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Stokes-Cahn-Hilliard formulations and simulations of two-phase flows with suspended rigid particles

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Abstract

Three formulations of the Stokes-Cahn-Hilliard (SCH) system are investigated for the simulation of rigid particles in two-phase flows. Using the SCH framework, we assume that the interface between the two fluids is diffuse, whereas the interface between the fluids and the particle is assumed to be sharp. To describe the sharp boundary of the particle, a moving, boundary-fitted mesh is used which is refined near the fluid-fluid interface.

The three formulations, the “stress form” and two “potential forms”, are first investigated in the absence of traction boundary conditions, by simulating a retracting droplet in a closed, cylindrical container. We show that the three formulations perform similar in terms of accuracy, although the velocity is slightly more accurate for the potential forms. When investigating mesh-convergence, superconvergence of the velocity and chemical potential is observed in the three forms. In equilibrium, the stress form shows higher parasitic currents near the interface. **When comparing the pressure as it is defined in the stress form, the potential forms show higher peaks in the pressure near the interface.** The three methods are stable when simulating a stationary droplet for a long period of time.

We proceed by simulating a suspended rigid, spherical particle in the Cahn-Hilliard fluid, where a traction boundary conditions is applied to the particle boundary. For the potential forms, an additional integral term arises on the particle boundary. When investigating mesh-convergence, we observe superconvergence of the location and velocity of the particle, the

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velocity field and the chemical potential if the stress form is used. However, subconvergence is observed for these variables when using the potential forms. The three methods are stable when simulating a particle that is captured at a fluid-fluid interface for a long period of time.

1 Introduction

During the last few decades, diffuse-interface modeling has gained in popularity in numerical analysis of two-phase flows and has been successfully applied in numerous situations, see Anderson et al. [1] for an extensive review. The idea that the interface is diffuse can be traced back to the 19th century, when van der Waals used it to describe the interface between a vapour and a liquid phase [2]. The model was later reformulated for binary liquids by Cahn and Hilliard [3]. The general idea is to define an order parameter that can distinguish the two phases (or fluids), and to modify the free energy in terms of this order parameter. A function called the “phase field” may be introduced to play the role of the order parameter (hence the alternative name “phase-field modeling”), but for a binary liquid it is more straightforward to use the composition. Limiting ourselves to binary liquids, we follow the framework of Cahn and Hilliard, who modified the free energy by adding terms that depend on the gradient of the composition. Due to these additional terms in the free energy, extra terms appear in the momentum balance, i.e. the interfacial tension, yielding the fully coupled Navier-Stokes-Cahn-Hilliard system of equations. In the remainder of this paper, inertia of the fluids and particles is assumed to be negligible, yielding the Stokes-Cahn-Hilliard (SCH) system of equations. Different formulations exist to include the interfacial tension term in the momentum balance, which have been called the “potential form” and the “stress form” by Jacqmin [4], which is the nomenclature we will adopt.

A well-known problem in diffuse-interface modeling is the occurrence of parasitic currents at the interface [5]. These currents occur near the interface, even if the system is in equilibrium. Jacqmin, who uses a potential form, mentions that “it is easy to generate phase-field numerical implementations that are dissipative of energy, and that therefore are free of parasitic flows” [4]. A similar result was found by Jamet et al. for single-component systems [5]. Yet, in the literature different forms of the capillary force can be found, such as the stress form in [6, 7] and the

potential form in [8, 9, 10, 11, 12]. See [13] for an elaborate overview.

To the authors knowledge, there is no literature on the SCH model with traction boundary conditions applied to moving boundaries of the flow. This is however an interesting problem, e.g. when describing a two-phase flow with particles, a problem that was already studied by Choi and Anderson [14] using the SCH model in combination with the extended finite element method. Millet and Wang [15] approached this problem by solving the Cahn-Hilliard equations, and directly adding the capillary forces on particles in the fluids. **Hydrodynamic interactions were included through Stokes drag at low Reynolds numbers.** Aland et al. [16], use a coarse-grained, continuum model to model particles at interfaces. Furthermore, Aland et al. show in [17] that a “diffuse-domain” approach can be used to model particles in two phase-flows in detail, by adding the capillary forces explicitly on the particle. We use a different approach, where the particles are described using a sharp-interface model, whereas the fluids are described using the SCH model.

We will show in this paper that a thorough understanding of the different forms is necessary to correctly prescribe traction boundary conditions in the SCH framework. Furthermore, we will analyze three different forms of the capillary force and compare them in terms of accuracy and stability in problems with and without traction boundary conditions.

2 Problem Statement

To study and compare the accuracy and stability of the potential and the stress forms of the SCH model, two axisymmetric problems are analyzed where interfaces play a role: 1) drop retraction in a closed, cylindrical container (see Fig. 1) and 2) a solid, spherical particle captured at a fluid-fluid interface that is brought out of equilibrium by applying a force on the particle (see Fig. 2). In both problems, it is assumed that the fluids are density- and viscosity-matched, with the density and viscosity given by ρ and η , respectively. The interfacial tension between the two fluids is given by γ . Furthermore, the contact angle between the fluid-fluid interface and solid boundaries is assumed to be 90° . Axisymmetry is assumed in both problems, and the axial coordinate is given by z and the radial coordinate by r , using the convention (z, r) .

Fig. 1: Problem description of drop retraction.

Fig. 2: Problem description of a particle at an interface.

In case of a static contact angle of 90° , the maximum force on the particle that the surface tension can support can be determined and equals $F_{z,\max} = b\gamma\pi$, as is shown in Fig. 3.

Fig. 3: The maximum force on the particle that the interfacial tension can support. The angle between the line connecting the center point of the particle to the point where the fluid-fluid interface meets the particle boundary and the horizontal is given by ϕ , which is similar to the angle between the horizontal and the fluid-fluid interface where it meets the particle. The length of the contact line is given by $l_{\text{int}} = 2\pi b'$, where $b' = b \cos \phi$. Multiplying the vertical component of γ by the length of the contact line, yields $F_z(\phi) = 2\pi b' \gamma \sin \phi = 2\pi \gamma b \cos \phi \sin \phi$, which has a maximum at $\phi = 45^\circ$ of $F_{z,\max} = b\gamma\pi$.

3 Governing equations

3.1 Cahn-Hilliard Theory

The diffuse-interface model was used by van der Waals to describe the interface between a liquid phase and a vapor phase [2] and was later reformulated by Cahn and Hilliard for binary liquids [3]. The model is based on the assumption that the Helmholtz free energy is not only a function of the local composition c , but also of the composition of the immediate environment, which can be taken into account by considering derivatives of c . By taking a Taylor expansion of the free energy around the critical point of a binary fluid, Cahn and Hilliard found

$$f(c, \nabla c) = f_0(c) + \frac{\kappa}{2} |\nabla c|^2, \quad (1)$$

where f is the specific Helmholtz free energy, κ is the gradient energy parameter and f_0 is the specific free energy of the fluids in the bulk and is given by a double-well function, with minima in the bulk values $\pm c_B$. Without loss of generality, we use the “ c^4 ” approximation for f_0 , which is also known as the Ginzburg-Landau free energy, and is given by [18]

$$f_0(c) = -\frac{1}{2}\alpha c^2 + \frac{1}{4}\beta c^4, \quad (2)$$

where α and β are parameters that can be used to change the shape of the double-well function. When minimizing the free energy given by Eq. (1), one can see that the first term is minimal if the two phases are completely separated, whereas the second term will be minimal for a critical fluids since in that case the gradients of c vanish. The combination of the two terms yields a system with a diffuse interface.

