

# A Dissipative Particle Dynamics Model for Thixotropic Materials Exhibiting Pseudo-yield Stress Behaviour<sup>☆</sup>

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## Abstract

Many materials (e.g., gels, colloids, concentrated cohesive sediments, etc.) exhibit a stable solid form at rest, and liquify once subjected to an applied stress exceeding a critical value – a yield-stress behaviour. This can be qualitatively explained by the forming and destruction of the fluid microstructure [1], and it may be modelled as a thixotropic and yield stress material. In this paper, we propose a mesoscopic model which is able to mimic a thixotropic and yield stress behaviour using a particle-based technique known as dissipative particle dynamics (DPD). The DPD technique satisfies conservation of mass and momentum and it has been applied successfully for a number of problems involving complex-structure fluids, such as polymer solutions, suspensions of rigid particles, droplets, biological fluids, etc. In this work, an indirect linkage dissipative particle model (ILDPM) is proposed based on qualitative microstructural physics, which results in a non-Newtonian fluid with observed yield stress and thixotropic properties. The model comprises of two types, or species, of DPD particles – with only repulsive conservative force between the same species, and with repulsive force at short range and attractive force at long range between different species. Numerical results show that the proposed DPD fluid can represent some observed complex behaviours, such as yield stress and thixotropic

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## 1. Introduction

Many engineering flow processes involve complex structure liquids, for examples, foodstuffs, polymers, cosmetic products, crude oil, cohesive sediment mixtures, etc. Such complex liquids hardly flow if the applied load is lower than  
5 a certain value, but they are liquefied and flow easily at loading higher than this value. This critical stress threshold associated with this loading is called the “yield stress”, and there are experimental evidences that these fluids do flow in a very viscous manner at a stress level lower than the yield stress, thus supporting modelling of the true fluid-to-solid transition by a very viscous transition at low  
10 strain rates. Moreover, the fluid at this low stress level is thixotropic as well; we refer the reader to an excellent recent review by Bonn [2] on this topic. The current view of this research area, which we share, of the yield stress as a very viscous transition remains an attractive and useful engineering idea, and that it is a result of a network microstructure generated by inter-particle interactions,  
15 which may rupture into smaller clusters by large enough applied stresses and then restores at a low stress level. The network needs time to build up and to disintegrate, and the rheology of the mixture therefore has a time scale; this results in a macroscopic thixotropic and yield stress behaviour, which has been considered one of the most complex phenomena in rheology [1]. For example,  
20 in cohesive sediment mixtures, which provide the motivation of this work, clay particles flocculate and then break up under flow conditions. At a low concentration some clay particles may form clusters and once the concentration reaches a threshold value, those clusters link up and form a network, leading to a yield stress behaviour.

25 In continuum mechanics, such yield-stress materials are modelled by a rel-

evant constitutive equation (the simplest model would be the Bingham model [3] or one of its variants). These models, representing a true solid-to-fluid transition behaviour without thixotropic complications, are transformed into sets of algebraic equations by means of discretisation and their flows are then solved

30 numerically by traditional numerical methods, such as finite volume (FVM), finite element (FEM) or boundary element (BEM) methods. Because of the solid-to-fluid transition at small applied stresses, a “numerical singularity”, associated with the indeterminate nature of the stress, occurs at low strain rates when dealing with these types of constitutive equations. To mitigate this numerical

35 difficulty, Papanastasiou [4] proposed a modified version of Bingham model, which is in fact a generalised Newtonian model with high viscosity at low enough strain rate; thus replacing the transition to solid-like behaviour by a very viscous behaviour at low strain rates. Mathematically, Papanastasiou’s model may be regarded as a regularisation of the Bingham model [5]. Papanastasiou’s model

40 has been widely used in many engineering applications [6]. However, it has been pointed out that, in practice, many substances, such as food products, crude oils, cohesive sediment mixtures display flow characteristics that may not be described by the Bingham models, or its viscous (regularised) approximations (we refer to these models as Bingham-type models). This is primarily because their

45 apparent viscosities do not only depend on applied shear stresses, but also the duration for which the fluids have been subjected to the flow processes, as well as their previous kinematic history. Those fluids may be classified as thixotropic fluids. To describe the behaviour of thixotropic fluids, another approach which based on structural kinematics theory has been proposed (for instance, [1]; [7];

50 [8]; [9]). In these models, the time dependent rheological behaviour is quantified by a non-dimensional structural parameter  $\lambda$ . The scalar  $\lambda$  indicates the integrity of a particulate network, i.e.,  $\lambda = 0$ : no network and  $\lambda = \lambda_0$ : a network is fully formed ( $\lambda_0$  is the maximum value of  $\lambda$ , usually set to unity). The viscosity and the stresses are thus functionals of  $\lambda(t)$ ;  $t$  is the time. Both the

55 Bingham-type (now depend on  $\lambda(t)$ ) and a relevant thixotropic model (for  $\lambda(t)$ ) are simultaneously solved together with the Navier-Stokes equations to obtain

a specific solution, subjected to a relevant set of boundary/initial conditions.

In the last two decades the DPD method [10] has been developed as an alternative and promising mesoscopic approach for modelling complex fluids. Different from spatial macroscopic schemes, the DPD method is originally based on a coarse-grained representation of the fluid and everything in it. DPD method often looks similar to Molecular Dynamics (MD), with built-in thermal equilibrium via a fluctuation-dissipation theorem [11]. However, DPD particles interact through soft potentials and thus the simulation can be carried out on length and time scales far beyond those associated with MD. It has been shown that mean quantities (e.g., density and linear momentum) formed from the microstate of a DPD system (consisting of DPD particles positions and velocities) satisfy mass and momentum conservations [12]. Therefore, the method may be regarded as a particle-based method for solving complex fluid problems [13]. From this point of view, a single DPD particle may be regarded as similar to a smoothed particle hydrodynamics (SPH) particle (in its implementation), in which a certain volume of fluid is represented as a Lagrangian particle in the SPH method. Especially, in one of DPD variants called the smoothed DPD method (sDPD) [14], the interaction forces have a specific form which comprised from the SPH discretisation of Navier-Stokes equations. Many applications of DPD method or its variants in the simulations of complex fluids have been reported, e.g., sphere colloidal suspensions ([15]; [16]; [17]; [18]; [19]; [20]), colloidal suspensions of spheres, rods, and disks [21], viscoelastic fluid [22], ferromagnetic colloidal suspension [23], magnetic colloidal dispersions [24], soft matter and polymeric applications [25], [26], lipid bilayer [27], flows of DNA suspensions [28], polymer chains [29], red blood cell modelling [30], [31]; this list is not meant to be exhaustive.

The continuum approaches including Bingham-type models and/or structural kinetics models may be classified as top-down approach, which starts with the macro behaviour of the systems. In this paper, we report a DPD method, as a bottom-up approach, built on a microstructure interaction model, which yields the desired observed macro properties. An indirect linkage model for

dissipative particles through a micro interaction forces is proposed to mimic the formation of particulate network in a natural way. The continual forming,  
90 breaking and recovering of the microstructures results in yield stress together with the desired thixotropic behaviours.

The number density, fluid velocities and stresses in our DPD model are calculated by ensemble averaging the instant data (e.g., particle configuration, particle velocities, etc.), or by time-averaging over a number of time steps (with  
95 the assumption of the ergodic theorem). The stress-strain rate relation of the fluid is studied for Couette and Poiseuille flows. It is found that the numerical results can fit well to Papanastasiou's model at steady state. The DPD model also replicates thixotropic behaviour in a natural way, in unsteady flows, through the continual forming and rupturing of DPD microstructure network  
100 under applied stresses.

The remainder of the paper is organised in the following manner. An overview of the standard DPD fluid is provided in Section 2. Then, Section 3 gives a brief review of Bingham model, its variants and some prominent structural kinetics models. The proposed indirect linkage dissipative model is then  
105 presented in Section 4. Subsequently, in Section 5 and 6, a material preparing process is described, akin to an experimental process, and numerical results are discussed. Section 7 gives some concluding remarks.

## **2. Dissipative Particle Dynamics (DPD)**

### *2.1. DPD fluid*

DPD is a particle-based simulation method that satisfies mass and momentum conservations. It is a promising method, originally devised for simulations at mesoscopic length and time scale for material with a complex microstructure. The DPD method produces field results that satisfy the Navier-Stokes equations, in the same manner of other standard continuum methods (e.g., FEM, FVM, BEM), and therefore it can be regarded as a particle-based discretisation of the Navier-Stokes equations in mesoscale where thermal fluctuations are accounted

for. In DPD method, the fluid and all its component phases (if any) are defined by the assemblage of  $N$  particles, each of mass  $m_i, i = \{1, 2, \dots, N\}$  located at position  $\mathbf{r}_i$ , with velocity  $\mathbf{v}_i$ . With the assumption of identical mass, without much loss of generality,  $m_i = m$ , the DPD particles interact with each other and undergo their Langevin motions [32]:

$$\frac{d\mathbf{r}_i}{dt} = \mathbf{v}_i, \quad (1)$$

$$m \frac{d\mathbf{v}_i}{dt} = \mathbf{f}_i + \mathbf{f}_e, \quad (2)$$

where  $\mathbf{f}_e$  is external forces on particle  $i$  (e.g., gravity force),  $\mathbf{f}_i = \sum_{j \neq i} \mathbf{f}_{ij}$  ( $\mathbf{f}_{ii} = 0$ ) the interaction force on particle  $i$  by all other particles  $j$ , pairwise additive. It is noted that the sum runs over all other particles within a certain cut-off radius  $r_c$ . The interaction force  $\mathbf{f}_{ij}$  consists of three parts, a conservative force,  $\mathbf{f}_{ij}^C$ , a dissipative force,  $\mathbf{f}_{ij}^D$ , and a random force,  $\mathbf{f}_{ij}^R$ :

$$\mathbf{f}_{ij} = \mathbf{f}_{ij}^C + \mathbf{f}_{ij}^D + \mathbf{f}_{ij}^R. \quad (3)$$

110 Expressions of interaction forces are listed in Table 1 in which  $a_{ij}$  is conservative force strength;  $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ ;  $r_{ij} = |\mathbf{r}_{ij}|$ ;  $\hat{\mathbf{r}}_{ij} = \mathbf{r}_{ij}/|\mathbf{r}_{ij}|$ ;  $w^C, w^D, w^R$  weight functions of conservative, dissipative and random forces, respectively;  $\mathbf{v}_{ij} = \mathbf{v}_i - \mathbf{v}_j$ ;  $\gamma$  a coefficient related to the system viscosity;  $\xi_{ij}$  a Gaussian variable with zero mean and variance equal to  $\delta t^{-1}$ , where  $\delta t$  is the time step, and  $\sigma$  is the magnitude of the random force.

Table 1: List of interaction forces and their formulas. Note that the balance between dissipative and random forces must obey the fluctuation-dissipation theorems  $\sigma = \sqrt{2\gamma k_B T}$  and  $w^D = (w^R)^2$  [32].

