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1	Investigating the influence of ultrasound processing on drying kinetics and moisture
2	migration measurement in lactobacillus cultured and uncultured beef jerky
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17 Abstract

18 Low Frequency-Nuclear Magnetic Resonance (LF-NMR) was employed to elucidate changes in water distribution in cultured and uncultured beef jerky samples subjected to ultrasound pre-treatment. 19 20 Ultrasound pre-treatment at frequencies of 25, 33 and 45 kHz for 30 min, followed by marination (18 h) was carried out for both uncultured and cultured (Lactobacillus sakei) jerky samples. Water mobility 21 and distribution of water during drying were measured using LF-NMR. Among the various kinetic 22 models assessed, the Wang and Singh model provided the closest fit to the drying experimental data, 23 24 with high R2 (\geq 0.994), low RMSE (\leq 0.023) and low AICc (<-74.535) values for both cultured and uncultured samples. Distributed exponential analysis of T2 transversal relaxation times measured by 25 LF-NMR curves revealed the presence of three distinct peaks attributed to; bound water, water present 26 within the dense myofibrillar protein matrix and free-water at a relaxation time range of 0-10 ms (T2b), 27 10–100 ms (T21) and >100 ms (T22), respectively. Results presented in this study demonstrates that 28 the ultrasound effect on drying behaviour was frequency dependent and that LF-NMR can be employed 29 to evaluate moisture mobility and drying degree of beef jerky. 30

31 1. Introduction

32

Beef jerky is a nutrient dense ready-to-eat meat snack, possessing characteristics of a typical 33 intermediate moisture content product with a relatively long shelf-life. Commercially, beef jerky is 34 prepared using a hurdle-technology approach which involves employment of interventions, such as; 35 reducing water activity (a_w) and addition of preservatives such as organic acids, spices and curing 36 (nitrate/nitrite) salts. The development of whole-muscle and/or restructured jerky from a range of meats 37 by employing various curing ingredients (e.g. as organic acids, spices, sugars, NaCl and nitrate/nitrite 38 salts), curing methods and drying conditions have been widely reported (Choi, Jeong, Han, Choi, Kim, 39 40 Lee, et al., 2008; Jang, Kim, Hwang, Song, Kim, Ham, et al., 2015; Kucerova, Hubackova, Rohlik, & Banout, 2015). Most recently, the application of starter culture (e.g. lactic acid bacteria) to improve 41

flavour and quality of jerky products, while preventing the growth of spoilage bacteria, has been
reported (Biscola, Todorov, Capuano, Abriouel, Gálvez, & Franco, 2013; O'Connor, Ross, Hill, & Cotter,
2015; Zhao, Zhao, Lu, Huang, He, Tan, et al., 2016).

The application of ultrasound has been reported to enhance mass transfer rates during brining/curing of 45 meat, primarily by disrupting the continuity of cellular membranes due to various physical and chemical 46 effects of ultrasound (C Ozuna, Cárcel, García-Pérez, Peña, & Mulet, 2015). Ultrasound, in combination 47 with vacuum application has been shown to enhance the drying rate of beef and chicken meat (Baslar, 48 Kilicli, Toker, Sağdıc, & Arici, 2014). Ultrasound pre-treatment is widely reported to accelerate drying of 49 a range of food products (Awad, Moharram, Shaltout, Asker, & Youssef, 2012), which can affect texture 50 and water activity of products. Additionally, ultrasound treatment has shown promise in improving meat 51 52 tenderisation, depending on the ultrasonic intensities and processing times employed.

Moisture content is the main factor influencing the quality, safety and shelf life of meat-based 53 jerky. Conventionally, the moisture content of commercial forms of jerky is determined by oven drying 54 methods and sensory assessments. However, these methods are tedious, time-consuming, expensive 55 and require trained and skilled personnel. Thus, there is a great scientific and industrial interest to 56 develop a rapid, non-destructive and online method for determination of moisture content and drying 57 degree in order to ensure consistent jerky quality. Low-field nuclear magnetic resonance (LF-NMR) is a 58 59 sensitive, fast and non-invasive technique which has been widely adopted as an analytical technique for the characterization of water mobility and distribution within food matrices (Agudelo-Laverde et al., 60 2014; Troutman et al., 2001; Haiduc and van Duynhoven, 2005). The state and distribution of water in 61 food matrices, including meat, can be determined by LF-NMR and can provide useful information 62 about interactions between water and myofibrillar meat proteins, as it is governed by exchange of water 63 protons and exchangeable protons in proteins (Bertram, Engelsen, Busk, Karlsson, & Andersen, 2004). 64 LF-NMR has been successfully employed to study the effectiveness of various processing techniques. 65 including; brining, cooking, freezing and thawing on water distribution and mobility (Bertram, Kohler, 66