The generalized chemical potential of the system is found by taking the variational derivative of the free energy with respect to the concentration

$$\mu = \frac{\delta f}{\delta c} = -\alpha c + \beta c^3 - \kappa \nabla^2 c \quad \text{in } \Omega. \quad (3)$$

The evolution of the system can be described by assuming that material diffuses from areas of high chemical potential to areas of low chemical potential, effectively lowering the total free

energy. This yields the Cahn-Hilliard equation which is given by

$$\frac{Dc}{Dt} = \nabla \cdot (M \nabla \mu) \quad \text{in } \Omega, \quad (4)$$

where M is a diffusion coefficient called the mobility, which can be a function of c but is assumed to be constant in this paper, and $D()/Dt = \partial()/\partial t + \mathbf{u} \cdot \nabla()$ is the material derivative.

For a planar interface in equilibrium ($\mu = 0$), where c only depends on coordinate z , Eq. (3) and Eq. (4) can be solved analytically, which yields a composition profile given by

$$c(z) = c_B \tanh\left(\frac{z}{\sqrt{2}\xi}\right), \quad (5)$$

where $c_B = \sqrt{\alpha/\beta}$ is the composition in the bulk, and $\xi = \sqrt{\kappa/\alpha}$ defines the interface thickness.

The surface energy γ , which is by definition the excess energy present in the system due to the presence of the interface, can be found by [19]

$$\gamma = \int_{-\infty}^{\infty} \rho \kappa \left(\frac{dc}{dz}\right)^2 dz. \quad (6)$$

Substituting the analytical solution for a planar interface given by Eq. (5) in Eq. (6) yields the following expression for the surface energy of a planar interface

$$\gamma = \frac{2\sqrt{2}}{3} \frac{\rho \kappa c_B^2}{\xi}. \quad (7)$$

3.2 Balance equations

It is assumed that inertia can be neglected and that the fluids are incompressible. Therefore, the momentum balance and mass balance become

$$-\nabla \cdot \boldsymbol{\sigma} = \mathbf{f}, \quad \text{in } \Omega, \quad (8)$$

$$\nabla \cdot \mathbf{u} = 0 \quad \text{in } \Omega. \quad (9)$$

where $\boldsymbol{\sigma}$ is the stress tensor and $\mathbf{f}$ is an external volume force acting on the fluid, which is assumed to be zero in this paper. Since a term which depends on ∇c is added to the free energy, additional terms arise in the momentum balance, i.e. the interfacial tension. This

capillary stress can be derived **using a variational procedure, and yields a stress** $-\kappa\nabla c\nabla c$ **plus an isotropic term which can be absorbed in the pressure** [6]. Furthermore, we follow [20, 21], where an isotropic term is added to the capillary stress to ensure that the capillary stresses are parallel to the interface, which yields

$$\boldsymbol{\tau}_c = \rho\kappa \left(|\nabla c|^2 \mathbf{I} - \nabla c \nabla c \right), \quad (10)$$

where $\boldsymbol{\tau}_c$ is the capillary stress tensor, $\nabla c \nabla c$ is a dyadic product and $\mathbf{I}$ is the unit tensor. It is assumed that the fluids are Newtonian with equal viscosity η , therefore the viscous stress tensor $\boldsymbol{\tau}_v$ is given by

$$\boldsymbol{\tau}_v = 2\eta \mathbf{D}, \quad (11)$$

where $\mathbf{D} = (\nabla \mathbf{u} + (\nabla \mathbf{u})^T)/2$ is the symmetric part of the velocity gradient tensor and η is the viscosity. The stress tensor is found by adding the **viscous stress, the capillary stress and a pressure contribution**, which yields

$$\boldsymbol{\sigma} = -g_0 \mathbf{I} + \boldsymbol{\tau}_v + \boldsymbol{\tau}_c, \quad (12)$$

where g_0 is a **pressure field that enforces** the incompressibility constraint given by Eq. (9). Using Eq. (12) in combination with Eq. (10) and Eq. (8) yields the *stress form of the SCH system* [4].

The term $\nabla \cdot \boldsymbol{\tau}_c$ can be rewritten to obtain the potential form of the SCH system:

$$\begin{aligned} \nabla \cdot \boldsymbol{\tau}_c &= \nabla \cdot \rho\kappa \left(|\nabla c|^2 \mathbf{I} - \nabla c \nabla c \right) \\ &= \rho\mu \nabla c - \rho \nabla \left(f_0 - \frac{\kappa}{2} |\nabla c|^2 \right). \end{aligned} \quad (13)$$

Using this expression, we can rewrite Eq. (8) to

$$-\nabla \cdot \hat{\boldsymbol{\sigma}} = \rho\mu \nabla c, \quad (14)$$

with $\hat{\boldsymbol{\sigma}} = \boldsymbol{\tau}_v - g_1 \mathbf{I}$, where a **pressure field** $g_1 = g_0 + \rho f_0 - \rho\kappa |\nabla c|^2/2$ **has been** introduced. In the remainder of this paper, this potential form will be referred to as *potential form 1*.

Furthermore, using the relation $\nabla(\mu c) = c\nabla\mu + \mu\nabla c$, Eq. (13) can be rewritten to obtain

$$\nabla \cdot \boldsymbol{\tau}_c = -\rho c \nabla \mu - \rho \nabla \left(f_0 - \frac{\kappa}{2} |\nabla c|^2 - \mu c \right), \quad (15)$$

which can be used to rewrite Eq. (8) to

$$-\nabla \cdot \bar{\boldsymbol{\sigma}} = -\rho c \nabla \mu, \quad (16)$$

with $\bar{\boldsymbol{\sigma}} = \boldsymbol{\tau}_v - g_2 \mathbf{I}$, where a **pressure field** $g_2 = g_0 + \rho f_0 - \rho \kappa |\nabla c|^2 / 2 - \rho \mu c = g_1 - \rho \mu c$ **has been** introduced. In the remainder of this paper, this potential form will be referred to as *potential form 2*.

3.2.1 Particle at an interface

In the particle at an interface problem it is assumed that external forces and torques on the particle are exactly balanced by the hydrodynamic stresses, which arise due to the particle velocity U , and capillary stresses. This yields the following boundary condition on Γ_p , stating that the resultant of forces on the particle is equal to zero

$$\int_{\Gamma_p} \boldsymbol{\sigma} \cdot \mathbf{n} dS = \mathbf{F}_{\text{ext}} \quad (17)$$

where $\mathbf{n}$ is the normal on Γ_p , pointing away from the fluid, and $\mathbf{F}_{\text{ext}} = (F, 0)$ is a constant external force that is applied on the particle. Due to axisymmetry, the radial component of $\mathbf{F}_{\text{ext}}$ is zero.

To complete the set of balance equations, a kinematic equation is used to update the location of the particle, which is given by

$$\frac{dZ}{dt} = U, \quad (18)$$

where Z is the z -coordinate of the center point of the particle and U is the particle velocity in z -direction, which is an unknown that is solved as part of the system of equations, as will be explained in Section 4.1.

3.3 Initial and boundary conditions

3.3.1 Drop retraction

Due to the time derivative in Eq. (4), an initial composition field $c(t = 0) = c^0$ has to be specified. For the drop retraction problem, the initial interface has the shape of a capsule (or pill). Despite its discontinuous radius of curvature, this shape is chosen as the initial shape since the signed distance to the interface $d(\mathbf{x})$, where $\mathbf{x}$ are the coordinates of any point in the domain, can easily be determined. Eq. (5) can then be used to impose a well-defined initial c field by assuming $z = d(\mathbf{x})$.

For the stability analysis in Section 5.1.3, the initial shape is spherical, with a radius a' . It is known that in the Cahn-Hilliard framework, a system might lower its free energy by decreasing the drop size while slightly shifting the bulk values of the composition. Yue et al. [22] approximated the equilibrium radius and shifts of c for a spherical droplet, which we will use to create a droplet that conserves its mass.

