Pairwise Force Smoothed Particle Hydrodynamics model for multiphase flow: surface tension and contact line dynamics

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Abstract

We present a novel formulation of the Pairwise Force Smoothed Particle Hydrodynamics Model (PF-SPH) and use it to simulate two- and three-phase flows in bounded domains. In the PF-SPH model, the Navier-Stokes equations are discretized with the Smoothed Particle Hydrodynamics (SPH) method and the Young-Laplace boundary condition at the fluid-fluid interface and the Young boundary condition at the fluid-fluid-solid interface are replaced with pairwise forces added into the Navier-Stokes equations. We derive a relationship between the parameters in the pairwise forces and the surface tension and static contact angle. Next, we demonstrate the accuracy of the model under static and dynamic conditions. Finally, to demonstrate the capabilities and robustness of the model we use it to simulate flow of three fluids in a porous medium.

1. Introduction

Flow of two or more fluid phases in a bounded domain is a complex problem, commonly described by the multiphase Navier-Stokes equations.

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11 These equations have two nonlinearities: one is the standard inertial term,
 12 and another is due to the Young-Laplace and/or Young boundary condi-
 13 tions at the fluid-fluid and/or fluid-fluid-solid interfaces, respectively. Eu-
 14 lerian grid-based Computational Fluid Dynamics methods have been used
 15 to directly solve the multiphase Navier-Stokes equations, but they require
 16 computationally expensive interface tracking techniques. The phase-field
 17 (Liu et al., 2007), level-set (Fedkiw, 2003), and volume-of-fluid (Huang and
 18 Meakin, 2008; Raeini et al., 2012; Tomin and Lunati, 2013) methods have
 19 been proposed to simplify interface tracking by introducing an additional
 20 field variable defining the position of the interface. In these methods, the
 21 explicit tracking of the fluid-fluid interface is avoided at the cost of solving
 22 an additional hyperbolic equation for the field variable.

23 In addition to the non-linearity caused by moving fluid-fluid interfaces,
 24 the standard Navier-Stokes description presents two specific challenges for
 25 modeling multiphase flow in bounded domains: 1) there is an unphysical
 26 discontinuity in the fluid velocity across the fluid-fluid-solid interface (Dus-
 27 san V, 1979); and 2) a model relating the contact angle to the velocity of the
 28 contact line should be prescribed. Under static conditions, the static contact
 29 angle θ_0 can be easily measured and prescribed as a parameter for a given
 30 fluid-fluid-solid system. Under dynamic conditions, the contact angle θ may
 31 have a complex, hysteretic behavior and is difficult to measure.

32 Different approximate models (e.g creeping flow and lubrication flow (Huh
 33 and Scriven, 1971)) for dealing with velocity discontinuity and phenomeno-
 34 logical models (Tanner, 1979; de Gennes, 1985; Cox, 1986) for dynamic con-
 35 tact angles have been proposed. Numerical solution of the resulting equa-

36 tions, especially in geometrically complicated domains, can be difficult to
 37 obtain. As a result, very few Eulerian models, based on a direct solution
 38 of the Navier-Stokes equations, have been used to model multiphase flow
 39 in domains with geometrically complicated boundaries such as, for example,
 40 porous media. A review of numerical methods for pore-scale multiphase flows
 41 can be found in Meakin and Tartakovsky (2009).

42 Here we present a novel formulation of the Pairwise Force Smoothed Parti-
 43 cle Hydrodynamics Model (PF-SPH) for the multiphase Navier-Stokes equa-
 44 tions. Smoothed Particle Hydrodynamics (SPH) is a Lagrangian method
 45 for solving conservation equations, including mass, momentum and energy
 46 conservation equations (Monaghan, 2005). SPH approximations of spatial
 47 derivatives are two-step. Differential operators are approximated with inte-
 48 grals and integrals are approximated with Riemann sums. SPH is an attrac-
 49 tive alternative to the Eulerian methods for solving interfacial problems. In
 50 SPH, each fluid phase is discretized with its own set of particles (discretiza-
 51 tion points) that are advected according to the equation of motion. There
 52 are two main approaches in SPH to impose the Young-Laplace boundary
 53 condition (and the surface tension) at the fluid-fluid interface and the Young
 54 condition (contact angle) at the fluid-fluid-solid interface. The first approach
 55 is based on the Continuum Boundary Force (CBF) method (Brackbill et al.,
 56 1992; Morris, 2000; Hu and Adams, 2006; Adami et al., 2010). The second
 57 approach is the so-called Pairwise Force Smoothed Particle Hydrodynamics
 58 method (Tartakovsky and Meakin, 2005a,b, 2006; Tartakovsky et al., 2009a,
 59 2007). In the both approaches, the boundary conditions are replaced with
 60 forces that have non-zero net contributions only on SPH particles near the

61 corresponding boundaries. In the CBF approach, these forces are directly
 62 computed from related to the boundary conditions. This approach has the
 63 advantage that the surface tension and contact angle can be prescribed di-
 64 rectly. But the negative consequence of this is that, as Eulerian methods,
 65 the CBF-SPH approach has to deal with nonphysical stress singularity and
 66 requires a phenomenological model for the dynamic contact angle. In the
 67 PF-SPH method, the forces responsible for the Young-Laplace and Young
 68 boundary conditions are assumed to be a pairwise molecular-like forces. As
 69 in the Molecular Dynamics method, in the PF-SPH model the behavior of
 70 the fluid-fluid and fluid-fluid-solid interfaces is the result of a force balance.
 71 Therefore, the PF-SPH method avoids the nonphysical stress singularity and
 72 does not require any knowledge of the contact angle dynamics. The PF-SPH
 73 method has been extensively used for modeling two-phase flows including
 74 liquid-gas flow and flow of two liquids in porous media. Until now, the
 75 analytical relationship between the pair-wise forces and the continuum hy-
 76 drodynamic parameters such as surface tension and the contact angle has not
 77 been established and the main disadvantage of the original PF-SPH model
 78 had been the need to calibrate the parameters in the pairwise forces with
 79 respect to the desired surface tension and static contact angle. The first
 80 step in establishing the correlation between the parameters in the PF-SPH
 81 model and the surface tension and contact angle has been made in Bandara
 82 et al. (2013) based on the theory of capillarity of Maxwell (1890). A good
 83 agreement between PF-SPH model with the parameters estimated based on
 84 the Maxwell theory and multiphase microfluidic laboratory experiments has
 85 been observed in Bandara et al. (2013), but some important questions with

86 regard to the applicability of the Maxwell theory to the SPH method have
 87 remained unanswered. In his work, Maxwell has derived a relationship be-
 88 tween molecular pairwise forces and the surface tension, based on a “static
 89 view of the matter” (Rowlinson and Widom, 2002). In the SPH method,
 90 pressure gradient and viscous forces are implicitly many-body forces as they
 91 depend on local particle density and the contribution of these forces to the
 92 surface tension cannot be readily explained by the Maxwell theory. Also
 93 being a continuum approximation of the discrete molecular system, Maxwell
 94 theory does not contain estimates of the resulting errors, which is important
 95 in applying it to a numerical method such as SPH.

96 In this paper we derive relationships between parameters in the pairwise
 97 forces and surface tension and static contact angle using thermodynamics
 98 considerations. We start by relating SPH forces to the stress in the fluid
 99 using Hardy formula (Hardy, 1982) and expressing surface tension as an in-
 100 tegral of the difference of the normal and tangent components of the stress
 101 taking along a line normal to the interface using the Gibbs thermodynamics
 102 theory (Rowlinson and Widom, 2002). To obtain a closed form relationship
 103 between SPH forces and pressure and surface tension, we approximate (dese-
 104 crate) Hardy stress, given as a Riemann sum over all SPH particles, with an
 105 integral. This approximation is similar to the SPH approximation of spatial
 106 derivatives, and, therefore, the accuracy of the resulting relationship between
 107 the pairwise forces and the surface tension is similar to the accuracy of the
 108 SPH discretization of spatial derivatives. Next, we demonstrate the accu-
 109 racy of the PF-SPH method under static and dynamic conditions. Finally,
 110 to demonstrate the capabilities and robustness of the method, we use it to

111 simulate pore-scale flow of three fluids in a porous medium. To the best of
 112 our knowledge, this is the first model able to simulate pore-scale flow of three
 113 and more fluids.

114 2. Governing Equations

115 Flow of two or more fluids is traditionally described by the Navier-Stokes
 116 equations written for each fluid phase α in the form of the continuity equa-
 117 tion:

$$\frac{D\rho_\alpha}{Dt} = -\rho_\alpha (\nabla \cdot \mathbf{v}_\alpha), \quad (1)$$

118 and the momentum conservation equation:

$$\rho_\alpha \frac{D\mathbf{v}_\alpha}{Dt} = -\nabla P_\alpha + \nabla \cdot \boldsymbol{\tau}_\alpha + \rho_\alpha \mathbf{g}, \quad (2)$$

119 where $\mathbf{v}_\alpha$ is the velocity vector; $\boldsymbol{\tau}_\alpha = [\mu_\alpha(\nabla \mathbf{v}_\alpha + \nabla \mathbf{v}_\alpha^T)]$ is the viscous stress
 120 tensor; ρ_α is the density; P_α is the pressure; μ_α is the dynamic viscosity; $\mathbf{g}$
 121 is the gravitational acceleration; and $D/Dt = \partial/\partial t + \mathbf{v}_\alpha \cdot \nabla$ denotes a total
 122 derivative.

123 For generality, we treat the fluids as compressible and prescribe an equa-
 124 tion of state $P_\alpha = f(\rho_\alpha)$ for each phase to close Eqs. (1) and (2). It is
 125 common to assume the no-slip boundary conditions for the fluid velocity at
 126 the fluid-solid boundary

$$v_{\alpha,n} = 0 \quad \text{and} \quad v_{\alpha,\tau} = 0; \quad (3)$$

127 the dynamic Young-Laplace boundary condition for pressure and velocity at
 128 the fluid-fluid-interface

$$(P_\alpha - P_\beta)\mathbf{n} = (\boldsymbol{\tau}_\alpha - \boldsymbol{\tau}_\beta) \cdot \mathbf{n} + \kappa \sigma_{\alpha\beta} \mathbf{n}; \quad (4)$$

129 and to prescribe the contact angle θ at the contact line (fluid-fluid-solid
130 interface). Here, $v_{\alpha,n}$ and $v_{\alpha,\tau}$ are the normal and tangent components of
131 velocity; α and β denote the non-wetting and wetting phases, respectively;
132 κ is the curvature of the interface; and $\sigma_{\alpha\beta}$ is the surface tension between
133 α and β fluids. The normal vector $\mathbf{n}$ is pointed away from the non-wetting
134 phase. At equilibrium the (static) contact angle, θ_0 , is given by the Young
135 formula

$$\sigma_{\alpha\beta} \cos \theta_0 + \sigma_{s\alpha} = \sigma_{s\beta}. \quad (5)$$

136 where $\sigma_{s\alpha}$ and $\sigma_{s\beta}$ are the superficial surface tension between the solid and
137 the fluid phases α and β , respectively. Under dynamic conditions, the fluid-
138 fluid-solid contact line moves and the dynamic contact angle depends on the
139 direction of flow and the magnitude of the velocity of the contact line. The
140 advancing contact angle, θ_a , formed by advancing wetting fluid is greater
141 than θ_0 and the receding contact angle, θ_r , formed by receding wetting fluid
142 is smaller than θ_0 . A number of phenomenological models exist for dynamic
143 contact angles, including the exponential model (Meakin and Tartakovsky,
144 2009):

$$\theta_a - \theta_0 = a_a u^{b_a}, \quad \theta_r - \theta_0 = a_r u^{b_r}. \quad (6)$$

145 It should be noted that the combination of the the no-slip boundary condition
146 away from the contact line and the assumption that the contact line moves
147 with non-zero velocity leads to a nonphysical divergence of the viscous stress
148 (Dussan V, 1979). A review of grid-based Eulerian methods for modeling the
149 contact line dynamics can be found in Bonn et al. (2009).

150 3. PF-SPH multiphase flow model

151 In the multiphase PF-SPH model (Tartakovsky and Meakin, 2006), vari-
 152 ous fluid and solid phases are represented by separate sets of particles. These
 153 particles serve as discretization points for the Navier-Stokes equations. The
 154 SPH method is based on a meshless interpolation scheme that allows estima-
 155 tion of a vector or scalar function $A(\mathbf{r})$ (e.g. velocity, pressure, density) at
 156 position $\mathbf{r}$ in terms of the values of the function at the discretization points
 157 with positions $\mathbf{r}_j$:

$$A(\mathbf{r}) \approx \int A(\mathbf{r}') W(|\mathbf{r} - \mathbf{r}'|, h) d\mathbf{r}' \approx \sum_{j=1}^N \frac{A_j}{n_j} W(|\mathbf{r} - \mathbf{r}_j|, h), \quad (7)$$

where $A_j = A(\mathbf{r}_j)$, N is the total number of SPH particles, $n_j = \rho_j/m_j$ is the number density associated with point j , m_j is the mass of particle j , and $\rho_j = \rho(\mathbf{r}_j)$ is the fluid density. The function W is the SPH smoothing kernel with compact support h ($W(|\mathbf{r}| > h) = 0$). Due to the compactness of W , the summation in Eq. (7) can be replaced with summation over just the particles within the distance h from $\mathbf{r}$. The smoothing kernel W must have at least a continuous first derivative and satisfy the normalization condition $\int W(\mathbf{r} - \mathbf{r}', h) d\mathbf{r}' = 1$, where integration is performed over the entire domain of A . In the limit $h \rightarrow 0$, W is required to reduce to the Dirac delta function: $\lim_{h \rightarrow 0} W(\mathbf{r} - \mathbf{r}', h) = \delta(\mathbf{r} - \mathbf{r}')$. In this work, we use the fourth-order weighting function, which in two spatial dimensions has the form (Morris et al., 1997)

$$W(|\mathbf{r}|, h) = \frac{189}{478\pi h^2} \tilde{W}^{(4)}(|\mathbf{r}|, h), \quad (8)$$

where

$$\tilde{W}^{(4)}(|\mathbf{r}|, h) = \begin{cases} \left(3 - \frac{3|\mathbf{r}|}{h}\right)^5 - 6\left(2 - \frac{3|\mathbf{r}|}{h}\right)^5 + 15\left(1 - \frac{3|\mathbf{r}|}{h}\right)^5 & 0 \leq |\mathbf{r}| < \frac{1}{3}h \\ \left(3 - \frac{3|\mathbf{r}|}{h}\right)^5 - 6\left(2 - \frac{3|\mathbf{r}|}{h}\right)^5 & \frac{1}{3}h \leq |\mathbf{r}| < \frac{2}{3}h \\ \left(3 - \frac{3|\mathbf{r}|}{h}\right)^5 & \frac{2}{3}h \leq |\mathbf{r}| < h \\ 0 & |\mathbf{r}| \geq h. \end{cases} \quad (9)$$

158 The number density n_j (the number of SPH particles within volume h^d
 159 centered at $\mathbf{r}_j$) and h define the resolution of the SPH discretization scheme.
 160 For a sufficiently smooth A , the integral approximation (7) is second-order
 161 accurate in h . Approximating the integral in Eq (7) with the sum introduces
 162 an additional error in the SPH discretization scheme that scales as $n^{-1/d}$
 163 (Quinlan et al., 2006).

