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Abstract The present work is concerned with the simulation and optimal control of two-phase flows. We provide stable time discretization schemes for the simulation based on both, smooth and non-smooth free energy densities, which we combine with a practical, reliable and efficient adaptive mesh refinement concept for the spatial variables. Furthermore, we consider optimal control problems for two-phase flows and, among other things, derive first order optimality conditions. In the presence of smooth free energies we encounter classical Karush-Kuhn-Tucker (KKT) conditions, while in the case of non-smooth free energies we can prove C(larke)-stationarity. Moreover, we propose a dual weighted residual concept for spatial mesh adaptivity which is based on the newly derived stationarity conditions. We also address future research directions, including closed-loop control concepts and model order reduction techniques for simulation and control of variable density multiphase flows.

1 Introduction

In the present work we

a) develop and analyze numerical discretization concepts for the simulation of two-phase flow problems with variable fluid densities that guarantee a locally refined resolution of the local processes at the interface and preserve the thermodynamically consistency of the underlying models;

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b) formulate and analyze optimal control problems for two-phase flows and develop robust and reliable solution strategies for optimal control of two-phase flows governed by diffuse interface models.

Concerning a) we extend the work [61] to the thermodynamically consistent model for two-phase flow with different densities proposed in [5]. This allows to accurately resolve the transition region between the fluid phases, thus yielding quantifiable simulation of the physical processes located in the interfacial region. Special care is taken to preserve the thermodynamical consistency on the time discrete and fully (i.e., space and time) discrete level. In addition, residual–based a posteriori error estimation of the flow is incorporated into our approach. The benchmark for rising bubble dynamics from [79] is used as a validation.

In many applications, one is interested in steering the underlying multiphase flow towards a desired phase pattern, e.g., at a specific (final) time and/or to a desired flow profile, e.g., yielding rotation-free flows. Particular applications can be found in polymer science, where membrane formation or blending are of importance. In the former case, the porosity pattern of the membrane determines the membrane's use and quality, whereas in the latter context particular material properties of the new blends can be obtained. Moreover, for instance, in the case of immiscible polymer blends, on a macroscopic scale pure phases are present, but close to the interface certain diffusion processes take place. The latter are modeled by diffusive interfaces with certain partial differential equations (PDEs) posed within the narrow band of the diffuse interface. In this context, it appears that the double-obstacle potential is well suited for modeling the interface (indeed, the double-well potential allows unphysical violations of the constraints for the concentration, whereas a logarithmic potential would not allow to reach a pure phase). We also mention that further applications of surface active agents obeying PDEs on an (diffuse) interface include drug delivery, industrial emulsification, or liquid/liquid extraction and hydrodesulfurization of crude oil. These topics are anayzed within objective b).

Mathematically, the underlying state system, i.e. the multi-phase flow model, is given by a coupled Cahn-Hilliard-Navier-Stokes (CHNS) system, where the Cahn-Hilliard part models the phase separation and the Navier-Stokes system captures the dynamics of the fluid. Task (b), which is mentioned above, hence requires to establish existence of solutions to the underlying control problems, stability and sensitivity of the CHNS system subject to perturbations, and the derivation of first-order optimality or stationarity conditions. In this context, the semi-discretization (in time) of the forward model (CHNS) has to additionally guarantee consistency properties of the resulting adjoint system in order to enable the derivation of certain energy-type estimates which yield existence of a solution of the semi-discrete forward problem and the associated adjoint. Such a discretization technique is proposed in [45, 67], where the regularization approach presented in the first reference yields the desired stationarity conditions of ε-almost C-stationary type and facilitates the implementation of a solution algorithm. Concerning the control action in the context of optimal control problems, we use Dirichlet boundary control as in [72] and control of the amplitudes of given distributed control actions [58].

Objective b) is further concerned with the design and analysis of numerical solvers for the underlying control problems. Due to the extreme computational cost caused by solving the Navier-Stokes system coupled to a non-linear and non-smooth parabolic system for the phase separation for each time step, this especially requires an efficient mesh refinement technique for the underlying finite elements method. Therefore it is our goal to incorporate suitable error estimates which take the special structure of the optimal control problem into account by estimating the discretization error with respect to the objective functional, i.e. the quantity of interest.

1.1 Related Work

We commence with an overview of the state-of-the-art for the numerical treatment and analysis for optimal control problems for variable density two-phase flows.

We consider the diffuse interface approach for the simulation of two-phase flows. In contrast to so called sharp interface approaches, the interface between immiscible fluids in this approach is assumed to have a positive but small thickness. In sharp interface approaches this interface is assumed to be a lower dimensional manifold that is represented during numerical simulation, either explicitly or implicitly. Here we only refer to [48], [79] and [104] and the references therein. We further stress that several projects inside Special Priority Programme (SPP) 1506 of the German Research Foundation (DFG) worked on numerical realizations for sharp interface models and we refer to the corresponding proceedings for further readings.

In the following we restrict ourselves to diffuse-interface approaches.

1.1.1 Work related to the simulation of two-phase flows using the diffuse interface approach

Since the pioneering work [31] and the famous model 'H' in [78] many authors have dealt with the investigation of two-phase flows using diffuse interface models with equal density fluids. In parallel, several attempts where made to generalize the model 'H' to the case of different densities.

For stable discretization schemes for the Cahn–Hilliard equation we refer to [54, 55] and for the Cahn–Hilliard Navier–Stokes system to [84]. Multigrid solvers for the Cahn–Hilliard equation are proposed e.g. in [85, 87], and residual based error estimation is proposed in [60, 61]. For a fully coupled solver for model 'H' we refer to [41].

Focusing now on models with different densities, one notes that one of the main limitations of model 'H' is that it is only thermodynamically consistent in situations where both fluids (roughly) have the same density. Indeed, in [38, 105] it is shown that the model is also consistent in the situation of different densities if the kinetic energy of the fluid is defined by using $\sqrt{\rho}|v|^2$ instead of $\rho|v|^2$, where ρ is the distributed density of the fluid and v is the velocity field. The notion of a distributed

density is based on φ and by using the densities of the individual fluid components, a global density field is defined by attaching to every point of the computational domain the density of the fluid.

In [10], solutions to the model in [38] using a discretization that sequentially couples the Cahn–Hilliard and the Navier–Stokes equations are compared to results for a rising bubble benchmark proposed in [79] using sharp interface numerics. The authors obtained good agreement of the numerical results. A critical point in numerical treatment of these equations is the discretization of interfacial forces that appear from surface tension. In [9], different stabilization schemes are proposed for the discretization of this terms and a CFL-like condition for admissible time step sizes is derived. In [86] a completely new model for this forces is proposed.

A first thermodynamically consistent model for two-phase flow with different densities is considered in [92]. Here the velocity is not solenoidal so that analytical and numerical investigations of this scheme explicitly have to consider the pressure. Further the pressure enters the equation for the two-phase structure leading to a strong coupling of the resulting equations. In [57], a time discretization scheme is proposed that preserves the consistence with thermodynamics and numerical examples are provided.

Another diffuse interface model for fluids of different densities is proposed in [26]. Here the velocity is solenoidal, but the model is not consistent with thermodynamics. In [10], results for an implementation of this model are compared with results for sharp interface models for a benchmark of rising bubble simulations as proposed in [79].

In [5], a thermodynamically consistent model for two-phase flows is proposed. It contains a solenoidal velocity field and can be regarded as an extension of model 'H', as it resembles its structure and only differs from it by using variable densities and by an additional term in the convective term in the Navier–Stokes equation. The latter term vanishes in the case of equal densities. In [5], three variants of this model are proposed that can also handle, e.g., non-Newtonian fluids or additional particles that are transported across the interface but do not interact with it. The existence of a weak solution for the case of constant mobility is shown in [3] for the logarithmic free energy that guarantees a-priori bounds on the range of the phase field. In [4], the existence of weak solutions is established for general smooth free energies together with a degenerate mobility that also guarantees these bounds. The existence of a weak solution for non-Newtonian fluids is discussed in [2] for a polynomially bounded free energy and constant mobility. In the latter work, also an extension of the model of [5] is proposed that allows to use nonlinear but smooth relations between the phase field φ and the density field $\rho(\varphi)$ for the case where $|\varphi|$ is not bounded by one, which appears due to a smooth free energy. We note here that by convention $\varphi \equiv -1$ if a pure fluid phase is reached and $\varphi \equiv 1$ if the respective other pure fluid phase is present.

In [49], the existence of generalized solutions is shown for the case of a polynomially bounded free energy and constant free energy. Depending on the densities of the individual fluids, expressed using the Atwood number, these generalized solu-

tions are weak solutions for some time horizon. The analysis is based on proceeding to the limit in a numerical scheme.

In [10], a discretization of this model is used to simulate the rising bubble benchmark from [79]. These authors obtained good agreements with results from sharp interface numeric. The free energy density is chosen as the smooth polynomially free energy and is linearized using Taylor expansion. The Navier–Stokes equation and the Cahn–Hilliard equation are used sequentially coupled by using the velocity field from the old time instance in the Cahn–Hilliard equation and solving the Navier–Stokes equation afterwards using the phase field and chemical potential obtained from the Cahn–Hilliard equation. We note that the important property of thermodynamically consistency is not preserved in the numerical realization by this approach.

Throughout the following publications, thermodynamically consistent time discretization schemes for the model from [5] are proposed.

A first thermodynamically consistent scheme is proposed in [51] that strongly couples the Chan–Hilliard and the Navier–Stokes equation. It is implemented in a splitting scheme, where the Navier–Stokes and the Cahn–Hilliard system as solved on each time instance subsequently until convergence. This allows the treatment of the (typically dominant) convection in the Cahn–Hilliard equation using a higher order finite volume scheme.

In [53] and [50], a thermodynamically consistent splitting scheme is proposed. Here the time discretization is used to sequentially couple the Cahn–Hilliard and Navier–Stokes equation, such that the two systems can be solved one after the other. In [50], additional various discretization methods for the polynomially bounded free energy are proposed and convergence of the scheme for vanishing discretization parameter to the model of [5] is shown.

In [67], for the purpose of optimal control of two-phase flow a stable time discretization is proposed. A scheme that also preserves the thermodynamical consistency in the fully discrete setting is proposed in [45]. The scheme is linear except for the usual non-linearity resulting from the free energy that is polynomially bounded. The mobility is not degenerate. The authors provide rigorous residual based error estimation to formulate an adaptive scheme. The consistency in the fully discrete setting is obtained by a suitable post processing step after the marking of cells for refinement and coarsening.

Based on the model of [5] in [46], a thermodynamically consistent model for two-phase flow with different densities is proposed that can also handle additional surface active agents, so called surfactants. These particles adhere to the interface, following some advection-diffusion equation and some sorption laws. On the interface they lower locally the surface tension of the interface. Thus, this model especially contains a locally varying surface tension and a partial differential equation on a diffuse interface. This work also contains numerical results, where especially the results of [40] on the simulation of partial differential equations on evolving interfaces that are given by a diffuse interface approach are used.

For a model that allows phase transition we refer to [8].

In the situation of multi phase flows with more then two fluid components a vector-valued phase field equation is used. Here we only refer to [24, 27].

Finally we stress that results concerning the simulation of one-phase flows stay valid or are a good starting point for two-phase flow and we only refer to the book [48] for the former.

For further reading on diffuse interface models, we refer to the scientific contibutions of H. Abels (Universität Regensburg), S. Aland (TU Dresden), and G. Grün (Universität Erlangen-Nürnberg) and H. Garcke (Universität Regensburg).

1.1.2 Work related to adaptive concepts for two-phase flows using the diffuse interface approach

The special structure of the phase field that models the spatial distribution makes it necessary to use an appropriately adapted spatial discretization. This is commonly achieved by heuristic mesh refinement. As the interfacial region is known to be characterized by $|\varphi| < 1$ typically local refinement based on the modulus of φ is used, see e.g. [10,23,84]. On the other hand, as at the center of the interfacial region we have $|\nabla\varphi| \approx \frac{1}{\pi\varepsilon}$, the value $|\nabla\varphi|$ is used as an indicator for the interface in [51]. The first variant leads to a homogeneously refined mesh across the interface, while in the second case most refinement takes place around the zero level line of φ where $|\nabla\varphi|$ takes its maximum. We refer to [60], for a comparison of different refinement and marking strategies.

In [61], reliable and efficient residual based error estimation is proposed for the Cahn–Hilliard system with a relaxed non-smooth double-obstacle free energy. In [60], the former work is extended to the simulation of two-phase flow based on model 'H' and it is further extended to the simulation of variable density two-phase flow based on the model of [5] in [45], where additionally arbitrary polynomially bounded free energies are used. We note that based on results of [32] in [43] for a Cahn–Hilliard type model it is argued that an estimator based on the jumps of normal derivatives in general will result in well adapted meshes.

A-posteriori error estimation for the Cahn–Hilliard systems with non-smooth double obstacle free energy is proposed in [15, 16]. There, also residual based error estimation is proposed and reliability of the derived estimator is shown.

1.1.3 Work related to MPECs

The presence of a non-smooth homogeneous free energy density associated with the underlying Ginzburg-Landau energy in the Cahn-Hilliard system gives rise to an optimal control problem governed by a variational inequality. Hence the problem falls into the realm of so-called mathematical programs with equilibrium constraints (MPECs) in function spaces, cf. [67,72]. The main difficulty in dealing with MPECs is that the feasible set which can be characterized by the solution operator of a variational inequality is usually non smooth and non convex and therefore violates the typical constraint qualifications known in classical optimization theory. As a result, stationarity conditions for this problem class are no longer unique. In finite dimen-

sions, MPECs and the associated difficulties are already fairly well understood, see, e.g., the monographs [93, 98, 101] and the references therein.

In contrast, the literature on infinite dimensional MPECs is comparatively scarce. In [21, 94, 95], the authors use the conical derivative of the solution operator of the variational inequality to derive a stationarity system for the control problem, which one would classify now as strong stationarity. A different approach is introduced in [13], where the variational inequalities are approximated by variational equations and optimality conditions are derived by a passage to the limit in the approximation process. This technique typically yields a weaker stationarity system only. Further contributions to the topic include [14, 22, 42, 80, 106] most of which use regularization-penalization methods.

A first step towards the systematization and completion of stationarity concepts in function space was undertaken in [68], where the concept of ε-almost C-stationarity is introduced, paving the way for various contributions in the recent past. Here, we mention [69] where an abstract first-order optimality system is derived by means of variational analysis. In [109], the MPEC is approximated by a sequence of non smooth problems similar to the virtual control approach from [88].

1.1.4 Work related to adaptive concepts for optimal control problems

In Subsection 1.1.2, we already discussed the importance of an adequate mesh refinement technique for solving the Cahn–Hilliard Navier–Stokes system numerically. In this subsection, we briefly comment on the available literature on adaptive finite element methods (AFEMs) for optimal control problems. Whereas AFEMs for partial differential equations have been studied in great detail over the last decades, see, e.g., [12, 99] and the references therein, the research on AFEMs for variational inequalities and optimal control problems is comparatively recent. It started in the beginning of the century with the works by [17, 18] which pioneered a new approach to error control and mesh adaptivity in the numerical solution of unconstrained optimal control problems governed by elliptic differential equations. Here the mesh adaptation is driven by weighted residual-based a posteriori error estimates which are derived by global duality arguments and include the error in the state, the adjoint state and the control. This general approach facilitated the control of the error with respect to any quantity of physical interest such as, e.g., the given objective functional. In the following years the approach was successfully transferred to optimal control problems with control constraints, see, e.g., [62, 63, 108], as well as state constraints, see, e.g., [19] and [64] where additional error terms coming from data oscillations have been considered.

Here we also mention the works by [37, 56, 59] where reliable a-posteriori error bounds for optimal control problems governed by point wise gradient constraints on the state were derived and an adaptive solution algorithm was presented.

In [90, 91] and [100], some other approaches are depicted which directly utilize the residuals of the associated first order optimality systems of the optimal control problems. The first paper presents a-posteriori error estimates for finite element

approximation of distributed optimal control problems with convex constraint sets, whereas the second one provides a reliable a-posteriori error estimator for optimal control problems with state and control constraints containing the L^∞-error of the state, the H^{-2}-residual of the adjoint equation as well as the L^2-residual of the variational inequality if the Slater condition is satisfied. Very recently, [103] presented a-posteriori error estimates for control-constrained, linear-quadratic optimal control problems using an altered norm motivated by the objective functional in order to measure the error.

In contrast to PDE-constrained optimal control problems, the literature on goal-oriented mesh adaptivity methods appears rather scarce with respect to MPECs in function spaces. However, in [30, 65] the method was successfully applied to the optimal control of elliptic variational inequalities.