Since it is assumed that the contact angle of the fluid-fluid interface with the solid boundaries is 90° , the following boundary conditions are imposed for c

$$\frac{\partial c}{\partial n} = 0 \quad \text{on } \Gamma, \quad (19)$$

where $\partial/\partial n$ is the partial derivative in the direction normal to the boundary. Furthermore, we assume that there is no in- or out flux of c , which yields the following boundary condition for μ

$$\frac{\partial \mu}{\partial n} = 0 \quad \text{on } \Gamma. \quad (20)$$

No-slip is assumed on the walls of the cylinder, therefore the boundary condition for the velocity is given by

$$\mathbf{u} = \mathbf{0} \quad \text{on } \Gamma. \quad (21)$$

Due to the axisymmetry, there is an additional symmetry condition for u_r on the center line of

the cylinder, which is given by

$$u_r = 0 \quad \text{on } r = 0. \quad (22)$$

To set the **pressure level, the value of g_0 , g_1 and g_2 is** set to zero in the upper left corner of the domain for the stress form, potential form 1 and potential form 2, respectively.

3.3.2 Particle at an interface

In the particle at an interface problem, c^0 is defined by using Eq. (5) directly, yielding a horizontal interface at $z = 0$. To solve Eq. (18), the particle location at $t = 0$ needs to be specified. As is shown in Fig. 2, we assume that the particle starts at the origin: $Z(t = 0) = Z^0 = 0$. Furthermore, there is an additional boundary given by Γ_p . Again, we assume a 90° contact angle and no in- or out flux of c , which yields the boundary conditions

$$\frac{\partial c}{\partial n} = 0 \quad \text{on } \Gamma_p, \quad (23)$$

$$\frac{\partial \mu}{\partial n} = 0 \quad \text{on } \Gamma_p. \quad (24)$$

For the velocity in z -direction, an additional boundary condition is given to enforce no-slip on the particle boundary, whereas the velocity in r -direction is set to zero due to symmetry

$$u_z = U \quad \text{on } \Gamma_p, \quad (25)$$

$$u_r = 0 \quad \text{on } \Gamma_p. \quad (26)$$

To set the **pressure level, the value of g_0 , g_1 and g_2 is** set to zero in the bottom left corner of the domain for the stress form, potential form 1 and potential form 2, respectively.

3.4 Arbitrary Lagrange Euler formulation

The drop retraction problem does not have any moving boundaries, and is solved on a fixed, Eulerian mesh. However, the particle at an interface problem does have moving boundaries, due to the movement of the particle. To describe the particle, a boundary-fitted mesh is used that is moved in time. Since the mesh is not stationary, the governing equations are rewritten

in the arbitrary Lagrange Euler (ALE) formulation [23]. In the ALE formulation the grid that describes the fluid domain is not stationary, as in the Eulerian formulation, but it also does not follow material points, as in the Lagrangian formulation. Instead, the grid movement is in between Eulerian and Lagrangian. To compensate for the the moving grid, the grid velocity needs to be subtracted from the advective term in the material derivative. Since we neglected inertia, the only advective term appears in the Cahn-Hilliard equation, Eq. (4), which is rewritten to

$$\frac{\delta c}{\delta t} + (\mathbf{u} - \mathbf{u}_{\text{grid}}) \cdot \nabla c = \nabla \cdot M \nabla \mu \quad \text{in } \Omega(t), \quad (27)$$

where $\delta/\delta t$ is the grid derivative and $\mathbf{u}_{\text{grid}}$ is the grid velocity.

4 Numerical method

The finite element method is used to solve the SCH system on Ω . A mixed formulation is used and both the chemical potential μ and the composition c are regarded as unknowns. Isoparametric, triangular P_2/P_1 (Taylor-Hood) elements are used for the velocity/pressure, whereas isoparametric and triangular P_2 elements are used for both the composition and the chemical potential. To describe the sharp interface of the particle, a moving, boundary-fitted mesh is used, combined with adaptive meshing similar to [24]. The numerical method is explained for the particle at an interface problem. the drop retraction problem is solved in a similar way, but on a stationary mesh that is refined around the interface. The weak forms of the different formulations, which are used to solve the equations in space, are derived in Section 4.1, whereas the time integration is explained in Section 4.2.

4.1 Weak form

The weak form of the Cahn-Hilliard equation is given by: Find c and μ such that

$$\left(r, \frac{\delta c}{\delta t} + (\mathbf{u} - \mathbf{u}_{\text{grid}}) \cdot \nabla c\right) + M(\nabla r, \nabla \mu) = 0 \quad (28)$$

$$(s, -\beta c^3 + \alpha c) - \kappa(\nabla s, \nabla c) + (s, \mu) = 0. \quad (29)$$

for all admissible test functions r and s and where $(\cdot, \cdot)$ is an inner product on $\Omega(t)$ defined by

$$(a, b) = \int_{\Omega} ab \, dV, \quad (\mathbf{a}, \mathbf{b}) = \int_{\Omega} \mathbf{a} \cdot \mathbf{b} \, dV, \quad (\mathbf{A}, \mathbf{B}) = \int_{\Omega} \mathbf{A} : \mathbf{B} \, dV, \quad (30)$$

for scalars a and b , vectors $\mathbf{a}$ and $\mathbf{b}$ and tensors $\mathbf{A}$ and $\mathbf{B}$. In Eqs. (28) and (29) the boundary conditions for c and μ given by Eqs. (19) and (20) were used directly to set the boundary terms on $\Gamma_p \cup \Gamma$ to zero.

To describe the moving particle, we follow the approach of [25] and combine the balance of momentum and the motion of the particle in one weak form which avoids having to calculate the stresses on the particle explicitly. Note, that the particle velocity U is an unknown that is solved as part of the system of equations. The no-slip boundary condition on the particle is enforced using a Lagrange multiplier field λ .

The weak form of the stress form is given by: Find $\mathbf{u}$, g_0 , U and λ such that

$$(\mathbf{D}_v, 2\eta\mathbf{D}) - (\nabla \cdot \mathbf{v}, g_0) + \langle v_z - V, \lambda \rangle_{\Gamma_p} = -(\mathbf{D}_v, \rho\kappa(|\nabla c|^2 \mathbf{I} - \nabla c \nabla c)) + VF \quad (31)$$

$$(q, \nabla \cdot \mathbf{u}) = 0 \quad (32)$$

$$\langle \chi, u_z - U \rangle_{\Gamma_p} = 0, \quad (33)$$

for all admissible test functions $\mathbf{v} = (v_z, v_r)$, q , χ and V . Furthermore, $\mathbf{D}_v = (\nabla \mathbf{v} + (\nabla \mathbf{v})^T)/2$ and $\langle \cdot, \cdot \rangle_{\Gamma_p}$ is an inner product on Γ_p , defined in Section 4.4. Since the stress form is used, the boundary condition for the stress on the particle, given by Eq. (17) can be substituted directly into the integral over Γ_p that follows after integration by parts.

The weak form of the potential form 1 is given by: Find $\mathbf{u}$, g_1 , U and λ such that

$$(\mathbf{D}_v, 2\eta\mathbf{D}) - (\nabla \cdot \mathbf{v}, g_1) + \langle v_z - V, \lambda \rangle_{\Gamma_p} = (\mathbf{v}, \rho\mu\nabla c) + VF - (\mathbf{v}, ((\rho f_0 + \frac{1}{2}\rho\kappa|\nabla c|^2)\mathbf{I} - \rho\kappa\nabla c \nabla c) \cdot \mathbf{n})_{\Gamma_p} \quad (34)$$

$$(q, \nabla \cdot \mathbf{u}) = 0 \quad (35)$$

$$\langle \chi, u_z - U \rangle_{\Gamma_p} = 0, \quad (36)$$

for all admissible test function $\mathbf{v}$, q , χ and V and where $(\cdot, \cdot)_{\Gamma_p}$ is an inner product on Γ_p , similar to the inner products defined in Eq. 30.

