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Highlights

- Molten metal pyrolysis recycles aluminium-laminated plastics.
- The polypropylene layer pyrolyses to waxes.
- A 4,000 t/y or larger plants is economic; 20% internal rate of return (IRR).

Recycling of aluminium laminated pouches and Tetra Pak cartons by molten metal pyrolysis – pilot-scale experiments and economic analysis

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Abstract

Aluminium laminated (AL) pouch packages and aluminium laminated Tetra-Pak cartons are considered unrecyclable, reducing their otherwise excellent lifecycle performance. This paper describes experimental results on pilot plant trials to recycle AL packages with a molten metal pyrolysis reactor. The experimental evidence shows that both package formats can be recycled and that clean aluminium can be recovered. However, the recovered aluminium from Al pouches may require mechanical cleaning as the consumer's information is printed onto the

aluminium, leaving a carbon residue on the recovered aluminium. On the other hand, over 90% of the polypropylene plastic layer on the AL packaging pyrolysed into waxes, pointing to excellent kinetics. Moreover, an economic analysis of a 4,000 t/y commercial-scale plant demonstrates that a molten metal AL recycling plant is economically viable, achieving an internal rate of return (IRR) of over 20%.

Keywords: composite recycling, aluminium recycling, extended user responsibility, commercial-scale plant

1. Introduction

The aluminium laminated (AL) packaging meets many of the packaging requirements of the food industry (Yam, 2009). AL packaging is inert, lightweight, tough and the aluminium layer with a thickness of 6-150 µm provides a life-long barrier against aroma loss. It also protects the food against ingress of moisture, air, microorganisms, UV light and other food spoiling agents (Kerry, 2012), resulting in an extending shelf life (Kerry, 2012; Nonclercq, 2016). These advantages resulted in annual AL packaging growth rates of 10-15%, and in 2015 about 190,000 tonnes of aluminium were consumed for its production in Europe alone (Slater and Crichton, 2011).

Two popular AL food packaging presentations are the AL plastic pouch and the flow wrap. The AL plastic pouch is often used for baby food, coffee, pet foods, pasta sauce and cereals, whereas the flow wrap is used for crisps, chocolate bars and other snack foods (Nonclercq, 2016). The typical AL plastic pouch is made from three primary materials or layers: (1) a polyethylene terephthalate (PET) layer of ca. 12 µm thickness also displaying packaging information on the outside, (2) an aluminium foil of ca. 7 µm thickness and (3) an inside layer of polyethylene (PE) with a thickness of ca. 75 µm providing food compatibility and other properties (Nonclercq, 2016). The typical AL flow wrap comprises a polypropylene (PP) layer of 35 µm

thickness providing strength and stiffness. The aluminium layer of 0.04 µm thickness is typically vapour deposited onto the PP plastic. A primer applied to the aluminium layer allows printing onto the aluminium (Nonclercq, 2016).

A more rigid AL packaging is the Tetra Pak carton due to the inclusion of a paperback layer. This package is layered from the inside to the outside with PP, aluminium foil, PP, paperback and, finally, PP with the package information printed on it (Website, 2021b). A Tetra Pak carton composition is typically 63 wt.% cardboard or paper, 30 wt.% plastic, and 7 wt.% aluminium (Korkmaz et al., 2009).

AL packaging also has lifecycle advantages over other packaging formats performing the same function. Many authors' life cycle assessments (LCA) (Bayus et al., 2016; Bukowski and Richmond, 2018; Nonclercq, 2016; Xie et al., 2011) have shown the superiority of AL packaging over AL packaging other packaging formats. An LCA output for an AL pouch compared to other packaging formats, given in Table S1 in the supplemental information (Bukowski and Richmond, 2018), shows the superiority of AL packaging over different designs.

The major disadvantage of AL packaging is that it is considered unrecyclable (Horodytska et al., 2018; Slater and Crichton, 2011; Velzen et al., March 2020), which in Europe is often indicated on the packaging by the sign "currently not recycled" i.e., the material is landfilled. But if AL packaging was recyclable, its LCA could be improved even further (Bayus et al., 2016; Bukowski and Richmond, 2018; Nonclercq, 2016; Xie et al., 2011). Especially the recyclability of aluminium could have a significant positive impact, as the recycling of aluminium from secondary sources reduces the energy consumption compared to its production from ore by 95% (Warrings and Fellner, 2018). Moreover, aluminium can be recycled repeatedly without losing its properties, making it an ideal circular economy material (European Aluminium, 2020).

The literature describes various methods to recycle AL packaging, of which none is currently employed on an industrial scale. Fávaro et al. (Fávaro et al., 2013) studied the possibility of delaminating AL packages with solvents to remove the plastic layers. Under supercritical conditions of 255°C and a pressure of 11.65 MPa, metallic aluminium was obtained. Potential drawbacks of this process are the high operating pressures and solvent use, which in combination may result in an expensive process. Mu'min et al. (Mu'min et al., 2017) investigated wet torrefaction to recycle aluminium from AL packaging waste. Again, high operating pressures may result in an expensive process.

Pyrolysis has long been identified as an ideal process to recycle mixed plastics (Czajczyńska et al., 2017; López et al., 2010; Ragaert et al., 2017; Sharuddin et al., 2016; Siddiqui and Redhwi, 2009; Singh et al., 2017). Plastic pyrolysis is a depolymerisation process executed at temperatures above 400°C, in an oxygen-free environment and typically at ambient pressures (Boateng, 2008). Products of mixed plastic pyrolysis are pyrolysis oil (ca. 80-95%), pyrolytic carbon (ca. 1-5%; also referred to as char or ash in the literature) and gases (5-15%) depending on pyrolysis temperature, catalyst, and the mixed plastic composition (López et al., 2010; Sharuddin et al., 2016; Siddiqui and Redhwi, 2009). The pyrolysis oil may be upgraded to diesel or other petroleum products, the char would be landfilled, and the gases (methane, propane etc.) would heat the process. The capability of pyrolysis to treat mixed plastics, i.e., PET, PE and PP, is important in AL packaging recycling as these plastics are present in AL packaging. Moreover, because it is difficult to obtain a clean AL packaging waste stream from municipal waste without other plastics, this advantage of pyrolysis becomes ever more critical.

Furthermore, pyrolysis can also treat contaminated plastics, e.g., plastic packaging with food residues, paper, ink and other contaminants. Irawan et al. (Irawan et al., 2018) pyrolysed AL packaging in a conventional pyrolysis reactor (Boateng, 2008). Their study found that the recovered aluminium could be separated from the char easily. A potential drawback of conventional rotary kiln pyrolysis reactors is their relatively poor kinetics compared to directly

heated reactors (Riedewald et al., 2021). Furthermore, increasing operating temperatures result in decreased char yields, whereas the aluminium yield remains the same. The possibility of recycling AL packaging with microwave-induced pyrolysis was investigated by Palafox and Chase on a bench-scale reactor (Ludlow-Palafox and Chase, 2001), whereas Slater and Crichton (Slater and Crichton, 2011) report on experiments performed on a larger scale. At both scales, clean aluminium together with hydrocarbons from the pyrolysis of the plastics was obtained. Microwave reactors have the advantage over rotary kilns in that they are directly heated reactors. Still, they have the drawback of using a microwave generator – a potentially expensive piece of equipment. Moreover, the non-condensable gases cannot be combusted for internal heat generation. Instead, they would have to be combusted in an electrical generator, generating electricity to drive the microwave generator. The capital and operational costs of generating electricity in this fashion may, however, be prohibitive.

The capability of pyrolysis to treat paper, plastic, and aluminium is also crucial for recycling Tetra Pak cartons. Korkmaz et al. (Korkmaz et al., 2009) pyrolysed 1 to 2 cm large Tetra Pak pieces in a laboratory-scale tube reactor. While the yields, gas, and wax composition are reported, no information is conveyed to separate the aluminium and carbon. Robertson (Robertson, 2021) reviewed various Tetra Pak recycling methods, including pyrolysis, whereas Zawadiak et al. (Zawadiak et al., 2017) provide an overview of the current techniques of Tetra Pak recycling. In summary, pyrolysis can recycle both AL and Tetra Pak packaging formats. One of the advantages of pyrolysis is that pyrolysis can operate with residual food contamination still present on the packaging (Slater and Crichton, 2011).

This paper proposes pyrolysing AL and Tetra Pak packaging by direct heat transfer with molten metal resulting in a rapid process already shown on whole tyres (Rathsack et al., 2015), which were fully pyrolysed within 15 minutes - significantly faster than processing tyres in indirectly heated rotary kilns or similar reactors. This demonstrates the advantages of direct heat transfer or, in this case, processing on molten metals. In this paper, Al plastic pouches and Tetra-Pak

aluminium laminated packages were treated in a pilot plant. An AL recycling process must be capable of treating both of these packaging types, as they represent the vast majority of aluminium laminated food packages. Moreover, in practice, separation of these two packages types may not always be possible.

The molten metal pyrolysis reactor does not have the disadvantages of other reactor types. First, the non-condensables can be used for internal heat generation, resulting in a self-sustaining process. Second, the molten metal reactor is a directly heated reactor i.e., the AL packages are in direct contact with the molten metal resulting in a fast process. Finally, this paper presents an economic analysis of a future commercial-scale AL recycling process named ALPyro although only limited design and economic data are available. Such a financial analysis is essential to obtain investment for a demonstration plant phase before commercialisation of the process can commence. But significant amounts of private capital (about €2-4 million depending on the size of the demonstration unit and other parameters) will be required, and a private investor will need to know the potential financial returns of this process, which the economic analysis provides.

