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1 **A 3D-CFD-heat-transfer-based model for the microbial inactivation of pasteurized food**
2 **products**

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9 **ABSTRACT**

10 This study proposes to couple a 3D-CFD and heat transfer finite elements model with the
11 microbial inactivation approach proposed by Geeraerd et al. (2000). The CFD-heat transfer
12 model was developed using thermophysical properties for both heating fluid (water) and the
13 processed sample (ground beef). The kinetic microbial parameters were estimated using
14 experimental data from the inactivation of *Escherichia coli* K12 in a packaged sample. The
15 proposed inactivation model was tested under more severe dynamic conditions than usual
16 (heating rates from 1 to 13 °C/min). The inactivation kinetic parameters were found
17 independent of the heating rate applied. In addition, the results reveal that the Geeraerd et al.
18 (2000) model without shoulder is sufficient to fit the experimental data. Such a model could
19 be beneficial in simulating microbial inactivation for food products, thus ensuring food safety
20 by limiting, as far as possible, overtreatment.

21 *Keywords: Microbial inactivation; Modeling; 3D-CFD; Heat transfer; Pasteurization.*

Nomenclature

a, b	dimensions of the elliptical sample (mm)
C_c	critical component related to the physiological state of the cells (-)
C_p	apparent specific heat ($\text{kJ kg}^{-1} \text{K}^{-1}$)
D_{ref}	decimal reduction time (s)
g	gravitational constant (m/s^2)
h_{air}	heat transfer coefficient of air ($\text{W m}^{-2} \text{K}^{-1}$)
k_{max}	specific inactivation rate (s^{-1})

L	sample length (mm)
LL	maximum likelihood
n	number of experimental data points
N	microbial population (CFU /g)
N_0	initial microbial population (CFU /g)
N_{simu}	simulated microbial population (CFU /g)
N_{exp}	experimental microbial population (CFU /g)
p	number of independently adjusted parameters within the model
P	absolute pressure (Pa)
Q	volumetric heat generation term (W/m^3)
R	radius (mm)
T	temperature ($^{\circ}\text{C}$)
T_0	initial temperature of product ($^{\circ}\text{C}$)
T_{∞}	ambient temperature ($^{\circ}\text{C}$)
t_i	simulated time (s)
T_{ref}	microbial inactivation reference temperature ($^{\circ}\text{C}$)
$T_{\text{cylinder cell}}$	temperature of lateral surfaces of the cylinder ($^{\circ}\text{C}$)
\vec{u}	velocity field (m/s)
u, v, w	spatial components of velocity field
V	volume of the meat sample (m^3)
x, y, z	spatial coordinates in the three dimensions (m)
z	thermal resistance constant ($^{\circ}\text{C}$)

Greek letters

ρ	density (kg/m^3)
μ	dynamic viscosity (Pa s)
σ	standard deviation of experimental data
λ	thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)

22 **1. Introduction**

23 Prediction of microbial inactivation during thermal treatment is crucial not only to ensure the
24 safety of food products but also to avoid overtreatment (Boillereaux, Curet, Hamoud-Agha, &
25 Simonin, 2013). Although conventional processing technologies produce safe products, they
26 can also lead to significant changes to the sensory and nutritional attributes of foods. In

27 today's quality conscious world, much attention is given to producing foods that retain
28 superior sensorial quality but must also remain safe (Bott, 2014; Stratakos & Koidis, 2015).
29 Consequently, the determination of the microbial destruction level that a thermal treatment
30 **can** deliver to a product requires both understanding of: (i) the heat transfer within the food
31 product and (ii) the destruction kinetics of the microorganism of interest (Valdramidis et al.,
32 2005). The necessity of such an approach is reinforced regarding the thermal heterogeneities
33 that occur during a pasteurization process.

34 A reliable inactivation parameter estimation is essential for building predictive models (Chen,
35 2013) and is necessary to establish **safe** minimal cooking-cooling conditions (Marcotte, Chen,
36 Grabowski, Ramaswamy, & Piette, 2008). The determination of these parameters has been the
37 subject of numerous studies, either under static (Ahmed, Conner, & Huffman, 1995; Smith,
38 Maurer, Orta-Ramirez, Ryser, & Smith, 2001) or dynamic conditions (Garre et al., 2018;
39 Hassani, Cebrián, Mañas, Condón, & Pagán, 2006; Juneja & Marks, 2003; Valdramidis,
40 Geeraerd, Bernaerts, & Van Impe, 2006; Valdramidis, Geeraerd, & Impe, 2007). Different
41 models have also been proposed to describe the microbial inactivation kinetics.

42 Numerous studies focusing on the estimation of microbial kinetic parameters are performed in
43 isothermal and dynamic conditions, but under ideal laboratory conditions (capillary tubes,
44 liquid inoculum and small sample mass). In real **time** industrial processing, food products are
45 often submitted to time-varying temperatures. Moreover, due to several factors affecting heat
46 transfer, i.e. geometry, thermophysical properties or external convection, significant thermal
47 heterogeneities can occur. In the literature, the effect of capillary tube diameters on the
48 inactivation parameters of *E. Coli*, in solid food, was investigated from a two-dimensional
49 heat transfer approach (Chung, Wang, & Tang, 2007).

50 A recent study by Garre et al., (2018) presented a mathematical model to describe the non-
51 isothermal microbial inactivation process. The model was dedicated to *Escherichia coli*
52 inactivation under linear heating rates of 1, 5, 15, 35 and 40 °C/min, from 35 to 70 °C. The
53 authors developed a mathematical model taking into account the thermotolerance of the
54 microbial cells. The model was able to describe the experimental data using a unique set of
55 model parameters.

56 Another investigation, Hamoud-Agha, Curet, Simonin, & Boillereaux, (2013) proposed to
57 estimate D_{ref} and z -value of Bigelow's (1921) equation. These parameters were estimated for
58 *E. coli* K12 inoculated into a model food under dynamic conditions (homogeneous

59 temperature distribution due to a very small solid sample). These parameters were included in
60 the Geeraerd et al., (2000) inactivation model coupled with a 3D-heat transfer model under
61 microwaves to illustrate the resulting inactivation heterogeneity.

62 The literature is rich concerning CFD and heat transfer modelling dedicated to pasteurization
63 of food products (Bhuvaneswari & Anandharamakrishnan, 2014; Cordioli, Rinaldi, Copelli,
64 Casoli, & Barbanti, 2014; Denys, Pieters, & Dewettinck, 2003; Dimou & Yanniotis, 2011).
65 However, except in Hamoud-Agha et al., (2013), it is rare to find these models coupled with
66 microbial inactivation equations and even less with real products under external dynamic
67 conditions. These elements reflect the originality of this work proposal. In this study, a 3D-
68 CFD and heat transfer approach is coupled with the Geeraerd et al. (2000) inactivation model.
69 External temperature conditions (heating rates from 1 to 13 °C/min) are applied to
70 *Escherichia coli* K12 inactivation in pre-packed ground beef. Model parameters are estimated
71 from experimental data using the Levenberg-Marquardt algorithm (Marquardt, 1963).

72 **2. Materials and methods**

73 ***2.1 Experimental section***

74 ***2.1.1 Sample preparation***

75 Raw ground beef (Beef Steak 5% fat, Charal[®], Cholet, France) was obtained from a local
76 retail store. The proportion of lean meat and fat was determined on the basis of information on
77 the package. Ground beef was provided in vacuum-packed portions of 100 g. The samples
78 were quickly frozen in a -20 °C freezer (Servathin, Carrières-sous-Poissy, France) and stored
79 in a freezing chamber at -20 ± 1 °C. Before each experiment, a bag of the sample was thawed
80 for 18 h in a cooling chamber (4 °C).