The weak form of the potential form 2 is given by: Find $\mathbf{u}$, g_2 , U and λ such that

$$(\mathbf{D}_v, 2\eta\mathbf{D}) - (\nabla \cdot \mathbf{v}, g_2) + \langle v_z - V, \lambda \rangle_{\Gamma_p} = -(\mathbf{v}, \rho c \nabla \mu) + VF \\ - (\mathbf{v}, ((\rho f_0 + \frac{1}{2}\rho\kappa|\nabla c|^2 - \mu c)\mathbf{I} - \rho\kappa\nabla c \nabla c) \cdot \mathbf{n})_{\Gamma_p} \quad (37)$$

$$(q, \nabla \cdot \mathbf{u}) = 0 \quad (38)$$

$$\langle \chi, u_z - U \rangle_{\Gamma_p} = 0, \quad (39)$$

for all admissible test function $\mathbf{v}$, q , χ and V .

Note, that the last term on the right hand side of Eqs. (34) and (37) is added to satisfy the boundary condition for the stress on Γ_p , given by Eq. (17) (see Appendix A for the derivation).

4.2 Time integration

Due to the interfacial tension term in the momentum equation and the convection term in the Cahn-Hilliard equations, the SCH system of equations is fully coupled. One could opt to solve this system at once, but this would yield one system with a large number of unknowns, which is computationally expensive. Another approach is to solve the Cahn-Hilliard and Stokes systems sequentially within one time step, yielding two systems with substantially less degrees of freedom in each system, decreasing the computation time per time step significantly. After specifying an initial concentration field c^0 , as was explained in Section 3.3, an initial velocity field $\mathbf{u}^0$ and particle velocity U^0 can be found by solving the equations of motion. Note, that for the potential forms, the chemical potential at $t = 0$ is needed, which is found by substituting c^0 into Eq. (29) and solving for μ . The particle position at t^1 is found using a first-order, explicit Euler scheme:

$$Z^1 = Z^0 + \Delta t U^0. \quad (40)$$

For subsequent time steps, a second-order, explicit Adams-Bashforth scheme is used to evaluate Z^{n+1}

$$Z^{n+1} = Z^n + \Delta t \left(\frac{3}{2} U^n - \frac{1}{2} U^{n-1} \right). \quad (41)$$

With the new particle location Z^{n+1} , the mesh is updated to represent the domain $\Omega(t^{n+1})$, as will be explained in Section 4.3. The Cahn-Hilliard equations are then solved first on $\Omega(t^{n+1})$.

For the first time step, Eq. (27) is discretized using a first-order, semi-implicit Euler scheme. Combining with the discretized form of Eq. (3) yields the following system of equations

$$\frac{c^1 - c^0}{\Delta t} + (\mathbf{u}^0 - \mathbf{u}_{\text{grid}}^1) \cdot \nabla c^1 = \nabla \cdot (M \nabla \mu^1), \quad (42)$$

$$-\alpha c^1 + \beta (c^1)^3 - \kappa \Delta c^1 = \mu^1, \quad (43)$$

where $\mathbf{u}^0$ is used as a first-order prediction for $\mathbf{u}^1$. For subsequent time steps, a second-order, semi-implicit Gear scheme is used to evaluate c^{n+1} and μ^{n+1} :

$$\frac{\frac{3}{2}c^{n+1} - 2c^n + \frac{1}{2}c^{n-1}}{\Delta t} + (\hat{\mathbf{u}}^{n+1} - \mathbf{u}_{\text{grid}}^{n+1}) \cdot \nabla c^{n+1} = \nabla \cdot (M \nabla \mu^{n+1}), \quad (44)$$

$$-\alpha c^{n+1} + \beta (c^{n+1})^3 - \kappa \Delta c^{n+1} = \mu^{n+1}, \quad (45)$$

where $\hat{\mathbf{u}}^{n+1} = 2\mathbf{u}^n - \mathbf{u}^{n-1}$ is a second-order prediction for the velocity. Note, that Eq. (45) is non-linear and a Newton-Raphson iteration is used to find a solution. The solution is considered converged if the maximum difference in c with the previous solution in the Newton-Raphson iteration is less than 10^{-9} . Using c^{n+1} and μ^{n+1} , the weak forms of the balance of momentum, given in Section 4.1, are solved for $\mathbf{u}^{n+1}$, U^{n+1} , λ^{n+1} and the **pressures** g_0^{n+1} , g_1^{n+1} and g_2^{n+1} for the stress form, potential form 1 and potential form 2 respectively. Furthermore, c^{n+1} can be used to add the integral on Γ_p in Eqs. (34) and (37).

Note, that the Cahn-Hilliard equations (28) and (29) yield an asymmetric system, which is solved using the direct MA41 solver from the HSL library [26]. The flow equations (31)-(39) yield symmetric systems, which are solved using the direct MA57 solver for symmetric matrices from the HSL library [26].

4.3 Motion of the mesh

The equations defined in Section 4.1 are solved on a boundary fitted mesh, where the elements are aligned with the particle boundary. To apply the ALE scheme, we need to define the motion of the mesh. In order to do this, the particle boundary is moved such that it coincides with the new particle position Z^{n+1} , while keeping the other boundaries stationary. This is done by solving a Poisson problem on the mesh at t^n , where the particle displacement, given by $\Delta Z = Z^{n+1} - Z^n$, is used as a boundary condition on Γ_p , whereas the boundary conditions on

the remaining boundaries are set to zero. This yields the following set of equations:

$$\nabla \cdot (K_e \nabla (\Delta z_m)) = 0 \quad \text{in } \Omega, \quad (46)$$

$$\Delta z_m = 0 \quad \text{on } \Gamma, \quad (47)$$

$$\Delta z_m = \Delta Z \quad \text{on } \Gamma_p, \quad (48)$$

where z_m are the nodal mesh z -coordinates and K_e is a diffusion coefficient which is proportional to the inverse of the element size, which ensures that larger elements will deform more than smaller elements, minimizing mesh distortion [23]. The mesh displacement field can be used to find the coordinates of the new mesh according to

$$z_m^{n+1} = z_m^n + \Delta z_m. \quad (49)$$

An example of an initial, undeformed mesh is shown in Fig. 4, whereas a deformed mesh is shown in Fig. 5.

Fig. 4: Initial mesh that is refined around the particle boundary and around the diffuse interface.

Fig. 5: Deformed mesh that moves with the particle, but further away is stationary. The mesh deformation is exaggerated for clarity.

For the first time step, the mesh velocity in each node is obtained using a first-order back-

wards differencing scheme:

$$u_m^1 = \frac{z_m^1 - z_m^0}{\Delta t}. \quad (50)$$

whereas for subsequent time steps, a second-order backwards differencing scheme is used to find the mesh velocity:

$$u_m^{n+1} = \frac{\frac{3}{2}z_m^{n+1} - 2z_m^n + \frac{1}{2}z_m^{n-1}}{\Delta t}. \quad (51)$$

Solving the SCH system requires a high mesh resolution around the interface, where the gradients of c and μ are high. Since the gradients far away from the interface are much smaller, adaptive meshing is employed, and meshes are used that are refined around the interface, but coarse far away from the interface. The refinement is done using Gmsh [27], which allows the use of refinement points: points that are not connected to the mesh and which have the purpose to create local refinements. Each point is associated with an element size h_{fine} and a distance d_{fine} . In the circle with radius d_{fine} around the refinement point, the element size of that point is used to create the mesh. On the particle boundary, element size h_{particle} is prescribed, whereas far away from the interface, the elements have size h_{coarse} . By using many refinement points on the interface, a band of finer elements is created as is shown in Fig. 4.

4.3.1 Remeshing and projection

As the mesh is deformed in time, elements may become too deformed to yield accurate solutions. Furthermore, the diffuse interface may have moved out of the refined region. To ensure the mesh quality and resolution, a new mesh is generated by using the known position and radius of the particle. Refinement points are added on the interface, defined by the contour line $c = 0$, to make sure that the new mesh is refined near the interface and coarse far away from the interface. Two criteria are used to invoke remeshing.