2 An energy conserving adaptive discretization scheme for variable density two-phase flows

The subsequent investigations are based on the following diffuse interface model for two-phase flows as proposed in [5]:

$$\begin{aligned}
\rho\partial_t v + ((\rho v + J)\cdot\nabla)\, v - \operatorname{div}(2\eta Dv) + \nabla p &= \mu\nabla\varphi + \rho g && \forall x\in\Omega, \forall t\in I, && (1)\\
-\operatorname{div}(v) &= 0 && \forall x\in\Omega, \forall t\in I, && (2)\\
\partial_t\varphi + v\cdot\nabla\varphi - \operatorname{div}(m\nabla\mu) &= 0 && \forall x\in\Omega, \forall t\in I, && (3)\\
-\sigma\varepsilon\Delta\varphi + \Psi'(\varphi) - \mu &= 0 && \forall x\in\Omega, \forall t\in I, && (4)\\
v(0,x) &= v_0(x) && \forall x\in\Omega, && (5)\\
\varphi(0,x) &= \varphi_0(x) && \forall x\in\Omega, && (6)\\
v(t,x) &= 0 && \forall x\in\partial\Omega, \forall t\in I, && (7)\\
\nabla\mu(t,x)\cdot\overrightarrow{\nu}_\Omega = \nabla\varphi(t,x)\cdot\overrightarrow{\nu}_\Omega &= 0 && \forall x\in\partial\Omega, \forall t\in I, && (8)
\end{aligned}$$

where $J = -\frac{d\rho}{d\varphi} m\nabla\mu$. Here $\Omega\subset\mathbb{R}^n$, $n\in\{2,3\}$, denotes an open and bounded domain with outer normal $\overrightarrow{\nu}_\Omega$, $I=(0,T]$ with $0<T<\infty$ a time interval, φ denotes the phase field, μ the chemical potential, v the volume averaged velocity, p the pressure, and $\rho=\rho(\varphi)=\frac{1}{2}((\rho_2-\rho_1)\varphi+(\rho_1+\rho_2))$ the mean density, where $0<\rho_1\le\rho_2$ denote the densities of the involved fluids. The viscosity is denoted by η and can be chosen arbitrarily, fulfilling $\eta(-1)=\tilde{\eta}_1$ and $\eta(1)=\tilde{\eta}_2$, with individual fluid viscosity $\tilde{\eta}_1,\tilde{\eta}_2$. Here we restrict to Newtonian fluids, but non-Newtonian fluids are covered by the model as well. The mobility is denoted by $m=m(\varphi)$. The gravitational force is denoted by g. By $Dv=\frac{1}{2}(\nabla v+(\nabla v)^t)$ we denote the symmetrized gradient. The scaled surface tension is denoted by σ and the interfacial width is proportional to ε. The scaling of the physical surface tension is required due to the

diffuse interface approach, see [5, Sec. 4.3.4]. The free energy density is denoted by Ψ and fulfills $\operatorname{argmin}(\Psi) = \pm 1$. The initial data is given by $(\varphi_0, v_0) = (\varphi_a, v_a)$.

The above model couples the Navier–Stokes equations (1)–(2) to the Cahn–Hilliard model (3)–(5) in a thermodynamically consistent way, i.e. the energy inequality from Theorem 1 holds for the total energy of the system, which is the sum of a Ginzburg–Landau energy for the interface between the two fluids and the kinetic energy of the fluids.

Theorem 1. *Let v, φ, μ be a sufficiently smooth solution to* (1)–(8)*. Then it holds that*

$$\frac{d}{dt}\left(\int_\Omega \frac{\rho}{2}|v|^2 + \frac{\sigma\varepsilon}{2}|\nabla\varphi|^2 + \frac{\sigma}{\varepsilon}\Psi(\varphi)\,dx\right) = -\int_\Omega 2\eta|Dv|^2 + m|\nabla\mu|^2\,dx + \int_\Omega \rho g v\,dx.$$

Furthermore, the existence of weak solutions to system (1)–(8) for specific choices of data, i.e. Ψ, m, η, is shown in [2–4, 49].

As mentioned above, it is our goal to extend the existing theory by a more application driven perspective focusing on the development of efficient numerical solvers for the problem. For this purpose, we subsequently present some advanced numerical solution techniques for the simulation of the problem itself, as well as for generic optimal control problems which contain the system (1) – (8) as constraints. This will be accompanied by a rigorous analysis of the underlying problems concerning, e.g., the thermodynamical consistency of the discretization scheme, mesh stability, existence of solutions, characterization of stationarity conditions, and a-priori error estimation.

The rest of this section is organized as follows. In Section 2.1 we propose a discretization scheme for the numerical treatment of (1) – (8) that preserves the consistency with thermodynamics in the fully discrete setting and that is nearly linear. This scheme is proposed and analytically investigated for polynomially bounded free energies. Residual based error estimation is proposed for the spatial discretization. Further optimal control and instantaneous control of two-phase fluids are proposed.

In Section 3, we analyse a general optimal control problem associated to a semi-discrete version of (1) – (8) where the Ginzburg-Landau energy is characterized by the double-obstacle potential. This includes the existence of feasible points, as well as global optimal solutions, and culminates in the derivation of ε-almost C-stationarity conditions via a Yosida regularization technique with a subsequent passage to the limit with the Yosida parameter.

Section 4 provides a rigorous derivation of a goal-oriented dual-weighted error estimator for the problem considered in Section 3 and presents some numerical results along with the details of the numerical implementation of the solution algorithm for the optimal control problem.

Notation

Let $\Omega \subset \mathbb{R}^n$, $n \in \{2,3\}$ denote a bounded domain with sufficiently smooth boundary $\partial\Omega$ and outer normal $\overrightarrow{\nu}_\Omega$. Let $I = (0,T]$ denote a time interval.

We use the conventional notation for Sobolev and Hilbert Spaces, see e.g. [6]. By $L^p(\Omega)$, $1 \le p \le \infty$, we denote the space of measurable functions on Ω, whose modulus to the power p is Lebesgue-integrable. $L^\infty(\Omega)$ denotes the space of measurable functions on Ω, which are essentially bounded. For $p = 2$ we denote by $L^2(\Omega)$ the Hilbert space of square integrable functions on Ω with inner product $(\cdot,\cdot)$ and norm $\|\cdot\|$. For a subset $D \subset \Omega$ and functions $f, g \in L^2(\Omega)$ we denote by $(f,g)_D$ the inner product of f and g restricted to D, and by $\|f\|_D$ the respective norm. By $W^{k,p}(\Omega)$, $k \ge 1, 1 \le p \le \infty$, we denote the Sobolev space of functions admitting weak derivatives up to order k in $L^p(\Omega)$. If $p = 2$ we write $H^k(\Omega)$ to acknowledge the Hilbertian structure of the space. The subset $H_0^1(\Omega)$ denotes $H^1(\Omega)$ functions with vanishing boundary trace. For $k \in \mathbb{N}$, we further set

$$
\begin{aligned}
H^k_{0,\sigma}(\Omega;\mathbb{R}^n) &:= \left\{ f \in H^k(\Omega;\mathbb{R}^n) \cap H^1_0(\Omega;\mathbb{R}^n) : \operatorname{div} f = 0, \text{ a.e. on } \Omega \right\};\\
\overline{H}^k(\Omega) &:= H^k_{(0)}(\Omega) := \left\{ f \in H^k(\Omega) : \int_\Omega f dx = 0 \right\};\\
\overline{H}^k_{\partial_n}(\Omega) &:= \left\{ f \in \overline{H}^k(\Omega) : \partial_n f_{|\partial\Omega} = 0 \text{ on } \partial\Omega \right\},\ k \ge 2;
\end{aligned}
$$

where 'a.e.' stands for 'almost everywhere' and the boundary condition is supposed to hold true in the trace sense. We stress that the subscript σ used here is not related to the surface tension, but using σ here is common notation.
Unless otherwise noted, $\langle\cdot,\cdot\rangle := \langle\cdot,\cdot\rangle_{\overline{H}^{-1},\overline{H}^1}$ represents the duality pairing between $\overline{H}^1(\Omega)$ and $\overline{H}^{-1}(\Omega)$. For $u \in L^q(\Omega)^n$, $q > n$, and $v, w \in H^1(\Omega)^n$ we introduce the trilinear form

$$
a(u,v,w) = \frac{1}{2}\int_\Omega ((u\cdot\nabla)v)\, w\, dx - \frac{1}{2}\int_\Omega ((u\cdot\nabla)w)\, v\, dx. \tag{9}
$$

Note that it holds that $a(u,v,w) = -a(u,w,v)$, and especially $a(u,v,v) = 0$.

2.1 A stable time discretization for smooth free energies

In this section we summarize the fully practical scheme for the numerical treatment of (1)–(8) as proposed in [45]. Here we assume that the free energy density Ψ is smooth, see Assumption A2. To emphasize this, in the following we denote the free energy density by W, and we introduce the splitting $W = W_+ + W_-$, where W_+ denotes the convex part of W and W_- denotes the concave part.

Assumption 1 *Concerning the data of our problem we invoke the following assumptions:*

A1 There exist constants $\overline{\rho} \geq \underline{\rho} > 0$, $\overline{\eta} \geq \underline{\eta} > 0$, *and* $\overline{m} \geq \underline{m} > 0$ *such that the following relations are satisfied:*

- $\overline{\rho} \geq \rho(\varphi) \geq \underline{\rho} > 0$,
- $\overline{\eta} \geq \eta(\varphi) \geq \underline{\eta} > 0$,
- $\overline{m} \geq m(\varphi) \geq \underline{m} > 0$.

Especially we assume that the mobility is non degenerate. In addition, we assume that ρ, η, *and* m *are continuous.*

A2 $W : \mathbb{R} \to \mathbb{R}$ *is continuously differentiable.*

A3 W *and the derivatives* W'_+ *and* W'_- *are polynomially bounded, i.e. there exists* $C > 0$ *such that* $|W(x)| \leq C(1+|x|^q)$, $|W'_+(x)| \leq C(1+|x|^{q-1})$ *and* $|W'_-(x)| \leq C(1+|x|^{q-1})$ *holds for some* $q \in [1,4]$ *if* $n = 3$ *and* $q \in [1,\infty)$ *if* $n = 2$,

A4 W'_+ *is Newton (sometimes called slantly) differentiable (see e.g. [66]) regarded as nonlinear operator* $W'_+ : H^1(\Omega) \to \left(H^1(\Omega)\right)^*$ *with Newton derivative* G *satisfying*

$$(G(\varphi)\delta\varphi, \delta\varphi) \geq 0$$

for each $\varphi \in H^1(\Omega)$ *and* $\delta\varphi \in H^1(\Omega)$.

To ensure Assumption A1 we introduce a cut-off mechanism to guarantee the bounds on ρ defined in Assumption A1 independently of φ. Note that $\eta(\varphi)$ and $m(\varphi)$ can be chosen arbitrarily fulfilling the stated bounds. We define the mass density as a smooth, monotone and strictly positive function $\rho(\varphi)$ fulfilling

$$\rho(\varphi) = \begin{cases} \frac{\rho_2-\rho_1}{2}\varphi + \frac{\rho_1+\rho_2}{2} & \text{if } -1 - \frac{\rho_1}{\rho_2-\rho_1} < \varphi < 1 + \frac{\rho_1}{\rho_2-\rho_1}, \\ \text{const} & \text{if } \varphi > 1 + \frac{2\rho_1}{\rho_2-\rho_1}, \\ \text{const} & \text{if } \varphi < -1 - \frac{2\rho_1}{\rho_2-\rho_1}. \end{cases}$$

For a discussion we refer to [49, Remark 2.1].

Remark 1. We stress that in the following we require the affine linearity of $\rho(\varphi) := \frac{1}{2}\left((\rho_2-\rho_1)\varphi + (\rho_1+\rho_2)\right)$ to derive our numerical scheme. Also, we note that it is sufficient to have the affine-linearity of ρ for all values of φ that appear in a simulation. This is the case in all of our numerical examples. Concerning the necessity of the cut-off procedure we also note Remark 5. For the case of a nonlinear dependence between φ and ρ we also refer to [2], where an extension of model (1)–(8) to non-linear functions $\rho(\varphi)$ is proposed. Extending our results to this case is subject to future work.

Remark 2. Assumptions A2–A4 are for example fulfilled by the polynomial free energy

$$W^{poly}(\varphi) = \frac{1}{4}\left(1-\varphi^2\right)^2.$$

Another free energy fulfilling these assumptions is the relaxed double-obstacle free energy given by

$$W^{rel}(\varphi) = \frac{1}{2}\left(1-\varphi^2+s\lambda^2(\varphi)\right),$$

with

$$\lambda(\varphi) := \max(0,\varphi-1)+\min(0,\varphi+1),$$

where $s \gg 0$ denotes the relaxation parameter. W^{rel} is introduced in [61] as Moreau–Yosida relaxation of the double-obstacle free energy

$$W^{obst}(\varphi) = \begin{cases} \frac{1}{2}\left(1-\varphi^2\right) & \text{if } |\varphi| \le 1, \\ 0 & \text{else,} \end{cases}$$

which is proposed in [96] to model phase separation and is first analytically investigated in [25].

In the numerical examples of this section we use the free energy $W \equiv W^{rel}$. For this choice the splitting into convex and concave part reads

$$W_+(\varphi) = s\frac{1}{2}\lambda^2(\varphi), \qquad\qquad W_-(\varphi) = \frac{1}{2}(1-\varphi^2).$$

We note that the minima of W^{rel} are at $\pm\frac{s}{s-1} \neq 1$. One might rescale the argument of W^{rel} to obtain $\operatorname{argmin}(W) = \pm 1$. As s is typically large and thus $\frac{s}{s-1} \approx 1$ we refrain from this rescaling.

With the above assumptions we introduce a weak formulation of (1)–(8) that we use to derive our discrete scheme. For this purpose, we restrict ourselves to solenoidal velocity fields in what follows, i.e., we neglect the pressure p.

For a sufficiently smooth solution (φ,μ,v) of (1)–(8) and using the linearity of ρ it holds that

$$\partial_t\rho + \operatorname{div}(\rho v + J) = 0,$$

see [5, p. 14]. Using this mass balance we can rewrite (1) as

$$\partial_t(\rho v) + \operatorname{div}(\rho v \otimes v) + \operatorname{div}(v \otimes J) - \operatorname{div}(2\eta Dv) + \nabla p = \mu\nabla\varphi + \rho g. \tag{10}$$

We also observe that the term $\rho v + J$ in (1) is not solenoidal (which might lead to difficulties both in the analytical and the numerical treatment) and that the trilinear form $(((\rho v + J)\cdot\nabla)u, w)$ thus is not anti-symmetric. To obtain a weak formulation yielding an anti-symmetric convection term we use a convex combination of (1) and (10) to define a weak formulation.

This leads to the following weak formulation.

Definition 1. We call v, p, φ, μ a weak solution to (1)–(8) if $v(0) = v_0$, $\varphi(0) = \varphi_0$, and

$$\frac{1}{2}\int_\Omega (\partial_t(\rho v) + \rho\partial_t v)\, w\, dx + \int_\Omega 2\eta Dv : Dw\, dx$$
$$+a(\rho v + J, v, w) - (p, \mathrm{div} w) = \int_\Omega \mu\nabla\varphi w + \rho g w\, dx \quad \forall w \in H_0^1(\Omega)^n, \qquad (11)$$
$$-(\mathrm{div} v, q) = 0 \quad \forall q \in L^2_{(0)}(\Omega) \qquad (12)$$
$$\int_\Omega (\partial_t\varphi + v\cdot\nabla\varphi)\,\Phi\, dx + \int_\Omega m(\varphi)\nabla\mu\cdot\nabla\Phi\, dx = 0 \quad \forall \Phi \in H^1(\Omega), \qquad (13)$$
$$\sigma\varepsilon\int_\Omega \nabla\varphi\cdot\nabla\Psi\, dx + \frac{\sigma}{\varepsilon}\int_\Omega W'(\varphi)\Psi\, dx - \int_\Omega \mu\Psi\, dx = 0 \quad \forall \Psi \in H^1(\Omega), \qquad (14)$$

is satisfied for almost all $t \in I$.

Using this weak formulation, the energy equality in Theorem 1 can be derived by using $w \equiv v$, $q \equiv p$, $\Phi \equiv \mu$, and $\Psi \equiv \varphi$ and summation over the resulting equations.

2.1.1 The energy stable scheme

For a numerical treatment we next discretize the weak formulation (11)–(14) in time and space. We aim at an adaptive discretization of the domain Ω, and thus obtain a different spatial discretization in every time step.

