^2$ day |
| Water vapour permeability | P | $3.01x10^{-12}$ | (mol m)/(m$^2$ s Pa) |

* Package assumptions (a film surrounding the product)

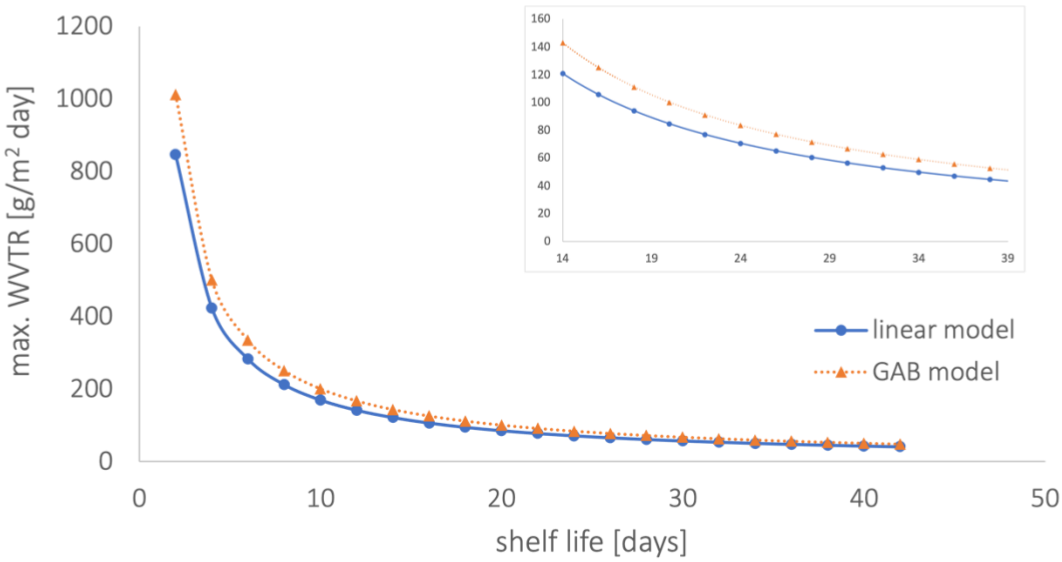


*Figure 7-2: Predicted maximal WVTR for a defined shelf life with a cut of point of 10% moisture loss.*

#### *7.3.1.1 Linear Model*

The required minimum transmission rate based on the linear model approach was calculate based on equation 7-2.8. The slope ($b_{lin}$), crossing with the y-axis ($C_{lin}$) and the pseudo-equilibrium moisture content ($m_{e'}$) of the linear relation was calculated based on the sorption isotherm, and the help of the initial and critical moisture content as described below and shown in Figure 7-3. The slop of the linear relation is 57.14 Kg water/Kg solids/unit $a_w$. The pseudo-equilibrium moisture content by extending the linear portion of the sorption isotherm gives a value of -10.14. The point of the intersection with the y-axis was estimated to be equal to -53. From, this the corresponding WVTR could be calculated for a specific required shelf life. Important to note are the boundary conditions for the linear model as the linear is only valid for higher activity ranges above 0.88 (Table 7-5). In the case of a high moisture RtE seafood product higher water activity ranges are more relevant, as the initial and critical moisture content are within the water activity range above 0.9.

Defined expression for missing unknown:

$$C_{lin} = m - (b_{lin} a_w) \quad \text{eq. 7.3-1}$$

$$b_{lin} = \frac{m_2 - m_1}{a_{w_2} - a_{w_1}} \quad \text{eq. 7.3-2}$$

$$m_e = b_{lin} a_{w_e} + C_{lin} \quad \text{eq. 7.3-3}$$

Table 7-5: Boundary conditions of the linear model.

| | |
|---|---|
| $m_b < \mathrm{m} < m_i$ | $0.44 < \mathrm{m} < 3.2$ |
| $a_{w,equ} < a_w < a_{w,in}$ | $0.88 < a_w < 0.98$ |