Böcker, Ofstad, & Andersen, 2006; Damez & Clerjon, 2013; C. Li, Liu, Zhou, Xu, Qi, Shi, et al., 2012;
Ojha, Keenan, Bright, Kerry, & Tiwari, 2016; Sánchez-Alonso, Moreno, & Careche, 2014). This
technique has also been suggested as an alternative method for the conventional determination of
drying degree upon the quality of chicken jerky (M. Li, Wang, Zhao, Qiao, Li, Sun, et al., 2014).

The objective of this study was to investigate the use of ultrasound as a pre-treatment prior to hot air convective drying of cultured and uncultured beef jerky. Modelling approaches were used to assess the influence of ultrasound frequency on the drying kinetics of beef jerky samples. Another objective of this study was to demonstrate a feasibility of using LF-NMR to determine water mobility and distribution of water during drying of cultured and uncultured beef jerky samples. Correlation analysis of transverse relaxation times and the moisture contents of dried beef jerky at different drying intervals were also determined to evaluate the drying degree of cultured and uncultured beef jerky samples.

78

79 2. Materials and methods

80 **2.1. Sample preparation and ultrasonic pre-treatment**

Beef used in this study was *Musculus Semitendinosus* which was obtained from a local supplier (Dublin 81 Meat Company, Blanchardstown, Co. Dublin, Ireland). Meat was stored at 4°C, sliced to 0.2 cm in 82 thickness using a meat slicer and were further cut by knife into slices of uniform dimensions (Length= 83 10 cm, Width = 4 cm). The beef slices were cured using two different curing solutions: (I) Cultured, 84 containing 70% water, L. sakei DSM 15831 culture, 1.5% salt, 1.0% sugar, 0.05% sodium nitrite and (II) 85 Uncultured, containing 70% water, 1.5% salt, 1.0% sugar, 0.05% sodium nitrite (based on raw meat 86 87 weight; v/w). The ingredients were thoroughly mixed, and samples from both cultured and uncultured treatment groups were subjected to ultrasonic (US) pre-treatments at frequencies of 25 kHz (Model: 88 Elma IT H5), 33 kHz (Model: Jencons-PLS S1000) and 45 kHz (Model: Elma IT H5) for 30 min at 89 comparable output power of circa 65 W along with a control (no US pre-treatment). US pre-treatments 90

91 were performed in ultrasonic bath systems maintained at a temperature of 30°C. All samples were
92 subsequently cured for 18 h at 4°C.

93

94 2.2. Drying of Beef Jerky

95 Cultured and uncultured cured beef jerky slices were dried using a hot air drying oven (Gallendkamp 96 Plus II, Weiss Technik, UK) at a temperature of 60°C for 4 h and using an air velocity which was 97 maintained at 0.3 m/s. Beef jerky samples were placed in trays and were transferred to the hot air 98 drying oven. Two slices from each treatment were withdrawn after every 30 min for 4 h and 99 subsequently weight using precise weighing balance (Sartorius, Germany), after weight determination 90 slices were placed back to the oven.

101

102 **2.3. Mathematical modelling**

Moisture content, on a dry basis, is the weight of moisture present in the product per unit weight of dry matter in the product. For drying experiments, where weight losses were recorded, the instantaneous moisture contents at any given time can be obtained from Eq.1:

106
$$M = \frac{(M_o+1)W_o}{W_t} - 1$$
 Eq. 1

Where W_o is the initial weight (g) of jerky sample after a curing period of 18 h, W_t is the weight (g) of sample at time t (min) and M_o is the initial moisture content (g water/g dry solids), respectively. The initial moisture content was determined using the hot air oven method as per AOAC. The data obtained experimentally for control and ultrasound pre-treated beef jerky slices from both uncultured and cultured groups were plotted as a dimensionless variable moisture ratio (MR) *versus* time as calculated from Eq. 2:

113 Moisture ratio (MR) =
$$\frac{(M_t - M_e)}{(M_o - M_e)}$$
 Eq.2

114

Where M_t is the moisture content at any time t, M_e the equilibrium moisture content and M_o is the initial moisture content and all expressed as g water/g dry solids. The value of the equilibrium moisture content (Me) is relatively small compared to M_t or M_o. Thus, Eq. (1) can be simplified as MR =Mt/Mo (Ju, El-Mashad, Fang, Pan, Xiao, Liu, et al., 2016; Xie, Mujumdar, Fang, Wang, Dai, Du, et al., 2017). Moisture diffusivity (D_f) for beef jerky samples were calculated by using Eq. 3 by analogy to the analytical solution to the Fick's second law of diffusion assuming negligible shrinkage, constant temperature, and constant moisture diffusivity (Zielinska & Michalska, 2016).

122
$$MR = \frac{8}{\pi^2} exp\left[-\frac{\pi^2 D_f t}{4L^2}\right]$$
 Eq.3

123

124 Where, D_f is the effective moisture diffusivity (m²/min), L is the thickness of the sliced beef (m).

125

Six empirical models were employed to describe drying kinetics were Henderson and Pabis, Wang and Singh, Page, Lewis (Newton), Weibull and Peleg (Table 1). The regression coefficient (R^2), Root mean square error (RMSE) and AICc (Akaike information criterion) values were calculated using Eq. 4 – 6, respectively. R², RMSE and AICc values were used as the primary criteria for measuring best model fit.

$$R^{2} = \frac{\sum_{i=1}^{N} (MR_{i} - MR_{pred,i}) \times \sum_{i=1}^{N} (MR_{i} - MR_{exp,i})}{\sqrt{\left[\sum_{i=1}^{N} (MR_{i} - MR_{pred,i})^{2}\right] \times \left[\sum_{i=1}^{N} (MR_{i} - MR_{exp,i})^{2}\right]}}$$
Eq.4

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^{N} \left(MR_{exp,i} - MR_{pred,i} \right)^2}$$
Eq.5

131
$$AICc = 2n - 2log_e(\mathcal{L}(\hat{\theta}|y) + \frac{2n(n+1)}{N-n-1})$$
 Eq. 6

Where, $MR_{exp,i}$ is moisture content observed experimentally and $MR_{pre,i}$ is predicted moisture content; *SSE* is the sum of squared error, $2log_e(\mathcal{L}(\hat{\theta}|y))$ is the log-likelihood at its maximum point of the model estimated, *N* and *n* represent the number of observations and parameters assessed, respectively.

137 **2.2. LF-NMR transverse relaxation measurements**

LF-NMR transverse relaxation measurements were carried out using a method described by McDonnell, Allen, Duggan, Arimi, Casey, Duane, et al. (2013) using a Maran Ultra instrument (Oxford Instruments, Abington, Oxfordshire, UK) resonating at a frequency of 23.2 MHz. Transverse relaxation (T₂) times were measured using Carr-Purcell-Meiboom-Gill (CPMG) pulse sequence with the resultant relaxation decays analysed by tri–exponential unsupervised fitting using RI Win–DXP software (Version 1.2.3 Oxford Instrument, Abington, Oxfordshire, UK).

144

145 2.4. Statistical data analysis

Analysis of variance (ANOVA) was performed using SAS procedure (SAS Version 9.1.3, statistical Analysis Systems). Tukey's multiple comparison was used to compare treatment means. Pearson's correlation coefficients were analysed to determine a relationship between moisture content (MC, %) and TD-NMR relaxation parameters. Correlation coefficients and significance values were determined using PROC CORR (SAS Version 9.1.3).