164 The SPH discretization scheme (7) can be used to discretize the continuity
 165 equation (1). Alternatively, density $m_i n_i$ can be found directly from Eq (7)
 166 by replacing A_j with n_j :

$$n_i = \sum_{j=1}^N W(|\mathbf{r} - \mathbf{r}_j|, h). \quad (10)$$

167 The PF-SPH discretization of the momentum conservation equation (2),
 168 subject to the boundary condition (4), is given as:

$$\frac{D(m_i \mathbf{v}_i)}{Dt} = \mathbf{F}_i^P + \mathbf{F}_i^\mu + \mathbf{F}_i^{int} + \mathbf{F}^b. \quad (11)$$

169 Here, $\mathbf{F}^b$ is the body force and $\mathbf{F}_i^P$ and $\mathbf{F}_i^\mu$ are the “pressure gradient” and
 170 “viscous” forces given by an SPH discretization of the corresponding terms
 171 in the momentum conservation equation, and the subscript α is omitted for

172 convenience. In multiphase fluid flows, wetting and non-wetting behaviors at
 173 the fluid-fluid-solid interface and pressure jump across the fluid-fluid bound-
 174 ary are a physical consequence of interactions between molecules of different
 175 fluid and solid phases. In the PF-SPH model, these physical phenomena are
 176 modeled by the molecular-like pairwise interaction forces,

$$\mathbf{F}_i^{int} = \sum_{j=1}^N \mathbf{F}_{ij}^{int} \quad \text{and} \quad \mathbf{F}_{ij}^{int} = -F_{\alpha\beta}^{int}(r_{ij}) \frac{\mathbf{r}_{ij}}{r_{ij}}, \quad (12)$$

177 acting between particle i of the α phase and particle j of the β phase, where
 178 β can be a fluid or solid phase. In the above equations, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ and
 179 $r_{ij} = |\mathbf{r}_{ij}|$. Like the pairwise molecular forces, the interaction forces $\mathbf{F}_{ij}^{int}$
 180 should be short-range repulsive ($F_{\alpha\beta}^{int}(r_{ij} \leq r^*) < 0$, $r^* < h$) and long-
 181 range attractive ($F_{\alpha\beta}^{int}(r^* < r_{ij} \leq h) > 0$). For computational efficiency, the
 182 interaction force between two particles should be zero (or decay rapidly) for
 183 $r_{ij} \geq h$. The SPH equations are closed with an equation of state

$$P_\alpha = f_\alpha(n_\alpha). \quad (13)$$

184 The SPH discretization scheme (7) can be used to obtain $\mathbf{F}_i^P$ and $\mathbf{F}_i^\mu$
 185 in different forms (Monaghan, 2005). Since our focus is on the surface ten-
 186 sion, which is not affected by the viscous force, here we consider only one
 187 form of $\mathbf{F}_i^\mu$, which is commonly used to model incompressible and weakly
 188 compressible fluids (Morris et al., 1997):

$$\mathbf{F}_i^\mu = \sum_{j=1}^N \mathbf{F}_{ij}^\mu, \quad \mathbf{F}_{ij}^\mu = \frac{4\mu_i\mu_j}{\mu_i + \mu_j} \frac{\mathbf{v}_{ij}}{n_i n_j r_{ij}} \frac{dW(r_{ij}, h)}{dr_{ij}}, \quad (14)$$

189 where $\mathbf{v}_{ij} = \mathbf{v}_i - \mathbf{v}_j$.

190 Two common forms of

$$\mathbf{F}_i^P = \sum_{j=1}^N \mathbf{F}_{ij}^P \quad (15)$$

191 are

$$\mathbf{F}_{ij}^P = - \left(\frac{P_j}{n_j^2} + \frac{P_i}{n_i^2} \right) \frac{\mathbf{r}_{ij}}{r_{ij}} \frac{dW(r_{ij}, h)}{dr_{ij}} \quad (16)$$

192 and

$$\mathbf{F}_{ij}^P = - \frac{P_i + P_j}{n_i n_j} \frac{\mathbf{r}_{ij}}{r_{ij}} \frac{dW(r_{ij}, h)}{dr_{ij}}. \quad (17)$$

193 It can be seen that for incompressible flows ($n_i = n_j = \text{const}$) or for
 194 (weakly) compressible flows with the ideal gas equation of state $P_i = cn_i$,
 195 these two expressions are equivalent. On the other hand, for compressible
 196 fluids with non-linear equations of state, these two expressions may produce
 197 different results. In Section 5.2 we show that Eq (16) produces an artificial
 198 surface tension. Therefore, in any SPH multiphase model (including the PF-
 199 SPH model), it is preferable to discretize the pressure gradient term using
 200 Eq (17).

201 The PF-SPH model (11) was originally proposed in Tartakovsky and
 202 Meakin (2006) with $F_{\alpha\beta}^{int}$ in the form:

$$F_{1,\alpha\beta}^{int}(r_{ij}) = \begin{cases} -s_{\alpha\beta} \cos\left(\frac{3\pi}{2h} r_{ij}\right) & r_{ij} \leq h \\ 0 & r_{ij} > h, \end{cases} \quad (18)$$

203 where $s_{\alpha\beta}$ is the strength of the interaction force between particles of phases α
 204 and β . The conservation of linear momentum requires $s_{\alpha\beta} = s_{\beta\alpha}$. Under this
 205 condition, the PF-SPH momentum equation (11) conserves linear momentum
 206 exactly. The immiscible behavior of the non-wetting and wetting phases is
 207 achieved by setting $s_{\alpha\alpha} > s_{\alpha\beta}$ and $s_{\beta\beta} > s_{\alpha\beta}$. The wetting behavior of fluids
 208 is determined by the relative values of $s_{\alpha\alpha}$, $s_{\beta\beta}$, $s_{\alpha\beta}$, $s_{s\beta}$ and $s_{s\alpha}$. Here, $s_{s\alpha}$

209 and $s_{0.2\beta}$ are the strength of the pairwise forces acting between particles of
 210 the α fluid and solid and the β fluid and solid, respectively. For the β fluid
 211 to wet the surface in the presence of the α fluid (for the static contact angle
 212 to be less than 90 degrees), the following condition should be satisfied:

$$s_{s\beta}/s_{\beta\beta} > 1, \quad s_{s\alpha}/s_{\alpha\alpha} < 1. \quad (19)$$

213 In Kordilla et al. (2013), $F_{\alpha\beta}$ was represented as a combination of two
 214 SPH weighting functions:

$$F_{4,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta} \left[-A\tilde{W}(r_{ij}, h^*) + \tilde{W}(r_{ij}, h) \right], \quad (20)$$

215 where A defines the ratio of magnitudes of the repulsive and attractive parts
 216 of the force $\mathbf{F}_{ij}^{int}$ and h^* and h ($h^* < h$) determine ranges of the repulsive
 217 and attractive parts of the force, respectively. In this paper we consider
 218 four different forms of $F_{\alpha\beta}^{int}$, which are listed in Table 1. In Section 5.3, we
 219 demonstrate that the form of $\mathbf{F}_{ij}^{int}$ and the choice of the parameters affect
 220 the distribution of SPH particles. The particle distribution is an important
 221 consideration in any variant of the SPH method, as it affects the accuracy of
 222 the SPH discretization.

223 There are several approaches in SPH to impose the no-slip boundary
 224 condition at the fluid-solid boundary away from the fluid-fluid-solid interface.
 225 In this work, we approximately impose the no-slip boundary condition by
 226 assigning zero velocity to the solid particles and including the solid particles
 227 in evaluation of the $\mathbf{F}_i^P$, $\mathbf{F}_i^\mu$ and $\mathbf{F}_i^{int}$ forces acting on the fluid particles. The
 228 particle density of the solid particles is evaluated in the same way as the
 229 density of fluid particles (i.e. from Eq(10)). When computing $\mathbf{F}_i^P$, the EOS
 230 of solid particle j is assumed to be the same as the EOS of fluid particle i .

231 In the expression of $\mathbf{F}_i^\mu$, the viscosity of solid particle j is assumed to be the
 232 same as the viscosity of fluid particle i . For single-phase flows, more accurate
 233 implementations of the no-slip BC have been proposed (Morris et al., 1997;
 234 Holmes et al., 2011). Extension of these methods for the multiphase PF-SPH
 235 method is a subject of our future research.

236 We integrate the SPH equations using the “velocity Verlet” algorithm
 237 (Allen and Tildesley, 1989; Tartakovsky and Meakin, 2005a,b; Kordilla et al.,
 238 2013):

$$\mathbf{r}_i(t + \Delta t) = \mathbf{r}_i(t) + \Delta t \mathbf{v}_i(t) + \frac{\Delta t^2}{2m_i} \mathbf{f}_i(t) \quad (21)$$

$$\mathbf{v}_i(t + \Delta t) = \mathbf{v}_i(t) + \frac{\Delta t}{2m_i} [\mathbf{f}_i(t) + \mathbf{f}_i(t + \Delta t)]. \quad (22)$$

239 Here, $\mathbf{f}_i(t + \Delta t) = \mathbf{F}_i^P(t + \Delta t) + \mathbf{F}_i^\mu(t + \Delta t) + \mathbf{F}_i^{int}(t + \Delta t) + \mathbf{g}$, $\mathbf{F}_i^P(t + \Delta t)$ and
 240 $\mathbf{F}_i^{int}(t + \Delta t)$ are computed using particle positions at time $(t + \Delta t)$, $\mathbf{F}_i^\mu(t + \Delta t)$
 241 is found using particle positions at time $(t + \Delta t)$ and velocities at time t , and
 242 the time step Δt satisfies the constraints (Morris et al., 1997)

$$\begin{aligned} \Delta t &\leq 0.25 \min_i \left(\frac{h}{3|\mathbf{v}_i|} \right), \\ \Delta t &\leq 0.25 \min_i \left(\sqrt{\frac{m_i h}{3|\mathbf{f}_i|}} \right), \\ \Delta t &\leq 0.25 \min_i \left(\frac{\rho_i h^2}{9\mu} \right). \end{aligned} \quad (23)$$

243 In the past, the PF-SPH multiphase model has been validated under both
 244 static and dynamic conditions for different forms of $\mathbf{F}_{ij}^{int}$ (Tartakovsky and
 245 Meakin, 2006; Kordilla et al., 2013). The first step in these validation studies
 246 was to determine the surface tension and the static contact angle for a pre-
 247 scribed set of parameters in $\mathbf{F}_{ij}^{int}$. The surface tension was determined from

248 the Young-Laplace equation by simulating a bubble of one fluid surrounded
 249 by another fluid. The static contact angle was measured by simulating a
 250 bubble resting on a horizontal surface. In the second step of the validation
 251 studies, the two-phase flow was simulated under dynamic conditions and the
 252 results were compared with the corresponding analytical solutions and/or
 253 experimental results. It was also shown that the PF-SPH model is able to
 254 model multiphase flows with density ratios as large as 1000 and viscosity
 255 ratios as large as 100 (Tartakovsky et al., 2007). Until now, the main dis-
 256 advantage of the PF-SPH multiphase model was the need to calibrate the
 257 parameters in $\mathbf{F}_{ij}^{int}$ with respect to surface tension and static contact angle.

258 In Section 4, we demonstrate that the parameters in $\mathbf{F}_{ij}^{int}$ can be related
 259 to macroscopic parameters such as σ and θ_0 via closed-form analytical ex-
 260 pressions. Therefore, the validation cases presented in this paper mainly
 261 focus on testing these analytical expressions. We also demonstrate that the
 262 PF-SPH model is able to reproduce the Tanner Law for a bubble spreading
 263 on the surface. In addition, we study flow of a droplet between two parallel
 264 plates to demonstrate that the resulting velocity and contact angles satisfy
 265 the analytical solution. We show that the receding and advancing contact
 266 angles change as a power of the droplet velocity, the behavior also observed in
 267 experiments. In the last section, we present simulations of three-phase flow
 268 in porous media. The latter numerical examples demonstrate robustness of
 269 the PF-SPH model, i.e. these simulations show that the PF-SPH model is
 270 able to simulate complex flows in domains with complex geometry.

271 4. Direct calculation of surface tension from the total stress

272 In SPH, as in any Lagrangian particle model, the total stress in the fluid
 273 at point $\mathbf{x}$ can be found according to the Hardy formula (Hardy, 1982):

$$\mathbf{T}(\mathbf{x}) = \mathbf{T}_{(c)}(\mathbf{x}) + \mathbf{T}_{(int)}(\mathbf{x}), \quad (24)$$

274 where $\mathbf{T}_{(c)}(\mathbf{x})$ is the convection stress,

$$\mathbf{T}_{(c)}(\mathbf{x}) = - \sum_{j=1}^N m_j (\bar{\mathbf{v}}(\mathbf{x}) - \mathbf{v}_j) \otimes (\bar{\mathbf{v}}(\mathbf{x}) - \mathbf{v}_j) \tilde{\psi}_\eta(\mathbf{x} - \mathbf{r}_j), \quad (25)$$

275 and $\mathbf{T}_{(int)}(\mathbf{x})$ is the interaction stress:

$$\mathbf{T}_{(int)}(\mathbf{x}) = \frac{1}{2} \sum_{i=1}^N \sum_{j=1}^N \mathbf{f}_{ij} \otimes (\mathbf{r}_j - \mathbf{r}_i) \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - s\mathbf{r}_i - (1-s)\mathbf{r}_j) ds, \quad (26)$$

276 where $\bar{\mathbf{v}}(\mathbf{x}) = \sum_j m_j \mathbf{v}_j \tilde{\psi}_\eta(\mathbf{x} - \mathbf{r}_j) \left(\sum_j m_j \tilde{\psi}_\eta(\mathbf{x} - \mathbf{r}_j) \right)^{-1}$ is the average veloc-
 277 ity and $\mathbf{f}_{ij}$ is the total force acting on the particles. In the absence of gravity,
 278 in the PF-SPH model this force is given as

$$\mathbf{f}_{ij} = \mathbf{F}_{ij}^P + \mathbf{F}_{ij}^\mu + \mathbf{F}_{ij}^{int}. \quad (27)$$

279 The summation here is over all particles, and $\otimes$ denotes dyadic product
 280 of vectors. The choice of the weighting function $\tilde{\psi}(\mathbf{x})$ is fairly arbitrary,
 281 and here we assume that $\tilde{\psi}(\mathbf{x})$ is the product of one-dimensional functions
 282 $\psi_{\eta,l} = \frac{1}{\eta} \psi(\mathbf{x}_{(l)})$, where $l = 1, 2, 3$ denotes a component of the vector. The
 283 function $\psi_{\eta,l}(\mathbf{x})$ has the compact support η , or becomes sufficiently small for
 284 $|\mathbf{r}| > \eta$. The support η should be reater than h and smaller than the size of
 285 the domain. In our calculations, we set $\eta = 2h$. At the static equilibrium
 286 ($\mathbf{v}_i = 0$ for $i = 1, \dots, N$), $\mathbf{T}_{(c)}(\mathbf{x}) = 0$ and $\mathbf{T}(\mathbf{x})$ reduces to $\mathbf{T}(\mathbf{x}) = \mathbf{T}_{(int)}(\mathbf{x})$.

287 In a bulk fluid (away from the fluid-fluid interface), $\mathbf{T}(\mathbf{x})$ becomes a diagonal
 288 tensor with the diagonal components $T_{xx} = T_{yy} = T_{zz} = -p$, where p is the
 289 “total” pressure. It should be noted that in the PF-SPH model, p is the sum
 290 of P and the virial pressure generated by the $\mathbf{F}_{ij}^{int}$ forces.

