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# Multiscale modelling of flow, heat transfer and swelling during thermo-mechanical treatment of starch suspensions

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## Abstract

During thermal treatment of starch-based liquid food products, many coupled phenomena occur at different scales. Heat transfer affects local temperature evolution, which determines starch transformation (swelling and eventually amylose release and/or granules breakage). This transformation modifies the rheology, which in turn affects the fluid flow and heat transfer. This two-way coupling of flow, transfer and transformation was previously simulated in heat exchangers by Computational Fluid Dynamics (CFD), through purely Eulerian or Eulerian/Lagrangian approaches, assuming that the starch granules follow locally the fluid velocity. Moreover, the transformation was only characterized by a deterministic model for the mean particle size evolution (swelling). The present work takes into account directly the trajectory and the swelling of each starch granule in interaction with the surrounding fluid through an approach combining CFD with Discrete Element Method (DEM) simulations. Due to lubrication forces, the granules tend to avoid one another and when they come in contact, an elastic rebound is assumed. This approach is applied here to a waxy maize starch suspension during heating in a Couette rheometer. A shear-induced random walk of the particles is observed. Swelling first occurs near the heated wall, then swollen granules migrate toward zones of lower solid fraction. This multiscale model (rheometer scale: 1 mm, starch granule scale: 10  $\mu\text{m}$ ) can be used for the analysis of such rheometry tests in which the sample cannot be assumed as homogeneous.

*Keywords:* Starch Granule, Swelling kinetics, DEM, CFD, Modelling

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## 1. Introduction

Starch is one of the most abundantly available naturally occurring biopolymers on earth. It is predominantly found in grains and tubers and is the source of 70-80 % of the calories consumed by humans worldwide in various forms of food products (Bertolini, 2010). Starch also finds many applications apart from food in industries such as bio-medical, packaging, paper, textile and pharmaceutical industries. Starch in its native form occurs as granules which consists of two polysaccharides, amylose and amylopectin. Both amylose and amylopectin have  $\alpha$ -glucose as the monomer but differ in the way they are bonded. Amylose has  $\alpha(1-4)$  bonds and is linear whereas amylopectin has  $\alpha(1-6)$  bonds and is highly branched. The granule structure, molecular weight distribution, degree of amylopectin branching and the ratio of amylose to amylopectin varies according to the botanical origin of the starch granule. This, in turn, can have drastic consequences in the physical and chemical properties of the granules such as rheology and gelatinization behaviour (Banks et al., 1974; Hoover, 2001; Srichuwong et al., 2005; Visser et al., 1997). Some kinds of starch are chemically modified to have enhanced structural and functional properties of the starch granules (Debnath et al., 2013; Alcázar-Alay and Meireles, 2015). By 2025, the world starch production is projected to be around 156 million metric tons, including native and modified starches (Reportlinker.com, 2020). As a result starch has been in the past, a subject of perennial research interest for researchers from diverse backgrounds and will be in the near future as well.

Gelatinization is the process in which starch granules undergo an irreversible transformation when subjected to thermomechanical treatment in the presence of water. During this process, starch granules swell by imbibing water and the overall viscosity of the suspension increases. It is also sometimes referred to as starch pasting in literature. Starch granules are semi-crystalline with concentric rings of crystalline and amorphous phases (Ratnayake and Jackson, 2008). On the microscopic scale, the process corresponds to the melting of crystallites, loss of birefringence, leaching of amylose into the continuous phase and sometimes rupture of the granular structure (Atwell, 1988; Biliarderis, 1992). The evolution of viscos-

29 ity during gelatinization is also influenced by the process parameters such as concentration,  
30 shear rate, thermal history, pH and presence of other ingredients (Thomas and Atwell, 1999;  
31 Malumba et al., 2018). For high amylopectin cereal starches, such as waxy maize starch, the  
32 granules tend to hydrate with ease, swell rapidly and rupture to a great extent which results  
33 in loss of paste viscosity. As starch is used as a viscosity enhancer in many food applications  
34 such as in ketchup and also in non-food applications such as enhanced oil recovery in oil  
35 drilling, the rupture of the granules which leads to loss in suspension viscosity should be  
36 avoided, this can be avoided by chemically modifying the starch.

### 37 *1.1. Coupling starch swelling, flow and heat transfer*

38 It is of particular interest to understand the coupled evolution of temperature, starch  
39 gelatinization (notably swelling) and rheology in heat exchangers. Pioneer work was done  
40 in this field by Yang and Rao (1998) and more recently by Plana-Fattori et al. (2013) and  
41 Plana-Fattori et al. (2016). In these studies, the rheology of partially gelatinized suspen-  
42 sion is related to the temperature evolution directly (Yang and Rao, 1998) or via a swelling  
43 kinetic model where the parameters of this model are estimated empirically. Then fluid  
44 flow and heat transfer are studied after solving the governing equations in interaction with  
45 the rheology/swelling model. The simulations were in relatively good agreement with some  
46 experimental measurement. However, these studies had several limitations: the size distri-  
47 bution of the granules was ignored (only average diameter evolution was considered), the  
48 variability from a granule to another in terms of swelling was ignored, the granules followed  
49 exactly the fluid flow, inside a fluid parcel (on a given trajectory) all the granules were  
50 assumed to have the same behaviour. It is known that particles migrate towards a region of  
51 lower shear rate, lower volume fraction and lower viscosity (Shauly et al., 2000). It is also  
52 known, that even without a gradient of these variables, there is shear-induced self-diffusion  
53 of the particles due to elastic collisions and lubrication interaction (Foss and Brady, 1999).  
54 This is well documented in literature (Eckstein et al., 1977; Leighton and Acrivos, 1987;  
55 Breedveld et al., 1998; Phillips et al., 1992). Shear induced self diffusivity is proportional  
56 to the shear rate and to the squared particle radius. The proportionality parameter de-

57 pends on the particle volume fraction ( $\phi$ ) and different correlations were proposed (linear  
58 or quadratic). Eckstein et al. (1977) found that this parameters increases almost linearly  
59 from 0 to 0.02 when the volume fraction rises from 0 to 0.2. This general trend was con-  
60 firmed by Breedveld et al. (1998) who compared different studies. Inertial effect can also be  
61 involved depending on the particle Reynolds number. Also, particle migratory self-diffusion  
62 can induce an increase of momentum and heat transfer, in other words, this phenomenon  
63 can increase the apparent viscosity and conductivity.

64 The main question we would like to answer in this study is, are these various granular  
65 phenomena of appreciable importance in the case of starch suspension subjected to fluid flow  
66 and heat transfer? To answer this question, an approach combining Computational Fluid  
67 Dynamics (CFD) and the Discrete Element Method (DEM) is here proposed, similar to one  
68 implemented in Qian et al. (2014) for simulating granular filters. One specificity is that the  
69 suspension evolves because of the starch granule swelling. We apply this modelling approach  
70 to the case of a small gap Couette rheometer during fast heating of the external cylinder.  
71 In this case, we also have shear and heat transfer with momentum and energy exchanges  
72 between fluid and particles, possible particle migration and self-diffusion (random walk) but  
73 the fluid flow is simpler than in a heat exchanger.

#### 74 *1.2. Starch swelling modelling*

75 Many models exist in the literature to predict the average degree of swelling depending  
76 on the thermal history of a given starch suspension (Lagarrigue et al., 2008; Malumba et al.,  
77 2013; Plana-Fattori et al., 2016). More recently, Desam et al. (2018a,b, 2020) proposed a  
78 mechanistic model based on the Flory-Rehner theories, for the starch swelling of various  
79 grades. However, this deals at the population level assuming same model parameters for  
80 all granules and hence, does not take into account the inter-granule variability. The inter-  
81 granule variability has biological origins and the alternating crystalline and amorphous phase  
82 of the starch granule is bound to slightly vary even among starch granules from the same  
83 plant. Thus, modelling the average behavior may not be sufficient to understand some  
84 mechanisms such as granule migration during thermal processing in presence of high velocity

85 and temperature gradients. Therefore, in our previous work we have described the granules  
86 swelling kinetics at the individual granule scale using a semi-mechanistic model (Palanisamy  
87 et al., 2020) for modified waxy maize starch. According to the microscopic observation of  
88 starch granules during heating in the presence of water, we observed variability in swelling  
89 kinetics from granule to granule. Each granule has a specific delay time associated with the  
90 onset of swelling. Also, the granules which began to swell at a later instant tended to swell  
91 slower compared to the granules which began to swell earlier for a given time-temperature  
92 profile. Below a minimal temperature, no swelling was observed. Experiments showed that  
93 high temperatures resulted in lower delay time and faster increase in granule diameter. These  
94 phenomena were cast mathematically in a model (Palanisamy et al., 2020). Nayouf et al.  
95 (2003) indicated that the volume fraction of granule can be estimated if the mean granule  
96 size can be accurately determined. As discussed in the next section, volume fraction of  
97 starch is a critical parameter used in the closure relations for predicting viscosity in mixture  
98 models. This inter granular level kinetic model of starch granule swelling is coupled here,  
99 with a CFD-DEM model for prediction of fluid flow, heat transfer and granule trajectories.

### 100 *1.3. Starch suspension rheology*

101 As discussed in the previous sections, starch has many different varieties and is used  
102 as a viscosity enhancer in various fields. Rheology measurements are generally carried out  
103 at a constant temperature and after complete gelatinization. However, having the evolu-  
104 tion of rheology during gelatinization is quintessential for robust process design and can be  
105 of interest to design control strategies as well. This is done notably with the Brabender  
106 farinograph method to characterize different starches before processing. This method in-  
107 volves measuring the torque required to move the impeller at a predetermined rpm while  
108 measuring the temperature of the sample independently throughout the gelatinization pro-  
109 cess. However, this method does not allow measuring directly the usual rheological material  
110 functions but gives only an indication of the viscosity in arbitrary Brabender units (Elias-  
111 son, 2004). To carry out Couette rheometry of starch suspensions during heating, at a rate  
112 comparable to the industrial processes (around  $1\text{ }^\circ\text{C}/\text{s}$ ), Lagarrigue (2002) developed a high

113 heating/cooling rate rheometer. She took into account (via a thermal model) the thermal  
114 gradient in the rheometer gap and was able to propose a model predicting the rheology of  
115 the starch suspension as a function of its thermal history.