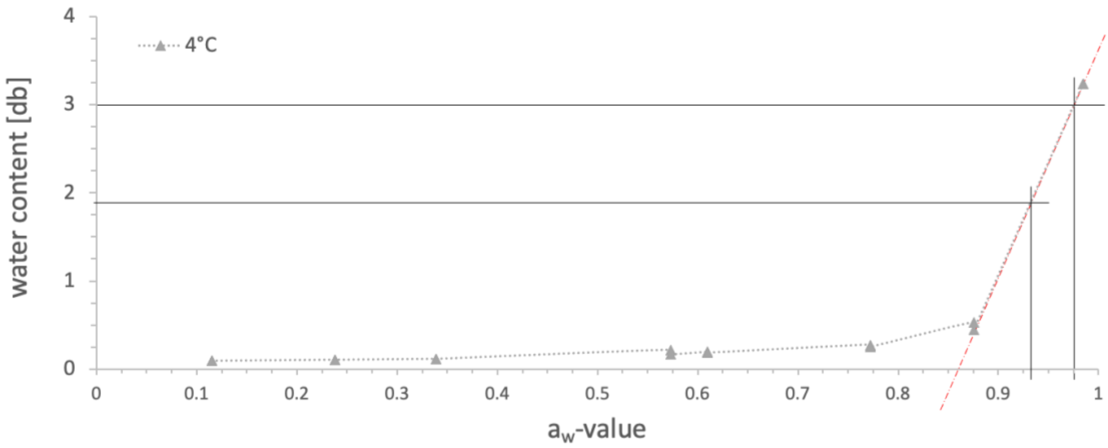


*Figure 7-3: Boundary conditions for the linear model.*

#### *7.3.1.2 GAB model*

The required minimum transmission rate based on the GAB model was estimated by solving the equation 7.2-11 by numerical integration using Euler's method with a steps size of 180 sec (Figure 7-4). The critical moisture content set to 10% weight loss was reached after 21 days when the water vapour permeability is equal to $1.05x10^{-12}$ mol m/m$^2$sPa. This corresponds to a film water transmission rate of 95.3 g/m$^2$ day. When considering a shelf life of 4 weeks, the required WVTR is 77.2 g/m$^2$ days.

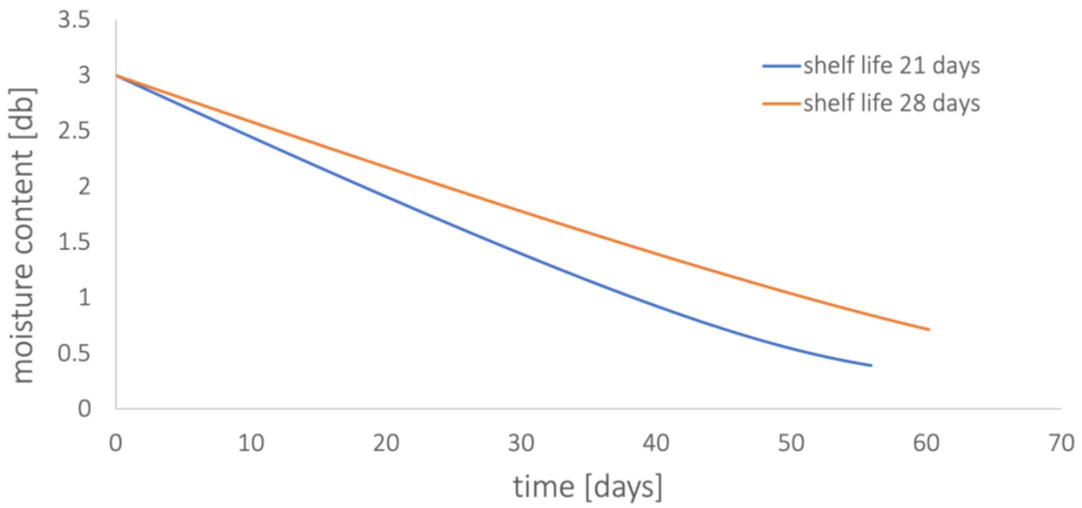


*Figure 7-4: Plot of numerically predicted moisture content [Kg water/Kg solids] of baked fish product in a permeable package versus storage time [days] using Euler's method.*