$\mathbf{f}_{ij}$	Weight function	Form
$\mathbf{f}_{ij}^C$	$w^C(r_{ij}) = 1 - \frac{r_{ij}}{r_c}$	$a_{ij} w^C \hat{\mathbf{r}}_{ij}$
$\mathbf{f}_{ij}^D$	$w^D(r_{ij}) = \left(1 - \frac{r_{ij}}{r_c}\right)^k$	$-\gamma w^D (\hat{\mathbf{r}}_{ij} \cdot \mathbf{v}_{ij}) \hat{\mathbf{r}}_{ij}$
$\mathbf{f}_{ij}^R$	$w^R(r_{ij}) = \sqrt{\left(1 - \frac{r_{ij}}{r_c}\right)^k}$	$\sigma w^R \zeta_{ij} \hat{\mathbf{r}}_{ij}$

From the system state, one can define the local fluid density:

$$\rho(\mathbf{r}, t) = \left\langle \sum_i m \delta(\mathbf{r} - \mathbf{r}_i) \right\rangle = md(\mathbf{r}, t), \quad (4)$$

where the symbol  $\langle \rangle$  indicates an ensemble average (which can be equated to a suitable time average over some iteration steps by the ergodic theorem), and  $d(\mathbf{r}, t)$  is the number density. The local linear momentum is calculated by

$$\rho(\mathbf{r}, t) \mathbf{u}(\mathbf{r}, t) = \left\langle \sum_j m \mathbf{v}_j \delta(\mathbf{r} - \mathbf{r}_j) \right\rangle. \quad (5)$$

These quantities have been shown to satisfy conservation laws ([12]; [32]):

$$\frac{\partial}{\partial t} \rho(\mathbf{r}, t) + \nabla \cdot (\rho(\mathbf{r}, t) \mathbf{u}(\mathbf{r}, t)) = 0, \quad \nabla = \partial / \partial \mathbf{r}, \quad (6)$$

and

$$\frac{\partial}{\partial t} (\rho \mathbf{u}) + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) = \nabla \cdot \boldsymbol{\sigma}. \quad (7)$$

Thus, DPD may be regarded as a particle-based method for solving continuum flow problems Eqs.(6)-(7). The macroscopic properties including fluid density  $\rho$ , stress  $\boldsymbol{\sigma}$  and consequentially viscosity  $\mu$  are calculated by appropriate averages over all sampled data in each bin. The relevant stress tensor  $\boldsymbol{\sigma}$  is calculated by Irving-Kirkwood expression ([33]). In one dimensional shear flow, the fluid viscosity can be found from the shear stress and the shear rate,  $\mu = S_{xz} / \dot{\gamma}$ . In this particle-based point of view, a DPD particle may be thought of as a volume of fluid with a built-in behaviour (e.g., a non-Newtonian viscous compressible fluid volume), rather than a cluster of fluid molecules. Yield stress and thixotropic behaviours may also be captured by this particle-based method. The nonlinear relationship between stress and strain rate needs not to be specified a-priori, but can be obtained after post-processing step.

### 3. Brief review of yield stress fluid models

#### 3.1. Bingham models

130 In continuum mechanics, Bingham plastic is a non-Newtonian fluid behaviour characterised by the existence of a threshold stress called the apparent yield stress ( $S_0$ ), which must be exceeded for the fluid to deform (shear) or flow. Such behaviour can be modelled by a simple Bingham rheological constitutive model ([3], [5]):

$$\mathbf{S} = 2 \left\{ \mu + \frac{S_0}{\mathbb{III}_D^{1/2}} \right\} \mathbf{D}, \quad |\mathbf{S}| > S_0. \quad (8)$$

In the above,  $\mathbf{S}$  is the extra stress;  $\mathbb{III}_D^{1/2}$  denoted the generalised strain rate;  $\mathbb{III}_D$  the second invariant of strain rate tensor  $\mathbf{D}$ ,

$$\mathbf{D} = \frac{1}{2} (\nabla \mathbf{u} + \nabla \mathbf{u}^T), \quad (9)$$

where  $\nabla \mathbf{u}^T$  the velocity gradient tensor (by definition).  $|\mathbf{S}|$  the magnitude of the extra stress tensor and is defined as

$$|\mathbf{S}| = \sqrt{\frac{1}{2} [\mathbf{S} : \mathbf{S}]}. \quad (10)$$

Below the stress threshold, the stress is indeterminate, and while this is an engineering simplification, this discontinuity in behaviour is a major problem in numerical implementation. Papanastasiou [4] proposed a modified version of Bingham model to overcome this numerical difficulty by introducing an exponential regularisation for the stress and strain rate relation in Eq.(8); more details of this regularisation can be found in [5]. Here the stress is written as

$$\mathbf{S} = 2 \left\{ \mu + \frac{S_0 \left[ 1 - e^{(-n\mathbb{III}_D^{1/2})} \right]}{\mathbb{III}_D^{1/2}} \right\} \mathbf{D}, \quad (11)$$



135 where  $n$  is a stress growth parameter which determines how fast the devel-  
 opment of stress. It is noted that  $n$  does not have physics meaning, although  
 it has the dimension of time, and it is set to a large enough value that does  
 not cause numerical instability. In one dimensional problems,  $\mathbb{III}_D^{1/2}$  becomes the  
 shear rate  $\dot{\gamma}$ . At high shear rate where the second term in the curly brackets  
 140 approaches zero, this model asymptotes to  $2\mu\mathbf{D}$ , a Newtonian fluid with viscos-  
 ity  $\mu$ . At low strain rate, the stress asymptotes to  $2(\mu + nS_0)\mathbf{D}$ . Consequently,  
 for large  $n$ , this fluid model is close to Bingham model, in the sense that the  
 deformation at low strain rate is small because of the large viscosity, but the  
 true solid-to-fluid transition (as embodied in the Bingham model) is lost, and  
 145 is now replaced by a very viscous transition.

### 3.2. Structural kinetics model

#### Qualitative concept of particulate networks

A well-dispersed system, such as a dense cohesive sediment suspension, dis-  
 plays rheological characteristics which cannot be simply described by mathe-  
 150 matical expression of the form of Eqs.(8) or (11). In fact their viscosity and  
 yield stress are not only functions of the applied shear rate ( $\dot{\gamma}$ ), but also their  
 previous shear history.

Similar to Bingham-type fluids, this type of non-Newtonian fluids exhibit a  
 yield stress  $S_0$ : it will flow in a whole like a solid body when externally applied  
 155 stresses are less than  $S_0$ . Naturally, when the magnitude of the external stress  
 exceeds  $S_0$ , the fluid may exhibit shear-thinning effect. Such a fluid at rest  
 consists of small attractive particles which form a cluster to produce a structure  
 network of sufficient connectivity - this structure can resist any applied stress  
 less than  $S_0$  without deformation. In contrast, if the applied stress magnitude  
 160 is greater than  $S_0$ , the structure network breaks down and thus results in a  
 decreasing resistance to deformation and flow. The reversed may also happen,  
 i.e., the fluid may restore some of its network connectivity, and the yield stress  
 value of the recovered state may be equal or lower than that of the initial state.

#### Thixotropy model

To quantify the structure dependent rheological behaviour, a non-dimensional structural parameter  $\lambda$  has been used to characterise any fluid parameter, for example viscosity or yield stress may be expressed as functionals of this parameter, i.e.,  $\mu(\lambda(t))$ , or  $S_0(\lambda(t))$  [9]. This parameter is a measurement of the degree of structure in the dispersed system, having a value in the range 0 (fully broken) to  $\lambda_0$  (fully structured), commonly assumed to be unity. In a one-dimensional problem, a simple model of  $\lambda$ 's evolution may be described by a first-order rate equation:

$$\frac{d\lambda}{dt} = \frac{\partial\lambda}{\partial t} + u_x \frac{\partial\lambda}{\partial x} = a(\lambda_0 - \lambda) - b\dot{\gamma}\lambda, \quad (12)$$

where  $u_x$  the  $x$ -component of the fluid velocity vector (the only relevant component in this 1D problem);  $a$  and  $b$  two coefficients determined from experiments. The first term on the right side of (12) represents the network coalescence with the associated constant  $a$ ; and in the second term, the constant  $b$  represents the disintegration of the particulate network due to the flows. The equilibrium value of the structural parameter  $\lambda_e$  can be obtained by equating the right side of (12) to zero:

$$\lambda_e = \frac{a\lambda_0}{a + b\dot{\gamma}} = \frac{\lambda_0}{1 + \beta\dot{\gamma}}, \quad (13)$$

165 where  $\beta = b/a$ .

A rheological equation for a thixotropic yield stress fluid has been proposed by [9] (in 1D)

$$S_{xz} = (\lambda_0 + \lambda - \lambda_e)S_0 + (\mu_\infty + c\lambda)\dot{\gamma}. \quad (14)$$

At equilibrium, when the rate of disintegration equals the rate of recovery, the equilibrium flow (EF) curve is characterised by evaluating Eq.(14) at equilibrium point  $\lambda = \lambda_e$  and using the relation (13) :

$$S_{xz}^e = \lambda_0 S_0 + \mu_\infty \dot{\gamma} + \frac{c\lambda_0 \dot{\gamma}}{1 + \beta\dot{\gamma}}. \quad (15)$$

Thixotropic fluids may exhibit a family of stress/strain rate curves called constant structure (CS) curves [34], each one corresponding to a (constant) value

of its structure parameter. The EF curve and CS curves intersect at points on which the structure is equilibrium (Figure 1). Consider a CS curve which cut across the EF curve at equilibrium point E. At points above the EF curve (e.g., point A), if the shear rate is kept constant, the structure will break resulting in a reduction in the shear stress  $S^A$  to equilibrium value  $S_e^A$ . In contrast, at points below the EF curve, such as point B, the structure will continually build up and the shear stress  $S^B$  would raise until the system reaches the equilibrium state ( $S = S_e^B$ ). When the EF curve is known and can be described by Eq.(15), a simple procedure allows the determination of the CS curves (either numerically or experimentally). For example, consider an  $i$ th data point of the EF curve, one has a CS curve which have a single value of structural parameter  $\lambda_i$ . The value of  $\lambda_i$  is the same of  $\lambda_E$  at the crossover point of this CS and the EF curves. Substitution of this value into Eq.14 gives the CS curve of the  $i$ th point,

$$S = (\lambda_E + \lambda_e \beta \dot{\gamma}) S_0 + (\mu_\infty + c \lambda_E) \dot{\gamma}. \quad (16)$$

#### 4. Proposed indirect linkage dissipative particle (ILDLP) model

Inspired from this qualitative concept of a particulate network, we propose the following DPD model consisting of *two types DPD particles*, represented by the ensemble of  $N = N^{(a)} + N^{(b)}$  particles, each of the set of  $N^{(s)}$  particles  
170 represents number of particles in each species  $s = a$  or  $b$  of the fluid. Among  $DPD^{(a)}$  and  $DPD^{(b)}$  particles, there are three types of interactions:  $DPD^{(a)}$  to  $DPD^{(a)}$  or (a,a),  $DPD^{(b)}$  to  $DPD^{(b)}$  or (b,b), and  $DPD^{(a)}$  to  $DPD^{(b)}$  or (a,b). Here  $\mathbf{f}_{ij}^{(a,b)}$  is the pairwise additive interparticle force by particle  $j(b) \in N^{(b)}$  on particle  $i(a) \in N^{(a)}$  (the notation  $i(a)$  reads particle  $i$  in species  $a$ ). To  
175 avoid a phase separation between (a) and (b), we propose an indirect linkage by introducing cohesive forces between  $DPD^{(a)}$  and  $DPD^{(b)}$  particles. The indirect linkage dissipative particle model (ILDLP) thus comprises from (a, a) or (b, b) interactions with repulsive force (the “standard” model for DPD fluid), and long-range attractive plus short-range repulsive ((a, b) or (b, a)) - the network of

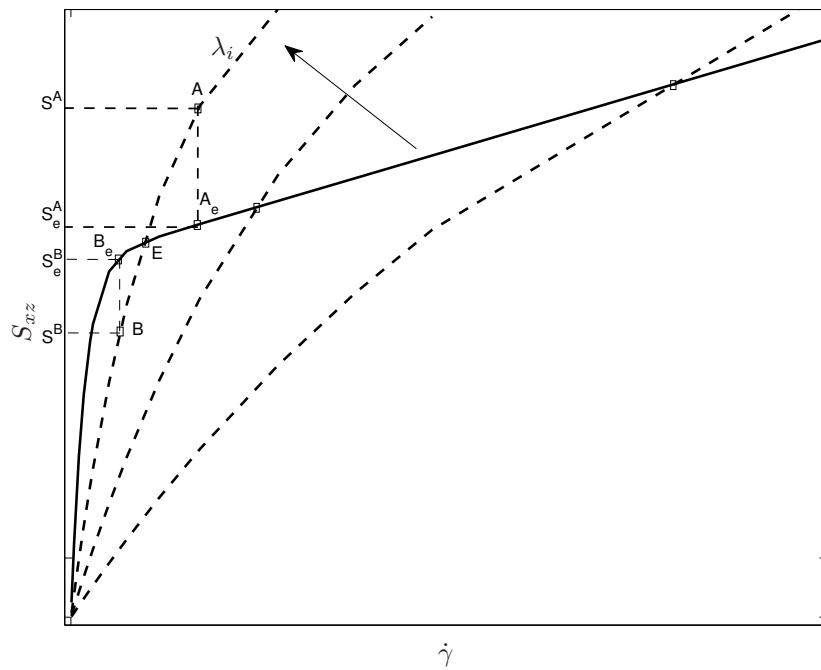


Figure 1: Equilibrium flow curve (solid line) and constant structure curves (dash lines). The arrow indicates that the structural levels increase (not to scale).

