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1 **A deterministic approach for predicting the transformation of starch suspensions in**  
2 **tubular heat exchangers**

3

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17

18

19 **Abstract**

20 Numerical modeling of fluid flow, heat transfer and transformation is conducted for an  
21 aqueous suspension of starch granules running throughout a four-heating-section tubular  
22 exchanger. The numerical model considers the transformation kinetics and the rheological  
23 behavior of the starch suspension as determined from laboratory work; further, the model  
24 includes the geometrical characteristics of the heat exchanger, as well as the operating  
25 conditions which were considered in running it. Model predictions are compared with results  
26 from experimental work, after sampling the starch suspension under thermal processing and  
27 later characterizing it using laboratory techniques. The numerical model predicts the bulk  
28 swelling state of the starch suspension at a level of agreement ( $-41\%$  in volume mean  
29 diameter increase) which is better than the one reached after assuming plug-flow and radially-  
30 independent temperature ( $-66\%$ ). The inclusion of two-way coupling between the relevant  
31 phenomena constitutes therefore a positive improvement.

32

33

## 34 **1. Introduction**

35 Manufacturing food products in a reproducible manner requires understanding the  
36 mechanisms which drive the transformation of the ingredients along the processing pathway.  
37 For instance, in dairy desserts the milk is mixed with starch, to which carrageenan is added as  
38 a gelling agent (Doublier and Durand, 2008). Experimental work at laboratory scale has been  
39 conducted in order to study the influence of thermal treatment on the structure of whey  
40 proteins (Tolkach and Kulozik, 2007; Nicolai et al., 2011), of starch granules (Ratnayake and  
41 Jackson, 2008; Liu et al., 2009; Schirmer et al., 2013) and of carrageenan chains (Piculell,  
42 2006; Nunez-Santiago et al., 2011). The interactions occurring between these ingredients have  
43 been studied and, for a variety of blends, the rheological behavior was analyzed along the  
44 batch thermal history (Verbeke et al., 2006; Doublier and Durand, 2008; Noisuwan et al.,  
45 2009; Huc et al., 2014; Matignon et al., 2014a, 2014b, 2015). The preparation of dairy  
46 desserts at industrial scale includes, among other steps, thermal processing under continuous  
47 flow. The phenomena of fluid flow, heat transfer and product transformation are strongly  
48 coupled within heat exchangers: transformation affects the characteristics of the product (e.g.,  
49 particle sizes), with consequences on its rheological behavior and hence on the velocity field  
50 inside the processing unit. In turn, fluid flow influences heat transfer while the temperature  
51 level drives the transformation rate. As a consequence, the final structure of the liquid product  
52 can depend not only on the interactions between the ingredients but also on the geometry and  
53 on the thermal and dynamical operating conditions associated with the heat exchanger.

54 To predict the transformation of a liquid food product along its thermo-dynamical  
55 history is a challenging task. Physics-based and observation-based modeling approaches  
56 constitute complementary tools for food product, processes, and equipment designers.  
57 Physics-based models especially can provide a level of insight that is often not possible

58 experimentally (Datta, 2008). The phenomena of fluid flow, heat transfer, and product  
59 transformation need to be considered in the case of the thermal processing of liquid food  
60 products under continuous flow. The rheological behavior of liquid food products is typically  
61 non-Newtonian and temperature-dependent; therefore, it is difficult to derive analytical  
62 solutions for the conservation equations of mass, momentum and energy.

63 Computational Fluid Dynamics (CFD) has been employed to study a number of  
64 problems in food engineering and related disciplines (Norton and Sun, 2006; Galeazzo et al.,  
65 2006; Lemus-Mondaca et al., 2011; Khatir et al., 2013; Norton et al., 2013; Rocha et al.,  
66 2013; Defraeye, 2014). However, limited efforts were devoted to coupled problems involving  
67 liquid food products whose rheological behavior evolves as the transformation progresses  
68 along the processing pathway. Bouvier et al. (2014) studied the thermal denaturation-  
69 aggregation of whey proteins flowing under turbulent flow in a plate heat exchanger; those  
70 authors obtained promising agreement between model predictions and observations of the  
71 transformation state reached by the liquid product, after assuming a simple rheological  
72 behavior for the whey protein solutions under consideration (Newtonian, temperature-  
73 dependent). Liao et al. (2000) considered a more complex rheological behavior and developed  
74 a numerical model coupling fluid flow, heat transfer and gelatinization of waxy rice starch  
75 paste; the apparent viscosity of the product was described through a power law, and both the  
76 consistency coefficient and the behavior index were assumed to be temperature-dependent.  
77 We can argue that describing the rheological behavior as a function of the local temperature is  
78 not a robust strategy. It can be shown that the characteristics of the liquid product, and hence  
79 its rheological behavior at a given position in the heat exchanger, depend not only on the local  
80 conditions but also on the thermal (and eventually dynamical) histories associated with the  
81 fluid parcels running at this position (Chantoiseau et al., 2012; Plana-Fattori et al., 2013). The  
82 two-way coupling between product transformation, fluid flow and heat transfer phenomena

83 via the rheological behavior constitutes a robust strategy for predicting the transformation of a  
84 liquid product along its thermo-dynamical history.

85 In the logical continuation of previous efforts (Plana-Fattori et al., 2013), this study  
86 presents a numerical model for predicting the transformation of a starch suspension running  
87 throughout an existing four-heating-section tubular exchanger. We focus on a given starch  
88 suspension (type and concentration), and the parameters relevant to the numerical model are  
89 estimated through our own experimental protocol at laboratory scale. Both the swelling  
90 kinetics and the rheological behavior are represented more realistically than in that previous  
91 study; on the one hand, a threshold temperature is assumed for the swelling kinetics rate  
92 (rather than an Arrhenius-like approximation); on the other hand, the progressive shear-  
93 thinning behavior of the starch suspension is described. Later, we compare model predictions  
94 of the swelling state reached by the suspension, with observations obtained at pilot scale by  
95 running the tubular heat exchanger whose geometry and operating conditions are considered  
96 in the numerical model. The objective of this study was double: firstly to implement the  
97 coupled model for thermal processing at pilot scale by applying key model parameters  
98 obtained from experimental work at laboratory scale, and secondly to assess the improvement  
99 that this coupled model represents relatively to a simpler approach, based on the assumption  
100 of plug-flow and radially-independent temperatures. Results constitute a meaningful step  
101 towards the implementation of numerical models able to represent the variety of dynamical  
102 and thermal conditions experienced by realistic food fluid parcels during their pathway in the  
103 processing unit. They will be useful for the design of starch suspension thermo-mechanical  
104 treatments.

105

## 106 **2. Coupled physical problem**

107 We are interested in the phenomena of fluid flow, heat transfer and liquid product

108 transformation occurring within a tubular heat exchanger which is running under stationary  
 109 conditions. Conservation equations for mass, momentum and energy can be expressed as:

$$110 \quad \vec{\nabla} \cdot (\rho \vec{u}) = 0 \quad (1)$$

$$111 \quad \rho (\vec{u} \cdot \vec{\nabla}) \vec{u} = \vec{\nabla} \cdot \left( -p \vec{I} + \eta \left( \vec{\nabla} \vec{u} + (\vec{\nabla} \vec{u})^T \right) - \frac{2}{3} \eta (\vec{\nabla} \cdot \vec{u}) \vec{I} \right) \quad (2)$$

$$112 \quad \rho C_P (\vec{u} \cdot \vec{\nabla}) T = \vec{\nabla} \cdot (\lambda \vec{\nabla} T) \quad (3)$$

113 where  $\vec{u}$  is the velocity (magnitude in m/s),  $p$  is the pressure (Pa),  $T$  is the temperature (K);  $\rho$   
 114 is the product's density ( $\text{kg} \cdot \text{m}^{-3}$ ),  $\eta$  its apparent viscosity (Pa.s),  $C_P$  its specific heat capacity  
 115 ( $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ ), and  $\lambda$  its thermal conductivity ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ ).

116 In this exploratory study focus is on the transformation of an aqueous suspension of  
 117 chemically-modified waxy maize starch. Under heating, this liquid product undergoes a quite  
 118 simple transformation: starch granules swell without either rupture of swollen granules or  
 119 release of soluble species in water (Matignon et al., 2015). Hence, the volume occupied by the  
 120 granules can indicate the transformation state regarding its influence on the apparent viscosity  
 121 of the starch suspension. At a given time in the thermal history of a suspension droplet, the  
 122 solid volume fraction  $\Phi$  can be estimated as:

$$123 \quad \Phi = \Phi_0 (D/D_0)^3 \quad (4)$$

124 where  $D$  ( $\mu\text{m}$ ) is the volume mean diameter of starch granules; values  $D_0$  and  $\Phi_0$  correspond  
 125 to the condition of untreated starch, before any thermal treatment. The transformation state  
 126 associated with the starch suspension can also be indicated by the dimensionless value of the  
 127 volume mean diameter (also known as swelling degree,  $S$ ):

$$128 \quad S = (D - D_0) / (D_{MAX} - D_0) \quad (5)$$

129 where  $D_{MAX}$  is the volume mean diameter after the maximum thermal treatment here  
 130 considered.. In the case of a modified waxy starch suspension in water, the variation in the

131 swelling degree over time can be described using a second-order kinetics equation  
132 (Lagarrigue et al., 2008):

$$133 \quad \frac{dS}{dt} = V\{T\}(I-S)^2 \quad (6)$$

134 where  $V$  is the temperature-dependent swelling kinetics rate ( $s^{-1}$ ). Equations (4, 5, 6)  
135 summarize the product swelling for a travelling droplet in the starch suspension. The  
136 following conservation equation puts in evidence the inherent coupling which occurs between  
137 starch swelling, fluid flow and heat transfer at every position of the processing unit:

$$138 \quad \vec{u} \cdot \vec{\nabla} S = V\{T\}(I-S)^2 + \vec{\nabla} \cdot (d_S \vec{\nabla} S) \quad (7)$$

139 where  $d_S$  is a diffusion coefficient ( $m^2.s^{-1}$ ). Equation (7) states that the swelling state results  
140 from a) convective transport, b) temperature-dependent swelling of starch granules, and c)  
141 diffusion around the position under consideration. The coefficient  $d_S$  considered in this  
142 equation expresses the Brownian (random) diffusion affecting the starch granules in the  
143 suspension as well as some diffusion between parallel pathways along the exchanger.