2. Materials and Methods

2.1. Methods

Fig. 1 presents the process and instrumentation diagram of the multipurpose molten metal pyrolysis experimental setup. Various inner containers can be used, as well as various molten metals allowing the processing of various materials and in-situ separation. A 6" 316 L stainless-steel container was filled with 1,300 g of tin, equating to a molten tin level of 1 cm. This container was placed inside the pyrolysis reactor, which operates in batch mode. The pyrolysis reactor was manufactured from an 8" (125 mm) ANSI schedule 10, 316L stainless steel pipe and a length resulting in a vessel height of 300 mm. A 3 mm thick 316L stainless steel plate was

the bottom wall of the vessel. By unbolting the top flange, the vessel can be opened. All interconnecting pipes were ½" Swagelok tubes. The pyrolysis vapours were condensed by a 30 cm long ambient temperature water-cooled ½" tube. The condenser was sloped by 20 degrees draining into a condensate collection vessel. The formation of an explosive atmosphere within the pyrolysis vessel vapour space and downstream equipment was prevented by initially nitrogen inerting the equipment and continuously sweeping the pyrolysis vessel with a nitrogen flow of ca. 120 liters per hour during the experiment. The operating pressure of the entire system was slightly above atmospheric (~10 mbar), preventing air ingress due to the hydrostatic pressure induced by the bubbler. The space between the reactor walls and the container was filled with 2,600 g of LiCl-KCl salt providing heat transfer between the heating pads and the container.

Three ceramic electrical heating pads heated the reactor. The vessel was insulated with 100 mm thick glass wool. But the top flange and the vessel sides below 80 mm of the flange were not insulated. An Ashcroft temperature gauge (range of 0 to 500ºC, with a 1 % ASME B40.3, Grade A accuracy inserted into a thermowell manufactured from 316L stainless steel) was used to measure the operating temperature. The operating temperature was controlled to an estimated ±5°C by a control system (Stork Copperheat 50 KVA Heat Treatment Module, Model no. 16050). The temperature feedback to the Heat Treatment Module control system was provided by a thermocouple located between the outside surface of the reactor and one of the heating pads (denoted TIC in Fig. 1).

The ½" pipe connecting the reactor to the condenser was also insulated. The preferred pipe connections were metal to metal fittings (Swagelok) as they are vapour tight and can be opened for inspection.

For the experiments, the operating temperature was 450°C as such a temperature appears to be ideal in maximising the yield of pyrolysis oil or wax from plastic pyrolysis (López et al., 2010;

Sharuddin et al., 2016; Siddiqui and Redhwi, 2009), one of the revenue streams for an operator. Once the operating temperature of 450°C was reached, it was maintained for 60 minutes to ensure the pyrolysis reaction completes. On reaching this point, the heat was turned off, and the equipment cooled naturally to ambient. Once the reactor cooled down to 100°C, the nitrogen flow was switched off. Finally, the reactor was opened, and the pyrolysed material in the container was removed for analysis.

The condensate collection vessel was a 100 mL laboratory glass bottle. The interceptor vessel was a 1,000 mL laboratory glass bottle (Fig. 4). The bubbler was a 250 mL laboratory glass bottle filled with 100 mL of water to make the flow of nitrogen through the experiment visible and to provide a small amount of overpressure throughout the experiment preventing air ingress.

A Sartorius ED4202S laboratory scale (max 4,200 g; d = 0.01 g) was used to weigh the various materials.

The temperature gauge located after the condenser was a GenWare electric temperature gauge with a range of -40 to 230°C. The temperature of the top flange and the area below the top flange was measured with a GenWare infrared-thermometer having a temperature range from -32 to 550°C and a 0.5 second response time.

A carbon adsorber, a drum filled with 25 kg activated carbon supplied by Silcarbon Aktivkohle GmbH, Germany, abated the non-condensable vapours from the experiment before discharging the gas stream to the air extract system of the building.

After each experiment, the reactor, pipes and condenser were visually inspected for evidence of wax or other deposits, but none were found.

2.2. Materials

Two different AL packages were used. One waste package was Tetra Pak (Fig. 2). The Tetra Pak carton was cut into five pieces to fit the reactor, as shown in Fig. 2, but the Sugarcane (Website, 2021a) top and screw cap were removed and discharged as this is not present on all Tetra Pak cartons. The other packages were coffee packages from Cork Coffee Roasters and Whiskas cat food pouches (Fig. 3); both packages were washed and dried.

The tin was 99.95% pure, presented in 3-5 mm lumps, and was purchased from Amazon. 316L stainless steel is a suitable material of construction for molten tin for the temperature ranges of these experiments (Maaß and Peißker, 2011).

The heat transfer salt was a eutectic mixture of technical grade lithium chloride (LiCl) and potassium chloride (KCl) salts (41.8 mol% KCl and 58.2 mol% LiCl (LiCl-KCl)). LiCl-KCl salt was chosen for these experiments, as it is stable, non-toxic and inert at the operating temperatures (Sohal et al., March 2010). Additionally, 316L stainless steel is a suitable material of construction for molten LiCl-KCl (Ignatiev and Surenkov, 2012). Leverton Clarke, Basingstoke, United Kingdom, supplied the LiCl. Scientific & Chemical Supplies Ltd, Cork, Ireland, provided the KCl.

The nitrogen was certified 99.999 % pure.

3. Results and Discussions

3.1. Experimental results

The emphasis of this research was on the solid residue, i.e., the aluminium and carbon from the AL and Tetra Pak packages remaining in the reactor. The pyrolysis oil, waxes and gases generated were of secondary importance, as the composition, yields, and optimum operating

temperatures are known for (1) PE, PP and PET (Brems et al., 2011; Honus et al., 2018a; Honus et al., 2018b; López et al., 2010; Sharuddin et al., 2016; Siddiqui and Redhwi, 2009), (2) paper (Li et al., 2005; Ma et al., 2017; Rutkowski, 2013; Zhou et al., 2013) and (3) mixtures of paper and plastics (Fekhar et al., 2020; Johansson et al., 2018).

The experiments on Tetra Pak cartons demonstrate that aluminium recycling from a Tetra Pak carton is straightforward. The aluminium can be recovered clean, as shown in Fig. 2. The black material is pyrolytic carbon from the pyrolysis of the paperback. The pyrolytic carbon does not adhere to the aluminium foil. Moreover, it is very brittle, making the separation of the pyrolytic carbon from the aluminium easy to do in a follow-on operation. The yield of pyrolysis carbon from the pyrolysis of the plastics, on the other hand, is negligible, as the yield of pyrolytic carbon from PE, PP and PET at 450°C is between 0-3% (Aguado et al., 2002; López et al., 2010).

The experiments on the AL pouch packages also demonstrate that the aluminium can be recycled with the molten metal process. But, as shown in Fig. 3, the recovered aluminium foil is not as clean as the aluminium from the Tetra Pak's. This is because the consumer's information is printed onto the aluminium, leaving a carbon residue from the print on the aluminium. As a result, a mechanical cleaning step may be required to remove the carbon residue from the recovered aluminium foil. Such a mechanical cleaning step, if necessary, appears feasible, as the carbon residue can be wiped off quite easily. In contrast, the side of the aluminium foil forming the inside of the package is completely clean. It is clean, as the pyrolysis of PE, PP or PET only results in negligible amounts of carbon (Aguado et al., 2002; López et al., 2010).
Another option to ease the removal of the carbon residue from the aluminium would be for the manufacturer to change the formulation of the paint, which would be feasible in the context of a recycling/circular economy framework.
As already stated in the introduction, the aluminium layer may be of various thicknesses and grades (Slater and Crichton, 2011). Hence in practice, the recovered aluminium would need to

be bailed and send to an aluminium smelter for re-melting closing the recycling loop (Schlesinger, 2017).

The pyrolysis of the PP plastic layers of the Tetra Pak and the PET and PE of the AL plastic resulted in waxes rather than oils. This result is consistent with the literature stating that at a pyrolysis temperature of 450°C, PP generates up to 92% waxes (Aguado et al., 2002; Honus et al., 2018a; Honus et al., 2018b; Korkmaz et al., 2009). This must be considered for the design of the experiment and for a full-scale plant as waxes, should they solidify have the potential to block the condenser (Park et al., 2002) or other pieces of equipment resulting in a high-pressure event causing safety concerns.

The experimental setup (Fig.1) included a glass interceptor vessel giving a visual impression of the state of the vapour stream after the condenser. The fluid velocity reduces within this vessel, allowing wax particles to sink to the bottom, visible by the top section of the glass vessel being clear of fog, as shown in Fig. 4.

A sharp 90-degree bend at the entrance to the interceptor vessel caused wax particles suspended in the nitrogen flow to collide with the wall and collect there. Intermittently the waxes blocked the gas flow, the pressure would slowly rise to about 150 mbar, which would blow the waxes into the interceptor vessel (Fig. 4, right), and the process would start again. This wax collection and blowout took place three times within about 5 minutes, and one might speculate that the plastic pyrolysis reaction took place during that timeframe indicating fast kinetics. But, if the kinetics are indeed that fast, it must be proven by a follow-on experiment designed to ascertain the kinetics of the molten metal process, which could, for instance, be executed on a future demonstrator scale unit.

Waxes also condensed on the inside surfaces of the vessel, i.e., on the gasket below the top flange (Fig. 1). The outside surface temperature of the top flange was about 130°C; hence only the longer chain waxes condensed there. After each experiment, a visual inspection of the condenser and the pipes found no build-up of waxes or other materials there.

A wax mass balance suggests that the heating of the plastic was rapid as virtually all of the plastic converted into waxes, confirming the wax yields of others, i.e., over 90% (Aguado et al., 2002; Honus et al., 2018a; Honus et al., 2018b; Korkmaz et al., 2009). Secondary reactions, i.e., cracking of waxes into oil and gases, did not occur as high surface temperatures, high-temperature hot spots, or long residence times are avoided by the molten metal reactor. Hence, the results of others on plastic pyrolysis (Honus et al., 2018a; Honus et al., 2018b; Korkmaz et al., 2009) are valid to design a full-scale molten metal pyrolysis recycling plant and were used for the economic analysis, which will be presented in the next section.

3.2. Economic analysis of a full-scale plant

Performing an economic analysis at this process development stage is difficult as not more than 5% of the engineering is completed. Therefore, the accuracy of the cost estimate is -20 to +30% (AACE, 2005). Performing such an economic analysis at this stage of the process development is essential for the reasons given in the introduction.

3.2.1 Description of the full-scale plant

The ALPyro process is a continuous recycling process and is shown schematically in Fig. 5. The waste material is in direct contact with the molten metal. Zinc rather than tin is used, as it is more cost-effective. But by using zinc, the operating temperature must be at least 450°C, as the melting point of zinc is 419°C. However, an operating temperature of 450°C maximises the yield of pyrolysis oil, a revenue stream, as stated above. All equipment subject to condensation of waxes is kept at a temperature above 80°C allowing the waxes to remain liquid and drain off.