116 The properties of starch vary based on its botanical origin and processing it has under-  
117 gone. Hence, suspension rheology also varies widely, it can be either shear thinning or shear  
118 thickening depending on the variety and concentrations (Plana-Fattori et al., 2016; Crawford  
119 et al., 2013). Origins of such varied behaviour of starch suspensions can be deciphered by  
120 leveraging the literature existing on viscosity predictions of broad class of materials known  
121 as non-brownian particulate suspensions.

122 Researchers over the years have developed two methods of modelling the suspension  
123 rheology namely, two-fluid model and mixture model (Vollebregt et al., 2010). In a two-  
124 fluid model the conservation equations are written for both particle phase and the fluid phase  
125 with the interaction terms between the phases. In the mixture approach, the suspension is  
126 modelled as an equivalent single phase. Both two-fluid model and mixture model regard the  
127 liquid and solid phases as interpenetrating continua.

128 The **two-fluid model** is obtained by averaging the equations of motion over the liquid  
129 and the particle phases. This results in a set of equations with a closure problem as it  
130 contains a number of indeterminate terms which are not related to the averaged variables  
131 but associated with the details in the microscopic length scale. Multiple methods exist for  
132 coming up with this closure problem. Two of which are effective medium theory and fully  
133 resolved CFD-DEM simulation. The latter is an Eulerian-Lagrangian method which is com-  
134 putationally expensive. Effective medium theory approach is favourable for thermodynamic  
135 and computational point of view. The indeterminate terms of the two-fluid model are rep-  
136 resented by the effective stress tensors of the phases and interaction force exchanged by the  
137 phases. The expressions for the effective stress tensors and fluid-particle interaction forces  
138 are important as they indicate the origins of these terms.

139 According to Jamshidi et al. (2019), the liquid effective stress tensor is the sum of four  
140 terms. The first is the product of the liquid viscosity( $\mu$ ), the liquid volume fraction ( $1-\phi$ ) and  
141 the mean liquid shear rate ( $\dot{\gamma}$ ). The fourth term is similar to Reynolds stress and becomes

142 important in turbulent case. The second and third terms together are called the ‘particle-  
143 particle stress’. The ‘particle-presence stress’ takes notably into account that the presence  
144 of particles distort the fluid streamlines and generate larger local velocity gradients, which  
145 increase viscous dissipation. This occurs even if the particles follow the liquid i.e. with zero  
146 slip velocity with the surrounding fluid. For dense mixtures, it considers also lubrication  
147 films between a significant number of particles in which viscous dissipation occurs.

148 For non-Brownian, neutrally buoyant, spherical particles dispersed in a Newtonian fluid  
149 in laminar flow and low volume fractions (typically  $\phi < 0.05$ ), Jamshidi et al. (2019) recov-  
150 ered the expression obtained by Einstein (1905) for the part of the ‘particle-presence stress’  
151 due to streamline distortion (in this derivation the lubrication interactions are neglected).  
152 Jamshidi et al. (2019) further claim that the approach often used in two-fluid modelling for  
153 the fluid/fluid interaction, which considers only the first term of the liquid effective stress  
154 tensor is incorrect and should be avoided notably because it predicts a decrease in effective  
155 viscosity as the solid concentration rises.

156 For dense suspensions and soft particle pastes, besides fluid/fluid interactions, a critical  
157 point is how to treat the particle/fluid and particle/particle interactions (Vollebregt et al.,  
158 2010; Van der Sman and Vollebregt, 2012). In particular, shear induced diffusion is a phe-  
159 nomenon of main importance. It has been shown that particle-particle interaction leads to  
160 osmotic pressure of the suspension as derived by Seth et al. (2011). These authors studied  
161 through simulations the variation of osmotic pressure and suspension shear modulus as a  
162 function of the particle properties. It was shown that gradient in osmotic pressure of a sus-  
163 pension contributes to the shear induced particle migration (Vollebregt et al., 2010). Further  
164 Van der Sman and Vollebregt (2012) went on to define an effective granular temperature  
165 based on osmotic pressure for sheared suspensions.

166 In the **mixture approach**, the suspension is considered like a single phase with an  
167 apparent viscosity also called suspension viscosity. The latter includes not only the effect  
168 of streamlines distortion due to the simple presence of the particles but also the parti-  
169 cle/particle and fluid/particle interactions. Analytical and empirical correlations were pro-  
170 posed (e.g. Batchelor and Green (1972)). The four most important factors that dictate a



171 suspension viscosity are (a) suspending medium’s viscosity, (b) granule volume fraction, (c)  
 172 granule deformability, and (d) imposed shear rate (Chen et al., 2007). Granules deformabil-  
 173 ity (granule’s Young’s modulus) is of particular importance for large volume fractions when  
 174 particle-particle interactions becomes predominant (Li et al., 2020; Evans and Lips, 1990).  
 175 The governing momentum equation for the mixture can also be obtained by adding the two  
 176 momentum equations of the two-fluid approach, this allows also developing approximate  
 177 expressions for the suspension viscosity (Jamshidi et al., 2019).

178 For concentrated suspensions, when fluid/fluid interactions becomes negligible compared  
 179 to contact and lubrication forces, DEM simulations can also predict the rheology including  
 180 shear-thickening (Ness and Sun, 2016).

181 Despite these efforts to understand how rheology evolution is connected to the trans-  
 182 formation of the starch suspension during heating, there is no unequivocal single answer to  
 183 explain or model all the experimentally observed phenomena.

## 184 **2. Modelling**

### 185 *2.1. Starch swelling modelling*

186 The previously developed individual starch granule swelling model (Palanisamy et al.,  
 187 2020) is presented briefly here. In this study we use chemically stabilized cross-linked waxy  
 188 maize starch (C\*Tex 06205), provided by Cargil (Baupte, France). This has a  $D_{10}$  greater  
 189 than  $8 \mu m$  and thus is non-colloidal in nature.

190 For a constant reference temperature  $T_{ref}$ , the onset delay time  $t_{onset}$  follows an expo-  
 191 nential distribution for which the cumulative distribution function is:  $1 - e^{-t_{onset}/\tau_{ref}}$ , where  
 192  $\tau_{ref}$  is the characteristic latency time at  $T_{ref}$ . To traduce this, a parameter  $\alpha$  called ‘diffi-  
 193 culty of swelling’ was introduced. For each granule,  $\alpha$  is selected at random from uniform  
 194 distribution in the range  $[0;1]$ . This way, for constant temperature  $T_{ref}$ , the delay time  $t_{onset}$   
 195 follows:

$$t_{onset} = -\tau_{ref} \cdot \ln(1 - \alpha) \quad (1)$$

196 Regarding the swelling onset, equation 1 arises from an experiment carried out a constant

197 temperature of 62 °C during one hour (Palanisamy et al., 2020).

198 For variable temperature, the time delay in onset of swelling is given by solving equations  
199 2 and 3, which means that the delay in onset of swelling is higher at lower temperatures  
200 that at higher temperatures.

$$\ln(1 - \alpha) = - \int_0^{t_{onset}} \frac{dt}{\tau} \quad (2)$$

$$\tau = \max \left( 0, \left( \frac{T_{ref} - T_{min}}{T - T_{min}} \right) \right) \tau_{ref} \quad (3)$$

201 Dimensionless diameter  $S$  is given by

$$S(t) = \frac{D(t) - D_i}{D_f - D_i} \quad (4)$$

202 where  $D(t)$  is the diameter at an instant  $t$ ,  $D_i$  is the initial diameter of the granule,  $D_f$  is  
203 the final diameter of the granule.

204 After the delay time  $t_{onset}$  is overcome, the swelling kinetics is given by:

$$\frac{dS}{dt} = K(1 - S) \quad (5)$$

205 with

$$K = K_{ref}(1 - \alpha)^{0.5} \left( \frac{T - T_{min}}{T_{ref} - T_{min}} \right)^2 \quad (6)$$

206 where  $K_{ref}$  is the rate constant at  $T_{ref}$  for a granule which has no difficulty to swell ( $\alpha = 0$ ).

207 For chemically stabilized cross-linked waxy maize starch (C\*Tex 06205) provided by  
208 Cargill (Baupte, France) the following parameters were estimated:  $T_{min} = 60$  °C,  $\tau_{ref} = 190$   
209 s and  $K_{ref} = 0.0023s^{-1}$  at  $T_{ref} = 62$  °C. From the above equations, one can describe the  
210 swelling kinetics of these starch granules.

211 The following assumptions regarding the starch population were made. Initial diameters  
212 and swelling ratio are from a random normal distribution:  $D_i \subset \mathcal{N}(14.77\mu m, 4.44\mu m)$ ,  
213  $D_f/D_i \subset \mathcal{N}(2.34, 0.33)$ .

214 The details and experimental basis of this swelling model for individual starch granules

215 of cross-linked waxy maize starch were presented in previous work (Deslandes et al., 2019;  
 216 Palanisamy et al., 2020). The model was validated by microscopic observation for different  
 217 temperature ramps (up to 90 °C) and also by comparing size distribution obtained by  
 218 Malvern mastersizer after different non-linear heat treatments. These model parameters  
 219 should be re-calibrated for other waxy cross-linked starches and the model should be modified  
 220 for other types of starches and is not applicable if the granule ruptures.

## 221 2.2. CFD-DEM Modelling

222 A two-phase modelling approach is used as by Jamshidi et al. (2019) except that the  
 223 solid phase (starch granules) is not treated as a continuous phase but as a discrete one: each  
 224 particle is characterized by its centre position and velocity. This approach is similar to the  
 225 one used by Qian et al. (2014).

226 An Eulerian approach is used for solving flow and heat transfer of the continuous fluid (in  
 227 this case pure water). In each representative elementary volume (REV) there are a number  
 228 of solid particles (in this case starch granules).

229 For the fluid phase, in a general case, the momentum equation holds:

$$(1 - \phi)\rho_f \frac{\partial \vec{v}_f}{\partial t} + (1 - \phi)\rho_f \vec{v}_f \cdot \vec{\nabla} \vec{v}_f = -\vec{\nabla} \cdot \bar{\bar{S}} + (1 - \phi)\rho_f \vec{g} + \vec{f}_{p \rightarrow f} \quad (7)$$

230 where  $\vec{f}_{p \rightarrow f}$  is the sum of the forces exerted on the fluid by the particles per unit volume  
 231 (notably drag forces), and  $\bar{\bar{S}}$  is the fluid effective stress tensor.