For the discretization in time, let $0 = t_0 < t_1 < \ldots < t_{k-2} < t_{k-1} < t_k < \ldots < t_K = T$ denote an equidistant subdivision of the interval $\bar{I} = [0,T]$ with $\tau_{k+1} - \tau_k \equiv \tau$. From here onwards the superscript k denotes the corresponding variables at time instance t_k, and we abbreviate $\rho^k := \rho(\varphi^k)$, $\eta^k := \eta(\varphi^k)$, and $m^k := m(\varphi^k)$.

For the discretizaton in space let $\mathscr{T}^k := \bigcup_{i=1}^{N_T} T_i$ denote a conforming triangulation of $\overline{\Omega}$ with closed simplices $T_i, i = 1,\ldots,N_T$ and edges $E_i, i = 1,\ldots,N_E$, $\mathscr{E}^k := \bigcup_{i=1}^{N_E} E_i$. Here, k refers to the time instance t_k. On $\mathscr{T}^k$ we define the following finite element spaces:

$$V_1^k = \{v \in C(\mathscr{T}^k)\,|\, v|_T \in P^1(T)\,\forall T \in \mathscr{T}^k\},$$
$$V_2^k = \{v \in C(\mathscr{T}^k)^n\,|\, v|_T \in (P^2(T))^n\,\forall T \in \mathscr{T}^k, v|_{\partial\Omega} = 0\},$$

where $P^l(S)$ denotes the space of polynomials up to order l defined on S.

We further introduce an H^1-stable projection operator $\mathscr{P}^k : H^1(\Omega) \to V_1^k$ satisfying

$$\|\mathscr{P}^k v\|_{L^p(\Omega)} \le \|v\|_{L^p(\Omega)} \text{ and } \|\nabla\mathscr{P}^k v\|_{L^r(\Omega)} \le \|\nabla v\|_{L^r(\Omega)}$$

for $v \in H^1(\Omega)$ with $r \in [1,2]$ and $p \in [1,6)$ if $n = 3$, and $p \in [1,\infty)$ if $n = 2$. Possible choices are the H^1-projection, the Clément operator or, by restricting the preimage

to $C(\overline{\Omega}) \cap H^1(\Omega)$, the Lagrangian interpolation operator, see e.g. [29]. For triangulations with specific properties, also the L^2-projection fulfills these assumption, see [28].

Using these spaces we state the fully discrete counterpart of (11)–(14): Let $k \geq 2$, given $\varphi^{k-2} \in V_1^{k-2}$, $\varphi^{k-1} \in V_1^{k-1}$, $\mu^{k-1} \in V_1^{k-1}$, $v^{k-1} \in V_2^k$, find $v_h^k \in V_1^k$, $p_h^k \in V_k^1 \cap L^2_{(0)}(\Omega)$, $\varphi_h^k \in V_1^k$, $\mu_h^k \in V_1^k$ such that for all $w \in V_2^k$, $q \in V_1^k \cap L^2_{(0)}(\Omega)$, $\Phi \in V_1^k$, $\Psi \in V_1^k$ it holds that:

$$\begin{aligned}\frac{1}{2\tau}(\rho^{k-1}v_h^k - \rho^{k-2}v^{k-1} + \rho^{k-2}(v_h^k - v^{k-1}), w) + a(\rho^{k-1}v^{k-1} + J^{k-1}, v_h^k, w)\\ +(2\eta^{k-1}Dv_h^k, Dw) - (p_h^k, \mathrm{div} w) - (\mu_h^k \nabla\varphi^{k-1} + \rho^{k-1}g, w) = 0,\end{aligned} \tag{15}$$

$$-(\mathrm{div} v_h^k, q) = 0, \tag{16}$$

$$\frac{1}{\tau}(\varphi_h^k - \mathscr{P}^k\varphi^{k-1}, \Phi) + (m^{k-1}\nabla\mu_h^k, \nabla\Phi) + (v_h^k\nabla\varphi^{k-1}, \Phi) = 0, \tag{17}$$

$$\sigma\varepsilon(\nabla\varphi_h^k, \nabla\Psi) + \frac{\sigma}{\varepsilon}(W_+'(\varphi_h^k) + W_-'(\mathscr{P}^k\varphi^{k-1}), \Psi) - (\mu_h^k, \Psi) = 0, \tag{18}$$

where $J^{k-1} := -\frac{d\rho}{d\varphi}m^{k-1}\nabla\mu^{k-1}$, $\varphi^0 = P\varphi_0$ denotes the L^2 projection of φ_0 onto V_1^0, $v^0 = P^L v_0$ denotes the Leray projection of v_0 onto H^0_σ, see [35]. The functions $\varphi_h^1 \in V_1^1$, $\mu_h^1 \in V_1^1$ and $v_h^1 \in V_2^1$ are obtained by a one-step-scheme for equation (1)–(8), see [45].

Remark 3. Due to the appearance of ρ^{k-2} in (15) the scheme (15)–(18) is a two-step scheme and thus needs additional initialization. By using ρ^{k-2} we obtain a scheme that is only nonlinear in the discretization of the convex part of the free energy density. However we can replace (15) by

$$\begin{aligned}&\frac{1}{2\tau}\int_\Omega \left(\rho_h^k v_h^k - \rho^{k-1}v^{k-1}\right)w + \rho^{k-1}(v_h^k - v^{k-1})w\,dx\\ &+a(\rho^{k-1}v^{k-1} + J^{k-1}, v_h^k, w) + \int_\Omega 2\eta^{k-1}Dv_h^k : Dw\,dx\\ &\qquad -(p_h^k, \mathrm{div} w) - \int_\Omega \mu_h^k\nabla\varphi^{k-1}w + \rho^{k-1}gw\,dx = 0 \quad \forall w \in H^k_\sigma,\end{aligned} \tag{19}$$

which leads to a one-step-scheme that then also is nonlinear in the Navier–Stokes equation. For the scheme including (15) the unique solution can be found by Newton's method, while - up to now - this has not been proven for the scheme including (19).

We further argue that if we have initial data φ_{-1} at some virtual time instance $t_{-1} := -\tau$ together with v_0, we can solve (17)–(18) to obtain φ_h^0 and μ_h^0 and then proceed with the scheme (15)–(18).

To guarantee mass conservation for φ_h^k we implement the term $(v_h^k \nabla \varphi^{k-1}, \Phi)$ in (17) by using integration by parts as $-(v_h^k \varphi^{k-1}, \nabla \Phi)$.

The following is proven in [45].

Theorem 2. *There exist unique* $v_h^k \in V_2^k$, $p_h^k \in V_1^k \cap L^2_{(0)}(\Omega)$, $\varphi_h^k \in V_1^k$, $\mu_h^k \in V_1^k$ *solving* (15)–(18).

Remark 4. As (15)–(18) is linear in all terms but the free energy density W_+ which is Newton differentiable and convex by Assumption A4 using the same arguments as for Theorem 2 one can show that Newton's method can be used to find the unique solution from Theorem 2.

The time discretization is chosen such that the following fully discrete correspondence to the energy equality in Theorem 1 holds.

Theorem 3. *Let* $(\varphi_h^k, \mu_h^k, v_h^k, p_h^k)$ *be a solution to* (15)–(18). *Then for* $k \geq 2$*:*

$$\begin{aligned}
&\frac{1}{2}\int_\Omega \rho^{k-1}\left|v_h^k\right|^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\varphi_h^k|^2 dx + \frac{\sigma}{\varepsilon}\int_\Omega W(\varphi_h^k)\,dx \\
&+\frac{1}{2}\int_\Omega \rho^{k-2}|v_h^k - v^{k-1}|^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\varphi_h^k - \nabla\mathscr{P}^k\varphi^{k-1}|^2 dx \\
&+\tau\int_\Omega 2\eta^{k-1}|Dv_h^k|^2 dx + \tau\int_\Omega m^{k-1}|\nabla\mu_h^k|^2 dx \qquad (20)\\
\leq\ &\frac{1}{2}\int_\Omega \rho^{k-2}\left|v^{k-1}\right|^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\mathscr{P}^k\varphi^{k-1}|^2 dx + \frac{\sigma}{\varepsilon}\int_\Omega W(\mathscr{P}^k\varphi^{k-1})\,dx \\
&+\tau\int_\Omega \rho^{k-1} g v_h^k.
\end{aligned}$$

So we observe that the energy

$$E(\varphi_h^k, v_h^k) := \frac{1}{2}\int_\Omega \rho^{k-1}|v_h^k|^2 dx + \sigma\int_\Omega \frac{\varepsilon}{2}|\nabla\varphi^k|^2 + \frac{1}{\varepsilon}W(\varphi^k)\,dx$$

decays as in the continuous case due to diffusion and increases due to outer forces, like the gravitational force. We further have some addition loss of energy due to numerical dissipation. We further observe that $E(\varphi_h^k, v_h^k)$ is in fact bounded by $E(\mathscr{P}^k\varphi_h^{k-1}, v_h^{k-1})$ which is not necessarily bounded by the energy at time t_{k-1}, i.e. $E(\varphi_h^{k-1}, v_h^{k-1})$. This will be dealt with during our adaptive scheme to obtain a guaranteed decrease of the total energy. We formulate this in the following assumption.

Assumption 2 *For every solution* $v_h^{k-1}, p_h^{k-1}, \varphi_h^{k-1}, \mu_h^{k-1}$ *to* (15)–(18) *at time* t_{k-1}*, we assume that the following inequality is satisfied*

$$E(\mathscr{P}^k\varphi_h^{k-1}, v_h^{k-1}) \leq E(\varphi_h^{k-1}, v_h^{k-1}),$$

which is equivalent to

$$\sigma\int_\Omega \frac{\varepsilon}{2}|\nabla\mathscr{P}^k\varphi^{k-1}|^2 + \frac{1}{\varepsilon}W(\mathscr{P}^k\varphi^{k-1})\,dx \leq \sigma\int_\Omega \frac{\varepsilon}{2}|\nabla\varphi^{k-1}|^2 + \frac{1}{\varepsilon}W(\varphi^{k-1})\,dx.$$

We can use the unique solution from Theorem 2 in a Galerkin approach to investigate the limit $h \to 0$ in the scheme (15)–(18). This leads to the following time discrete scheme:
Given $\varphi^{k-2} \in H^1(\Omega) \cap L^\infty(\Omega)$, $\varphi^{k-1} \in H^1(\Omega) \cap L^\infty(\Omega)$, $\mu^{k-1} \in W^{1,q}(\Omega)$, $q > n$, $v^{k-1} \in H_0^1(\Omega)^n$,
find $v^k \in H_0^1(\Omega)^n$, $p^k \in L^2_{(0)}(\Omega)$, $\varphi^k \in H^1(\Omega)$, $\mu^k \in H^1(\Omega)$ satisfying

$$\begin{aligned}
&\frac{1}{2\tau}\int_\Omega \left(\rho^{k-1}v^k - \rho^{k-2}v^{k-1}\right) w + \rho^{k-2}(v^k - v^{k-1}) w \, dx \\
&\quad + a(\rho^{k-1}v^{k-1} + J^{k-1}, v^k, w) + \int_\Omega 2\eta^{k-1} Dv^k : Dw \, dx \\
&\qquad - (p^k, \mathrm{div} w) - \int_\Omega \mu^k \nabla\varphi^{k-1} w - \rho^{k-1} g w \, dx = 0 \quad \forall w \in H_0^1(\Omega)^n, \quad (21)\\
&\qquad - (\mathrm{div} v^k, q) = 0 \qquad \forall q \in L^2_{(0)}(\Omega), \quad (22)\\
&\frac{1}{\tau}\int_\Omega (\varphi^k - \varphi^{k-1})\Phi \, dx + \int_\Omega (v^k \cdot \nabla\varphi^{k-1})\Phi \, dx \\
&\qquad + \int_\Omega m^{k-1}\nabla\mu^k \cdot \nabla\Phi \, dx = 0 \quad \forall \Phi \in H^1(\Omega), \quad (23)\\
&\sigma\varepsilon \int_\Omega \nabla\varphi^k \cdot \nabla\Psi \, dx - \int_\Omega \mu^k \Psi \, dx \\
&\quad + \frac{\sigma}{\varepsilon}\int_\Omega ((W_+)'(\varphi^k) + (W_-)'(\varphi^{k-1}))\Psi \, dx = 0 \quad \forall \Psi \in H^1(\Omega), \quad (24)
\end{aligned}$$

where $J^{k-1} := -\frac{d\rho}{d\varphi} m^{k-1}\nabla\mu^{k-1}$.

Theorem 4. *Let $\varphi^{k-2} \in H^1(\Omega) \cap L^\infty(\Omega)$, $\varphi^{k-1} \in H^1(\Omega) \cap L^\infty(\Omega)$, $\mu^{k-1} \in W^{1,q}(\Omega)$, $q > n$, $v^{k-1} \in H_0^1(\Omega)^n$, be given. Then there exists a unique solution $(v^k, p^k, \varphi^k, \mu^k) \in H_0^1(\Omega)^n \times L^2_{(0)}(\Omega) \times H^1(\Omega) \times H^1(\Omega)$ to* (21)–(24)*. In fact it holds $\varphi^k \in H^2(\Omega) \subset H^1(\Omega) \cap L^\infty(\Omega)$ and $\mu^k \in W^{1,q}(\Omega), q > n$.*

Also the energy inequality stays valid in the time discrete setting.

Theorem 5. *Let $(\varphi^k, \mu^k, v^k, p^k)$ be a solution to* (21)–(24)*. Then for $k \geq 2$:*

$$\begin{aligned}
&\frac{1}{2}\int_\Omega \rho^{k-1}\left|v^k\right|^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\varphi^k|^2 dx + \frac{\sigma}{\varepsilon}\int_\Omega W(\varphi^k) dx \\
&\quad + \frac{1}{2}\int_\Omega \rho^{k-2}|v^k - v^{k-1}|^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\varphi^k - \nabla\varphi^{k-1}|^2 dx \\
&\qquad + \tau\int_\Omega 2\eta^{k-1}|Dv^k|^2 dx + \tau\int_\Omega m^{k-1}|\nabla\mu^k|^2 dx \qquad (25)\\
&\leq \frac{1}{2}\int_\Omega \rho^{k-2}\left|v^{k-1}\right|^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\varphi^{k-1}|^2 dx + \frac{\sigma}{\varepsilon}\int_\Omega W(\varphi^{k-1}) dx \\
&\qquad + \tau\int_\Omega \rho^{k-1} g v^k.
\end{aligned}$$

Remark 5. Let W denote the relaxed double-obstacle free energy W^{rel} introduced in Remark 2 with relaxation parameter s. Let $(v_s, p_s, \varphi_s, \mu_s)_{s\in\mathbb{R}}$ denote the sequence of solutions of (21)–(24) for a sequence $(s_l)_{l\in\mathbb{N}}$ with $s_l \to \infty$. From the linearity of (21) and [61, Prop. 4.2] it follows that there exists a subsequence, still denoted by $(v_s, p_s, \varphi_s, \mu_s)_{s\in\mathbb{R}}$, such that

$$(v_s, p_s, \varphi_s, \mu_s)_{s\in\mathbb{R}} \to (v^\star, p^\star, \varphi^\star, \mu^\star) \quad \text{in } H^1(\Omega),$$

where $(v^\star, p^\star, \varphi^\star, \mu^\star)$ denotes the solution of (21)–(24), where W^{obst}, denoted in Remark 2, is chosen as free energy. Especially $|\varphi^*| \leq 1$ holds true.

In the following argumentation we concentrate on the phase field only. From the regularity $\varphi_s \in H^2(\Omega)$ together with a-priori estimates on the solution of the Poisson problem and the energy inequality of Theorem 5, we obtain the existence of a strongly convergent subsequence $\varphi_{s'} \to \varphi^*$ in $C^{0,\alpha}(\overline{\Omega})$, where we use the compact embedding $H^2(\Omega) \hookrightarrow C^{0,\alpha}(\overline{\Omega})$ for $2\alpha < 4-n$.

Thus, for s large enough we have $|\varphi_s| \leq 1+\theta$ with θ arbitrarily small. In fact, using arguments from [70] in [82] a rate

$$\|\max(|\varphi_s|-1,0)\|_{L^\infty(\Omega)} \leq Cs^{-1}$$

is argued for the pure Cahn–Hilliard system. This is also observed in the numerical tests of these works.

2.2 A-posteriori error estimation

For an efficient solution of (15)–(18) we next describe an a-posteriori error estimator based mesh refinement scheme that is reliable and efficient up to terms of higher order and errors introduced by the projection. We propose an all-in-one adaptation concept for the fully coupled Cahn–Hilliard Navier–Stokes system, where we exploit the energy inequality of Theorem 3. Further we describe how we can guarantee that the total energy can not increase in absence of outer forces, i.e. how we can guarantee the validity of Assumption 2.