The AL waste is provided in a feeding bin and transported by a feeding screw into the pyrolysis reactor, i.e., onto molten zinc. On the molten zinc, the waste pyrolyses into hydrocarbon vapours, gases and solids. Any water present in the feedstock also evaporates. The solids, composed of aluminium and pyrolytic carbon, float on the molten zinc.

A discharge screw removes the recovered aluminium and the pyrolytic carbon into the recovered aluminium product hopper. The pyrolysis vapours are removed from the pyrolysis reactor via a vapour removal line and condensed to pyrolysis oil, waxes, or both. A fan provides the suction or driving force for the vapour removal. The non-condensable gases, i.e., methane, propane may be sent to the burners providing the heat for the pyrolysis process, ensuring that the process runs without any additional energy.

3.2.2 Economic analysis

The economics of an ALPyro plant is assessed by a number of financial measures such as the internal rate of return (IRR), the net present value (NPV) and the breakeven point. Of these measures, the IRR is the most helpful measure indicating the relative return of an investment without taking the scale of the project into account (Crundwell, 2008). In general, larger infrastructure projects such as waste plastic pyrolysis plants are considered profitable by private investors if the IRR exceeds 15% (Riedewald et al., 2021). Here a minimum IRR of 20% is demanded, as the financial uncertainty is still considerable. The evaluation period of the IRR and the NPV is ten years. The IRR of the ALPyro plant is 21%, the NPV €280,000, and the breakeven point is reached after 3.15 years. Table S3 in the supplemental material presents the business performance of the plant over 10 years, giving a much better and comprehensive impression of the financial performance of the ALPyro plant than any of the financial measures. The following explains how the most relevant factors influencing the financial performance of the plant were estimated.

The plant's location is assumed to be in Europe, close to significant areas of population such as London, Birmingham, Berlin or Amsterdam. The costs for a warehouse for temporary waste storage is not included. Instead, it is assumed that the plant is located close to a plastic sorting plant and that warehouse space exists. A storage capacity for 8 hours' worth of feedstock may be worth considering, but this is not included in the cost estimate. This surge capacity would be used when the AL recycling plant is idle and temporarily store AL waste delivered to the site from other waste sorting facilities.

Moreover, the AL recycling plant is part of a larger facility, meaning that management, engineering and other support is available. Further, it is assumed that AL pouch packages and Tetra Pak cartons are treated together.

Market: The AL recycling plant can only be sized if the addressable amount of waste AL is known. About 14,000 t/y of aluminium laminated plastic is collected in the UK and available for recycling (Slater and Crichton, 2011). But waste transport costs (Riedewald et al., 2021) will limit UK plant sizes to about 4,000 t/y.

Yields: The yields from AL plastic depend on the package specification, the amount of plastic and aluminium. But for this cost estimate, the yields as stated in Table 1 were used.

Revenues: As reasoned in (Riedewald et al., 2021), the waxes or the pyrolysis oil is sold as Heavy Bunker Oil to an oil refinery, achieving a revenue figure of €210/t for the ALPyro operator. The operator pays the transport cost of the oil to the refinery, and this is reflected in the oil price.

The AL waste treatment operator is paid a fee, the tipping fee, for accepting the waste. In Europe, as in many other countries, products are subject to the Extended Producer Responsibility (EPR) strategy. EPR refers to legislation (Pouikli, 2020) mandating that the producer must ensure that the product is disposed of responsibly towards its end of life. Moreover, increasingly the legislator is demanding recycling rates. Less clear is what EPR means in terms of revenue for a waste operator, as the EPR fee depends on the country, type of waste and other factors (Hogg et al., April 2020; Watkins et al., 2017). For simplicity, the Belgian incineration or landfill fee of €125/t (CEWEP, 2017) was taken as the EPR revenue stream for the AL waste pyrolysis operator. This revenue figure makes the tipping fee the most significant income stream for an AL recycling operator accounting for 62% of the revenue. In comparison, pyrolysis oil accounts for 21% and aluminium for 17%. Fig. 6 summarises the financial flows of the plant.

Capital Expenditure (CAPEX): The CAPEX, including the engineering effort, amounts to €3.5 million and was estimated with factorial techniques. Specifically, the VDI standard 2225 (VDI, 1997) was used to estimate the cost of the tank farm, condenser & overheads, the utilities, civil

and pipe runs. But, as the hourly throughput of the plant is well within conventional pyrolysis plants, the factorial costs can be compared with historical costs and cost adjustments were made if deemed necessary. A molten zinc pyrolysis reactor with a AL waste package throughput of 4,000 t/y or 2.2 t/h requires a molten zinc surface of 8 m length and 2 m width based on a 10-minute reaction time (Riedewald et al., 2021). The plant operates 8 h/day, 5 days a week and 340 days per year with a 2-week scheduled annual maintenance shutdown. The molten zinc depth is 1.5 m resulting in a relatively small molten zinc reactor (Maaß and Peißker, 2011). The reactor is filled with 24 m^3 of zinc at a cost of €269,000. For the carbon-aluminium mechanical separation equipment, a budget of €120,000 is allocated. Lastly, a contingency of 8% CAPEX was included. Table S2 in the supplemental material gives a more detailed breakdown of the CAPEX requirements.

Operating Expenditure (OPEX): The OPEX is estimated to be €278,000 per annum consisting of direct (operating materials or consumables, salaries, maintenance, etc.), indirect (depreciation, taxes, etc.), and general costs (administration, permits, insurance, etc.). The annual maintenance costs are estimated to be €45,000 and are a function of the equipment and the CAPEX (Peters et al., 2004). The consumables (nitrogen, compressed air, water, wastewater) are estimated to amount to €13,600/y and were factored down from the data given in (Riedewald et al., 2021). Also included are zinc losses, estimated to amount to €3,100/y. The zinc losses are associated with zinc drag out by the recovered aluminium and minor losses due to evaporation. The pyrolytic carbon from the plastic and paper pyrolysis is landfilled at a cost of €130/t. The plant is fully automated; only one operator is required to run the plant at a cost of €75,000/y. The insurance cost of the plant is estimated at 1% of CAPEX (Peters et al., 2004). €20,000/y is allowed for professional and licence fees. Moreover, an annual licence fee of €40,000 is included.

Sensitivity analysis: The revenues comprised of EPR, Al scrap, wax or pyrolysis oil, the CAPEX and the OPEX costs were increased by 10%, summarised in Table 2, to establish the most relevant factors influencing the IRR.

As the EPR contributes 62% to the overall revenue from the plant, any variation in its revenue will significantly influence the financial returns of the plant. The sensitivity analysis shows that an increase in the tipping fee by 10% will result in an IRR of 33% (Table 2). Therefore, any EPR increase would be beneficial for the financial returns of the plant. Furthermore, it is stabilising the financial returns as market forces do not influence the EPR as the wax or aluminium scrap revenues are. Increasing either the revenues of the wax or the revenue of the aluminium scrap by 10%, on the other hand, will only raise the IRR to 25%. Table S4 in the supplemental information contains the business performance of the plant with an increased EPR of 10% over 10 years.

Increasing the CAPEX by +10% decreases the IRR from 21 to 9%. Clearly, CAPEX spending must be tightly controlled to ensure that the plant remains economic. Increasing the OPEX by +10% reduces the IRR to 19%. Although this decrease is not as significant as the CAPEX, an operator must nevertheless tightly control the operating costs (salaries, business expenditures, etc.) to ensure that the plant is economical to operate. Table S5 in the supplemental information contains the business performance of the plant with an increased CAPEX of 10% over 10 years.

The IRR of the plant described above is calculated as 21% making it a desirable investment. The above cost estimate assumes that the pyrolytic carbon from the plastic and paper pyrolysis is landfilled at €130/t. But if the pyrolytic carbon could be "sold" to a coal-fired power station or to a municipal waste incinerator at no cost, i.e., revenue-neutral (€0.0/t rather than €130/t), the IRR of the plant would increase to 37% - a very attractive investment opportunity. And, indeed, the pyrolytic carbon may hold this value as the pyrolytic carbon from paper pyrolysis has a calorific value of 26 MJ/kg (Korkmaz et al., 2009), while the carbon obtained from plastic

pyrolysis has a calorific value of 20-26 MJ/kg (Jamradloedluk and Lertsatitthanakorn, 2014; Walendziewski, 2005), about the same as coal but without containing any sulphur (Walendziewski, 2005).

A plant operating a two-shift schedule would operate with half the hourly throughput, i.e., 1.1 t/h. Such a plant achieves an IRR of 20%. Hence, the difference in IRR between the 1 and 2-shift plant is only 1%, which is insignificant and within the estimate accuracy. Consequently, only a more detailed study can decide if a 1 or 2 shift operation is more economical.

Another possibility is to combine the large mixed plastic plant estimated in (Riedewald et al., 2021) with an aluminium laminated plastic recycling plant resulting in a more integrated recycling facility. This option was, however, not investigated for the current paper.

4. Conclusion

This paper provides pilot-scale experimental evidence that a directly heated molten metal reactor can recycle AL packaging, i.e., aluminium-laminated and Tetra Pak packaging. In addition, an economic analysis of a 4,000 t/y commercial-scale AL packaging recycling plant indicates that the molten metal process is economically viable as the IRR is over 20%. In combination, these two results suggest that the molten metal process may be a practical and profitable alternative to other reactors such as rotary kilns or fluidised beds. However, the experiments have two limitations. First, the experiments were in batch rather than in continuous mode. And second, the kinetics are uncertain. But, the AL packaging recycling experiments, although in batch mode, demonstrated that the reaction is fast, as only waxes condensed, meaning that secondary reactions due to long residence times were minimised.

The next step in the development of the AL packaging recycling process is a continuously operated demonstrator. Such a plant would establish the kinetics of the continuous AL

packaging recycling process among other parameters such as continuous operation and quality and quantity of the recycled products. Once the kinetics are known, the engineering design of a commercial plant could proceed into much more detail, therefore also improving the accuracy of the cost estimate. The current accuracy of the cost estimate is only -20 to +30% and, therefore, too low for a meaningful interpretation and assessment of the net present value (NPV), the payback period, and other financial indicators. Furthermore, large throughput reactors are possible because of the molten metal reactor's easy scale-up, i.e., doubling the molten metal surface area doubles the throughput.