81 ***2.1.2 Preparation of bacteria inoculum***

82 The strain of *Escherichia coli* K12 (CIP 54.117, N° 11.612) used in this study was provided
83 by the Pasteur Institute, France, in the form of a freeze-dried sample. The strain was
84 rehydrated in 0.2 mL and inoculated in 5 mL of Brain Heart Infusion (BHI) (Panreac
85 Applichem – Darmstadt, Germany) at 37 °C for 18 h at 150 rpm (Incubator Shaker Series –
86 Model Excella E24, New Jersey, USA). A pre-culture was prepared with 0.1 mL of the
87 preceding suspension and inoculated in blood agar (Columbia + 5% Sheep blood - Bio-Rad -

88 Marnes-la-Coquette, France) at 37 °C, for 18 h. A clone of the pre-culture was then dissolved
89 in 2 mL of sterile water (0.85% NaCl - Medium, BioMerieux). 0.1 mL of this solution was
90 isolated in several Petri dishes of the blood agar with a sterile Pasteur pipette and incubated
91 **under** the same conditions. The colonies of this second isolation were removed with a sterile
92 swab and dissolved in 1 mL of the beef serum (Eurobio Serum Bovin, France) supplemented
93 with 20% (v/v) glycerol and stored frozen at -80 °C (Thermo Scientific TSE - USA).

94 To reactivate the strain, 0.1 mL of the stocked culture was inoculated into the blood agar
95 (Columbia agar + 5% sheep blood – 43041, BioMérieux, France) and incubated at 37 °C for
96 18 h. Fresh colonies were carefully removed with a sterile swab and dissolved in 5 mL of
97 medium suspension (Api 20150, BioMérieux, France) until a cellular concentration of
98 0.5 McFarland was reached (Densitometer Densimat, BioMérieux, Italy). The optical density
99 of 0.5 McFarland standard is comparable to that of a bacterial suspension concentration of
100 approximately 1.5×10^8 CFU/mL (CLSI, 2012).

101 *2.1.3 Inoculation of ground beef*

102 Raw ground beef samples (36 g) were removed aseptically from their refrigerated packaging
103 (7 °C) and transferred into sterile polypropylene sampling pouches with 4 g of bacterial
104 suspension. Ground beef was mixed manually to evenly distribute the inoculum throughout
105 the sample, producing an initial cell concentration of approximately 10^7 CFU/g. For heat
106 treatment, 3 g of inoculated ground beef samples were aseptically placed in packaging of
107 11.6 mm wide x 60 mm **long**.

108 *2.1.4 Heat treatment for microbial inactivation*

109 As illustrated in Figure 1, each sample was inserted **into** the cylindrical vessel and submitted
110 to a linear heating ramp in a programmable Peltier-based effect water bath (Hart Scientific
111 AOIP, FC 9105, USA). After the heating treatment, the sample was quickly cooled to 20 °C
112 by ice immersion. The temperature was measured with a thermocouple (type-K) **positioned**
113 near the bottom of the sample. The data were collected by a data logger (AOIP Datalog,
114 91133 Ris Orangis, France) with output recordings every 1 second. To obtain the inactivation
115 kinetics curves, eight final temperatures (50, 52, 54, 56, 58, 60, 62 and 64 °C) were
116 considered. Six different heating rates (1, 3, 5, 7, 10 and 13 °C/min) were applied. Triplicates
117 were carried out for each experiment.

118 2.1.5 *Microbial enumeration and expression of results*

119 The method of analysis was according to ISO 16649-1 (2001). This standard procedure
120 specifies a horizontal method for the enumeration of β -glucuronidase-positive *Escherichia*
121 *coli* in products intended for human consumption. It is based on a colony-count technique at
122 44 °C in a solid medium containing a chromogenic ingredient for detection of the β -
123 glucuronidase enzyme (ISO 16649-1, 2001).

124 Untreated inoculated ground beef and treated ground beef were aseptically removed from the
125 packaging and transferred to a sterile filtered stomacher bag (400 mL, BagPage, St. Nom,
126 France). The samples were diluted with 1:9 (w/w) of buffered peptone water (BK131HA,
127 Biokar Diagnostics, France) and stomached for 2 min at 230 rpm (Stomacher Lab Blender
128 InterscienceBagMixer® - 400, Grosseron, St. Herblain, France). Decimal serial dilutions were
129 performed in buffered peptone water. 1 mL of the initial suspension or decimal dilution was
130 inoculated **onto** duplicate plates of tryptone-bile-glucuronic (TBX) medium (BK146HA,
131 Biokar Diagnostics, France) and incubated between 18 h to 24 h at 44 °C. The presence of
132 blue colonies is considered to be β -glucuronidase-positive *Escherichia coli*.

133 Each dish containing less than 150 typical CFU was numbered and the result was calculated
134 as the arithmetic mean from two parallel plates. The results expressed to the base 10
135 logarithm of the microbial population for each temperature measurement were collected. The
136 detection limit was fixed to 1 CFU/g of the ground beef.

137 2.2 *Model design section*

138 2.2.1 *Heat transfer and computational fluid dynamics (CFD)*

139 The experimental methodology described in section 2.1.4 was considered in a CFD-heat
140 transfer model, with the following assumptions:

- 141 • *Assumption 1:* Water in the cylindrical vessel and the packaged sample were at the
142 same initial homogeneous temperature (7 °C).
- 143 • *Assumption 2:* The ground beef sample was considered as **being** homogeneous and
144 isotropic.
- 145 • *Assumption 3:* Specific heat, density and thermal conductivity of the ground beef were
146 considered as **being** constant within the temperature range (Pan & Paul Singh, 2001;
147 Tsai, Unklesbay, Unklesbay, & Clarke, 1998).

- 148 • *Assumption 4:* The geometry of the packaged sample was an ellipsoid-cylinder and the
 149 cross-section was considered homogeneous along the z-coordinate.
- 150 • *Assumption 5:* Axial symmetry was supposed for both thermal problems and fluid
 151 mechanics, limiting the size to one quarter of the experimental apparatus.
- 152 • *Assumption 6:* The mass transfer and the shrinkage of the sample were assumed **to be**
 153 negligible (in-package thermal treatment).
- 154 • *Assumption 7:* The package thickness was sufficiently thin to neglect its impact on the
 155 heat transfer (low thermal resistance).

156 The model is aimed at predicting the temperature profile when a heating rate is **applied** to the
 157 wall of the water bath heating cell. The model consists of two main computational domains:
 158 the first relates to liquid water which partially fills the cylindrical cell, the second is the
 159 ground beef sample located at the centre of the cell. As illustrated in Figure 2, the sample was
 160 immersed in water and its upper surface was exposed to air surrounding the medium (natural
 161 convection). The lateral surfaces of the cylinder are modelled with a first type boundary
 162 condition ($T = T_{\text{cylinder cell}}$).