291 Consider two fluid phases α and β separated by a flat interface perpen-
 292 dicular to the z direction. From symmetry considerations, the off-diagonal
 293 components of $\mathbf{T}$ are zero, the diagonal components depend only on z , and
 294 $T_{xx}(z) = T_{yy}(z) = T_{\tau}(z)$. Then, based on the Gibbs treatment of thermody-
 295 namics, the surface tension can be found as (Rowlinson and Widom, 2002)

$$\sigma_{\alpha\beta} = \int_{-\infty}^{+\infty} [T_{\tau}(z) - T_n(z)] dz, \quad (28)$$

296 where $T_n(z) = T_{zz}(z)$ and $T_{\tau}(z)$ are the normal and tangent components
 297 of the stress. In a two-dimensional space for two fluids separated by a flat
 298 interface parallel to the x direction, similar arguments yield Eq. (28) with

$$T_n(y) = T_{yy} \quad \text{and} \quad T_{\tau}(y) = T_{xx}(y). \quad (29)$$

299 Fig 1 shows components of the stress tensor obtained from an SPH sim-
 300 ulation of two fluids separated by a flat front located at $y = 16h$ after the
 301 fluid system reached equilibrium. In this simulation, at time zero, the SPH
 302 particles are randomly placed in a domain $16h \times 32h$ with the average par-
 303 ticle density $n_{eq} = 36$, which is achieved by dividing the domain into $1h$ by
 304 $1h$ square subdomains and randomly placing 36 particles in each subdomain.
 305 The SPH particles in the lower half of the domain (with the y -coordinate less
 306 than $16h$) are labeled as α particles and the rest of the particles are labeled
 307 as β particles. Then, the positions of particles are evolved according to the

Figure 1: T_{xx} and T_{yy} components of stress as a function of y obtained from a simulation of two fluids separated by flat interface located at $y = 16$. In the simulation, the pairwise force $F_{2,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta} [-A\psi_{\varepsilon_0}(r_{ij}) + \psi_{\varepsilon}(r_{ij})]$ is used with $s_{ww} = s_{nn} = 2$, $n_w = n_n = 36$, $s_{nw} = 0.00001$, $\varepsilon = h/3.5$, $\varepsilon_0 = \varepsilon/2$ and $A = \varepsilon^2/\varepsilon_0^2$.

SPH momentum conservation equation. Fig 1(a) displays the components of $\mathbf{T}(x = 8, y)$ and Fig 1(b) shows the components of $\mathbf{T}(x, y)$ averaged over the x direction, $\bar{\mathbf{T}}(y) = \frac{1}{N} \sum_{i=1}^N \mathbf{T}(x_i, y)$. The stress is computed using the two-dimensional version of Eq (26) after the the system reached equilibrium. Due to particle disorder, both components of $\mathbf{T}(\mathbf{x})$ vary with y and, as follows from the equilibrium condition $\nabla \cdot \mathbf{T} = 0$, x , which violates the condition (29). This means that on the particle level the interface is not flat. On the other hand, the components of the average stress $\bar{\mathbf{T}}(\mathbf{x})$ satisfy the condition for stress at a flat interface; i.e., $\bar{T}_{yy}$ is constant and $\bar{T}_{xx}$ depends only on y . Then, the surface tension is equal to the area between the two curves in Fig 1(b).

Next, we simulate a two-dimensional bubble of the fluid α surrounded by the fluid β using the same parameters as in the simulation with a flat front described above. The bubble is located in the center of a $32h \times 32h$ domain,

Figure 2: Tangent (T_{xx}) and normal (T_{yy}) components of stress as a function of the radial coordinate ($y - 8$) obtained from a simulation of a droplet of fluid α separated by fluid β . The center of the two-dimensional droplet is at $x = y = 16$ and the radius is equal to 8. In the simulation, the pairwise force $F_{2,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta} [-A\psi_{\varepsilon_0}(r_{ij}) + \psi_{\varepsilon}(r_{ij})]$ is used with $s_{ww} = s_{nn} = 2$, $n_w = n_n = 36$, $s_{nw} = 0.00001$, $\varepsilon = h/3.5$, $\varepsilon_0 = \varepsilon/2$ and $A = \varepsilon^2/\varepsilon_0^2$.

322 and has radius $8h$. Fig 2 shows the stress components $T_{xx}(x = 16, y) =$
323 $T_{\tau}(x = 16, y)$ and $T_{yy}(x = 16, y) = T_n(x = 16, y)$ as a function of y after the
324 system reached equilibrium. The T_{yy} -component at point $(x = 16, y = 24)$ is
325 normal to the interface and, therefore, is equal to the negative total pressure
326 $-p$. It can be seen that p changes gradually across the interface from the
327 larger value inside the bubble to the smaller value outside of the bubble. The
328 transition zone is on the order of 2η . As before, the surface tension is given
329 by the area between the T_{xx} and T_{yy} curves.

330 In Appendix A we perform an analytical integration in Eq (28) to obtain
331 closed-form relationships between the surface tension and parameters in the
332 SPH forces.

333 5. Relationship between the pairwise forces and macroscopic pa- 334 rameters

335 5.1. Virial pressure due to pairwise forces

336 Under static conditions, the force $\mathbf{f}_{ij}$ acting between particle i of phase α
337 and particle j of phase β can be written as:

$$\mathbf{f}_{ij} = \phi_{\alpha\beta}(r_{ij}) \frac{\mathbf{r}_{ij}}{r_{ij}} = -[F_{\alpha\beta}^P(r_{ij}) + F_{\alpha\beta}^{int}(r_{ij})] \frac{\mathbf{r}_{ij}}{r_{ij}}, \quad (30)$$

338 where $\phi_{\alpha\beta}(r_{ij}) = -[F_{\alpha\beta}^P(r_{ij}) + F_{\alpha\beta}^{int}(r_{ij})]$ and $\phi_{\alpha\beta}^P$ is given, depending on the
339 choice of discretization, by:

$$F_{\alpha\beta}^P(z) = \left(\frac{P_\alpha}{n_\alpha^2} + \frac{P_\beta}{n_\beta^2} \right) \frac{dW(z)}{dz}, \quad (31)$$

340 OR

$$F_{\alpha\beta}^P(z) = \frac{P_\alpha + P_\beta}{n_\alpha n_\beta} \frac{dW(z)}{dz}. \quad (32)$$

341 In Appendix B, we demonstrate that away from the interface (at the
342 normal distance from the interface greater than η), the stress component
343 $\mathbf{T}_{(int)}$ is a diagonal tensor $\mathbf{T}_{(int)} = -p\mathbf{I}$, where p is the total pressure that
344 can be found analytically, by approximating the double sum in Eq (26) with
345 the double integral, as:

$$p_\alpha = -\frac{2}{3}\pi n_\alpha n_\alpha \int_0^\infty z^3 [F_{\alpha\alpha}^P(z) + F_{\alpha\alpha}^{int}(z)] dz \quad (33)$$

346 in three spatial dimensions, and

$$p_\alpha = -\frac{1}{2}\pi n_\alpha n_\alpha \int_0^\infty z^2 [F_{\alpha\alpha}^P(z) + F_{\alpha\alpha}^{int}(z)] dz \quad (34)$$

347 in two dimensions. Here, the subscript α emphasizes that p_α is the pressure
 348 in the α phase. It can be easily verified that

$$-\frac{2}{3}\pi n_\alpha n_\alpha \int_0^\infty z^3 F_{\alpha\alpha}^P(z) dz = P_\alpha \quad (35)$$

349 in three spatial dimensions and

$$-\frac{1}{2}\pi n_\alpha n_\alpha \int_0^\infty z^2 F_{\alpha\alpha}^P(z) dz = P_\alpha \quad (36)$$

350 in two spatial dimensions. The last two expressions show that the pressure
 351 generated by the forces $\mathbf{F}_i^P$ is exactly equal to the pressure prescribed by the
 352 EOS, thus confirming the consistency of Eqs (33) and (34). In the following,
 353 we refer to the pressure generated by forces $\mathbf{F}_{ij}^{int}$ as the virial pressure.

354 Table 1 gives functional expressions for the virial pressure generated by
 355 various forms of $\mathbf{F}_{ij}^{int}$ in two and three spatial dimensions. The virial pressure
 356 generated by $F_{1,\alpha\beta}^{int}$ is always negative, regardless of the choice of parameters.
 357 The virial pressure due to the pairwise forces with $F_{k,ij}^{int}$, $k = 2, 3, 4$ in Table 1
 358 (which are given by a combination of two Gaussian or polynomial functions),
 359 can be positive or negative and can be eliminated by choosing a certain
 360 value of the parameter A . For example, in two spatial dimensions, the virial
 361 pressure generated by $F_{2,ij}^{int}$ can be eliminated by setting $A = \varepsilon^4/\varepsilon_0^4$. At the
 362 same time, the parameter A affects the distribution of the SPH particles, both
 363 in the bulk and near the fluid-fluid interface. Our simulations show that a
 364 negative virial pressure is needed to generate a stable interface between two
 365 fluids and a uniform particle distribution. Table 1 lists the values of the
 366 parameter A used in our simulations.

Table 1: Virial pressure, $K_{\alpha\alpha} = -\xi n_\alpha^2 s_{\alpha\alpha}$, and specific energy, $T_{\alpha\beta} = \lambda n_\alpha n_\beta s_{\alpha\beta}$, due to different pairwise forces $\mathbf{F}_{ij}^{int} = -F_{k,\alpha\beta}^{int}(r_{ij}) \frac{\mathbf{r}_{ij}}{r_{ij}}$ ($k = 1, 2, 3, 4$) in two and three spatial dimensions. In $F_{2,\alpha\beta}^{int}$ and $F_{3,\alpha\beta}^{int}$, $\psi_\epsilon(r_{ij}) = e^{-\frac{r_{ij}^2}{2\epsilon^2}}$, $\epsilon = h/3.5$ and $\psi_{\epsilon_0}(r_{ij}) = e^{-\frac{r_{ij}^2}{2\epsilon_0^2}}$.

$$F_{1,\alpha\beta}^{int}(r_{ij}) = -s_{\alpha\beta} \cos\left(\frac{3\pi}{2h} r_{ij}\right) \text{ if } 0 \leq r_{ij} \leq h; F_{1,\alpha\beta}^{int}(r_{ij}) = 0 \text{ if } r_{ij} > h$$

$$3\text{D: } \xi = \frac{16}{81} \frac{h^4}{\pi^3} \left[\frac{9}{4} \pi^3 - 6\pi - 4 \right]$$

$$2\text{D: } \xi = \frac{h^3(-8+9\pi^2)}{27\pi^2}$$

$$3\text{D: } \lambda = \frac{3}{4\pi^4} [2^7 - 3^2 \times 2^4 \pi^2 + 3^3 \pi^4] \left(\frac{h}{3}\right)^5$$

$$2\text{D: } \lambda = \frac{8}{81} \frac{h^4}{\pi^4} \left[\frac{9}{4} \pi^3 - 6\pi - 4 \right]$$

$$F_{2,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta} [-A\psi_{\epsilon_0}(r_{ij}) + \psi_\epsilon(r_{ij})], \epsilon/\epsilon_0 = 2$$

$$3\text{D: } A = (\epsilon/\epsilon_0)^3, \text{ and } 2\text{D: } A = (\epsilon/\epsilon_0)^2$$

$$3\text{D: } \xi = \frac{4\pi}{3} [-A\epsilon_0^4 + \epsilon^4]$$

$$2\text{D: } \xi = \frac{\pi\sqrt{\pi}}{2\sqrt{2}} [-A\epsilon_0^3 + \epsilon^3]$$

$$3\text{D: } \lambda = (2\pi)^{3/2} \frac{3}{32} (-A\epsilon_0^5 + \epsilon^5)$$

$$2\text{D: } \lambda = \frac{2}{3} (-A\epsilon_0^4 + \epsilon^4).$$

$$F_{3,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta} r_{ij} [-A\psi_{\epsilon_0}(r_{ij}) + \psi_\epsilon(r_{ij})], \epsilon/\epsilon_0 = 2$$

$$3\text{D: } A = (\epsilon/\epsilon_0)^4, \text{ and } 2\text{D: } A = (\epsilon/\epsilon_0)^3$$

$$3\text{D: } \xi = \sqrt{2}\pi^{3/2} [-A\epsilon_0^5 + \epsilon^5]$$

$$2\text{D: } \xi = \pi [-A\epsilon_0^4 + \epsilon^4]$$

$$3\text{D: } \lambda = \pi (-A\epsilon_0^6 + \epsilon^6)$$

$$2\text{D: } \lambda = \frac{1}{2} \sqrt{2\pi} (-A\epsilon_0^5 + \epsilon^5)$$

$$F_{4,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta} \left[-A\tilde{W}_{h_0}^{(4)}(r_{ij}) + \tilde{W}_h^{(4)}(r_{ij}) \right], h/h_0 = 2$$

$$3\text{D: } A = (h/h_0)^3, \text{ and } 2\text{D: } A = (h/h_0)^2$$

$$3\text{D: } \xi = \frac{2771}{10206} \pi [-Ah_0^4 + h^4]$$

$$2\text{D: } \xi = \frac{5}{9} \pi [-Ah_0^3 + h^3]$$

$$3\text{D: } \lambda = \frac{7}{324} \pi (-Ah_0^5 + h^5)$$

$$2\text{D: } \lambda = \frac{2771}{20421} (-Ah_0^4 + h^4)$$

367 *5.2. Surface tension and contact angles*

368 Approximating double summation in the definition of stress $\mathbf{T}$ with dou-
 369 ble integral, substituting this in the expression (28) for $\sigma_{\alpha\beta}$, and integrating
 370 the latter analytically, we demonstrate that the surface tension between fluids
 371 α and β can be found as (Appendix A):

$$\sigma_{\alpha\beta} = \mathcal{T}_{\alpha\alpha} + \mathcal{T}_{\beta\beta} - 2\mathcal{T}_{\alpha\beta}, \quad (37)$$

372 where $\mathcal{T}_{\alpha\beta}$ is the specific interfacial energy between α and β fluid phases and
 373 $\mathcal{T}_{\alpha\alpha}$ and $\mathcal{T}_{\beta\beta}$ are the specific energies in α and β phases, respectively.

374 For three fluids in contact with each other, the surface tensions between
 375 each pair of fluids and contact angles, formed by the boundaries of each fluid,
 376 satisfy at the equilibrium the following conditions:

$$\begin{aligned} \sigma_{13}\cos\theta_1 + \sigma_{23}\cos\theta_2 + \sigma_{12} &= 0 \\ \sigma_{12}\cos\theta_1 + \sigma_{23}\cos\theta_3 + \sigma_{13} &= 0 \\ \sigma_{12}\cos\theta_2 + \sigma_{13}\cos\theta_3 + \sigma_{23} &= 0. \end{aligned}$$

377 Two fluids in contact with the solid surface form the contact angle, an
 378 angle between the solid surface and fluid-fluid interface bounding the α -fluid.
 379 At the equilibrium, this angle is commonly referred to as the static contact
 380 angle, θ_0 , and is given by Young's equation (5).