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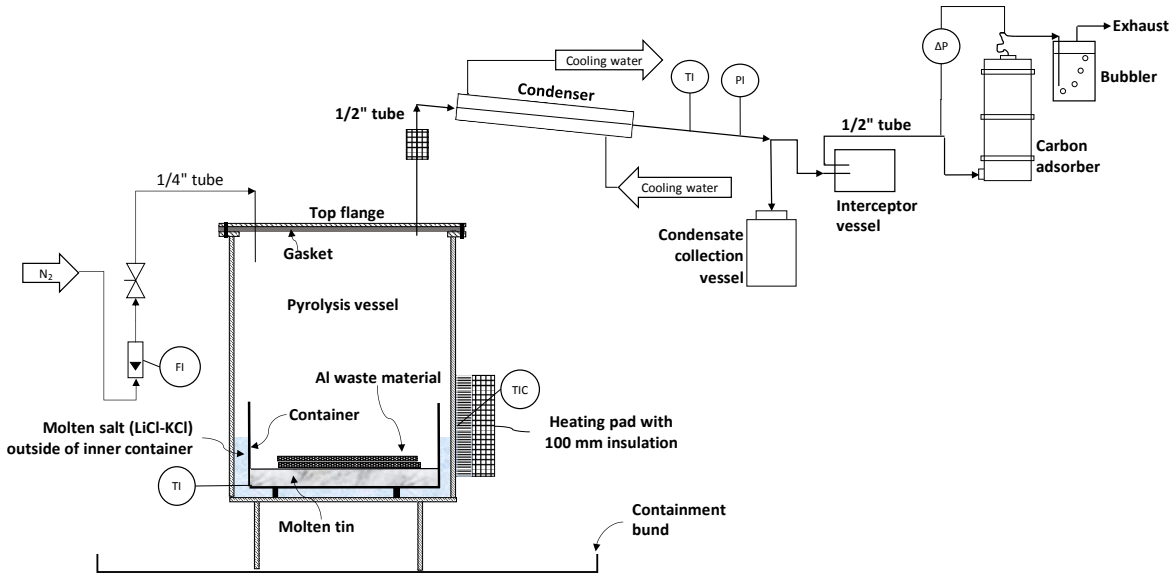


Fig. 1. Process and instrumentation diagram of the reactor and venting system for the pyrolysis experiments; (TI = temperature indicator, TIC = temperature indicator controller, PI = pressure indicator, ΔP = differential pressure gauge, FI = N_2 flow indicator, N_2 = nitrogen).

Tetra Pak	Reactor loaded with Tetra Pak	Recovered Tetra Pak (red circle)

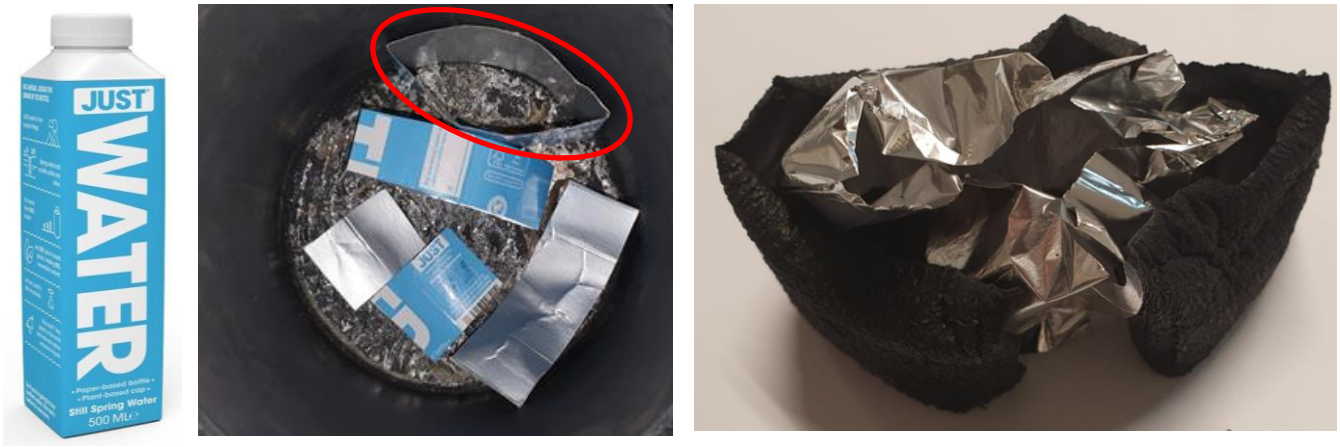


Fig. 2. Left: Original Tetra Pak; middle: the container inside the reactor loaded with five pieces cut from the Just Water Tetra Pak; right: Recycled clean aluminium with pyrolytic carbon from paper recovered from the reactor. The original Tetra Pak piece is in the red circle.

Original AL packages	Treated coffee AL package	Treated Whiskas food pouches



Fig. 3. AL pouch packages from Cork Coffee Roasters and Whiskas cat food pouches before and after recovery from the molten metal pyrolysis reactor.



Fig. 4. Wax fog and waxes in the interceptor vessel during one of the Tetra Pak pyrolysis experiments; left: during the experiment; right: waxes at the bottom of the interceptor vessel.

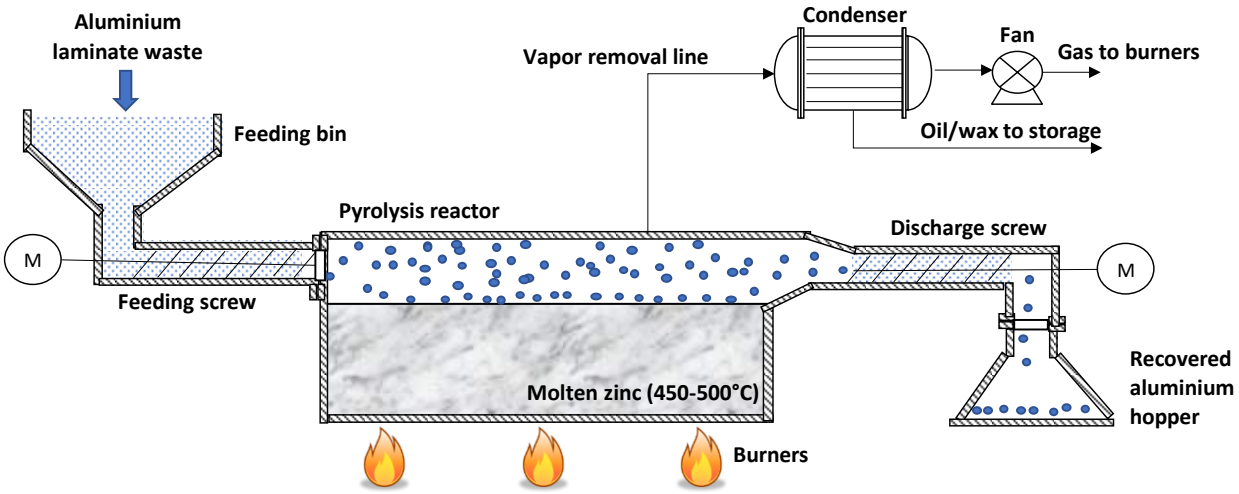


Fig. 5. Schematic of the proposed full-scale ALPyro aluminium laminate process.

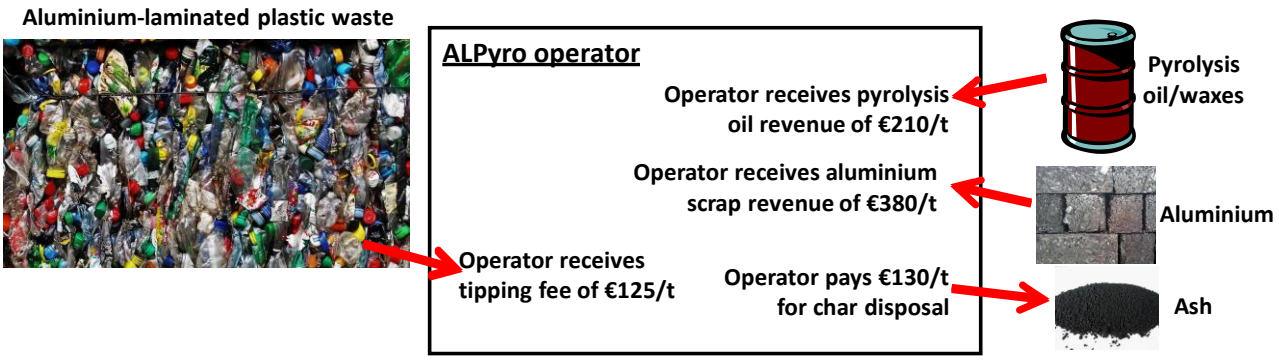


Fig. 6. Financial balance of an ALPyro plant with the revenues for the pyrolysis oil, aluminium, tipping fees and char disposal costs used in this study.

Table 1. Yields used for the cost estimate.

Material	Yield	Reference
Aluminium	90 kg per ton of AL plastic waste	(Slater and Crichton, 2011)
Pyrolysis oil/wax	200 kg per ton of AL plastic waste	(Slater and Crichton, 2011)
Char	13 wt.% of AL plastic waste	(Riedewald et al., 2021)

Table 2. Summary of the financial measures of the ALPyro plant compared to the results of the sensitivity analysis.