180  $DPD^{(b)}$  can be encouraged to form by adding a long-range attractive component  
to the conservative forces, which is taken from [35]. It is important to note that  
 $DPD^{(b)}$  particles attract  $DPD^{(a)}$  directly, and  $DPD^{(b)}$  indirectly.  $DPD^{(b)}$   
particles form an indirect network through layers of  $DPD^{(a)}$  particles (Figure  
2). In this model, we view  $DPD^{(a)}$  and  $DPD^{(b)}$  as a solvent and suspended  
185 phase, respectively.

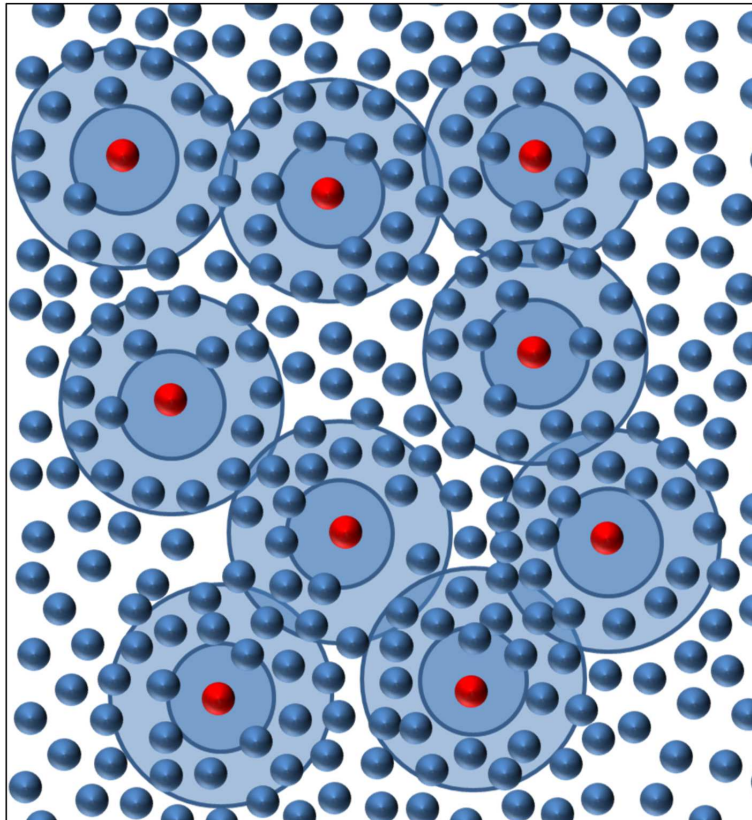


Figure 2: Indirect linkage DPD network: Microstructure of the proposed model-two species of DPD ( $DPD^{(a)}$  (blue colour)) and  $DPD^{(b)}$  (red colour)). A  $DPD^{(b)}$  attracts some  $DPD^{(a)}$  within its long range (circles of larger radius) and repulses other  $DPD^{(b)}$  and  $DPD^{(a)}$  in short range (circles of smaller radius).

Assuming that each DPD particle of a species  $s$  has a mass of  $m_i^{(s)}$  located at position  $\mathbf{r}_i^{(s)}$ , with velocity  $\mathbf{v}_i^{(s)}$ . The DPD particles interact with each other in their Newton's second law motions:

$DPD^{(a)}$ :

$$\frac{d\mathbf{r}_i^{(a)}}{dt} = \mathbf{v}_i^{(a)}, \quad m_i^{(a)} \frac{d\mathbf{v}_i^{(a)}}{dt} = \sum_{j \neq i} \mathbf{f}_{ij}^{(a,b)} + \sum_{j \neq i} \mathbf{f}_{ij}^{(a,a)}. \quad (17)$$

$DPD^{(b)}$ :

$$\frac{d\mathbf{r}_i^{(b)}}{dt} = \mathbf{v}_i^{(b)}, \quad m_i^{(b)} \frac{d\mathbf{v}_i^{(b)}}{dt} = \sum_{j \neq i} \mathbf{f}_{ij}^{(b,a)} + \sum_{j \neq i} \mathbf{f}_{ij}^{(b,b)}. \quad (18)$$

Similar to the conventional DPD,  $\mathbf{f}_{ij}$  consists of three parts, a conservative force  $\mathbf{f}_{ij}^C$ , a dissipative force  $\mathbf{f}_{ij}^D$ , and a random force  $\mathbf{f}_{ij}^R$ . In Eqs.(17)-(18), the sum runs over all other particles except  $i$  (note, by definition  $\mathbf{f}_{ii}^{(s,s)} = \mathbf{0}$ ). These forces are built-in with a certain cut-off radius  $r_c$ ; outside this cut-off radius, the interactions are zero. Here one may allow the cut-off radius to be different for different type of forces. The dissipative and random forces are taken the same forms listed in Table 1. Conservative force  $\mathbf{f}_{ij}^C$  of  $DPD^{(a)}$  and  $DPD^{(b)}$  interaction is calculated according to the model proposed by [35]

$$\mathbf{f}_{ij}^C = -a_{ij}(Aw^{Cr}(r, r_r) - Bw^{Ca}(r, r_a))\hat{\mathbf{r}}_{ij}, \quad (19)$$

where A and B are coefficient of  $w^{Cr}(r, r_r)$  and  $w^{Ca}(r, r_a)$ , respectively.  $r_r$  is cut-off radius of repulsive component and  $r_a$  is that of attractive component and

$$w^{Cr} = \begin{cases} 18 \left( \frac{A}{r_r^3} - \frac{B}{r_a^3} \right) r^2 - 12 \left( \frac{A}{r_r^2} - \frac{B}{r_a^2} \right) r, & r < \frac{r_r}{2} & (a) \\ -6 \left( \frac{A}{r_r^3} + \frac{B}{r_a^3} \right) r^2 + 12 \left( \frac{A}{r_r^2} + \frac{B}{r_a^2} \right) r - 6 \frac{A}{r_r}, & \frac{r_r}{2} \leq r < \frac{r_a}{2} & (b) \\ -6 \left( \frac{A}{r_r^3} - \frac{B}{r_a^3} \right) r^2 + 12 \left( \frac{A}{r_r^2} - \frac{B}{r_a^2} \right) r - 6 \left( \frac{A}{r_r} - \frac{B}{r_a} \right), & \frac{r_a}{2} \leq r < r_0 & (c) \\ 0, & r > r_0 & (d) \end{cases} \quad (20)$$

$$w^{Ca} = \begin{cases} 0, & r < r_0 & (a) \\ -6 \left( \frac{A}{r_r^3} - \frac{B}{r_a^3} \right) r^2 + 12 \left( \frac{A}{r_r^2} - \frac{B}{r_a^2} \right) r - 6 \left( \frac{A}{r_r} - \frac{B}{r_a} \right), & r_0 \leq r < r_r & (b) \\ 6 \frac{B}{r_a^3} r^2 - 12 \frac{B}{r_a^2} r + 6 \frac{B}{r_a}, & r_r \leq r < r_a & (c) \end{cases} \quad (21)$$

where value of  $r_0$  is the solution of quadratic equation (20(c)).

The conservative, dissipative and random forces are taken as follows

$$\mathbf{f}_{ij}^{C(a,a)} = \mathbf{f}_{ij}^{C(b,b)} = \mathbf{f}_{ij}^{Cr}, \quad (22)$$

$$\mathbf{f}_{ij}^{C(a,b)} = \mathbf{f}_{ij}^{C(b,a)} = \mathbf{f}_{ij}^{Cr} + \mathbf{f}_{ij}^{Ca}, \quad (23)$$

$$\mathbf{f}_{ij}^{D(a,a)} = \mathbf{f}_{ij}^{D(b,b)} = \mathbf{f}_{ij}^{D(a,b)} = \mathbf{f}_{ij}^D, \quad (24)$$

$$\mathbf{f}_{ij}^{R(a,a)} = \mathbf{f}_{ij}^{R(b,b)} = \mathbf{f}_{ij}^{R(a,b)} = \mathbf{f}_{ij}^R. \quad (25)$$

With the same mass, i.e.,  $m_i^{(a)} = m_i^{(b)} = m$ , Eqs.(17)-(18) can be rewritten as

$$m \frac{d\mathbf{v}_i}{dt} = \begin{cases} \sum_{j \neq i} (\mathbf{f}_{ij}^{Cr} + \mathbf{f}_{ij}^D + \mathbf{f}_{ij}^R), & i, j \in N^{(a)} \text{ or } i, j \in N^{(b)} \\ \sum_{j \neq i} (\mathbf{f}_{ij}^{Cr} + \mathbf{f}_{ij}^{Ca} + \mathbf{f}_{ij}^D + \mathbf{f}_{ij}^R), & i \in N^{(a)}, j \in N^{(b)} \text{ or } i \in N^{(b)}, j \in N^{(a)} \end{cases} \quad (26)$$

where  $\mathbf{f}_{ij}^{Ca} = a_{ij} B w^{Ca} \hat{\mathbf{r}}_{ij}$  and  $\mathbf{f}_{ij}^{Cr} = -a_{ij} A w^{Cr} \hat{\mathbf{r}}_{ij}$ .

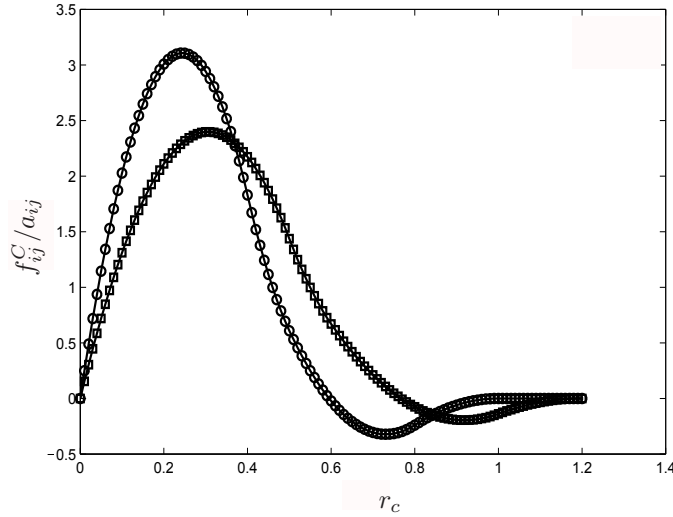


Figure 3: Conservative forces with short range repulsion and long range attraction ( $-\circ$ ,  $r_a = 1.0$ ) and ( $-\square$ ,  $r_a = 1.2$ ).

It can be seen from Figure 3 that the conservative force between two different

200 species particles is repulsive when their separation distance is less than a radius  
 value of  $r_0$ , e.g., 0.5952, for  $A = 2, B = 1, r_r = 1.0, r_a = 1.2$ ; and when their  
 separation distance is between 0.5952 and 1, this force describes a long range  
 attraction (negative). If purely repulsive conservative forces (for example, when  
 setting  $B = 0$  in Eq.(19), for  $DPD^{(b)}$ ), are applied for both type particles, the  
 205 resulting DPD fluid is thus simply Newtonian. When the attractive component  
 in conservative forces of  $DPD^{(b)}$  are turned on, a structure network is formed  
 resulting in high DPD fluid's resistance to applied stresses. The advantages of  
 indirectly linking between  $DPD^{(b)}$  include (i) simple and straight forward to  
 implement, (ii) uniform distribution for  $DPD^{(a)}$  and  $DPD^{(b)}$  (without phase  
 210 separation), and (iii) creating a DPD structure network which results in yield  
 stress and thixotropic behaviour. In next sections, numerical experiments are  
 carried out for this proposed model.