We define the following error terms:

$$e_v := v_h^k - v^k, \qquad e_p := p_h^k - p^k, \tag{26}$$

$$e_\varphi := \varphi_h^k - \varphi^k, \qquad e_\mu := \mu_h^k - \mu^k, \tag{27}$$

as well as the discrete element residuals

$$
\begin{aligned}
r_h^{(1)} :=& \frac{\rho^{k-1}+\rho^{k-2}}{2} v_h^k - \rho^{k-2} v^{k-1} + \tau (b^{k-1}\nabla) v_h^k + \frac{1}{2}\tau \mathrm{div}(b^{k-1}) v_h^k \\
& - 2\tau \mathrm{div}\left(\eta^{k-1} D v_h^k\right) + \tau \nabla p_h^k - \tau \mu_h^k \nabla \varphi^{k-1} - \rho^{k-1} g, \\
r_h^{(2)} :=& \varphi_h^k - \mathscr{P}^k \varphi^{k-1} + \tau v_h^k \nabla \varphi^{k-1} - \tau \mathrm{div}(m^{k-1} \nabla \mu_h^k), \\
r_h^{(3)} :=& \frac{\sigma}{\varepsilon}\left(W_+'(\varphi_h^k) + W_-'(\mathscr{P}^k \varphi^{k-1})\right) - \mu_h^k,
\end{aligned}
$$

where $b^{k-1} := \rho^{k-1} v^{k-1} + J^{k-1}$. Furthermore we define the error indicators

$$
\begin{aligned}
\eta_T^{(1)} :=& h_T \| r_h^{(1)} \|_T, \quad \eta_E^{(1)} := h_E^{1/2} \| 2\eta^{k-1} \left[D v_h^k \right]_{\overrightarrow{\nu}_E} \|_E, \\
\eta_T^{(2)} :=& h_T \| r_h^{(2)} \|_T, \quad \eta_E^{(2)} := h_E^{1/2} \| m^{k-1} \left[\nabla \mu_h^k \right]_{\overrightarrow{\nu}_E} \|_E, \\
\eta_T^{(3)} :=& h_T \| r_h^{(3)} \|_T, \quad \eta_E^{(3)} := h_E^{1/2} \| \left[\nabla \varphi_h^k \right]_{\overrightarrow{\nu}_E} \|_E.
\end{aligned} \tag{28}
$$

Here $[\cdot]_{\overrightarrow{\nu}_E}$ denotes the jump of a discontinuous function across an edge E of $\mathscr{T}^k$ in normal direction $\overrightarrow{\nu}_E$ pointing from the triangle with lower global number to the triangle with higher global number. Thus $\eta_E^{(j)}$, $j = 1,2,3$, measures the jump of the corresponding variable across the edge E, while $\eta_T^{(j)}$, $j = 1,2,3$, measures the triangle-wise residuals.

Theorem 6. *There exists a constant $C > 0$ only depending on the domain Ω and the regularity of the mesh $\mathscr{T}^k$ such that*

$$
\begin{aligned}
\underline{\rho}\|e_v\|^2 + \tau\underline{\eta}\|\nabla e_v\|^2 + \tau\underline{m}\|\nabla e_\mu\|^2 + \sigma\varepsilon\|\nabla e_\varphi\|^2 + \frac{\sigma}{\varepsilon}(W_+'(\varphi_h^k) - W_+'(\varphi^k), e_\varphi) \\
\leq C\left(\eta_\Omega^2 + \eta_{h.o.t} + \eta_C\right),
\end{aligned}
$$

holds with

$$
\begin{aligned}
\eta_\Omega^2 =& \frac{1}{\tau\underline{\eta}} \sum_{T\in\mathscr{T}^k} \left(\eta_T^{(1)}\right)^2 + \frac{\tau}{\underline{\eta}} \sum_{E\in\mathscr{E}^k} \left(\eta_E^{(1)}\right)^2 \\
& \frac{1}{\tau\underline{m}} \sum_{T\in\mathscr{T}^k} \left(\eta_T^{(2)}\right)^2 + \frac{\tau}{\underline{m}} \sum_{E\in\mathscr{E}^k} \left(\eta_E^{(2)}\right)^2 \\
& \frac{1}{\sigma\varepsilon} \sum_{T\in\mathscr{T}^k} \left(\eta_T^{(3)}\right)^2 + \sigma\varepsilon \sum_{E\in\mathscr{E}^k} \left(\eta_E^{(3)}\right)^2, \\
\eta_{h.o.t.} =& \tau(div(e_v), e_p), \\
\text{and } \eta_C =& (\mathscr{P}^k\varphi^{k-1} - \varphi^{k-1}, e_\mu) - \frac{\sigma}{\varepsilon}(W_-'(\mathscr{P}^k\varphi^{k-1}) - W_-'(\varphi^{k-1}), e_\varphi).
\end{aligned}
$$

Remark 6.

- The term $\eta_{h.o.t.}$ is of higher order. By approximation results it can be estimated in terms of h_T to a higher order then the orders included in $\eta_T^{(i)}$, $\eta_E^{(i)}$, $i = 1,2,3$. Thus it is neglected in the numerics.
- The term η_C arises due to the transfer of φ^{k-1} from the old grid $\mathscr{T}^{k-1}$ to the new grid $\mathscr{T}^k$ through the projection $\mathscr{P}^k$. In our numerics we use Lagrangian interpolation $\mathscr{I}^k$ as projection operator. We note that $\mathscr{I}^k\varphi^{k-1}$ and φ^{k-1} do only differ in regions of the domain where coarsening in the last time step took place, if bisection is used as refinement strategy. Since it seems unlikely that elements being coarsened in the last time step are refined again in the present time step, this term is neglected in the numerics. We note that this term might be further estimated to obtain powers of h_T by approximation results for the Lagrange interpolation, see e.g. [29].
- Due to these two terms involved the estimator is not fully reliable.
- Neglecting these two terms the estimator can be shown to be efficient by the standard bubble technique, see e.g. [7, 61].
- An adaptation of the time step size is not considered in the present work, since it would conflict with the time discretization over three time instances. In our numerics we have to choose time steps small enough to sufficiently well resolve the interfacial force $\mu_h^k \nabla \varphi^{k-1}$.

In the numerical part, this error estimator is used together with the mesh adaptation cycle described in [61]. The overall adaptation cycle

$$\text{SOLVE} \rightarrow \text{ESTIMATE} \rightarrow \text{MARK} \rightarrow \text{ADAPT}$$

is performed once per time step. For convenience of the reader we state the marking strategy here.

Algorithm 1 (Marking strategy)

- *Fix* $a_{\min} > 0$ *and* $a_{\max} > 0$*, and set* $\mathscr{A} = \{T \in \mathscr{T}^{k+1} \,|\, a_{\min} \leq |T| \leq a_{\max}\}$.
- *Define indicators:*
  1. $\eta_T = \frac{1}{\tau\underline{\eta}} \left(\eta_T^{(1)}\right)^2 + \frac{1}{\tau\underline{m}} \left(\eta_T^{(2)}\right)^2 + \frac{1}{\sigma\varepsilon} \left(\eta_T^{(3)}\right)^2$,
  2. $\eta_{TE} = \sum_{E \subset T} \left[\frac{\tau}{\underline{\eta}} \left(\eta_{TE}^{(1)}\right)^2 + \frac{\tau}{\underline{m}} \left(\eta_{TE}^{(2)}\right)^2 + \sigma\varepsilon \left(\eta_{TE}^{(3)}\right)^2\right]$.
- *Refinement: Choose* $\theta^r \in (0,1)$*,*
  1. *Find a set* $R^T \subset \mathscr{T}^{k+1}$ *with* $\theta^r \sum_{T \in \mathscr{T}^{k+1}} \eta_T \leq \sum_{T \in R^T} \eta_T$,
  2. *Find a set* $R^{TE} \subset \mathscr{T}^{k+1}$ *with* $\theta^r \sum_{T \in \mathscr{T}^{k+1}} \eta_{TE} \leq \sum_{T \in R^{TE}} \eta_{TE}$.
- *Coarsening: Choose* $\theta^c \in (0,1)$*,*
  1. *Find the set* $C^T \subset \mathscr{T}^{k+1}$ *with* $\eta_T \leq \frac{\theta^c}{N} \sum_{T \in \mathscr{T}^{k+1}} \eta_T \, \forall T \in C^T$,
  2. *Find the set* $C^{TE} \subset \mathscr{T}^{k+1}$ *with* $\eta_{TE} \leq \frac{\theta^c}{N} \sum_{T \in \mathscr{T}^{k+1}} \eta_{TE} \, \forall T \in C^{TE}$.
- *Mark all triangles of* $\mathscr{A} \cap (R^T \cup R^{TE})$ *for refining.*
- *Mark all triangles of* $\mathscr{A} \cap (C^T \cup C^{TE})$ *for coarsening.*

Ensuring the validity of the energy estimate

To ensure the validity of the energy estimate during the numerical computations we ensure that the total energy does not increase triangle-wise. For the following considerations we restrict to bisection as refinement strategy combined with a hierarchically organised mesh, such that coarsening inverses prior refinement steps, see e.g. [102]. Note that in such a situation local coarsening, e.g. substituting four triangles by two triangles, only appears if all four triangles are marked for coarsening. We call such a set of four (resp. two cells at $\partial\Omega$), triangles a nodeStar, following [33].

By using this strategy, we do not harm the Assumption 2 on triangles that are refined. We note that this assumption can only be violated on patches of triangles where coarsening appears.

After marking triangles for refinement and coarsening and before applying refinement and coarsening to $\mathscr{T}^k$ we make a post-processing of all triangles that are marked for coarsening.

Let M^C denote the set of triangles marked for coarsening obtained by the marking strategy described in Algorithm 1. To ensure the validity of the energy estimate (20) we perform the following post processing steps:

Algorithm 2 (Post processing)

1. *For each triangle $T \in M^C$:*
   if T is not part of a nodeStar
   then set $M^C := M^C \setminus T$.
2. *For each nodeStar $S \in M^C$:*
   if Assumption 2 is not fulfilled on S
   then set $M^C := M^C \setminus S$.

The resulting set M^C only contains triangles yielding nodeStars on which Assumption 2 is fulfilled.

2.2.1 A numerical example

The proposed scheme is implemented in C++ using a mesh based on [33] and using the package [36]. We note that the linear systems that are solved during Newton's method have a saddle point structure, where the diagonal blocks again have saddle point structure. For saddle point problems efficient preconditioning techniques are available, we refer to [20, 83] for details and [45] for the actual reailzation in the present setting. In the following we use $\mathscr{P}^k \equiv \mathscr{I}^k$, where $\mathscr{I}^k$ denotes the Lagrangian interpolation, associated with the space V_1^k. This leads to a small deviation in the total mass, see [45].

To test the validity of the energy inequality in the fully discrete setting, we use the classic example of spinodal decomposition [31] as test case. The parameters are chosen as: $\rho_1 = \rho_2 = \eta_1 = \eta_2 = 1$, $g \equiv 0$, and $m \equiv 10^{-3}\varepsilon$, $\varepsilon = 0.01$, $\sigma = 0.01$ and $\tau = 10^{-5}$.

In Figure 1 we show the time evolution of the monotonically decreasing Ginzburg–Landau energy (left plot). We obtain the expected rate of $E \sim t^{-1/3}$ and also observe a time span where $E \sim t^{-1}$ holds, as predicted in [97].

Next we investigate the validity of the energy inequality; see Figure 1 (right plot). The plot depicts the time evolution of the term

$$\begin{aligned}\zeta =& \frac{1}{2}\int_\Omega \rho^{k-1}\left|v_h^k\right|^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\varphi_h^k|^2 dx + \frac{\sigma}{\varepsilon}\int_\Omega W(\varphi_h^k)\,dx \\ &+ \frac{1}{2}\int_\Omega \rho^{k-2}(v_h^{k-1}-v^k)^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\varphi_h^k - \nabla\mathscr{I}^k\varphi^{k-1}|^2 dx \\ &+ \tau\int_\Omega 2\eta^{k-1}|Dv_h^k|^2 dx + \tau\int_\Omega m^{k-1}|\nabla\mu_h^k|^2 dx \\ &- \left(\frac{1}{2}\int_\Omega \rho^{k-2}\left|v^{k-1}\right|^2 dx + \frac{\sigma\varepsilon}{2}\int_\Omega |\nabla\mathscr{I}^k\varphi^{k-1}|^2 dx \right. \\ &\left. + \frac{\sigma}{\varepsilon}\int_\Omega W(\mathscr{I}^k\varphi^{k-1})\,dx + \int_\Omega \rho^{k-1} g v_h^k\right).\end{aligned}$$

The post processing of Algorithm 2 guarantees that this term is always negative as we observe in the plot.

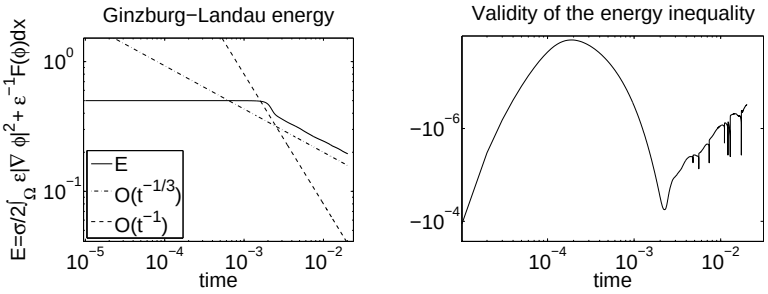

Fig. 1 Time evolution of the Ginzburg–Landau energy (left), and validity of the energy inequality (right).

We also simulated the benchmark for rising bubble dynamics from [79]: Here we observed results that are closer to sharp interface numeric when we use the relaxed double obstacle free energy W^{rel} then when using the polynomially free energy W^{poly}, see Remark 2, which clearly shows the benefits of using the relaxed double-obstacle free energy.

3 Optimal control of variable density two-phase flow

In this section, we focus on a double-obstacle potential type free energy density which yields an optimal control problem for a family of coupled systems in each time instant of a variational inequality of fourth order and the Navier–Stokes equation. By proposing a suitable time discretization, we establish the existence of solutions to the primal system and of optimal controls for the original problem as well as for a family of regularized problems which is introduced to handle the constraint degeneracy. The latter correspond to Moreau–Yosida type approximations of the double-obstacle potential. We further show the consistency of these approximations and derive first-order optimality conditions for the regularized problems. Through a limit process with respect to the regularization parameter, we obtain a stationarity system for the original problem which corresponds to a function space version of C-stationarity. The following results have been derived in [67].

3.1 The semi-discrete CHNS-system and the optimal control problem

The presence of a non smooth free energy density gives rise to a degenerate constraint system with the overall problem falling into the realm of mathematical programs with equilibrium constraints (MPECs). This evokes a variety of new challenges when it comes to the analytical treatment of the problem, cf. Section 1.1.3, and demands for a slightly different set of assumptions and definitions as used in previous section which is provided in the following.

First note that, assuming integrability in time, from (2), (3), (7), and (8), it follows that

$$\int_\Omega \partial_t \varphi dx = -\int_\Omega v\nabla\varphi dx + \int_\Omega \operatorname{div}(m(\varphi)\nabla\mu)dx = 0.$$

Hence, utilizing (6) the integral mean of φ satisfies

$$\frac{1}{|\Omega|}\int_\Omega \varphi dx \equiv \frac{1}{|\Omega|}\int_\Omega \varphi_a dx =: \overline{\varphi_a},$$

i.e., it is constant in time. By assuming $\overline{\varphi_a} \in (-1,1)$, we exclude the uninteresting case $|\overline{\varphi_a}| = 1$. This can be achieved by considering the shifted system (1) – (8), where φ is replaced by its projection onto $\overline{L}^2(\Omega)$. Consequently, we need to work with shifted variables such as, e.g. $m(y+\overline{\varphi_a})$, which we again denote by $m(y)$ in a slight misuse of notation.

As in the previous section, we assume throughout that the mobility and viscosity coefficients are strictly positive as specified in Assumption 3 below. Furthermore, we extend the connection between φ and ρ to all of $\mathbb{R}$, as our studies include cer-

tain double-well type potentials which allow for values of φ outside the physically relevant interval $[-1,1]$.

Assumption 3 *1. The coefficient functions $m,\eta \in C^2(\mathbb{R})$ in (1) and (3) as well as their derivatives up to second order are bounded, i.e. there exist constants $0 < b_1 \leq b_2$ such that for every $x \in \mathbb{R}$, it holds that $b_1 \leq \min\{m(x),\eta(x)\}$ and*

$$\max\{m(x),\eta(x),|m'(x)|,|\eta'(x)|,|m''(x)|,|\eta''(x)|\} \leq b_2.$$

2. The initial state satisfies $(v_a,\varphi_a) \in H^2_{0,\sigma}(\Omega;\mathbb{R}^n) \times \left(\overline{H}^2_{\partial_n}(\Omega) \cap \mathbb{K}\right)$ where

$$\mathbb{K} := \left\{ v \in \overline{H}^1(\Omega) : \psi_1 \leq v \leq \psi_2 \text{ a.e. in } \Omega \right\},$$

with $-1-\overline{\varphi_a} =: \psi_1 < 0 < \psi_2 := 1-\overline{\varphi_a}$.