151

152 **3. Result and Discussion**

153 **3.1. Drying kinetics**

The effects of ultrasound frequencies on drying kinetics of marinated (uncultured and cultured) beef jerky slices are shown in Figure 1(a) & 1(b), respectively. In general, the moisture ratio (MR) decreased

132

156 exponentially with time for control and ultrasound pre-treated samples from both cultured and uncultured groups. A variable effect was observed on the drying curves, depending upon culture 157 treatment and ultrasonic frequency, as can be deduced from Figure 1. In general, a fast decrease in the 158 MR [-] was observed for all treatments at initial stages followed by a slow decrease with drying time 159 [min] at a drying temperature of 60°C. The moisture content decreased gradually for all samples, while 160 a fast decrease in moisture content was observed at a frequency of 45 kHz, followed by the control, 25 161 kHz and 33 kHz, respectively, for cultured samples. In the case of uncultured samples, control samples 162 showed the fastest decrease in moisture content, followed by 45 kHz, 33 kHz and 25 kHz. Previous 163 studies have shown that ultrasound pre-treatment can enhance drying rate for various food matrices 164 (Fernandes, Rodrigues, García-Pérez, & Cárcel, 2015; García-Pérez, Cárcel, Benedito, & Mulet, 2007). 165 166 However, the effect of ultrasound assisted drying depends largely on food matrix being dried, ultrasonic processing parameters and drying temperature. For example, ultrasound pre-treatment of various food 167 matrices showed a significant decrease in drying time, whereas in some cases, minor improvements 168 were reported (F. A. N. Fernandes, M. I. Gallão, & S. Rodrigues, 2008; A Mulet, Carcel, Sanjuan, & 169 Bon, 2003). Generally, during the drying process, migration of moisture is fast due to the evaporation of 170 surface moisture and decreases exponentially with an increase in drying time due to resistance offered 171 by the matrix to moisture movement. In a study conducted by Başlar, Kılıçlı, Toker, Sağdıç, and Arici 172 (2014), a significant decrease in drying time for ultrasound-assisted, vacuum-drying of chicken and 173 beef meat samples was observed. There are several supporting studies which show that ultrasound 174 enhances drying rate, owing to various mechanisms, thus modifying the diffusion boundary due to 175 acoustic pressure waves, oscillating viscosities, compressions and expansions of materials leading to 176 the formation of micro channels on surfaces which is required for fluid movement (Cárcel, García-177 Pérez, Benedito, & Mulet, 2012; A Mulet, Cárcel, Benedito, Rosselló, & Simal, 2003; Yao, 2016). 178 Variation in drying rate in this study may be due to the diffusion of marination solution into the meat 179 matrix due to the formation of micro channels on surfaces. Studies have shown that ultrasound 180

application can increase brine diffusion rate into a range of meat matrices (J. A. Cárcel, J. Benedito, J.
Bon, & A. Mulet, 2007; A. Mulet, Cárcel, Sanjuán, & Bon, 2003; César Ozuna, Puig, García-Pérez,
Mulet, & Cárcel, 2013). This may occur due to ultrasound assisted microinjection of brine into meat
through the formation of microjets as a result of asymmetric cavitation near the solid surface of the
product (Mason & Lorimer, 2002). However, it has been reported that no linear increase in diffusion of
brine solution into meat matrices was observed with respect to ultrasonic intensity (McDonnell, Lyng,
Arimi, & Allen, 2014).

The successful application of ultrasound on meat drying rates has been reported, however, the 188 mechanism of action is not yet clear. In this study, the effect of ultrasound frequency on drying rate for 189 both uncultured and cultured samples was probably due to the effect of ultrasound on lactobacillus 190 191 culture and diffusion of marination solution into the beef jerky samples. A significant moisture change was observed in marinated beef jerky samples after 18 h marination for ultrasonic pre-treated samples 192 compared to fresh beef (72.0%). For uncultured samples treated, at the lowest ultrasound frequency 193 (25 kHz), a gain of 6.04% was observed whereas for 33 kHz and 45 kHz pre-treatments moisture gains 194 of 5.60 % and 6.15%, respectively, were observed. In the case of cultured samples, no significant 195 moisture gain was observed for the control group, whereas moisture gains of 5.12%, 4.11% and 3.58% 196 were observed for ultrasound pre-treatments 33 kHz, 25 kHz and 45 kHz, respectively. 197

198 The observed changes were mainly due to uptake of marination solution. Similar gains in moisture have been reported for ultrasound pre-treatment prior to drying of fruit (F. A. Fernandes, M. I. Gallão, & S. 199 Rodrigues, 2008; Oliveira, Gallão, Rodrigues, & Fernandes, 2011). However, in some cases, solid 200 losses during ultrasound pre-treatments were also reported (Kadam, Tiwari, & O'Donnell, 2015; 201 Oliveira, Gallão, Rodrigues, & Fernandes, 2011). A concentration gradient of soluble solids between 202 beef slices and the marination solution resulted in water gain after pre-treatment and subsequent 203 incubation. Increase in moisture uptake has been reported for marinated beef products, including; pork. 204 poultry and beef, depending on composition of marination solution. Aktas and Kaya (2001) observed an 205