232 For non-Brownian, neutrally bouyant, spherical particles dispersed in a Newtonian fluid  
 233 in laminar flow and low volume (typically  $\phi < 0.05$ ), Jamshidi et al. (2019) obtained the  
 234 following approximation.

$$\bar{\bar{S}} = p\bar{\bar{I}} - (1 + 2.5\phi)\mu\bar{\bar{\gamma}} \quad (8)$$

235 where  $\bar{\bar{\gamma}}$  is twice the deformation rate tensor. we can use the deformation rate tensor of  
 236 the fluid because particle velocity rapidly tends to the surrounding fluid velocity (gradient  
 237 of the mean particle velocity is close to the fluid velocity gradient).

238 In the gap of a Couette rheometer only the variations in radial directions are considered;  
 239 taking a small gap, i.e.  $(R_e - R_i) \ll R_e$ , a Cartesian approximation can be used. The  
 240 radial position in the gap is referred as  $x$  (unit vector  $\vec{u}_x$ ). On the internal wall  $x = 0$ , on  
 241 the external wall  $x = e$ . The flow direction is referred as  $y$ .

242 For the fluid, only momentum balance in  $y$  direction is considered and only the shear  
 243 stress  $\tau_{xy}$  is involved. Fluid velocity is assumed to be in  $y$  direction (unit vector  $\vec{u}_y$ ) its  
 244 magnitude is referred as  $v_f$ ; hence:

$$(1 - \phi)\rho_f \frac{\partial v_f}{\partial t} + \frac{\partial \tau_{xy}}{\partial x} = f_{y,p \rightarrow f} \quad (9)$$

245 where  $f_{y,p \rightarrow f}$  is the sum of the forces exerted on the fluid by the particles located in the REV  
 246 per unit volume in  $y$  direction. Since there is no variation in pressure nor in fluid velocity  
 247 in  $y$  direction, the convective term of momentum and the pressure gradient term vanish.

248 In this equation  $\tau_{xy}$  accounts for the fluid/fluid interaction. Therefore, this term should  
 249 take into account the increased viscous dissipation due the distortion of the streamlines  
 250 caused by the simple presence of the particles. According to equation 8Jamshidi et al.  
 251 (2019), for dilute suspensions, it can be approximated by Einstein's correlation. Unfortu-  
 252 nately, as per our knowledge, there is no available expression for higher volume fraction,  
 253 which only integrates the additional stress due to streamlines distortion. As a case in point,  
 254 the correlation of Batchelor and Green (1972) applies for higher volume fractions but in-  
 255 cludes lubrication interactions as well which are here directly calculated in the DEM part  
 256 of the model. Thus, we intend to use Einstein (1905) expression for the effective viscosity  
 257 even though it certainly underestimate the additional stress due to streamline distortion  
 258 for intermediate solid volume fractions of  $0.05 < \phi < 0.3$ . At least, this relation (equation  
 259 11) takes into account that higher the particle volume fraction, the more the streamline are  
 260 distorted, which increases viscous dissipation.

$$\tau_{xy} = -\mu_e(T_f, \phi) \frac{\partial v_f}{\partial x} \quad (10)$$

261

$$\mu_e(T, \phi) = \mu(T_f) \cdot (1 + 2.5\phi) \quad (11)$$

262 The energy balance for the fluid is :

$$(1 - \phi)\rho_f C_{pf} \frac{\partial T_f}{\partial t} + \frac{\partial q}{\partial x} = Q_{p \rightarrow f} + Q_{diss} \quad (12)$$

263 where

$$q = -\lambda \frac{\partial T_f}{\partial x} \quad (13)$$

264 where  $Q_{p \rightarrow f}$  is the sum of the heat fluxes from the particles to the fluid and  $Q_{diss}$  is the  
265 viscous dissipation of mechanical energy into heat due to the viscous forces.

$$Q_{diss} = \mu_e(T_f, \phi) \left( \frac{\partial v_f}{\partial x} \right)^2 \quad (14)$$

266 The external wall has zero velocity and imposed temperature evolution, say

$$267 \quad x = e : v_f = 0 \text{ and } T_f = T_{e(t)}.$$

268 The internal wall has a given velocity, this corresponds to a strain controlled rheometer  
269 (constant global shear rate):

$$270 \quad x = 0 : v_f = v_i = \dot{\gamma} \cdot e$$

271 The internal wall is also characterized by a given thermal inertia per unit surface ( $\frac{m_{iw} C_{p,iw}}{A_{iw}}$ ):

$$272 \quad x = 0 : \frac{m_{iw} C_{p,iw}}{A_{iw}} \frac{dT_{iw}}{dt} = \lambda \frac{\partial T_f}{\partial x}$$

273 Periodic boundary conditions apply in  $y$  and  $z$  direction.274 The trajectory of each particle of index  $i$  is given by :

$$m_i \frac{d^2 \vec{x}_i}{dt^2} = \vec{F}_{f \rightarrow i} + \sum_j \vec{F}_{j \rightarrow i} \quad (15)$$

275 with  $\vec{F}_{f \rightarrow i} = 6\pi\mu r_i (v_f \cdot \vec{u}_y - \vec{v}_i)$  (Stokes viscous drag force). Here,  $v_f$  is the fluid velocity  
276 interpolated at particle centre position  $\vec{x}_i$  (piecewise linear velocity profile is considered)  
277 and  $r_i$  is the radius of granule  $i$ .

278 The maximal observed particle/fluid relative velocity was about 1 mm/s in our case, the

279 maximal granule diameter is around 50  $\mu\text{m}$ , and the lowest liquid viscosity is 0.32 mPa.s.  
 280 Therefore, Reynolds number was always lower than 0.15, inertial effect is negligible and  
 281 Stokes law is applicable. Therefore we can write that

$$F_{p \rightarrow f} = - \sum_{i \in REV} \frac{\overrightarrow{F_{f \rightarrow i}}}{V_{REV}} \cdot \overrightarrow{u_y} \quad (16)$$

282 Lubrication forces in the normal direction are expressed by the following expression. Only  
 283 the first-order normal force is considered, a more detailed lubrication interaction (Kim and  
 284 Karrila, 2013) will be integrated in further work.

$$\overrightarrow{F_{j \rightarrow i}} = 6\pi\mu r_i \frac{\beta^2}{(1 + \beta)^3} \frac{r_i + r_j}{h_{ij,eff}} ((\overrightarrow{v_i} - \overrightarrow{v_j}) \cdot \overrightarrow{n_{ij}}) \overrightarrow{n_{ij}} \quad (17)$$

$$285 \quad \overrightarrow{x_{ij}} = \overrightarrow{x_j} - \overrightarrow{x_i}$$

$$286 \quad d_{ij} = |\overrightarrow{x_{ij}}|$$

$$287 \quad \overrightarrow{n_{ij}} = \overrightarrow{x_{ij}} / d_{ij}$$

$$288 \quad h_{ij} = d_{ij} - (r_i + r_j)$$

289  $\overrightarrow{n_{ij}}$  is the centre to centre unit vector and  $h_{ij}$  is the surface to surface distance.

$$290 \quad h_{ij,eff} = \max(h_{ij}, 0.005(r_i + r_j))$$

$$291 \quad \beta = r_j / r_i$$

292 No gravity, no Brownian or repulsive forces were considered.

293 If two particles come in contact ( $h_{ij} < 0$ ) an instantaneous elastic rebound is assumed.  
 294 Friction between particles are assumed negligible, this assumption should be reconsidered,  
 295 especially for high volume fraction, since starch granules cannot be considered as smooth  
 296 spheres. They exchange momentum  $\overrightarrow{P}$  in the normal direction  $\overrightarrow{n_{ij}}$  given by:

$$\overrightarrow{P_{j \rightarrow i}} = \frac{2m_i m_j}{m_i + m_j} ((\overrightarrow{v_j} - \overrightarrow{v_i}) \cdot \overrightarrow{n_{ij}}) \overrightarrow{n_{ij}} \quad (18)$$

$$297 \quad m_i \delta \overrightarrow{v_i} = \overrightarrow{P_{j \rightarrow i}}; \quad m_j \delta \overrightarrow{v_j} = \overrightarrow{P_{i \rightarrow j}} \quad (19)$$

298 where  $\delta \overrightarrow{v_i}$  and  $\delta \overrightarrow{v_j}$  are the difference in velocities before and after the collision for particles

299  $i$  and  $j$  which are applied during next time step.

300 Elastic rebound was assumed on the internal and external walls ( $x = 0$  and  $x = e$ ).  
 301 Periodic boundary conditions were assumed in  $y$  and  $z$  direction for both the particle and  
 302 fluid phases.

303 Seth et al. (2006) have used a similar approach of elastohydrodynamic lubrication for  
 304 microgels. However, they have used Hertzian contact model when particles touch each other  
 305 in place of the instantaneous shocks in the model presented here. In their case with a  
 306 high concentration suspension, a detailed contact model was essential because the volume  
 307 fraction was higher than the random close packing fraction. In our case, the maximum  
 308 volume fraction is about 30 %, much lower than the random close packing value of 68 %  
 309 which was determined for our poly-disperse suspension using the methodology given in Farr  
 310 and Groot (2009). Thus, the current approach (instantaneous elastic collision) seems a valid  
 311 approximation.

