

Self-Propulsion via Nontransitive Phase Coexistence in Chemically Active Mixtures

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Chemical activity is central in active matter, where local fuel consumption can lead to self-propulsion and phase separation. However, phase separation can also originate from passive physical interactions. To understand the influence of chemical activity on phase separation, we study mixtures where solvent species interconvert while solutes segregate. We demonstrate that such reactions affect phase separation by altering the chemical potential balance and by introducing an osmotic pressure difference at interfaces. However, the system does not permit a pseudopressure balance, and bulk compositions depend on which phases are in contact. Consequently, phase coexistence is no longer transitive, which enables self-propulsion and more complex dynamics.

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Chemically active mixtures are common in many active matter systems [1–4]. If symmetries are broken, e.g., in anisotropic catalytic colloids, reactions can generate sustained fluxes leading to self-propulsion [5–11]. Such self-propulsion gives rise to rich collective dynamics, including phase separation, even in the absence of attractive interactions [1–4,12]. When interactions already favor phase separation, chemical activity and multiple coexisting phases can further intertwine, which is particularly relevant to condensates in cells [13–24]. While multiphase coexistence is generally understood in passive systems [25–30], the role of active reactions in shaping such coexistence remains largely unexplored.

Multicomponent mixtures with chemical reactions exhibit a wealth of spatial-temporal behaviors [20,21,31–40]. To characterize the basic influences of chemical activity on phase coexistence, we consider “weak” active reactions, which only convert solvent components with similar interaction properties and do not interrupt the phase separation of solutes. In this Letter, we first derive an effective chemical potential balance from two-phase coexistence, and show that the osmotic pressure cannot be balanced due to activity. We next generalize to more phases and reveal that transitivity of phase coexistence is broken, which can lead to self-propulsion.

We describe the state of a general mixture by volume fraction fields ϕ_α satisfying incompressibility $\sum_\alpha \phi_\alpha = 1$,

where α iterates over all components. The free energy of the mixture reads,

$$F = \frac{k_B T}{\nu} \int \left[f + \frac{w^2}{2} \sum_\alpha |\nabla \phi_\alpha|^2 + \xi \left(1 - \sum_\alpha \phi_\alpha \right) \right] d\mathbf{r}, \quad (1)$$

where $k_B T$ is the thermal energy, ξ is a Lagrange multiplier to enforce incompressibility, and ν denotes the molecular volume, which is equal for all components for simplicity. Local interactions are described by the bulk free energy density $f = \sum_\alpha \phi_\alpha \log \phi_\alpha + \frac{1}{2} \sum_{\alpha,\beta} \chi_{\alpha\beta} \phi_\alpha \phi_\beta$, which combines translational entropies and interactions quantified by Flory parameters $\chi_{\alpha\beta}$ with $\chi_{\alpha\alpha} = 0$ [41]. The gradient term in Eq. (1) limits the width of interfaces to roughly w [42].

Without reactions, the system’s behavior is governed by the minimum of F . This implies that the chemical potentials $\mu_\alpha = m_\alpha - w^2 \nabla^2 \phi_\alpha - \xi$ are homogeneous across the system, where $m_\alpha = \partial f / \partial \phi_\alpha$ represents the bulk contribution. Focusing on bulk regions in thermodynamically large systems, coexisting phases must balance all chemical potentials μ_α and the pressure $\sum_\alpha \mu_\alpha \phi_\alpha - f$. Cancelling the Lagrange multiplier ξ , this is equivalent to the balance of exchange chemical potentials with respect to one reference component, $\hat{\mu}_\alpha = \mu_\alpha - \mu_{\text{ref}} = m_\alpha - m_{\text{ref}}$, and the osmotic pressure of one arbitrary solute A, $p = m_A + f - \sum_\alpha \phi_\alpha m_\alpha$ (Supplemental Material [43]).

With active reactions, we describe the dynamics of the system by

$$\frac{\partial \phi_\alpha}{\partial t} = \Lambda_\alpha \nabla^2 \mu_\alpha + R_\alpha, \quad (2)$$

which captures that densities change due to diffusive fluxes driven by gradients in chemical potentials (proportional to mobility constants Λ_α), and reaction fluxes R_α , which