3. The density ρ depends on the order parameter φ via

$$\rho(\varphi) = \max\left\{ \frac{\rho_1+\rho_2}{2} + \frac{\rho_2-\rho_1}{2}(\varphi+\overline{\varphi_a}), 0 \right\} \geq 0.$$

We note that the pure phases are attained at x when $\varphi(x) = \psi_1$ or $\varphi(x) = \psi_2$. The *max*-operator in Assumption 3.3 ensures that the density remains always non-negative and maintains the affine connection of ρ and φ if φ is contained in the interval $[\psi_1,\psi_2]$. This is necessary to derive appropriate energy estimates.

With these assumptions we now state the semi-discrete Cahn–Hilliard Navier–Stokes system. For the sake of generality, we additionally introduce a distributed force on the right-hand side of the Navier-Stokes equation, which will later serve the purpose of a distributed control. As before, $\tau > 0$ denotes the time step-size and $K \in \mathbb{N}$ the total number of time instants in the semi-discrete setting. We further set $\sigma := \frac{1}{\varepsilon}$ and $\kappa := \frac{\tilde{\kappa}}{\varepsilon^2}$ in order to keep the notation as short as possible.

Definition 2 (Semi-discrete CHNS-system). Let $\Psi_0 : \overline{H}^1(\Omega) \to \mathbb{R}$ be a convex functional with subdifferential $\partial\Psi_0$. Fixing $(\varphi_{-1},v_0) = (\varphi_a,v_a)$ we say that a triple

$$(\varphi,\mu,v) = ((\varphi_i)_{i=0}^{K-1},(\mu_i)_{i=0}^{K-1},(v_i)_{i=1}^{K-1})$$

in $\overline{H}^2_{\partial_n}(\Omega)^K \times \overline{H}^2_{\partial_n}(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1}$ solves the semi-discrete CHNS system with respect to a given control $u = (u_i)_{i=1}^{K-1} \in L^2(\Omega;\mathbb{R}^n)^{K-1}$, denoted as $(\varphi,\mu,v) \in S_\Psi(u)$, if it holds for all $\phi \in \overline{H}^1(\Omega)$ and $\psi \in H^1_{0,\sigma}(\Omega;\mathbb{R}^n)$ that

$$\left\langle \frac{\varphi_{i+1}-\varphi_i}{\tau}, \phi \right\rangle + \langle v_{i+1}\nabla\varphi_i, \phi\rangle + (m(\varphi_i)\nabla\mu_{i+1}, \nabla\phi) = 0, \tag{29}$$

$$(\nabla\varphi_{i+1}, \nabla\phi) + \langle \partial\Psi_0(\varphi_{i+1}), \phi\rangle - \langle \mu_{i+1}, \phi\rangle - \langle \kappa\varphi_i, \phi\rangle = 0, \tag{30}$$

$$\begin{aligned}\left\langle \frac{\rho(\varphi_i)v_{i+1}-\rho(\varphi_{i-1})v_i}{\tau}, \psi \right\rangle_{H^{-1}_{0,\sigma},H^1_{0,\sigma}} &- (v_{i+1}\otimes\rho(\varphi_{i-1})v_i, \nabla\psi) \\ + \left(v_{i+1}\otimes\frac{\rho_2-\rho_1}{2}m(\varphi_{i-1})\nabla\mu_i, \nabla\psi\right) &+ (2\eta(\varphi_i)\varepsilon(v_{i+1}), \varepsilon(\psi)) \\ &- \langle \mu_{i+1}\nabla\varphi_i, \psi\rangle_{H^{-1}_{0,\sigma},H^1_{0,\sigma}} = \langle u_{i+1}, \psi\rangle_{H^{-1}_{0,\sigma},H^1_{0,\sigma}}.\end{aligned} \tag{31}$$

The first two equations are supposed to hold for every $0 \le i+1 \le K-1$ and the last equation holds for every $1 \le i+1 \le K-1$.

Remark 7. In general, the subdifferential of a convex function Ψ_0 can be a multi-valued, see, e.g., [39]. In this case, by equation (30) there exists $\beta \in \partial\Psi_0(\varphi_{i+1})$ such that

$$(\nabla\varphi_{i+1}, \nabla\phi) + \langle \beta, \phi\rangle - \langle \mu_{i+1}, \phi\rangle - \langle \kappa\varphi_i, \phi\rangle = 0,\ \forall \phi \in \overline{H}^1(\Omega).$$

Remark 8. We note that in the above system the boundary conditions specified in (7) and (8) are incorporated in the respective function spaces. Furthermore, the definition already includes the inherent regularity properties of φ and μ which anticipates the results obtained in Lemma 2 below.

We point out that, as also noted in Remark 3, this semi-discretization of (1) – (8) in time involves three time instants $(i-1, i, i+1)$ and (φ_0, μ_0) is characterized in an initialization step by the (decoupled) Cahn-Hilliard system only. At the subsequent time instants, however, the strong coupling of the Cahn-Hilliard and Navier-Stokes system which, in the case of non-matched densities, is additionally enforced through the presence of the relative flux J is preserved. As a result, well-posedness of the time discrete scheme can be guaranteed and energy estimates mirroring the physical fact of decreasing energies can be argued as seen below.

Finally, we present the optimal control problem for the semi-discrete CHNS system. For its formulation, let $U_{ad} \subset L^2(\Omega;\mathbb{R}^n)^{K-1}$ and $\mathscr{J} : \mathscr{X} \to \mathbb{R}$ be a Fréchet differentiable function, with

$$\mathscr{X} := \overline{H}^1(\Omega)^K \times \overline{H}^1(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times L^2(\Omega;\mathbb{R}^n)^{K-1}.$$

Further requirements on U_{ad} and $\mathscr{J}$ are made explicit in connection with the existence result, Theorem 4.1, below.

Definition 3. The optimal control problem is given by

$$\begin{aligned}&\min\ \mathscr{J}(\varphi,\mu,v,u) \text{ over } (\varphi,\mu,v,u) \in \mathscr{X} \\ &\text{s.t. } u \in U_{ad},\ (\varphi,\mu,v) \in S_\Psi(u).\end{aligned} \tag{P_Ψ}$$

In many applications, $\mathscr{J}$ is given by a tracking-type functional and U_{ad} by unilateral or bilateral box constraints.

3.2 Existence of feasible points

In this subsection, we investigate the existence of feasible points for the optimization problem (P_Ψ). For this purpose, we first study the solvability of the semi-discrete Cahn–Hilliard Navier–Stokes system. Since we will later on approximate the double-obstacle potential by a sequence of smooth potentials of double-well type, we consider here the following two types of free energy densities.

Assumption 4 *The functional $\Psi_0 : \overline{H}^1(\Omega) \to \mathbb{R}$ is convex, proper and lower-semi-continuous. It has one of the two subsequent properties:*

1. *Either it is given by $\Psi_0(\varphi) := \int_\Omega \psi_0(\varphi(x))dx$ where $\psi_0 : \mathbb{R} \to \overline{\mathbb{R}} := \mathbb{R} \cup \{+\infty\}$ represents the double-obstacle potential,*

$$\psi_0(z) := i_{[\psi_1;\psi_2]} := \begin{cases} +\infty \text{ if } z < \psi_1, \\ 0 \quad \text{if } \psi_1 \le z \le \psi_2, \\ +\infty \text{ if } z > \psi_2. \end{cases}$$

2. *Or it satisfies:*
   a. *Ψ_0 is Fréchet differentiable with $\{\Psi_0'(\varphi)\} = \partial\Psi_0(\varphi) \subset L^2(\Omega)$ for every $\varphi \in \overline{H}^1(\Omega)$;*
   b. *There exists $B_u \in \mathbb{R}$ such that $\Psi_0(\varphi) \le B_u$ for every $\varphi \in \mathbb{K}$.*

   Note that these conditions are satisfied for double-well type potential.

Additionally, we assume that the functional $\Psi(\varphi) := \Psi_0(\varphi) - \int_\Omega \frac{\kappa}{2}\varphi(x)^2 dx, \kappa > 0$, is bounded from below by a constant $B_l \in \mathbb{R}$.

As a first observation, we state that the chosen time discretization does not break the thermodynamical consistency of the system. In the subsequent lemma, $(\varphi_i, \varphi_{i-1}, \mu_i, v_i)$ characterizes the state of the system at a given time step i. Then the total energy of the next time step is non-increasing if the external force u_{i+1} is set to zero.

Lemma 1 (Energy estimate for a single time step). *Let $\varphi_i, \varphi_{i-1}, \mu_i \in \overline{H}^1(\Omega)$, $v_i \in H^1_{0,\sigma}(\Omega;\mathbb{R}^n)$, $u_{i+1} \in (H^1_{0,\sigma}(\Omega;\mathbb{R}^n))^*$ be given such that*

$$\rho(\varphi_i), \rho(\varphi_{i-1}) > 0. \tag{32}$$

In case of the double-obstacle potential suppose additionally that $\varphi_i, \varphi_{i-1} \in \mathbb{K}$.

Then, if $(\varphi_{i+1}, \mu_{i+1}, v_{i+1}) \in \overline{H}^1(\Omega) \times \overline{H}^1(\Omega) \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)$ solves the system (29)–(31) for one time step, the following energy estimate holds true:

$$\begin{aligned}
&\int_\Omega \frac{\rho(\varphi_i)\,|v_{i+1}|^2}{2}dx+\int_\Omega \frac{|\nabla\varphi_{i+1}|^2}{2}dx+\Psi(\varphi_{i+1})\\
&\quad+\int_\Omega \rho(\varphi_{i-1})\frac{|v_{i+1}-v_i|^2}{2}dx+\int_\Omega \frac{|\nabla\varphi_{i+1}-\nabla\varphi_i|^2}{2}dx\\
&\quad+\tau\int_\Omega 2\eta(\varphi_i)\,|\varepsilon(v_{i+1})|^2\,dx+\tau\int_\Omega m(\varphi_i)\,|\nabla\mu_{i+1}|^2\,dx+\int_\Omega \kappa\frac{(\varphi_{i+1}-\varphi_i)^2}{2}\\
&\qquad\le \int_\Omega \frac{\rho(\varphi_{i-1})\,|v_i|^2}{2}dx+\int_\Omega \frac{|\nabla\varphi_i|^2}{2}dx+\Psi(\varphi_i)+\langle u_{i+1},v_{i+1}\rangle_{H^{-1}_{0,\sigma},H^1_{0,\sigma}},
\end{aligned}\tag{33}$$

Remark 9. Note that in case of the double-obstacle potential the positivity of the density in (32) is always satisfied , since $\rho(\varphi_i)\ge\rho(\psi_1)>0$. For double-well type potentials, however, the assumption is necessary, since φ may attain arbitrary values in $\mathbb{R}$. Nevertheless, it can be argued that the order parameter attains values in a neighborhood of the interval $[\psi_1,\psi_2]$, if the double-well type potential approximates the double-obstacle potential in a certain sense, cf. Lemma 9.

Besides reflecting an important physical property of the Cahn-Hilliard-Navier-Stokes system, the energy estimate also constitutes a valuable ingredient in the proof of existence of solutions to the system (29)–(31). As it serves to verify the boundedness constraint of Schaefer's fixed point theorem, also called the Leray-Schauder principle, which, in combination with arguments from monotone operator theory, yields the following result concerning the solvability of the semi-discrete system (29)–(31) for single time steps, cf. [67].

Theorem 7 (Existence of solutions to the CHNS system for a single time step). *Let the assumptions of Lemma 1 be satisfied. Then the system* (29)–(31) *admits a solution* $(\varphi_{i+1},\mu_{i+1},v_{i+1})\in\overline{H}^1(\Omega)\times\overline{H}^1(\Omega)\times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)$ *for one time step.*

In our setting, the control force u_{i+1} is contained in $L^2(\Omega;\mathbb{R}^n)$. As a result the corresponding solution possesses higher regularity properties which can be shown via a bootstrap argument and well-known regularity results for the stationary Stokes equation.

Lemma 2 (Regularity of solutions). *Let the assumptions of Lemma 1 be satisfied, and suppose additionally that* $\varphi_i\in H^2(\Omega)$.

Then it holds that $\varphi_{i+1},\mu_{i+1}\in\overline{H}^2_{\partial_n}(\Omega)$ *and* $v_{i+1}\in H^2(\Omega;\mathbb{R}^n)$*, provided that* $(\varphi_{i+1},\mu_{i+1},v_{i+1})\in\overline{H}^1(\Omega)\times\overline{H}^1(\Omega)\times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)$ *satisfies the system* (29)–(31)*. Moreover, there exists a constant* $C=C(N,\Omega,b_1,b_2,\tau,\kappa)>0$ *such that*

$$\begin{aligned}
&\|\varphi_{i+1}\|_{H^2}+\|\mu_{i+1}\|_{H^2}+\|v_{i+1}\|_{H^2}\\
&\qquad\le C(\|\varphi_{i+1}\|+\|\mu_{i+1}\|+\|\varphi_i\|+\|v_{i+1}\|_{H^1}\,\|\varphi_i\|_{H^2}+\|\Psi_0'(\varphi_{i+1})\|).
\end{aligned}\tag{34}$$

In case of the double-obstacle potential, it also holds that $\varphi_{i+1}\in\mathbb{K}$ *and the term* $\|\Psi_0'(\varphi)\|$ *in the above inequality is dropped.*

Repeated applications of these statements for each time step $i = 0, .., K-2$ directly verifies the existence of feasible points in the case of a double-obstacle potential.

Theorem 8 (Existence of feasible points). *Let $u \in L^2(\Omega;\mathbb{R}^n)^{K-1}$. Let $\overline{\Psi}_0$ be the double-obstacle potential defined in Assumption 4.1.*

Then the system (29)-(31) admits a solution $(\varphi,\mu,v) \in \overline{H}^2_{\partial_n}(\Omega)^K \times \overline{H}^2_{\partial_n}(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1}$.

In order to approach the case of the double-well type potentials, we need to ensure the positivity of the density as explained in Remark 9. Using a technique from [70], the subsequent lemma guarantees that the order parameter of a solution to the system (29)–(31) for the double-well type potentials under consideration is always greater than $\psi_1 - \varepsilon$ for some small $\varepsilon > 0$.

Theorem 9. *Let $u \in L^2(\Omega;\mathbb{R}^n)^{K-1}$ be given and $\left\{\Psi_0^{(k)}\right\}_{k\in\mathbb{N}}$ a sequence of functions which satisfies the following two conditions:*

1. *For every $k \in \mathbb{N}$ $\Psi_0^{(k)}$ fulfills Assumption 4.*
2. *If $\left\{\hat{\varphi}^{(k)}\right\}_{k\in\mathbb{N}}$ is a sequence in $\overline{H}^1(\Omega)$ such that there exists a constant $C > 0$ with $\Psi_0^{(k)}\left(\hat{\varphi}^{(k)}\right) \leq C$ for $k \in \mathbb{N}$, then*

$$\left\|\max(-\hat{\varphi}^{(k)} + \psi_1, 0)\right\|_{L^1} \to 0, \text{ as } k \to \infty.$$

Furthermore, let $\left\{(\varphi^{(k)},\mu^{(k)},v^{(k)})\right\}_{k\in\mathbb{N}}$ be a sequence of solutions to the systems (29)–(31) *with $\Psi_0 = \Psi_0^{(k)}$. Then*

$$\left\|\max(-\varphi^{(k)} + \psi_1, 0)\right\|_{L^\infty} \to 0, \text{ as } k \to \infty.$$

Employing the previous theorem, we can verify that the semi-discrete CHNS system (29)-(31) has a solution if the double-well type potential under consideration is close enough to the double-obstacle potential.

Theorem 10 (Existence of feasible points). *Let $u \in L^2(\Omega;\mathbb{R}^n)^{K-1}$. Let $\left\{\Psi_0^{(k)}\right\}_{k\in\mathbb{N}}$ be a sequence which satisfies the conditions of Theorem 9.*

Then there exists $k^* \in \mathbb{N}$ such that the system (29)-(31) admits a solution (φ,μ,v) for every $\Psi_0 \in \left\{\Psi_0^{(k)}\right\}_{k\geq k^*}$.