206 increase in moisture uptake for beef Longissimus dorsi muscle after marination at 4°C for 24 h. In this study, moisture uptake was observed for ultrasound pre-treated samples, whereas no significant 207 change in moisture uptake was observed for control samples. Research carried out by J. Cárcel, J. 208 209 Benedito, J. Bon, and A. Mulet (2007) on ultrasound-assisted brine diffusion of pork muscle showed no significant change in moisture uptake in samples subjected to static brining and found that moisture 210 uptake was dependent on ultrasonic intensity at a constant frequency of 20 kHz. Limited studies with 211 muscle-based foods have, like this present study, also highlighted moisture uptake as a result of 212 ultrasound pre-treatment in the case of Halal and non-Halal chicken breast (Leal-Ramos, Alarcon-Rojo, 213 Mason, Paniwnyk, & Alarjah, 2011). 214

215

3.2. Drying models

217

Non-linear regression analysis was carried out for six drying models as a function of drying time and 218 moisture ratio and various statistical parameters (R², RMSE and AICc) were determined to measure the 219 goodness of model fit. Model and statistical parameters (of drying models are listed in Table 1. For all 220 models R² ranged from 0.941 to 0.998, RMSE ranged from 0006 to 0.075 and AICc values ranged from 221 -105.40 to -50.43. For beef jerky samples investigated, the Wang and Singh model had the closest fit 222 to the drying experimental data, as evident from the high R^2 values (≥ 0.994) and the low RMSE (\leq 223 0.023) and low AICc (<-74.535) values for both cultured and uncultured jerky samples. Model 224 parameters (a and b) obtained by fitting the Wang and Singh model indicated that the relative 225 magnitude of the parameter accurately reflects drying behaviour. Drying constant values (a) were in 226 the range of -5.98×10^{-3} min⁻¹ to -3.2×10^{-3} min⁻¹ for uncultured and -6.73×10^{-3} min⁻¹ to -3.39×10^{-3} 227 min⁻¹ for cultured jerky samples, whereas, drying constant values (b) varied from -4.22×10^{-7} min⁻² to 228 9.28×10^{-6} min⁻² for uncultured and 1.23×10^{-6} min⁻² to 1.22×10^{-5} min⁻² cultured samples. Model 229 parameter (a) was lowest in the case of 45 kHz and highest for 33 kHz for cultured samples, whereas, 230

231 in the case of uncultured samples it was lowest for control samples and highest for 25 kHz samples. The lower (a) values reflect the higher moisture removal rates. A similar trend was also observed for 232 drying kinetics when fitted to other models. Various models have been proposed to model drying 233 kinetics of various food products, including; beef and chicken (Başlar, Kılıçlı, Toker, Sağdıç, & Arici, 234 2014). Drying behaviour can be predicted using a range of models, however, in this study the Wang 235 and Singh model was found to be the best fit. Best model fit can be judged based on various statistical 236 parameters, however; AICc and RMSE values were the criteria used for model section, because R² 237 alone cannot be judged for model fitting. AICc tends to have performance advantages over other 238 criteria for model fitting (Burnham, Anderson, & Huyvaert, 2011). AICc value rise with an increase in the 239 number of model parameters and the lower the AICc value, the better is the model performance. AICc 240 241 criteria has been adopted by several researchers to test the performance of drying kinetics models (Buttchereit, Stamer, Junge, & Thaller, 2010; Gowen, Abu-Ghannam, Frias, & Oliveira, 2008; Kadam, 242 Tiwari, & O'Donnell, 2015). The D_f value of the of cultured and uncultured beef samples ranged 243 between 0.90 to 1.33×10^{-8} m².min⁻¹ and 0.83 to 1.45×10^{-8} m².min⁻¹, respectively, as shown in Figure 244 2. The highest Df value was observed for control uncultured samples, and cultured samples pre-treated 245 at 45 kHz. Df value was found to increase with an increase in ultrasonic frequency in the case 246 uncultured samples, however, values remained significantly lower for control jerky samples in all cases. 247 Calculated Df values were within the range (10-8 to 10-10 m²/s) of those previously reported for drying of 248 biological materials (Baslar, Kılıclı, Toker, Sağdıc, & Arici, 2014; Zogzas, Maroulis, & Marinos-Kouris, 249 1996). 250