Parameter	Original IRR	Original NPV	Original Breakeven	Parameter change	Revised IRR	Revised NPV	Revised Breakeven
Revenues							
ERP	21	€280,000	3.15	+10%	33%	€599,000	2.97
Al scrap					25%	€367,000	3.10
Wax or pyrolysis oil					25%	€387,000	9.09
OPEX					19%	€288,000	3.15
CAPEX					9%	€6,000	3.15

Recycling of aluminium laminated pouches and Tetra Pak cartons by molten metal pyrolysis – pilot-scale experiments and economic analysis

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Abstract

Aluminium laminated (AL) pouch packages and aluminium laminated Tetra-Pak cartons are considered unrecyclable, reducing their otherwise excellent lifecycle performance. This paper describes experimental results on pilot plant trials to recycle AL packages with a molten metal pyrolysis reactor. The experimental evidence shows that both package formats can be recycled and that clean aluminium can be recovered. However, the recovered aluminium from Al pouches may require mechanical cleaning as the consumer's information is printed onto the

aluminium, leaving a carbon residue on the recovered aluminium. On the other hand, over 90% of the polypropylene plastic layer on the AL packaging pyrolysed into waxes, pointing to excellent kinetics. Moreover, an economic analysis of a 4,000 t/y commercial-scale plant demonstrates that a molten metal AL recycling plant is economically viable, achieving an internal rate of return (IRR) of over 20%.

Keywords: composite recycling, aluminium recycling, extended user responsibility, commercial-scale plant

1. Introduction

The aluminium laminated (AL) packaging meets many of the packaging requirements of the food industry (Yam, 2009). AL packaging is inert, lightweight, tough and the aluminium layer with a thickness of 6-150 µm provides a life-long barrier against aroma loss. It also protects the food against ingress of moisture, air, microorganisms, UV light and other food spoiling agents (Kerry, 2012), resulting in an extending shelf life (Kerry, 2012; Nonclercq, 2016). These advantages resulted in annual AL packaging growth rates of 10-15%, and in 2015 about 190,000 tonnes of aluminium were consumed for its production in Europe alone (Slater and Crichton, 2011).

Two popular AL food packaging presentations are the AL plastic pouch and the flow wrap. The AL plastic pouch is often used for baby food, coffee, pet foods, pasta sauce and cereals, whereas the flow wrap is used for crisps, chocolate bars and other snack foods (Nonclercq, 2016). The typical AL plastic pouch is made from three primary materials or layers: (1) a polyethylene terephthalate (PET) layer of ca. 12 µm thickness also displaying packaging information on the outside, (2) an aluminium foil of ca. 7 µm thickness and (3) an inside layer of polyethylene (PE) with a thickness of ca. 75 µm providing food compatibility and other properties (Nonclercq, 2016). The typical AL flow wrap comprises a polypropylene (PP) layer of 35 µm

thickness providing strength and stiffness. The aluminium layer of 0.04 µm thickness is typically vapour deposited onto the PP plastic. A primer applied to the aluminium layer allows printing onto the aluminium (Nonclercq, 2016).

A more rigid AL packaging is the Tetra Pak carton due to the inclusion of a paperback layer. This package is layered from the inside to the outside with PP, aluminium foil, PP, paperback and, finally, PP with the package information printed on it (Website, 2021b). A Tetra Pak carton composition is typically 63 wt.% cardboard or paper, 30 wt.% plastic, and 7 wt.% aluminium (Korkmaz et al., 2009).

AL packaging also has lifecycle advantages over other packaging formats performing the same function. Many authors' life cycle assessments (LCA) (Bayus et al., 2016; Bukowski and Richmond, 2018; Nonclercq, 2016; Xie et al., 2011) have shown the superiority of AL packaging over AL packaging other packaging formats. An LCA output for an AL pouch compared to other packaging formats, given in Table S1 in the supplemental information (Bukowski and Richmond, 2018), shows the superiority of AL packaging over different designs.

The major disadvantage of AL packaging is that it is considered unrecyclable (Horodytska et al., 2018; Slater and Crichton, 2011; Velzen et al., March 2020), which in Europe is often indicated on the packaging by the sign "currently not recycled" i.e., the material is landfilled. But if AL packaging was recyclable, its LCA could be improved even further (Bayus et al., 2016; Bukowski and Richmond, 2018; Nonclercq, 2016; Xie et al., 2011). Especially the recyclability of aluminium could have a significant positive impact, as the recycling of aluminium from secondary sources reduces the energy consumption compared to its production from ore by 95% (Warrings and Fellner, 2018). Moreover, aluminium can be recycled repeatedly without losing its properties, making it an ideal circular economy material (European Aluminium, 2020).

The literature describes various methods to recycle AL packaging, of which none is currently employed on an industrial scale. Fávaro et al. (Fávaro et al., 2013) studied the possibility of delaminating AL packages with solvents to remove the plastic layers. Under supercritical conditions of 255°C and a pressure of 11.65 MPa, metallic aluminium was obtained. Potential drawbacks of this process are the high operating pressures and solvent use, which in combination may result in an expensive process. Mu'min et al. (Mu'min et al., 2017) investigated wet torrefaction to recycle aluminium from AL packaging waste. Again, high operating pressures may result in an expensive process.

Pyrolysis has long been identified as an ideal process to recycle mixed plastics (Czajczyńska et al., 2017; López et al., 2010; Ragaert et al., 2017; Sharuddin et al., 2016; Siddiqui and Redhwi, 2009; Singh et al., 2017). Plastic pyrolysis is a depolymerisation process executed at temperatures above 400°C, in an oxygen-free environment and typically at ambient pressures (Boateng, 2008). Products of mixed plastic pyrolysis are pyrolysis oil (ca. 80-95%), pyrolytic carbon (ca. 1-5%; also referred to as char or ash in the literature) and gases (5-15%) depending on pyrolysis temperature, catalyst, and the mixed plastic composition (López et al., 2010; Sharuddin et al., 2016; Siddiqui and Redhwi, 2009). The pyrolysis oil may be upgraded to diesel or other petroleum products, the char would be landfilled, and the gases (methane, propane etc.) would heat the process. The capability of pyrolysis to treat mixed plastics, i.e., PET, PE and PP, is important in AL packaging recycling as these plastics are present in AL packaging. Moreover, because it is difficult to obtain a clean AL packaging waste stream from municipal waste without other plastics, this advantage of pyrolysis becomes ever more critical.

Furthermore, pyrolysis can also treat contaminated plastics, e.g., plastic packaging with food residues, paper, ink and other contaminants. Irawan et al. (Irawan et al., 2018) pyrolysed AL packaging in a conventional pyrolysis reactor (Boateng, 2008). Their study found that the recovered aluminium could be separated from the char easily. A potential drawback of conventional rotary kiln pyrolysis reactors is their relatively poor kinetics compared to directly

heated reactors (Riedewald et al., 2021). Furthermore, increasing operating temperatures result in decreased char yields, whereas the aluminium yield remains the same. The possibility of recycling AL packaging with microwave-induced pyrolysis was investigated by Palafox and Chase on a bench-scale reactor (Ludlow-Palafox and Chase, 2001), whereas Slater and Crichton (Slater and Crichton, 2011) report on experiments performed on a larger scale. At both scales, clean aluminium together with hydrocarbons from the pyrolysis of the plastics was obtained. Microwave reactors have the advantage over rotary kilns in that they are directly heated reactors. Still, they have the drawback of using a microwave generator – a potentially expensive piece of equipment. Moreover, the non-condensable gases cannot be combusted for internal heat generation. Instead, they would have to be combusted in an electrical generator, generating electricity to drive the microwave generator. The capital and operational costs of generating electricity in this fashion may, however, be prohibitive.

The capability of pyrolysis to treat paper, plastic, and aluminium is also crucial for recycling Tetra Pak cartons. Korkmaz et al. (Korkmaz et al., 2009) pyrolysed 1 to 2 cm large Tetra Pak pieces in a laboratory-scale tube reactor. While the yields, gas, and wax composition are reported, no information is conveyed to separate the aluminium and carbon. Robertson (Robertson, 2021) reviewed various Tetra Pak recycling methods, including pyrolysis, whereas Zawadiak et al. (Zawadiak et al., 2017) provide an overview of the current techniques of Tetra Pak recycling. In summary, pyrolysis can recycle both AL and Tetra Pak packaging formats. One of the advantages of pyrolysis is that pyrolysis can operate with residual food contamination still present on the packaging (Slater and Crichton, 2011).

This paper proposes pyrolysing AL and Tetra Pak packaging by direct heat transfer with molten metal resulting in a rapid process already shown on whole tyres (Rathsack et al., 2015), which were fully pyrolysed within 15 minutes - significantly faster than processing tyres in indirectly heated rotary kilns or similar reactors. This demonstrates the advantages of direct heat transfer or, in this case, processing on molten metals. In this paper, Al plastic pouches and Tetra-Pak

aluminium laminated packages were treated in a pilot plant. An AL recycling process must be capable of treating both of these packaging types, as they represent the vast majority of aluminium laminated food packages. Moreover, in practice, separation of these two packages types may not always be possible.

The molten metal pyrolysis reactor does not have the disadvantages of other reactor types. First, the non-condensables can be used for internal heat generation, resulting in a self-sustaining process. Second, the molten metal reactor is a directly heated reactor i.e., the AL packages are in direct contact with the molten metal resulting in a fast process. Finally, this paper presents an economic analysis of a future commercial-scale AL recycling process named ALPyro although only limited design and economic data are available. Such a financial analysis is essential to obtain investment for a demonstration plant phase before commercialisation of the process can commence. But significant amounts of private capital (about €2-4 million depending on the size of the demonstration unit and other parameters) will be required, and a private investor will need to know the potential financial returns of this process, which the economic analysis provides.

2. Materials and Methods

2.1. Methods

Fig. 1 presents the process and instrumentation diagram of the multipurpose molten metal pyrolysis experimental setup. Various inner containers can be used, as well as various molten metals allowing the processing of various materials and in-situ separation. A 6" 316 L stainless-steel container was filled with 1,300 g of tin, equating to a molten tin level of 1 cm. This container was placed inside the pyrolysis reactor, which operates in batch mode. The pyrolysis reactor was manufactured from an 8" (125 mm) ANSI schedule 10, 316L stainless steel pipe and a length resulting in a vessel height of 300 mm. A 3 mm thick 316L stainless steel plate was

the bottom wall of the vessel. By unbolting the top flange, the vessel can be opened. All interconnecting pipes were ½" Swagelok tubes. The pyrolysis vapours were condensed by a 30 cm long ambient temperature water-cooled ½" tube. The condenser was sloped by 20 degrees draining into a condensate collection vessel. The formation of an explosive atmosphere within the pyrolysis vessel vapour space and downstream equipment was prevented by initially nitrogen inerting the equipment and continuously sweeping the pyrolysis vessel with a nitrogen flow of ca. 120 liters per hour during the experiment. The operating pressure of the entire system was slightly above atmospheric (~10 mbar), preventing air ingress due to the hydrostatic pressure induced by the bubbler. The space between the reactor walls and the container was filled with 2,600 g of LiCl-KCl salt providing heat transfer between the heating pads and the container.