The first criterion tracks the deformation of each element, for which we use a similar approach as was used in [23]. To quantify the mesh deformation, both the change in area and change in aspect ratio are determined. The change in area of each element is denoted by f_1^e and is defined by

$$f_1^e = |\log(A^e/A_0^e)|, \quad (52)$$

where A^e is the element area and A_0^e is the element area of the **undeformed** mesh. The change

in aspect ratio of each element is denoted by f_2^e and is defined by

$$f_2^e = |\log(S^e/S_0^e)|, \quad (53)$$

with the aspect ratio defined as $S^e = (L_{\max}^e)^2/A_0^e$, where $L_{\max}^e$ is the maximum length of the sides of an element. Remeshing is invoked if either $f_1^e > 1.39$ or $f_2^e > 1.39$, which coincides with a change in area or aspect ratio by a factor 4.

The second criterion follows [24], where a band of elements is defined around the interface which are allowed to contain the interface. If the interface, defined by the line $c = 0$, moves beyond this band, remeshing is invoked. The distance to the interface of each nodal point could be used as a measure, but it is more practical to use the nodal values of c , since these are readily available. Elements for which any of the nodal values of $|c| < 0.34$ are added to a list of elements that are allowed to contain the interface.

To solve the time integration given by Eq. (44), the solutions c^n , c^{n-1} , $\mathbf{u}^n$ and $\mathbf{u}^{n-1}$ are needed on the new mesh. In order to obtain these variables on the new mesh, a projection problem is solved. To illustrate the projection, consider the composition c that is given on an old, deformed, mesh by $c^{\text{old}} = \sum_i \phi_i^{\text{old}} c_i^{\text{old}}$, where ϕ_i^{old} are the shape function on the deformed mesh and c_i^{old} are the old nodal values. The composition on the new mesh is given by $c^{\text{new}} = \sum_j \phi_j^{\text{new}} c_j^{\text{new}}$, where ϕ_j^{new} are the shape function on the new mesh and c_j^{new} are the new, unknown, nodal values. To find the nodal values c_j^{new} , the following problem is solved

$$\sum_j (\phi_k^{\text{new}}, \phi_j^{\text{new}}) c_j^{\text{new}} = (\phi_k^{\text{new}}, c^{\text{old}}), \quad (54)$$

where the integration on the right hand side is performed by sampling c^{old} in the integration points. A similar set of equations is solved to obtain the other field variables at the previous time steps.

Furthermore, the coordinates of the new mesh at the previous time steps are needed to determine the mesh velocity $\mathbf{u}_{\text{mesh}}^{n+1}$. To obtain the coordinates of the new mesh at previous time steps, the mesh coordinates of the old mesh at t^n are used as nodal values in the old mesh at t^{n+1} . These values are then projected onto the new mesh at t^{n+1} , and the new mesh at the previous time step can be obtained by replacing the nodal coordinates by the projected values,

as is shown in Fig. 6. A similar procedure can be used to find the mesh at t^{n-1} and the mesh velocity can now be found in the usual manner, by using Eq. (51).

Fig. 6: By projection of the coordinates, the new mesh is obtained at previous time steps. To obtain the new mesh at t^{n-1} , the nodal values of the old mesh at t^{n-1} are projected as well.

4.4 Particle constraint

The constraints given by Eqs. (33), (36) and (39) involve an inner product on the particle boundary Γ_p . Following [25], we define the discrete inner product, which is similar to enforcing the constraint using a collocation method and is given by

$$\langle \chi, u_z - U \rangle_{\Gamma_p} = \sum_{k=1}^{n_{\text{coll}}} \chi_k \cdot (u_z(\mathbf{x}_k) - U), \quad (55)$$

where n_{coll} is the number of collocation points, which coincide with the nodes on the particle boundary. Furthermore, χ_k and $\mathbf{x}_k$ are the Lagrange multiplier and nodal coordinate of the k -th collocation point respectively.

5 Results

5.1 Drop retraction

In order to investigate the accuracy and stability of the potential and stress forms in the absence of traction boundary conditions, we consider the problem of a droplet retracting to a spherical shape in a closed, cylindrical container. The initial shape of the drop is given by a cylinder of length $2a$ with two hemispherical caps with radius a , as was shown in Fig. 1. In the following simulations $a = 0.1$ and the dimensions of the outer cylinder are $L = 1$ and $R = 0.5$. Both fluids have a viscosity of $\eta = 1$ and a density of $\rho = 1000$. The coefficients in the Cahn-Hilliard equation are given by $\alpha = \beta = 1$, $\kappa = 10^{-4}$ and $M = 0.01$. These values yield bulk values of the concentration given by $c_B = \pm 1$ and an interface thickness of $\xi = 0.01$, which were defined in Eq. (5). According to Eq. (7), we can calculate the interfacial tension/energy of a planar interface, which is given by $\gamma = 20\sqrt{2}/3$.

The characteristic velocity in this problem, where flow is completely due to surface tension, is given by $v \sim \gamma/\eta$ [9]. When taking a as the characteristic length scale, the characteristic time scale for the flow is $t \sim a\eta/\gamma \approx 0.01$.

5.1.1 h -convergence

Simulations are performed on a mesh with $h_{\text{coarse}} = 0.2$ and $h_{\text{fine}} = 0.02$, which is shown in Fig. 7. This mesh is then uniformly refined to obtain meshes with an increasing number of nodes, which are shown in Table 1. The simulations are performed until $t = 2.5 \times 10^{-2}$, with a time step of $\Delta t = 5 \times 10^{-4}$.

Fig. 7: Mesh M1 used in the mesh convergence study for the drop retraction problem. The isoline $c = 0$ is shown for $t = 0$ in green (dotted) and for $t = 2.5 \times 10^{-2}$ in blue (solid). The line on which the relative errors are determined, is shown in red (dashed).

Table 1: The uniformly refined meshes used in the convergence study.

	h_{coarse}	h_{fine}	number of nodes
M1	0.2	0.02	960
M2	0.1	0.01	3763
M3	0.05	0.005	14901
M4	0.025	0.0025	59305
M5*	0.00625	0.000625	945313

The relative L^2 -errors are determined at $t = 2.5 \times 10^{-4}$ on the line Γ_ϵ , which is given by $(z, r) = (s, a/2)$, where s is a local coordinate along Γ_ϵ . For the **pressures**, the relative L^2 -error is given by ϵ_{g_0} , ϵ_{g_1} and ϵ_{g_2} , which are defined by

$$\epsilon_{g_0} = \frac{\left(\int_{\Gamma_\epsilon} (g_0^h - g_0^*)^2 ds \right)^{1/2}}{\left(\int_{\Gamma_\epsilon} g_0^{*2} ds \right)^{1/2}}, \quad \epsilon_{g_1} = \frac{\left(\int_{\Gamma_\epsilon} (g_1^h - g_1^*)^2 ds \right)^{1/2}}{\left(\int_{\Gamma_\epsilon} g_1^{*2} ds \right)^{1/2}}, \quad \epsilon_{g_2} = \frac{\left(\int_{\Gamma_\epsilon} (g_2^h - g_2^*)^2 ds \right)^{1/2}}{\left(\int_{\Gamma_\epsilon} g_2^{*2} ds \right)^{1/2}}, \quad (56)$$

where g_0^h , g_1^h and g_2^h are the solutions on one of the meshes given in Table 1 and g_0^* , g_1^* and

g_2^* are reference solutions which were computed on mesh M5^{*}, which was obtained after twice uniformly refining M4. Note, that the reference **solution of the pressures** is computed for each form separately, since these solutions are form-dependent. The relative L^2 -errors for the velocity, the composition and the chemical potential are denoted by ϵ_u , ϵ_c and ϵ_μ , respectively, and are defined by

$$\epsilon_u = \frac{\left(\int_{\Gamma_\epsilon} \|\mathbf{u}^h - \mathbf{u}^*\|^2 ds\right)^{1/2}}{\left(\int_{\Gamma_\epsilon} \|\mathbf{u}^*\|^2 ds\right)^{1/2}}, \quad \epsilon_c = \frac{\left(\int_{\Gamma_\epsilon} (c^h - c^*)^2 ds\right)^{1/2}}{\left(\int_{\Gamma_\epsilon} c^{*2} ds\right)^{1/2}}, \quad \epsilon_\mu = \frac{\left(\int_{\Gamma_\epsilon} (\mu^h - \mu^*)^2 ds\right)^{1/2}}{\left(\int_{\Gamma_\epsilon} \mu^{*2} ds\right)^{1/2}}, \quad (57)$$

where $\mathbf{u}^h$, c^h and μ^h are the solutions on one of the meshes given in Table 1 and $\mathbf{u}^*$, c^* and μ^* are reference solutions which were computed using the stress form. The integrals given in Eq. (56) and Eq. (57) are evaluated by dividing the line Γ_ϵ into 10000 intervals and using a mid-point rule on each interval.