3.3. Water mobility by TD-NMR relaxometry

A representative LF-NMR T₂ transverse measurement for uncultured and cultured samples after 18 h marination (i.e. before drying) and after the 4 h drying period is shown in Figure 3. Distributed exponential analysis of curve obtained for various samples revealed the presence of three distinct peaks obtained at relaxation time ranges of 0–10 ms (T_{2b}), 10–100 ms (T_{21}) and >100 ms (T_{22})

respectively. These peaks can be attributed to various fractions of water present in beef jerky samples. 256 The first peak obtained at the shortest relaxation time (T_{2b}) represents bound water which is closely 257 associated with macromolecules (mainly proteins). The second peak at T₂₁ represents water present 258 259 within the dense myofibrillar protein matrix, whereas, the third peak at T₂₂ can be attributed to freewater present outside the myofibrillar protein matrix. Presence of three water fractions at relaxation 260 times and their association with muscle proteins has been previously reported (Huff-Lonergan & 261 Lonergan, 2005; Pearce, Rosenvold, Andersen, & Hopkins, 2011). Ultrasound pre-treatment showed a 262 shift in peaks for uncultured samples compared to cultured samples after 18 h of marination or 0 h 263 drying (Figure 3a&b). In the case of cultured control samples, a higher level of bound water fraction was 264 observed with a decrease in ultrasound pre-treated (Figure 3a), whereas, a shift in peaks were 265 266 observed in the case of uncultured samples (Figure 3b). In this study, the largest fraction of water present in beef jerky samples was observed at T₂₁ for cultured (in the range of 84.74–78.87%) and 267 uncultured (90.51 to 66.47%) samples after 18 h of marination, whereas, during drying at 60°C, the 268 proportion of water obtained at T_{21} was found to decrease with an increase in water proportion at T_{2b} . 269 An increase in water fraction at T₂₁ indicates an increase in the number of protons in the intra-270 myofibrillar space. Whereas, an increased water fraction at T₂₂ population indicates a similar rise in 271 number of protons, thereby representing an increase in the extra myofibrillar water population (Pearce, 272 Rosenvold, Andersen, & Hopkins, 2011). An increase in the proportion of water at T_{2b} suggests a 273 reduction in myofibrillar moisture and an increase in the bound water fraction obtained at T_{2b} due to the 274 removal of myofibril and free-moisture during drying. Similar increases in the bound water fraction, 275 indicating moisture mobility, was reported for beef granules during drying within a temperature range of 276 40-60°C (X. Li, Ma, Tao, Kong, & Li, 2012). Analysis of variance showed that culture and drying time 277 were the significant factors for all three relaxation times, whereas, ultrasound frequency was a 278 significant factor for T_{21} (p=0.0001), T_{22} (p=0.0010) and an insignificant factor for T_{2b} . Interaction effects 279 of drying time with culture and ultrasound frequency were significant for relaxation time and water 280

proportion. Similar, changes for water population at T_{21} and T_{22} relaxation times were also reported for ultrasound-assisted brining of pork samples in a study which concluded that a reduction in the T_{21} population and an increase in the T_{22} population may be due to increased salt intake and a change in physical properties of meat during the curing process (Ojha, Keenan, Bright, Kerry, & Tiwari, 2016). The increased intake of curing solution owing to ultrasound pre-treatment can cause an enlarged electrostatic repulsion within myofibrils, thereby resulting in water mobility and osmotic dehydration (Vestergaard, Andersen, & Adler-Nissen, 2007).