## 5. Material preparation

### 5.1. Pre-processing

215 A pre-processing program is used to generate a system which consists of  
 $N$  particles with masses  $m$  characterised by the positions  $\mathbf{x}_i, i = 1, N$ . There  
 are three types of particles in this list: wall,  $DPD^{(a)}$  and  $DPD^{(b)}$  particles.  
 Initially (Figure 4a), the box is filled with  $DPD^{(b)}$  particles in the bottom and  
 with  $DPD^{(a)}$  on the top. This initial distribution of these particles does not  
 220 satisfy the thermodynamic equilibrium state, and a mixing procedure is thus  
 applied. At the beginning of this procedure, the particles are allowed to move  
 freely until a thermodynamic equilibrium state is reached and then a body force  
 $\mathbf{g} = (0.2, 0, 0)$  is applied for a hundred thousand time steps to mix  $DPD^{(a)}$   
 and  $DPD^{(b)}$ . Figure 4b shows that after mixing, the conservative interactions  
 225 produced a uniform  $DPD^{(a)}$  and  $DPD^{(b)}$  dispersion. The particle configuration  
 is written in a data file which is then read by the DPD solver program.



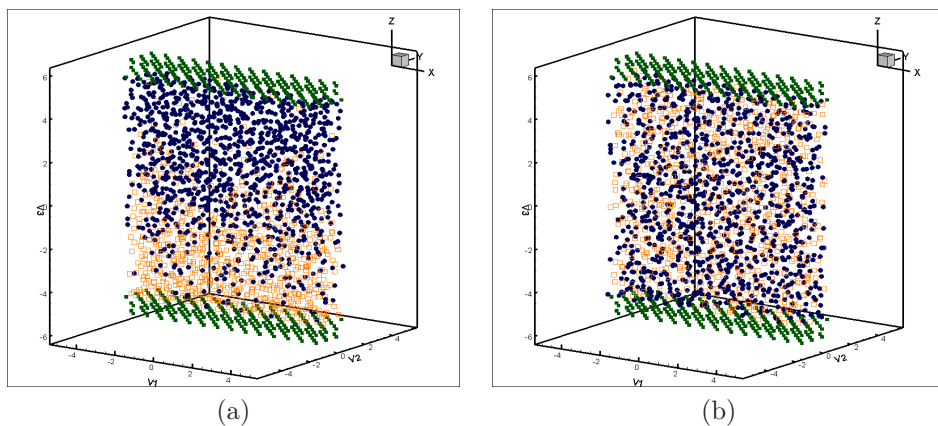


Figure 4: A thick slice along the center line ( $-0.5 \leq y \leq 0.5$ ): (a) Initial configuration of  $DPD^{(a)}$  ( $\bullet$ ) and  $DPD^{(b)}$  ( $\square$ ) - (b) Uniform distribution of two species of DPD, which are obtained over the period of 100000 time steps.

## 5.2. ILDP micro-networks

### 5.2.1. No flow

Before testing, all mixtures are set to a test kinetic energy  $k_B T = 1$  and then  
 230 let it run in no-flow condition for 40000 time steps, each of 0.01, to guarantee  
 that the microstructure network is fully built up. To track the formation of  
 the network, one may choose an  $i$ th particle of  $b$ -species and plot its position  
 $|r| = \sqrt{x^2(i) + y^2(i) + z^2(i)}$ . In this test, the dispersed system is allowed to  
 reach equilibrium and the coefficient of attractive forces  $\mathbf{f}^{Ca}$  are set to zero  
 235 for the first 10000 time steps. It is then turned on to activate any particulate  
 network. The value of  $|r|$  and the trajectory of  $i$ th particle are plotted in Figure  
 5a and 5b, respectively. It is observed that the fluctuation of  $i$ th particles is  
 reduced dramatically when the particulate network starts to form and the  $i$ th  
 particle is trapped in a cage formed by its neighbouring particles, as expected.

### 240 5.2.2. Shear flow testing

In this section, the dependence of structure to the applied shear rate is  
 investigated. The applied shear rate is a step function, for a time interval of  
 8000 time steps, each of 0.01. Initially, the DPD system is allowed to reach

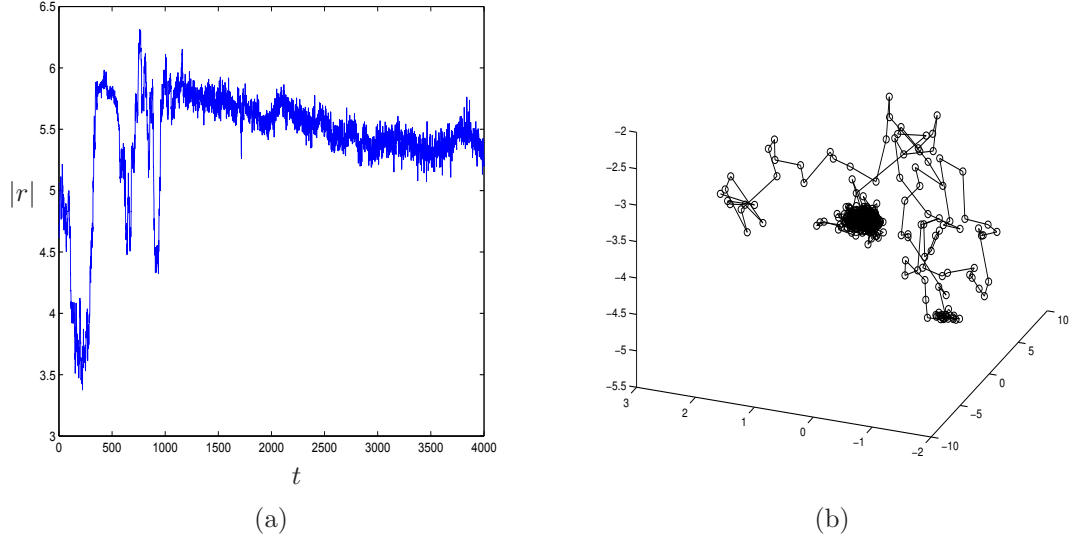


Figure 5: Structure forming: (a) The position  $|r|$  and (b) the trajectory of an  $i$ th  $DPD^{(b)}$  species particle, before and after the microstructure network built up.

equilibrium, the structure network is formed as displayed in Figure 6(a). Then,  
 245 a shear rate  $\dot{\gamma} = 1.0$  is applied; Figure 6(b) shows that the structure is totally  
 destroyed. In a general flow, the level of integrity of the structure network  
 depends on the local shear rate. After around 8000 time steps, the shear rate  
 is returned to zeros, and it is observed that the structure network is restored  
 (Figure 6(c)). Here, in order to have clearer configuration of the microstructure  
 250 for visualisation purpose a relative large cut-off radius for attractive forces is  
 employed ( $r_a = 1.33$ ).

## 6. Numerical experiments and discussion

The ILDP fluid are tested in Couette and Poiseuille flows. The simulations  
 are carried out on two domains, of  $10 \times 10 \times 10$  and  $20 \times 10 \times 20$  for Couette flow,  
 255 and  $10 \times 10 \times 10$  for Poiseuille flow; the DPD parameters are listed in Table 2.

For the conservative interaction of  $DPD^{(a)}$  and  $DPD^{(b)}$ , we fix the repulsive  
 and attractive coefficients at  $A = 2$  and  $B = 1$ , respectively (Figure 3). For

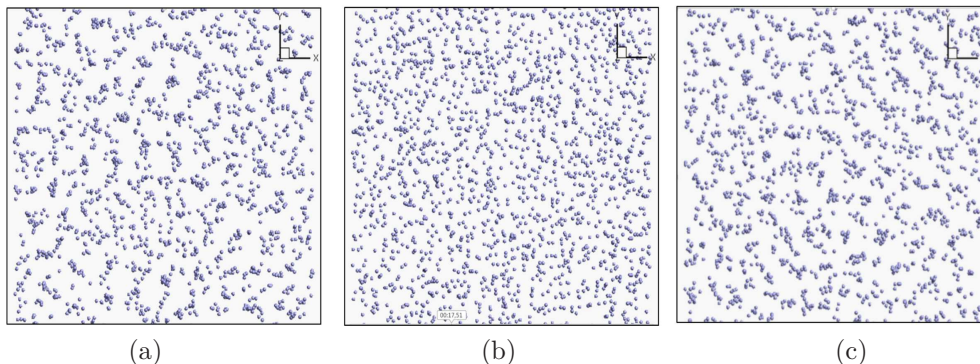


Figure 6: A snapshot of structure forming at initial state (a), and its breaking under an applied shear rate  $\dot{\gamma} = 1$  (b), and then its reforming when the shear rate is reset to zero again (c). It is note that only  $DPD^{(b)}$  particles are plotted.

the dissipative force listed in Table 1  $\mathbf{f}_{ij}^D$ , in a modified version of DPD [28], two parameters are set as  $1.0 \leq r_r \leq 1.5$  and  $k = 1/2$ , to enhance the dynamic response. We use the same setting  $k = 1/2$ , and fix the cut-off radius value  $r_r$  at 1.0 for both  $DPD^{(a)}$  and  $DPD^{(b)}$ . The concentration ratio,  $\phi$ , represents the amount of  $DPD^{(b)}$  to the total mixture,

$$\phi = \frac{N^{(b)}}{N^{(a)} + N^{(b)}}. \quad (27)$$

The simulation is run with 140,000 time steps in which 40,000 time steps is used to ensure the system reaches a fully built-up structure, and after some experiments, a time step  $10^{-2}$  is chosen for all simulations. Periodic boundary conditions are applied in  $x$ - and  $y$ -direction, i.e., particles that pass one periodic face reappear in the domain at the opposite face, and therefore effectively an infinitely large, but periodic DPD system is being considered. In  $z$ -direction, solid walls are represented by three layers of frozen particles. It is known that conventional solid boundary models for DPD lead to slip at the boundary even at moderate applied shear rate. To reduce this, a wall wetting model [36] is employed in the Couette and Poiseuille flows to mimic a hydrophilic behaviour. Figure 7 shows that the non-slip boundary condition is improved with the latter

wall model. In all the following simulations, the Verlet integration algorithm is employed to solve Eq.(26).

Table 2: A typical DPD parameters.

$a_{ij}$	$\sigma_{ij}$	$\gamma_{ij}$	$k_B T$	$m$	$d$	A	B
18.75	3.0	4.5	1.0	1.0	9.8	2	1

In a manner similar to a physical experiment, here one can measure the yield stress  $S_0$  by plotting the shear stress/shear rate curve, and then extrapolate the shear stress (on the Newtonian portion) onto the stress axis. This value on the stress axis gives us an approximation to the yield stress (Figure 8).

It is noted that the effective shear rate is calculated by ignoring the first and the last bin next to the top and bottom wall/boundaries particles. The shear rate used for plotting the shear stress/shear rate curve is thus the actual applied shear rate instead of the input shear rate. From the stress values calculated by post-processing, an average shear stress  $S_{xz}$  is computed and plotted against the actual shear rate, producing the flow curve at equilibrium for a specific set of parameters. It is useful here to use a continuum model, e.g., the Papanastasiou model for correlation. The curve fitting process using the optimisation Generalised Reduced Gradient algorithm (GRG) is carried out to fit DPD data into the continuum model.

### 6.1. Equilibrium flow curves

The range of input shear rates is  $\dot{\gamma} = \{0.01, 0.02, 0.03, 0.05, 0.07, 0.09, 0.10, 0.15, 0.20, 0.30, 0.40, 0.60, 0.80, 1.0\}$ , with a smaller interval at low shear rate for a better observation of the fluid behaviour at low shear rate. Here we let the system reaches equilibrium and then apply a constants shear rate (Figure 9). In this test it is noted that the steady state property of the fluid is of importance here.