144 Governing equations (1, 2, 3, 7) are solved in this study with the help of numerical  
145 modeling tools throughout a sequence of computational domains, as summarized in Section 4.  
146 The first step of our modeling approach is the description of the transformation kinetics and  
147 the rheological behavior of the liquid product as functions of the state variables.

148

### 149 **3. Estimation of model parameters**

#### 150 *3.1. Experimental work*

151 The liquid product is a 3.42 % w/w starch suspension in water. Chemically-stabilized  
152 and cross-linked waxy maize starch (acetylated adipate distarch, C\*Tex 06205) was kindly  
153 supplied by Cargill (Baupte, France). It consisted of at least 99 % of amylopectin and a  
154 maximum of 0.4 % w/w of endogenous proteins. The product was prepared by adding the



155 starch powder to a 0.1 M NaCl aqueous solution. Before any thermal treatment, the resulting  
156 liquid remained at 50 °C for 30 minutes under continuous gentle stirring. Test tubes of 50 mL  
157 were used to ensure an equivalent thermal history for all the fluid parcels contained in the  
158 sample, while being large enough to enable further analyses. Stirring was performed with a  
159 magnetized bar under permanent rotation (500 rpm) inside the test tube.

160 Thermal treatments consisted of a heating and a cooling step, both under continuous  
161 stirring. The role of the cooling step was to stop product transformation as rapidly as possible  
162 in order to associate different "heating times" with their corresponding "swelling states". The  
163 heating step started when the product sample was removed from the 50 °C water bath and  
164 immersed immediately in the second water bath, which was maintained at 90 °C. The cooling  
165 step started when the sample was removed from the hot water bath and immersed  
166 immediately in the third water bath, maintained at about 4 °C. The product's temperature was  
167 initially between 62 and 92 °C (depending on heating duration) and then rapidly fell. The  
168 cooling step was halted when the temperature reached 10 °C.

169 The product's temperature was recorded every 15 seconds during the whole heating  
170 and cooling steps. The choices regarding the temperature in the hot water bath (90 °C) and the  
171 heating duration (up to 64 minutes) enabled significant changes in the particle size  
172 distribution and apparent viscosity. Temperatures close to 90 °C are involved in industrial  
173 starch processing (see for instance Thomas and Atwell, 1999).

174 Figure 1A summarizes the thermal history obtained after each test: starting at 50 +/-  
175 0.2 °C, the product's temperature firstly increased during the prescribed heating time and then  
176 decreased during the cooling step. Tests are labeled in terms of heating time duration.

177 The particle size distribution associated with each sample was measured using a  
178 Malvern Mastersizer 2000 laser granulometer (Malvern Instruments Ltd, United Kingdom)  
179 equipped with a 300 mm Fourier cell (range: 0.05–879 µm). Drops of product were placed in

180 the cell (100 mL) until obscuration reached about 20 %, corresponding to a dilution of 1/100.  
181 Typical values were assumed for the real and imaginary parts of the refractive index of starch  
182 granules (1.53 and 0.01; Nayouf et al., 2003). Samples were analyzed twice within less than  
183 15 minutes at room temperature, and the volume mean diameter was retained for further  
184 application. For a given sample, the two estimates of the volume mean diameter were always  
185 closer than 0.5  $\mu\text{m}$ .

186 Rheological measurements were performed using a MCR 301 rheometer (Anton Paar,  
187 Austria) equipped with coaxial cylinders. The temperature was controlled with a Peltier  
188 system. The flow behavior of the suspension was measured at 20 °C. Increasing and  
189 decreasing shear scans were performed stepwise following a logarithmic scale, firstly from 1  
190 to 500  $\text{s}^{-1}$  and then from 500 to 1  $\text{s}^{-1}$ . Each shear scan was performed twice, and the whole  
191 sequence took less than 15 minutes. The range 1 to 500  $\text{s}^{-1}$  includes the maximum shear rate  
192 expected at the heat exchanger.

193

### 194 3.2. Kinetics parameters

195 Figure 1B presents the volume mean diameter as a function of the heating time. This  
196 display offers evidence that the first four minutes of heating play a major role in driving the  
197 swelling state of the starch suspension. Between 1 and 8 min of heating, replicating a test  
198 associated with a given heating time can allow volume mean diameters differing by some  
199 micrometers. After 64 minutes of heating, the replicates agreed by better than 1 °C in  
200 temperature and better than 0.5  $\mu\text{m}$  in volume mean diameter.

201 The influence of temperature on the swelling kinetics rate is a challenging issue when  
202 it comes to representing the transformation of starch suspensions under heating. Indeed, as  
203 revealed by studies performed since the 1940s, starch granules can suddenly increase many  
204 times in size within a small range of temperature at about 65 °C while the viscosity of the

205 suspension also increases rapidly (Freund, 1944). Such a sudden influence of temperature on  
 206 the swelling kinetics rate can be approximated by:

$$207 \quad V\{T\} = 0 \quad \text{if } T < Ta, \quad (8a)$$

$$208 \quad V\{T\} = Va (T - Ta) \quad \text{if } T > Ta, \quad (8b)$$

209 where  $Ta$  is a threshold temperature and  $Va$  is a constant value ( $s^{-1} \cdot ^\circ C^{-1}$ ). Recent observations  
 210 have shown that the changes in starch granule size become significant above a certain  
 211 temperature (e.g. Malumba et al., 2013); from this perspective, equations (8) constitute a first-  
 212 order approximation for such a change in the swelling kinetics rate.

213 The volume mean diameter  $D$  after a given time  $t$  depends on the whole thermal  
 214 history of the sample, since the beginning of the thermal treatment (integration of equation 6):

$$215 \quad D\{t\} = D_0 + (D_{MAX} - D_0) \left( 1 - 1 / (1 + X\{t\}) \right) \quad (9a)$$

$$216 \quad X\{t\} = \int_0^t V\{T\{t'\}\} dt' \quad (9b)$$

217 where  $V\{T\{t'\}\}$  is estimated through equation (8) for temperature  $T$  at time  $t'$ . Equations (9)  
 218 can be used to estimate the parameters  $Va$  and  $Ta$ , after exposing the starch suspension to a  
 219 number of thermal treatments.

220 All the available tests are shown in Figure 1. Parameters  $Va$  and  $Ta$  were estimated  
 221 from tests corresponding to the heating times of 1 min (three replicates), 1.5 min, 2 min (three  
 222 replicates), 4 min (two replicates) and 8 min. Sample temperatures corresponding to these five  
 223 heating times were about 62, 70, 72, 83, and 89 °C respectively. Tests corresponding to the  
 224 heating times of zero (no hot bath) and 64 minutes allowed assessing the extreme values  $D_0$   
 225 = 16.3  $\mu m$  and  $D_{MAX} = 43.3 \mu m$ . The values for parameters  $Va$  and  $Ta$  were estimated by  
 226 minimizing the sum of squared deviations between the experimental values of  $D$  and those  
 227 calculated from equation (9), considering their respective thermal histories; we obtained  $Ta =$   
 228 61.1 °C and  $Va = 3.79 \cdot 10^{-3} s^{-1} \cdot ^\circ C^{-1}$ .

229            Figures 1B and 1C compare volume mean diameter values obtained from experiment  
230 and predicted through equation (9), assuming the longest thermal history (heating time 64  
231 min). Predictions agreed with experiment by better than 3  $\mu\text{m}$ , and often better than 1  $\mu\text{m}$ .  
232 The averaged relative error between predictions and experimental values is about 6 % for  
233 heating times from 0 to 64 minutes.

234            It must be noted that the swelling kinetics described by equations (6) and (8) is  
235 empirical; these equations describe how the thermal history of suspension droplets can be  
236 translated into changes affecting the volume mean diameter of the starch suspension as seen  
237 through laser granulometry. No attempt was made to study the heat and mass transfers  
238 occurring between starch granules and the surrounding water. We can anyway estimate the  
239 heat penetration time from granule surface down to the center, by considering typical values  
240 for the thermal diffusivity of starch (about  $2 \cdot 10^{-7} \text{ m}^2/\text{s}$ ; see for instance Rodriguez and de la  
241 Cruz, 2003). The result obtained (a few milliseconds) is four orders of magnitude smaller than  
242 the mean residence time of the starch suspension in the heat exchanger considered (about 50  
243 seconds). Therefore, we can assume that starch granules exhibit a uniform temperature, and  
244 that it varies during their pathway throughout the heat exchanger in the same manner as the  
245 surrounding water temperature.