163 2.2.2 Heat transfer modelling

164 Heat transfer is based on the general heat equation, which depends on thermophysical
 165 properties. For the ground beef sample, the heat equation is reduced to:

$$166 \quad \rho_{(sample)} C_{p(sample)} \frac{\partial T}{\partial t} = \text{div} (\lambda_{(sample)} \nabla T) \quad (1)$$

167 For liquid water, a convective term is added to take into account the variation of the
 168 thermophysical properties of water as a function of temperature:

$$169 \quad \rho_{(water)} C_{p(water)} \frac{\partial T}{\partial t} + \rho_{(water)} C_{p(water)} \vec{u} \nabla T = \text{div} (\lambda_{(water)} \nabla T) \quad (2)$$

170 Where \vec{u} is the velocity field at any point of the 3D sample domain (u, v, w components).

171 The initial and boundary conditions are defined with the following mathematical expressions:

$$172 \quad T = T_0, \quad t = 0, \quad \forall (x, y, z), \quad T_0 = 7^\circ\text{C} \quad (3)$$

$$173 \quad \lambda_{(sample/water)} \left. \frac{\partial T}{\partial z} \right|_{z=L} = h_{air} (T - T_\infty), \quad \forall (x, y), \forall t > 0, \quad T_\infty = 20^\circ\text{C} \quad (4)$$

174 The boundary condition expressed by equation 4 is applied to the upper surface of water and
 175 sample.

$$176 \quad T_{cylindrical\ cell} = T_0 + \frac{dT}{dt} t \begin{cases} \text{for } (x, y) \in [0; R] \text{ and } Z = 0, \quad \forall t > 0 \\ \text{for } x, y = R, \quad \forall t > 0, \text{ for } Z \in [0, L] \end{cases} \quad (5)$$

177 Figure 2 displays the coordinate system used to describe these mathematical expressions.

178 The convective heat transfer coefficient is due to natural convection between the surrounding
 179 air medium and the upper surface of the product. Free convection occurs only at the upper
 180 surface of both the water and the sample (Figure 2). An empirical correlation was conducted
 181 to compute the heat transfer coefficient on the upper cooled side of the cylinder. The air heat
 182 transfer coefficient (h_{air}) at the surface of the vertical cylinder with its axis running vertically
 183 is the same as that from a vertical plate, so long as the thermal boundary layer is thin
 184 (Churchill & Chu, 1975).

185 2.2.3 Fluid flow modelling

186 The fluid flow is due to natural convection occurring within the liquid water. This convective
 187 flow is modelled from the Navier Stokes equations (Newtonian fluid with incompressible
 188 flow). Both the continuity and momentum equations are described as follows:

$$189 \quad \begin{cases} \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0 & (continuity) \\ \rho_{water} \frac{d\vec{u}}{dt} = \rho_{water} g - \nabla P + \mu_{water} \Delta \vec{u} & (momentum) \end{cases} \quad (6)$$

190 with

$$191 \quad \frac{d\vec{u}}{dt} = \frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla) \vec{u} \quad (7)$$

192 The associated initial and boundary conditions are given as:

$$193 \quad P = P_{atm} + \rho_{water} g z \quad \text{at } t = 0, \quad \forall x, y, \quad \text{for } z \in [0; L] \quad (8)$$

$$194 \quad \vec{u} = 0 \quad \text{at } R = x, y, \quad \forall t > 0, \quad \text{for } z \in [0; L] \quad (9)$$

195 For validation purposes, coupled heat transfer and fluid flow were solved for a heating rate of
196 10 °C /min to compare the simulation to experimental results in the same conditions.

197 2.2.4 Thermophysical properties

198 Thermophysical properties of the sample used for modelling purposes are listed in Table 1.
199 The apparent specific heat (C_p) of ground beef (5% fat) was determined using a differential
200 scanning calorimetry (μ DSC VII Evo - Setaram Instrumentations, France) at a constant
201 pressure. Samples (~ 500 mg) were weighed in aluminium cells and sealed. A hermetically
202 sealed empty cell was used as a reference. The samples were equilibrated at 20 °C and then
203 heated to 65 °C, at a heating rate of 1 °C/min. The measurements were made in triplicate and
204 the average values were determined.

205 The density and the thermal conductivity of sample were taken from Pan & Paul Singh
206 (2001). The authors reported densities of 1006-1033 kg/m³ and thermal conductivities of
207 0.35-0.41 W m⁻¹ °C⁻¹ of ground beef (4.8% fat). These values do not change significantly as a
208 function of temperature range (between 5 and 75 °C).

209 Thermophysical properties of water were considered as temperature dependent (Green &
210 Perry, 2007). These properties were directly implemented in the model using polynomial
211 interpolation functions (Table 2).

212 2.2.5 Computational details for CFD simulations

213 Based on the finite element method, the model solved the partial differential equations
214 numerically, with the computational code COMSOL® Multiphysics 5.3a. In this study, the
215 geometry consisted of irregular shapes (ellipsoid-cylinder with axial symmetry) justifying the
216 use of the finite element method to simulate the coupled CFD-heat transfer phenomenon.

217 As shown in Figure 3, the generated mesh was designed for a 3D configuration, to ensure
218 good accuracy for the numerical resolution (522 830 tetrahedral elements with appropriate
219 boundary layers at the near-cell wall zone). Additional mesh sensitivity studies were
220 performed to obtain accurate number of mesh elements in relation to the computational cost.
221 The mesh independency to the numerical results was also verified. First, the fluid flow was
222 solved as a stationary study in order to initialize the velocity field for a better consistent initial
223 value.

224 Due to the temperature-dependent thermophysical properties of water, the coupled equations
 225 (Eq. 6 and 7) within fluid and heat transfer within the meat sample (Eq. 1) and water (Eq. 2)
 226 were solved as a time-dependent study with a direct solver (strong coupling). To ensure a
 227 good consistent initial value, the initial time step for the resolution was fixed at 10^{-7} s. Based
 228 on these computational details, the computational time did not exceed 4 hours 9 minutes and
 229 12 seconds on a Dell® Precision TM Workstation computer, equipped with 2×Intel® Xeon
 230 processors (8 cores), at 2.5 GHz, with 256 GB of RAM, running on Windows®8 Professional,
 231 64 bits.

232 Several simulations were performed depending on the heating rate being tested (from 1 to
 233 13 °C/min).

234 2.2.6 Microbial inactivation model

235 The dynamic non log-linear model developed by Geeraerd et al. (2000) was selected in this
 236 study. The reduced model, without the tailing effect, consists of two coupled ordinary
 237 differential equations as follows:

$$238 \quad \frac{dN}{dt} = -k_{\max} \left(\frac{1}{1 + C_c} \right) N \quad (10)$$

$$239 \quad \frac{dC_c}{dt} = -k_{\max} C_c \quad (11)$$

240 where N represents the microbial cell density (CFU/g). In equation 10, $\frac{dN}{dt}$ is forced to zero
 241 once the limit of detection (1 CFU/g) is reached. k_{\max} denotes the specific inactivation rate (s^{-1})
 242 and C_c is related to the physiological state of cells (-). A single value for $C_c(0)$ can be used
 243 for all experiments, since the methodology of inoculation is standardized. Consequently, a
 244 similar initial physiological state is expected for cells

245 The temperature effect on kinetic parameters (k_{\max}) was expressed using Bigelow's (1921)
 246 equation:

$$247 \quad k_{\max}(T) = \frac{\ln 10}{D_{ref}} \exp \left(\frac{\ln 10}{z} (T - T_{ref}) \right) \quad (12)$$

248 where D_{ref} denotes the decimal reduction time (s) at the reference temperature T_{ref} and z -value
 249 the thermal resistance constant ($^{\circ}\text{C}$).

250 2.2.7 Parameter estimation procedure

251 The approach consists in estimating the model parameters of Equation 12 by minimizing the
 252 residual sum of squares (RSS):

$$253 \quad RSS = \sum_{i=1}^n \left(\log \frac{\frac{1}{V} \int N_{simu}(t_i) dv}{N_0} - \log \frac{\overline{N}_{exp}(t_i)}{N_0} \right)^2 \quad (13)$$

254 where t_i are the times at which the thermocouple located in the sample reached the expected
 255 the temperatures (respectively 50, 52, 54, 56, 58, 60, 62 and 64 $^{\circ}\text{C}$), n is the number of
 256 experimental data points, N_{simu} is the volume-averaged value of the simulated cell density at t_i .
 257 N_{exp} is the volume-averaged value of the triplicate experimental cell density at t_i . V is the
 258 volume of the sample and N_0 is the initial cell density.