An ILDP fluid with DPD parameters listed in Table 2,  $\phi = 0.2$ , and a cut-off radius  $r_a = 1.3$  is selected for this measurement. The viscosity as a function of

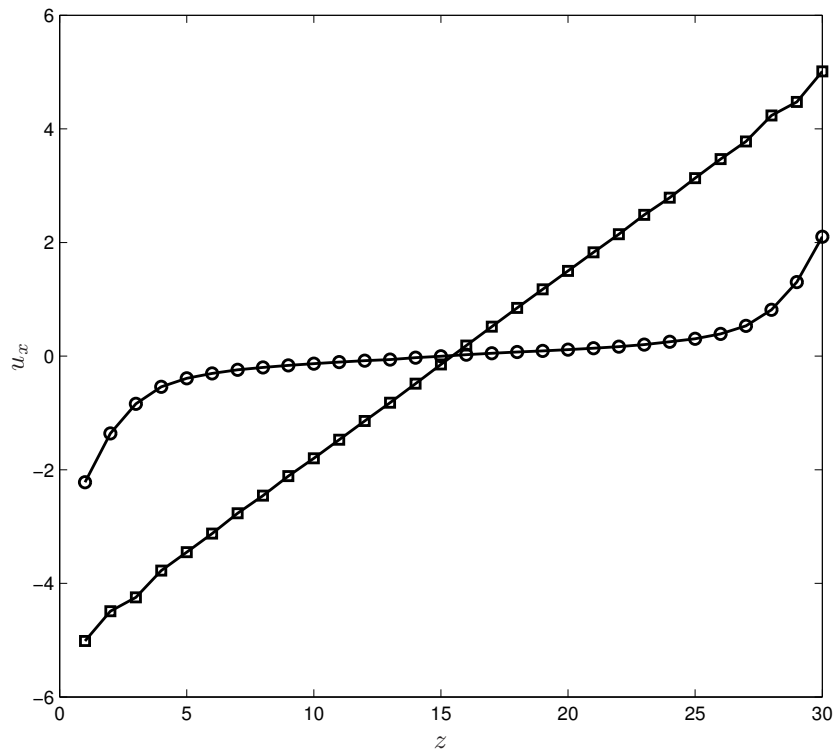


Figure 7: Velocity profile of Couette flow of an ILDP fluid (domain size  $10 \times 10 \times 10$ ,  $\hat{\gamma} = 1$ ) with two wall models: conventional solid boundary models ( $-o$ ) and wall wetting model ( $-\square$ ).

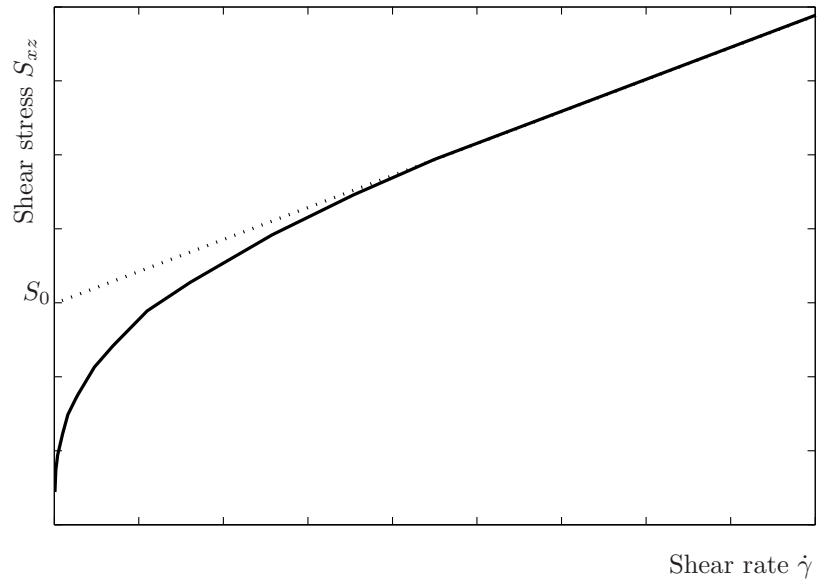


Figure 8: A typical nonlinear curve of shear stress versus shear rate. (Not to scale)

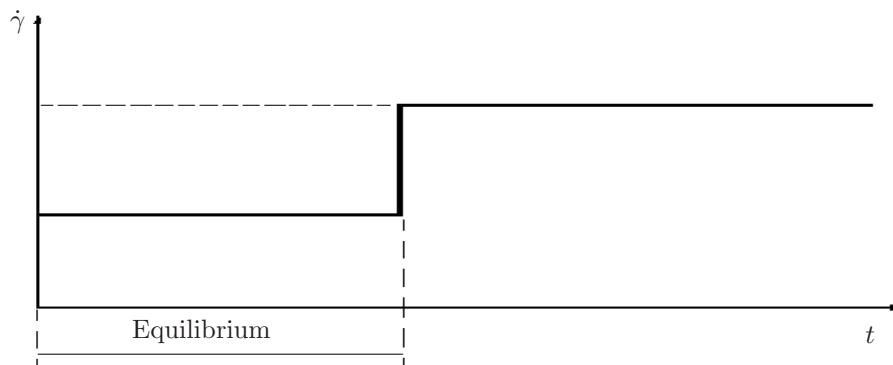


Figure 9: Couette flows: A typical applied shear rate. It is noted that the DPD system is allowed to reach equilibrium and the structures are fully built up after 40000 time steps.

shear rate is plotted in Figure 10. It is known that the flow breaks down the structure network into smaller clusters which results in reducing the apparent viscosity and thus shear-thinning behaviour appears. Numerical data show that  
 295 viscosity decreases dramatically from 600 at applied shear rate 0.03 to 18.8 at applied shear rate 2.0. The Couette velocity profile, temperature and density fluctuation are presented in Figure 11. It can be seen that linear profile of velocity is obtained, and uniform distribution of density and temperature at the chosen time step  $dt = 0.01$ .

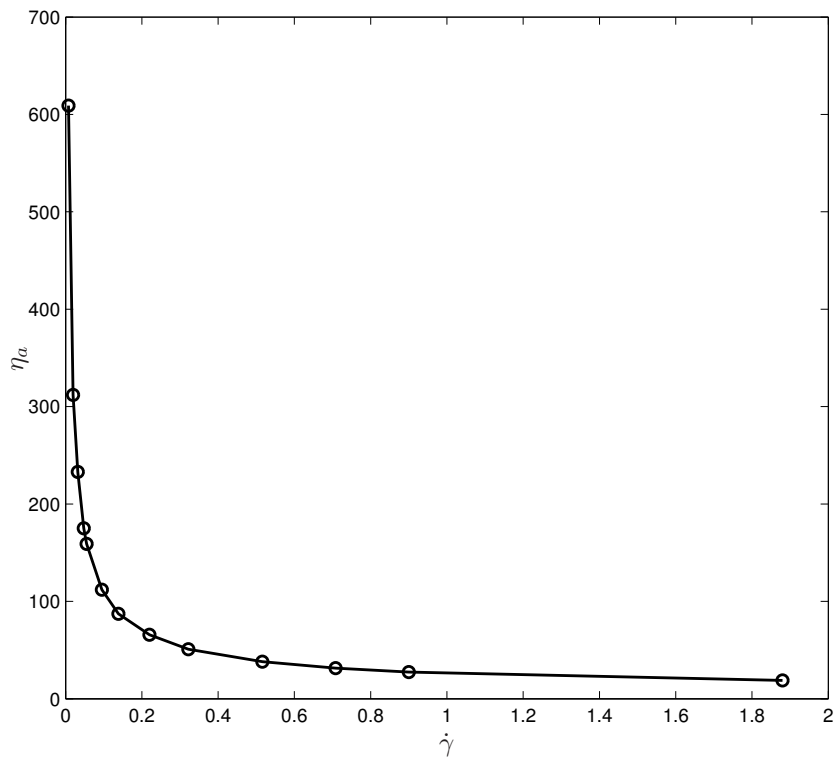


Figure 10: Couette flows (domain size  $20 \times 10 \times 20$ ): apparent viscosity  $\eta_a$  plotted as a function of shear rate.

300 We give an example of using the flow curve of an ILDP fluid to model a foodstuff product, in this case a salad dressing. The steady shear data for

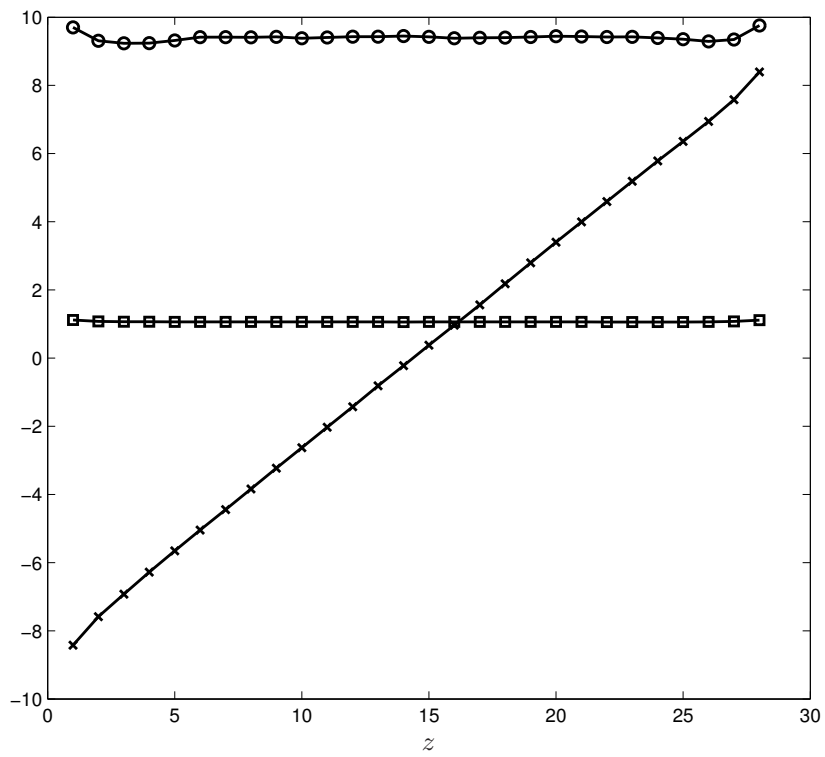


Figure 11: Couette flow (domain size  $20 \times 10 \times 20$ ,  $\dot{\gamma} = 0.5$ ): Profiles of velocity  $u_x$  ( $\times$ ), temperature  $k_B T$  ( $\square$ ) and number density  $d$  ( $\circ$ ).



this salad dressing has been obtained at temperature 295K using a concentric cylinder viscometer ( $R_1 = 20.04$  mm;  $R_2 = 73$  mm;  $h = 60$  mm) [37]. Table 3 shows salad dressing data with shear stresses and the shear rates normalised to  $S_0 = 10.1Pa$ ,  $\dot{\gamma}_0 = 11.396s^{-1}$ , respectively. The DPD data has been collected by the same procedure with the example above with the set of DPD parameters listed in Table 2,  $\phi = 0.2$ . The degree of fit between experiment and numerical data is good, as shown in Figure 12.

Table 3: Couette flow: salad dressing data (dimensionless data).

$\dot{\gamma}/\dot{\gamma}_0$	$S_{xz}/S_0$
0.07	0.40
0.24	0.66
0.49	0.83
1.00	1.07
1.99	1.34
3.00	1.61
4.00	1.79
5.02	1.96
6.03	2.08
7.03	2.20
8.03	2.32

Numerical tests are performed on an ILDP fluid of varying kinetic energy  $k_B T = \{0.7, 0.8, 0.9, 1.1, 1.2, 1.3, 1.4, 1.5, 2.0\}$ . Smaller value of  $k_B T$  reduces the fluctuations in the systems. Non-linear shear stress/shear rate curves are plotted in Figure 13, at different  $k_B T$ . Clearly, the whole family of curves shifts up (i.e., increasing the parameter  $n$  in Papanastasiou's model) with decreasing the kinetic energy  $k_B T$ . It is observed that below the value  $k_B T$  of 0.7, numerical results show that the fluid is not flowing.

We also match the DPD behaviour with the Papanastasiou's model (11). Figure 14 shows the comparison. It can be seen that the data obtained by the DPD model can be fitted well to Papanastasiou's model - the GRG algorithm provides a viscosity of 12.21, stress growth parameter  $n = 20$ , and a yield stress of  $S_0 = 9.98$ .