Three ceramic electrical heating pads heated the reactor. The vessel was insulated with 100 mm thick glass wool. But the top flange and the vessel sides below 80 mm of the flange were not insulated. An Ashcroft temperature gauge (range of 0 to 500ºC, with a 1 % ASME B40.3, Grade A accuracy inserted into a thermowell manufactured from 316L stainless steel) was used to measure the operating temperature. The operating temperature was controlled to an estimated ±5°C by a control system (Stork Copperheat 50 KVA Heat Treatment Module, Model no. 16050). The temperature feedback to the Heat Treatment Module control system was provided by a thermocouple located between the outside surface of the reactor and one of the heating pads (denoted TIC in Fig. 1).

The ½" pipe connecting the reactor to the condenser was also insulated. The preferred pipe connections were metal to metal fittings (Swagelok) as they are vapour tight and can be opened for inspection.

For the experiments, the operating temperature was 450°C as such a temperature appears to be ideal in maximising the yield of pyrolysis oil or wax from plastic pyrolysis (López et al., 2010;

Sharuddin et al., 2016; Siddiqui and Redhwi, 2009), one of the revenue streams for an operator. Once the operating temperature of 450°C was reached, it was maintained for 60 minutes to ensure the pyrolysis reaction completes. On reaching this point, the heat was turned off, and the equipment cooled naturally to ambient. Once the reactor cooled down to 100°C, the nitrogen flow was switched off. Finally, the reactor was opened, and the pyrolysed material in the container was removed for analysis.

The condensate collection vessel was a 100 mL laboratory glass bottle. The interceptor vessel was a 1,000 mL laboratory glass bottle (Fig. 4). The bubbler was a 250 mL laboratory glass bottle filled with 100 mL of water to make the flow of nitrogen through the experiment visible and to provide a small amount of overpressure throughout the experiment preventing air ingress.

A Sartorius ED4202S laboratory scale (max 4,200 g; d = 0.01 g) was used to weigh the various materials.

The temperature gauge located after the condenser was a GenWare electric temperature gauge with a range of -40 to 230°C. The temperature of the top flange and the area below the top flange was measured with a GenWare infrared-thermometer having a temperature range from -32 to 550°C and a 0.5 second response time.

A carbon adsorber, a drum filled with 25 kg activated carbon supplied by Silcarbon Aktivkohle GmbH, Germany, abated the non-condensable vapours from the experiment before discharging the gas stream to the air extract system of the building.

After each experiment, the reactor, pipes and condenser were visually inspected for evidence of wax or other deposits, but none were found.

2.2. Materials

Two different AL packages were used. One waste package was Tetra Pak (Fig. 2). The Tetra Pak carton was cut into five pieces to fit the reactor, as shown in Fig. 2, but the Sugarcane (Website, 2021a) top and screw cap were removed and discharged as this is not present on all Tetra Pak cartons. The other packages were coffee packages from Cork Coffee Roasters and Whiskas cat food pouches (Fig. 3); both packages were washed and dried.

The tin was 99.95% pure, presented in 3-5 mm lumps, and was purchased from Amazon. 316L stainless steel is a suitable material of construction for molten tin for the temperature ranges of these experiments (Maaß and Peißker, 2011).

The heat transfer salt was a eutectic mixture of technical grade lithium chloride (LiCl) and potassium chloride (KCl) salts (41.8 mol% KCl and 58.2 mol% LiCl (LiCl-KCl)). LiCl-KCl salt was chosen for these experiments, as it is stable, non-toxic and inert at the operating temperatures (Sohal et al., March 2010). Additionally, 316L stainless steel is a suitable material of construction for molten LiCl-KCl (Ignatiev and Surenkov, 2012). Leverton Clarke, Basingstoke, United Kingdom, supplied the LiCl. Scientific & Chemical Supplies Ltd, Cork, Ireland, provided the KCl.

The nitrogen was certified 99.999 % pure.

3. Results and Discussions

3.1. Experimental results

The emphasis of this research was on the solid residue, i.e., the aluminium and carbon from the AL and Tetra Pak packages remaining in the reactor. The pyrolysis oil, waxes and gases generated were of secondary importance, as the composition, yields, and optimum operating

temperatures are known for (1) PE, PP and PET (Brems et al., 2011; Honus et al., 2018a; Honus et al., 2018b; López et al., 2010; Sharuddin et al., 2016; Siddiqui and Redhwi, 2009), (2) paper (Li et al., 2005; Ma et al., 2017; Rutkowski, 2013; Zhou et al., 2013) and (3) mixtures of paper and plastics (Fekhar et al., 2020; Johansson et al., 2018).

The experiments on Tetra Pak cartons demonstrate that aluminium recycling from a Tetra Pak carton is straightforward. The aluminium can be recovered clean, as shown in Fig. 2. The black material is pyrolytic carbon from the pyrolysis of the paperback. The pyrolytic carbon does not adhere to the aluminium foil. Moreover, it is very brittle, making the separation of the pyrolytic carbon from the aluminium easy to do in a follow-on operation. The yield of pyrolysis carbon from the pyrolysis of the plastics, on the other hand, is negligible, as the yield of pyrolytic carbon from PE, PP and PET at 450°C is between 0-3% (Aguado et al., 2002; López et al., 2010).

The experiments on the AL pouch packages also demonstrate that the aluminium can be recycled with the molten metal process. But, as shown in Fig. 3, the recovered aluminium foil is not as clean as the aluminium from the Tetra Pak's. This is because the consumer's information is printed onto the aluminium, leaving a carbon residue from the print on the aluminium. As a result, a mechanical cleaning step may be required to remove the carbon residue from the recovered aluminium foil. Such a mechanical cleaning step, if necessary, appears feasible, as the carbon residue can be wiped off quite easily. In contrast, the side of the aluminium foil forming the inside of the package is completely clean. It is clean, as the pyrolysis of PE, PP or PET only results in negligible amounts of carbon (Aguado et al., 2002; López et al., 2010).
Another option to ease the removal of the carbon residue from the aluminium would be for the manufacturer to change the formulation of the paint, which would be feasible in the context of a recycling/circular economy framework.
As already stated in the introduction, the aluminium layer may be of various thicknesses and grades (Slater and Crichton, 2011). Hence in practice, the recovered aluminium would need to

be bailed and send to an aluminium smelter for re-melting closing the recycling loop (Schlesinger, 2017).

The pyrolysis of the PP plastic layers of the Tetra Pak and the PET and PE of the AL plastic resulted in waxes rather than oils. This result is consistent with the literature stating that at a pyrolysis temperature of 450°C, PP generates up to 92% waxes (Aguado et al., 2002; Honus et al., 2018a; Honus et al., 2018b; Korkmaz et al., 2009). This must be considered for the design of the experiment and for a full-scale plant as waxes, should they solidify have the potential to block the condenser (Park et al., 2002) or other pieces of equipment resulting in a high-pressure event causing safety concerns.

The experimental setup (Fig.1) included a glass interceptor vessel giving a visual impression of the state of the vapour stream after the condenser. The fluid velocity reduces within this vessel, allowing wax particles to sink to the bottom, visible by the top section of the glass vessel being clear of fog, as shown in Fig. 4.

A sharp 90-degree bend at the entrance to the interceptor vessel caused wax particles suspended in the nitrogen flow to collide with the wall and collect there. Intermittently the waxes blocked the gas flow, the pressure would slowly rise to about 150 mbar, which would blow the waxes into the interceptor vessel (Fig. 4, right), and the process would start again. This wax collection and blowout took place three times within about 5 minutes, and one might speculate that the plastic pyrolysis reaction took place during that timeframe indicating fast kinetics. But, if the kinetics are indeed that fast, it must be proven by a follow-on experiment designed to ascertain the kinetics of the molten metal process, which could, for instance, be executed on a future demonstrator scale unit.

Waxes also condensed on the inside surfaces of the vessel, i.e., on the gasket below the top flange (Fig. 1). The outside surface temperature of the top flange was about 130°C; hence only the longer chain waxes condensed there. After each experiment, a visual inspection of the condenser and the pipes found no build-up of waxes or other materials there.

A wax mass balance suggests that the heating of the plastic was rapid as virtually all of the plastic converted into waxes, confirming the wax yields of others, i.e., over 90% (Aguado et al., 2002; Honus et al., 2018a; Honus et al., 2018b; Korkmaz et al., 2009). Secondary reactions, i.e., cracking of waxes into oil and gases, did not occur as high surface temperatures, high-temperature hot spots, or long residence times are avoided by the molten metal reactor. Hence, the results of others on plastic pyrolysis (Honus et al., 2018a; Honus et al., 2018b; Korkmaz et al., 2009) are valid to design a full-scale molten metal pyrolysis recycling plant and were used for the economic analysis, which will be presented in the next section.

3.2. Economic analysis of a full-scale plant

Performing an economic analysis at this process development stage is difficult as not more than 5% of the engineering is completed. Therefore, the accuracy of the cost estimate is -20 to +30% (AACE, 2005). Performing such an economic analysis at this stage of the process development is essential for the reasons given in the introduction.

3.2.1 Description of the full-scale plant

The ALPyro process is a continuous recycling process and is shown schematically in Fig. 5. The waste material is in direct contact with the molten metal. Zinc rather than tin is used, as it is more cost-effective. But by using zinc, the operating temperature must be at least 450°C, as the melting point of zinc is 419°C. However, an operating temperature of 450°C maximises the yield of pyrolysis oil, a revenue stream, as stated above. All equipment subject to condensation of waxes is kept at a temperature above 80°C allowing the waxes to remain liquid and drain off.

The AL waste is provided in a feeding bin and transported by a feeding screw into the pyrolysis reactor, i.e., onto molten zinc. On the molten zinc, the waste pyrolyses into hydrocarbon vapours, gases and solids. Any water present in the feedstock also evaporates. The solids, composed of aluminium and pyrolytic carbon, float on the molten zinc.