312 The temperature evolution of each particle is given by:

$$m_i C_{pp} \frac{dT_i}{dt} = Q_{f \rightarrow i} + Q_{ld} \quad (20)$$

313 with  $Q_{f \rightarrow i} = 4\pi r_i^2 h (T_f - T_i)$ ,  $h = Nu \frac{\lambda}{2r_i}$  and  $Nu \approx 2$ . Here,  $Q_{ld}$  is the dissipation term for  
 314 the lubrication forces and drag. The same is calculated using the following expression:

$$Q_{ld} = \frac{1}{2} \sum_j [\overrightarrow{F_{j \rightarrow i}} \cdot (\vec{v}_i - \vec{v}_j)] + \overrightarrow{F_{f \rightarrow i}} \cdot (v_f \vec{u}_y - \vec{v}_i) \quad (21)$$

315 Initially the granules are randomly distributed, the fluid and particle velocity varies  
 316 linearly in the gap and the temperature is uniform and equal to the minimal swelling tem-  
 317 perature (60 °C)

318 Elsewhere researchers have often used fabric tensor to quantify anisotropy (Ness and  
 319 Sun, 2016; Barreto et al., 2009). However, fabric tensor is typically used for the cases of  
 320 quasi-static systems and we have a dynamic system with almost no true contacts. This is  
 321 why we consider ‘quasi-contact’ when the centre-centre distance between particle  $i$  and  $j$  is

322 less than  $1.2(r_i + r_j)$  where  $r_i$  and  $r_j$  are the radius of granules  $i$  and  $j$ . There exists two  
 323 definitions of fabric tensor and multiple definitions for the fabric deviator. Here, we follow  
 324 the definitions as reported in Barreto et al. (2009). They are as follows

$$\Omega_{ij} = \frac{1}{N_c} \sum_{k=1}^{N_c} n_i^k n_j^k \quad (22)$$

325 where  $N_c$  is the total number of quasi-contacts and  $n_i^k$  denotes the component of the nor-  
 326 malized centre to centre vector in direction  $i$  for the  $k$ -th quasi-contact.

327 Thus, we have the fabric tensor which is a  $3 \times 3$  matrix. The eigenvectors give the  
 328 dominant directions of these quasi-contacts. The eigenvalues ( $\Omega_1, \Omega_2 < \Omega_1, \Omega_3 < \Omega_2$ )  
 329 are called major, intermediate and minor magnitudes of the fabric tensor. In an isotropic  
 330 situation  $\Omega_1 = \Omega_2 = \Omega_3$ . The deviator fabric is defined by the following equation as reported  
 331 in Barreto et al. (2009). The higher its value the more the configuration is anisotropic.

$$\Omega_d = \frac{1}{\sqrt{2}} [(\Omega_1 - \Omega_2)^2 + (\Omega_2 - \Omega_3)^2 + (\Omega_1 - \Omega_3)^2]^{0.5} \quad (23)$$

### 332 2.3. Numerical solving

333 The simulated domain was of 1 mm in  $x$ -direction ( $R_e - R_i$  of the Couette rheometer)  
 334 and  $200 \mu\text{m}$  in the two other directions.

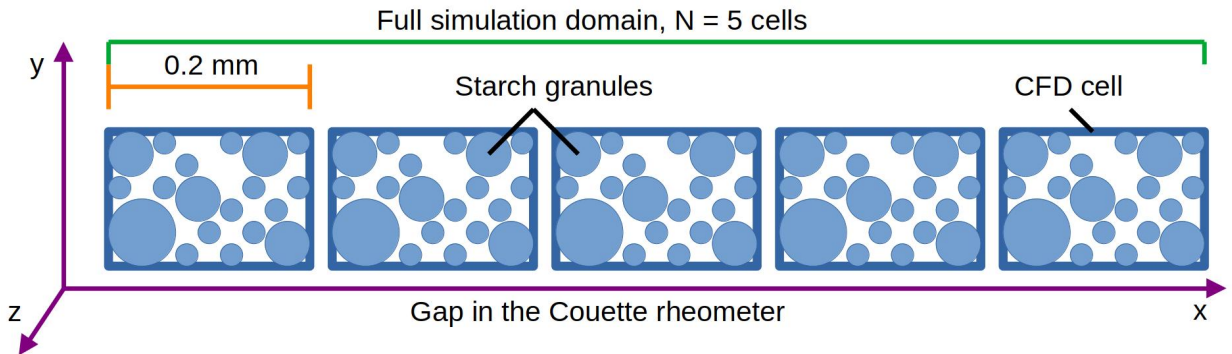


Figure 1: Simulation domain

335 Particle centroid method is used for coupling the DEM to the CFD. As the name sug-



336 gested the entire particle is assigned to a particular cell based on the location of the centroid  
337 of the particle. This method is known to be accurate only when the fluid cell size is sig-  
338 nificantly larger than the diameter of the particle. Thus, the fluid equations were solved  
339 with the finite volume method with  $N = 5$  cells and initially 100 granules in each cell as  
340 a compromise. Also, if  $N$  is too low, the fluid velocity and temperature profile are not  
341 accurately represented and if  $N$  is too high, the number of granules in each cell becomes  
342 too low to be representative. To ensure periodic condition in  $y$  and  $z$  direction, for each  
343 particle at position  $(x,y,z)$  inside the domain, 4 mirror particles (same radius and velocity)  
344 were considered at  $(x, y + \Delta y, z), (x, y - \Delta y, z), (x, y, z + \Delta z)$  and  $(x, y, z - \Delta z)$ .

345 First, for each granule, its initial radius ( $r_0$ ), its maximum swelling ratio ( $r_{max}/r_0$ ) and  
346 its difficulty of swelling ( $\alpha$ ) are chosen from the distributions as defined in section 2.1. Then  
347 its position is randomly chosen to avoid any overlap. This is ensured by checking for overlap  
348 at every step and if overlap occurs a new position is chosen until there is no overlap. This  
349 methodology works as the initial volume fraction of the granules ( $\phi$ ) is very low.

350 At each time step, the swelling of each granule is calculated (according to the temperature  
351 history of the granule and difficulty of the swelling parameter) in order to re-evaluate its  
352 radius and mass. All the different forces and heat fluxes are then estimated. Finally, the  
353 velocities, temperatures (fluid and particles) and particle positions are actualized by an  
354 explicit first order scheme. Fluid velocity is assumed to be in  $y$  direction. Particle positions  
355 and velocities are calculated in 3D.

356 The maximum time step is dictated by the characteristic Stokes time ( $\rho r^2/\mu$ );  $\Delta t = 10^{-5}$   
357 s was chosen (smallest granules having radius of about  $3 \mu\text{m}$ ). The code was implemented in  
358 Matlab (MATLAB, 2018). For the fluid (Eulerian part), the velocity and the temperature  
359 profiles were considered as piecewise linear and an explicit first order scheme was used.

360 The presented results correspond mainly to one simulation of  $N = 5$  cubic cells with a  
361 side length of 0.2 mm each. In each cell, 100 granules were initially randomly located. This  
362 allows comparing what happens with the granules depending on their initial position in the  
363 gap. The average shear rate was of  $10 \text{ s}^{-1}$  (internal wall velocity 10 mm/s). The precise  
364 results depend on the initial position and radius of the granules, four sets of random initial

365 conditions were simulated. The results were comparable and the reported analyze(trends,  
 366 dominant phenomena, etc..) applies also for the other runs. A comparison between two runs  
 367 for the evolution of some key variables is presented and the variability of some parameter  
 368 between the four runs is also presented.

369 The simulation duration was of 1 min, during which the external wall temperature in-  
 370 creased linearly from 60 to 90 °C. The mean volume fraction was initially 2.6 % and 30.0 %  
 371 at the end of the simulation. If all the granules were completely swollen the volume fraction  
 372 would have been 32.8 %, so on average, 91 % of the possible swelling is achieved after 1 min  
 373 of heating.

374 The liquid properties are those of pure water:

$$375 C_p = 4180 J.K^{-1}.kg^{-1}$$

$$376 \lambda_f = 0.6 W.m^{-1}.K^{-1}$$

$$377 \mu = 2.414 \cdot 10^{-5} \cdot 10^{(247.8/(T_f+133))} \text{ Pa.s where } T_f \text{ is in } ^\circ\text{C}.$$

378 The starch gelatinization enthalpy was neglected. In fact, the enthalpy of gelatinization  
 379 is less than 10 kJ per kg of dry starch (Desam et al., 2018a; Li et al., 2020), which corresponds  
 380 to 0.3 kJ per kg of suspension. This is negligible compared to the sensible heat increase  
 381 from 60 to 90 °C, which is about 125 kJ per kg of suspension.

#### 382 2.4. Results and Discussion

383 Figure 2a presents at 5 different instants the temperature profile of the fluid vs position  
 384 in the gap. Similarly, Figure 2b shows the temperature profile of the 500 granules and of  
 385 surrounding fluid vs position in the gap at a time of 30 s from the start of simulation. The  
 386 temperature of the particles is always very close to the local fluid temperature as shown  
 387 in Figure 2b. This is always the case irrespective of the time. This can be explained  
 388 by the very short characteristic time to reach thermal equilibrium. If we consider the  
 389 energy balance of one particle (taking into account only the heat exchange with the fluid):

$$390 m_p C_{pp} \frac{dT_i}{dt} = hS(T_f - T_i).$$

391 A characteristic time appears as follows:  $t_{th} = m_p C_{pp} / (h.S)$ , where  $S$  is the surface area  
 392 of the particle and  $h$  is obtained considering  $Nu = 2$ . This results in a  $t_{th}$  which is less

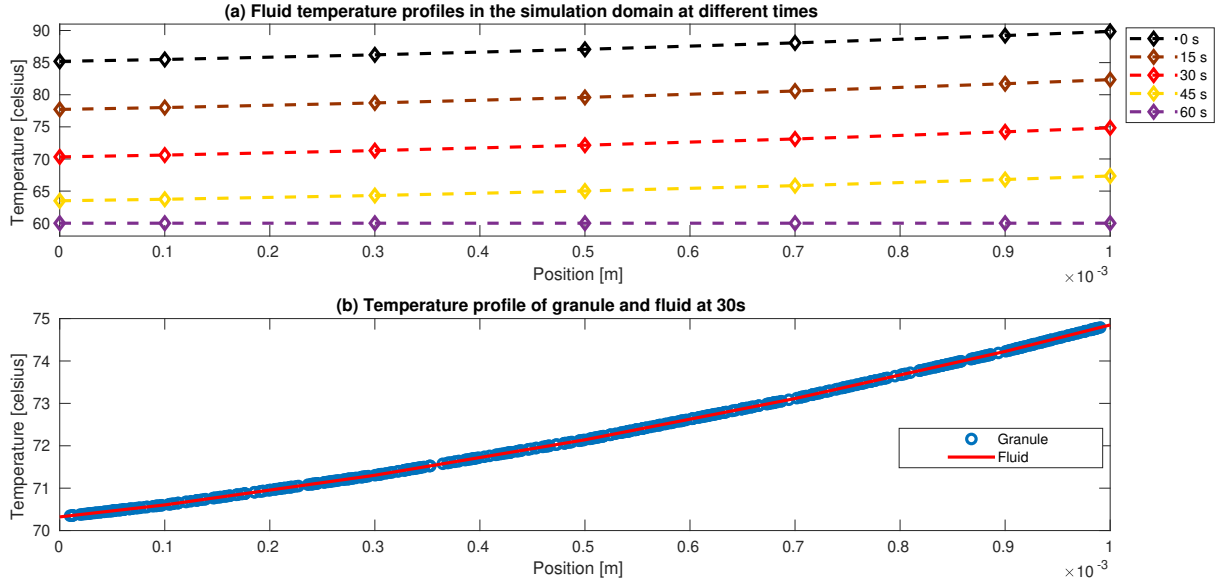


Figure 2: Temperature of granules and fluid

393 than 1 ms. The profile is nearly linear, the external/internal temperature difference reaches  
 394 about 5 °C. The viscous dissipation of mechanical energy into heat is negligible compared  
 395 to the conductive heat flux even at the end of the simulation. From Figure 7b (presented  
 396 later) it appears that the dissipation is less than  $0.2 \text{ W/m}^3$ , this is much lower than the  
 397 sensible heat increase of the fluid whose temperature increases by  $\Delta T = 30 \text{ °C}$  in  $\Delta t = 60$   
 398 s. In our case,  $\rho_f C_{pf} \Delta T / \Delta t$  is about  $2 \cdot 10^6 \text{ W/m}^3$ .