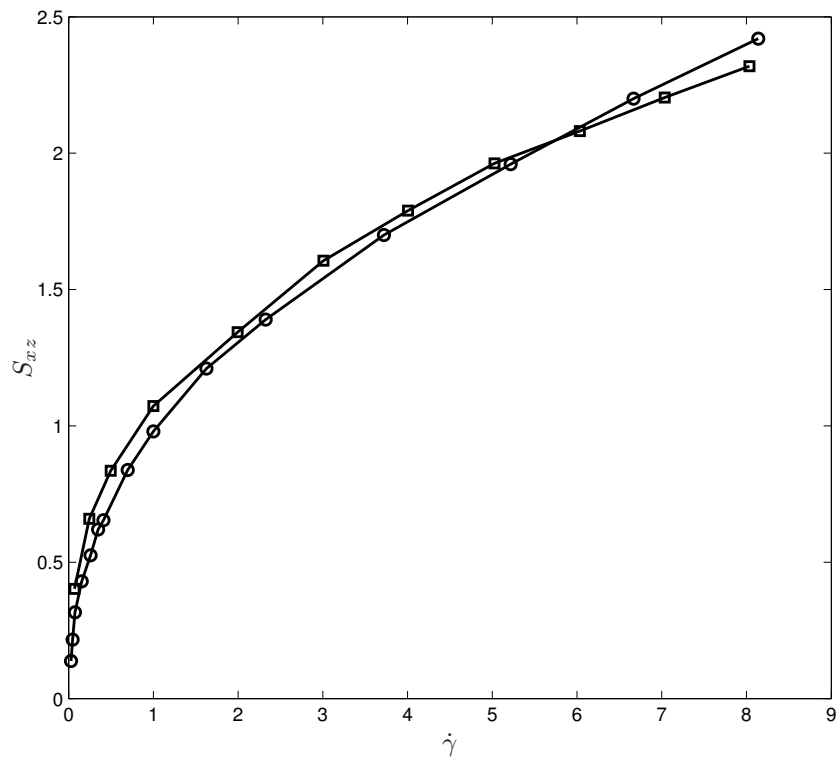


Figure 12: Couette flow: Shear stress/shear rate data for salad dressing data;  $\square$  experiment,  $\circ$  DPD results.

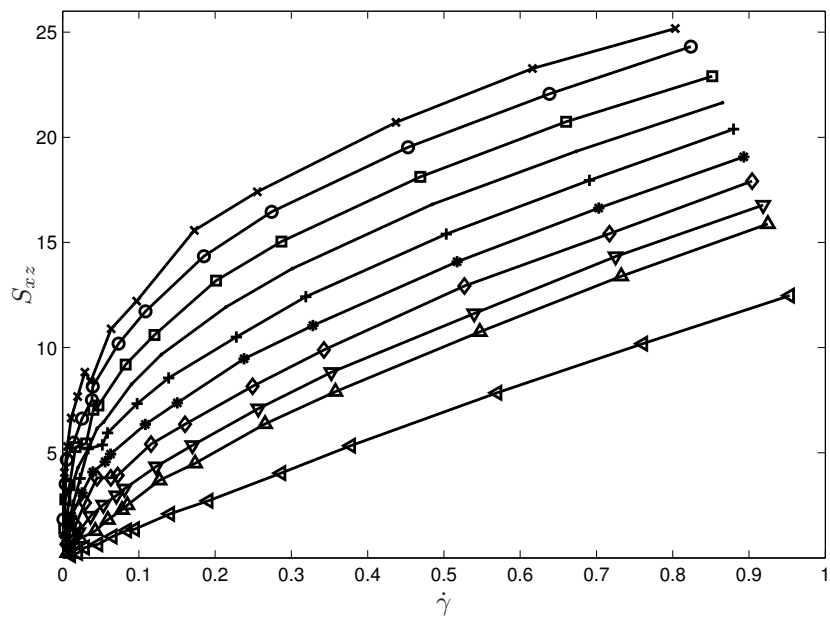


Figure 13: Couette flow: Non-linear shear stress - shear rate behaviour at different values of  $k_B T = \{0.7, 0.8, 0.9, 1.0, 1.1, 1.2, 1.3, 1.4, 1.5, 2.0\}$ , from top to bottom.

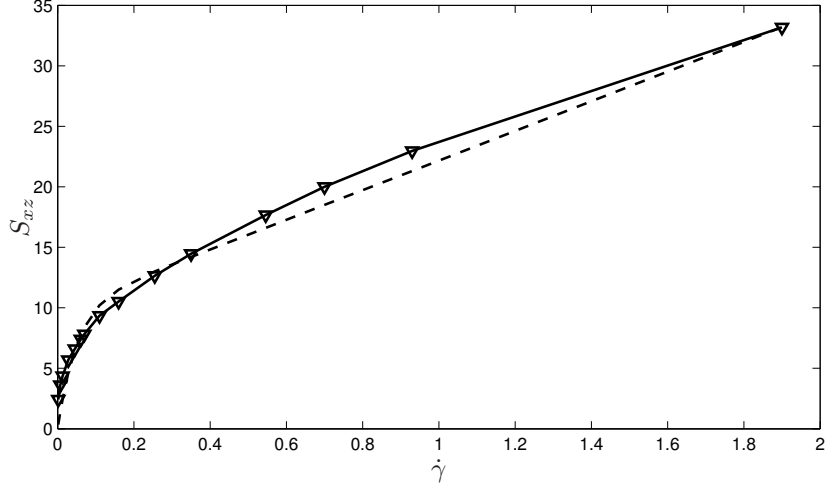


Figure 14: Couette flows: DPD data (Table 2,  $\phi = 0.2$ ,  $r_a = 1.3$ )-solid line-can be fitted well to Papanastasiou's model-dash line-(Eq.11), with a low-shear viscosity of  $\eta + nS_0 = 211.81$  (slope of the rheogram when shear rate smaller than 0.1) and a high-shear viscosity of  $\eta = 12.21$  (slope of the rheogram when shear rate larger than 0.1)(all dimensionless).

### 6.1.1. Effect of range of attractive force

In this part,  $r_a$ , the parameter controlling the cut-off radius of attractive forces between two species is investigated to observe its influence on the macroscopic behaviour. The flow curves for  $r_a$  from 1.1 to 1.32 are shown in Figure 15.

325 For the case that  $r_a = 1.1, 1.2$ , a small value of attractive cut-off radius, corresponds to insufficient bonds needed to form the microstructure network. Hence the fluid behaves such a Newtonian-like fluid. It can be seen from the Figure 15, the fluid behaves more viscous at small shear rate for a larger attractive cut-off radius. This is understandable as a larger cut-off radius for attractive force

330 means a larger effective zone, the  $DPD^{(b)}$  particles can attract more neighboring  $DPD^{(a)}$  particles, therefore increasing numbers of bonds, making the fluid harder to flow. This would make yield stress a higher value, as a larger stress is needed to break these bonds in order to make the fluid start flowing. The plastic viscosity is also increased due to the fact that the particulate network

335 keeps growing which results in more resistant to the flow. However, it should

be noted that if  $r_a$  is increased too large (e.g.  $r_a/r_r \geq 1.3$  for  $\phi = 0.2$  and DPD parameters listed in Table 2), some bulky clusters may be formed which make the system non-homogeneous.

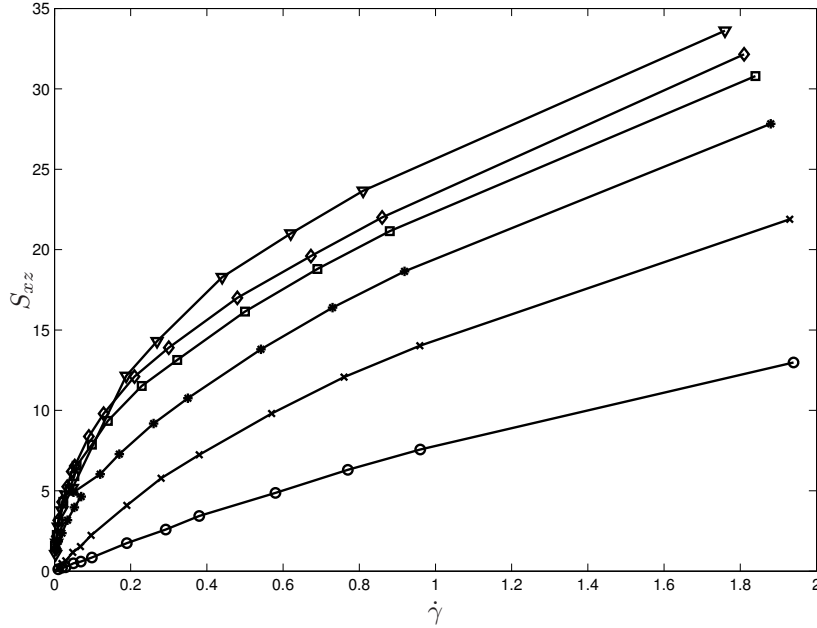


Figure 15: Couette flow: Non-linear shear stress - shear rate behaviour at different values of  $r_a = \{1.32, 1.3, 1.28, 1.26, 1.2, 1.1\}$ , from top to bottom.

### 6.1.2. Effect of concentration ratio, $\phi$

340 The concentration  $\phi$  is increased from 0.05 to 0.2 and the result is presented on Figure 16. As expected, at low concentration of  $DPD^{(b)}$  particles, the fluid behaves like a Newtonian fluid as the connections between two species are not sufficient to create a particulate network. Again, at larger concentrations of  $DPD^{(b)}$  particles, the fluid behaves more like a very high viscous material at  
 345 small shear rate, thus is leaning towards a Bingham plastic material. Due to the increasing amounts of interaction among particles, more bonds are formed, thus hinder the flow. This structure also results in larger stress needed to cause

the material to shear, i.e., larger stress for flow to start, thus a higher yield stress trend is observed. It is observed that if  $\phi$  is roughly larger than 0.2 with  
 350  $r_a = 1.3$  the material become too viscous to have a proper flow.

However, one may expected that a smaller  $r_a$  allows a larger range of  $\phi$  available for the model. In an attempt to verify this, some numerical experiments are carried out for  $\phi = 0.28$  and three values of  $r_a$  including  $\{1.23, 1.24, 1.25\}$  (smaller than 1.3). From Figure 17, at  $r_a = 1.25$ , the fluid shows a clear non-  
 355 linear behaviour from the shape of the flow curve, and resembles a pseudo-yield stress material. Thus with a concentration  $\phi = 0.28$ , a decreasing of  $r_a$  roughly from 1.3 to 1.25 is needed to obtain a proper flow. It can be seen that with material of DPD parameters  $\phi = 0.28, r_a = 1.25$ , the shear stress versus shear rate curve exhibits clearer yield stress behaviour than that of  $\phi = 0.2, r_a = 1.3$   
 360 (lower concentration but higher attractive cut-off radius).

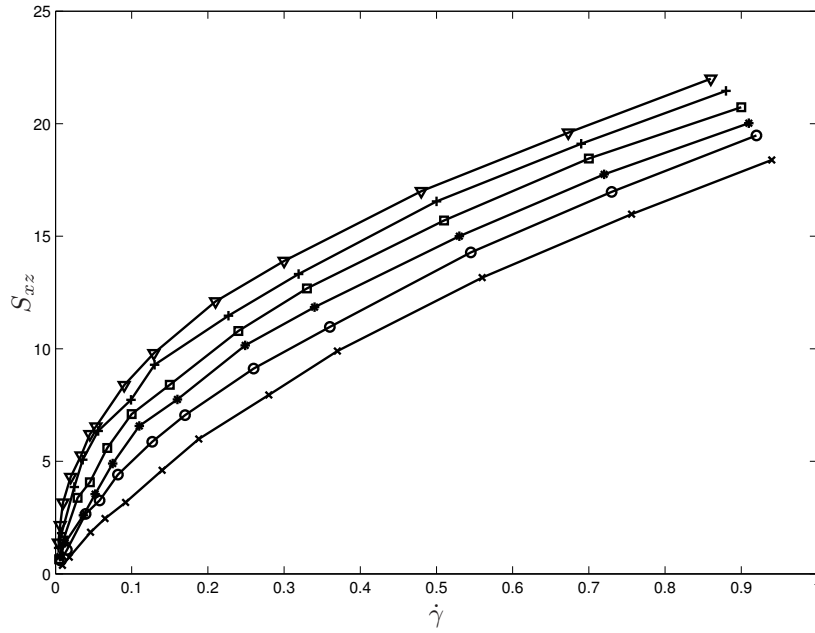


Figure 16: Couette flow: Non-linear shear stress - shear rate behaviour at different values of  $\phi = \{0.200, 0.175, 0.150, 0.125, 0.100, 0.050\}$ , from top to bottom.