381 In Appendix A, we derive an expression for $\mathcal{T}_{\alpha\beta}$ in terms of $\phi_{\alpha\beta}$. In three
 382 dimensions $\mathcal{T}_{\alpha\beta}$ is given by

$$\mathcal{T}_{\alpha\beta} = -\frac{1}{8}\pi\rho_\alpha\rho_\beta \int_0^\infty z^4 \phi_{\alpha\beta}(z) dz, \quad (\alpha, \beta) = n, w, s, \quad (38)$$

383 and in two dimensions, by

$$\mathcal{T}_{\alpha\beta} = -\frac{1}{3}\rho_\alpha\rho_\beta \int_0^\infty z^3 \phi_{\alpha\beta}(z)dz, \quad (\alpha, \beta) = n, w, s. \quad (39)$$

384 In the derivation of $\mathcal{T}_{\alpha\beta}$, we assume that the radii of the front curvature are
 385 much larger than h (i.e., the front can be locally approximated as flat) and the
 386 thickness of each phase (the dimension in the direction normal to the inter-
 387 face) is larger than h , the range of interaction of the pairwise forces. For the
 388 radii of the front curvature larger than h , the surface tension should be inde-
 389 pendent on the front curvature, and, therefore, Eq (39) should hold for fronts
 390 with with any well-resolved front curvature. It is also assumed that the num-
 391 ber density within each phase is constant. For the three-dimensional space,
 392 our expression is identical to that of Maxwell (1890), who considered fluids
 393 made of particles which interact with each other via pairwise forces. There-
 394 fore, in Appendix A, we present derivations for only the two-dimensional
 395 problem.

396 In the following, we calculate $\mathcal{T}_{\alpha\beta}$ under static conditions ($\mathbf{v}_i = [0, 0, 0]^T$),
 397 as this allows us to derive an analytical closed-form expression for $\mathcal{T}_{\alpha\beta}$ as a
 398 function of the parameters in $\mathbf{F}_{ij}^{int}$. Since the surface tension is the same under
 399 both dynamic and static conditions, the resulting relationship between the
 400 surface tension and parameters in the SPH model is applicable under both
 401 the dynamic and static conditions.

402 We can write $\mathcal{T}_{\alpha\beta}$ as

$$\mathcal{T}_{\alpha\beta} = \mathcal{T}_{\alpha\beta}^P + \mathcal{T}_{\alpha\beta}^{int}, \quad (\alpha, \beta) = n, w, s, \quad (40)$$

403 where $\mathcal{T}_{\alpha\beta}^P$ is the component of energy due to the $\mathbf{F}_{ij}^P$ forces and $\mathcal{T}_{\alpha\beta}^{int}$ is
 404 the component of energy due to the $\mathbf{F}_{ij}^{int}$ forces. Furthermore, we write the

405 surface tension as

$$\sigma_{\alpha\beta} = \sigma_{\alpha\beta}^P + \sigma_{\alpha\beta}^{int}, \quad (41)$$

406 where $\sigma_{\alpha\beta}^P$ and $\sigma_{\alpha\beta}^{int}$ are the contributions to the surface tension from the
407 pressure gradient and pairwise forces, respectively, and are given by

$$\sigma_{\alpha\beta}^P = \mathcal{T}_{\alpha\alpha}^P + \mathcal{T}_{\beta\beta}^P - 2\mathcal{T}_{\alpha\beta}^P \quad \text{and} \quad \sigma_{\alpha\beta}^{int} = \mathcal{T}_{\alpha\alpha}^{int} + \mathcal{T}_{\beta\beta}^{int} - 2\mathcal{T}_{\alpha\beta}^{int}. \quad (42)$$

408 Substituting Eqs (31) and (32) into Eq (38) in three dimensions and (39)
409 in two dimensions yields the following expressions for the partial surface
410 energy due the pressure gradient force:

$$\mathcal{T}_{\alpha\beta}^P = \gamma n_\alpha n_\beta \left(\frac{P_\alpha}{n_\alpha^2} + \frac{P_\beta}{n_\beta^2} \right) \quad (43)$$

411 OR

$$\mathcal{T}_{\alpha\beta}^P = \gamma(P_\alpha + P_\beta) \quad (44)$$

412 depending on the SPH discretization of the pressure gradient. Here $\gamma =$
413 $\frac{1}{8}\pi \int_0^\infty z^4 \frac{dW}{dz} dz$ and $\gamma = \frac{1}{3} \int_0^\infty z^3 \frac{dW}{dz} dz$ in three and two dimensions, respec-
414 tively. Note that $\gamma < 0$ in both two and three dimensions.

415 The partial surface energy due to the interaction force is given by

$$\mathcal{T}_{\alpha\beta}^{int} = \frac{1}{8}\pi n_\alpha n_\beta \int_0^\infty z^4 F_{\alpha\beta}^{int}(z) dz = \lambda n_\alpha n_\beta s_{\alpha\beta}, \quad (45)$$

416 and

$$\mathcal{T}_{\alpha\beta}^{int} = \frac{1}{3}n_\alpha n_\beta \int_0^\infty z^3 F_{\alpha\beta}^{int}(z) dz = \lambda n_\alpha n_\beta s_{\alpha\beta} \quad (46)$$

417 in three and two spatial dimensions, respectively. The parameter λ depends
418 on the range and specific form of $F_{\alpha\beta}^{int}$ and the number of spatial dimensions.

419 Knowing the “partial” surface energies, the “partial” surface tensions due
 420 to $\mathbf{F}_{ij}^P$ and $\mathbf{F}_{ij}^{int}$ can be found. In the following we consider contribution of
 421 partial surface tensions, $\sigma_{\alpha\beta}^P$ and $\sigma_{\alpha\beta}^F$, to the “total” surface tension, $\sigma_{\alpha\beta}$,
 422 starting with $\sigma_{\alpha\beta}^P$. First, we note that for incompressible fluids $\sigma_{\alpha\beta}^P$ based on
 423 Eq (43) is

$$\sigma_{\alpha\beta}^P = -2\gamma(P_\alpha + P_\beta - \frac{n_\beta}{n_\alpha}P_\alpha - \frac{n_\alpha}{n_\beta}P_\beta) \quad (47)$$

424 and $\sigma_{\alpha\beta}^P$ based on Eq (44) is

$$\sigma_{\alpha\beta}^P = 0. \quad (48)$$

425 The pressure gradient term in the Navier-Stokes equations does not gener-
 426 ate the surface tension (the surface tension should be prescribed, for example,
 427 via the Young-Laplace boundary condition) and the SPH discretization (17)
 428 explicitly enforces this condition. On the other hand, the SPH discretization
 429 (16) produces “numerical” surface tension if different resolution n is used
 430 to discretize different fluid phases. Therefore, in the multiphase SPH flow
 431 models it is necessary to discretize the pressure gradient term according to
 432 Eq. (17). We should note that for compressible fluids with linear EOS such
 433 as the ideal gas EOS

$$P_i = k_b T n_i, \quad (49)$$

434 these two discretizations of the pressure gradient become identical. In this
 435 EOS, k_b is the Boltzmann constant and T is temperature.

436 The ideal gas equation of state can be modified as

$$P_i = c_{\alpha\beta} n_i. \quad (50)$$

437 Substituting the EOS (50) in Eq (44) results in the force

$$c_{\alpha\beta} \frac{n_\alpha n_\beta}{n_\alpha n_\beta} \frac{dW}{dz} \frac{\mathbf{r}_{ij}}{r_{ij}} \quad (51)$$

438 between particle i of phase α and particle j of phase β with the strength $c_{\alpha\beta}$,
 439 which can be different for interaction between particles of different phases.
 440 Since the EOS (50) produces purely repulsive forces, to generate surface
 441 tension between α and β phases one should set $c_{\alpha\beta} > c_{\alpha\alpha}$ and $c_{\alpha\beta} > c_{\beta\beta}$,
 442 resulting in the surface tension:

$$\sigma_{\alpha\beta}^P = 2\gamma(n_\alpha c_{\alpha\alpha} + n_\beta c_{\beta\beta} - n_\beta c_{\alpha\beta} - n_\alpha c_{\alpha\beta}). \quad (52)$$

443 For incompressible and weakly compressible fluids, we can assume that $n_\alpha =$
 444 $n_\beta = n_{eq}$ resulting in

$$\sigma_{\alpha\beta}^P = 2\gamma n_{eq}(c_{\alpha\alpha} + c_{\beta\beta} - 2c_{\alpha\beta}). \quad (53)$$

445 Tartakovsky et al. (2009b) proposed to use the van der Waals (vdW) EOS

$$P = \frac{k_b T n}{1 - bn} - a_{\alpha\beta} n^2 \quad (54)$$

446 to generate surface tension in the PF-SPH model. Substituting the vdW
 447 EOS into Eq (44) yields

$$\mathcal{T}_{\alpha\beta}^P = \gamma \left(\frac{k_b T n_\alpha}{1 - bn_\alpha} + \frac{k_b T n_\beta}{1 - bn_\beta} - a_{\alpha\beta}(n_\alpha^2 + n_\beta^2) \right) \quad (55)$$

448 and

$$\sigma_{\alpha\beta}^P = -2\gamma (a_{\alpha\alpha} n_\alpha^2 + a_{\beta\beta} n_\beta^2 - a_{\alpha\beta}(n_\alpha^2 + n_\beta^2)). \quad (56)$$

449 To generate surface tension with the vdW EOS (54), it is sufficient to have
 450 $a_{\alpha\alpha} > a_{\alpha\beta}$ and $a_{\beta\beta} > a_{\alpha\beta}$. In the following, we will refer to equations of
 451 state with parameters different for interaction between SPH particles of the
 452 same fluids and particles of different fluids (such as Eqs (56) and (52))
 453 as *heterogeneous* equations of state. Equations of state with parameters

454 independent of the type of SPH particles will be referred to as *homogeneous*
 455 equations of state.

456 The partial surface tension due to $\mathbf{F}_{ij}^{int}$ is given by:

$$\sigma_{\alpha\beta}^{int} = \lambda(n_{\alpha}^2 s_{\alpha\alpha} + n_{\beta}^2 s_{\beta\beta} - 2n_{\alpha}n_{\beta}s_{\alpha\beta}) = \lambda\bar{\sigma}_{\alpha\beta}, \quad (57)$$

457 where $\bar{\sigma}_{\alpha\beta} = \bar{s}_{\alpha\alpha} + \bar{s}_{\beta\beta} - 2\bar{s}_{\alpha\beta}$ and $\bar{s}_{\alpha\beta} = n_{\alpha}n_{\beta}s_{\alpha\beta}$.

458 Eqs (56) and (57) show that the surface tension in the compressible and
 459 weakly-compressible SPH multiphase models can be generated with only the
 460 “pressure gradient” forces (i.e. without the pairwise forces $\mathbf{F}_{ij}^{int}$) by using
 461 a heterogeneous EOS. There are several limitations of this approach. The
 462 pressure gradient forces may not be sufficient to generate surface tension in
 463 free-surface models (i.e. models where only liquid phase is modeled and the
 464 gas phase is disregarded) (Tartakovsky and Meakin, 2005a). In addition,
 465 to prescribe the boundary condition (5), one needs to specify heterogeneous
 466 equations of state at the fluid-solid interface. Most importantly, in weakly
 467 compressible SPH, equations of state play an important role of controlling
 468 fluid compressibility, which limits the freedom of choice of the parameters
 469 in an EOS to define the surface tension, according to, for example, Eq (52)
 470 or (56). Finally, this approach cannot be used for modeling incompressible
 471 fluids, and is only limited to the compressible and weakly compressible SPH
 472 models as it requires prescribing a heterogeneous EOS. Because of these
 473 limitations, in the PF-SPH method, it is more common to generate surface
 474 tension using the pairwise force $\mathbf{F}_{ij}^{int}$ rather than heterogeneous equations of
 475 state. In the following, we will use the homogeneous EOS (Eq (49) with
 476 constant T) in combination with the $\mathbf{F}_{ij}^{int}$ forces to generate surface tension.
 477 We demonstrated above that, for homogeneous equations of state, $\sigma_{\alpha\beta}^P = 0$

478 and, therefore,

$$\sigma_{\alpha\beta} = \sigma_{\alpha\beta}^{int}. \quad (58)$$

479 For three fluids resting in contact with each other, the contact angles as
 480 a function of the parameters in the pairwise forces can be found from Eqs
 481 (38) and (57):

$$\begin{aligned} \cos \theta_1 &= \frac{\sigma_{23}^2 - \sigma_{13}^2 - \sigma_{12}^2}{2\sigma_{13}\sigma_{12}} = \frac{\bar{\sigma}_{23}^2 - \bar{\sigma}_{13}^2 - \bar{\sigma}_{12}^2}{2\bar{\sigma}_{13}\bar{\sigma}_{12}} \\ \cos \theta_2 &= \frac{\sigma_{13}^2 - \sigma_{12}^2 - \sigma_{23}^2}{2\sigma_{23}\sigma_{12}} = \frac{\bar{\sigma}_{13}^2 - \bar{\sigma}_{12}^2 - \bar{\sigma}_{23}^2}{2\bar{\sigma}_{23}\bar{\sigma}_{12}} \\ \cos \theta_3 &= \frac{\sigma_{12}^2 - \sigma_{23}^2 - \sigma_{13}^2}{2\sigma_{23}\sigma_{13}} = \frac{\bar{\sigma}_{12}^2 - \bar{\sigma}_{23}^2 - \bar{\sigma}_{13}^2}{2\bar{\sigma}_{23}\bar{\sigma}_{13}}. \end{aligned} \quad (59)$$

482 Substituting Eq (57) into the Young-Laplace equation gives an expression
 483 for the static contact angle θ as a function of the parameters in the pairwise
 484 forces:

$$\cos \theta_0 = \frac{\mathcal{T}_{\beta\beta}^{int} - \mathcal{T}_{\alpha\alpha}^{int} - 2\mathcal{T}_{\beta s}^{int} + 2\mathcal{T}_{\alpha s}^{int}}{\mathcal{T}_{\alpha\alpha}^{int} + \mathcal{T}_{\beta\beta}^{int} - 2\mathcal{T}_{\alpha\beta}^{int}} = \frac{\bar{s}_{\beta\beta} - \bar{s}_{\alpha\alpha} - 2\bar{s}_{\beta s} + 2\bar{s}_{\alpha s}}{\bar{s}_{\alpha\alpha} + \bar{s}_{\beta\beta} - 2\bar{s}_{\alpha\beta}}. \quad (60)$$

485 Eqs (57)-(60) show that the surface tension produced by the PF-SPH
 486 model (Eqs (11) and (12)) depends on the form of $F_{\alpha\beta}^{int}$, but the equilibrium
 487 contact angles for a three-fluid system and two fluids in contact with a solid
 488 depend only on $s_{\alpha\beta}$ and the number density.

489 In general, for a given numerical resolution an infinite combination of
 490 parameters $s_{\alpha\beta}$ can result in the same values of the surface tension and
 491 contact angles. In the following, we suggest a model with a reduced number
 492 of parameters, which allows a unique parameterization of the SPH model.
 493 We also demonstrate that the PF-SPH model produces surface tension and

494 contact angles that are in good agreement with the prescribed surface tension
 495 and contact angles.

496 *5.3. Parameterization of the PF-SPH model with respect to surface tension* 497 *and static contact angle*

498 In this section we present parameterization of the SPH model with respect
 499 to the surface tension, $\sigma_{\alpha\beta}$, and the static contact angle θ_0 .