### 7.3.2 Determining the WVTR of the film materials

To determine the barrier requirements of existing food products the barrier properties can be measured directly. Ideally, the entire food package with the product needs to be tested, however this method is resource and time intensive especially at a developing stage. Therefore, it is often more useful to test the barrier properties of a film section under the simulated storage conditions. The water vapour transmission rate of four films was predicted at 4 and 14°C, under different RH conditions. The film transmission rate especially for biobased materials can be affected by the storage temperature (Macedo, 2011). For the selection of a suitable film material the WVTR was tested at cooled storage conditions and the results are shown in Table 7-6. The table was extended with WVTR rate values provided by the supplier obtained under standard conditions at 38°C and 90% RH, and 25°C and 75% RH. The results show that three of the selected materials provide the required WVTR rate, namely the monolayer PP film, the RK+ and NK film. The NVS film based on cellulose with a coating has a high-water vapour transmission and is not suitable for a baked fish product. All other tested films provide a WVTR below the calculated minimum barrier requirements. The transmission rate of the RK+ film showed a slight reduction in permeability under lower temperatures with 4.5 g/m$^2$ day at 38°C and 3.5 at 4°C at RH gradient from 90 to 0%. The NK film, based on cellulose with a high-water barrier coating showed an increased WVTR value with decreasing temperature from 20 to 72.1 g/m$^2$ day at 38°C to 4°C at 90% RH.

Table 7-6: Water vapour transmission rate [g/m$^2$ day] of four film materials at defined temperature and relative humidity's.

| Temperature [°C] | $RH_{out}$ [%] | $RH_{in}$ [%] | $WVTR_{BOPP}$ | $WVTR_{RK+}$ | $WVTR_{NK}$ | $WVTR_{NVS}$ |
|---|---|---|---|---|---|---|
| 38 | 90 | 0 | 3.7* | 4.5* | 20* | 600.0* |
| 25 | 75 | 0 | - | 0.7* | - | 200* |
| 14 | 90 | 0 | 2.5 | 2.6 | 45.3 | 284.5 |
| 14 | 90 | 75 | - | 6.2 | - | 190.2 |
| 14 | 75 | 0 | 2.8 | 1.4 | 24.3 | 280.1 |
| 4 | 90 | 0 | 3.9 | 3.5 | 72.1 | 570.5 |
| 4 | 90 | 75 | - | 3.9 | - | 643.8 |
| 4 | 75 | 0 | 2.8 | 24.3 | - | 371.1 |

*The table was extended with values provides by the film suppliers tested under standard conditions according to ASTM E96, ASTM F1770

### 7.3.3 Environmental consideration

The first priority of the food packaging selection should be the product safety and protection to avoid food waste. The environmental carbon footprint of fish products can be as high as 31 $CO_2$-eq./Kg product (Almeida et al., 2021), emphasizing the importance that products are used as their full potential as source of nutrients. Another important consideration for the selection of packaging film materials are environmental aspects. The aim should be to minimize the environmental burden and aligning to circular economy principles. Therefore, different environmental aspects of the film materials were summarized in Table 7-7 and evaluated.

The carbon footprint of the materials varies from 3.1 to 5.05 Kg $CO_2$ eq./Kg film, with the lowest value for the mono material polypropylene based film. The applied edible coating material showed only a marginal environmental impact. The carbon footprint of 1 Kg coating formulation based on chitosan coating developed by Bremenkamp et al. (2023) was estimated to be 0.84 Kg $CO_2$ eq./Kg coating, whereby for 1 Kg coated product the impact is 0.032 Kg $CO_2$ eq./Kg product (Uxía Vázquez, 2022). The PP and RK+ films are based on fossil raw materials, whereby the NK and NVS films are based on biobased resources. Biobased resources are renewable, and the material can stay in the same material cycle when composting of the material is ensured. The end of life options for the PP monolayer films are recycling or incineration and for the RK+ multi-layer film it is only incineration. PVdC coating is an additional environmental burden due to toxic component development during incineration (Yasuhara et al., 2005) and should be avoided when necessary. The recyclability of a multi-layer material depends on selected laminated layers. Some materials are compatible, either due to possible separation within the recycling process or similar material structures (Tullo, 2016). In general, materials based on fewer components generates a high quality recyclate.