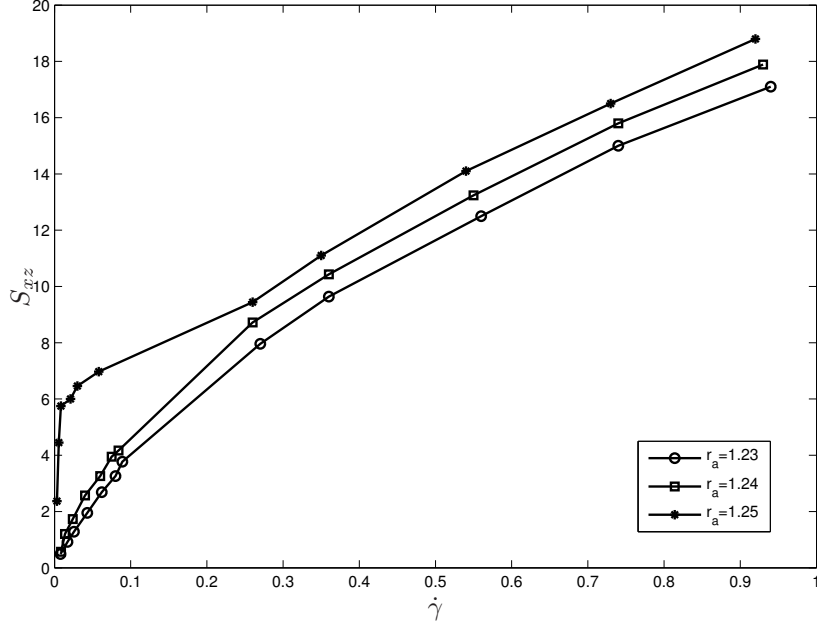


Figure 17: Couette flow: Non-linear shear stress - shear rate behaviour of  $\phi = \{0.28\}$  and  $r_a = \{1.23, 1.24, 1.25\}$ .

### 6.2. Constants structure curves

In this section we investigate the transient response of the ILDP fluid, shown in Figure 18, where an overshoot in the shear stress can be observed. It is noted that there is no overshoot when the attractive force component is set to zero (i.e., when  $B = 0$ ). The ILDP yield stress fluid is also thixotropic and can be represented by a family of constant structure (CS) curves, in the same manner as described by [9] and [34].

To measure the CS curves, we carry out a step-change in a shear rate experiment (Figure 19), as suggested by [34]. It is known that each reference shear rate has one structure level corresponding to a CS curve; therefore, the test is carried out by choosing one reference shear rate and then increase or decrease around this reference value. For example, the ILDP fluid is placed between two

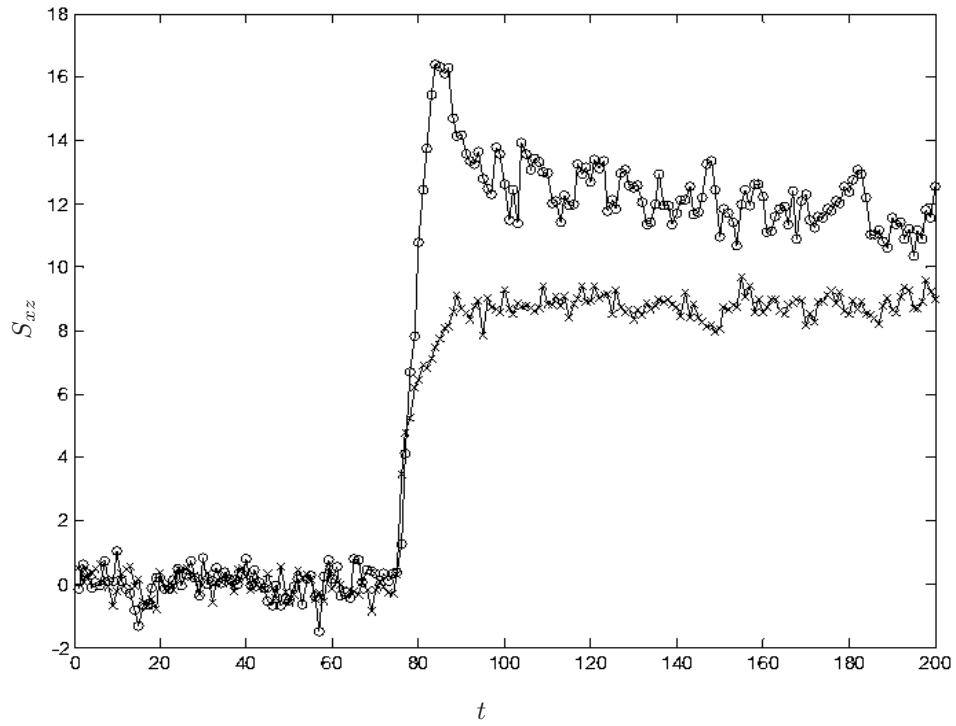


Figure 18: Thixotropic behaviour: Time response of ILDP fluid ( $-\circ$ ) and that of a DPD Newtonian fluid ( $-\times$ ) in a simple shear flow with a constant shear rate  $\dot{\gamma} = 0.3$ .

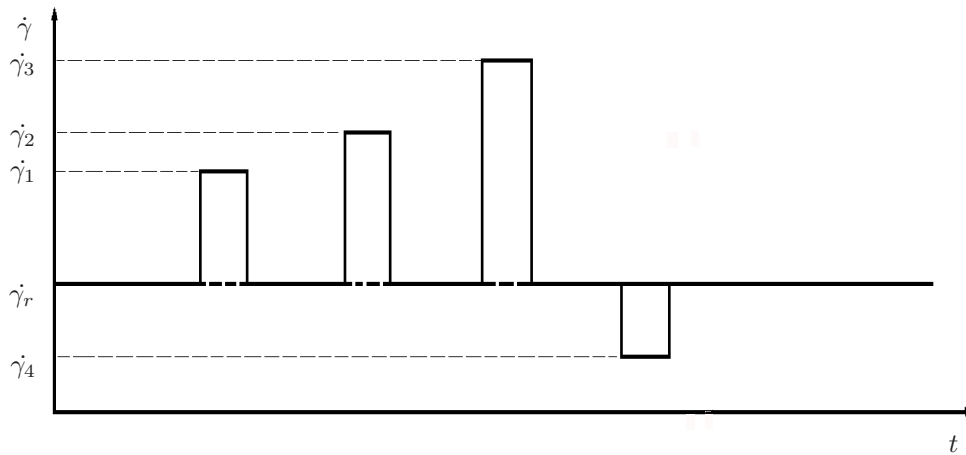


Figure 19: Thixotropic behaviour: Shear rate experiments with step-change to obtain the family of constant structure curves.



parallel plates and subjected to a reference shear rate  $\dot{\gamma} = \dot{\gamma}_r$  until the equilibrium state is reached. A higher shear rate  $\dot{\gamma} = \dot{\gamma}_1$  is then applied and the peak  
375 value of shear stress  $S_1$  is noted before  $\dot{\gamma}$  is set back to  $\dot{\gamma}_r$ . The same procedures  
are carried out with  $\dot{\gamma} = \{\dot{\gamma}_2, \dot{\gamma}_3, \dots, \dot{\gamma}_n\}$  and  $S = \{S_2, S_3, \dots, S_n\}$  are noted.  
The CS curve for the shear rate  $\dot{\gamma}_r$  is the plot of  $S$  versus  $\dot{\gamma}$ . In this report,  
we repeat this procedure for three reference shear rates  $\dot{\gamma}_r = \{0.05, 0.36, 1.91\}$   
and then plotted the set of CS curves. To quantify the time dependent rheo-  
380 logical behaviour, the non-dimensional structure parameter  $\lambda$  as introduced in  
[9] is adopted. Here the maximum value of the degree of structure  $\lambda_0$  is set  
to one (i.e., fully structured); the value of  $\lambda_e$  of each reference shear rate is  
determined from Eq.13. To construct the CS curves in Toorman's model (16),  
there are four parameters  $S_0, \mu_\infty, c$  and  $\beta$  are needed. By fitting, one can find  
385 these four parameters  $S_0 = 5.13, \mu_\infty = 12.16, c = 73.57$  and  $\beta = 60.57$  from the  
Bingham flow curve with a high shear viscosity of 11.66 and a yield stress of  
6.72 (which corresponds to a DPD fluid having parameters listed in Table 2 and  
 $r_a = 1.24, \phi = 0.4$ ). The fixed value of  $\lambda_i$  on a CS curve is to be identified as  
 $\lambda_E$  at the crossover point between this CS and the EF curves. A substitution  
390 of those values ( $S_0, \mu_\infty, c, \beta$ ) and  $\lambda_i$  of each CS curve in Eq.16 give a family of  
CS curves.

In Figure 20, a comparison of the DPD data and CS curves obtained from  
Toorman's model is shown. It can be seen that the ILDP fluid responds in a  
similar manner to Toorman's model for a structure dependent thixotropic fluid.

### 395 6.3. Poiseuille flow

In this part, we explore ILDP fluid in Poiseuille flow. As shown in Figure  
21, an ILDP fluid is placed between two parallel plates under a body force field  
 $\mathbf{g} = (g_x, 0, 0)$  to simulate Poiseuille flow (in the  $x$ -direction). Periodic boundary  
conditions are applied to fluid boundaries in the  $x$ - and  $y$ -directions. Velocity  
400 component in  $x$ -direction,  $u_x$ , is plotted versus  $z$ -coordinate to construct the  
velocity profile.  $z_0$  is the transition line at which the material yields.

We consider the same ILDP fluid with parameters tabulated in (Table 2).

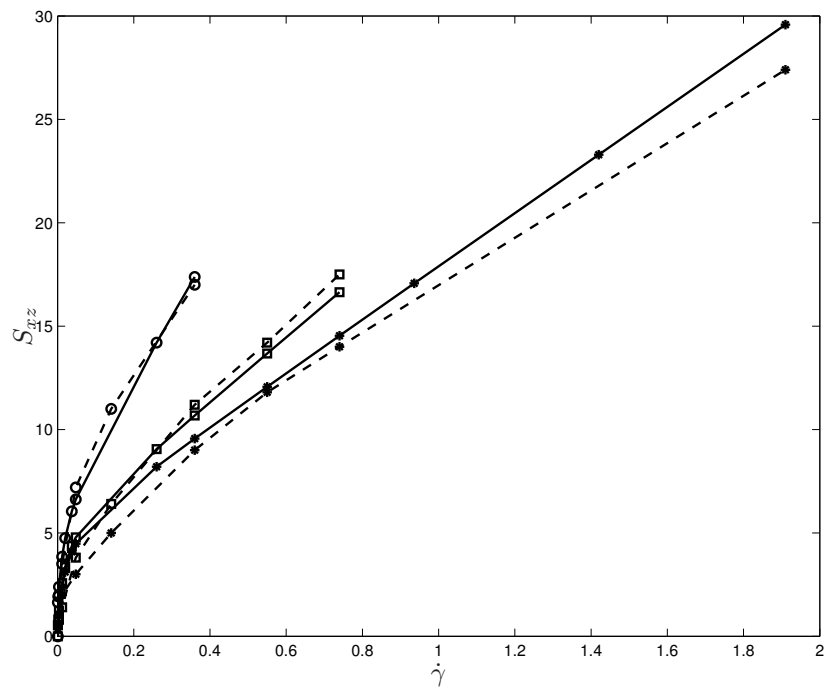


Figure 20: Shear stress plotted as a function of shear rate: constant structure curves (*CSC*). Dash line: constant structure curves obtained from DPD data for reference shear rates  $\dot{\gamma}_r = 0.05(-\circ), 0.36(-\square), 1.91(-*)$  (from top to bottom); solid line: Toorman's model.

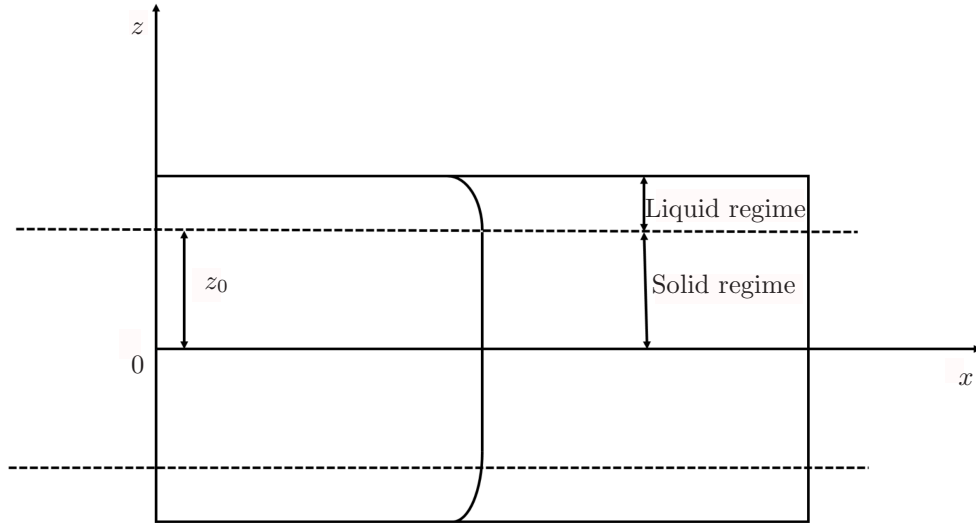


Figure 21: Poiseuille flow between two parallel plates: unyield and yield regions for velocity profile  $u_x$  along  $z$ -direction.