399 Figure 3 presents at 9 different time instants, the fluid and particles velocity profiles  
 400 ( $v_y$  vs  $x$ ). Cumulative particle density is superimposed, i.e. the fraction of particles whose  
 401 centre is at a position lower than  $x$ . For a uniform distribution, this line would correspond  
 402 to a straight line. From this we can see that near the wall there is a region where particles  
 403 are absent and this region increases slightly as the swelling increases. This is arising due to  
 404 the wall effects as there exists a minimum distance of particle radius between the centre of  
 405 the particle and the wall (around  $20 \mu\text{m}$ ).

406 At a given time, since the curvature is ignored, the shear stress is almost constant  
 407 throughout the gap, i.e. versus  $x$  in a quasi steady state approximation. This is not exactly  
 408 true since the particles are growing and moving during the time, but it should apply on

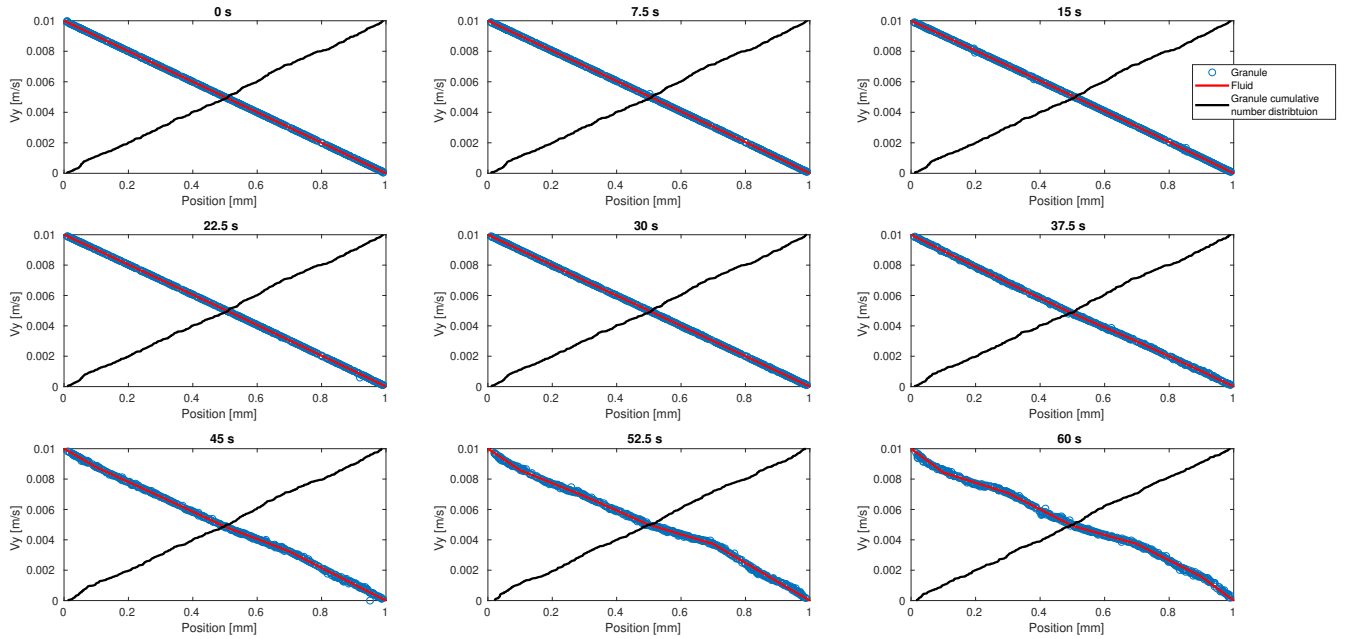


Figure 3: velocity of granules and fluid

409 average. Therefore, if we assume that the shear stress is almost the same across the rheometer  
 410 gap, at the beginning of the simulation, one could expect a higher velocity gradient near  
 411 the heated wall where the viscosity of the continuous phase is lower. This effect is observed  
 412 at  $t = 15$  s, the local shear rate varies monotonically between  $9.8 \text{ s}^{-1}$  to  $10.2 \text{ s}^{-1}$  from  
 413 internal to an external wall. However, this variation is very small because of the limited  
 414 temperature difference. At the later instances when granules swell first near the external  
 415 wall, the opposite effect is expected because the suspension viscosity will be higher due to  
 416 the increased volume fraction of the granules. A lower shear rate is effectively observed  
 417 near the heated wall ( $10.7 \text{ s}^{-1}$ ) than near the internal wall ( $12.2 \text{ s}^{-1}$ ) at  $t = 45$  s. But in  
 418 the same time, the particle density decreases near the wall. This explains that the velocity  
 419 gradient is higher than the average one ( $10 \text{ s}^{-1}$ ) near the wall and reaches its minimum  
 420 value ( $8 \text{ s}^{-1}$ ) near the centre of the gap. This phenomenon of depletion of particles near the  
 421 wall and subsequent higher shear rate is discussed in literature Coussot (2016). In addition,  
 422 the interaction between a particle and the smooth wall generates no friction contrary to the

423 interaction between particles. This highlights that the boundary effects are not completely  
 424 negligible even for a low ratio of particle diameter to gap width, which is about 4 % in  
 425 our case. In future works the impact of wall lubrication forces and friction forces can be  
 426 interesting to investigate. It is also observed that the particle velocity fluctuations increase  
 427 with time. This is because the granules swell and occupy more volume and they interact  
 more and more in a random manner.

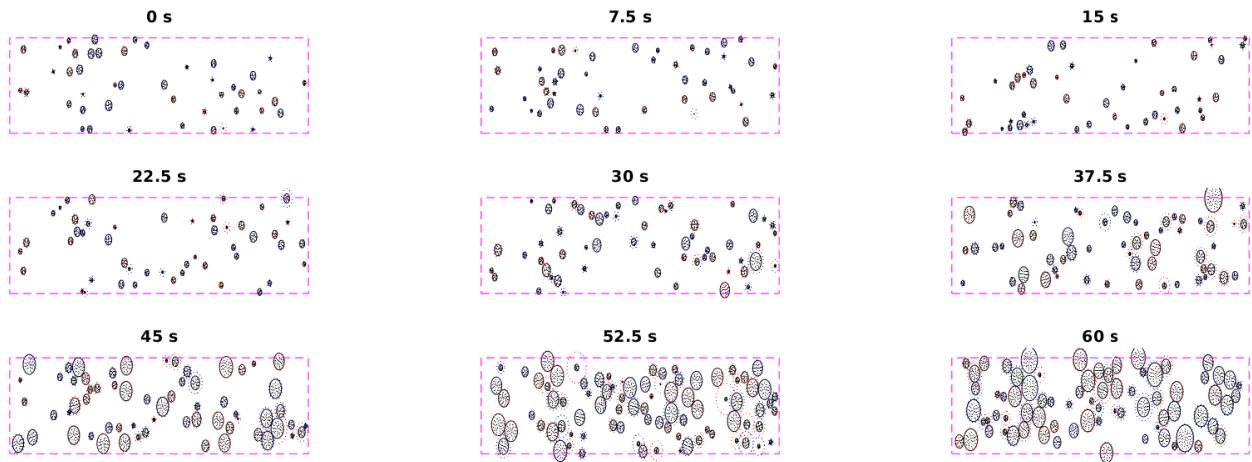


Figure 4:  $xy$  cross section of the simulation domain

428  
 429 Figure 4, shows a  $xy$  section (at middle  $z$  position) of the gap at 9 different instants.  
 430 The granule swelling begins significantly after about 30 s. As we expected a higher degree of  
 431 swelling is observed near the external heated wall (right side of the graphs) but the difference  
 432 through the gap appears small, perhaps because of the particle movements. It should be  
 433 noted that the swelling model was developed by observing a very dilute suspension without  
 434 any motion or collision and here, we employ the same in presence of particle interactions,  
 435 these interactions could modify the swelling kinetics. This should be investigated further.  
 436 In the figure at 60 s, we see a small grouping of particles.