A discharge screw removes the recovered aluminium and the pyrolytic carbon into the recovered aluminium product hopper. The pyrolysis vapours are removed from the pyrolysis reactor via a vapour removal line and condensed to pyrolysis oil, waxes, or both. A fan provides the suction or driving force for the vapour removal. The non-condensable gases, i.e., methane, propane may be sent to the burners providing the heat for the pyrolysis process, ensuring that the process runs without any additional energy.

3.2.2 Economic analysis

The economics of an ALPyro plant is assessed by a number of financial measures such as the internal rate of return (IRR), the net present value (NPV) and the breakeven point. Of these measures, ~~The~~ the IRR is the most helpful measure indicating~~es~~ the relative return of an investment without taking the scale of the project into account (Crundwell, 2008). In general, larger infrastructure projects such as waste plastic pyrolysis plants are considered profitable by private investors if the IRR exceeds 15% (Riedewald et al., 2021). Here a minimum IRR of 20% is demanded, as the financial uncertainty is still considerable. The evaluation period of the IRR and the NPV is ten years. The IRR of the ALPyro plant is 21%, the NPV €280,000, and the breakeven point is reached after 3.15 years. Table S3 in the supplemental material presents the business performance of the plant over 10 years, giving a much better and comprehensive impression of the financial performance of the ALPyro plant than any of the financial measures. The following explains how the most relevant factors influencing the financial performance of the plant were estimated.

The plant's location is assumed to be in Europe, close to significant areas of population such as London, Birmingham, Berlin or Amsterdam. The costs for a warehouse for temporary waste storage is not included. Instead, it is assumed that the plant is located close to a plastic sorting plant and that warehouse space exists. A storage capacity for 8 hours' worth of feedstock may be worth considering, but this is not included in the cost estimate. This surge capacity would be used when the AL recycling plant is idle and temporarily store AL waste delivered to the site from other waste sorting facilities.

Moreover, the AL recycling plant is part of a larger facility, meaning that management, engineering and other support is available. Further, it is assumed that AL pouch packages and Tetra Pak cartons are treated together.

Market: The AL recycling plant can only be sized if the addressable amount of waste AL is known. About 14,000 t/y of aluminium laminated plastic is collected in the UK and available for recycling (Slater and Crichton, 2011). But waste transport costs (Riedewald et al., 2021) will limit UK plant sizes to about 4,000 t/y.

Yields: The yields from AL plastic depend on the package specification, the amount of plastic and aluminium. But for this cost estimate, the yields as stated in Table 1 were used.

Revenues: As reasoned in (Riedewald et al., 2021), the waxes or the pyrolysis oil is sold as Heavy Bunker Oil to an oil refinery, achieving a revenue figure of €210/t for the ALPyro operator. The operator pays the transport cost of the oil to the refinery, and this is reflected in the oil price.

The AL waste treatment operator is paid a fee, the tipping fee, for accepting the waste. In Europe, as in many other countries, products are subject to the Extended Producer Responsibility (EPR) strategy. EPR refers to legislation (Pouikli, 2020) mandating that the producer must ensure that the product is disposed of responsibly towards its end of life. Moreover, increasingly the legislator is demanding recycling rates. Less clear is what EPR means in terms of revenue for a waste operator, as the EPR fee depends on the country, type of waste and other factors (Hogg et al., April 2020; Watkins et al., 2017). For simplicity, the Belgian incineration or landfill fee of €125/t (CEWEP, 2017) was taken as the EPR revenue stream for the AL waste pyrolysis operator. This revenue figure makes the tipping fee the most significant income stream for an AL recycling operator accounting for 62% of the revenue. In comparison, pyrolysis oil accounts for 21% and aluminium for 17%. Fig. 6 summarises the financial flows of the plant.

Capital Expenditure (CAPEX): The CAPEX, including the engineering effort, amounts to €3.5 million and was estimated with factorial techniques. Specifically, the VDI standard 2225 (VDI, 1997) was used to estimate the cost of the tank farm, condenser & overheads, the utilities, civil

and pipe runs. But, as the hourly throughput of the plant is well within conventional pyrolysis plants, the factorial costs can be compared with historical costs and cost adjustments were made if deemed necessary. A molten zinc pyrolysis reactor with a AL waste package throughput of 4,000 t/y or 2.2 t/h requires a molten zinc surface of 8 m length and 2 m width based on a 10-minute reaction time (Riedewald et al., 2021). The plant operates 8 h/day, 5 days a week and 340 days per year with a 2-week scheduled annual maintenance shutdown. The molten zinc depth is 1.5 m resulting in a relatively small molten zinc reactor (Maaß and Peißker, 2011). The reactor is filled with 24 m^3 of zinc at a cost of €269,000. For the carbon-aluminium mechanical separation equipment, a budget of €120,000 is allocated. Lastly, a contingency of 8% CAPEX was included. Table S2 in the supplemental material gives a more detailed breakdown of the CAPEX requirements.

Operating Expenditure (OPEX): The OPEX is estimated to be €278,000 per annum consisting of direct (operating materials or consumables, salaries, maintenance, etc.), indirect (depreciation, taxes, etc.), and general costs (administration, permits, insurance, etc.). The annual maintenance costs are estimated to be €45,000 and are a function of the equipment and the CAPEX (Peters et al., 2004). The consumables (nitrogen, compressed air, water, wastewater) are estimated to amount to €13,600/y and were factored down from the data given in (Riedewald et al., 2021). Also included are zinc losses, estimated to amount to €3,100/y. The zinc losses are associated with zinc drag out by the recovered aluminium and minor losses due to evaporation. The pyrolytic carbon from the plastic and paper pyrolysis is landfilled at a cost of €130/t. The plant is fully automated; only one operator is required to run the plant at a cost of €75,000/y. The insurance cost of the plant is estimated at 1% of CAPEX (Peters et al., 2004). €20,000/y is allowed for professional and licence fees. Moreover, an annual licence fee of €40,000 is included.

Sensitivity analysis: The revenues comprised of EPR, Al scrap, wax or pyrolysis oil, the CAPEX and the OPEX costs were increased by 10%, summarised in Table 2, to establish the most relevant factors influencing the IRR.

As the EPR contributes 62% to the overall revenue from the plant, any variation in its revenue will significantly influence the financial returns of the plant. The sensitivity analysis shows that an increase in the tipping fee by 10% will result in an IRR of 33% (Table 2). Therefore, any EPR increase would be beneficial for the financial returns of the plant. Furthermore, it is stabilising the financial returns as market forces do not influence the EPR as the wax or aluminium scrap revenues are. Increasing either the revenues of the wax or the revenue of the aluminium scrap by 10%, on the other hand, will only raise the IRR to 25%. Table S4 in the supplemental information contains the business performance of the plant with an increased EPR of 10% over 10 years.

Increasing the CAPEX by +10% decreases the IRR from 21 to 9%. Clearly, CAPEX spending must be tightly controlled to ensure that the plant remains economic. Increasing the OPEX by +10% reduces the IRR to 19%. Although this decrease is not as significant as the CAPEX, an operator must nevertheless tightly control the operating costs (salaries, business expenditures, etc.) to ensure that the plant is economical to operate. Table S5 in the supplemental information contains the business performance of the plant with an increased CAPEX of 10% over 10 years.

The IRR of the plant described above is calculated as 21% making it a desirable investment. The above cost estimate assumes that the pyrolytic carbon from the plastic and paper pyrolysis is landfilled at €130/t. But if the pyrolytic carbon could be "sold" to a coal-fired power station or to a municipal waste incinerator at no cost, i.e., revenue-neutral (€0.0/t rather than €130/t), the IRR of the plant would increase to 37% - a very attractive investment opportunity. And, indeed, the pyrolytic carbon may hold this value as the pyrolytic carbon from paper pyrolysis has a calorific value of 26 MJ/kg (Korkmaz et al., 2009), while the carbon obtained from plastic

pyrolysis has a calorific value of 20-26 MJ/kg (Jamradloedluk and Lertsatitthanakorn, 2014; Walendziewski, 2005), about the same as coal but without containing any sulphur (Walendziewski, 2005).

A plant operating a two-shift schedule would operate with half the hourly throughput, i.e., 1.1 t/h. Such a plant achieves an IRR of 20%. Hence, the difference in IRR between the 1 and 2-shift plant is only 1%, which is insignificant and within the estimate accuracy. Consequently, only a more detailed study can decide if a 1 or 2 shift operation is more economical.

Another possibility is to combine the large mixed plastic plant estimated in (Riedewald et al., 2021) with an aluminium laminated plastic recycling plant resulting in a more integrated recycling facility. This option was, however, not investigated for the current paper.

4. Conclusion

This paper provides pilot-scale experimental evidence that a directly heated molten metal reactor can recycle AL packaging, i.e., aluminium-laminated and Tetra Pak packaging. In addition, an economic analysis of a 4,000 t/y commercial-scale AL packaging recycling plant indicates that the molten metal process is economically viable as the IRR is over 20%. In combination, these two results suggest that the molten metal process may be a practical and profitable alternative to other reactors such as rotary kilns or fluidised beds. However, the experiments have two limitations. First, the experiments were in batch rather than in continuous mode. And second, the kinetics are uncertain. But, the AL packaging recycling experiments, although in batch mode, demonstrated that the reaction is fast, as only waxes condensed, meaning that secondary reactions due to long residence times were minimised.

The next step in the development of the AL packaging recycling process is a continuously operated demonstrator. Such a plant would establish the kinetics of the continuous AL

packaging recycling process among other parameters such as continuous operation and quality and quantity of the recycled products. Once the kinetics are known, the engineering design of a commercial plant could proceed into much more detail, therefore also improving the accuracy of the cost estimate. The current accuracy of the cost estimate is only -20 to +30% and, therefore, too low for a meaningful interpretation and assessment of the net present value (NPV), the payback period, and other financial indicators. Furthermore, large throughput reactors are possible because of the molten metal reactor's easy scale-up, i.e., doubling the molten metal surface area doubles the throughput.