Figure 22 provides the plot of  $u_x$  with respect to the applied body force  $g_x = 0.3$ . As expected, the velocity profile of ILDP fluid (Figure 22b) is no longer parabolic as that of DPD-Newtonian fluid ( $\mathbf{f}^{Ca} = 0$ , Figure 22a). The plugged flow region near the centre is clearly visible - in this region the shear rate is low, consequentially the applied stress is smaller than the yield tress  $S_0$  leading to a plugged flow.

**Comparison velocity profile to the analytic solution of Bingham model:** with a body force of  $g_x = \{0.1, 0.2, 0.3, 0.4, 0.5\}$ , the phase transition point  $z_0$  changes from solid to liquid regime. With larger body force, the velocity profile becomes more parabolic, with a smaller plugged (no-yield) zone. At large enough pressure gradient, the plugged zone reduces to zero indicating that the applied stresses are large enough to break all particulate structures. We note that the analytical solution of non-dimensional Bingham model can be written

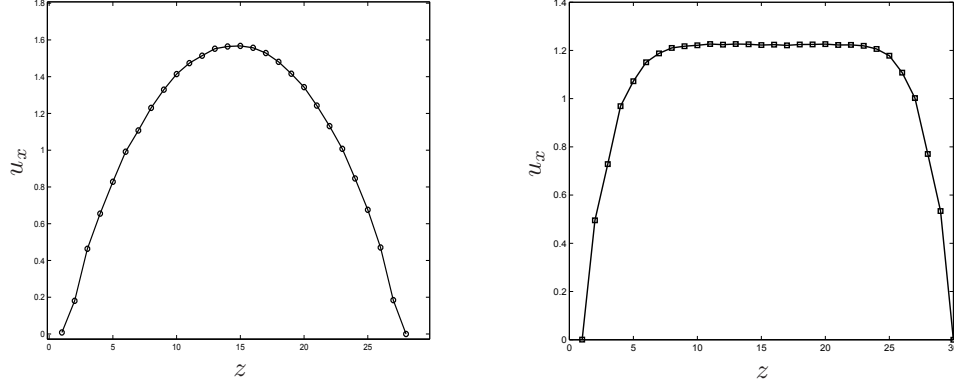


Figure 22: Poiseuille flow: Average velocity  $u_x$  : (a) DPD-Newtonian fluid— $\circ$  ( $f^{Ca} = 0$ ) - (b) ILDP fluid— $\square$  (Table 2,  $\phi = 0.2$ ).

as [38], [5]

$$u_x = \frac{1}{2}g_x(H^2 - z^2) - Bn(H - z) \quad z_0 < z < H \quad (28)$$

$$u_x = \frac{1}{2}g_x(H - z_0)^2 \quad 0 < z < z_0, \quad (29)$$

The phase transition point is determined by  $z_0 = Bn/g_x$  ( $Bn$  the Bingham number;  $g_x$  the non-dimensional pressure gradient). It can be seen that the line  $z = z_0$  is the threshold between the pre- and post-yield zones and it can be determined simply by tracking the point when the applied shear stresses larger than yield stress value  $S_0$ . For example in Figure 23, the ratio  $|Sxz/S_0|$  is larger than one at position  $z = 0.13$ , one can consider the line  $z_c = 0.13$  the phase transition line. Based on the analytical solution at the plateau zone, one can determine the value  $z_0$  from the plug zone:

$$\frac{u_x}{g_x} = \frac{1}{2}(H - z_0)^2. \quad (30)$$

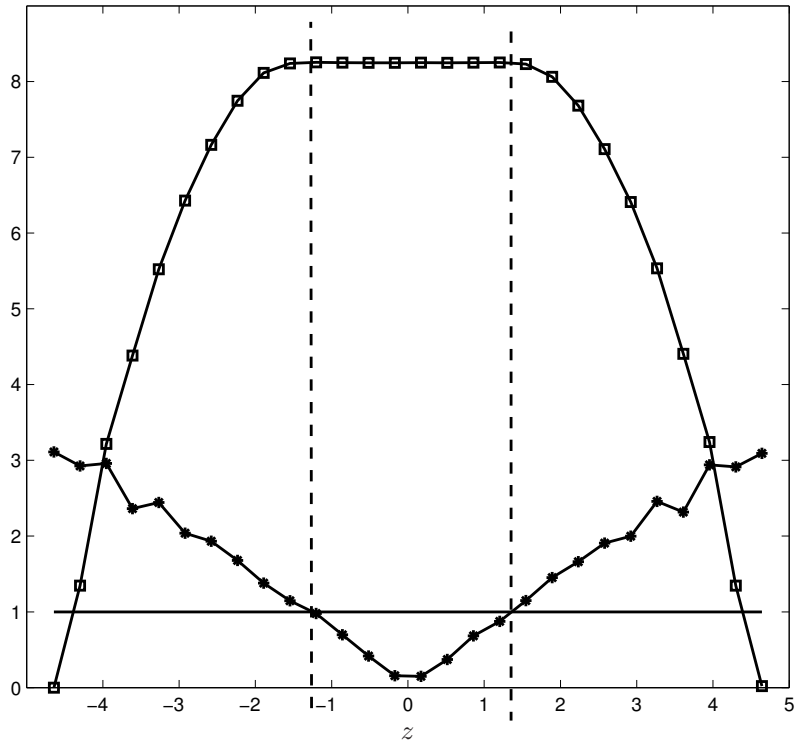


Figure 23: Poiseuille flow: Average velocity  $u_x$  ( $\square$ ) and variation of  $|Sxz/S_0|$  ( $*$ ) across the channel the  $x$ -component of applied body force 0.8.

This equation can be rewritten as

$$z_0 = H - \sqrt{2 \frac{u_x}{g_x}}. \quad (31)$$

The maximum value of velocity  $u_x$ , the  $x$ -component of body force  $g_x$  and the yield line  $z_0$ , calculated from Eq.31, are listed in Table 4. The non-dimensional velocity profiles of ILDP fluid and the analytical solution for Bingham number  $Bn = 0.2, 0.22, 0.25, 0.29, 0.34$  are shown in Figure 24. It can be seen that the ILDP results are comparable to those of analytical solutions. However, it should be noted that at the transition zones, the ILDP fluid exhibits a “softer” transition than that of a Bingham fluid (i.e., representing a transition here between high and low viscous flow regimes rather than the solid to liquid regimes transition in a truly yield stress fluid).

Table 4: DPD Poiseuille flow: maximum velocities with respect to  $g_x = \{0.2, 0.3, 0.4, 0.5, 0.6\}$  and transition line  $z_0$ .

$g_x$	$u_x$	$z_0$
0.2	1.2	0.34
0.3	2.22	0.29
0.4	3.13	0.25
0.5	3.80	0.22
0.6	4.30	0.20

## 7. Concluding Remarks

In the present study, the DPD method has been used as a bottom-up approach to obtain the desired macroscopic properties for a yield-stress and thixotropic material. We propose a simple way to construct a particulate structure network to mimic the natural processes in concentrated cohesive mixtures. The DPD model comprises of two DPD species with different conservative forces, one with only repulsive forces (between the same species) and the other with short range repulsive and long-range attractive forces (between different species) - we call this DPD fluid the Indirect Linkage Dissipative Particle (ILDP) fluid.

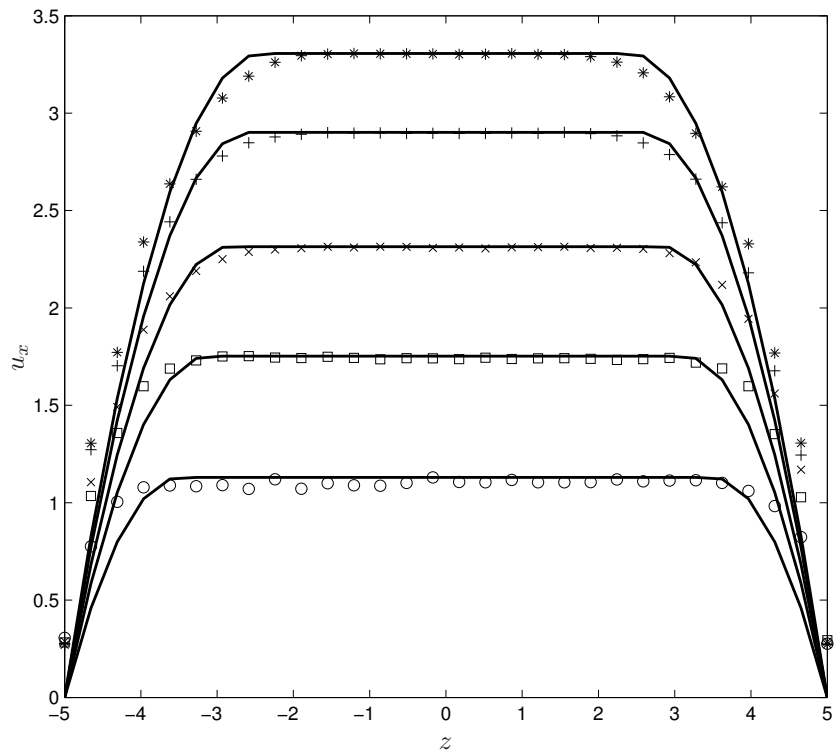


Figure 24: Poiseuille flow: Non-dimensional velocity profiles (\*, +, ×, □, ○) with respect to  $Bn = 0.2, 0.22, 0.25, 0.29, 0.34$  from top to bottom and the analytical steady state solutions of Bingham fluids (—).

The constitutive framework is fully specified with the microstructure that goes into the description of the DPD model. In particular, the numerical results demonstrate that

- 430 • The indirect linkage between  $DPD^{(b)}$  particles insures that the uniform distribution of two DPD species over the computational domains;
- An ILDP microstructure network is formed and ruptured under a certain applied stress and then recovers when the applied stress level sufficiently reduces;
- 435 • Yield stress and shear thinning effects are the consequences of the flocculation and break up of the microstructures;
- The shear stress/shear rate curves are shifted up (i.e., increasing Papanastasiou's parameter  $n$ ) with decreasing the kinetic energy of ILDP fluid.

The present model is able to produce nonlinear and thixotropy between  
440 shear stress and shear rate in viscometric flow that have been observed, or predicted by continuum methods. The ILDP velocity profile in Poiseuille flow is similar to analytical solution obtained with the Bingham's model. The model also produces the expected CS curves from high shear to low shear and vice versa flows, in qualitative agreement with Toorman's model. More detailed  
445 studies on the transient flows of thixotropic fluid with the ILDP model should be a welcome contribution, in particular in the area of highly concentrated and cohesive suspension mixtures.

The advantage of the DPD approach is that the multiphase properties of the system are reconstructed, without any reference to a particular constitutive  
450 equation. However, DPD model has many parameters to control such as conservative force coefficients including repulsive and attractive forces, random force and dissipative force coefficients, the Boltzmann temperature of the system, the ratio of  $DPD^{(a)}$  and  $DPD^{(b)}$  particles. A set of "standard" parameters have been well investigated, however. Another important parameter in addition to



<sup>455</sup> the DPD parameter is concentration of one phase. Different from top-down approaches where macroscopic properties (e.g., viscosity, yield stress) are inputs which are obtained from physical experiments, the DPD fluid properties are the results of microstructure interactions, and the need of numerical experiments to determine a proper set of parameters.

<sup>460</sup> The reported study was restricted to viscometric flows. For non-viscometric problems (e.g., contraction-expansion flows), compressibility effects may be present, which may effectively controlled by reducing DPD mass [19]).

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