259 To minimize the RSS, the Levenberg-Marquardt algorithm (*lsqnonlin* available in
 260 Matlab[®]7.10) was used. This algorithm is based on the least-squares minimization technique
 261 and is an improvement of the Gauss-Newton algorithm. In addition, confidence intervals at
 262 95% were calculated for the estimated parameters using *nlparci* function available in
 263 Matlab[®]7.10.

264 2.2.8 Methodology used for model comparison and selection

265 Considering the model proposed by Geeraerd et al. (2000), it appears that parameters $C_c(0)$,
 266 D_{ref} and z -value can be estimated to fit the experimental data. Nevertheless, it is necessary to
 267 evaluate the relevance of the number of adjusted parameters. In this contribution, a non-linear
 268 least squares approach based on minimization of the RSS (residual sum of squares) is used. It
 269 evaluates the well-known Akaike Information Criterion (AIC) (Akaike, 1974), which
 270 provides a means for model selection. AIC is established as follows, on the assumption that
 271 the residues distribution is Gaussian:

$$272 \quad AIC = 2p - 2 \ln(LL) \quad (14)$$

273 Where p is number of independently adjusted parameters within the model and LL is the
 274 maximum likelihood. If the AIC criterion is used only with the aim of comparing models, the
 275 following simplified expression is used:

$$276 \quad AIC^* = 2 p + \frac{RSS}{\sigma^2} \quad (15)$$

277 When the residual sum of squares are almost equal for two different models, the minimum
 278 AIC estimation selects the model with the lowest number of parameters according to the
 279 “principle of parsimony” (Yamaoka, Nakagawa, & Uno, 1978). In addition, a variance σ^2 was
 280 calculated for each heating rate from the experimental results obtained in triplicate.

281 **3. Results and discussion**

282 ***3.1 Specific heat of ground beef***

283 The apparent specific heat (C_p) of ground beef at 5% fat was determined from 20 to 65 °C. As
 284 can be observed in Figure 4, the C_p values were found to be quite constant on the temperature
 285 range. The average measured value ($C_p = 3.69 \pm 0.01 \text{ kJ kg}^{-1} \text{ K}^{-1}$) is close to the reported one
 286 in the literature for meat product (Zhang, Lyng, Brunton, Morgan, & McKenna, 2004). The
 287 C_p average value was integrated directly into the heat transfer model.

288 ***3.2 Heat transfer and CFD model validation***

289 To validate the modelling approach, the experimental temperatures measured as a function of
 290 time in both the water and the sample were compared to the one simulated from the CFD-heat
 291 transfer model. Figure 5 illustrates the case for a heat treatment at 10 °C/min. The standard
 292 deviation of the experimental data was about ± 0.3 °C, corresponding to the measuring
 293 accuracy of the thermocouple used. Figure 5 depicts the linear setting temperature. The
 294 agreement between experimental and model temperatures of the water and the sample is
 295 noticeable. Non-negligible thermal delays (about 26 s and 57 s) can be observed respectively
 296 between the water and the sample and the set temperature and the sample, justifying the
 297 interest of such an approach when heating rates are applied.

298 The CFD modelling enabled prediction of the velocity fields due to the natural convection
 299 problem within a static enclosure. The Navier-Stokes equation was solved by considering a
 300 laminar flow. The norm of the velocity field, for example at 10 °C/min, is depicted in Figure 6

301 for different processing times. Fluid motion occurs (illustrated by black arrows in Figure 6),
302 because of the temperature dependent thermophysical properties of water. Recirculation of the
303 heating fluid leads to velocity gradients, with a maximum value of 2.5 mm/s around the
304 sample (norm of the velocity field). The Reynold number was calculated considering this
305 maximum fluid velocity of 10 °C/min with a value of 9.4. This result clearly demonstrates a
306 natural convection of water within a confined space, in which the flow regime is laminar.

307 Once the CFD approach is validated, the model illustrates the temperature gradients when
308 heating rates are applied. Figure 7 illustrates the temperatures at the final time of simulation
309 for the six different heating rates considered. Simulation was stopped when the simulated
310 temperature of the corresponding point to the thermocouple position reached 64 °C.

311 For all treatments the hottest zone was located at the surface of the sample, except for the
312 upper central part, due to contact with ambient air (cooling). The coldest zone was along the
313 central axis of the product. The temperature gap between the hottest and coldest zones
314 increased with the applied heating rates, so 4.3, 6.2, 8.1, 9.8, 12.4, and 15.8 °C for 1, 3, 5, 7,
315 10 and 13 °C/min, respectively.

316 To confirm the effect of temperature heterogeneities on the volumetric microbial inactivation,
317 equations 10, 11 and 12 were used. The simulation of the 3D local temperature distribution in
318 the meat sample (CFD and heat transfer model solved by COMSOL® and transposed to
319 MATLAB®) was performed to quantify the 3D local microbial inactivation. For example,
320 under a thermal treatment at 10 °C/min, the numerical result illustrated a thermal cartography
321 with hot and cold points. These temperature heterogeneities lead to heterogeneous local
322 microbial inactivation represented by the hatched area in Figure 8.

323 In Figure 8, a total inactivation can be seen for the hottest point after 5.8 minutes. For the
324 coldest point, the maximum inactivation achieved was of 2 Log₁₀, at the end of treatment.
325 However, it is not possible to experimentally validate these local microbial inactivation data,
326 at every geometrical point of the domain. Consequently, the use of a mathematical model to
327 describe the local microbial inactivation during a pasteurization process remains a relevant
328 tool. More, it confirms the importance of considering the volumetric distribution of
329 temperature in the meat product to accurately predict microbial inactivation.

3.3 Estimation of kinetic parameters of inactivation

330 Various results can be found in the literature concerning evaluation of kinetic parameters
331 (D_{ref} , T_{ref} and z -value) under isothermal conditions. In a study of heat resistance of
332 *Escherichia coli* 0157:H7 inoculated into 7% fat ground beef, Ahmed et al. (1995) obtained a
333 D_{ref} , by linear regression, of 684 s (at 55 °C) and 27 s (at 60 °C), and a z -value equal to
334 4.78 °C. In the same strain, but in ground beef with 4.8% fat, Smith et al. (2001) calculated by
335 first-order kinetics a D_{ref} equal to 73 s (at 58 °C) and 19 s (at 61 °C), for a z -value of 3.79 °C.
336 These variations might be attributed to differences in the strains of *E. coli* used, the fat content
337 of meat, along with differences in recovery methods and sample size (Smith et al., 2001;
338 Stringer, George, & Peck, 2000). In both studies, the values of D_{ref} differ greatly whereas z -
339 value remains in the same order of magnitude.

341 In this study, ground beef with 5% fat was used and the heating rates **described above** were
342 applied. Despite these dynamical conditions, in a first approach, the parameters $D_{ref} = 73$ s, z -
343 value = 3.79 °C and $T_{ref} = 58$ °C obtained by Smith et al. (2001) in isothermal conditions and
344 $C_c(0) = 0.23$, as reported in Hamoud-Agha et al. (2013), were used in **the** Geeraerd et al.
345 (2000) model. Results are illustrated in Figure 9. Black circles illustrate the experimental data
346 in triplicate, whereas the **predictions** obtained using the aforementioned parameters are
347 represented by broken blue lines. **Note** that the model underestimates the experimental
348 inactivations, but this underestimation has the same order of magnitude whatever the heating
349 rate.