Table 7-7: Environmental summary of the investigated film materials.

| Name | Composition | Layers | Biomass content [%] | Carbon footprint [Kg $CO_2$ eq./Kg film] * | End of Life (theoretical) |
|---|---|---|---|---|---|
| BOPP | PP | Mono | 0 | 3.1 | recyclable |
| RK+ | PVdC coating<br>OPP<br>PVdC coating | Multi | 0 | - | - |
| NK | Coating*5<br>Cellulose<br>Coating*5 | Multi | 90 | 5.05 | Industrial compostable*2<br>Home compostable *3<br>Anaerobic digestion*4 |
| NVS | Coating*5<br>Cellulose<br>Coating*5 | Multi | 89% | 4.9 | Industrial compostable *2<br>Home compostable *3<br>Anaerobic digestion *4 |

*based on the supply information (Futamura), peer reviewed LCA 2019, GaBi software Impact 2002+, Global warming 500 yr – midpoint method, *1evaluated based on sorting, and recycling facilities in Ireland *2 EN 13432, EN 14995, AS4736, ASTM D6400, ISO 17088, *3 AS5810, *4 ISO 15985 *5 composition is unknown

Nevertheless, it should be distinguished between theoretical and actual food packaging end of life scenarios. The actual food packaging waste life cycle can vary significantly between countries and depends on available waste collection, sorting and recycling facilities. A view on the Irish packaging waste generation showed that, in the EU Ireland is one of the larger waste producers per person (EPA, 2021a). The shares of the packaging waste in declining order are paper & cardboard, plastic, glass, wood, metals, and others. In 2019, 62% of the packaging waste was recycled, 33% were incinerated with energy recovery, and 5% end up in landfill. Big differences in the rate of recycling across the material shares were reported, with the lowest recycling rate for plastics (Dwyer, 2021; EPA, 2021b, 2022). 319 082 t of plastic packaging waste were generated in 2019, and only 28% were recycled. Whereby, 5.7% were processed in Ireland, 76.3% in other EU member states and 18% outside the EU (EPA, 2022; REPAK, 2020). The remaining plastic waste of the total plastic packaging generated was mainly incinerated for energy use (69%) and a minor part ended up in landfills (3%) (EPA, 2021b). The low recycling rate of plastics can be related to the collection system in place, as until 2021 only clean and dry rigid plastics were allowed in the recycling bins, and therefore considered for recycling. Plastic films and all plastics with product contaminations were placed into the general household bin, which is directly send to incineration or landfill without any sorting step (mywaste). Improvements have been made in the Irish sorting facilities with state of art optical sorting equipment that can identify different plastic types based on reflection and refraction of light beams, allowing also soft plastics in recycling bin since September 2021 (Figure 7-5). Guidelines for design for recycling gives detailed packaging design recommendations to improve the recyclability of plastic packaging for a specific country or region, for Ireland the DfR Guideline of REPAK (2020) is a useful reference.

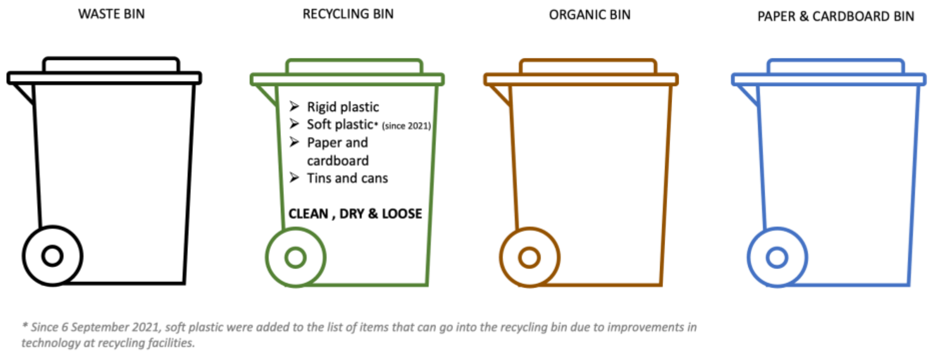


*Figure 7-5: Comment recycling bins in Ireland.*

Considering the environmental considerations of the listed film materials, the use of a biobased compostable material, as the NatureFlex$^{TM}$ NK film, could be an interesting eco-solution for products sold in Ireland. With the technical improvement of the available waste sorting system in Ireland, also NIR separable mono-material films could be an option. Nevertheless, here it needs to be guaranteed that the often-contaminated films with products end up in the recycling bin and therefore are considered for sorting for recycling.