A plot of moisture content (MC, %) and T₂₂ relaxation time (free-water) indicated that a change in 288 289 relaxation time is related to the MC of cultured and uncultured beef jerky samples (Figure 4). Similarly, (2014) showed a relationship between T_{21} and T_{22} with water holding capacity of tofu. Hence, moisture 290 population data obtained from NMR can be used for indirect prediction of key moisture related 291 measurements. In this study, a strong positive correlation was observed between MC and T_{22} (*r*=0.790, 292 p < 0.0001) and proportion of water at T₂₂ (P₂) (r = 0.709, p < 0.0001) indicating that the MC of beef jerky 293 samples is mainly associated with free-water. Correlation analysis also showed a strong positive 294 relationship between drying time (h) and various water fractions and relaxation times (Table 2), with the 295 exception of T_{2b}, whereas, a significant negative relationship was observed between water fraction 296 associated with T_{2b}. This is probably due to a shift in relaxation time during the drying process. 297

298 4. Conclusion

This study demonstrates that ultrasound pre-treatment have significant effect on drying behaviour and moisture mobility of cultured and uncultured beef jerky samples. However, improvement in drying rates for both cultured and uncultured samples was not evident from the drying models generated. Significant increases in moisture gain after ultrasonic pre-treatment promoted brine uptake due to the combined effect of cavitation and concentration gradient phenomena. Among several drying models tested to predict the drying behaviour of beef jerky samples, the Wang and Singh drying model was found to be

- the best model as demonstrated by high R², low RMSE and AICc values. LF-NMR results showed
- moisture mobility during drying process with strong correlation with MC of jerky samples. LF-NMR can
- 307 be employed to elucidate changes in water distribution and moisture content of beef jerky samples.

308 Nomenclature

- 309
- 310 LF-NMR: Low Frequency-Nuclear Magnetic Resonance
- 311 W_o: Initial weight [g]
- 312 Wt: Weight [g] at time t
- 313 t: time [min]
- 314 M_o: Initial moisture content [g water/g dry solids]
- M_t : is the moisture content at any time t,
- 316 Me: Equilibrium moisture content
- 317 D_f: Effective moisture diffusivity [m²/min],
- 318 L: The thickness of the sliced beef [m]
- 319 MR: Moisture ratio [–]
- 320 *R*²: The regression coefficient,
- 321 RMSE: Root mean square error
- 322 AICc: Akaike information criterion
- 323 MC: Moisture content [%]
- 324 $T_{2b} T_{21}$ and T_{22} : Relaxation time (ms)
- 325
- 326
- 327 **5. References**

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Figure 1. Moisture ratio [MR] vs. drying time [min] for a) uncultured and (b) cultured beef jerky slices pre-treated at various ultrasonic frequencies [Control (◊), 25

462 kHz (\Box), 33 kHz (Δ) and 45 kHz (o) respectively].





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- 470 Figure 3. Distribution of multi exponentially fitted transverse relaxation (T₂) data for
- 471 uncultured (a b) and cultured (c d) beef jerky slices pre-treated at various ultrasonic
- 472 frequencies [Control (\diamond), 25 kHz (\Box), 33 kHz (Δ) and 45 kHz (o) respectively].

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Figure 4. Relationship between relaxation time (T_{22}) and moisture content of beef samples during drying of cultured (\bullet) and uncultured (\bullet) control (a) and ultrasound pre-treated beef jerky samples at 25 kHz (b), 33 kHz (c) and 45 kHz (d).

Table 1: Model parameters obtained from fitting drying models to beef jerky samples along with key statistical parameters