500 Eq (57) shows that for α and β fluids to be immiscible (i.e., for $\sigma_{\alpha\beta}$ to
 501 be positive), it is sufficient for $s_{\alpha\beta}$, $s_{\beta\beta}$, and $s_{\alpha\alpha}$ to satisfy the condition
 502 $\bar{s}_{\alpha\alpha} + \bar{s}_{\beta\beta} > 2\bar{s}_{\alpha\beta}$. To reduce the number of parameters and satisfy the above
 503 condition we set $\bar{s}_{\alpha\alpha} = \bar{s}_{\beta\beta} = 10^\kappa \bar{s}_{\alpha\beta}$ with $\kappa > 1$. Then, for a fluid-fluid-solid
 504 system, Eq (60) reduces to:

$$\bar{s}_{s\alpha} - \bar{s}_{s\beta} = \frac{1}{2} \frac{\sigma_{\alpha\beta}}{\lambda} \cos \theta_0. \quad (61)$$

505 Next, to define the parameters uniquely, we set $\bar{s}_{s\alpha} = \bar{s}_{\alpha\alpha} - \beta$ and $\bar{s}_{s\beta} =$
 506 $\bar{s}_{\alpha\alpha} + \beta$ and assume that: 1) the initial discretization of the α and β phases
 507 is the same (i.e. $n_\alpha = n_\beta = n_{eq}$); 2) fluids are low compressible and n_α and
 508 n_β can be treated as a constant; and 3) $10^{-\kappa} \ll 1$ (e.g. in (Tartakovsky
 509 and Meakin, 2006) $\kappa = 4$ was used). Then, the parameters in the PF-SPH
 510 model can be related to the surface tension, static contact angle and SPH
 511 discretization parameters h and n as

$$s_{\alpha\alpha} = s_{\beta\beta} = \frac{1}{2} n_{eq}^{-2} \frac{\sigma_{\alpha\beta}}{\lambda} \quad (62)$$

512 and

$$s_{s\alpha} = \frac{1}{2} n_{eq}^{-2} \frac{\sigma_{\alpha\beta}}{\lambda} \left(1 + \frac{1}{2} \cos \theta_0 \right) \quad \text{and} \quad s_{s\beta} = \frac{1}{2} n_{eq}^{-2} \frac{\sigma_{\alpha\beta}}{\lambda} \left(1 - \frac{1}{2} \cos \theta_0 \right). \quad (63)$$

513 Similar expressions for $s_{\alpha\alpha}$, $s_{s\alpha}$ and $s_{s\beta}$ have been obtained in Bandara et al.
 514 (2013) based on the Maxwell theory of the capillarity (Maxwell, 1890).

515 The pairwise force $\mathbf{F}_{ij}^{int} = -F_{\alpha\beta}^{int}(r_{ij})\frac{\mathbf{r}_{ij}}{r_{ij}}$ with $F_{\alpha\beta}^{int} = F_{2,\alpha\beta}^{int}$, $F_{3,\alpha\beta}^{int}$ or $F_{4,\alpha\beta}^{int}$
 516 in Table 1 has several additional parameters: the parameter A , defining the
 517 relative magnitude of the repulsive and attractive parts of the force; and
 518 ε and ε_0 in $F_{\alpha\beta}^{int} = F_{2,\alpha\beta}^{int}$ and $F_{3,\alpha\beta}^{int}$, and h_0 in $F_{4,\alpha\beta}^{int}$, defining the relative
 519 range of the repulsive and attractive parts of the force. Based on physical
 520 considerations and by drawing an analogy with inter-molecular forces, we
 521 conclude that ε_0 should be smaller than ε . Besides that, the values of the
 522 parameters determine the distribution of the SPH particles. By construction,
 523 the force $\mathbf{F}_{ij}^{int} = -F_{4,\alpha\beta}^{int}(r_{ij})\frac{\mathbf{r}_{ij}}{r_{ij}}$ has the compact support h , which requires
 524 only neighboring particles within the distance h from any particle i to be
 525 included in summation in Eq (12) for $\mathbf{F}_i^{int}$. The functions $F_{2,\alpha\beta}^{int}$ and $F_{3,\alpha\beta}^{int}$
 526 are a combination of two Gaussian functions with the standard deviations
 527 ε_0 and ε ($\varepsilon_0 < \varepsilon$), and, for computational efficiency (i.e., in Eq (12) to
 528 sum only over particles within the distance h of particle i), one should chose
 529 $\varepsilon < h/3$ for these functions to nearly vanish for $r_{ij} > h$. In the presented
 530 below simulations, we set $\varepsilon = h/3.5$. The parameter A in $F_{2,\alpha\beta}^{int}$, $F_{3,\alpha\beta}^{int}$, and
 531 $F_{4,\alpha\beta}^{int}$ is selected such as to generate negative virial pressure. The values of A ,
 532 $\varepsilon_0/\varepsilon$, and h_0/h , used in the simulations, are shown in Table 1. Fig 3a shows
 533 $F_{k,\alpha\beta}(z)$ ($k=1,2,3,4$) as a function of z , corresponding to the surface tension
 534 $\sigma_{\alpha\beta} = 2$ (in the model units). All pairwise forces have support $h = 1$ (in the
 535 model units), or decay rapidly for $z > 1$. The virial pressure, generated by
 536 these forces, is shown in Fig 3b as a function of particle density n .

537 To study the effect of the functional form of $F_{k,\alpha\beta}^{int}(z)$ and the correspond-

Figure 3: a) $F_{k,\alpha\beta}(z)$ ($k = 1, 2, 3, 4$) for the surface tension $\sigma_{\alpha\beta} = 2$ (in the model units). (b) Virial pressure generated by the forces $\mathbf{F}_{k,ij} = -F_{k,\alpha\beta}(r_{ij})\frac{\mathbf{r}_{ij}}{r_{ij}}$ in a bulk fluid (away from the interface) as a function of particle number density n for $n_{eq} = 16$.

ing parameters on the distribution of the SPH particles and the accuracy
of the proposed parameterization of the PF-SPH model, we simulate flu-
ids α and β separated by a flat interface and a bubble of the fluid α sur-
rounded by the fluid β in two spatial dimensions using the force $\mathbf{F}_{ij}^{int}$ with
 $F_{k,\alpha\beta}^{int}$ ($k = 1, 2, 3, 4$). In these simulations, the particles are placed randomly
in the computational domain by dividing the domain in square subdomains
with the size h^2 and randomly placing n_{eq} particles in each subdomain. Then,
the simulations proceed as described in Section 4.

From the simulations of two fluids separated by a flat interface, we com-
pute the resulting surface tension $\sigma_{\alpha\beta}^r$ using Eq (28). Table 2 gives the relative
error between the prescribed and resulting surface tension $|\sigma_{\alpha\beta} - \sigma_{\alpha\beta}^r|/\sigma_{\alpha\beta}$
for different forces and particle number densities. In most cases, the error
is below 10%. For $n_{eq} = 16$ (which is commonly used in two-dimensional
simulations with the quintic spline $W^{(4)}$), the error is less than 5% for all
tested $F_{\alpha\beta}^{int}$. We can see that there is no strong dependence of the error on

Table 2: The relative error between the prescribed and resulting surface tension $|\sigma_{\alpha\beta} - \sigma_{\alpha\beta}^r|/\sigma_{\alpha\beta}$ for different forces $F_{k,\alpha\beta}^{int}$ ($k = 1, 2, 3, 4$) and particle number densities.

n_{eq}	$F_{1,\alpha\beta}^{int}$	$F_{2,\alpha\beta}^{int}$	$F_{3,\alpha\beta}^{int}$	$F_{4,\alpha\beta}^{int}$
9	0.442	0.075	0.004	1.13
16	0.034	0.044	0.026	0.014
25	0.053	0.067	0.071	0.141
36	0.064	0.067	0.068	0.039
49	0.072	0.078	0.069	0.024

Figure 4: Simulation of two fluids separated by a flat front. $F_{1,\alpha\beta}^{int}$ with $\bar{s}_{\alpha\alpha} = 16^2 \times 0.1$ and $\bar{s}_{\alpha\beta} = 16^2 \times 0.00001$. a) $n_{eq} = 16$ and b) $n_{eq} = 49$.

Figure 5: Simulation of two fluids separated by flat front. $F_{3,\alpha\beta}^{int}$ with $\bar{s}_{\alpha\alpha} = 16^2 \times 2$ and $\bar{s}_{\alpha\beta} = 16^2 \times 0.00001$. a) $n_{eq} = 16$ and b) $n_{eq} = 49$.

553 n_{eq} . In general, the error in the SPH method should decrease with increasing
 554 n_{eq} . Furthermore, the proposed parameterization of the PF-SPH model is
 555 based on an integral approximation of Eq (26), and, for uniformly distributed
 556 SPH particles, the accuracy of this approximation should increase with in-
 557 creasing n_{eq} . One possible explanation for the error in Table 2 in some cases
 558 increasing with increasing n_{eq} is that, for the parameters used in $F_{\alpha\beta}^{int}$, the
 559 particles become more disordered with increasing n_{eq} . For example, Fig 4
 560 shows simulations of two fluids separated by a flat front with the pairwise
 561 force $F_{1,\alpha\beta}^{int}$ at two resolutions, $n_{eq} = 16$ and 49. In the simulations at both
 562 resolutions some clustering of particles is observed, but the clustering is more
 563 pronounced in the simulation with $n_{eq} = 49$ than $n_{eq} = 16$. Simulations with
 564 the pairwise force $F_{3,\alpha\beta}^{int}$ and two resolutions, $n_{eq} = 16$ and 49, are shown in
 565 Figure 5. Here, particles are uniformly distributed in the simulation with
 566 $n_{eq} = 16$, but form a gap near the interface in the simulation with $n_{eq} = 49$.

567 It might be possible to improve the particle distribution in the simulation
 568 with $n_{eq} = 49$ by adjusting the ratio $\varepsilon/\varepsilon_0$. We determine that for $n_{eq} = 16$,
 569 the resolution typically used in two-dimensional SPH simulations with the
 570 quintic spline W , the best results in terms of particle distribution and the
 571 error are obtained with $\varepsilon/\varepsilon_0 = 2$ in $F_{2,\alpha\beta}^{int}$ and $F_{3,\alpha\beta}^{int}$ and $h/h_0 = 2$ in $F_{4,\alpha\beta}^{int}$.

572 Some of the simulations of a fluid bubble are shown in Figure 6. Figure
 573 6(a) depicts the particle distribution obtained from the simulation with $F_{1,\alpha\beta}^{int}$.
 574 Figures 6(b) and 6(c) show the particle distributions obtained from the sim-
 575 ulations with $F_{3,\alpha\beta}^{int}$, $A=(\varepsilon_0/\varepsilon)^4$, and $\varepsilon_0 = 0.5\varepsilon$ and $\varepsilon_0 = 0.4\varepsilon$, respectively.
 576 It can be seen that the pairwise force with $F_{3,\alpha\beta}^{int}$ and $\varepsilon_0 = 0.5\varepsilon$ produces a
 577 more uniform distribution of particles than the other two simulations. Sim-
 578 ilar simulations of the bubble with $F_{2,\alpha\beta}^{int}$, $\varepsilon_0 = 0.5\varepsilon$ and $A = (\varepsilon_0/\varepsilon)^3$ and
 579 $F_{4,\alpha\beta}^{int}$, $h_0 = 0.5h$ and $A = (\varepsilon_0/\varepsilon)^3$ produced a uniform particle distribution
 580 similar to the one shown in Figure 6(b).

581 Next, we simulate bubbles of different sizes and demonstrate that the
 582 PF-SPH model, with the parameters estimated from Eqs (62) and (63), pro-
 583 duces correct surface tension and static contact angle. First, we compute
 584 the surface tension using the pressures inside and outside of the bubble and
 585 radius of the bubble at the equilibrium, R , using the Young-Laplace law

$$P_1 - P_2 = \frac{\sigma}{R}, \quad (64)$$

586 where P_1 and P_2 are the fluid pressures inside and outside of the bubble and
 587 σ is the surface tension. Figs 7(a) and (b) show $P_1 - P_2$ versus $1/R$ ob-
 588 tained from the simulations with $F_{1,\alpha\beta}^{int}$ and $F_{3,\alpha\beta}^{int}$, respectively. These figures
 589 demonstrate that $P_1 - P_2$ varies linearly as a function of $1/R$. According to
 590 the Young-Laplace law, we compute the surface tension as the slope of the

Figure 6: Particle distribution obtained from simulations of a bubble of one fluid surrounded by another fluid with: (a) cosine pairwise force; (b) the pairwise force $F_{3,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta}r_{ij} [-A\psi_{\varepsilon_0}(r_{ij}) + \psi_{\varepsilon}(r_{ij})]$ with $\varepsilon_0 = 0.5\varepsilon$; and (c) $F_{3,\alpha\beta}^{int}(r_{ij})$ with $\varepsilon_0 = 0.4\varepsilon$.

Figure 7: $P_1 - P_2$ versus $1/R$ obtained from SPH simulations of a bubble of fluid 1 with radius R surrounded by fluid 2. The results are obtained with: a) Cosine pairwise function with the parameters: $\bar{s}_{11} = \bar{s}_{22} = 16^2 \times 0.1$ and $\bar{s}_{12} = 16^2 \times 0.00001$; and b) $F_{3,\alpha\beta}^{int}(r_{ij})$ pairwise force with parameters: $\bar{s}_{11} = \bar{s}_{22} = 16^2 \times 2$ and $\bar{s}_{12} = 16^2 \times 0.00001$, $\varepsilon = h/3.5$, $\varepsilon_0 = \varepsilon/2$ and $A = \varepsilon^3/\varepsilon_0^3$. In (a) a good agreement is observed between results obtained with $n = 16$ and 36 . The surface tension, found from a linear regression analysis, is 2.45 , agreeing well (within 1%) with the value $\sigma = 2.43$ predicted by Eq (57). In (b) the surface tension, found from a linear regression analysis, is 1.91 that agrees within 5% with the value $\sigma = 1.83$ predicted by Eq (57).

Figure 8: Sketch of a droplet (the segment of the circle above the horizontal line) on the flat surface (the horizontal line). Contact angle θ is determined from the height a of the droplet, and the lateral size $2b$.

591 lines formed by the $P_1 - P_2$ versus $1/R$ data. For the considered parameters
 592 (listed in the caption of Fig 7), the surface tension, found from the SPH
 593 simulations, are within 5% of the value predicted by Eq (57). The simula-
 594 tions with $F_{1,\alpha\beta}^{int}$ are conducted at two different resolutions (particle number
 595 density n), and Fig 7(a) shows that the results of the SPH simulations and,
 596 hence, the accuracy of Eq (57) are practically independent of the resolution.
 597 We also computed the surface tension from the “bubble” simulations using
 598 Eq (28) and found it to closely agree with the surface tension found from the
 599 Young-Laplace formula.

600 Next, we demonstrate that the static contact angle, obtained in the SPH
 601 model, agrees with Eq (60). We simulate a droplet of fluid 1 in contact
 602 with the solid surface and surrounded by fluid 2. We start with a droplet,
 603 forming the contact angle $\pi/2$ with the solid surface, and let the droplet
 604 relax to an equilibrium with the solid and surrounding fluid. The accurate
 605 direct measurement of the contact angle from a simulated droplet can be
 606 complicated. Instead, we measure the hight of a droplet, a , and the width of

Figure 9: Simulations of a bubble on a solid wall with $F_{3,\alpha\beta}^{int}$. The surface tension between two fluids is $\sigma = 1.83$ in model units. Left figure: Bubble of a non-wetting fluid on a flat surface surrounded by a wetting fluid in the absence of gravity. Prescribed and simulated static contact angles are $\theta = 2.34$ and 2.35 rad, correspondingly. Right figure: Bubble of a wetting fluid on a flat surface surrounded by a non-wetting fluid in the absence of gravity. Prescribed and simulated static contact angles are $\theta = 1.04$ and 0.99 rad, correspondingly. In both these figures the black circle is fitted to the simulated droplet to demonstrate that in the absence of gravity the droplet forms a segment of a circle. These simulations are performed with particle number density $n = 36$.