246

### 247 *3.3. Rheological parameters*

248            Figure 2 shows the apparent viscosities of pasted starch suspensions gathered through  
249 Couette rheometry at 20 °C. These measurements were obtained after the tests corresponding  
250 to heating times of zero (no hot bath), 1, 2, 4 and 8 minutes. The starch suspension became  
251 shear-thinning as granule swelling progressed, suggesting describing the apparent viscosity  
252 through an Ostwald power law with a flow behavior index depending on the swelling state. In  
253 addition, the apparent viscosity increased with the heating time, suggesting that the

254 consistency coefficient can be expressed as a function of the swelling state too. Finally, as a  
 255 consequence of the relatively low mass concentration of starch in the suspension (3.42 %), the  
 256 apparent viscosity at a given swelling state is expected to decrease with the local temperature,  
 257 following a behavior similar to that exhibited by pure water.

258 The apparent viscosity associated with the starch suspension is hereinafter described  
 259 using the following approximations:

$$260 \quad \eta\{\dot{\gamma}, \Phi, T\} = K\{\Phi, T\} \dot{\gamma}^{n\{\Phi\}-1} \quad (10a)$$

$$261 \quad K\{\Phi, T\} = k_1 \exp(k_2 \Phi) \eta_{water}\{T\} \quad (10b)$$

$$262 \quad n\{\Phi\} = n^* + (1 - n^*) \exp(-k_3 (\Phi - \Phi_0)) \quad (10c)$$

263 where  $K$  is the consistency coefficient (Pa.s<sup>n</sup>),  $n$  is the flow behavior index,  $\dot{\gamma}$  is the shear rate  
 264 (s<sup>-1</sup>) and  $\eta_{water}$  is the dynamic viscosity associated with pure water (Pa.s). Equation (10c)  
 265 expresses that the flow behavior index exhibits an asymptotic pattern towards the value  $n^*$  as  
 266 the solid volume fraction increases. Equations (10) express the mean dependence of the  
 267 apparent viscosity with the shear rate, the solid volume fraction, and the temperature, while  
 268 requiring only few parameters. More-detailed descriptions could reconcile the Newtonian  
 269 plateau and the shear-thinning behavior (see for instance the grey squares in Figure 2);  
 270 however, such an improvement would require a higher number of parameters.

271 The parameters required by equation (10) were estimated from the experimental  
 272 values shown in Figure 2. Best fit values were estimated by minimizing the sum of squared  
 273 deviations between the predictions and experimental values of apparent viscosity; we  
 274 obtained  $k_1 = 0.662 \text{ s}^{n-1}$ ,  $k_2 = 13.6$ ,  $k_3 = 4.91$  and  $n^* = 0.55$ . Lines in Figure 2 indicate the  
 275 predictions of the apparent viscosities corresponding to the five thermal treatments; the  
 276 averaged relative error between predictions and experimental values is about 17 %.

277 It must be noted that the rheological behavior described by equations (10a) to (10c) is

278 empirical, with no considerations on the changes affecting starch granules under thermal  
279 treatment. Equations (10a) to (10c) associate the apparent viscosity of the starch suspension  
280 and the volume mean diameter of starch granules provided by Couette rheometry and laser  
281 granulometry respectively; changes in size are explicitly described via the volume mean  
282 diameter, while changes in other starch granules' characteristics (like shape and deformability)  
283 are implicitly accounted for in the mean general behavior described through equations (10) by  
284 modifying the values of constants  $k_1$ ,  $k_2$  and  $k_3$ .

285

#### 286 **4. Numerical model**

287 The heat exchanger under consideration is represented in Figure 3A. It is constituted  
288 by four tubular heating sections, one holding section, and a number of short curved tubes  
289 which are here grouped into three "bends". The liquid product executes counter-flow with  
290 respect to super-heated water along the heating sections; the product travels through the  
291 holding section and also the bends under thermally-insulated conditions. Figure 3B  
292 summarizes how the heat exchanger is represented in the numerical model: a sequence of  
293 two-dimensional, axi-symmetrical computational domains. Considering a volume flow rate of  
294 15 L/h, the mean residence time is about 50 seconds.

295 Table 1 summarizes the boundary conditions assumed for the radial  $u_r$  and axial  $u_z$   
296 components of the velocity, for the temperature  $T$ , and for the swelling degree  $S$ . Some of  
297 these boundary conditions are closely related to the operating conditions which were specified  
298 in running the heat exchanger, namely the volume flow rate ( $\dot{V}$ ), and the temperature at key  
299 positions: at the exchanger's inlet ( $T_1$ ), at the second heating section's outlet ( $T_2$ ), and at the  
300 fourth heating section's outlet ( $T_3$ ). These three temperatures allowed us to estimate, through  
301 global energy budget considerations, the averaged inward heat flux to be applied as boundary  
302 condition at the walls of the first and the second heating sections (about  $6930 \text{ W/m}^2$ ) and at

303 the walls of the third and the fourth heating sections (about 4210 W/m<sup>2</sup>).

304 Looking for the resolution of equations which govern the coupled phenomena, every  
305 computational domain has to be subdivided into a number of small, non-overlapping cells.  
306 The rectangular geometry of these domains suggests the adoption of rectangular cells  
307 characterized by dimensions  $dr$  and  $dz$  along the domain's radius and axis respectively. We  
308 chose cells whose length is four times their width ( $dz / dr = 4$ ), as a compromise for  
309 representing gradients along both directions. Three grids were successively assumed, each of  
310 them associated with an increasing number of identical cells in the radial direction:  $nr = 2^5 =$   
311  $32$ ,  $2^6 = 64$ , and  $2^7 = 128$ . For the last grid, we obtain  $nz = 6400$  in the case of our heating  
312 sections (length 800 mm); considering all the domains required to represent the heat  
313 exchanger, the final number is over 4.3 million cells. The mesh resolution corresponding to  $nr$   
314  $= 2^7$  is the highest allowed by computer memory capabilities (Intel Xeon CPU X5650 @ 2.67  
315 GHz, 64-bits, 32-Gb RAM computer).

316 Governing equations (1, 2, 3, and 7) are solved under steady-state conditions by  
317 applying the finite-element method as implemented in the simulation package COMSOL  
318 Multiphysics, version 4.4 (Zimmermann, 2006). The solution of the large linear system  
319 resulting from the linearization of the coupled equations is reached with the help of the  
320 Multifrontal Massively Parallel Sparse Direct Solver (MUMPS; Amestoy et al., 2001, 2006).  
321 Satisfactory model convergence was reached after assuming the diffusion coefficient  $d_S =$   
322  $10^{-8} \text{ m}^2 \cdot \text{s}^{-1}$  in equation (7).

323 Lastly, we are aware that curved tubes situated between two successive heating or  
324 holding sections can induce some mixing which tends to homogenize temperature and liquid  
325 product properties. Such an influence can be significant, as put in evidence through numerical  
326 modeling (Hajmohammadi et al., 2013) as well as analysis of experimental residence time  
327 distributions (Ndoye et al., 2012). In developing our numerical model, the curved tubes

328 ("bends") situated between successive heating or holding sections are assumed to play the role  
329 of thermally-insulated perfectly-mixed reactors. On the one hand, the mass-weighted value of  
330 the product temperature remains unchanged while the fluid parcels travel from the bend's inlet  
331 to its outlet. On the other hand, additional transformation is allowed should the product  
332 temperature exceed the threshold value  $Ta = 61.1$  °C (see Section 3).

333

## 334 **5. Results**

335 Figure 4 presents the main results obtained with the numerical model throughout the  
336 heating and the holding sections of the heat exchanger. Grey rectangles indicate the curved  
337 tubes (bends) situated between two successive sections. The results were obtained using  $nr =$   
338  $2^7 = 128$  rectangular cells in the radial direction when building the mesh in all the  
339 computational domains.

340 The temperature field is exhibited in display A. In the heating sections (domains He1,  
341 He2, He3, and He4), the starch suspension is warmed from the wall: its temperature increases  
342 more rapidly in the vicinity of the wall than at the axis of symmetry.

343 The starch suspension undergoes progressive transformation should its temperature  
344 exceed the threshold value  $Ta = 61.1$  °C. The volume mean diameter and the solid volume  
345 fraction are exhibited in displays B and C respectively. In the heating sections, their patterns  
346 match the temperature field. In the bends, the product is mixed and subject to additional  
347 transformation. Indeed, the fluid leaving the bends is characterized by mass-weighted values  
348 of solid volume fraction of 3.58, 5.36, and 8.21 %, which are gradually higher than the values  
349 at the outlets of the preceding heating sections: 3.57, 4.82, and 7.63 %, respectively. No  
350 transformation occurs in the holding section because it succeeds a bend whose outlet  
351 temperature (60.1 °C) is lower than the threshold value  $Ta$ .