For more details on the proof of the above theorem, we refer to [67]. In Definition 4 below, we propose a specific regularization which satisfies the conditions of Theorem 9 and Theorem 10, respectively.

3.3 Existence of globally optimal points and convergence of minimizers

In the previous section, the existence of feasible points for the optimal control problem (P_Ψ) is verified. Our next goal is to investigate the existence of an optimal solution to (P_Ψ). We commence by presenting the following lemma which states some important properties of the corresponding control-to-state operator.

Lemma 3 (Regularity and boundedness of the state). *There exists a positive constant $C = C(N,\Omega,b_1,b_2,\tau,\kappa,v_a,\varphi_a,u) > 0$ such that for every solution $(\varphi,\mu,v) \in \overline{H}^2_{\partial_n}(\Omega)^K \times \overline{H}^2_{\partial_n}(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1}$ of Theorem 8 and Theorem 10 it holds that*

$$\|v\|^2_{(H^2)^K} + \|\mu\|^2_{(H^2)^K} + \|\varphi\|^2_{(H^2)^{K+1}} \leq C. \tag{35}$$

Furthermore, the operator $L^2(\Omega,\mathbb{R}^n)^{K-1} \ni u \longmapsto C(N,\Omega,b_1,b_2,\tau,\kappa,v_a,\varphi_a,u) \in \mathbb{R}$ is bounded.

With the help of Lemma 3 it is possible to verify the existence of globally optimal points via standard arguments from optimization theory if some classical assumptions on the objective functional and the constraint set U_{ad} are imposed.

Theorem 11 (Existence of global solutions). *Suppose that the objective functional $\mathscr{J} : \overline{H}^2_{\partial_n}(\Omega)^K \times \overline{H}^2_{\partial_n}(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times L^2(\Omega;\mathbb{R}^n)^{K-1} \to \mathbb{R}$ is convex and weakly lower-semi-continuous and U_{ad} is non-empty, closed and convex. Assume that either U_{ad} is bounded or $\mathscr{J}$ is partially coercive, i.e. for every sequence $\left\{(\varphi^{(k)},\mu^{(k)},v^{(k)},u^{(k)})\right\}_{k\in\mathbb{N}}$ with $\lim_{k\to\infty}\left\|u^{(k)}\right\| = +\infty$ it holds that $\lim_{k\to\infty}\mathscr{J}(\varphi^{(k)},\mu^{(k)},v^{(k)},u^{(k)}) = +\infty$.*

Then the optimization problem (P_Ψ) admits a global solution.

Next, we turn our focus to the consistency of the regularization, i.e. the convergence of a sequence of solutions to $(P_{\Psi^{(k)}})$ with $\Psi^{(k)}$ a double-well potential approaching the double-obstacle potential in the limit as $k \to \infty$, to a solution of (P_Ψ) with Ψ the double-obstacle potential. For this purpose, we consider a sequence of functionals $\left\{\Psi^{(k)}\right\}_{k\in\mathbb{N}}$ satisfying Assumption 4.2 and a corresponding limit functional $\overline{\Psi}$.

The following theorem provides conditions under which a sequence of globally optimal solutions to $(P_{\Psi^{(k)}})$ converge to a global solution of $(P_{\overline{\Psi}})$, as $k \to \infty$.

Theorem 12 (Consistency of the regularization). *Let the assumptions of Theorem 11 be fulfilled. The objective $\mathscr{J} : \overline{H}^1(\Omega)^K \times \overline{H}^1(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times L^2(\Omega;\mathbb{R}^n)^{K-1} \to \mathbb{R}$ is supposed to be upper-semicontinuous, and let $\left\{\Psi^{(k)}\right\}_{k\in\mathbb{N}}$ be a sequence of potentials satisfying Assumption 4.2. Assume further that $\overline{\Psi}$ is given such that for every sequence $\left\{(x^{(k)},y^{(k)})\right\}_{k\in\mathbb{N}} \subset \overline{H}^1(\Omega) \times \overline{H}^{-1}(\Omega)$ with*

$y^{(k)} = \Psi^{(k)\prime}(x^{(k)})$ and $(x^{(k)}, y^{(k)}) \to (x^{(\infty)}, y^{(\infty)})$ strongly in $\overline{H}^1(\Omega) \times \overline{H}^{-1}(\Omega)$ it holds that $y^{(\infty)} \in \partial\overline{\Psi}(x^{(\infty)})$.

Then a sequence $\left\{(\varphi^{(k)}, \mu^{(k)}, v^{(k)}, u^{(k)})\right\}_{k\in\mathbb{N}}$ of global solutions to $(P_{\Psi^{(k)}})$ in $\overline{H}^2(\Omega)^K \times \overline{H}^2(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times U_{ad}$ converges to a global solution of $(P_{\overline{\Psi}})$, provided that $\left\{\mathscr{J}(\varphi^{(k)}, \mu^{(k)}, v^{(k)}, u^{(k)})\right\}_{k\in\mathbb{N}}$ is assumed bounded, whenever U_{ad} is unbounded.

In summary, the optimal control problems under consideration are well-posed and admit globally optimal solutions. Furthermore, the chosen regularization approach is consistent in the sense of Theorem 12.

3.4 Stationarity conditions

At this point, we turn our attention to the derivation of stationarity conditions for the optimal control problem. In this case of smooth potential functions which satisfy Assumption 4.2, stationarity or first-order optimality conditions for the problem (P_Ψ) can be derived by applying classical results from Zowe and Kurcyusz concerning the existence of Lagrange multipliers. The latter approach is employed in the following theorem.

Theorem 13 (First-order optimality conditions for smooth potentials). *Let $\mathscr{J} : \overline{H}^1(\Omega)^K \times \overline{H}^1(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times L^2(\Omega;\mathbb{R}^n)^{K-1} \to \mathbb{R}$ be Fréchet differentiable and let Ψ_0 satisfy Assumption 4.2 such that Ψ_0' maps $\overline{H}^2_{\partial_n}(\Omega)$ continuously Frèchet-differentiably into $L^2(\Omega)$. Further, let $\bar{z} := (\bar{\varphi}, \bar{\mu}, \bar{v}, \bar{u})$ be a minimizer of (P_Ψ).*

Then there exist $(p,r,q) \in \overline{H}^1(\Omega)^K \times \overline{H}^1(\Omega)^{K-1} \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1}$, with $p = (p_{-1}, \dots p_{K-2})$, $r = (r_{-1}, \dots r_{K-2})$, $q = (q_0, \dots q_{K-2})$, such that

$$
\begin{aligned}
-\frac{1}{\tau}(p_i - p_{i-1}) + m'(\varphi_i)\nabla\mu_{i+1}\cdot\nabla p_i - \operatorname{div}(p_i v_{i+1}) - \Delta r_{i-1} \\
+\Psi_0''(\varphi_i)^* r_{i-1} - \kappa r_{i+1} - \frac{1}{\tau}\rho'(\varphi_i)v_{i+1}\cdot(q_{i+1}-q_i) \\
-(\rho'(\varphi_i)v_{i+1} - \frac{\rho_2-\rho_1}{2}m'(\varphi_i)\nabla\mu_{i+1})(Dq_{i+1})^\top v_{i+2} \\
+2\eta'(\varphi_i)\varepsilon(v_{i+1}) : Dq_i + \operatorname{div}(\mu_{i+1}q_i) \;=\; \frac{\partial \mathscr{J}}{\partial \varphi_i}(\bar z), \quad (36)
\end{aligned}
$$

$$
\begin{aligned}
-r_{i-1} - \operatorname{div}(m(\varphi_{i-1})\nabla p_{i-1}) - \operatorname{div}(\frac{\rho_2-\rho_1}{2}m(\varphi_{i-1})(Dq_i)^\top v_{i+1}) \\
-q_{i-1}\cdot\nabla\varphi_{i-1} \;=\; \frac{\partial \mathscr{J}}{\partial \mu_i}(\bar z), \quad (37)
\end{aligned}
$$

$$
\begin{aligned}
-\frac{1}{\tau}\rho(\varphi_{j-1})(q_j - q_{j-1}) - \rho(\varphi_{j-1})(Dq_j)^\top v_{j+1} \\
-(Dq_{j-1})(\rho(\varphi_{j-2})v_{j-1} - \frac{\rho_2-\rho_1}{2}m(\varphi_{j-2})\nabla\mu_{j-1}) \\
-\operatorname{div}(2\eta(\varphi_{j-1})\varepsilon(q_{j-1})) + p_{j-1}\nabla\varphi_{j-1} \;=\; \frac{\partial \mathscr{J}}{\partial v_j}(\bar z), \quad (38)
\end{aligned}
$$

$$
\Big(\frac{\partial \mathscr{J}}{\partial u_k}(\bar z) - q_{k-1}\Big)_{k=1}^{K-1} \in \big[\mathbb{R}_+(U_{ad}-\bar u)\big]^+, \quad (39)
$$

for all $i = 0,\dots,K-1$ and $j = 1,\dots,K-1$. Here, $\big[\mathbb{R}_+(U_{ad}-\bar u)\big]^+$ denotes the polar cone of the set $\{r(w-u)|w\in U_{ad}\wedge r\in\mathbb{R}^+\}$. Furthermore, we use the convention that p_i, r_i, q_i are equal to 0 for $i \geq K-1$ along with q_{-1} and φ_i, μ_i, v_i for $i \geq K$.

In [67], it was further shown that the adjoint state (p,r,q) is bounded independently of the regularization parameter. This enables the derivation of a slightly weaker form of stationarity for certain non-smooth potentials via a limiting process which is given in the following theorem.

Theorem 14 (Stationarity conditions). *Suppose that the following assumptions are satisfied.*

1. *$\mathscr{J}'$ is a bounded mapping from $\overline{H}^1(\Omega)^K \times \overline{H}^1(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times U_{ad}$ into the space $(\overline{H}^1(\Omega)^K \times \overline{H}^1(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times L^2(\Omega;\mathbb{R}^n)^{K-1})^*$ and $\frac{\partial \mathscr{J}}{\partial u}$ satisfies the following weak lower-semicontinuity property*

$$
\Big\langle \frac{\partial \mathscr{J}}{\partial u}(\hat z), \hat u \Big\rangle \;\leq\; \liminf_{n\to\infty} \Big\langle \frac{\partial \mathscr{J}}{\partial u}(\hat z^{(n)}), \hat u^{(n)} \Big\rangle,
$$

   for $\hat z^{(n)} = (\hat\varphi^{(n)}, \hat\mu^{(n)}, \hat v^{(n)}, \hat u^{(n)})$ converging weakly in $\overline{H}^2_{\partial_n}(\Omega)^K \times \overline{H}^2_{\partial_n}(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times U_{ad}$ to $\hat z = (\hat\varphi, \hat\mu, \hat v, \hat u)$.
2. *For every $n\in\mathbb{N}$ let $\Psi_0^{(n)} : \overline{H}^2_{\partial_n}(\Omega) \to \overline{\mathbb{R}}$ be a convex, lower-semicontinuous and proper functional satisfying the assumptions of Theorem 13.*

3. Let $(\varphi^{(n)},\mu^{(n)},v^{(n)},u^{(n)}) \in \overline{H}^2_{\partial_n}(\Omega)^K \times \overline{H}^2_{\partial_n}(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times U_{ad}$ *be a minimizer for* $(P_{\Psi^{(n)}})$ *and let further* $(p^{(n)},r^{(n)},q^{(n)}) \in \overline{H}^1(\Omega)^K \times \overline{H}^1(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1}$ *be given as in Theorem 13.*

Then there exists an element (φ,μ,v,u,p,r,q) *and a subsequence denoted by* $\left\{(\varphi^{(m)},\mu^{(m)},v^{(m)},u^{(m)},p^{(m)},r^{(m)},q^{(m)})\right\}_{m\in\mathbb{N}}$ *with*

$$\begin{aligned}
\varphi^{(m)} &\to \varphi \text{ weakly in } \overline{H}^2_{\partial_n}(\Omega)^K,\ \mu^{(m)} \to \mu \text{ weakly in } \overline{H}^2_{\partial_n}(\Omega)^{K-1},\\
v^{(m)} &\to v \text{ weakly in } H^2(\Omega;\mathbb{R}^n)^{K-1},\ u^{(m)} \to u \text{ weakly in } L^2(\Omega;\mathbb{R}^n)^{K-1},\\
p^{(m)} &\to p \text{ weakly in } \overline{H}^1(\Omega)^K,\ r^{(m)} \to r \text{ weakly in } \overline{H}^1(\Omega)^{K-1},\\
q^{(m)} &\to q \text{ weakly in } H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1},\ {\Psi_0^{(m)}}''(\varphi_{i+1}^{(m)})^* r_i^{(n)} \to \lambda_i \text{ weakly in } \overline{H}^1(\Omega)^*,
\end{aligned}$$

for all $i = -1,\dots,K-2$ *such that for* $z = (\varphi,\mu,v,u)$ *and* $\tilde{q}_k := q_{k-1}$ *it holds that*

$$\begin{aligned}
-\frac{1}{\tau}(p_i - p_{i-1}) + m'(\varphi_i)\nabla\mu_{i+1}\cdot p_i - \operatorname{div}(p_i v_{i+1}) - \Delta r_{i-1}&\\
+\lambda_{i-1} - \kappa r_{i+1} - \frac{1}{\tau}\rho'(\varphi_i)v_{i+1}\cdot(q_{i+1}-q_i)&\\
-(\rho'(\varphi_i)v_{i+1} - \frac{\rho_2-\rho_1}{2}m'(\varphi_i)\nabla\mu_{i+1})(Dq_{i+1})^\top v_{i+2}&\\
+2\eta'(\varphi_i)\varepsilon(v_{i+1}) : Dq_i + \operatorname{div}(\mu_{i+1}q_i) &= \frac{\partial \mathscr{J}}{\partial \varphi_i}(z), \quad (40)\\
-r_{i-1} - \operatorname{div}(m(\varphi_{i-1})\nabla p_{i-1}) - \operatorname{div}(\frac{\rho_2-\rho_1}{2}m(\varphi_{i-1})(Dq_i)^\top v_{i+1})&\\
-q_{i-1}\cdot\nabla\varphi_{i-1} &= \frac{\partial \mathscr{J}}{\partial \mu_i}(z), \quad (41)\\
-\frac{1}{\tau}\rho(\varphi_{j-1})(q_j - q_{j-1}) - \rho(\varphi_{j-1})(Dq_j)^\top v_{j+1}&\\
-(Dq_{j-1})(\rho(\varphi_{j-2})v_{j-1} - \frac{\rho_2-\rho_1}{2}m(\varphi_{j-2})\nabla\mu_{j-1})&\\
-\operatorname{div}(2\eta(\varphi_{j-1})\varepsilon(q_{j-1})) + p_{j-1}\nabla\varphi_{j-1} &= \frac{\partial \mathscr{J}}{\partial v_j}(z), \quad (42)\\
\frac{\partial \mathscr{J}}{\partial u}(z) - \tilde{q} &\in \left[\mathbb{R}_+(U_{ad}-u)\right]^+. \quad (43)
\end{aligned}$$

We point out that a tracking-type functional, like, e.g.,

$$\mathscr{J}(\varphi,\mu,v,u) := \sum_{i=0}^{K-1}\frac{1}{2}\left\|\varphi_i - \varphi_d^i\right\|^2 + \frac{\xi}{2}\|u\|^2_{(L^2)^{(K-1)}},\ \xi > 0, \tag{44}$$

with desired states $\varphi_d^i \in L^2(\Omega)$, satisfies the assumptions of Theorem 14.

If the set U_{ad} is bounded, Theorem 14 holds also true for a sequence of stationary points for $(P_{\Psi^{(n)}})$. If it is unbounded, then the result can still be transferred to sequences of stationary points by assuming that the sequence $\left\{u^{(n)}\right\}_{n\in\mathbb{N}}$ is bounded in $L^2(\Omega;\mathbb{R}^n)^{K-1}$.

In order to apply the developed theory, in particular Theorem 14, to the initially stated optimal control problem associated to the double-obstacle potential, we provide the following definition which characterizes the sequence of approximating double-well type potentials. For this purpose, let ψ_0 be defined as in Assumption 4.1 and set $\gamma := \partial\psi_0 \subset \mathbb{R}\times\mathbb{R}$.