350 A second approach consisted in estimating the model parameters **from the dynamic**
351 **experiments**. First, the triplet parameters $C_c(0)$, D_{ref} and z -value were estimated. **Poschet et al.,**
352 **(2005) showed that the uncertainties on D_{ref} and z -value are minimal when T_{ref} is equal to the**
353 **middle of the lethal experimental temperature range. As D_{ref} is closely linked to T_{ref} , this value**
354 **was fixed at 58 °C, as defined in Smith et al. (2001).** The $C_c(0)$ value was estimated with a
355 lower bound equal to zero. In table 3, $C_c(0)$ is not **significantly different from zero** for all
356 heating rates. Consequently, equation (11) can be removed and the Geeraerd et al. (2000)
357 model becomes a classical first order inactivation model.

358 With the Bigelow model, the D_{ref} and z -values were estimated (Table 4). It should be noted
359 that these AIC^* values are lower than the AIC^* values obtained in previous estimate procedure
360 with three parameters (Table 3). The confidence intervals for D_{ref} increase for extreme heating
361 rates (10 and 13 °C/min), certainly due to the large thermal heterogeneities. Another

362 **explanation** is based on Van Derlinden, Balsa-Canto, & Van Impe, (2012), where the authors
363 discussed the link between parameter identifiability and the shape of the temperature profile.
364 The results of z -value indicated in Table 4 are consistent with those presented by Smith et al.
365 (2001), for a similar product at 58 °C (z -value = 3.79 °C). Indeed, this value (3.79 °C) is
366 almost included in the confidence intervals and will thus be considered hereafter.

367 With the estimation limited to D_{ref} , the confidence intervals are widely reduced, even if they
368 remain significant for heating rates of 10 and 13 °C /min (12.52 s and 8.75 s). These results
369 are summarized in Table 5. Compared to Table 4, quite similar or lower values of the AIC*
370 were obtained. **It appears that adjusting D_{ref} is sufficient to fit the experiments without degrading
371 the AIC* values. Moreover, it also reduces the parameter uncertainties as verified with 95%
372 confidence intervals.**

373 Thereby, for one estimated parameter, the D_{ref} values remain in the same order of magnitude.
374 **Consequently, the average of the previously estimated D_{ref} values (Table 5) of 47 s was used.**
375 The predicted inactivation curves obtained in this case are presented in black continuous line
376 in Figure 9. They are very close to inactivation results obtained with each estimated D_{ref} (red
377 **dotted line** is superimposed on black continuous line in figure 9). **Hence, for D_{ref} values
378 ranging from 40 to 50 s, considering a unique value of 47 s does not significantly affect the
379 inactivation predictions.**

380 **Finally, results reveal that the k_{max} expression with temperature dependence proposed by
381 Bigelow is sufficient to fit experimental data obtained in dynamic conditions with large
382 temperature heterogeneities (>15 °C for 13 °C /min), provided that these heterogeneities are
383 considered in the prediction model. Constant values for T_{ref} , z and D_{ref} can be considered for
384 all heating rates in the range 1 to 13 °C /min. Additionally, T_{ref} and z -values of literature
385 (Smith et al., 2001) were used with only a fitting of D_{ref} .**

386 **4. Conclusion**

387 **The contribution of this work lies in the integration of experimental and numerical approaches
388 dedicated to the real-food pasteurization process including both thermal and inactivation
389 kinetics with high heating rates.**

390 The originality of this paper lies (*i*) in the integration of three-dimensional heat transfer
391 conditions in parameter estimations applied to the inactivation model and (*ii*) in the
392 application of extreme heating rates to challenge the validity of the Geeraerd modelling

393 approach developed for isothermal conditions. The heat transfer model was validated during
394 blank tests, and the volume-average of logarithmic inactivation was considered to be
395 comparable with experimental data. In addition, the experiments were carried out on real
396 ground beef, and not in capillary tubes, as usually proposed. The inactivation model was
397 coupled with heat transfer to design a 3D simulator to predict the spatial distribution of
398 logarithmic inactivation of *E. coli* in a meat sample.

399 Such a 3D simulator illustrates how the temperature gradients can provoke large inactivation
400 heterogeneities, and thus the necessity to consider them in realistic pasteurization conditions.
401 The satisfactory agreement between model and experiments allows coupling of the modelling
402 approach with process control procedures, allowing energy supply to reach the expected
403 microbial inactivation while limiting temperature heterogeneities. Such an objective could be
404 reached for example by using combined energy sources with volumetric heating to avoid
405 overtreatment at the surface of the food product (microwaves, ohmic heating).