Convergence plots are shown in Fig. 8. The errors of the pressures converge second-order, which is expected from the order of the elements. Due to the difference in the **definition of the pressures**, there is some difference in accuracy with the stress form being the least accurate and the second potential form being the most accurate. The convergence of the error of the composition field is third-order, which is also as expected from the order of the elements. However, errors of the velocity and chemical potential both converge fourth-order, which is one order higher than expected. Furthermore, the velocity is about 1.6 times more accurate in the potential forms **than** in the stress form. For the composition and chemical potential, there is no significant difference in the accuracy of the solutions of the different forms.

Note, that in the convergence plots for $\mathbf{u}$, c and μ , a reference solution was computed using the stress form. We checked if computing the reference solution using one of the potential forms would yield a different results, which it did not. It can therefore be concluded that the different forms indeed converge to the same values.

(a) h -convergence of g_0 (stress form), g_1 (potential form) 1 and g_2 (potential form 2).

(b) h -convergence of $\mathbf{u}$.

(c) h -convergence of c .

(d) h -convergence of μ .

Fig. 8: h -convergence for the drop retraction problem for the **pressures** (a), velocity (b), composition (c) and chemical potential (d).

5.1.2 Time-convergence

To study the time-convergence, the same problem as in the previous section was studied with time step sizes between $\Delta t = 5 \times 10^{-3}$ to $\Delta t = 1 \times 10^{-4}$. The mesh used in this time convergence study is M3, as was given in Table 1. Fig. 9 shows the convergence of the errors as they were defined in Eqs. (56) and (57). However, the reference solutions g_0^h , g_1^h , g_2^h , $\mathbf{u}^*$, c^* and μ^* are now computed using a small time step of $\Delta t = 1 \times 10^{-5}$ on mesh M3. Furthermore, the reference solutions are determined for each form separately, since the spatial error is not necessarily the same for each form. As expected from the order of our time-integration scheme, the time-convergence is second-order. Again, a difference can be observed in the accuracy of

the pressures, due to **their different definitions**. The accuracy of the velocity, composition and chemical potential is the same for the three forms.

Fig. 9: time-convergence of the drop retraction problem for the **pressures** (a), velocity (b), composition (c) and chemical potential (d).

5.1.3 Stability

In order to study the stability of the forms, simulations are performed of a stationary droplet. For these simulations, the interface thickness is 10 times smaller than in the mesh- and time-convergence studies. This is done by setting $\kappa = 10^{-6}$, yielding an interface thickness of $\xi = 10^{-3}$. To keep the interfacial tension the same, the density has been increased to $\rho = 10000$. Following a scaling law for the mobility of $M \sim \xi$ [4], we set the mobility to $M = 10^{-3}$. To ensure the droplet is in equilibrium from the beginning of the simulation, the radius of the

droplet is given by $a' = 0.1986$, and c is given by $c = -c_B \tanh(d(\mathbf{x})/\sqrt{2}\xi) + 0.002$, where $d(\mathbf{x})$ is the signed distance to the interface. These values of the droplet radius and composition field are the relaxed values of a droplet that starts with radius 0.2, according to [22]. The mesh on which the simulations are performed, is strongly refined with an element size of $h_{\text{fine}} = 0.001$ around the interface, which yields around 9 to 10 nodal points across the interface, which has been reported to be enough for convergence [6]. Further away from the interface, the element size is $h_{\text{coarse}} = 0.1$, as is shown in Fig. 10a. Fig. 10b shows the drop at $t = 0$, whereas Fig. 10c shows the droplet at $t = 50$. Note, that this spans 2500 times the characteristic flow time in this problem which is given by $t \sim a'\eta/\gamma \approx 0.02$. The droplet is stable for the three forms and can not be visually distinguished from one another.

Fig. 10: The initial mesh that is used to simulate a stationary droplet (a), c of the initial droplet (b) and c of the final droplet (c), which is visually indistinguishable for the **three** forms.

To further investigate the stability, Fig. 11 shows the surface area of the droplet, defined by the area of the iso-surface $c = 0$, and the droplet volume, defined by the volume inside the iso-surface $c = 0$. A decrease of both the surface area and volume is observed for the three forms, which is smallest for potential form 2 and largest for the stress form, which shows a decrease of around 0.3% in volume.

Fig. 11: The surface area of the droplet (a) and the volume of the droplet (b) versus time.

For a stationary droplet, it is expected that the kinetic energy, given by $\int ||\mathbf{v}||^2/2 dV$, vanishes in the entire domain. To investigate if this is indeed the case, we plot the kinetic energy as a function of time in Fig. 12. The kinetic energy for the stress form is about $O(10^{-9})$ from the beginning to the end of the simulation. However, the potential forms show a higher kinetic energy at the initial stages of the simulations, which then decreases to $O(10^{-12})$. Indeed, when we look at the velocity field for the different forms at $t = 50$ in Fig. 13, it can be seen that for potential form 1 and the stress form (which both contain ∇c terms in the momentum balance), spurious current occur near the interfacial region. However, these currents are two orders of magnitude larger for the stress form than for the first potential form. The velocity field for the second potential form is completely different, with areas of larger velocity occurring in the bulk. We can explain these observations by looking at the forcing terms. In the stress form, the $\nabla c \nabla c$ term causes higher inaccuracies near the interfacial region. This problem is less in the first potential form, which adds the $\mu \nabla c$ term. This force does, however, not vanish since the c field is slightly shifted in equilibrium, and μ does not go to zero. Instead, the chemical potential goes to a constant value, which explains that for potential form 2, which adds a $-c \nabla \mu$ term, no spurious current are observed near the interface. The spreading of the chemical potential is however a slow process since it is completely diffusion-driven and gradients of μ are still present in the domain that induce currents in the bulk.

Fig. 12: The **kinetic energy** in the whole domain for a stationary droplet versus time.

Fig. 13: Comparison of the velocity field at $t = 50$ for the first potential form (a), the second potential form (b) and the stress form (c). The close-up shows contour lines of the velocity magnitude around the interface. Spurious currents near the interface are clearly observed in the stress form and in the first potential form, whereas the velocity field does not vanish in the bulk for the second potential form.

The **pressure fields** at $t = 50$ are shown in Fig. 14. The stress form shows a higher g_0 inside the droplet than outside the droplet, i.e. the Laplace pressure. At the interface, some

irregularities of g_0 can be observed, with peaks with a maximum value of 116% of the **difference of g_0** across the interface. For potential form 1, g_1 has a very similar distribution, but is more regular across the interface. **For** potential form 2, g_2 is distributed over a larger region, and its absolute values are smaller. Using the definitions of the **pressures** given in Section 3.2, g_0 can be derived for the potential forms, **and a direct comparison can be made between the g_0 -fields obtained using the three different forms.** For potential form 1, g_0 is derived by solving the projection problem: Find g_0 such that

$$(r, g_0) = (r, g_1 - \rho f_0 + \rho \kappa |\nabla c|^2 / 2), \quad (58)$$

for all admissible test functions r . For potential form 2, g_0 is derived by solving the projection problem: Find g_0 such that

$$(r, g_0) = (r, g_2 - \rho f_0 + \rho \kappa |\nabla c|^2 / 2 + \rho \mu c), \quad (59)$$

for all admissible test functions r . After projection, the value of g_0 in the upper left corner is subtracted from the entire domain to ensure the level coincides with the stress form.