352 The apparent viscosity of the starch suspension is shown in display F. At the heat



353 exchanger's inlet, the apparent viscosity is associated with low values (about 0.7 mPa.s) and  
354 Newtonian behavior due to the occurrence of untreated starch granules only ( $\Phi = \Phi_0$  in  
355 equation 10c). Moving forward in the heat exchanger, the apparent viscosity is slightly  
356 affected before the onset of starch swelling ( $T = Ta$ ). Beyond this point, the transformation  
357 state progresses; as a result, the liquid becomes shear-thinning (the flow behavior index  
358 decreases with  $\Phi$  in equation 10c) while the apparent viscosity values rise (the consistency  
359 coefficient increases with  $\Phi$  in equation 10b). The swelling state is the highest at the wall of  
360 the fourth heating section's outlet (domain He4).

361         The apparent viscosity of the starch suspension is here assumed to decrease with  
362 temperature following the behavior associated with pure water (equation 10a). The apparent  
363 viscosity decreases at the axis of symmetry throughout the two first heating sections (He1 and  
364 He2) because the temperature rises under low transformation. Inversely, the temperature's  
365 influence on the apparent viscosity can be dominated by that associated with the swelling  
366 state when the latter is significant, like in the vicinity of the wall near the outlet of the fourth  
367 heating section.

368         The progressive transformation of the starch suspension affects its rheological  
369 behavior, which in turn drives the flow field in the heat exchanger. The velocity magnitude  
370 and the shear rate predicted by the numerical model are presented in displays D and E. At the  
371 heat exchanger's inlet, a fully-developed velocity profile is assumed. In the first heating  
372 section (domain He1), the velocity magnitude decreases at the axis from the inlet to the outlet,  
373 whereas it increases in the vicinity of the wall; such a result can be related to the apparent  
374 viscosity values at the wall, which decrease as a consequence of progressive warming under  
375 low product transformation. In the three other heating sections (He2, He3 and He4), on the  
376 contrary, the velocity magnitude increases at the axis and decreases near the wall as a  
377 consequence of product transformation and increase in apparent viscosity near the wall: fluid

378 parcels travelling in the vicinity of the wall are slowed down, while those running at the axis  
379 undergo acceleration. Such a feature is quite severe in the fourth heating section; relatively  
380 high shear rate values are obtained towards its outlet, along a tongue-like region which  
381 separates the vicinity of the heating wall (where velocity is vanishing) and the region near the  
382 axis (where velocity is the highest).

383 Figure 5 displays the radial distribution of the volume mean diameter at the outlet of  
384 the last heating section (domain He4) as predicted by numerical model simulation after  
385 assuming three mesh resolutions. Figure 5A shows results obtained with the highest mesh  
386 resolution here considered ( $nr = 128$ ), while Figure 5B presents the differences between  
387 results from a given mesh and those corresponding to  $nr = 128$ . Firstly, these differences are  
388 often negative (swelling is underestimated); secondly, they become stronger as the mesh  
389 becomes coarser; finally, their strongest values occur at about 2.5 mm from the axis, where  
390 the radial variation of the volume mean diameter is the highest. These findings can be  
391 explained by the progressively-finer representations of the gradients of state variables as the  
392 mesh resolution increases.

393 The influence of mesh resolution can also be assessed in terms of volume mean  
394 diameter which corresponds to the mass-weighted value of the solid volume fraction at the  
395 outlet of a given section of the heat exchanger. After assuming 32, then 64, and finally 128  
396 cells in the radial direction, the bulk values of the volume mean diameter predicted by the  
397 numerical model were respectively of 18.19, 18.26 and 18.30  $\mu\text{m}$  at the outlet of the second  
398 heating section, and 25.37, 25.42 and 25.44  $\mu\text{m}$  at the outlet of the fourth heating section.  
399 Differences between results corresponding to two successive meshes decrease as the latter  
400 become finer, suggesting a convergence-like behavior. We argue that a further leap in mesh  
401 resolution, from  $nr = 2^7 = 128$  to  $nr = 2^8 = 256$ , would allow closer results, with differences  
402 between respective predictions of the volume mean diameter smaller than 0.05  $\mu\text{m}$ .

403           Figures 4 and 5 put in evidence that we have satisfactorily solved the coupled  
404 problem: while Figure 5 shows that we reached some degree of convergence in solving the  
405 equations which govern the relevant phenomena, the two-dimensional fields displayed in  
406 Figure 4 are physically consistent. Assessing the performance of model predictions requires  
407 comparisons with experimental values. Such a comparison can be performed in terms of  
408 mass-weighted values of the swelling state resulting from numerical model simulations.

409           Experimental results were obtained after running the available heat exchanger under  
410 the operating conditions which were applied in numerical modeling simulations (see Table 1).  
411 The starch suspension was sampled three times at the outlet of both the second and the fourth  
412 heating sections, and immediately cooled down to about 4 °C to avoid additional product  
413 transformation. The size distribution analysis of the product samples provided volume mean  
414 diameter values of about 16.5 and 31.6  $\mu\text{m}$  respectively after the second and the fourth  
415 heating sections, with standard deviations of 0.4  $\mu\text{m}$ . Considering the numerical model  
416 simulations which were conducted with the highest mesh resolution allowed, the bulk values  
417 for the predicted volume mean diameter after those two heating sections were respectively  
418 18.30 and 25.44  $\mu\text{m}$  (+11 % and -19 % respectively compared to the experimental values).

419           The level of agreement of numerical coupled modeling with experiment can be  
420 compared with that obtained through a simpler modeling approach. The simplest way to  
421 characterize the path followed by fluid parcels along a heat exchanger consists in neglecting  
422 the radial dependence of both velocity and temperature. In such a one-dimensional model, it is  
423 assumed that fluid parcels move according to a plug-flow velocity field, and that they are  
424 exposed to a temperature which depends only on the distance from the exchanger inlet. The  
425 swelling state of the starch suspension at selected positions of the heat exchanger was  
426 estimated through equations (9). For this, the swelling kinetics rate was represented as in the  
427 numerical coupled model; in building the thermal history, the bulk temperature was assumed

428 to vary linearly along the heating sections, and to be constant along the bends and the holding  
429 section. At the outlet of the second heating section, the predicted volume mean diameter is  
430 identical to the value assumed as boundary condition at the exchanger's inlet; this result is  
431 easily understandable because the temperature value at the second heating section's outlet  
432 (60.1 °C) is lower than the threshold value  $T_a = 61.1$  °C. Starch swelling takes place along the  
433 following sections according to this one-dimensional model; at the outlet of the fourth heating  
434 section, the predicted volume mean diameter is 21.5  $\mu\text{m}$  (–32 % compared to the  
435 experimental value).

436 We can summarize that: a) when the suspension bulk temperature does not reach the  
437 threshold value  $T_a$ , the plug-flow and radially-independent temperature approach is unable to  
438 predict any starch swelling, contrarily to the numerical coupled model; b) when that threshold  
439 is overcome, the simpler approach underestimates the swelling state reached by the starch  
440 suspension. Starch swelling can be compared in terms of volume mean diameter value  
441 increase between the exchanger's inlet and outlet: we obtain  $(31.6-16.3) = 15.3$   $\mu\text{m}$  from  
442 experiment at pilot scale,  $(25.4-16.3) = 9.1$   $\mu\text{m}$  from the numerical coupled model, and  
443  $(21.5-16.3) = 5.2$   $\mu\text{m}$  from the simpler approach. In other words, the numerical model  
444 predicts the bulk swelling state of the starch suspension at a level of agreement (–41 % in  
445 volume mean diameter increase) which is better than that reached after assuming plug-flow  
446 and radially-independent temperature (–66 %).

447 Finally, a number of reasons can contribute to explain the differences between  
448 predictions from the numerical coupled model and respective experimental values. Firstly, the  
449 curved tubes (bends) were assumed, in numerical model simulations, to play the role of  
450 thermally-insulated perfectly-mixed reactors; actually, we expect the occurrence of slight  
451 cooling and incomplete mixing, whose consequences on product transformation are difficult  
452 to anticipate. Secondly, starch swelling under heat treatment was assumed to follow a single

453 temperature-dependent swelling kinetics (equation 6), independently on the granule size. The  
454 influence of starch granule size on the swelling onset has been scarcely studied (Myllarinen et  
455 al., 1998; see references in Sasaki and Matsuta, 1998); however, no generalization seems  
456 possible regarding the starch type. On the other hand, we cannot discard some experimental  
457 bias in sampling the starch suspension while it flows in the heat exchanger.

458

## 459 **6. Conclusions and Forthcoming Work**

460

### 461 Conclusions

462 The key parameters describing the swelling kinetics and the rheological behavior of a  
463 starch suspension were estimated from a limited number of experiments at laboratory scale,  
464 after studying small volumes of product under heat treatment and continuous agitation. The  
465 application of these parameters allowed model predictions of the final swelling state reached  
466 by the starch suspension which did agree, to some extent, with observations conducted while  
467 running the heat exchanger. We can therefore conclude that kinetics and rheological model  
468 parameters can be transferred from laboratory to pilot scale.

469 The plug-flow hypothesis implies no influence of product viscosity on the velocity  
470 field, and therefore no coupling between fluid flow, heat transfer and transformation. Such a  
471 simpler approach provides worse estimates of the final swelling condition reached by starch  
472 granules in suspension, relatively to those obtained with the numerical coupled model. We  
473 conclude that the consideration of two-way coupling between the relevant phenomena  
474 constitutes a positive improvement in looking for realistic predictions of the product's  
475 transformation level.

476

477 Forthcoming work

478           This study indicates three key issues that deserve attention in implementing numerical  
479 coupled models to represent the transformation of realistic food fluid parcels during their  
480 pathway in thermal processing units.