Definition 4. Let a mollifier $\zeta \in C^1(\mathbb{R})$ with $\operatorname{supp}\zeta \subset [-1,1]$, $\int_{\mathbb{R}}\zeta = 1$ and $0 \le \zeta \le 1$ a.e. on $\mathbb{R}$, and a function $\theta : \mathbb{R}^+ \to \mathbb{R}^+$, with $\theta(\alpha) > 0$ and $\frac{\theta(\alpha)}{\alpha} \to 0$ as $\alpha \to 0$, be given. For the Yosida approximation γ_α with parameter $\alpha > 0$ of γ define

$$\zeta_\alpha(s) := \tfrac{1}{\alpha}\zeta\left(\tfrac{s}{\alpha}\right), \quad \widetilde{\gamma}_\alpha := \gamma_\alpha * \zeta_{\theta(\alpha)}, \quad \psi_{0\alpha}(s) := \textstyle\int_0^s \widetilde{\gamma}_\alpha(t)\,dt,$$
$$\Psi_{0\alpha}(c) := \textstyle\int_\Omega (\psi_{0\alpha}\circ c)(t)\,dt.$$

Moreover, we set $\alpha_n := n^{-1}$, $\Psi_0^{(n)} := \Psi_{0\alpha_n}$.

Utilizing Theorem 14 with respect to the approximating sequence from Definition 4 yields a stationarity system for the optimal control problem of the semi-discrete CHNS system with the double-obstacle potential. Through a careful limiting analysis the system can be extended by additional complementarity conditions which are presented in the subsequent theorem which can be found in [67].

Theorem 15 (Limiting ε-almost C-stationarity). *Let $\Psi_0^{(n)}$, $n \in \mathbb{N}$ be the functionals of Definition 4, and let the tuples $(\varphi^{(m)},\mu^{(m)},v^{(m)},u^{(m)},p^{(m)},r^{(m)},q^{(m)})$, (φ,μ,v,u,p,r,q) and $\mathscr{J}$ be as in Theorem 14. Moreover, let $\Lambda : \mathbb{R}\to\mathbb{R}$ be a Lipschitz function with $\Lambda(\psi_1) = \Lambda(\psi_2) = 0$. For*

$$a_i^{(m)} := {\Psi_0^{(m)}}'(\varphi_i^{(m)}), \quad \lambda_i^{(m)} := {\Psi_0^{(m)}}''(\varphi_i^{(m)})^* r_{i-1}^{(m)}$$

for $i = 0,\dots,K$, and for a_i denoting the limit of $a_i^{(m)}$, it holds that

$$(a_i, \Lambda(\varphi_i))_{L^2} = 0, \qquad \langle\lambda_i, \Lambda(\varphi_i)\rangle = 0, \tag{45}$$
$$(a_i, r_{i-1})_{L^2} = 0, \qquad \liminf(\lambda_i^{(m)}, r_{i-1}^{(m)})_{L^2} \ge 0. \tag{46}$$

Moreover, for every $\varepsilon > 0$ there exist a measurable subset M_i^ε of $M_i := \{x\in\Omega : \psi_1 < \varphi_i(x) < \psi_2\}$ with $|M_i \setminus M_i^\varepsilon| < \varepsilon$ and

$$\langle\lambda_i, v\rangle = 0 \qquad \forall v \in \overline{H}^1(\Omega),\ v|_{\Omega\setminus M_i^\varepsilon} = 0.$$

In combination with the results from Theorem 14, the last theorem states stationarity conditions corresponding to a function space version of C-stationarity for MPECs, cf. [68, 69]. More precisely, the resulting stationarity system is of limiting

ε-almost C-stationarity type. For the underlying problem class, this is currently the most (and, to the best of our knowledge, only) selective stationarity system available.

4 Goal oriented adaptivity for optimal control of two-phase flow

The specific semi-discretization in time for the coupled CHNS system with non-matched fluid densities of the previous section represents a first step towards a numerical investigation/realization of the problem. Furthermore, the constructive nature of our derivation of the stationarity conditions facilitates the implementation of a solution algorithm for the problem which solves each approximating problem by a Newton method applied to a suitable finite element discretization in space. For this purpose, it is necessary to solve a sequence of large-scale nonlinear optimization problems. As already mentioned in Subsection 2.2 for the primal system, this might cause an immense numerical expense. Hence, we aim to reduce the computational effort by developing a beneficial adaptation process for the underlying space mesh which incorporates the fact that, for optimal control problems, one is usually interested in an accurate estimation of the target quantity, i.e., the objective functional.

For this purpose, we present an adaptive finite elements solver for the optimal control problem of the Cahn–Hilliard Navier–Stokes-system. This includes an adequate error estimator which consists of dual-weighted primal residuals, primal-weighted dual residuals and complementarity errors. It is based on the notion of a modified Lagrangian associated with the MPEC and uses the associated saddle-point condition for optimal points to characterize the error in the objective function between the continuous solution and a fully discretized problem.

The next subsection is devoted to the derivation of the error estimator, whereas Subsection 4.2 deals with the numerical details and showcases some of the obtained results.

4.1 Goal-oriented error estimator

In order to treat the problem numerically and to derive the aforementioned error estimates, it is necessary to establish a fully discretized version of the problem. Hereby, we follow the so called first optimize, then discretize approach in that we directly discretize the optimality conditions given in Section 3.4. The spacial discretization uses Taylor-Hood finite elements which are known to be LBB-stable in case of the Navier-Stokes equation, cf., e.g., [47, 107]. More precisely, the phase field and the chemical potential are discretized via piecewise linear and continuous finite elements, whereas the discretization of the velocity field utilizes piecewise quadratic and continuous finite elements. For more details on the chosen discretization ap-

proach we refer the reader to [58]. Furthermore, we consider the concrete objective functional given in (44).

The subsequent definition characterizes the MPCC-Lagrangian of the optimal control problem (P_Ψ), which is defined on the product function space

$$\begin{aligned}\mathscr{Y} :=& \overline{H}^1(\Omega)^K \times \overline{H}^1(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times \overline{L}^2(\Omega)^K \times L^2(\Omega;\mathbb{R}^n)^{K-1} \times \overline{H}^1(\Omega)^K \\ & \times \overline{H}^1(\Omega)^K \times H^1_{0,\sigma}(\Omega;\mathbb{R}^n)^{K-1} \times \overline{H}^1(\Omega)^K \times \left(\overline{H}^1(\Omega)^*\right)^K \times \left(\overline{H}^1(\Omega)^*\right)^K.\end{aligned}$$

In contrast to the classical Lagrange function, the MPCC-Lagrangian does not include a multiplier for the complementarity condition. It rather corresponds to the Lagrange function of certain tightend nonlinear problems associated to the MPEC, cf. ,e.g., [93, 101].

Definition 5. The MPCC-Lagrangian $L : \mathscr{Y} \to \mathbb{R}$ corresponding to (P_Ψ) (see Defintion 3) is given by

$$\begin{aligned}L(\varphi,\mu,v,a,u,p,r,q,\pi,\lambda^+,\lambda^-) :=& \mathscr{J}(\varphi,\mu,v,u) \\ &+ \sum_{i=-1}^{K-2}\left[\left\langle \frac{\varphi^{i+1}-\varphi^i}{\tau}, p^{i+1}\right\rangle + \left\langle v^{i+1}\nabla\varphi^i, p^{i+1}\right\rangle - \left\langle \operatorname{div}(m(\varphi^i)\nabla\mu^{i+1}), p^{i+1}\right\rangle\right] \\ &+ \sum_{i=-1}^{K-2}\left[\left\langle -\Delta\varphi^{i+1}, r^{i+1}\right\rangle + \left\langle a^{i+1}, r^{i+1}\right\rangle - \left\langle \mu^{i+1}, r^{i+1}\right\rangle - \left\langle \kappa\varphi^i, r^{i+1}\right\rangle\right] \\ &+ \sum_{i=0}^{K-2}\left[\left\langle \frac{\rho(\varphi^i)v^{i+1}-\rho(\varphi^{i+1})v^i}{\tau}, q^{i+1}\right\rangle_{H^{-1},H^1_0} + \left\langle \operatorname{div}(v^{i+1}\otimes\rho(\varphi^{i+1})v^i), q^{i+1}\right\rangle_{H^{-1},H^1_0}\right. \\ &- \left\langle \operatorname{div}(v^{i+1}\otimes\frac{\rho_2-\rho_1}{2}m(\varphi^{i+1})\nabla\mu^i), q^{i+1}\right\rangle_{H^{-1},H^1_0} + (2\eta(\varphi^i)\varepsilon(v^{i+1}),\varepsilon(q^{i+1})) \\ &\left. - \left\langle \mu^{i+1}\nabla\varphi^i, q^{i+1}\right\rangle_{H^{-1},H^1_0} - \left\langle u^{i+1}, q^{i+1}\right\rangle_{H^{-1},H^1_0}\right] \\ &- \sum_{i=0}^{K-1}\left\langle a^i,\pi^i\right\rangle - \sum_{i=0}^{K-1}\left\langle (\lambda^i)^+, \varphi^i-\psi_2\right\rangle - \sum_{i=0}^{K-1}\left\langle (\lambda^i)^-, \varphi^i-\psi_1\right\rangle.\end{aligned}$$

For the sake of readability, we subsequently collect the primal variables in $y := (\varphi,\mu,a,v)$ which describes the state of the optimal control problem and the adjoint variables in $\Phi := (p,r,q)$. Furthermore, $\mathscr{Y}_h$ denotes the discrete equivalent to $\mathscr{Y}$.

Remark 10. Note that if (y,u) is an ε-almost C-stationary point of (P_Ψ) with adjoints $(\Phi,\pi,\lambda^+,\lambda^-)$ then

$$L(y,u,\Phi,\pi,\lambda^+,\lambda^-) = \mathscr{J}(\varphi,\mu,v,u). \tag{47}$$

Based on the MPCC-Lagrangian we provide a first characterization of the difference of the objective values at stationary points of the semi-discrete and the fully discretized problem. Subsequently, the index δ denotes the difference of the discrete and the continuous variables, e.g. $(y_\delta,u_\delta,\Phi_\delta) := (y_h,u_h,\Phi_h) - (y,u,\Phi)$.

Theorem 16. *Let $(y,u,\Phi,\pi,\lambda^+,\lambda^-)$ be a stationary point of the optimal control problem (P_Ψ) and assume that $(y_h,u_h,\Phi_h,\pi_h,\lambda_h^+,\lambda_h^-) \in \mathscr{Y}_h$ satisfy the discretized stationarity system. Then it holds that*

$$\begin{aligned}\mathscr{J}(\varphi_h,\mu_h,v_h,u_h)-\mathscr{J}(\varphi,\mu,v,u) &= \frac{1}{2}\left(\sum_{i=0}^{K-1}\langle a_h^i,\pi^i\rangle-\sum_{i=0}^{K-1}\langle a^i,\pi_h^i\rangle\right)\\ &+\frac{1}{2}\left(\sum_{i=0}^{K-1}\langle (\lambda^i)^+,\varphi_h^i-\psi_2\rangle-\sum_{i=0}^{K-1}\langle (\lambda_h^i)^+,\varphi^i-\psi_2\rangle\right)\\ &+\frac{1}{2}\left(\sum_{i=0}^{K-1}\langle (\lambda^i)^-,\varphi_h^i-\psi_1\rangle-\sum_{i=0}^{K-1}\langle (\lambda_h^i)^-,\varphi^i-\psi_1\rangle\right)\\ &+\frac{1}{2}\nabla_x L(y_h,u_h,\Phi_h,\pi_h,\lambda_h^+,\lambda_h^-)((y_h,u_h,\Phi_h)-(y,u,\Phi)). \end{aligned} \tag{48}$$

The last term on the right-hand side of equation (48) assembles the weighted dual and primal residuals. Whereas the other terms display the mismatch in the complementarity between the discretized solution and the original one.

For each time step $i \in \{0,..,K-1\}$, the latter can be split into the following four parts

$$\begin{aligned}\eta_{CM1,i} &:= \frac{1}{2}\langle a_h^i,\pi^i-\pi_h^i\rangle,\ \eta_{CM2,i} := \frac{1}{2}\langle (\lambda_h^i),\varphi^i-\varphi_h^i\rangle,\\ \eta_{CM3,i} &:= \frac{1}{2}\langle a^i,\pi_h^i-\pi^i\rangle,\ \eta_{CM4,i} := \frac{1}{2}\langle (\lambda^i)^+,\varphi_h^i-\psi_2\rangle+\langle (\lambda^i)^-,\varphi_h^i-\psi_1\rangle.\end{aligned}$$

The so-called dual-weighted primal residual $\eta_{CHNS,i} := \eta_{CH1,i}+\eta_{CH2,i}+\eta_{NS,i}$ consists of the three parts coming from the respective primal equations (for $i=-1,..,K-2$)

$$\begin{aligned}\eta_{CH1,i+1} &:= \left\langle \frac{\varphi_h^{i+1}-\varphi_h^i}{\tau}, p_\delta^{i+1}\right\rangle + \langle v_h^{i+1}\nabla\varphi_h^i, p_\delta^{i+1}\rangle - \langle \mathrm{div}(m(\varphi_h^i)\nabla\mu_h^{i+1}), p_\delta^{i+1}\rangle,\\ \eta_{CH2,i+1} &:= \langle -\Delta\varphi_h^{i+1}, r_\delta^{i+1}\rangle + \langle a_h^{i+1}, r_\delta^{i+1}\rangle - \langle \mu_h^{i+1}, r_\delta^{i+1}\rangle - \langle \kappa\varphi_h^i, r_\delta^{i+1}\rangle,\\ \eta_{NS,i+1} &:= \left\langle \frac{\rho(\varphi_h^i)v_h^{i+1}-\rho(\varphi_h^{i-1})v_h^i}{\tau}, q_\delta^{i+1}\right\rangle_{H^{-1},H_0^1}\\ &\quad + \langle \mathrm{div}(v_h^{i+1}\otimes\rho(\varphi_h^{i-1})v_h^i), q_\delta^{i+1}\rangle_{H^{-1},H_0^1}\\ &\quad - \left\langle \mathrm{div}(v_h^{i+1}\otimes\frac{\rho_2-\rho_1}{2}m(\varphi_h^{i-1})\nabla\mu_h^i), q_\delta^{i+1}\right\rangle_{H^{-1},H_0^1}\\ &\quad + (2\eta(\varphi_h^i)\varepsilon(v_h^{i+1}),\varepsilon(q_\delta^{i+1}))\\ &\quad - \langle \mu_h^{i+1}\nabla\varphi_h^i, q_\delta^{i+1}\rangle_{H^{-1},H_0^1} - \langle u_h^{i+1}, q_\delta^{i+1}\rangle_{H^{-1},H_0^1}.\end{aligned}$$

Finally, the primal-weighted dual residuals can be defined for each $i \in \{0,..,K-1\}$ (with $\eta_{ADv,0} := 0$) in three steps by

$$\begin{aligned}
\eta_{AD\varphi,i} := \Big[& \varphi_h^i - \varphi_d^i - \frac{1}{\tau}(p_h^{i+1} - p_h^i) + m'(\varphi_h^i)\nabla\mu_h^{i+1}\cdot\nabla p_h^{i+1} - \operatorname{div}(p_h^{i+1}v_h^{i+1}) - \Delta r_h^i \\
& + \lambda_h^i - \kappa r_h^{i+1} - \frac{1}{\tau}\rho'(\varphi_h^i)v_h^{i+1}\cdot(q_h^{i+2} - q_h^{i+1}) \\
& - (\rho'(\varphi_h^i)v_h^{i+1} - \frac{\rho_2-\rho_1}{2}m'(\varphi_h^i)\nabla\mu_h^{i+1})(Dq_h^{i+2})^\top v_h^{i+2} \\
& + 2\eta'(\varphi_h^i)\varepsilon(v_h^{i+1}) : Dq_h^{i+1} + \operatorname{div}(\mu_h^{i+1}q_h^{i+1})\Big](\varphi_\delta^i), \\
\eta_{AD\mu,i} := \Big[& - r_h^i - \operatorname{div}(m(\varphi_h^{i-1})\nabla p_h^i) - \operatorname{div}(\frac{\rho_2-\rho_1}{2}m(\varphi_h^{i-1})(Dq_h^{i+1})^\top v_h^{i+1}) \\
& - q_h^i\cdot\nabla\varphi_h^{i-1}\Big](\mu_\delta^i), \\
\eta_{ADv,i} := \Big[& - \frac{1}{\tau}\rho(\varphi_h^{i-1})(q_h^{i+1} - q_h^i) - \rho(\varphi_h^{i-1})(Dq_h^{i+1})^\top v_h^{i+1} \\
& - (Dq_h^i)(\rho(\varphi_h^{i-2})v_h^{i-1} - \frac{\rho_2-\rho_1}{2}m(\varphi_h^{i-2})\nabla\mu_h^{i-1}) \\
& - \operatorname{div}(2\eta(\varphi_h^{i-1})\varepsilon(q_h^i)) + p_h^i\nabla\varphi_h^{i-1}\Big](v_\delta^i).
\end{aligned}$$

By these definitions and Theorem 16, the discretization error with respect to the objective function is then given by

$$\begin{aligned}
& \mathscr{J}(\varphi_h,\mu_h,v_h,u_h) - \mathscr{J}(\varphi,\mu,v,u) \\
& \qquad = \sum_{i=0}^{K-1}(\eta_{CM1,i} + \eta_{CM2,i} + \eta_{CM3,i} + \eta_{CM4,i} + \eta_{CH1,i} \\
& \qquad\qquad + \eta_{CH2,i} + \eta_{NS,i} + \eta_{AD\varphi,i} + \eta_{AD\mu,i} + \eta_{ADv,i}).
\end{aligned} \tag{49}$$

We point out that the integral structure of these error terms allows a patchwise evaluation on the underlying mesh. Apart from the weights φ_δ^i, μ_δ^i and v_δ^i and p_δ^i, q_δ^i, r_δ^i, respectively, the primal-dual-weighted error estimators only contain discrete quantities. In order to obtain a fully a-posteriori error estimator the weights are approximated involving a local higher-order approximation based on the respective discrete variables.