437 Figure 5 shows some parameters evolution as the simulation progresses. Here we show  
 438 two runs of the simulation with different initial properties(radius, swelling difficulty) and  
 439 positions of the starch granules (reference case at left). Figure 5a shows the evolution of

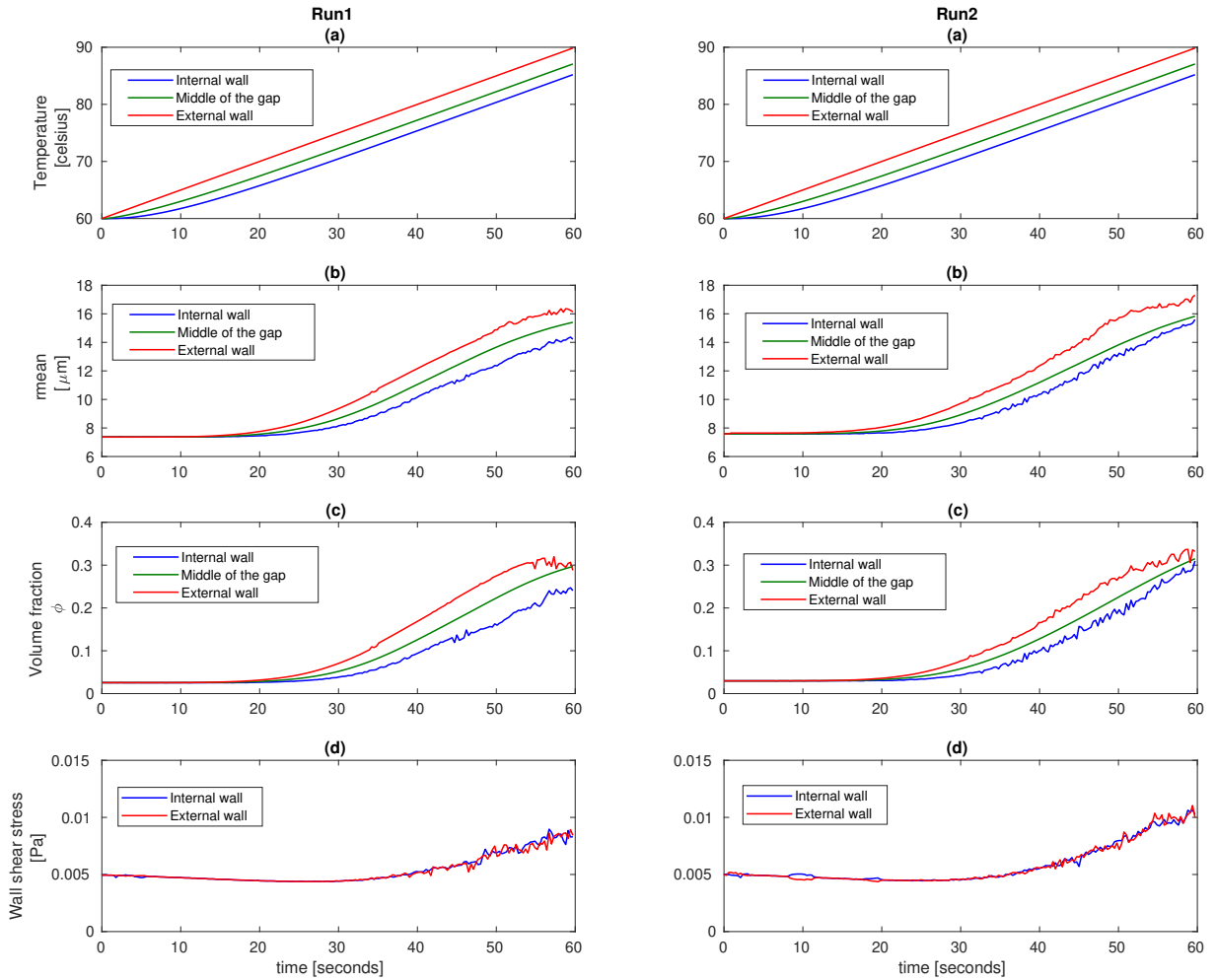


Figure 5: Time evolution of suspension parameters at different regions of the simulation domain

440 fluid temperature at the internal and external walls and in the middle of the gap. There is a  
 441 time lag for the temperature rise from external to internal wall of about 10 s. Fig 5b presents  
 442 the evolution of the mean granule radius over the entire gap, only for the internal cell and  
 443 only for the external cell. The granules begin to swell significantly near the external heated  
 444 wall when its temperature reaches around 70 °C. Similar to the temperature, there is also a  
 445 time delay for granule swelling from the external to the internal wall. Figure 5c presents the  
 446 evolution of the mean particle volume fraction  $\phi$  over all the gap, the internal cell and the  
 447 outer cell. These time series are directly related to those of granules radius. Near the heated

448 external wall, the volume fraction reaches a plateau whereas the mean value over the gap  
 449 and the value near the internal (colder) wall are still increasing at the end of heating time  
 450 of 1 min. Figure 5d shows the evolution of the wall shear stress. Shear stress first decreases  
 451 slightly because when the temperature is increased, liquid viscosity decreases. But when the  
 452 volume fraction reaches about 10 %, the shear stress increases significantly. The swelling  
 453 effect then becomes predominant compared to liquid viscosity decrease. This stress is mainly  
 454 due to the viscous friction between the fluid layers (represented here by the 5 cells). This  
 455 friction is proportional to the effective viscosity which increases with the particles volume  
 456 fraction. Another contribution of global stress is due to the movement of the particle relative  
 457 to the fluid because of the lubrication forces and the collisions. For example, one particle  
 458 located near the internal wall with a high velocity in the  $y$ -direction can move toward a  
 459 more external zone, and then via the Stokes forces, it will communicate some momentum  
 460 (in the  $y$ -direction) to this new fluid layer. Alternatively, it can also interact with slower  
 461 particles of this layer. For a given run, the shear stress on the internal and external wall  
 462 are almost equal at a given time. Random collisions can explain these fluctuations and  
 463 the small differences between internal and external values. Nevertheless, averagely constant  
 464 shear stress across the gap seems ensured.

465 From figure 5, it can be observed that the temperature evolution is the same for the  
 466 two runs. The mean radius and volume fraction evolutions over the whole gap are slightly  
 467 different between the two runs. This can be explained by a small difference in initial volume  
 468 fraction (run 1: 2.6 % ; run 2: 2.9 %) as well as differences in swelling difficulty and swelling  
 469 ratio which are randomly chosen. When we observe the internal or external cells, there  
 470 are more fluctuations because of the random walk of the particles. The most important  
 471 difference between the two runs is observed for the shear stress ( $\tau_{xy}$ ). Initially the time  
 472 series are almost the same, but when swelling becomes significant ( $t > 30$  s) the shear stress  
 473 becomes higher for run 2. This can be explained by a slightly higher volume fraction (at  
 474 the end of simulation run1: 30 %, run2: 32 %) this leads to a higher effective viscosity but  
 475 also to an increase of particle/particle interactions (collisions and lubrication).

476 <sup>☆</sup> average for  $55 \text{ s} < t < 60 \text{ s}$

Table 1: Variability between runs

	$\phi_{initial}(\%)$	$\phi_{final}(\%)$	$T_{iwfinal}$ ( $^{\circ}C$ )	$\tau_{xyfinal}^{\star}$ ( $mPa$ )	$D_{rwfinal}^{\star}$ ( $\mu m^2/s$ )	$\Omega_{d.final}^{\star}$	$d_{nn.final}$ ( $\mu m$ )
run1	2.6	30.0	85.20	8.1	79	0.060	31.2
run2	2.9	31.5	85.20	9.9	76	0.065	31.9
run3	2.5	24.5	85.20	7.3	56	0.073	30.9
run4	2.7	30.2	85.20	8.7	82	0.067	31.2
mean	2.7	29.1	85.20	8.5	73	0.066	31.3
std	0.2	3.1	< 0.01	1.1	12	0.005	0.4

477

478 Table 1 presents the variability of some parameters between the four runs (some of them  
479 will be presented later). The variability is mainly linked to the range of initial volume  
480 fraction. For example run 3 has the lowest initial value of  $\phi$  and logically the shear stress  
481 and the random walk diffusivity ( $D_{rw}$ ) are also the lowest. Variability could be reduced  
482 by increasing box sizes in  $y$  and  $z$  direction. To compare easier different runs, the particle  
483 properties could be fixed and only the initial position could be varied. To obtain precise  
484 quantitative values, average over large number of simulations could be taken. Nevertheless  
485 the trends are the same and the phenomena analyzed of run 1 applies also to the other runs.  
486 In the following only results for run 1 are presented.

487 To illustrate the movement of the particles inside the gap, Figure 6d presents the time  
488 evolution of the  $x$ -displacement of 20 particles (initially 4 particles in each of the 5 cells).  
489 At the beginning of the simulation, the granules are small, they interact only weakly by  
490 lubrication forces; random walk of low amplitude is observed. After about 40 s, when  
491 the volume fraction reaches about 10 % (see Fig 5c), the grain grain interactions induce a  
492 random walk movement which becomes more significant and the displacement sometimes  
493 exceed 100  $\mu m$ , which is not negligible compared to the gap width. Figure 6a presents  
494 the temperature evolution of these particles. In the beginning, the particle temperatures  
495 increase smoothly but when a random walk becomes significant, small fluctuations appear  
496 as can be observed in the zoom view in figure 6b and 6c. This means that the particle moves  
497 randomly between layers of different temperatures (more or less close to the heating wall).