607 the droplet, $2b$, (see Fig 8), and calculate the static contact angle as

$$\theta = \frac{\pi}{2} - \text{Arcsin} \left[\frac{b^2 - a^2}{b^2 + a^2} \right]. \quad (65)$$

608 Results of numerical simulations with $\theta_0 = 2.34$ and 1.04 rad with $F_{3,\alpha\beta}^{int}$
609 and $n_{eq} = 36$ are shown in Fig 9. For $\theta_0 = 2.34$, the difference between
610 prescribed and resulting static contact angle is 1%, and for $\theta_0 = 1.04$, the
611 error is 5%. Simulations with $n_{eq} = 16$ produced results with similar errors.
612 The same simulations with $F_{1,\alpha\beta}^{int}$ resulted in $\approx 5\%$ errors for $n_{eq} = 36$, but

Figure 10: The contact angle vs $\bar{s}_{1s} - \bar{s}_{2s}$ formed between fluids 1 and 2 and the solid phase s . Solid line is the angle obtained from Eq (60) and symbols are the contact angles obtained from the SPH simulations. $\bar{s}_{11} = \bar{s}_{22} = 16^2 \times 0.1$, $\bar{s}_{12} = 16^2 \times 0.00001$ and $n = 36$.

613 significantly larger errors (up to 30% depending on the prescribed static
614 contact angle) for $n_{eq} = 16$. The reason for the higher accuracy of the
615 simulations with $F_{3,\alpha\beta}^{int}$ relative to the simulations with $F_{1,\alpha\beta}^{int}$ is a more uniform
616 distribution of SPH particles in the former than in the latter. The effect of
617 the form of $F_{k,\alpha\beta}^{int}$ on the particle distribution can be seen, for example, in Fig
618 6. Fig 10 shows the static contact angle θ_0 as a function of $\bar{s}_{1s} - \bar{s}_{2s}$ obtained
619 from the PF-SPH simulations with $F_{3,\alpha\beta}^{int}$ and $n_{eq} = 36$ and calculated from
620 Eq (60). Good agreement (within 5%) between the SPH results and the
621 analytical expression is observed for the entire range of static contact angles
622 (θ_0 varying from 0 to π).

623 Next, we demonstrate the accuracy of the PF-SPH model under dynamic
624 conditions. To do this, we simulate flow of a droplet of a wetting fluid
625 between two parallel plates, driven by a body force. The simulation setup is
626 shown in Fig 11. The steady-state droplet's velocity, u , can be related to the
627 surface tension and the receding (θ_r) and advancing (θ_a) contact angles via
628 the analytical expression (Dragila and Weisbrod, 2004)

Figure 11: Simulation of a droplet flow between two parallel plates under the influence of the body force acting from left to right. In this simulation, $n_{eq} = 36$, density and viscosity of the liquid (fluid 1 denoted by blue particles) are $\rho_l = 36$ and $\mu_l = 1$, density and viscosity of the gas (fluid 2 denoted by green particles) are $\rho_g = 0.0036$ and $\mu_g = 0.001$. The surface tension and the static contact angles are $\sigma = 1.83$ and $\theta_0 = 1.069$ rad. The body force per unit mass is $g = 0.00089$ resulting in $Ca = \frac{u\mu_l}{\sigma} = 0.036$, where u is the steady-state velocity of the droplet. The pairwise force is $F_{3,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta}r_{ij}[-A\psi_{\varepsilon_0}(r_{ij}) + \psi_{\varepsilon}(r_{ij})]$ with $\varepsilon = h/3.5$, $\varepsilon_0 = \varepsilon/2$ and $s_{11} = s_{22} = 0.395$, $s_{12} = s_{11} \times 10^{-5}$, $s_{1s} = 1.24s_{11}$ and $s_{2s} = 0.76s_{11}$.

$$u = \frac{b^2}{12\mu} \left[\rho g + \frac{\sigma}{L} \left(\frac{1}{R_a} - \frac{1}{R_r} \right) \right], \quad (66)$$

where L is the length of the droplet, b is the distance between the two plates and R_a , and R_r are the radii of the curvatures formed by receding and advancing contact angles. We conduct four simulations with Capillary numbers $Ca = \frac{u\mu}{\sigma} = 0.074, 0.056, 0.036$ and 0.015 . The difference between u found from the simulations and Eq (66) for all considered Ca is less than 7%. Figure 12(a) shows the receding and advancing contact angles as functions of Ca obtained from the SPH simulations. It can be seen that the receding and advancing dynamic contact angles follow the power law described by Eq (6). Figure 12(b) shows $\theta_a^3 - \theta_0^3$ and $\theta_0^3 - \theta_r^3$ as functions of Ca . It can be seen that $\theta_a^3 - \theta_0^3$ scales as $Ca^{0.9}$, which agrees well with the Cox-Voinov relationship for the advancing contact angle $\theta_a^3 - \theta_0^3 = a_a Ca$ (a_a is the parameter related

Figure 12: Left panel: $\theta_a - \theta_0$ (diamonds) and $\theta_0 - \theta_r$ (circles) vs. Ca obtained from the simulations of a droplet flowing between two parallel plates. Right panel: $\theta_a^3 - \theta_0^3$ (diamonds) and $\theta_0^3 - \theta_r^3$ (circles) vs. Ca obtained from the same simulations. Here θ_a , θ_r and θ_0 are the advancing, receding and static contact angles formed by the bubble surface and the planes. The setup and parameters of the simulations are given in Fig 11. In the simulations, the capillary number is changed by changing g , the body force per unit length. Also shown is power law fit to the data.

to microscopic properties of the interface) (Cox, 1986).

It has been shown that under certain conditions, e.g., withdrawing of the plate from a bath of liquid, the Cox-Voinov relationship also holds for receding contact angles, i.e., $\theta_0^3 - \theta_r^3 = a_r Ca$ (a_r is the parameter related to microscopic properties of the interface) (Bonn et al., 2009). Figure 12(b) shows that for a bubble moving between two parallel walls the receding contact angle does not satisfy the Cox-Voinov relationship. To show that the Cox-Voinov relationship cannot hold for both the receding and advancing contact angles of the bubble, we can rewrite Eq (66) as $\cos \theta_r - \cos \theta_a = Ca \left[\frac{6L}{b} - \frac{bL\rho g}{2\mu u} \right]$. On the other hand, the Cox law implies that $\cos \theta_r - \cos \theta_a \approx aCa$, where a is the velocity independent constant and the relationships $\cos \theta_r - \cos \theta_0 \approx 5.63(\theta_0^3 - \theta_r^3)$ and $\cos \theta_0 - \cos \theta_a \approx 5.63(\theta_a^3 - \theta_0^3)$ for

652 $|\cos \theta| < 0.65$ are used (Sheng and Zhou, 1992). To verify that under proper
653 conditions the PF-SPH model can reproduce the Cox-Voinov relationship,
654 we simulate the withdrawal of a plate from the liquid bath. The simulation
655 setup is shown in Figure 13(a), and the receding angle is computed at the
656 angle formed by a circle, fitted to the interface as shown in the figure, and the
657 solid boundary. Figure 13(b) demonstrates that the receding contact angle
658 in this problem satisfies the Cox law, i.e., $\theta_0^3 - \theta_r^3 \sim Ca^\alpha$ with $\alpha = 0.93$ which
659 is close to the theoretical value $\alpha = 1$.

660 Finally, we model spreading of a droplet on the horizontal surface (Fig
661 14), and validate the model by comparing the height of the bubble H as
662 a function of time with the Tanner Law $H \sim t^{-1/\alpha}$ (Tanner, 1979). The
663 simulation is initialized by placing a droplet of the fully wetting fluid ($\theta_0 = 0$)
664 in the shape of a half circle with radius $8h$ on the horizontal surface. The
665 rest of the domain is occupied by the non-wetting fluid that has density 1000
666 times smaller and viscosity 100 times smaller than those of the wetting fluid.
667 Tanner has analytically demonstrated that, for a two dimensional bubble
668 formed by a wetting fluid, the exponent is equal to $\alpha = 7$. Fig 15 shows a
669 good agreement between H , as a function of time, found from the simulation
670 and Tanner Law.

671 5.4. Parameterization of the PF-SPH model for three (or more) fluid phases

672 For a three-fluid system (e.g., fluid 1, fluid 2, and fluid 3), the surface
673 tensions for each pair of fluids (i.e., σ_{12} , σ_{23} and σ_{13}) should be specified. For
674 the three fluids to stay in contact with each other at equilibrium, the surface

Figure 13: Left panel: Steady-state distribution of fluid phases obtained from a simulation of the withdrawal of a plate from the bath of liquid (fluid 1 denoted by green particles). Blue particles represents the gas phase (fluid 2). Red particles represent the solid phase. Zero velocity is imposed at the bottom fluid-solid interface and the constant velocity u (the velocity of the withdrawing plate) is prescribed at the left fluid-solid vertical boundary. The upper horizontal and right-hand-side vertical boundaries are assumed to be impermeable (zero normal velocity) and have zero normal viscous stress. The black line is a part of the circle fitted to the fluid-fluid interface. In this simulation, $n_{eq} = 36$, density and viscosity of the liquid are $\rho_l = 36$ and $\mu_l = 3$, density and viscosity of the gas are $\rho_g = 0.0036$ and $\mu_g = 0.03$. The surface tension and the static contact angles are $\sigma = 1.83$ and $\theta_0 = \pi/2$, and the capillary number $Ca = \frac{u\mu_l}{\sigma} = 0.03$. The pairwise force is $F_{3,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta}r_{ij} [-A\psi_{\varepsilon_0}(r_{ij}) + \psi_{\varepsilon}(r_{ij})]$ with $\varepsilon = h/3.5$, $\varepsilon_0 = \varepsilon/2$ and $s_{11} = s_{22} = s_{1s} = s_{2s} = 0.395$, and $s_{12} = s_{11} \times 10^{-5}$. Right panel: $\theta_0^3 - \theta_r^3$ vs. Ca , obtained from the simulations of the plate withdrawal from a bath of liquid. In the simulations, the capillary number is changed by changing u . Also shown is power law fit to the data.

Figure 14: Simulation of a droplet spreading on a horizontal surface under the influence of fluid-solid interactions and surface tension. Green liquid (fluid 1) is assumed to be fully wetting ($\theta_0 = 0$) and gravity is set to zero. In this simulation, $n_{eq} = 36$, density and viscosity of the liquid are $\rho_l = 36$ and $\mu_l = 1000$, density and viscosity of the gas (fluid 2 represented by blue particles) are $\rho_g = 0.0036$ and $\mu_g = 1$. The surface tension and the static contact angles are $\sigma = 183$ and $\theta_0 = 0$ rad. The pairwise force is $F_{3,\alpha\beta}^{int}(r_{ij}) = s_{\alpha\beta} r_{ij} [-A\psi_{\varepsilon_0}(r_{ij}) + \psi_{\varepsilon}(r_{ij})]$ with $\varepsilon = h/3.5$, $\varepsilon_0 = \varepsilon/2$ and $s_{11} = s_{22} = 39.5$, $s_{12} = s_{11} \times 10^{-5}$, $s_{1s} = 1.75s_{11}$ and $s_{2s} = 0.25s_{11}$.

675 tensions should satisfy the following condition:

$$\sigma_{12} < \sigma_{13} + \sigma_{23} \quad (67)$$

$$\sigma_{13} < \sigma_{12} + \sigma_{23}$$

$$\sigma_{23} < \sigma_{13} + \sigma_{12}$$

676 In the PF-SPH model for such three-fluid system, six parameters have to
 677 be specified: s_{11} , s_{22} , s_{33} , s_{12} , s_{13} and s_{23} . To satisfy the conditions (67), and
 678 to uniquely define the parameters $s_{\alpha\beta}$, it is sufficient to set

$$s_{\alpha\beta} \ll s_{\alpha\alpha}, \quad \alpha \neq \beta \quad \alpha, \beta = 1, 2, 3 \quad (s_{\alpha\beta} = s_{\beta\alpha}). \quad (68)$$

Figure 15: Hight of the droplet H (blue symbols), measured at the center of the drop and the half-length L (red symbols), measured from the center to the tip of the advancing film as function of time. The setup and parameters of this simulation are described in Fig 14. Also shown is power law fit to the data.

679 Then, from Eq (57),

$$\begin{aligned}
 \sigma_{12} &\approx \lambda n_{eq}^2 (s_{11} + s_{22}) \\
 \sigma_{13} &\approx \lambda n_{eq}^2 (s_{11} + s_{33}) \\
 \sigma_{23} &\approx \lambda n_{eq}^2 (s_{22} + s_{33}).
 \end{aligned} \tag{69}$$

680 It can be seen that for any choice of $s_{\alpha\alpha}$ ($\alpha = 1, 2, 3$), the surface tensions,
 681 defined according to Eq (69), will satisfy the condition (67). The parameters
 682 $s_{\alpha\alpha}$ can be uniquely related to the surface tension by solving the system of
 683 equations (69). Fig 16(b) shows the equilibrium distribution of three fluids
 684 obtained with $s_{11} = 2$, $s_{22} = 3$, $s_{33} = 4$, and $s_{12} = s_{13} = s_{23} = 10^{-5}s_{11}$. In
 685 this figure, fluid 1 is modeled with blue particles, fluid 2 with green particles,
 686 and fluid 3 with red particles. The initial distribution of the fluid phases in
 687 this simulation is shown in Fig 16(a). The contact angles measured at the
 688 equilibrium are $\theta_1 = 1.77$, $\theta_2 = 2.14$ and $\theta_3 = 2.36$. Theory (Eqs (57) and
 689 (59)) predicts equilibrium contact angles $\theta_1 = 1.83$, $\theta_2 = 1.97$ and $\theta_3 = 2.48$.
 690 The difference between prescribed and estimated contact angles is less than
 691 8%.

692 In the simulations shown in Figs 17(c)-17(d), the parameters $s_{\alpha\beta}$ ($\alpha, \beta =$
 693 $1, 2, 3$) are selected such as to violate the condition (67) (which should prevent
 694 all three fluids from staying in contact with each other). In Fig 17(c), we
 695 set $s_{12} = s_{23} = O(s_{\alpha\alpha})$ and $s_{13} \ll s_{\alpha\alpha}$, and in Fig 17(d), we set $s_{12} = s_{13} =$
 696 $O(s_{\alpha\alpha})$ and $s_{23} \ll s_{\alpha\alpha}$. The initial distribution of fluid phases is shown in Fig
 697 17(a). For the same initial configuration of three fluids, we demonstrate in
 698 Fig 17(b) that the fluids are able to stay in contact with each other when the

Figure 16: (a) Initial distribution of fluid 1 (blue fluid), fluid 2 (green) and fluid 3 (red); (b) equilibrium distribution of the three fluid phases. The pairwise force is given by $F_{3,\alpha\beta}^{int}(r_{ij})$ with $\varepsilon = h/3.5$ and $\varepsilon_0 = \varepsilon/2$, $\bar{s}_{11} = 16^2 \times 2$, $\bar{s}_{22} = 16^2 \times 3$, $\bar{s}_{33} = 16^2 \times 4$, $\bar{s}_{12} = \bar{s}_{13} = \bar{s}_{23} = 16^2 \times 0.00001$. Theory predicts equilibrium contact angles $\theta_1 = 1.83$, $\theta_2 = 1.97$ and $\theta_3 = 2.48$. The angles measured at the equilibrium from the simulation are $\theta_1 = 1.77$, $\theta_2 = 1.14$ and $\theta_3 = 2.36$.

699 (67) is satisfied. In Figs 17(c) and 17(d), the condition (67) is not satisfied,
700 and a three-fluid contact line (point in two spatial dimensions) cannot be
701 formed. The parameters used in these simulations are shown in the caption
702 of Fig 17.