481           Firstly, after representing the heat exchanger under consideration by a sequence of axi-  
482 symmetrical two-dimensional (2D) domains, the most obvious following step should be to  
483 solve the coupled problem by considering the actual three-dimensional (3D) geometrical  
484 characteristics of the equipment. In fact, the thermal mixing due to the secondary flow in  
485 curved tubes can be more or less effective depending on the flow rate and the fluid  
486 rheological behavior (e.g. Cvetkovski et al., 2015). Inclusion of secondary flow is certainly a  
487 positive improvement, and its implementation requires higher computer memory and time  
488 resources. The challenging task to solve a similar coupled problem by considering the tubular  
489 heat exchanger schematized in Figure 3A is already under way.

490           Secondly, additional efforts are required in representing the product transformation, as  
491 the following step towards more realistic predictions from the numerical model, even in the  
492 case of a relatively simple liquid product like a starch suspension. Towards such a goal,  
493 experiments were conducted with an optical microscope coupled to a warming plate in order  
494 to continuously observe the starch granules' behavior during thermal treatments. Indeed,  
495 visual observation of individual granules suggested us that, in the case of modified waxy  
496 maize starch, the swelling mechanism presents some stochastic nature while being associated  
497 with diffusion of surrounding water into the granule.

498           Finally, the whole strategy could be adapted to product formulations associated with  
499 increasing complexity and industrial relevancy, for instance by studying starch swelling in  
500 skim milk and later by adding carrageenan. The notion of swelling degree (associated with  
501 starch granules in water) could be replaced by that of "filling degree" associated with all  
502 objects floating in the continuous phase. Further attention should be paid to estimating the

503 relevant solid volume fraction. The main structure of the numerical model could be  
504 maintained, as the changes would only affect the key parameters describing transformation  
505 kinetics and rheological behavior.

506

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Table 1: Boundary conditions taken into account for the radial  $u_r$  and axial  $u_z$  components of the velocity, for the temperature  $T$ , and for the swelling degree  $S$ .

	fluid flow	heat transfer	transformation
inlet (a)(b)(c)	velocity: $u_r = 0, u_z\{r\}$	temperature: $T\{r\}$	swelling degree: $S\{r\}$
outlet	null pressure, no viscous stress	convective flux: $\frac{\partial T}{\partial z} = 0$	convective flux: $\frac{\partial S}{\partial z} = 0$
axis	symmetry: $u_r = 0, \frac{\partial u_z}{\partial r} = 0$	symmetry: $\frac{\partial T}{\partial r} = 0$	symmetry: $\frac{\partial S}{\partial r} = 0$
wall	no slipping: $u_r = u_z = 0$	flux density (d)(e)	no diffusion: $\frac{\partial S}{\partial r} = 0$

(a) first heating section: the suspension is assumed to be at the temperature  $T_1 = 43.9$  °C, containing untreated starch granules only ( $S = 0$ ); the axial velocity follows the fully-developed parabolic profile associated with the volume flow rate  $\dot{V} = 15$  L/h (mean velocity  $\bar{u}_z = 0.083$  m.s<sup>-1</sup>, Reynolds number of about 1040).

(b) other sections, when the preceding one is not a bend: the temperature, the swelling degree and the axial velocity follow the distributions  $T\{r\}$ ,  $S\{r\}$  and  $u_z\{r\}$ , which were predicted at the outlet of the preceding section.

(c) other sections, when the preceding one is a bend: the temperature assumes its mass-weighted (bulk) value  $\tilde{T}$  at the bend's outlet; the swelling degree assumes the value  $\tilde{S}$  which corresponds to the mass-weighted value  $\tilde{\Phi}$  of the solid volume fraction at the bend's outlet;

the axial velocity follows  $u_z\{r\} = \bar{u}_z \left( \frac{3\tilde{n} + 1}{\tilde{n} + 1} \right) \left( 1 - \frac{r}{R} \right)^{\frac{(\tilde{n} + 1)}{\tilde{n}}}$  where  $\tilde{n}$  is the flow behavior

index given through equation (10c) by replacing  $\Phi = \tilde{\Phi}$ .

(d) heating and holding sections: inward heat flux estimated from the section's global energy budget, by taking into account the temperatures  $T_1 = 43.9$  °C at the exchanger's inlet,  $T_2 = 60.1$  °C at the second heating section's outlet, and  $T_3 = 70.0$  °C at the fourth heating section's

| inlet.

(e) bends: thermal insulation.

## Figure Captions

Figure 1: Key results regarding the evolution of the starch suspension under thermal treatments: A) the sample thermal history; B) the mean volume diameter as function of the heating time, obtained from experiment and estimated through the second-order kinetics; C) predicted versus experimental values for the mean volume diameter.

Figure 2: Apparent viscosity values at 20 °C of the starch suspension, after selected thermal treatments. Lines indicate the corresponding predictions of apparent viscosity as a function of shear rate and solid volume fraction.

Figure 3: Three-dimensional (A) and two-dimensional axi-symmetrical (B) schematic representations of the heat exchanger under consideration. The heating sections are indicated in red, the curved tubes (bends) in blue, and the holding section in green. In scheme B, the dashed line and the red continuous lines indicate the axis of symmetry and the heating walls, respectively.

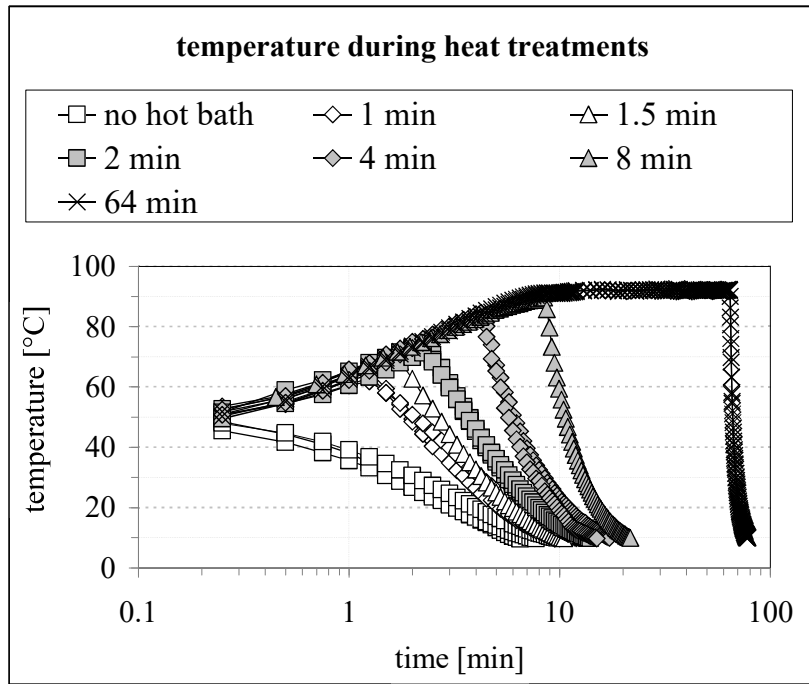
Figure 4: Numerical model predictions for the temperature, the mean volume diameter and the volume fraction associated with starch granules, the velocity magnitude, the shear rate, and the apparent viscosity throughout the heating and holding sections of the heat exchanger.

Figure 5: Mean volume diameter as predicted by the numerical model at the outlet of the fourth heating section: A) results obtained by assuming a mesh consisting of 128 rectangles in the radial direction; B) difference between results obtained by assuming successively 64, and 32 rectangles in the radial direction, and results shown in the display A.