## 7.4 Conclusion

A packaging film material was selected, for a synergetic packaging consisting of an edible coating and a film layer. The film material selection was performed based on product requirements, and environmental considerations. A mathematical approach was studied for modelling the required water barrier properties for controlling the water loss of a RtE baked fish product for a defined storage time. The results showed that a WVTR of around 81-95 g/m$^2$ day based on the GAB model approach and linear model approach is needed to stay below the accepted water loss within 21 days. Extending the shelf life to 28 or even 35 days gives a minimum allowed WVTR of 48-56 g/m$^2$ day. As the WVTR of the film materials is affected by storage temperature and relative humidity, the WVTR of the selected films were measured under realistic storage conditions for a RtE seafood product. It was seen that the WVTR for biobased film materials is affected by the storage temperature, highlighting the necessity of performing barrier measurements under realistic storage scenario before selecting a suitable film packaging material. The tested OPP, Futamure RK+ and NatureFlex$^{TM}$ NK film are fulfilling the required water vapour barrier properties. Based on the environmental consideration the biobased, compostable NatureFlex$^{TM}$ NK film is recommended for RtE coated products sold on the Irish market.

Chapter EIGHT

# Final Conclusion and Recommendation

## 8.1 General Conclusion

Food packaging offers numerous advantages that contribute to safety, quality, preservation, and convenience of food products. Besides, offering a range of benefits, food packaging also comes with certain challenges. The packaging industry is under pressure to reduce the dependency on fossil resources and finding solutions for the significant amounts of generated food packaging waste, to reduce the environmental impact of food packaging. Special emphasis in this work has been given to sustainable packaging developments for ready to eat (RtE) healthy and nutritious food products. The research carried out emphasises the importance of understanding the food life cycle to design complementary tailored packaging. Commercial RtE seafood products are frequently packed in primary packaging using synthetic plastic materials which fulfil necessary packaging properties. Nevertheless, the dependency on fossil resources and the end of life option of these packaging materials is challenging. This work exploited an alternative packaging strategy, by shifting the required packaging properties to two separated packaging layers, a biobased edible coating directly applied to the product surface in combination with a film layer.

Life cycle assessment (LCA) serves as a decision-making tool to enhance the environmental performance of packaging. The success of new packaging development lies in its ability to minimize the environmental impact of a current food-packaging system while maintaining core requirements such as product protection, transportation, and communication. A systematic investigation showed the variability in approaches taken to assess packaging's environmental performances. A) Packaging can be evaluated as an integrated part of the food supply chain, revealing its environmental contribution to the overall food-packaging impact. In the case for high impact food products as RtE seafood products, the packaging share of the overall impact was small, with the exception for products packed in heavy materials. This highlights the significance of preventing waste and ensuring products are consumed, preventing the worst-case scenario of unused packed food waste. However, it is important to recognize that the cumulative impact of all packaging units can be substantial, necessitating careful consideration. B) Consequential LCA of packaging materials is a valuable approach to compare materials, such as comparing mono-materials with multilayer options. Either comparing direct impact categories, or additionally taken indirect effects into account, for more accurate outcome. C) Comparisons of different packaging systems often include both direct and indirect environmental impacts. Although quantifying the relationship between food packaging and the broader food supply chain can be complex, this approach provides a comprehensive perspective. Studies targeting multiply indirect impacts are rarer compared to those focusing on singular indirect impacts. D) Reviewing LCA studies on novel packaging systems revealed that when solely considering direct impacts, novel systems sometimes exhibit higher environmental footprints. However, when factoring in direct impacts, especially those tied to waste reduction due to extended shelf life, the advantages tend to favour novel packaging. In summary, the diverse LCA approaches adopted serve specific goals, and when seen as sequential steps, they provide a holistic understanding of the environmental impact of novel food packaging. A comprehensive grasp of impacts and benefits is crucial for successful and sustainable packaging innovations.