Model	Parameter		Uncu	Itured		Cultured			
		Control	25 kHz	33 kHz	45 kHz	Control	25 kHz	33 kHz	45 kHz
Henderson and Pabis	а	1.036	1.057	1.048	1.056	1.068	1.045	1.040	1.031
$MR = a \exp(-kt)$	k	8.18× 10 ⁻³	5.53× 10 ⁻³	6.83× 10 ⁻³	7.12× 10 ⁻³	7.82× 10 ⁻³	6.56× 10 ⁻³	5.09× 10 ⁻³	8.94× 10 ⁻³
	R ²	0.987	0.950	0.982	0.963	0.980	0.984	0.974	0.994
	RMSE	0.038	0.069	0.043	0.065	0.049	0.040	0.046	0.026
	AICc	-64.230	-50.435	-60.915	-51.810	-58.080	-62.630	-59.260	-71.965
Wang and Singh	а	-5.98× 10 ⁻³	-3.2× 10 ⁻³	-4.8× 10 ⁻³	-4.59× 10 ⁻³	-5.34× 10 ⁻³	-4.71× 10 ⁻³	-3.39× 10 ⁻³	-6.73× 10 ⁻³
$MR = 1 + at + bt^2$	b	9.28× 10⁻ ⁶	-4.2× 10 ⁻⁷	5.28 × 10 ⁻⁶	3.74 × 10 ⁻⁶	6.71× 10 ⁻⁶	5.22× 10 ⁻⁶	1.23× 10-6	1.22× 10⁻⁵
	R^2	0.999	0.994	0.999	0.996	0.997	0.999	0.998	1.000
	RMSE	0.010	0.023	0.011	0.020	0.019	0.011	0.012	0.006
	AICc	-95.105	-74.535	-90.375	-78.980	-79.380	-90.780	-89.385	-105.400
Page	k	2.49 × 10 ⁻³	3.17× 10-4	1.38× 10-3	6.56× 10-4	1.05× 10 ⁻³	1.38× 10 ⁻³	8.35× 10-4	3.76× 10-₃
$MR = exp(-kt^n)$	п	1.250	1.545	1.319	1.4785	1.392	1.3	1.3465	1.1725
	R^2	0.997	0.984	0.997	0.991	0.998	0.998	0.991	0.999
	RMSE	0.019	0.039	0.018	0.032	0.016	0.016	0.027	0.010
	AICc	-79.925	-62.775	-79.875	-67.365	-82.830	-83.160	-71.390	-92.330
Lewis (Newton)	k	7.86× 10-3	5.13× 10-3	6.45× 10-3	6.68× 10 ⁻³	7.27× 10-3	6.22× 10 ⁻³	4.82× 10-3	8.64× 10 ⁻³
$\mathbf{MR} = \exp(-\mathbf{kt})$	R^2	0.984	0.941	0.977	0.957	0.972	0.980	0.969	0.993
	RMSE	0.042	0.075	0.049	0.071	0.058	0.045	0.051	0.029
	AICc	-66.115	-52.575	-62.025	-53.965	-58.060	-63.680	-61.235	-73.125
Weibull	а	0.9737	0.95545	0.9788	0.955	0.9886	0.9792	0.97	0.9864
$MR = a \exp(-kt^n)$	k	1.81× 10-3	1.02× 10-4	9.56× 10-4	2.64× 10-4	8.83× 10-4	9.80× 10-4	4.47× 10-4	3.20× 10-3
	R^2	1.323	1.7515	1.302	1.0445	1.425	1.303	1.403	1.2025
	RMSE	0.998	0.988	0.997	0.994	0.998	0.998	0.993	0.999
	AICo	0.016	0.034	0.016	0.026	0.015	0.013	0.023	0.009
Deler	AICC	-78.310	-60.655	-77.165	-66.835	-78.525	-81.595	-68.960	-89.970
Peleg MR $- 1 - t/(a + ht)$	q b	0.48645	312.1 -0.03865	199.00	214.1 0.23575	0 37325	203.05	293.45 0.12383	122.00
$\frac{1}{(u+bt)}$	\tilde{R}^2	0.997	0.994	0.998	0.996	0.995	0.998	0.998	0.997
	RMSE	0.017	0.023	0.014	0.021	0.024	0.014	0.012	0.017
	AICc	-81.455	-74.535	-85.955	-77.730	-73.750	-84.980	-89.590	-80.710

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479 Table 2. Correlation analysis showing a relationship between various parameters

	Time (h)	P ₀	P 1	P ₂	T _{2b}	T ₂₁	T ₂₂	MC (%)
Time (h)	1.000	0.507***	-0.437**	-0.762****	0.206 ^{ns}	-0.400*	-0.822****	-0.929****
P 0		1.000	-0.994****	-0.468**	0.144 ^{ns}	0.282 ^{ns}	-0.305 ^{ns}	-0.615****
P 1			1.000	0.366*	-0.136 ^{ns}	-0.323*	0.249 ^{ns}	0.557***
P ₂				1.000	-0.123	0.205 ^{ns}	0.565**	0.709****
T _{2b}					1.000	0.386 ^{ns}	0.214 ^{ns}	-0.205 ^{ns}
T 21						1.000	0.702****	0.340*
T 22							1.000	0.790****
MC (%)								1.000
480	ns:Not significant	; *P<0.05; **F	P<0.01; ***P<0.0	001; ****P<0.0	001			
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Research Highlights

- 1. Drying behaviour is ultrasonic frequency dependent
- 2. Ultrasound can enhance marination rates
- 3. LF-NMR can be employed for water mobility and drying degree of beef jerky.