Oscillations in g_0 of about 300% of the difference across the interface are observed for both potential forms. **The value of g_0** is shown along the line Γ_ϵ in Fig. 15. Across the interface, oscillations are observed in the three forms, but for the potential forms these oscillations are about an order of magnitude larger than for the stress form.

These oscillations can be explained by looking at the **definitions of the pressure** for the potential forms, repeated here for convenience: $g_1 = g_0 + \rho f_0 - \rho \kappa |\nabla c|^2 / 2$ and $g_2 = g_0 + \rho f_0 - \rho \kappa |\nabla c|^2 / 2 - \rho \mu c$. Only gradients of **the pressures** are computed in the simulations, and the f_0 and $|\nabla c|^2$ are not computed explicitly. If g_1 and g_2 seem to be smooth, this does not necessarily mean that f_0 and $|\nabla c|^2$ are smooth as well. It is clear from Fig. 15 that the mesh is not refined enough to accurately describe these terms, yielding large fluctuations in g_0 .

Fig. 14: The **pressure fields** at $t = 50$ for the stationary droplet. The close-up shows contour lines of the **pressure** around the interface.

Fig. 15: The **pressure as defined in the stress form** (g_0) for the three forms on the line Γ_ϵ (a) and a close-up around the droplet (b). For the potential forms, g_0 was derived from g_1 and g_2 using the definitions given in Section 3.2.

5.2 Particle at an interface

In order to investigate the accuracy and stability of the potential and stress forms in the presence of traction boundary conditions, we consider the problem of a particle at an interface. Simulations were performed using a particle with radius $b = 0.1$, in a cylinder with dimensions $L = 1$ and $R = 0.5$. The fluid- and diffuse-interface parameters are the same as those in Section

5.1. The force that is applied on the particle is equal to the maximum force that the interface can support, which was shown in Fig. 3 to be equal to $F = b\gamma\pi = \pi 2\sqrt{2}/3$.

Fig. 16: Mesh M1 used in the mesh convergence study for the particle at an interface problem at $t = 0$ (left) and $t = 0.1$ (right). The isoline $c = 0$ is shown in blue (solid). The line on which the relative errors are determined, is shown in red (dashed).

Table 2: The uniformly refined meshes used in the convergence study.

	h_{coarse}	h_{fine}	h_{particle}	number of nodes
M1	0.2	0.02	0.02	1356
M2	0.1	0.01	0.01	5315
M3	0.05	0.005	0.005	21045
M4	0.025	0.0025	0.0025	83753
M5*	0.00625	0.000625	0.000625	1334945

5.2.1 h -convergence

Simulations are performed on a mesh with $h_{\text{coarse}} = 0.2$ and $h_{\text{fine}} = h_{\text{particle}} = 0.02$ which is shown in Fig. 16. This mesh is then uniformly refined to obtain meshes with an increasing number of nodes, which are shown in Table 2. The simulations are performed until $t = 0.1$, with a time step of $\Delta t = 5 \times 10^{-4}$. The relative errors in the location and the velocity of the

particle, given by ϵ_Z and ϵ_U respectively, are determined at $t = 0.1$. The errors are defined as

$$\epsilon_Z = \frac{|Z - Z^*|}{|Z^*|} \quad \epsilon_U = \frac{|U - U^*|}{|U^*|} \quad (60)$$

where $|Z^*|$ and $|U^*|$ are reference solutions which were computed on $M5^*$ using the stress form. Convergence of the location and velocity of the particle is shown in Fig. 17. For the stress form, superconvergence of both the location and velocity of the particle is observed, similar to the convergence of $\mathbf{u}$ and μ for the drop retraction problem. However, for both potential forms, the convergence is significantly lower, close to second order. Note, that if the reference solution is computed using one of the potential forms, the same second-order convergence is obtained for the potential forms. However, due to the lower rate of convergence, that solution is not accurate enough to show convergence of the stress form.

Fig. 17: h -convergence of the particle at an interface problem for the **particle location** (a) and **particle velocity** (b).

We further inspect the accuracy by plotting the error inside the domain, which is again determined on a line Γ_ϵ , as is shown in Fig. 16. The errors are similar to the ones defined in Section 5.1.1, and again the integrals in Eqs. (56) and (57) are determined by dividing Γ_ϵ into 10000 intervals and using mid-point rule on each interval. Convergence plots are shown in Fig. 18. For the stress form, the errors for $\mathbf{u}$ and μ converge fourth-order, which is again one order higher than expected. The convergence of the error of the **pressures** and c is as expected, second-order and third-order respectively. This result is similar to what was seen in the drop

retraction problem. However, for both potential forms, the convergence is second-order for all field variables.

Fig. 18: h -convergence of the particle at an interface problem for the **pressures** (a), **velocity** (b), **composition** (c) and **chemical potential** (d).

5.2.2 Time-convergence

To study the time-convergence, the same problem as in the previous section was studied with time step sizes between $\Delta t = 5 \times 10^{-3}$ to $\Delta t = 2 \times 10^{-4}$. The mesh used in this time convergence study is M3, as was given in Table 2. Fig. 19 shows the convergence of the errors as they were defined in Eq. (60). However the reference solutions Z and U are now computed for a small time step of $\Delta t = 1 \times 10^{-5}$ on mesh M3. To isolate the time error, the reference solutions are determined for each form separately. The convergence plots presented in Fig. 19 indeed confirm

the second-order accuracy of the time-integration of the particle movement.

Fig. 19: time-convergence of the particle at an interface problem for the particle location (a) and particle velocity (b).

5.2.3 Stability

In order to study the stability of the forms in the presence of traction boundary conditions, the simulation from the previous section is continued until $t = 50$. For these simulations, the interface thickness is 10 times smaller than in the mesh- and time-convergence studies. This is done by setting $\kappa = 10^{-6}$, yielding an interface thickness of $\xi = 10^{-3}$. The mobility has been set to $M = 10^{-3}$. To keep the interfacial tension the same, the density has been increased to $\rho = 10000$. The mesh on which the simulations are performed, is strongly refined with an element size of $h_{\text{fine}} = 0.001$ around the interface and $h_{\text{particle}} = 0.01$. Further away from the interface, the element size is $h_{\text{coarse}} = 0.1$, as is shown in Fig 20a. Fig. 20b shows the particle at $t = 0$, whereas Fig. 20c shows the particle at $t = 50$. Since the particle comes to a complete stop, capillary forces completely dominate this problem. The characteristic flow time in this problem is then given $t \sim b\eta/\gamma \approx 0.01$, where b has been used as the characteristic length scale. Note, that the simulations span 5000 times the characteristic time scale. The simulations are stable for the three forms and the final c field can not be visually distinguished from each other for the different forms.

Fig. 20: The initial mesh that is used to simulate a particle that is captured by an interface (a), the initial location of the particle (b) and the final location of the particle (c).

The particle location and particle velocity are shown in Fig. 21. Clearly, the velocity drops significantly as the interface moves along the particle boundary and exerts an upwards force on the particle. At approximately $t = 5$, the particle is in equilibrium with the surface tension, and at the end of the simulation, the particle velocity has decreased to $O(10^{-6})$.

Fig. 21: The particle location (a) and the particle velocity (b) versus time (note the log scales in (b)).

Since the particle is in equilibrium with the surface tension, we expect the velocity field to vanish. Therefore, we investigate the **kinetic energy** in the domain, which is shown in Fig. 22. Clearly, it is the first potential form that has the highest decrease in kinetic energy to $O(10^{-14})$, whereas for the stress form, the total kinetic energy remains 6 orders of magnitude higher. On

close inspection of the velocity field, as shown in Fig. 23, the first potential form and the stress form show spurious currents around the interface, due to the ∇c term, whereas the second potential form has higher currents in the bulk. The observation that the first potential form has a higher decrease in **kinetic energy** can be explained by the amount of fluid with $c_B = -1$ being equal to the amount of fluid with $c_B = 1$. The system can not lower its free energy as much by decreasing the interfacial area while increasing the bulk values of c , as was the case for a droplet. **Furthermore, the curvature of the interface is smaller as compared to the case of the droplet.** Therefore, μ will be smaller and consequently $\mu \nabla c$ will be smaller as well, giving rise to less fluid motion and thus a lower kinetic energy.