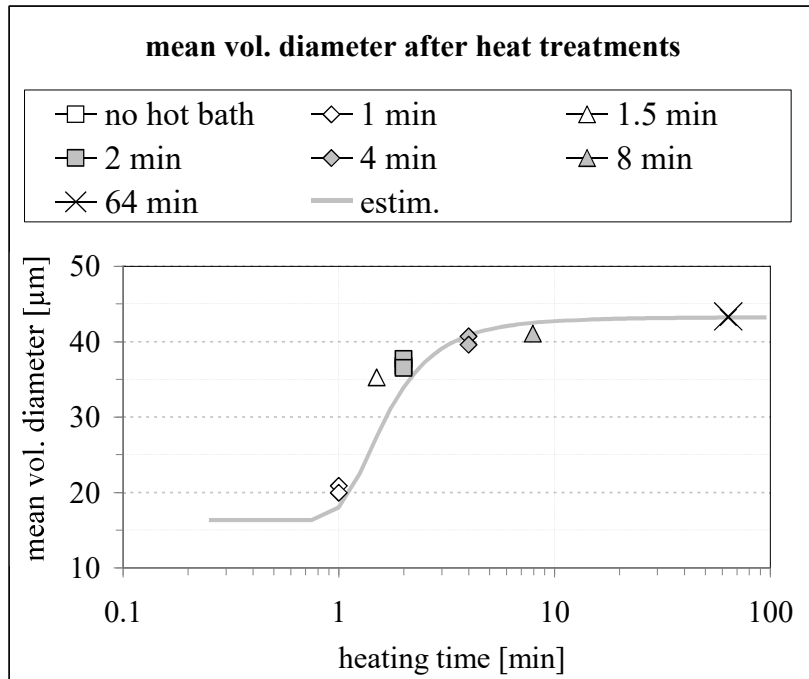


Figure

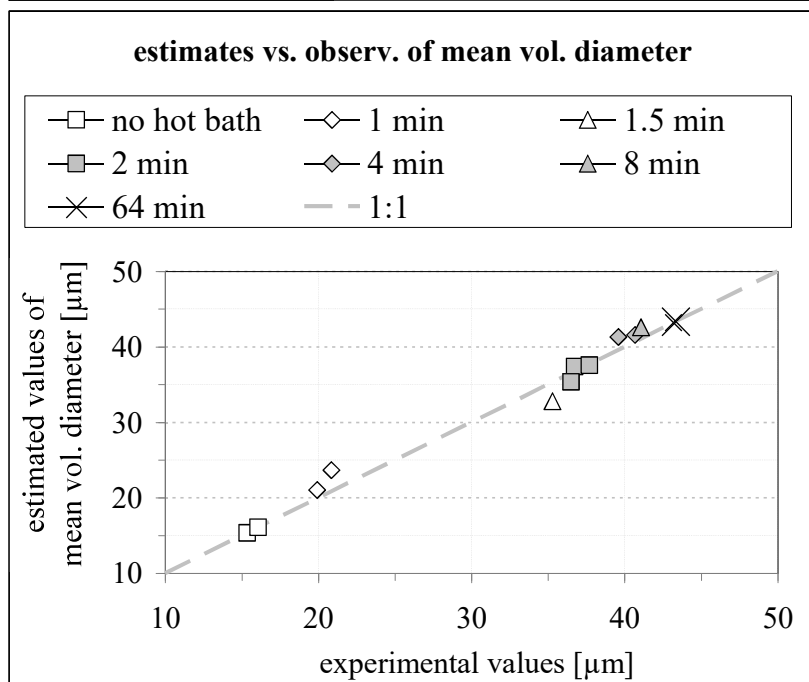
**A**



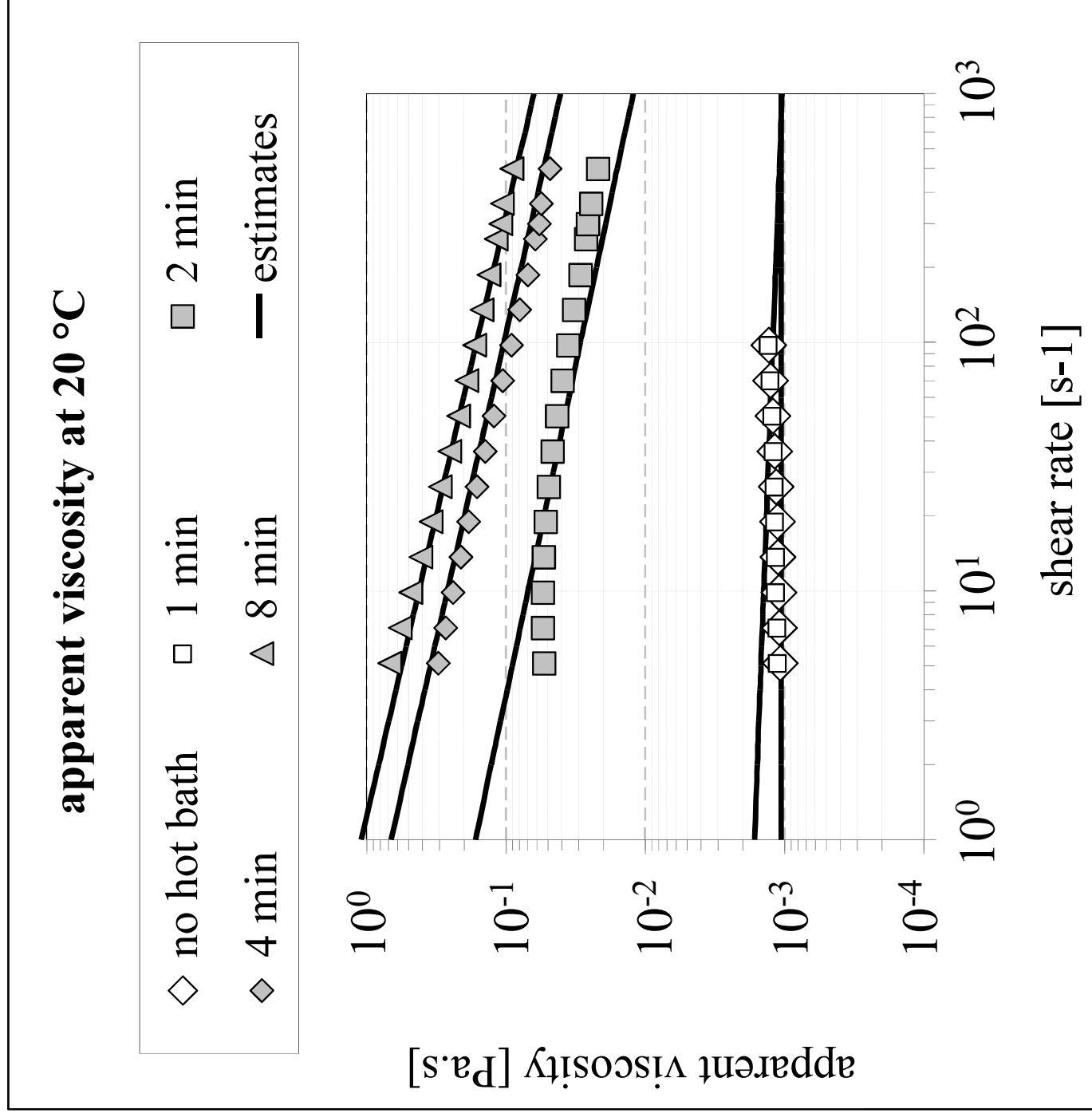
**B**



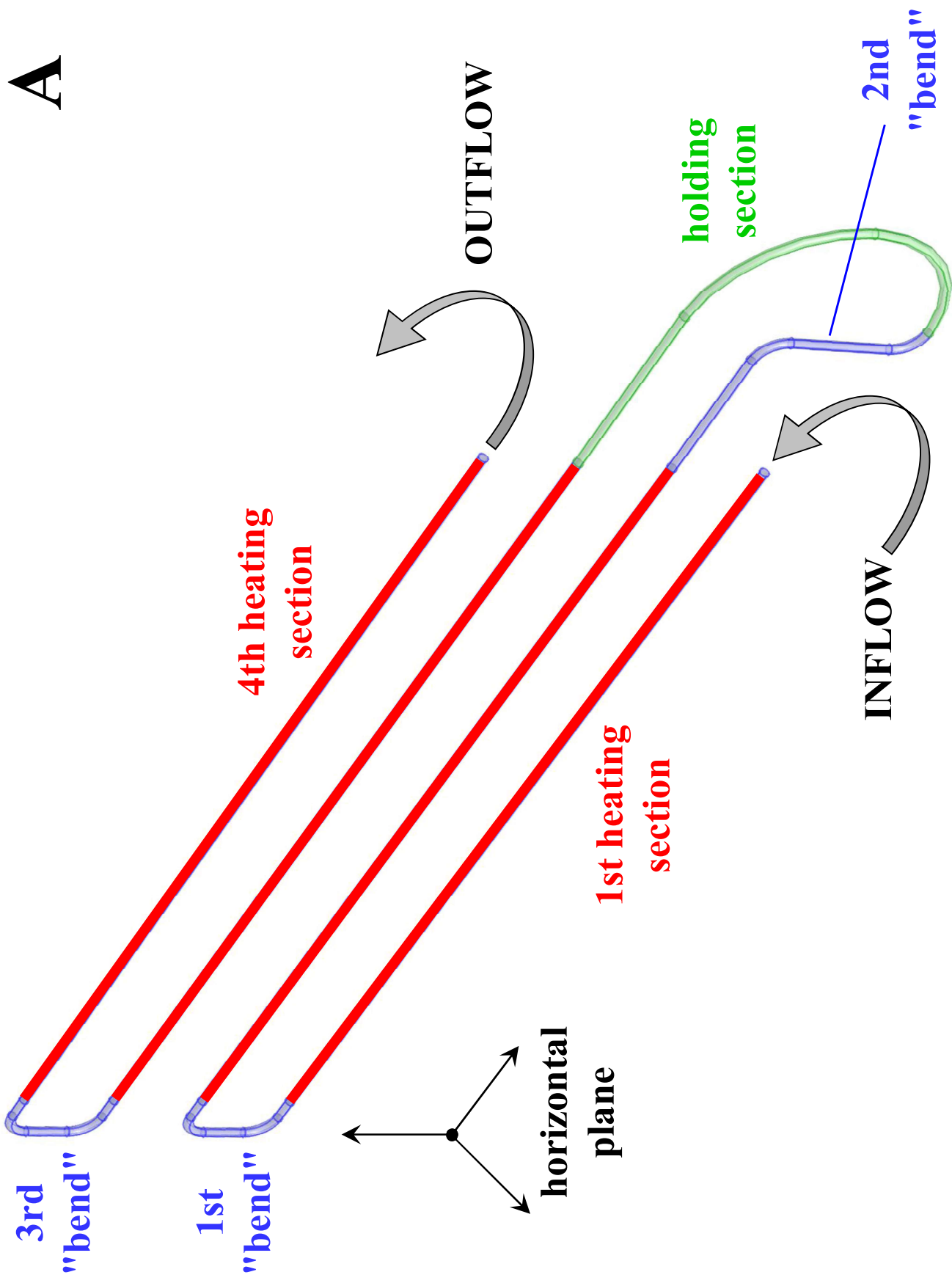