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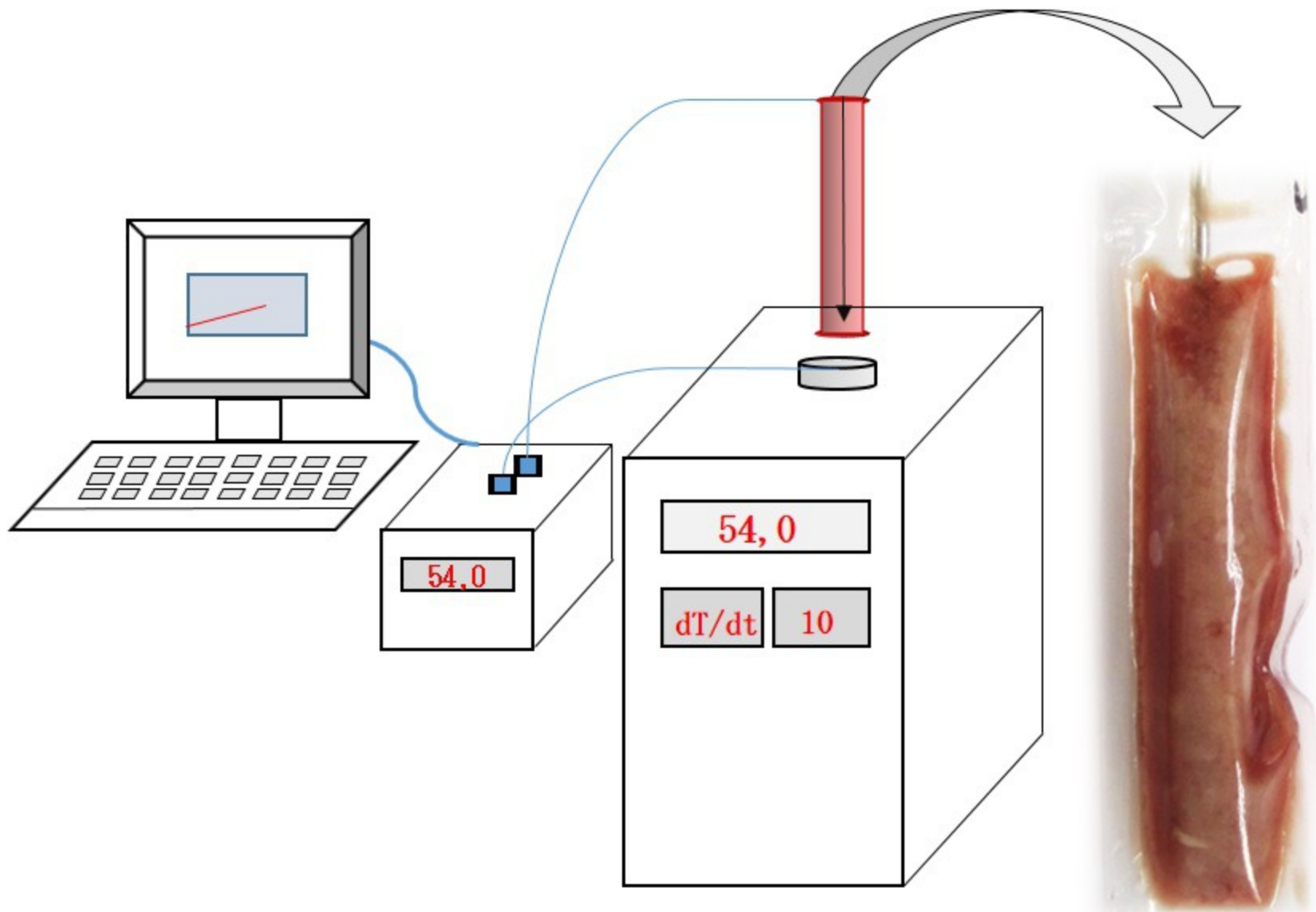
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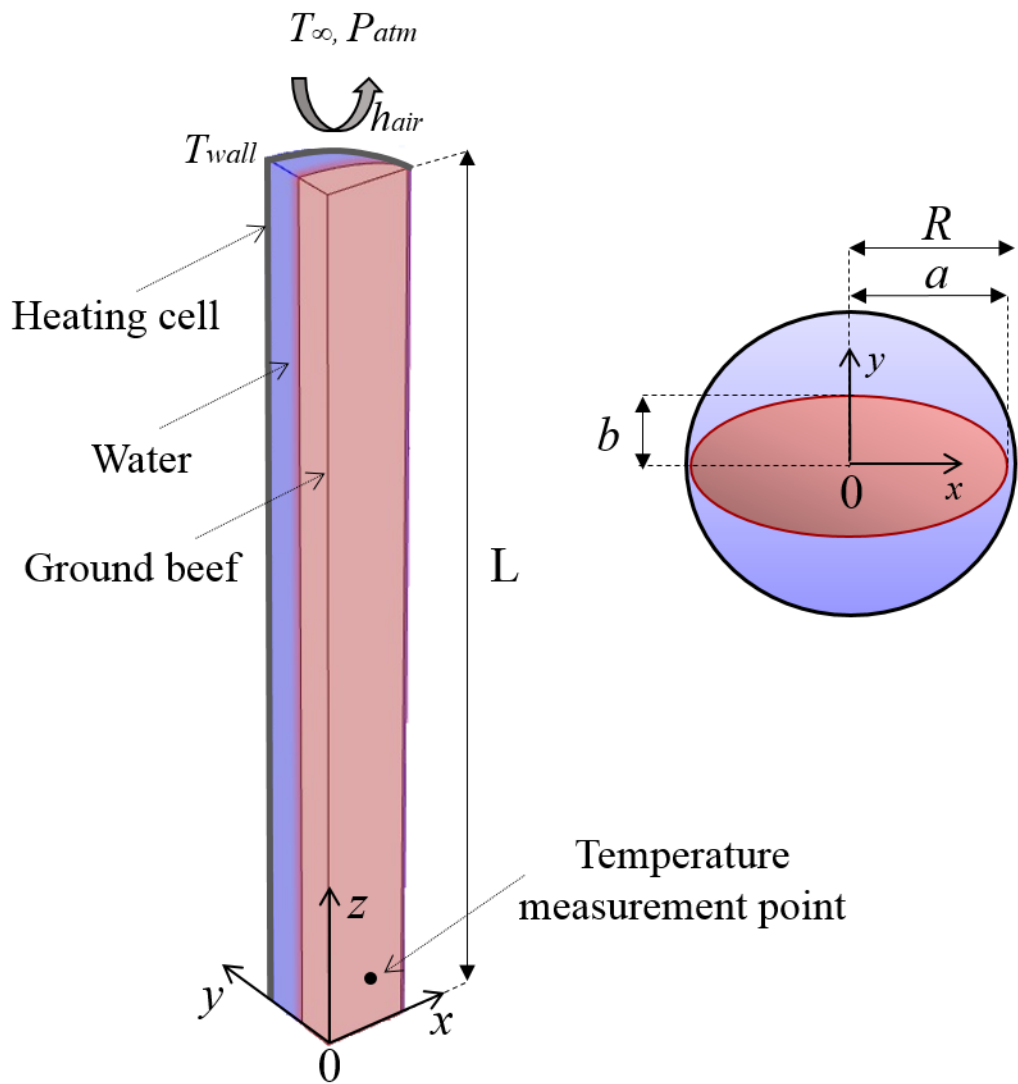
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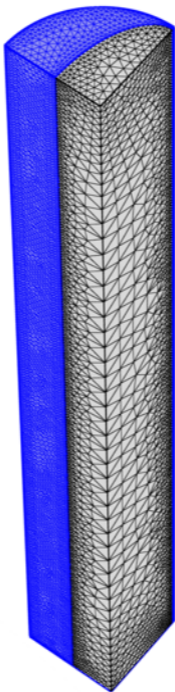
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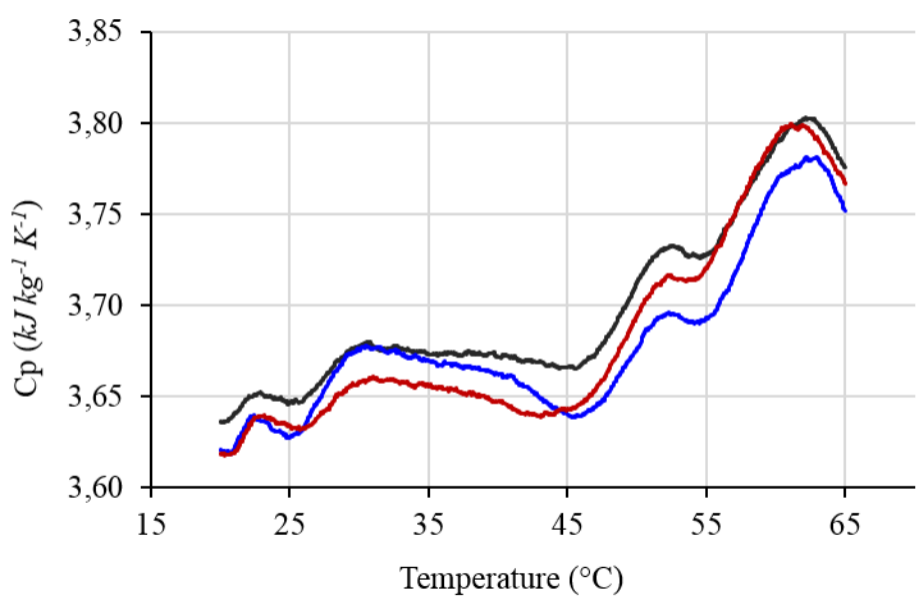
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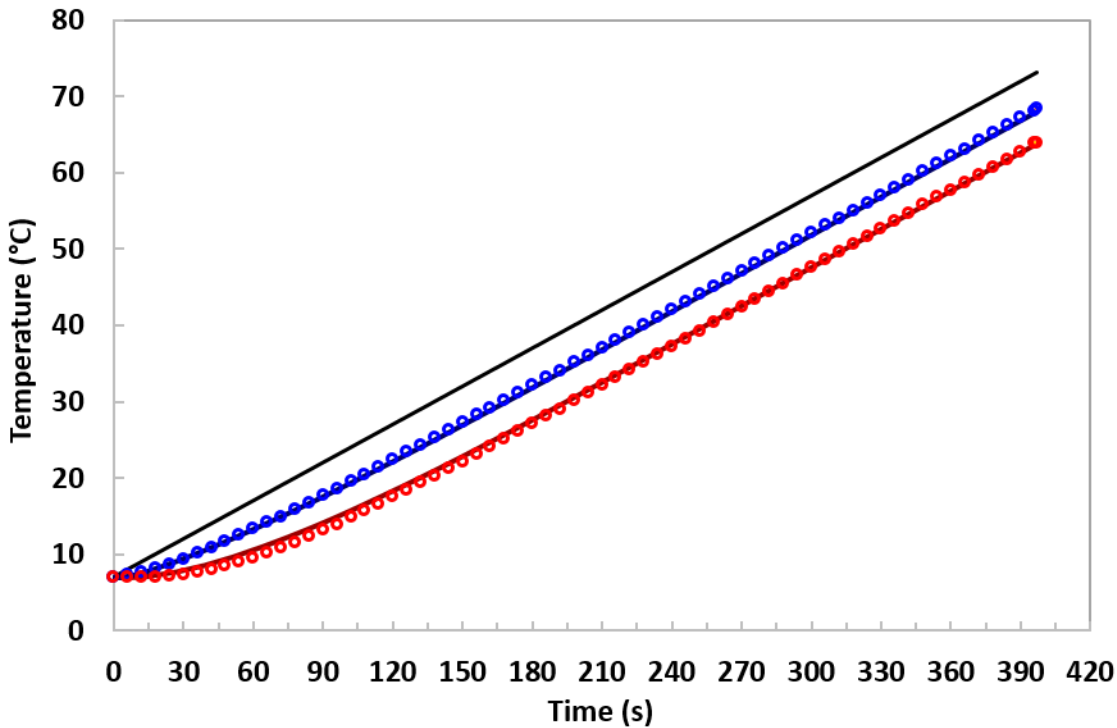
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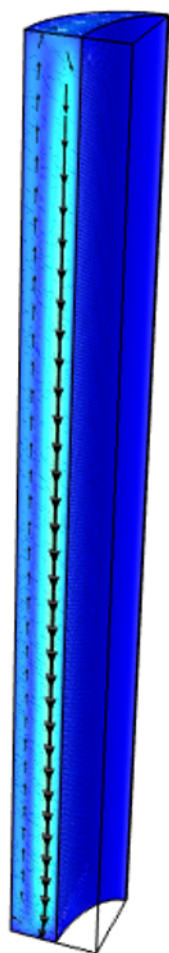
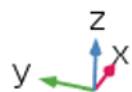