The growing consumer demand for healthy, safe, convenient, and sustainable food products has led to the exploration of edible coatings for RtE seafood products. These coatings, combined with film packaging, show promise in enhancing product safety and extending shelf life. While most studies have focused on microbiological control, particularly *Listeria monocytogenes*, there is potential to broaden research to address other degradation processes. Edible coatings offer sustainability benefits due to minimal waste generation, biobased composition, and shelf life extension. LCAs could further underscore their positive environmental impact. However, challenges of commercialising of edible coatings include sensory aspects, regulatory compliance, labelling, and consumer acceptance. Market opportunities lie in customization and personalization. Coating properties can be tailored using various methods, but balancing protection and sensory aspects is a challenge. Exploring different application techniques, such as spraying, could improve scalability. Standardized testing methods for coatings are needed to improve comparability between studies. In the RtE food sector, edible coatings are gaining traction with a focus on sustainability and customization. Overcoming challenges through research and development is crucial to ensure compatibility, performance optimization, regulatory adherence, and cost-effectiveness. Developing edible coatings for RtE seafood involves multidisciplinary efforts to address technological, engineering, environmental, and consumer factors. Integrating edible coatings into a comprehensive RtE seafood product-packaging approach holds significant potential to address challenges and maximize benefits.

Data on the degradation process of two RtE seafood products, an RtE baked fish product and an RtE minced fish patty, were generated to define critical quality parameters and predict the product shelf life. The moisture sorption behaviour of a RtE baked fish product was studied through sorption isotherm analysis. The research utilized a static gravimetric method across a range of water activity levels (0.12 to 0.98) and explored the impact of storage temperatures (4°C and 14°C). Different mathematical models were tested to establish the relation between equilibrium moisture content and water activity. The sorption isotherm curves of a RtE seafood product were found to follow a type II isotherm shape. The sorption isotherms could mathematically be modelled by Peleg and GAB model with a high conformity. The microbiological degradation, followed by water loss was critical for both tested products. Whereby, the fat oxidation is only critical for products with a higher lipid level (≥ 6%). Shelf life predictions ranged from 7 to 14 days for the RtE baked fish product, primarily limited by microbial growth, while the shelf life of RtE fish patty was predicted to be under 7 days due to fat oxidation. Evaluating the potential supply chain indicated a minimum required shelf life of 21 days for a RtE seafood product. Consequently, packaging attributes like oxygen, light, and water vapour barriers, along with antimicrobial properties, were identified as crucial for ensuring product safety and quality. These findings provide a foundational understanding an specialized food packaging concept that maintain product integrity.

Edible coatings made from biobased materials are environmentally friendly packaging options that can control food product degradation, and improve food safety and quality. This study examined chitosan and alginate-based coatings to control fat oxidation, water loss, and microbial growth in a RtE baked fish product. The research assessed the influence of coating composition on performance and analysed how coatings affected product shelf life under optimal and abused storage conditions. Results indicated that coating composition significantly impacted performance, with an optimal chitosan composition identified as 1% (w/v) chitosan, in 1% (v/v) acetic acid solution with 15% (w/w chitosan) glycerol, and for the alginate a coating solution with 1% (w/v) sodium alginate showed the best performance. Both chitosan and alginate coatings, applied individually or in bi-layers, formed invisible layers around the product. While the tested alginate coatings had limited effects, the chitosan coating substantially reduced microbial growth and showed an effect on the weight loss during storage, regardless of temperature conditions. A shelf life extension of chitosan coated samples compared to uncoated control sample stored at optimal temperature of 4°C could be achieved from 7 days to 21 days, and under abused conditions from 2 to 7 days. An effect of tested coatings at optimal and abused storage conditions could be seen, and it is therefore recommended using an edible coating as a safety measure against temperature abused during the food supply chain. Overall, the study highlights the potential of using an edible chitosan coating as a packaging solution to enhance safety and quality for RtE seafood products, even in challenging storage conditions.

This study emphasized the significance of employing hurdle technologies to support innovative approaches in the field of post-harvest processing. By combining novel technologies, advantages can be gained in terms of enhancing food safety and quality while reducing food waste. Specifically, the research investigated the synergistic effects of applying cold plasma treatment and an equilibrium modified atmosphere packaging (EMAP) for increasing the shelf life of cherry tomatoes stored at cold and ambient temperature. The study's objective was to examine how the combination of plasma-washing treatment and EMAP packaging influenced the quality characteristics of cherry tomatoes during storage, with a focus on microbiological quality and overall quality preservation. Various parameters were monitored to assess cherry tomato quality, including weight loss, soluble solid content, colour, pH, and microbial loads. Results highlighted the importance of the cold plasma and EMAP combination in controlling the quality of RtE cherry tomatoes. It was observed that the combination of cold plasma and EMAP design was important to ensure the quality of RtE cherry tomatoes. Cold plasma was identified as a great alternative to the common applied washing step with chemicals. In combination with an EMAP the shelf life was extended to 32 days and 11 days when stored at 10°C and 20°C based on the best before date of the supplier. This approach not only maintained quality characteristics but also reduced the reliance on cold storage, representing a promising advancement in fresh produce preservation techniques.