**C**



Figure

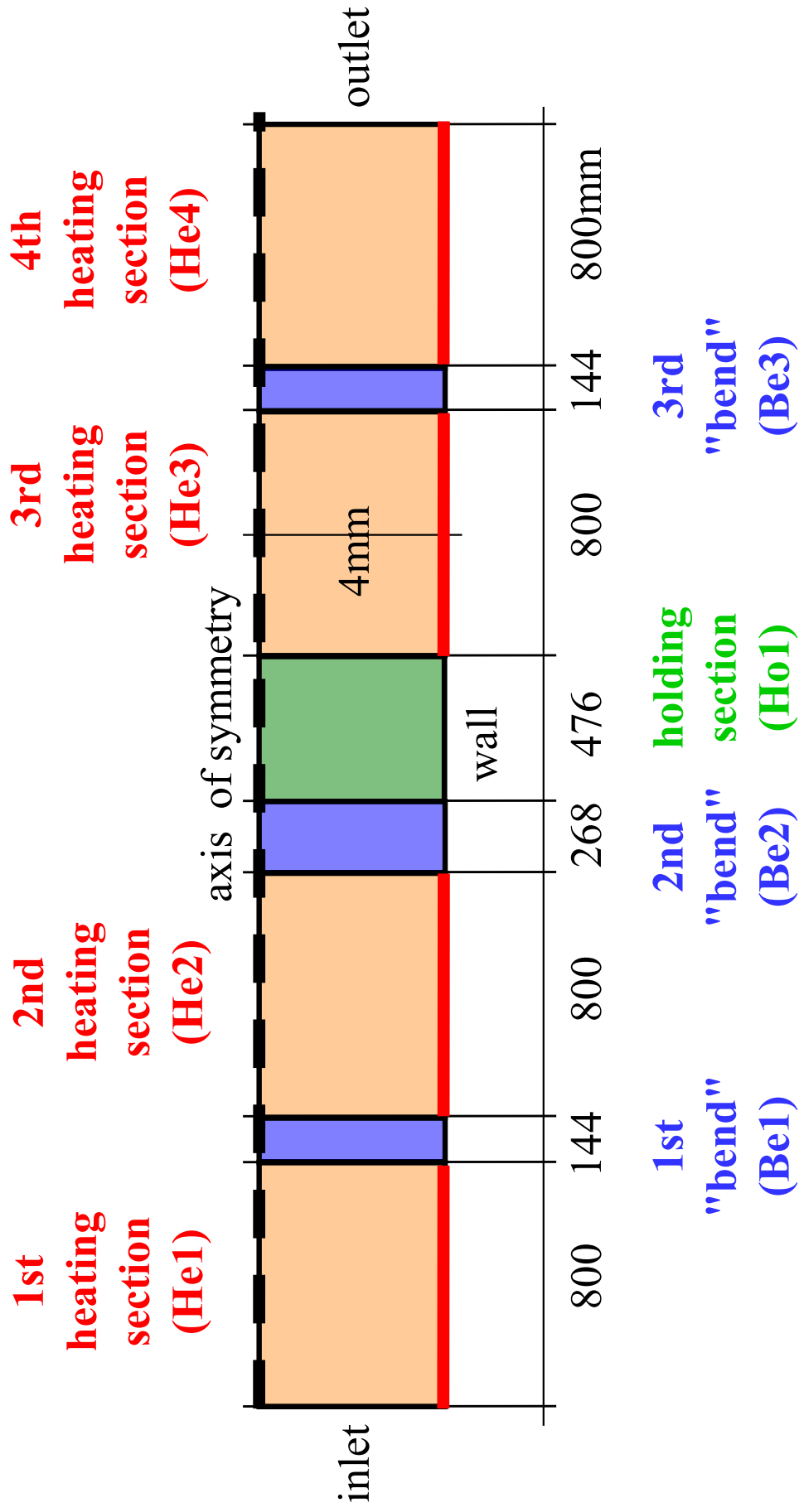


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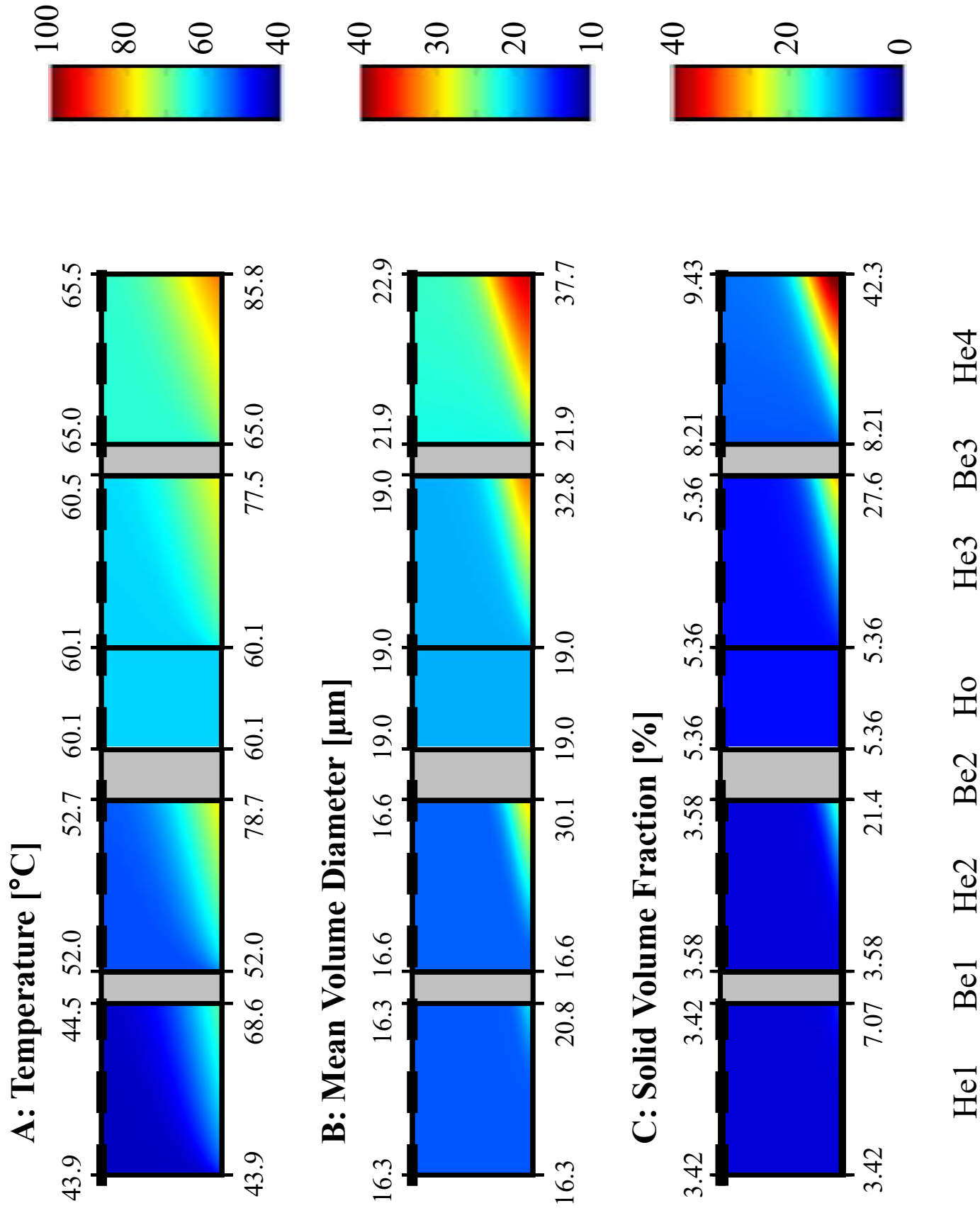