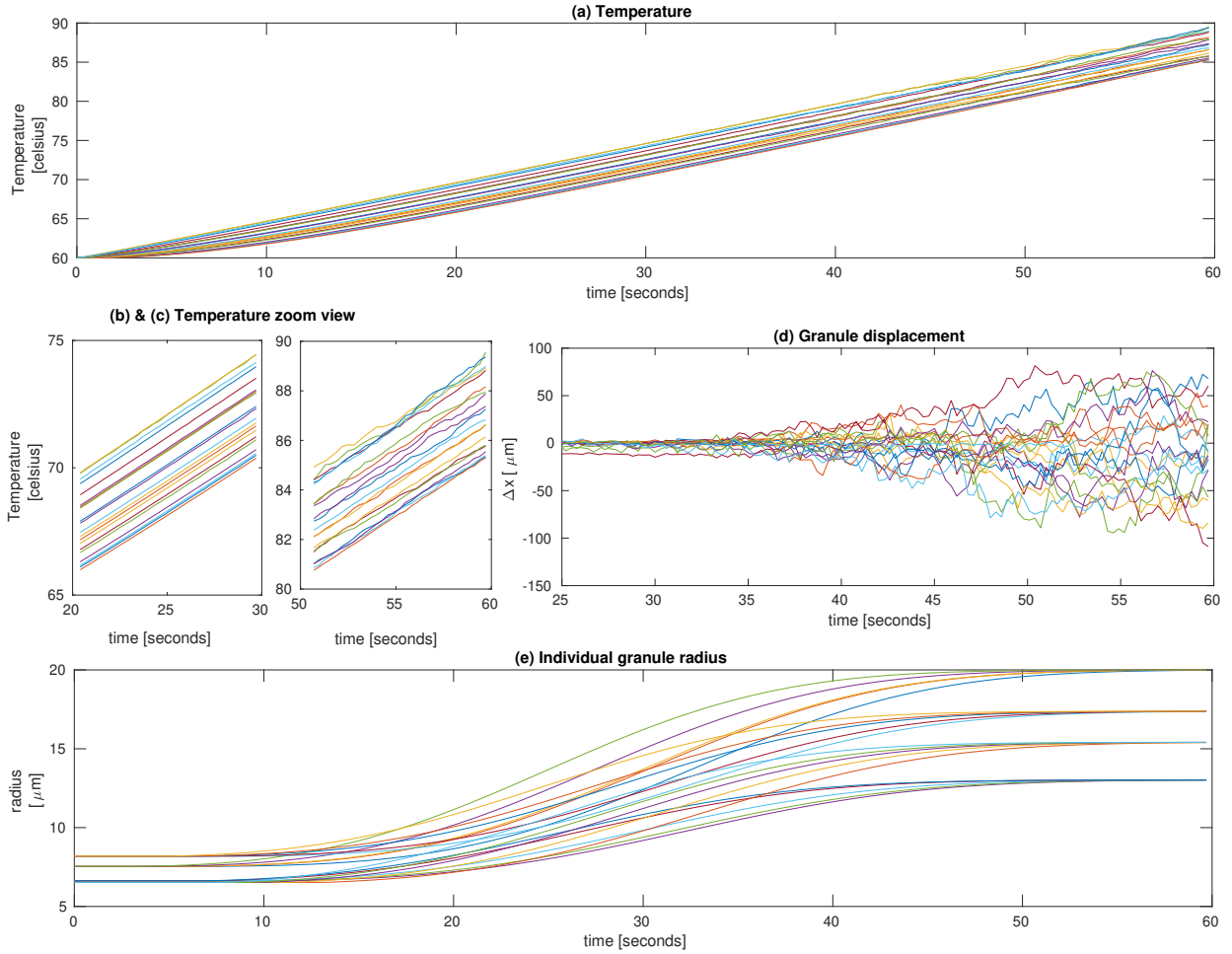


Figure 6: Sample granule properties evolution during simulation

498 When a particle moves towards a cold layer, heat is transferred from this particle to the  
 499 fluid. This participates in heat transfer from the external to the internal wall. Figure 6e  
 500 presents the radius evolution of these 20 particles. Initially, the four tracked particles in a  
 501 cell have the same radius and the difficulty to swell as the four particles in the other cells  
 502 to see the influence of the initial position in the gap of identical granules. As expected, the  
 503 particles close to the heating wall begin to swell earlier and faster.

504 Figure 7a presents the variance of displacement ( $\sigma^2$ ) in  $x$ -direction of all the particles  
 505 respectively from 35 to 40 s ( $\phi \approx 10\%$ ) and from 55 to 60 s ( $\phi \approx 30\%$ ). For one-dimensional  
 506 random walk it is expected that  $\sigma^2 \approx 2D_{rw}t$  where  $D_{rw}$  is the self-diffusivity of the particles.

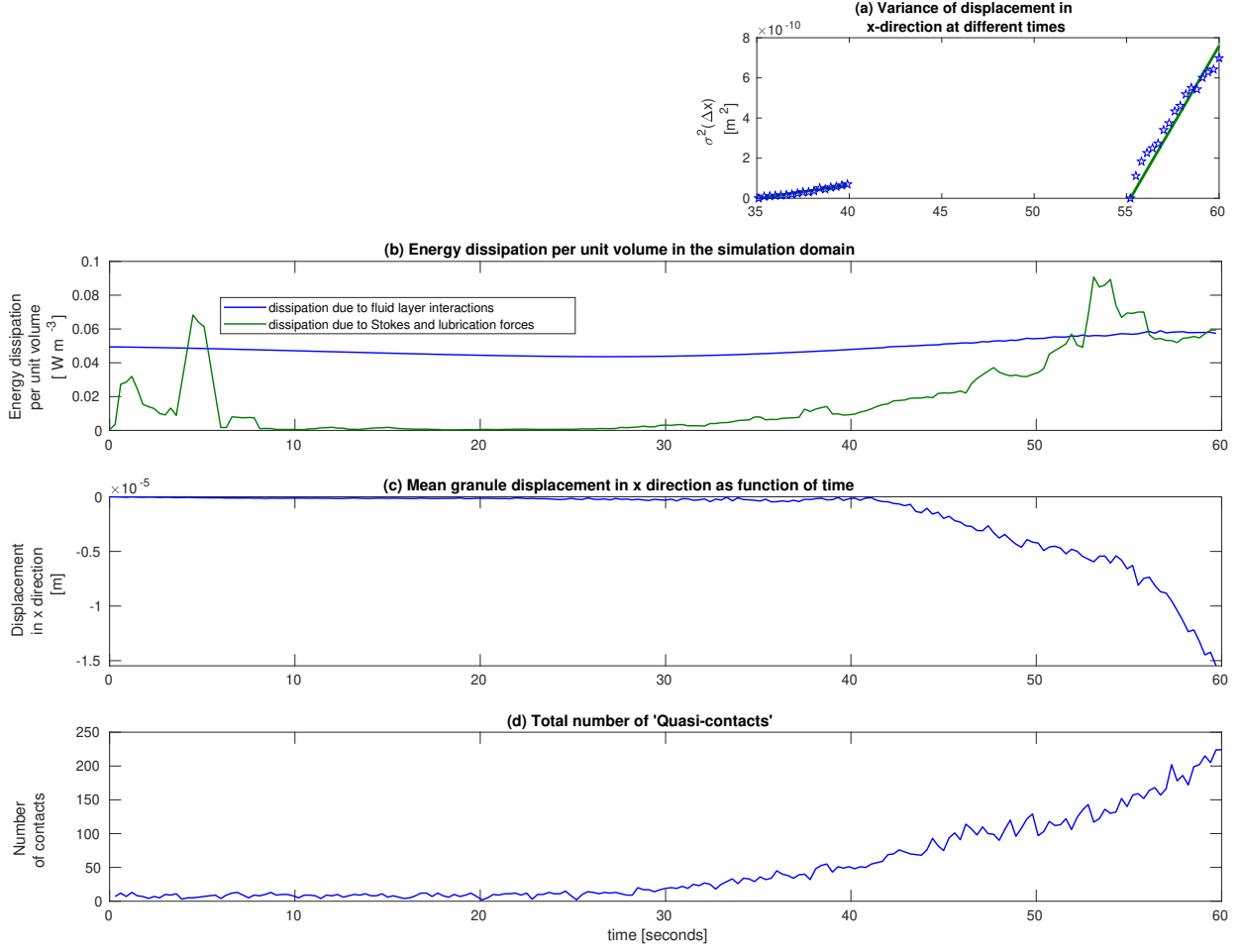


Figure 7: Granule population properties evolution with time

507 Different formulas are given in Leighton and Acrivos (1987), in a first approach the findings  
 508 of Eckstein et al. (1977) can be used.

$$\frac{D_{rw}}{r^2 \dot{\gamma}} \approx \frac{\phi}{10} \quad (24)$$

509 In our case  $\dot{\gamma} = 10 \text{ s}^{-1}$ ; when  $\phi \approx 10 \%$  most of the granules are unswollen, mean radius  
 510 is about  $7 \mu\text{m}$  and when  $\phi \approx 30 \%$  mean radius is about  $17 \mu\text{m}$ , based on this formula  
 511 the self-diffusivity is expected to be around  $4.9 \cdot 10^{-12} \text{ m}^2/\text{s}$  and  $8.7 \cdot 10^{-11} \text{ m}^2/\text{s}$  respectively.  
 512 This is close to our estimates of  $D_{rw}$ , which is  $5 \cdot 10^{-12}$  and  $1 \cdot 10^{-10} \text{ m}^2/\text{s}$  for these periods

513 respectively (values obtained by linear regression). This is much higher than the Brownian  
514 diffusivity of the particles due to the thermal agitation (collisions of the water molecules on  
515 the granules)  $D_B = \frac{k_B T}{6\pi\mu r} \approx 5 \cdot 10^{-14} \text{ m}^2 \text{ s}^{-1}$ .

516 The self-diffusivity due to the random walk of the particles is notably a function of the  
517 volume fraction (as shown here) and of the shear rate as shown in the literature (Leighton  
518 and Acrivos, 1987). Its value can be interesting to analyze in other situations, like heat  
519 treatment of starch suspension. For example, for a tubular heat exchanger (tube radius  $R_t$ ,  
520 residence time  $t_r$ ), it can reasonably be assumed that the particles follow the fluid flow only  
521 if  $\sqrt{D_{rw} t_r} \ll R_t$ .

522 As explained earlier, the random walk of the particles participates to heat transfer. It  
523 is expected that the apparent conductivity is increased by about  $\phi \rho_p C_{pp} D_{rw}$ . The order of  
524 magnitude of this additional conductivity (about  $10^{-4} \text{ W.m}^{-1}.\text{K}^{-1}$ ) is however low compared  
525 to the ordinary conductivity of water (about  $0.6 \text{ W.m}^{-1}.\text{K}^{-1}$ ). When this kind of heat  
526 transfer is switched off ( $Nu = 0$ ) there is almost no difference ( $< 0.15 \text{ }^\circ\text{C}$ ) in the heating  
527 rate of the internal wall.

528 To identify the part of the friction due to particle random walk, Figure 7b presents the  
529 evolution of the dissipation due to viscous friction between layers of fluid and the dissipa-  
530 tion due to lubrication and drag forces (per unit volume:  $\text{W.m}^{-3}$ ). The total dissipation  
531 is the product of the wall shear stress and the global shear rate. So this figure illustrates  
532 how the shear stress and similarly the mixture apparent viscosity evolve and which part is  
533 due to fluid/fluid interaction and particle/particle/fluid interactions. The fluid/fluid inter-  
534 action first decreases because the liquid viscosity decreases when temperature rises. Then  
535 it increases because the solid volume fraction increases which enhances the distortion of the  
536 fluid streamlines. The particle/particle/fluid interaction (lubrication and drag) is first very  
537 low ( $t < 30 \text{ s}$ ) except at the simulation starting. Initially, due to the random localisation,  
538 some particles are very close to another, this leads to some collisions. Then the lubrica-  
539 tion forces tend these particles to move away from each other. When the volume fraction  
540 becomes significant ( $t > 30 \text{ s}$ ) and the particles moves randomly across the gap the par-  
541 ticle/particle interaction increases significantly the friction, or in other terms the mixture

542 apparent viscosity.