703 6. Three-phase flow in porous media

704 To demonstrate the capabilities and robustness of the PF-SPH method,
705 we use it to simulate three-phase flow in a two-dimensional porous medium,
706 made of irregularly shaped unconnected immobile solid objects, shown in
707 Fig 18. The porous medium is periodic in the horizontal (x) direction and

Figure 17: Panel (a): initial distribution of fluid 1(blue fluid)), fluid 2 (green) and fluid 3 (red). Equilibrium distribution of the three fluid phases for the pairwise force $F_{3,\alpha\beta}^{int}(r_{ij})$ with : b) $\bar{s}_{11} = 16^2 \times 2$, $\bar{s}_{22} = 16^2 \times 3$, $\bar{s}_{33} = 16^2 \times 4$, $\bar{s}_{12} = \bar{s}_{23} = 16^2 \times 0.00001$ and $\bar{s}_{13} = 16^2 \times 1$; c) $\bar{s}_{11} = 16^2 \times 4$, $\bar{s}_{22} = 16^2 \times 2$, $\bar{s}_{33} = 16^2 \times 4$, $\bar{s}_{12} = \bar{s}_{23} = 16^2 \times 1.5$ and $\bar{s}_{13} = 16^2 \times 0.00001$; and d) $\bar{s}_{11} = 16^2 \times 2$, $\bar{s}_{22} = 16^2 \times 4$, $\bar{s}_{33} = 16^2 \times 4$, $\bar{s}_{12} = \bar{s}_{13} = 16^2 \times 1.5$ and $\bar{s}_{23} = 16^2 \times 0.00001$. In all simulations $\varepsilon = h/3.5$ and $\varepsilon_0 = \varepsilon/2$.

Figure 18: Initial distribution of fluid 1(blue fluid)), fluid 2 (green) and fluid 3 (red).

708 initially filled with Liquid 1. The top and bottom of the porous medium are
 709 connected to the atmosphere (i.e. the pressures at top and bottom boundaries
 710 are equal). At time zero, Liquid 1 is drained from the porous medium under
 711 gravity, acting in the vertical (y) direction, and gas (air) enters the pore
 712 spaces no-longer occupied by Liquid 1. The equilibrium distribution of Liquid
 713 1 and air is shown in Fig 18. Here, Liquid 1 is assumed to wet the solid phase
 714 in the presence of air. Next, we consider three scenarios in which Liquid 2
 715 is injected in the domain through the upper boundary. In the rest of this
 716 section we use the following notation: the subscripts 1, 2, g and s denote
 717 Liquid 1, Liquid 2, gas, and solid phases, respectively. In Scenario 1, we
 718 set $\sigma_{12} = \sigma_{1g} = \sigma_{2g} = 3.4$, and assume that Liquid 1 wets the solid in the
 719 presence of gas and Liquid 2 by setting the static contact angle at the solid
 720 boundary formed by Liquid 1 and gas to $\theta_0^{1g} = 0$ rad, by Liquid 2 and gas
 721 to $\theta_0^{2g} = 0$ rad, and by Liquid 1 and Liquid 2 to $\theta_0^{12} = 1$ rad. Liquids 1 and
 722 2 and gas are assumed to be able to stay in contact with each other at the

Figure 19: Saturations of three fluids obtained in Cases 1-3.

equilibrium and form angles $\theta_1 = \theta_2 = \theta_g = 2.09$. The second scenario is
 similar to the first scenario, except Liquid 2 wets the solid in the presence of
 Liquid 1. The static contact angle at the solid boundary formed by Liquid
 1 and Liquid 2 is set to $\theta_0^{1g} = \pi$ rad. The rest of the parameters are the
 same as in the first scenario. In the third scenario, the wetting behavior is
 the same as in the second scenario but it is assumed that the three fluids
 (Liquid 1, Liquid 2 and air) cannot stay in contact with each other. Here,
 we set $\sigma_{12} = 1.71$, $\sigma_{1g} = 5.15$, $\sigma_{2g} = 2.58$, $\theta_0^{1g} = 0$ rad, $\theta_0^{2g} = 0$ rad, $\theta_0^{12} = \pi$
 rad, $\theta_1 = \pi$, $\theta_2 = 0$, and $\theta_g = \pi$.

Fig 19 shows the saturations of the three fluids as functions of time ob-
 served in the three scenarios. The saturations are greatly affected by the
 properties of Liquid 2 (properties of Liquid 1 and air stay constant in Sce-
 nario 1 and 2 and change only slightly in Scenario 3). The residual (late time)
 saturation of Liquid 1 is greatest in Scenario 1 (where Liquid 1 is wetting in
 presence of Liquid 2 and the surface tension between Liquids 1 and 2, σ_{12} ,
 is relatively large) and the smallest in Scenario 3 (where Liquid 2 is wetting
 in presence of Liquid 1 and σ_{12} is relatively small). In all three scenarios,
 the saturation of Liquid 1 starts rapidly decreasing around time $t = 500$ (in

the model units). Fig 20 shows the distribution of fluid phases in Scenarios 1-3 at the time $t = 500$ and the late time ($t=2020$), when the fluids reached a near equilibrium distribution. In Scenarios 1 and 2, the surface tensions satisfy Eq (67); i.e., the three fluids should be able to stay in contact with each other at equilibrium. Figs 20(a), (b), (d) and (e) show that in these scenarios the displaced fluids (Liquid 1 (green) and air (blue)) stay mostly in contact with each other. Opposite to this, in Scenario 3 we can see many bubbles of the displaced air and Liquid 1 completely surrounded by Liquid 2 (the displacing fluid).

7. Conclusions

We presented a new formulation of the Pairwise Force Smoothed Particle Hydrodynamics (PF-SPH) model. In the PF-SPH model, the Young-Laplace and Young boundary conditions at the fluid-fluid and fluid-fluid-solid interfaces are replaced with pairwise forces $\mathbf{F}_{ij}^{int} = -F_{\alpha\beta}^{int}(r_{ij})\frac{\mathbf{r}_{ij}}{r_{ij}}$, which are added into the SPH momentum conservation equation. We derived new approximate relationships between the parameters in $F_{\alpha\beta}^{int}$ and macroscopic properties of a multiphase flow system. For two fluids in contact with a solid surface, the proposed relationships express the parameters in $F_{\alpha\beta}^{int}$ as a function of the surface tension and static contact angle. For a system of three (or more) fluids in which the fluids are able to stay in contact with each other at equilibrium, the parameters in the forces also depend on the equilibrium contact angles formed by any three fluids. We studied the effect of a functional form of $F_{\alpha\beta}^{int}$ on the distribution of the SPH particles and the accuracy of the proposed parameterization. We considered four forms of $F_{\alpha\beta}^{int}$, which

Figure 20: Distribution of Liquid 1 (green), Liquid 2 (red) and air (blue) at different times obtained from Scenarios 1-3. Panels (a) and (d): distribution of the fluid phases at time = 500 and 2020, respectively. Panels (b) and (e): distribution of the fluid phases at time = 500 and 2020, respectively. Panels (c) and (f): distribution of the fluid phases at time = 500 and 2020, respectively. In all simulations $n_{eq} = 19$. Scenario 1: $s_{11} = s_{22} = s_{gg} = 0.1$, $s_{12} = s_{1g} = s_{2g} = 10^{-5}$, $s_{1s} = 0.2$, $s_{2s} = 0.15$ and $s_{3s} = 10^{-5}$. Scenario 2: $s_{11} = s_{22} = s_{gg} = 0.1$, $s_{12} = s_{1g} = s_{2g} = 10^{-5}$, $s_{1s} = 0.2$, $s_{2s} = 0.3$ and $s_{3s} = 10^{-5}$. Scenario 3: $s_{11} = s_{22} = 0.2$, $s_{gg} = 0.1$, $s_{12} = 0.15$, $s_{1g} = 10^{-5}$, $s_{2g} = 0.075$, $s_{1s} = 0.2$, $s_{2s} = 0.3$ and $s_{3s} = 10^{-5}$.

are listed in Table 1, and demonstrated that $F_{\alpha\beta}^{int} = F_{1,\alpha\beta}^{int}$ leads to clumping of the SPH particles, while the other three forms of $F_{\alpha\beta}^{int}$ produce uniform distributions of the particles. The accuracy of SPH discretization schemes decreases as particles become disordered (and clumped together), therefore the choice of $F_{\alpha\beta}^{int}$ can affect the accuracy of the SPH model.

We demonstrated that away from the interface, $\mathbf{F}_{ij}^{int}$ generates an additional “virial” pressure and derived an analytical expression for this pressure, which can be used to compute the total pressure in PF-SPH simulations. We further demonstrated the accuracy of the PF-SPH model by simulating flow of a droplet between two parallel plates and spreading of a droplet on a horizontal surface and comparing the results of the simulations with the analytical solutions.

In numerical models of multiscale systems, it is difficult, if not impossible, to sufficiently resolve all length scales. For example, in a typical porous medium, the size of a pore is much smaller than the size of the porous domain, pore throats are much smaller than the pore size, and fluid bubbles could be much smaller than the average size of the pore throats. Consequently, in a numerical model of the multiphase flow in porous media, it is prohibitively expensive to achieve an adequate resolution of small fluid bubbles and other features, such as films of a wetting fluid surrounding solid phase. One attractive feature of the PF-SPH method is that the forces $\mathbf{F}_{ij}^{int}$ generate surface tension and contact angles in the same way as molecular forces do. Because of this, the PF-SPH model remains numerically stable even when some features of the simulated system (e.g. small bubbles) are not adequately resolved. This is in contrast to other numerical models, which

790 directly implement the Young-Laplace boundary condition. The direct im-
791 plementation of the Young-Laplace boundary condition requires calculations
792 of the normal vectors to, and curvature of, the interface. The calculation
793 of curvatures and normals can become very noisy if the interface between
794 two fluids is not adequately resolved, which can, in turn, lead to numerical
795 instabilities. We should note that, unlike molecular forces, the $\mathbf{F}_{ij}^{int}$ forces
796 are “soft”; i.e., the magnitude of $\mathbf{F}_{ij}^{int}$ is finite for the zero-distance between
797 the particles. Because of this, the $\mathbf{F}_{ij}^{int}$ forces do not impose time step con-
798 straints in addition to the time constraints (23) in the standard SPH method
799 (Monaghan, 2005).

800 We demonstrated that the PF-SPH model is capable of simulating com-
801 plex multiphase flows such as a three-phase flow in a porous medium. We
802 simulated flow of two liquids and gas in a two-dimensional porous medium,
803 made of solid obstacles of irregular shape. Liquids were assumed to be 1000
804 times denser and 100 times more viscous than the gas phase, the ratios typi-
805 cal for water/oil/gas systems. We studied the effect of wettability and surface
806 tensions between different phases on the fluid flow and residual saturation of
807 the fluid phases. Most of the grid-based multiphase flow models are limited
808 to no more than two fluid phases. To the best of our knowledge, the PF-SPH
809 model is one of a very few models that can reliably deal with three-phase
810 pore-scale flows.

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820 **Appendix A. Integral expression for surface tension**

821 For compactness, in this section we provide derivations only in two spacial
822 dimensions. Derivations in three-spacial dimensions are similar, and the final
823 results are identical to the result of Maxwell (1890), obtained for a fluid made
824 of particles (molecules), which do not move at the static equilibrium and
825 interact with each other via pairwise forces.

826 For simplicity, we assume that the fluids are separated by a flat interface.
827 Without loss of generality, we set the interface at $y = 0$, and assume that
828 the subspace $\mathcal{I}_1 = (x, y < 0)$ is occupied by phase 1, while phase 2 occupies
829 the subspace $\mathcal{I}_2 = (x, y > 0)$. According to Eq (28), the surface tension σ is
830 given as

$$\sigma = \int_{y_1}^{y_2} [T_{xx}(y) - T_{yy}(y)] dy, \quad (\text{A.1})$$

831 where $y_1 = -y'$, $y_2 = y'$, and $y' \gg \eta$.

832 Under static condition, the convection component of the stress is equal
833 to zero and the stress is given by Eq (26). For the considered here geometry,
834 T_{xx} can be written as

(A.2)

$$\begin{aligned}
T_{xx}(\mathbf{x}) &= -\frac{1}{2} \sum_{i \in \mathcal{I}_1} \sum_{j \in \mathcal{I}_1} \phi_{11}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}_i + (1-s)\mathbf{r}_j)) ds \\
&- \frac{1}{2} \sum_{i \in \mathcal{I}_2} \sum_{j \in \mathcal{I}_2} \phi_{22}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}_i + (1-s)\mathbf{r}_j)) ds \\
&- \frac{1}{2} \sum_{i \in \mathcal{I}_1} \sum_{j \in \mathcal{I}_2} \phi_{12}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}_i + (1-s)\mathbf{r}_j)) ds \\
&- \frac{1}{2} \sum_{i \in \mathcal{I}_2} \sum_{j \in \mathcal{I}_1} \phi_{21}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}_i + (1-s)\mathbf{r}_j)) ds,
\end{aligned}$$

835 where $\mathbf{r}_{ij(x)}$ is the x component of the vector $\mathbf{r}_{ij}$.

836 Noting that $\phi_{12}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} = \phi_{21}(r_{ji}) \frac{(\mathbf{r}_{ji(x)})^2}{r_{ji}}$, we rewrite the above expres-
837 sion as

(A.3)

$$\begin{aligned}
T_{xx}(\mathbf{x}) &= -\frac{1}{2} \sum_{i \in \mathcal{I}_1} \sum_{j \in \mathcal{I}_1} \phi_{11}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}_i + (1-s)\mathbf{r}_j)) ds \\
&- \frac{1}{2} \sum_{i \in \mathcal{I}_2} \sum_{j \in \mathcal{I}_2} \phi_{22}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}_i + (1-s)\mathbf{r}_j)) ds \\
&- \sum_{i \in \mathcal{I}_1} \sum_{j \in \mathcal{I}_2} \phi_{12}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}_i + (1-s)\mathbf{r}_j)) ds.
\end{aligned}$$

(A.4)

838 To evaluate the integral of T_{xx} , we approximate the sums in the definition
839 of the stress tensor by integrals:

$$T_{xx} = T_{xx}^{(11)} + T_{xx}^{(22)} + T_{xx}^{(12)}, \quad (\text{A.5})$$

840 where

$$T_{xx}^{(11)}(\mathbf{x}) \approx -\frac{1}{2}n_1^2 \int_{\mathcal{I}_1} \int_{\mathcal{I}_1} \phi_{11}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds d\mathbf{r}' d\mathbf{r}'',$$

(A.6)

841

$$T_{xx}^{(22)}(\mathbf{x}) \approx -\frac{1}{2}n_2^2 \int_{\mathcal{I}_2} \int_{\mathcal{I}_2} \phi_{22}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds d\mathbf{r}' d\mathbf{r}'',$$

(A.7)

842 and

$$T_{xx}^{(12)}(\mathbf{x}) \approx -n_1 n_2 \int_{\mathcal{I}_1} \int_{\mathcal{I}_2} \phi_{12}(r_{ij}) \frac{(\mathbf{r}_{ij(x)})^2}{r_{ij}} \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) d\mathbf{r}' d\mathbf{r}''.$$

(A.8)

843 Next, we introduce the notation:

$$\mathbf{r}' = (x', y'), \quad \mathbf{r}'' = (x'', y''),$$

(A.9)

$$\boldsymbol{\rho} = \mathbf{r}' - \mathbf{r}'' = (\rho_1, \rho_2),$$

$$\mathbf{R} = \frac{1}{2}(\mathbf{r}' + \mathbf{r}'') = (R_1, R_2),$$

844 and rewrite $T_{xx}^{(11)}$, $T_{xx}^{(22)}$, and $T_{xx}^{(12)}$ as

$$\begin{aligned} T_{xx}^{(11)} &= -\frac{1}{2}n_1^2 \int_{-\infty}^{\infty} dx' \int_{-\infty}^0 dy' \int_{-\infty}^{\infty} dx'' \int_{-\infty}^0 dy'' \phi_{11}(|\mathbf{r}' - \mathbf{r}''|) \frac{(x' - x'')^2}{|\mathbf{r}' - \mathbf{r}''|} \\ &\times \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds, \end{aligned}$$