4.2 The numerical realization

For a numerical realization we discretize problem (P_Ψ) in space using a sequence of meshes $(\mathscr{T}^i)_{i=1}^K$ and introduce fully discrete sequences of functions using linear

finite elements for φ, μ, and p and quadratic finite elements for v, yielding fully discretized variables φ_h, μ_h, p_h, and v_h. Note that we introduce the pressure p as a primal variable.

We further introduce the following approximation of Ψ_0

$$\Psi_0^s(\varphi) := \frac{s}{2}\left(\max(0,\varphi-1)^2 + \min(\varphi+1)^2\right),\ s>0.$$

The resulting fully discrete optimization problem is then solved using the steepest descent method for a sequence $s_n \to \infty$, mimicking the approach from Theorem 14. Especially we define the multipliers arising in Theorem 14 using Ψ_0 as given in Theorem 15.

The overall procedure is given in the subsequent Algorithm 1.

Data: Initial data: $\varphi_{-1}, \varphi_0, v_0, N_{\max}$
1 **repeat**
2 **for** $l = 1, \dots$ **do**
3 solve (P_Ψ) using steepest descent method;
4 **if** complementarity conditions (45), (46) are satisfied up to a tolerance tol_c **then**
5 break;
6 **else**
7 increase s_n;
8 **end**
9 **end**
10 calculate the error indicators and find the set $\mathscr{M}_r$ of cells to refine and the set $\mathscr{M}_c$ of cells to coarsen;
11 Adapt $(\mathscr{T}^i)_{i=1}^K$ using $\mathscr{M}_r$ and $\mathscr{M}_c$;
12 **until** $\sum_{i=1}^K |\mathscr{T}^i| < N_{\max}$;

Algorithm 1: The overall solution procedure

Here, the outer loop describes the refinement of the grids $(\mathscr{T}^i)_{i=1}^K$ using the error estimator given in (49). When the for-loop breaks, then we have found an approximate optimal control on the current sequence of grids that solves the system (40)–(46) sufficiently well in the sense that the complementarity conditions (45), (46) are satisfied up to a given tolerance tol_c. Then, in line 10, we evaluate the error indicators $\eta_T^i := \eta_{CM1,i}|_T + \eta_{CM2,i}|_T + \eta_{CM3,i}|_T + \eta_{CM4,i}|_T + \eta_{CH1,i}|_T + \eta_{CH2,i}|_T + \eta_{NS,i}|_T + \eta_{AD\varphi,i}|_T + \eta_{AD\mu,i}|_T + \eta_{ADv,i}|_T$ for all time steps i and for all cells $T \in \mathscr{T}^i$ and choose $\mathscr{M}_r$ as the set with smallest cardinality, such that

$$\sum_{T\in\mathscr{M}_r} \eta_T \geq \theta^r \sum_{i=1}^K \sum_{T\in\mathscr{T}^i} \eta_T$$

with a parameter $0 < \theta^r < 1$ using a greedy marking algorithm. We mark all cells in $\mathscr{M}_r$ for refinement. As in [61] we further choose $\theta^c \in (0,1)$ and define

$$M_c := \left\{ T \in (\mathscr{T}^i)_{i=1}^K \,|\, \eta_T \geq \frac{\theta^c}{N} \sum_{i=1}^K \sum_{T\in\mathscr{T}^i} \eta_T \right\},$$

where $N := \sum_{i=1}^{K} |\mathscr{T}^i|$. Thus, we use the well-known Dörfler marking procedure, where we refine a given proportion of the estimated error. We stress that we do not perform Dörfler marking on each time instance separately, but, as the representation (49) suggests, we perform a marking over all cells in the space-time cylinder. We point out that we have to use a locally refined initial grid in order to get a meaningful initial resolution of the interface. This prevents us from using a very coarse grid initially. As a consequence, we also need to introduce a coarsening strategy, where we mark cells for coarsening, if they contain an error that is smaller than θ^c times the mean error. We repeat this outer adaptation unless a given total amount of cells $N_{\max}$ is reached, summed over all cells, see line 12.

The inner loop, i.e. lines 2–9, solves (P_Ψ) using the steepest descent method from the GNU scientific library [1]. Thereafter we check whether the complementarity conditions are sufficiently well approximated by the current Moreau–Yosida relaxed system. For this we evaluate the terms (45)–(46) for all time instances. If the absolute value of all these terms is smaller then a given tolerance tol_c, we accept the solution and proceed with the adaptation step. If any of these terms has an absolute value larger than tol_c we increase parameter s_n and solve the optimality problem again.

Finally, we shortly illustrate the performance of our algorithm in an example where we aim to prevent a bubble from rising and split it into two bubbles. For more details we refer to [58].

We show the evolution of the total number of cells over the optimization procedure in Figure 2 (left). On the right we show the distribution of the cells over the simulation horizon. These plots clearly show the benefits of using the proposed adaptive concept for the optimization of two-phase fluids.

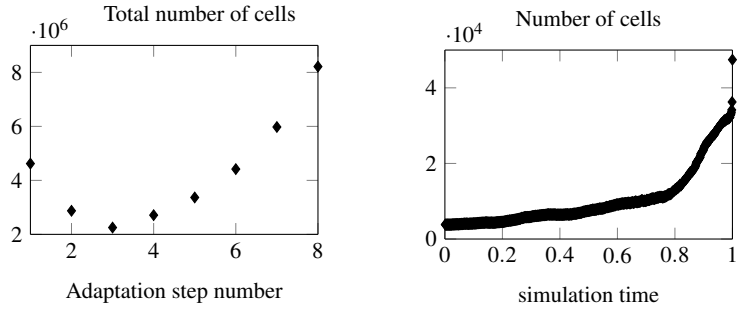

Fig. 2 The evolution of the total number of cells, i.e. $\sum_{i=1}^{K} NC(\mathscr{T}^i)$, where $NC(\mathscr{T}^i)$ denotes the number of cells of the triangulation $\mathscr{T}^i$ over the adaptation steps (left). We note, that we can not start with an arbitrary coarse mesh, as the interface as least has to be roughly resolved at the initialization of the optimization procedure. On the right we depict the distribution of the number of cells over the time horizon. We observe, that the mesh is refined most close to the final time instance, where our optimization aim is located.

5 Further aspects and future research directions

In the present section we briefly discuss further aspects and future research directions in simulation and control of variable density multiphase flows.

Optimal control of multiphase flows based on (15)-(18)

Based on the results in Section 2.1 we have a stable time discretization scheme at hand, for which we next state a time discrete optimal control problem. Here we restrict to control by volume forces that act on the fluid. In [44] additionally Dirichlet boundary control and control using the initial value φ_0, which can be seen as an inverse problem, are also investigated. Let us first state additional assumptions on the data.

Assumption 5 *Additionally to Assumption 1 we assume the following for the data:*

- *A5* *W is twice continuous differentiable and there exist $C > 0$ such that $|W_+''(\varphi)| \leq C(1+|\varphi|^2)$ and $|W_-''(\varphi)| \leq C(1+|\varphi|^2)$.*
- *A6* *For ease of presentation in the following we assume, that ρ and η are affine linear with respect to φ, and that m is constant.*

In the following in (1) we add an additional volume force Bu on the right hand side of the equation and understand this a control action, that can be chosen to influence the two-phase fluid. Let $U = L^2(0,T;\mathbb{R}^S)$ and $f_i \in L^2(\Omega)^n$, $i = 1,\dots,S$ be given volume forces. We define $B : U \to L^2(\Omega)^n$ by

$$(Bu)(t,x) = \sum_{i=1}^{S} u_i(t) f_i(x),$$

i.e. our control acts as amplitudes of given volume forces. Especially u is independent of the actual spatial discretization.

To state the time discrete optimization problem, we define $u^k := \tau^{-1} \int_{t_{k-1}}^{t_k} u(t)\,dt$. This can be regarded as an ansatz using piecewise constant ansatz functions and we stress, that this can be obtained by variational discretization [74] of the first order optimality system stated below. Additionally, for the ease of presentation, we assume, that sufficient initial data is available for the two-step scheme, i.e. φ^{-1}, φ^0, μ^0, and v^0 are given functions, see Remark 3.

Now we can formulate the optimal control problem under consideration.

$$\min_{u \in L^2(0,T,\mathbb{R}^S)} \mathscr{J}(\varphi_h, u) := \frac{1}{2}\|\varphi_h - \varphi_d\|^2_{L^2(\Omega)} + \frac{\xi}{2}\|u\|^2_U \qquad (\mathscr{P}_h)$$

$$\text{s.t.}$$

$$\text{accordingly modified equations } (15) - -(18)$$

Here and in the following we use the abbreviation $\varphi_h := (\varphi_h^k)_{k=1}^K$ and equivalently for the other variables.

Remark 11. Note that we apply a time continuous control $u \in L^2(0,T,\mathbb{R}^S)$ to the fully discrete system (15)– (18). Note that a simulation of two-phase flow has to obey certain CFL conditions, and thus the time step size τ has to b chosen depending on the actual a-priori unknown velocity field v. By not discretizing the control, we can base our numerical approach on a descent method with respect to the control and adjust the time step size τ during the optimization process without changing the actual control space U.

Based on the energy inequality from Theorem 3 one can show the following result by standard techniques.

Theorem 17. *There exists at least one solution to $(\mathscr{P}_h)$, i.e. at least one optimal control.*

By classical KKT-theory one can show the following first order optimality conditions.

Theorem 18. *Let $u^\star, v_h^\star, p_h^\star, \varphi_h^\star, \mu_h^\star$ be an optimal solution to $(\mathscr{P}_h)$. Then there exist adjoint variables $p_{v,h}^\star \in H_0^1(\Omega)^n$, $p_{p,h}^\star \in L^2_{(0)}(\Omega)$, $p_{\varphi,h}^\star \in H^1(\Omega)$, $p_{\mu,h}^\star \in H^1(\Omega)$ such that* (15)–(18) *and the following system is fulfilled for all $k = 1,\dots,K$ and all $w \in H_0^1(\Omega)^n$, $q \in L^2_{(0)}(\Omega)$, $\Psi \in H^1(\Omega)$, $\Phi \in H^1(\Omega)$, $\tilde{u} \in U$.*

$$
\begin{aligned}
-\frac{1}{\tau}\left((\frac{\rho^{k-1}+\rho^{k-2}}{2}w, p_{v,h}^k) - (\rho^{k-1}w, p_v^{k+1})\right) \\
-a(\rho^k w, v^{k+1}, p_v^{k+1}) - a(\rho^{k-1}v_h^{k-1} + J^{k-1}, w, p_{v,h}^k) \\
-(2\eta^{k-1}Dw, Dp_{v,h}^k) - (div(w), p_h^k) - (w\nabla\varphi^{k-1}, p_{\varphi,h}^k) = 0, \\
-(divp_v^k, q) = 0, \\
-a(J_{\mu_h^k}^k\Psi, v_h^{k+1}, p_{v,h}^{k+1}) + (\Psi\nabla\varphi^{k-1}, p_{v,h}^k) - (m\nabla\Psi, \nabla p_{\varphi,h}^k) + (\Psi, p_{\mu,h}^k) = 0, \\
\nabla p_{\varphi,h}^k \cdot \overrightarrow{v}_\Omega = 0, \\
\delta_{kK}(\varphi_h^k - \varphi_d, \Phi) - \frac{1}{\tau}\left(\rho'\frac{v^{k+1}p_v^{k+1} + v^{k+2}p_v^{k+2}}{2}, \Phi\right) + \frac{1}{\tau}\left(\rho' v^{k+1}p_v^{k+2}, \Phi\right) \\
-a(\rho'\Phi v_h^k, v^{k+1}, p_v^{k+1}) - (\eta'\Phi Dv^{k+1}, Dp_v^{k+1}) \\
+(\mu^{k+1}\nabla\Phi, p_v^{k+1}) + (\rho'\Phi g, p_v^{k+1}) \\
-\frac{1}{\tau}\left((\Phi, p_{\varphi,h}^k) - (P^{k+1}\Phi, p_\varphi^{k+1})\right) - (v^{k+1}\nabla\Phi, p_\varphi^{k+1}) \\
-\sigma\varepsilon(\nabla\Phi, \nabla p_{\mu,h}^k) - \frac{\sigma}{\varepsilon}(W_+''(\varphi_h^k)\Phi, p_{\mu,h}^k) - \frac{\sigma}{\varepsilon}(W_-''(P^{k+1}\varphi_h^k)P^{k+1}\Phi, p_\mu^{k+1}) = 0, \\
\xi\tau u^k + B^* p_{v,h}^k = 0 \in \mathbb{R}^S.
\end{aligned}
$$

Here by $J^k_{\mu_h^k}\Psi$ we abbreviate $-\frac{d\rho}{d\varphi}m\nabla\Psi$, i.e. the derivative og J^k with respect to μ_h^k in direction Ψ. From integration by parts we obtain the boundary data

$$\nabla p^k_{\mu,h}\cdot\overrightarrow{\nu}_\Omega = 0.$$

Here $B^\star p_v^k$ is defined as

$$B^\star p^k_{v,h} := ((f_l, p^k_{v,h})_{L^2(\Omega)^n})_{l=1}^S.$$

Here for notational convenience we introduce artificial variables v_h^{K+1}, v_h^{K+2}, $p_{v,h}^{K+1}$, and $p_{v,h}^{K+2}$, and set them as zero.

Remark 12. We note that the prolongation operator P^k enters the adjoint equation acting on the test function Φ.

Concerning a numerical example we refer to [44].

Model predictive control

The optimization problem $(\mathscr{P}_h)$ describes so called open-loop control, which relies on the assumption, that the controlled system is not subject to external disturbances. In many practical applications however, such disturbances are present, which require the design of appropriate regulators. From the mathematical point of view this leads to concepts of closed-loop control. Here we propose the use of so called model predictive control (MPC) [52] and especially of the variant called instantaneous control (IC) [34, 73]. For a further discussion and for the application of IC to two-phase flows we refer to [75] closed-loop control of single-density two-phase flows, and to [76] for the case variable density two-phase flows.

Optimal control with non-smooth free energy density

In Section 3, we established a function space version of C-stationarity for the optimal control problem in the case of non-smooth free energy densities. While this is already a beneficial form of stationarity, since most numerical solvers target this type of stationarity, recent results by [81] for parabolic variational inequalities suggest that it might be possible to obtain even stronger stationarity conditions such as strong stationarity or B-stationarity conditions. Their method does not require the differentiability of the constraint mapping but rather uses the Lipschitz continuity of the control-to-state operator, which can be established for the Cahn-Hilliard-Navier-Stokes system, in order to bound certain difference quotients. This enables the extraction of weakly convergent subsequences whose limit points prove to be

auspicious candidates for the directional derivative of the control-to-state operator. One of the main advantages of strong stationarity is that it also permits the application of novel numerical concepts such as the bundle-free implicit programming technique, cf. [71], which can be used to the design efficient solution algorithms for the optimal control problem.

Model order reduction

It turned out and was expected that the numerical effort for optimal control of two-phase fluids due to the involved Navier–Stokes equation is enormous. Here model order reduction techniques are a promising tool to dramatically decrease the overall cost of the optimization process. We aim at so called proper orthogonal decomposition (POD) [77, 89]. It is well known, that POD is well applicable for the solution of the Navier–Stokes equation. Based on a high resolution simulation, a small subspace of the finite element space is constructed during the optimization process. This drastically reduces the overall number of unknowns, thus speeding up the computations. First numerical experiments indicate that POD is also well applicable for the Cahn–Hilliard equation with smooth free energy. In the case of non-smooth free energy densities, adapted schemes as proposed in [11] are required to obtain the desired reduction of unknowns. Combining these building blocks and adding properly adapted dual weighted residual error estimation will provide a highly efficient solver for optimal control problems of two-phase fluids.

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