Fig. 22: The **kinetic energy** in the whole domain for a particle that is captured at an interface versus time. For the potential forms, larger oscillations are seen after remeshing due to the lower rate of convergence.

Fig. 23: Comparison of the velocity field at $t = 50$ for the first potential form (a), the second potential form (b) and the stress form (c). The close-up shows contour lines of the velocity magnitude around the interface. Spurious currents near the interface are clearly observed in the stress form and in the first potential form, whereas the velocity field does not vanish in the bulk for the second potential form.

The **pressure fields** at $t = 50$ are shown in Fig. 24. In the stress form, a Laplace pressure can be seen in the bottom fluid, due to the curvature of the interface. At the interface, g_0 is irregular, with peaks with a maximum of about 1440% of the difference across the interface. For potential form 1, g_1 has a very similar distribution, but is more regular across the interface. In potential form 2, g_2 is distributed over a larger region, and its absolute values are smaller. Using the definitions of g_1 and g_2 given in Section 3.2, g_0 can be derived for the potential forms, **and a direct comparison can be made between the g_0 -fields obtained using the three different forms.** This is done by solving a projection problem, as was explained in Section 5.1.3. In g_0 , oscillations as large as 6100% of the difference across the interface are found for both potential forms. **A plot of g_0** is shown along the line Γ_ϵ in Fig. 25. Across the interface, oscillations are observed in the three forms, but for the potential forms these oscillations are again about an order of magnitude larger than for the stress form. The larger relative oscillations in the particle at an interface problem can be attributed to the smaller curvature of the interface, and therefore smaller Laplace pressure.

Fig. 24: The **pressure fields** at $t = 50$, when the force on the particle is in equilibrium with the surface tension. The close-up shows contour lines of the **pressure** around the interface.

Fig. 25: The **pressure as defined in the stress form** (g_0) for the three forms on the line Γ_ϵ . For the potential forms, g_0 was derived from g_1 and g_2 using the definitions given in Section 3.2.

6 Discussion and Conclusion

We have compared the accuracy and stability of three formulations of the SCH system: the stress form and two potential forms. The results of the simulations of drop retraction indicate that the accuracy of the three forms is comparable in the absence of traction boundary conditions. Superconvergence is observed for the velocity and the chemical potential, which is one order higher than is expected from the order of the elements. Furthermore, the three forms are able to simulate a stationary droplet with a small interface thickness, without showing instabilities. Spurious currents around the interface are observed in the stress form and in potential form 1, which can be attributed to ∇c -terms in the capillary force. The spurious currents are about two order of magnitude higher for the stress form, but still low enough to accurately describe a stationary droplet over a long period of time. For potential form 2, a non-vanishing velocity field in the bulk is observed. **When comparing the pressure as it is defined in the stress form (g_0), oscillations at the interface are seen in all three methods. However, these oscillations are around an order of magnitude larger for the potential forms, where g_0 is derived from the pressures g_1 and g_2 by solving a projection problem.**

We investigated the behavior of the three forms in the presence of traction boundary conditions by simulating a particle at an interface, and showed that the stress form outperforms both potential forms in terms of accuracy. It seems that the integral on the particle boundary, necessary to impose the traction boundary condition for the potential forms, limits the mesh-convergence to second-order. Stability was investigated by simulating a particle with a force applied on it that is in equilibrium with the surface tension for a long duration. Again, the three forms are able to simulate the particle for a long duration using a small interface thickness, without showing instabilities. Similarly to the drop retraction problem, the stress form and potential form 1 show spurious currents at the interface, whereas a non-vanishing velocity field in the bulk is seen for potential form 2. Furthermore, oscillations are observed in g_0 at the interface, which are about an order of magnitude larger for the potential forms.

This analysis can help in choosing the right form of the capillary force. If traction boundary conditions are imposed on the flow, the stress formulation is preferred due to the simpler formulation and higher accuracy. However, if only Dirichlet boundary conditions are imposed on the flow, all forms perform well. The potential forms show lower spurious currents in equilibrium

and the velocity is slightly more accurate, whereas the stress form gives less fluctuations of g_0 near the interface, when comparing to the the potential forms (**where g_0 is obtained by solving a projection problem**). The results indicate that when simulating two-phase flows in the presence of traction boundary conditions, the stress form of the SCH is a straightforward way to increase mesh-convergence, which in turn allows computations on coarser meshes, which is especially interesting for simulations in 3D.

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Appendix

A Prescribing traction boundary conditions in the potential forms

We want to prescribe a traction on an arbitrary traction boundary:

$$\boldsymbol{\sigma} \cdot \mathbf{n} = \mathbf{t}_N \quad \text{on } \Gamma_N. \quad (61)$$

Using $\boldsymbol{\sigma} = -g_0 \mathbf{I} + \boldsymbol{\tau}_c + \boldsymbol{\tau}_v$, we can rewrite this to

$$(-g_0 \mathbf{I} + \boldsymbol{\tau}_v) \cdot \mathbf{n} = \mathbf{t}_N - \boldsymbol{\tau}_c \cdot \mathbf{n} \quad \text{on } \Gamma_N. \quad (62)$$

For the weak form of the potential form 1 we obtain: Find $\mathbf{v}$ and g_1 such that

$$(\mathbf{D}_v, \hat{\boldsymbol{\sigma}}) - (\mathbf{v}, \hat{\boldsymbol{\sigma}} \cdot \mathbf{n})_{\Gamma_N} = (\mathbf{v}, \rho \mu \nabla c), \quad (63)$$

for all admissible test function $\mathbf{v}$, with $\hat{\boldsymbol{\sigma}} = \boldsymbol{\tau}_v - g_1 \mathbf{I}$, where a **pressure** $g_1 = g_0 + \rho f_0 - \rho \kappa |\nabla c|^2 / 2$ **has been** introduced. We can now write

$$\begin{aligned} \hat{\boldsymbol{\sigma}} \cdot \mathbf{n} &= (-\rho f_0 + \rho \kappa |\nabla c|^2 / 2) \mathbf{I} \cdot \mathbf{n} + (-g_0 \mathbf{I} + \boldsymbol{\tau}_v) \cdot \mathbf{n} \\ &= (-\rho f_0 + \rho \kappa |\nabla c|^2 / 2) \mathbf{I} \cdot \mathbf{n} + \mathbf{t}_N - \boldsymbol{\tau}_c \cdot \mathbf{n} \\ &= \mathbf{t}_N - ((\rho f_0 + \rho \kappa |\nabla c|^2 / 2) \mathbf{I} - \rho \kappa \nabla c \nabla c) \cdot \mathbf{n}. \end{aligned} \quad (64)$$

Likewise, for the weak form of the potential form 2 we obtain

$$(\mathbf{D}_v, \bar{\boldsymbol{\sigma}}) - (\mathbf{v}, \bar{\boldsymbol{\sigma}} \cdot \mathbf{n})_{\Gamma_N} = -(\mathbf{v}, \rho c \nabla \mu), \quad (65)$$

with $\bar{\boldsymbol{\sigma}} = \boldsymbol{\tau}_v - g_2 \mathbf{I}$, where a **pressure** $g_2 = g_0 + \rho f_0 - \rho \kappa |\nabla c|^2 / 2 - \mu c$ **has been** introduced.

We can now write

$$\bar{\boldsymbol{\sigma}} \cdot \mathbf{n} = \mathbf{t}_N - ((\rho f_0 + \rho \kappa |\nabla c|^2 / 2 - \mu c) \mathbf{I} - \rho \kappa \nabla c \nabla c) \cdot \mathbf{n}. \quad (66)$$