(A.10)

845

$$\begin{aligned} T_{xx}^{(22)} &= -\frac{1}{2}n_2^2 \int_{-\infty}^{\infty} dx' \int_{-\infty}^0 dy' \int_{-\infty}^{\infty} dx'' \int_{-\infty}^0 dy'' \phi_{22}(|\mathbf{r}' - \mathbf{r}''|) \frac{(x' - x'')^2}{|\mathbf{r}' - \mathbf{r}''|} \\ &\times \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds, \end{aligned}$$

(A.11)

846 and

$$\begin{aligned} T_{xx}^{(12)} &= -\frac{1}{2}n_1^2 \int_{-\infty}^{\infty} dx' \int_{-\infty}^0 dy' \int_{-\infty}^{\infty} dx'' \int_{-\infty}^0 dy'' \phi_{12}(|\mathbf{r}' - \mathbf{r}''|) \frac{(x' - x'')^2}{|\mathbf{r}' - \mathbf{r}''|} \\ &\times \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds. \end{aligned}$$

(A.12)

847 Similarly, $T_{yy} = T_{yy}^{(11)} + T_{yy}^{(22)} + T_{yy}^{(12)}$, where

$$\begin{aligned} T_{yy}^{(11)} &= -\frac{1}{2}n_1^2 \int_{-\infty}^{\infty} dx' \int_{-\infty}^0 dy' \int_{-\infty}^{\infty} dx'' \int_{-\infty}^0 dy'' \phi_{11}(|\mathbf{r}' - \mathbf{r}''|) \frac{(y' - y'')^2}{|\mathbf{r}' - \mathbf{r}''|} \\ &\times \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds, \end{aligned} \quad (\text{A.13})$$

848

$$\begin{aligned} T_{yy}^{(22)} &= -\frac{1}{2}n_1^2 \int_{-\infty}^{\infty} dx' \int_{-\infty}^0 dy' \int_{-\infty}^{\infty} dx'' \int_{-\infty}^0 dy'' \phi_{22}(|\mathbf{r}' - \mathbf{r}''|) \frac{(y' - y'')^2}{|\mathbf{r}' - \mathbf{r}''|} \\ &\times \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds, \end{aligned} \quad (\text{A.14})$$

849 and

$$\begin{aligned} T_{yy}^{(12)} &= -\frac{1}{2}n_1^2 \int_{-\infty}^{\infty} dx' \int_{-\infty}^0 dy' \int_{-\infty}^{\infty} dx'' \int_{-\infty}^0 dy'' \phi_{12}(|\mathbf{r}' - \mathbf{r}''|) \frac{(y' - y'')^2}{|\mathbf{r}' - \mathbf{r}''|} \\ &\times \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds. \end{aligned} \quad (\text{A.15})$$

850 Splitting the stress components into contributions from different types of
851 forces, we obtain

$$\sigma = \int_{y_1}^{y_2} ((T_{11}^{11}(y) - T_{22}^{11}(y)) + (T_{11}^{22}(y) - T_{22}^{22}(y)) + (T_{11}^{12}(y) - T_{22}^{12}(y))) dy. \quad (\text{A.16})$$

852 Next, choose

$$\tilde{\psi}_\eta(\mathbf{x}) = \psi_\eta(x)\psi_\eta(y), \quad (\text{A.17})$$

853 where ψ_η is chosen so that

$$\psi_\eta(x) = \frac{1}{\eta} \psi\left(\frac{x}{\eta}\right), \quad \psi(x) = \begin{cases} 1 & \text{if } x \in (-1/2, 1/2), \\ 0 & \text{otherwise.} \end{cases} \quad (\text{A.18})$$

854 and

$$\Psi_\eta(y_1, y_2, R_2) = \int_{y_1}^{y_2} \psi_\eta(y - R_2) dy = \begin{cases} 0 & \text{if } R_2 < y_1 - \frac{1}{2}\eta, \\ \frac{1}{\eta}(-y_1 + R_2 + \frac{1}{2}\eta) & \text{if } y_1 - \frac{1}{2}\eta \leq R_2 < y_1 + \frac{1}{2}\eta, \\ 1 & \text{if } y_1 + \frac{1}{2}\eta \leq R_2 < y_2 - \frac{1}{2}\eta, \\ \frac{1}{\eta}(y_2 - R_2 + \frac{1}{2}\eta) & \text{if } y_2 - \frac{1}{2}\eta \leq R_2 < y_2 + \frac{1}{2}\eta, \\ 0 & \text{if } R_2 \geq y_2 + \frac{1}{2}\eta. \end{cases} \quad (\text{A.19})$$

855 Thus,

$$\tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) = \psi_\eta(x - (sx' + (1-s)x''))\psi_\eta(y - (sy' + (1-s)y'')), \quad (\text{A.20})$$

856 and we approximate

$$\begin{aligned} & \int_0^1 \tilde{\psi}_\eta(\mathbf{x} - (s\mathbf{r}' + (1-s)\mathbf{r}'')) ds \\ & \approx \int_0^1 \psi_\eta(x - (\frac{1}{2}x' + (1-\frac{1}{2})x''))\psi_\eta(y - (\frac{1}{2}y' + (1-\frac{1}{2})y'')) ds \\ & = \psi_\eta(x - R_1)\psi_\eta(y - R_2). \end{aligned} \quad (\text{A.21})$$

857 Inserting this into (A.10) and (A.13), and changing variables of integration

858 from $\mathbf{r}', \mathbf{r}''$ to $\boldsymbol{\rho}, \mathbf{R}$, we obtain

$$T_{xx}^{(11)} - T_{yy}^{(11)} \approx -\frac{1}{2}n_1^2 \int_{-\infty}^{\infty} dR_1 \int_{-\infty}^0 dR_2 \int_{S(\mathbf{R})} d\boldsymbol{\rho} \phi_{11}(|\boldsymbol{\rho}|) \frac{\rho_1^2 - \rho_2^2}{|\boldsymbol{\rho}|} \psi_\eta(x - R_1) \psi_\eta(y - R_2). \quad (\text{A.22})$$

The region $S(\mathbf{R})$ of integration with respect to $\boldsymbol{\rho}$ is defined by

$$\rho_1 \in (-\infty, \infty), \quad 2R_2 < \rho_2 < -2R_2.$$

This follows from $y' = R_2 + \frac{1}{2}\rho_2 < 0$, and $y'' = R_2 - \frac{1}{2}\rho_2 < 0$. Since $S(\mathbf{R})$ is independent of R_1 , integration with respect to R_1 can be carried out first.

Because of the normalization condition built into the choice of ψ_η ,

$$\int_{-\infty}^{\infty} \psi_\eta(x - R_1) dR_1 = 1,$$

859 and, thus,

$$T_{xx}^{(11)} - T_{yy}^{(11)} \approx -\frac{1}{2}n_1^2 \int_{-\infty}^0 dR_2 \int_{S(\mathbf{R})} d\boldsymbol{\rho} \phi_{11}(|\boldsymbol{\rho}|) \frac{\rho_1^2 - \rho_2^2}{|\boldsymbol{\rho}|} \psi_\eta(y - R_2).$$

Next, observe that, because of symmetry of $S(\mathbf{R})$, and also because the function under the integral depends only on ρ_1^2 and ρ_2^2 ,

$$\int_{S(\mathbf{R})} \phi_{11}(|\boldsymbol{\rho}|) \frac{\rho_1^2 - \rho_2^2}{|\boldsymbol{\rho}|} d\boldsymbol{\rho} = 4 \int_{\hat{S}(\mathbf{R})} \phi_{11}(|\boldsymbol{\rho}|) \frac{\rho_1^2 - \rho_2^2}{|\boldsymbol{\rho}|} d\boldsymbol{\rho},$$

where

$$\hat{S}(\mathbf{R}) = \{\boldsymbol{\rho} : \rho_1 > 0, 0 < \rho_2 < -2R_2\}.$$

860 Now we change the variable of integration in the $\boldsymbol{\rho}$ -integral to polar coordi-
861 nates t, ξ . Then,

$$T_{xx}^{(11)} - T_{yy}^{(11)} \approx -2n_1^2 \int_{-\infty}^0 dR_2 \int_{D^{(11)}(R_2, t)} dt d\xi \phi_{11}(t) t^2 \cos^2 2\xi \psi_\eta(y - (R_2 + t) \cos 2\xi)$$

where the region of integration $D^{(11)}(R_2, t)$ with respect to t, ξ is described as follows. If $t \in (0, -2R_2)$, then $\xi \in (0, \pi/2)$. If $t \geq -2R_2$, then $\xi \in (0, \hat{\xi})$ with

$$\hat{\xi} = \sin^{-1} \left(-\frac{2R_2}{t} \right).$$

862 To simplify notation, write

$$g(u) = \sin^{-1} u, \tag{A.24}$$

863 so that

$$\hat{\xi} = g \left(-\frac{2R_2}{t} \right) = -g \left(\frac{2R_2}{t} \right). \tag{A.25}$$

864 Similarly,

$$T_{xx}^{(22)} - T_{yy}^{(22)} \approx -2n_1^2 \int_{-\infty}^0 dR_2 \int_{D^{(22)}(R_2, t)} dt d\xi \phi_{22}(t) t^2 \cos^2 2\xi \psi_\eta(y - R_2) \quad (\text{A.26})$$

865 where the region of integration $D^{(22)}(R_2, t)$ with respect to t, ξ is described
866 as follows. If $t \in (0, 2R_2)$, then $\xi \in (0, \pi/2)$. If $t \geq 2R_2$, then $\xi \in (0, \hat{\xi})$ and

$$\hat{\xi} = \sin^{-1} \left(\frac{2R_2}{t} \right). \quad (\text{A.27})$$

867 The contribution from the interaction forces between particles of different
868 phases has the form

$$\begin{aligned} & T_{xx}^{(12)}(y) - T_{yy}^{(12)}(y) \\ &= -n_2 n_2 \int_{-\infty}^0 dR_2 \psi_\eta(y - R_2) \int_{D^{(12,1)}(R_2, t)} dt d\xi \phi_{12}(t) t^2 \cos 2\xi \quad (\text{A.28}) \\ &- n_1 n_2 \int_0^\infty dR_2 \psi_\eta(y - R_2) \int_{D^{(12,2)}(R_2, t)} dt d\xi \phi_{12}(t) t^2 \cos 2\xi. \end{aligned}$$

869 The last two integrals are equal, so we only need to calculate the first one.

870 Thus,

$$T_{xx}^{(12)}(y) - T_{yy}^{(12)}(y) = -2n_2 n_2 \int_{-\infty}^0 dR_2 \psi_\eta(y - R_2) \int_{D^{(12,1)}(R_2, t)} dt d\xi \phi_{12}(t) t^2 \cos 2\xi, \quad (\text{A.29})$$

871 where $D^{(12,1)}(R_2, t)$ is described as follows:

$$D^{(12,1)}(R_2, t) = \{(t, \xi) : -2R_2 < t < \infty, \pi + \hat{\xi} < \xi < 2\pi - \hat{\xi}\}, \quad (\text{A.30})$$

872 where

$$\hat{\xi} = \sin^{-1} \left(-\frac{2R_2}{t} \right). \quad (\text{A.31})$$

873 To calculate the contributions from particles of the same phase we first
874 compute

$$T_{xx}^{(11)}(y) - T_{yy}^{(11)}(y) = -n_1^2 \int_{-\infty}^0 dR_2 \psi_\eta(y - R_2) \int_{-2R_2}^{\infty} dt \phi_{11}(t) t^2 \sin 2\hat{\xi}.$$

875 Integrating in y we obtain

$$\int_{y_1}^{y_2} T_{xx}^{(11)}(y) - T_{yy}^{(11)}(y) dy = -\frac{1}{3} n_1^2 \int_0^{\infty} \phi_{11}(t) t^3 dt. \quad (\text{A.32})$$

876 Similarly,

$$\int_{y_1}^{y_2} T_{xx}^{(22)}(y) - T_{yy}^{(22)}(y) dy = -\frac{1}{3} n_2^2 \int_0^{\infty} \phi_{22}(t) t^3 dt. \quad (\text{A.33})$$

877 To compute the contribution from particles of different phases to the surface
878 tension, we note that

$$T_{xx}^{(12)}(y) - T_{yy}^{(12)}(y) = 2n_1 n_2 \int_{-\infty}^0 dR_2 \psi_\eta(y - R_2) \int_{2R_2}^{\infty} dt \phi_{12}(t) t^2 \sin 2\hat{\xi} \quad (\text{A.34})$$

879 Integrating in y we obtain

$$\int_{y_1}^{y_2} T_{xx}^{(12)}(y) - T_{yy}^{(12)}(y) dy = \frac{2}{3} n_1 n_2 \int_0^{\infty} \phi_{12}(t) t^3 dt. \quad (\text{A.35})$$

880 Combining equations (A.16), (A.32), (A.33), and (A.35) we find

$$\sigma = -\frac{1}{3} n_1^2 \int_0^{\infty} \phi_{11}(t) t^3 dt - \frac{1}{3} n_2^2 \int_0^{\infty} \phi_{22}(t) t^3 dt + \frac{2}{3} n_1 n_2 \int_0^{\infty} \phi_{12}(t) t^3 dt. \quad (\text{A.36})$$

881 **Appendix B. Integral expression for pressure**

882 Pressure in the i -th fluid phase ($i=1,2$) far away from the interface separating
883 fluids 1 and 2 is given by

$$p_i = -T_{xx}^{ii} = -T_{yy}^{ii}. \quad (\text{B.1})$$

884 For $i = 1$, T_{xx}^{11} can be found from Eq (A.23) as

$$T_{xx}^{(11)} \approx -4\frac{1}{2}n_1^2 \int_{-\infty}^0 dR_2 \int_{D^{(11)}(R_2, t)} dt d\xi \phi_{11}(t) t^2 \cos^2 \xi \psi_\eta(y - R_2) \quad (\text{B.2})$$

885 where the region of integration $D^{11}(R_2, t)$ is defined in Appendix Appendix
886 A.

887 We choose a negative y with large $|y|$ so that the support of $\psi_\eta(y - R_2)$
888 is located inside the interval $(-\infty, 0)$. Since the range of the force $\phi_{11}(t)$ is
889 finite, y can be moved further away from the interface if necessary, to ensure
890 that $\phi_{11}(t) = 0$ for all $t > -2R_2$. In that case, the region of integration
891 $D^{(11)}(R_2, t)$ is simply $\xi \in (0, 2\pi), t < -2R_2$, and the integration in t can be
892 extended to $(0, \infty)$ without changing the value of the integral. This yields

$$p_1 = \frac{1}{2}n_1^2 \int_{-\infty}^0 dR_2 \int_0^\infty \int_0^{2\pi} dt d\xi \phi_{11}(t) t^2 \cos^2 \xi \psi_\eta(y - R_2). \quad (\text{B.3})$$

893 Since support of $\psi_\eta(y - R_2)$ lies inside the interval of integration in R_2 , this
894 integral is equal to one. The integral with respect to ϕ can be easily computed
895 directly. The result is

$$p_1 = \frac{1}{2}\pi n_1^2 \int_0^\infty \phi_{11}(t) t^2 dt. \quad (\text{B.4})$$

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