543 At the end of the simulation( $\phi \approx 0.30$ ), the two contributions are of the same order.  
544 This highlights the importance of particle random walk in the rheology evolution of a starch  
545 suspension.

546 As shown previously, swelling begins near the heated wall. In addition to the random  
547 walk, a global particle migration from the zone of highest volume fraction (near the heated  
548 wall) towards zones of lower volume fraction (near the internal wall) is expected.

549 Figure 7c shows the mean displacement in the  $x$ -direction of the granules initially located  
550 in the cell near the heated wall vs time. The phenomenon starts effectively with the swelling  
551 but it remains small: only about  $15 \mu\text{m}$ . Part of this displacement is simply due to the  
552 fact that the centre of the particles is at least one radius from the wall. This is on average  
553  $7 \mu\text{m}$  initially and  $17 \mu\text{m}$  once the granules are swollen. Another part is due to diffusion  
554 in the direction opposite to the volume fraction gradient, but this gradient remains weak  
555 (Fig5c). It would be interesting in further work to relate particle migration to gradients in  
556 osmotic pressure as proposed by Vollebregt et al. (2010) but this is not easy in our case due  
557 to the heterogeneity across the gap. Thus, we can see that the granule migration towards  
558 the internal walls is significant only when the granules begin to swell. The temperature  
559 gradient and velocity gradients, which exist initially, do not lead to any significant diffusion  
560 unless the granules begin to swell.

561 The figure 7d shows the number of quasi contacts defined by the criteria that centre to  
562 centre distance between particles is less than  $1.2 \times (r_i + r_j)$ . Initially when the starch granules  
563 are in an unswollen state the number of quasi-contacts are too low to determine the fabric  
564 tensor. When this number exceeded 100, the fabric deviator was found to be less than 0.1.  
565 At the end of the simulation, the fabric tensor is as follows

$$\Omega_{ij,run1} = \begin{bmatrix} 0.3549 & -0.0009 & -0.0027 \\ -0.0009 & 0.3151 & -0.0025 \\ -0.0027 & -0.0025 & 0.3300 \end{bmatrix} \quad (25)$$

566 and the fabric deviator,  $\Omega_{d,run1} = 0.060$ .

567 One can see very faint anisotropy based on these metrics in the system as the non-  
 568 diagonal elements are very smaller than the unity. For perfect isotropy, all the non-diagonal  
 569 elements are equal to zero.

570 The quadratic mean velocity components of the particles (relative to the fluid) in  $x$ ,  $y$   
 571 and  $z$ -direction at the end of the simulation are respectively 0.097, 0.096 and 0.050 mm/s.  
 572 Particle agitation is anisotropic, this can be explained by the particular direction of shear  
 573 (in the velocity gradient only the term  $\frac{\partial v_y}{\partial x} \neq 0$ ). This finding is coherent with the results of  
 574 Breedveld et al. (1998) which found that the self diffusivity was 1.7 higher in the velocity  
 575 gradient direction than in the other direction. A characteristic distance before direction  
 576 change (like mean free path before the collision) can be obtained by dividing  $2D_{rw}$  by the  
 577 quadratic mean velocity. The order of magnitude is of some microns at the end of the  
 578 simulation.

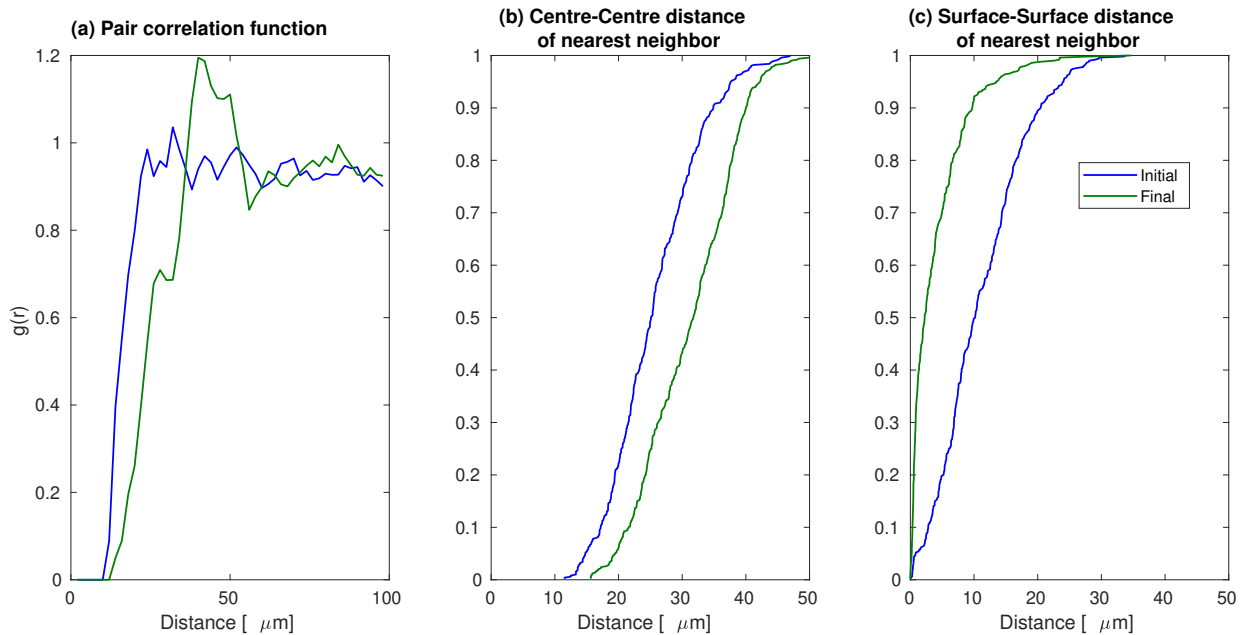


Figure 8: cumulative distributions of distance from neighbors

579 In figure 8a the pair correlation function or otherwise known as radial distribution func-  
 580 tion, which represents the probability of finding another particle at a given distance from

581 the centre of a representative particle. Initially the particles have a low volume fraction  
582 and hence the pair correlation function is fairly flat. At the end of the simulation particle  
583 swelling and increase in volume fractions lead to a peak at around  $45 \mu\text{m}$ .

584 Figure 8b shows the cumulative distribution of the centre to the centre distance to the  
585 nearest neighbour at the beginning and the end of the simulation. The centre to centre  
586 average distance increases from  $25.7 \mu\text{m}$  (initial random positioning) to  $31.2 \mu\text{m}$  at the  
587 end of the simulation. The particles tend to increase their mutual distance, by the swelling  
588 effect and by the lubrication forces. However, the final value is far from the maximal possible  
589 distance (hexagonal arrangement) :  $2^{1/6}(V_{box}/I)^{1/3} = 48 \mu\text{m}$  in our case. It was mentioned  
590 previously that the particles velocity fluctuations are not isotropic. Indeed, alignment in  $y$ -  
591 direction could reduce friction (diminish dissipation) as expected from the minimal entropy  
592 production principle proposed by Prigogine (Prigogine, 1978). From the visual observation  
593 of Figure 4, such trends are not obvious even at the final instant, two zones orientated in  
594 the  $y$ -direction of lower particle density seem to be present. Therefore, to quantify possible  
595 heterogeneity/anisotropy of particles repartition in addition to the fabric tensor discussed  
596 previously, the position of the nearest neighbour of all the particles were analyzed. The  
597 root mean squared average distance to the closest neighbour of each particle in  $x$ ,  $y$  and  
598  $z$  directions at the end of the simulation are respectively  $19.1$ ,  $17.5$  and  $18.8 \mu\text{m}$ . This  
599 indicates a small trend toward alignment in the  $y$ -direction (lower  $y$ -direction distance,  
600 higher  $x$ -direction distance). Isothermal simulations at different shear rates (not presented  
601 here) indicate that the alignment trend increases with the shear rate which could imply shear  
602 thinning. This has to be confirmed by further work. Figure 9c shows the distribution of the  
603 surface to the surface distance to the closest neighbour. The mean distance decreases from  
604  $11.1$  to  $4.0 \mu\text{m}$  as expected since the volume fraction increases from  $2.6$  to  $30 \%$ . For about  
605  $35 \%$  of the particles, the closet neighbour is at less than  $1 \mu\text{m}$  at the end of the simulation.

### 606 3. Conclusion

607 Starch granules swelling inside a heated Couette rheometer is simulated using a CFD-  
608 DEM model with a kinetic model for granule swelling. One can see, as expected that the

609 sample is not uniform across the rheometer gap and the same has been quantified through  
610 various parameters. The velocity profile is not always linear except at the start of the  
611 simulation. The particle temperature is very close to the temperature of the nearby fluid  
612 always. It is also showed that random walk of the particles (self diffusivity) arising due to  
613 the lubrication force is significant. There is a small migration of particles from the outer  
614 walls to the inner cells. The transport of heat by particles is negligible compared to heat  
615 transfer by the fluid.

616 A small anisotropy exists in the direction of shear for the position of the particles in the  
617 system. This study was limited to a volume fraction of about 30 % whereas volume fraction  
618 up to 70 % are common in processing. For higher volume fraction, thermal effects are  
619 still expected to be negligible. However, the particle-particle and particle-fluid interaction  
620 will dominate the fluid-fluid interaction. The lubrication/drag interactions and contact  
621 frictions will gain prominence; thus, warranting a pure DEM approach by neglecting the  
622 fluid phase at  $\phi$  close to  $\phi_{rcp}$ . This model should be enhanced, to account for the shear-  
623 thinning behaviour which is experimentally observed for gelatinized starch suspensions of  
624 intermediate solid volume fraction. Further work could integrate full lubrication terms  
625 along with angular velocity components as well as lubrication forces between the walls and  
626 particles. In conclusion, this work gives insights into the important phenomena that one  
627 needs to consider during thermo-mechanical treatment of starch suspensions.

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