mm/s



$t = 30$ s



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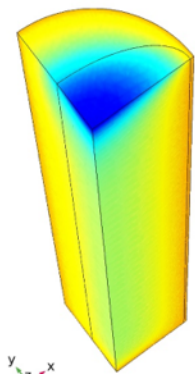
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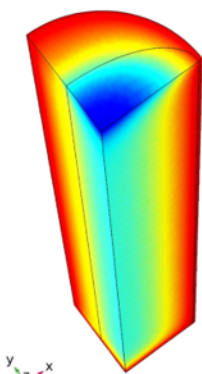
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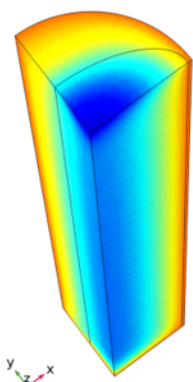
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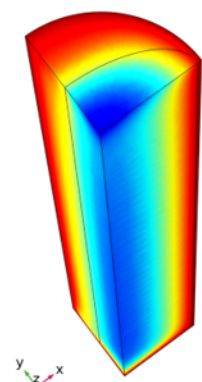
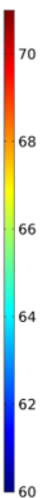
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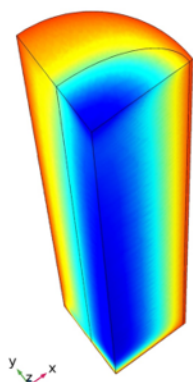
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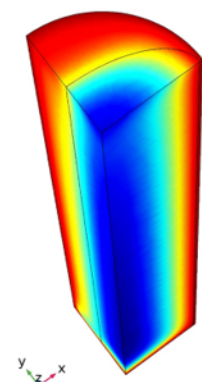
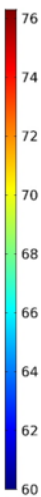
5 °C/min