In the pursuit of creating a comprehensive packaging concept, a packaging film material was chosen for a synergistic packaging approach, combining an edible coating and film layer.

This selection process factored in both product-specific needs and environmental concerns. A mathematical methodology was employed to model the necessary water barrier properties for effectively managing water loss in a RtE baked fish product within a specified storage duration. The results indicated that a water vapour transmission rate (WVTR) of approximately 81-95 $g/m^2$ day, as determined by both the GAB model and linear model approaches, is necessary to keep water loss under acceptable levels over a 21 day storage period. Extending the product's shelf life to 28 or 35 days further mandates a minimum WVTR of 48-56 $g/m^2$ day. The WVTR of film materials can be influenced by storage temperature and relative humidity, emphasizing the need to measure barrier properties under realistic storage conditions before selecting a suitable film material. Consequently, the WVTR of selected films was assessed under practical storage scenarios for a RtE seafood product. The findings underscored the impact of storage temperature on biobased film materials, highlighting the importance of evaluating barrier measurements in realistic conditions. Among the tested films, polypropylene, Futamura RK$^+$, and NatureFlex$^{TM}$ NK films met the required water vapor barrier criteria. While considering environmental factors, the biobased and compostable NatureFlex$^{TM}$ NK film was recommended for RtE coated products targeting the Irish market.

Results of this work indicated that it is highly recommended to investigate novel food packaging approaches to introduce new food packaging perspectives. A systematic quantitative approach is recommended to develop a synergetic tailored packaging for a specific food product. A rethinking of packaging from being hardly degradable to be easily degradable, visible to invisible, a threat to nature to being part of a natural life cycle should be driving new packaging developments.

## 8.2 Recommendation

The next step in the field of edible coatings for RtE seafood products should definitely be to broaden the study field, by combining food science, food technology, and packaging engineering aspects.

Edible coatings were selected based on the effect of controlling three critical quality parameters, microbiological growth, water loss and fat oxidation. The appearance was assessed visually with no discoverable differences. Nevertheless, it is recommended to consider sensory aspects, study consumer acceptance and food declaration.

A positive effect of chitosan coating regarding storage temperature was identified. Further studies could also be performed to validate the shelf life under dynamic and more realistic scenarios by simulating the distribution chain, and therefore gain more understanding about the enhanced product robustness to temperature changes with a chitosan coating.

The developed edible coating mainly prolonged the product shelf life by increasing the product safety, but the effect on the water loss and fat oxidation was limited. It would be interesting to further develop the coating by testing other materials or using the layer-by-layer or blending technique for combining properties of natural coating materials. An edible coating that can control multiple degradation processes would provide a more robust product protection and would even further reduce the requirements for the outside layer.

Other recommendation, could be to model food packaging to include an edible coating and an outside layer. How to select an outside packaging for a coated product, without overprotecting the product? Edible coatings for RtE seafood product showed high potential and other recommendations could involve further studies with other RtE food products using edible coating underpinning the positive findings of our study. Research looking into necessary steps and research gaps for upscaling from experimental level to pilot or industrial applications is also necessary. Here, it is further recommended to perform a LCA study reflecting upon a holistic view, elaborating, and proving the environmental advantages of a packaging system based on an edible coating and outside layer.

Looking back on the history of food packaging, it shows how important food packaging developments were for our current food supply chain and food safety. Challenges at the time were overcome by advances in technologies, and novel developments. Highlighting the importance for research in the field of food packaging overcoming the challenges of our time. Rethinking packaging with an open mind, learning from other successful developments, being open to change our behaviour and cooperation between interdisciplinary supply chain drivers will be important.

Chapter NINE

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