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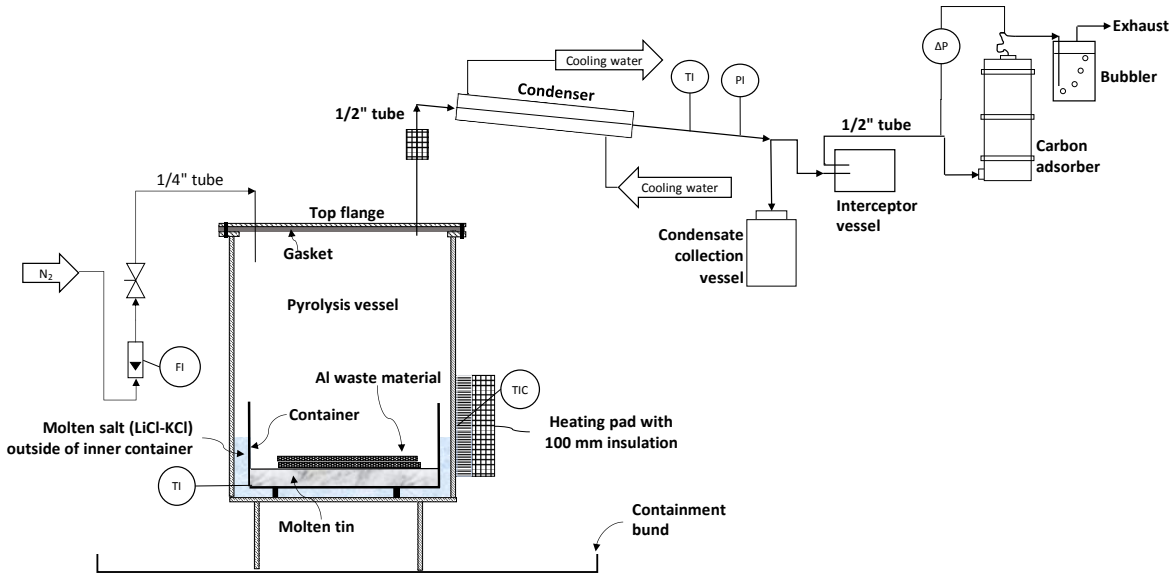


Fig. 1. Process and instrumentation diagram of the reactor and venting system for the pyrolysis experiments; (TI = temperature indicator, TIC = temperature indicator controller, PI = pressure indicator, ΔP = differential pressure gauge, FI = N_2 flow indicator, N_2 = nitrogen).

Tetra Pak	Reactor loaded with Tetra Pak	Recovered Tetra Pak (red circle)

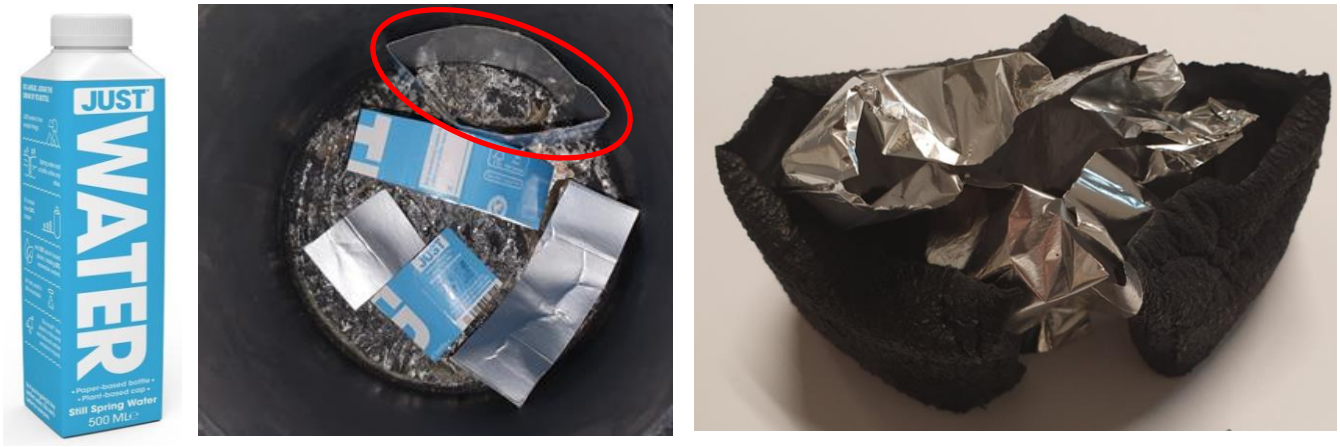


Fig. 2. Left: Original Tetra Pak; middle: the container inside the reactor loaded with five pieces cut from the Just Water Tetra Pak; right: Recycled clean aluminium with pyrolytic carbon from paper recovered from the reactor. The original Tetra Pak piece is in the red circle.

Original AL packages	Treated coffee AL package	Treated Whiskas food pouches



Fig. 3. AL pouch packages from Cork Coffee Roasters and Whiskas cat food pouches before and after recovery from the molten metal pyrolysis reactor.



Fig. 4. Wax fog and waxes in the interceptor vessel during one of the Tetra Pak pyrolysis experiments; left: during the experiment; right: waxes at the bottom of the interceptor vessel.

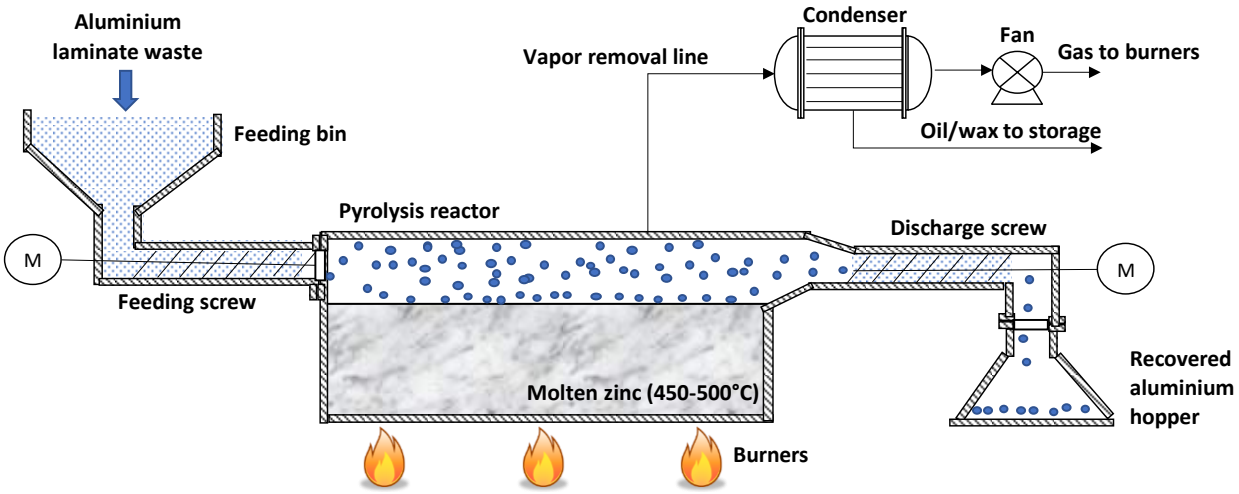


Fig. 5. Schematic of the proposed full-scale ALPyro aluminium laminate process.

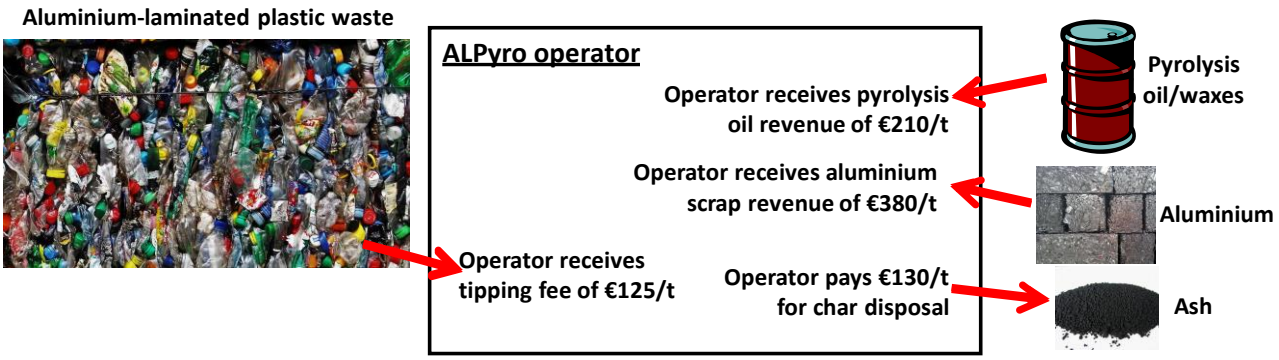


Fig. 6. Financial balance of an ALPyro plant with the revenues for the pyrolysis oil, aluminium, tipping fees and char disposal costs used in this study.

Table 1. Yields used for the cost estimate.

Material	Yield	Reference
Aluminium	90 kg per ton of AL plastic waste	(Slater and Crichton, 2011)
Pyrolysis oil/wax	200 kg per ton of AL plastic waste	(Slater and Crichton, 2011)
Char	13 wt.% of AL plastic waste	(Riedewald et al., 2021)

Table 2. Summary of the financial measures of the ALPyro plant compared to the r~~R~~esults of the sensitivity analysis.

Parameter	Original IRR	Original NPV	Original Breakeven	Parameter change	Revised IRR	Revised NPV	Revised Breakeven
Revenues							
ERP	21	€280,000	3.15	+10%	33%	€599,000	2.97
Al scrap					25%	€367,000	3.10
Wax or pyrolysis oil					25%	€387,000	9.09
OPEX					19%	€288,000	3.15
CAPEX					9%	€6,000	3.15

Declaration of interests

☒ Authors Daniel Vogt, Ian Povey, Killian Barton, Liam Lewis, Tom Caris, Silvia Santos, Maria O'Mahoney, Maria Sousa-Gallagher: Declaration of interest: None.

☒The authors Frank Riedewald, Yunus Patel and Edward Wilson declare the following financial interests/personal relationships which may be considered as potential competing interests:

We are Directors and shareholders of Composite Recycling Ltd, the company owning the patent of the ALPyro process.

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