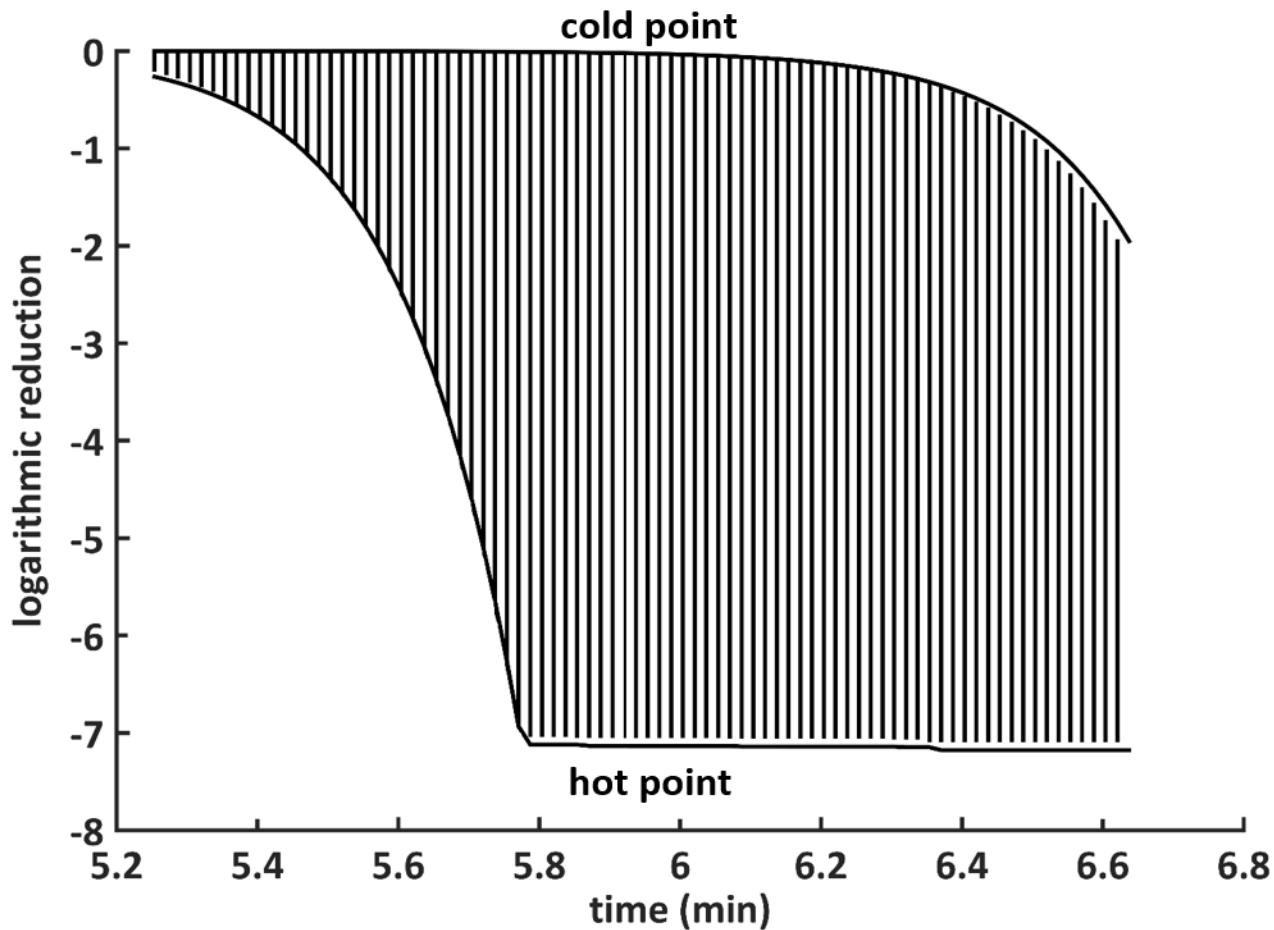
7 °C/min



10 °C/min



13 °C/min



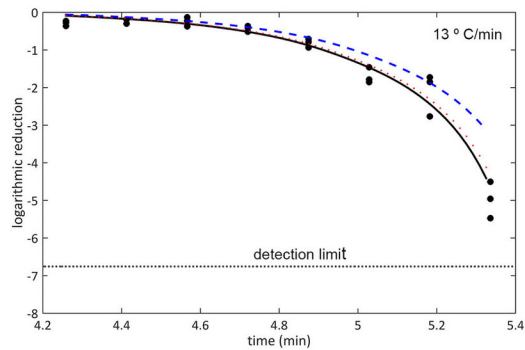
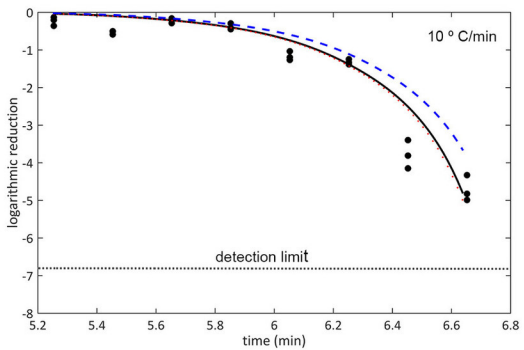
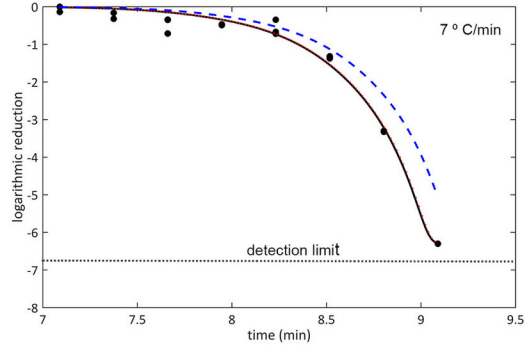
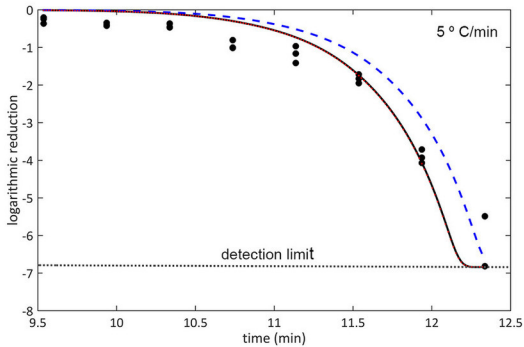
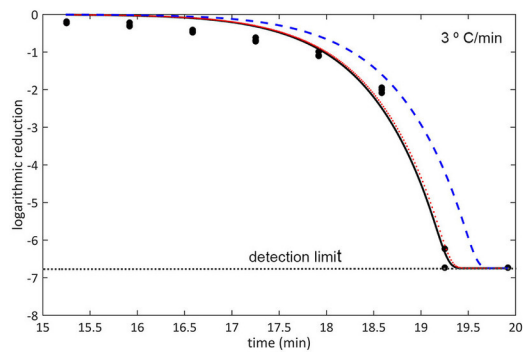
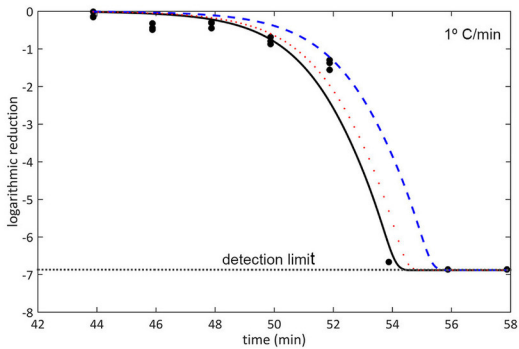


Table 1. Thermophysical properties of ground beef.

Properties	Value	Unit	Source
Apparent specific heat (C_p)	3.69 ± 0.01	$\text{kJ kg}^{-1} \text{K}^{-1}$	Experimental
Thermal conductivity (λ)	0.35	$\text{W m}^{-1} \text{°C}^{-1}$	Pan and Singh (2001).
Density (ρ)	1006	kg/m^3	Pan and Singh (2001).

Table 2. Thermophysical properties of water as a function of temperature (Kelvin).

Functions of the properties*	Unit
$C_p = 12010.15 - 80.41 \times T + 0.31 \times T^2 - 5.38 \times 10^{-4} \times T^3 + 3.62 \times 10^{-7} \times T^4$	$\text{J kg}^{-1} \text{K}^{-1}$
$\lambda = -0.87 + 8.95 \times 10^{-3} \times T - 1.58 \times 10^{-5} \times T^2 + 7.97 \times 10^{-9} \times T^3$	$\text{W m}^{-1} \text{°K}^{-1}$
$\rho = 838.47 + 1.40 \times T - 3.01 \times 10^{-3} \times T^2 + 3.72 \times 10^{-7} \times T^3$	kg /m^3

*Green & Perry, 2007

Table 3. Estimated values of D_{ref} , z -value and $C_c(0)$, for $T_{\text{ref}} = 58 \text{ °C}$. Standard deviation of experimental data (σ^2), 95% confidence intervals (CI) and simplified Akaike Information Criterion (AIC*).

Heating rate (°C /min)	σ^2	D_{ref} (s)	CI D_{ref}	z -value (°C)	CI z -value	$C_c(0)$	CI $C_c(0)$	AIC*
1	0.24	33.1	3.8	1.96	1.41	0	0.30	11.8
3	0.20	48.9	6.8	3.81	0.63	0	0.21	12.2
5	0.23	41.7	5.3	7.32	1.53	0.11	0.15	10.2
7	0.34	45.7	8.1	3.59	0.61	0.21	0.18	9.3
10	0.50	56.2	22.4	6.19	2.3	0.08	0.17	11
13	0.63	74.7	33.1	2.99	0.69	0.04	0.15	9.2

Table 4. Estimated values of D_{ref} and z -value ($T_{\text{ref}} = 58 \text{ }^\circ\text{C}$). Standard deviation of experimental data (σ^2), 95% confidence intervals (CI) and simplified Akaike Information Criterion (AIC*).

Heating rate ($^\circ\text{C}/\text{min}$)	σ^2	D_{ref} (s)	CI D_{ref}	z -value ($^\circ\text{C}$)	CI z -value	AIC*
1	0.24	33.1	3.8	1.96	1.41	8.8
3	0.20	48.9	6.8	3.81	0.63	9.2
5	0.23	40.8	5.1	6.70	1.41	7.2
7	0.34	31.4	12.4	4.80	0.81	7.2
10	0.50	62.1	23.2	5.98	2.65	8.2
13	0.63	73.1	32.6	3.02	0.71	6.6

Table 5. Estimated values of D_{ref} ($z = 3.79$ and $T_{\text{ref}} = 58 \text{ }^\circ\text{C}$). Standard deviation of experimental data (σ^2), 95% confidence intervals (CI) and simplified Akaike Information Criterion (AIC*).

Heating rate ($^\circ\text{C}/\text{min}$)	σ^2	D_{ref} (s)	CI D_{ref}	AIC*
1	0.24	40.1	0.03	9
3	0.20	47.5	0.38	6.2
5	0.23	50.0	0.59	8.2
7	0.34	50.0	0.48	3.6
10	0.50	50.1	12.52	6.8
13	0.63	46.7	8.75	3.6