Figure

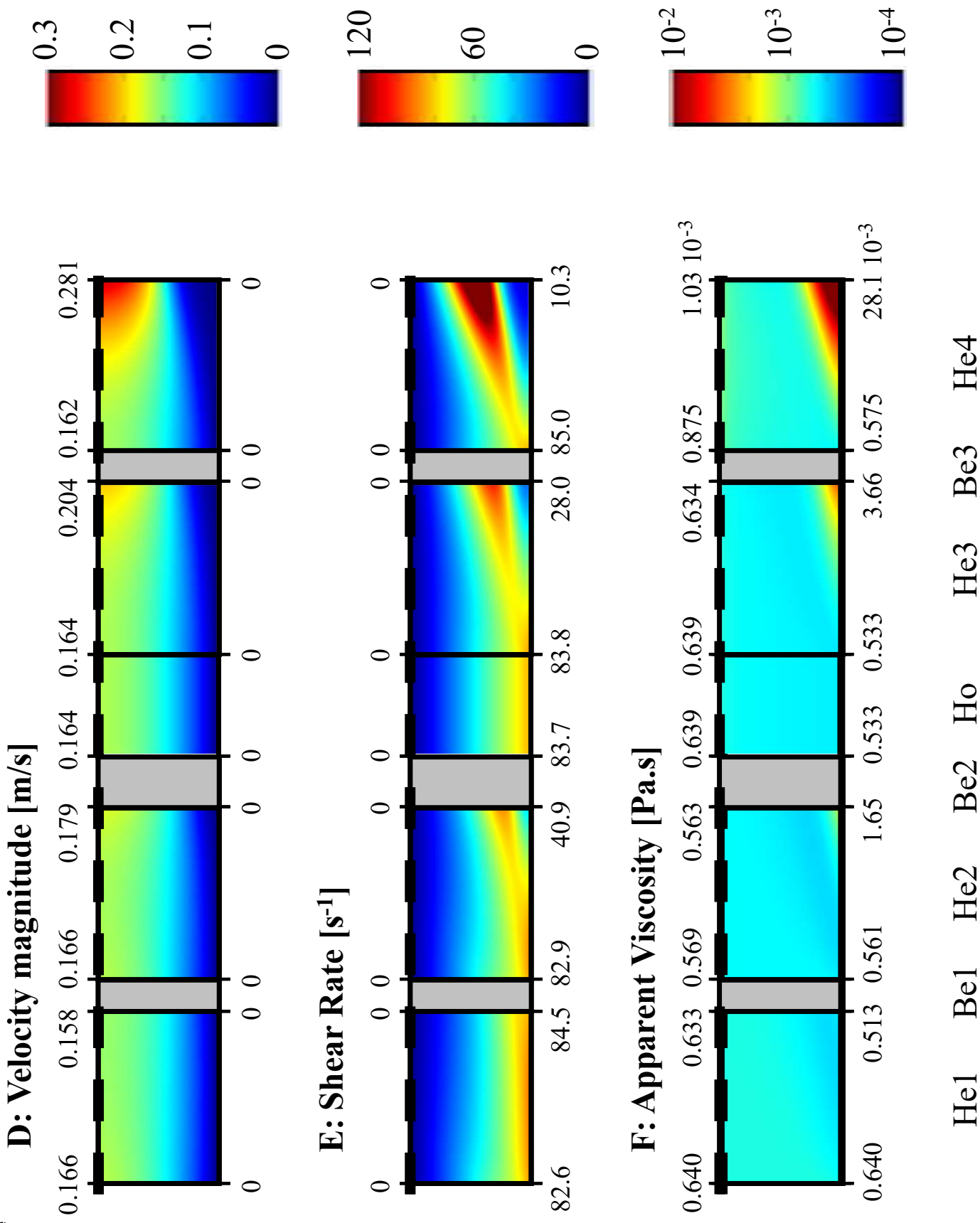
# B



Figure

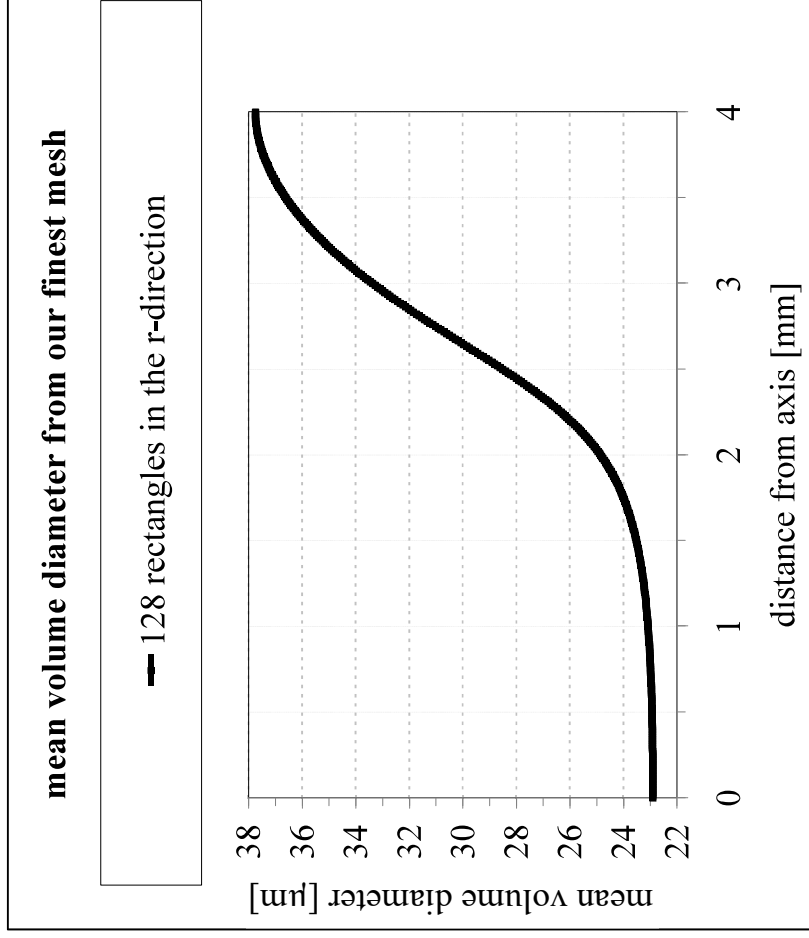


Figure



Figure

**A**



**B**

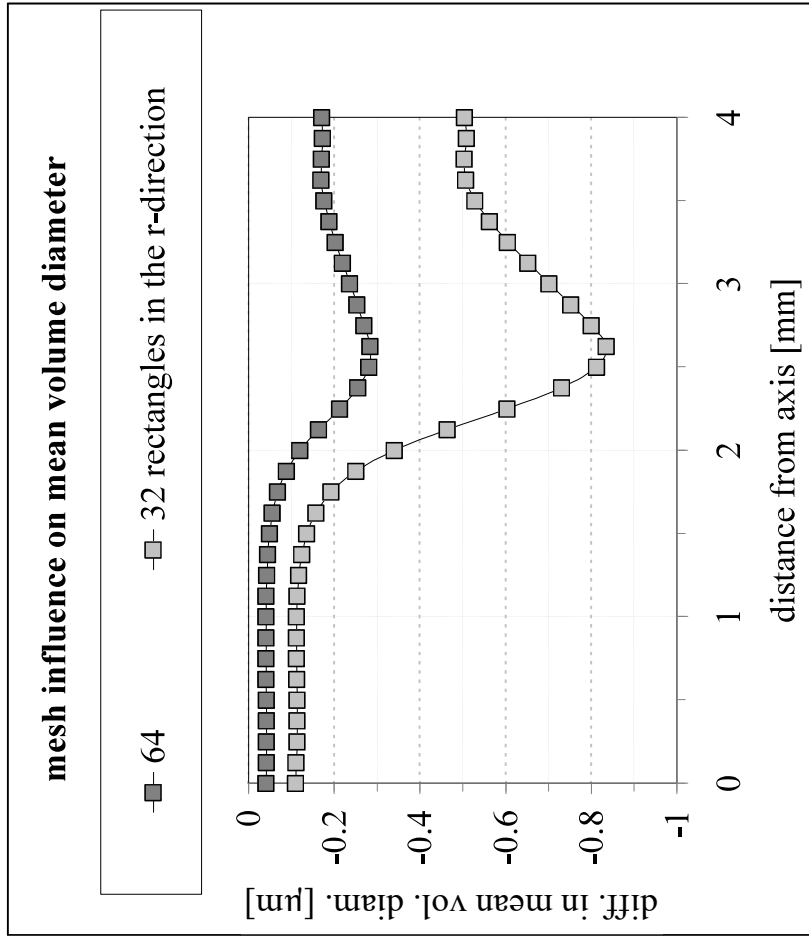


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axis	symmetry: $u_r = 0, \frac{\partial u_z}{\partial r} = 0$	symmetry: $\frac{\partial T}{\partial r} = 0$	symmetry: $\frac{\partial S}{\partial r} = 0$
wall	no slipping: $u_r = u_z = 0$	flux density (d)(e)	no diffusion: $\frac{\partial S}{\partial r} = 0$

(a) first heating section: the suspension is assumed to be at the temperature  $T_I = 43.9$  °C, containing untreated starch granules only ( $S = 0$ ); the axial velocity follows the fully-developed parabolic profile associated with the volume flow rate  $\dot{V} = 15$  L/h (mean velocity  $\bar{u}_z = 0.083$  m.s<sup>-1</sup>, Reynolds number of about 1040).

(b) other sections, when the preceding one is not a bend: the temperature, the swelling degree and the axial velocity follow the distributions  $T\{r\}$ ,  $S\{r\}$  and  $u_z\{r\}$ , which were predicted at the outlet of the preceding section.

(c) other sections, when the preceding one is a bend: the temperature assumes its mass-weighted (bulk) value  $\tilde{T}$  at the bend's outlet; the swelling degree assumes the value  $\tilde{S}$  which corresponds to the mass-weighted value  $\tilde{\Phi}$  of the solid volume fraction at the bend's outlet;

the axial velocity follows  $u_z\{r\} = \bar{u}_z \left( \frac{3\tilde{n} + 1}{\tilde{n} + 1} \right) \left( 1 - \frac{r}{R} \right)^{\frac{(\tilde{n} + 1)}{\tilde{n}}}$  where  $\tilde{n}$  is the flow behavior

index given through equation (10c) by replacing  $\Phi = \tilde{\Phi}$ .



(d) heating and holding sections: inward heat flux estimated from the section's global energy budget, by taking into account the temperatures  $T_1 = 43.9$  °C at the exchanger's inlet,  $T_2 = 60.1$  °C at the second heating section's outlet, and  $T_3 = 70.0$  °C at the fourth heating section's

| inlet.

(e) bends: thermal insulation.