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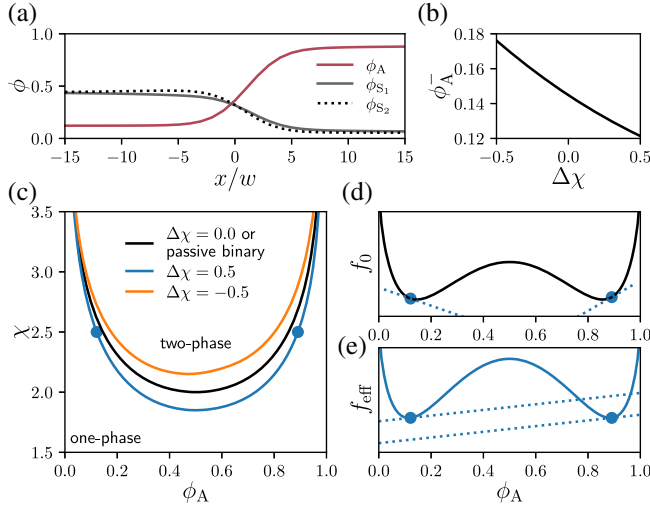


FIG. 1. Active reactions modify phase coexistence. (a) Stationary volume fractions ϕ_i between two coexisting phases satisfying Eq. (2) for $\chi = 2.5$ and $\Delta\chi = 0.5$. (b) Solute fraction in the dilute phase, ϕ_A^- , as a function of interaction difference $\Delta\chi$ between solvents. (c) Phase diagrams as a function of average composition ϕ_A and average interaction χ with solvents for various $\Delta\chi$. (d), (e) Equilibrium (d) and effective (e) bulk free energy as a function of ϕ_A . Dashed lines mark tangent lines corresponding to the blue dots in panel (c). (a)–(d) Parameters are $\Lambda_{S_1}/\Lambda = 0.5$, $\Lambda_{S_2}/\Lambda = 2$, $r_{12} = r_{21} = 0.1\Lambda w^{-2}$.

depend on composition [44]. While Eq. (2) can lead to spatial-temporal patterns [34–37], we here focus on reactions that only weakly modify phase coexistence. In particular, we consider mixtures containing two solvent components S_1 and S_2 that do not phase separate from each other (using $\chi_{S_1 S_2} = 0$ for now), but actively convert into each other ($R_{S_1} = -R_{S_2}$), while all other solute components are conserved ($R_\alpha = 0$ for $\alpha \neq S_1, S_2$). We thus ask how the conversion between the two solvent species can affect phase separation of other species.

We start by considering the simple case of one solute A that segregates from the two reacting solvents S_1 and S_2 to different degrees ($\chi_{AS_1} = \chi - \Delta\chi$ and $\chi_{AS_2} = \chi + \Delta\chi$). If χ is sufficiently large, an A-rich and a solvent-rich phase form, separated by an interfacial region of fixed size. To go beyond thermodynamic equilibrium, we choose the simple reaction flux $R_{S_1} = r_{21}\phi_{S_2} - r_{12}\phi_{S_1}$ with constant reaction rates r_{21} and r_{12} , which implies that these reactions are driven by active processes [18,45]. If $\Delta\chi$ is not too large, this system still permits macroscopically large phases, which are separated by interfacial regions. As an example, Fig. 1(a) shows stationary profiles at equal reaction rates ($r_{21} = r_{12}$) and asymmetric mobility ($\Lambda_{S_2} > \Lambda_{S_1}$). Reaction fluxes vanish away from interfaces ($R_{S_1} = R_{S_2} = 0$), implying a fixed solvent ratio, $\phi_{S_1}/\phi_{S_2} = r_{21}/r_{12}$. Introducing the total solvent fraction $\phi_S = \phi_{S_1} + \phi_{S_2}$, the bulk free energy thus simplifies to $f = \chi_{AS}^{\text{react}} \phi_A \phi_S + \phi_A \log \phi_A + \phi_S \log \phi_S$, where

$$\chi_{AS}^{\text{react}} = \chi + \delta r \Delta\chi, \quad \delta r = \frac{r_{12} - r_{21}}{r_{12} + r_{21}} \quad (3)$$

and we omit a linear term that has no influences on the thermodynamics. The bulk free energy thus recovers the form describing a passive binary mixture, lumping the two reacting solvents into one effective species S. The interaction parameter χ_{AS}^{react} represents a static view, capturing how reactions control the mean interaction of the solvent with the solute.

This initial analysis naively suggests that the chemical reactions merely alter phase coexistence by modifying the bulk free energy. However, while bulk free energies fully determine coexistence in equilibrium systems, this is not necessarily the case in nonequilibrium systems. To illustrate this, we consider symmetric reactions ($r_{12} = r_{21}$), where Eq. (3) implies $\chi_{AS}^{\text{react}} = \chi$. Consequently, χ_{AS}^{react} predicts that the bulk behavior is independent of the interaction difference $\Delta\chi$, but numerical simulations of Eq. (2) show that $\Delta\chi$ still affects the phase coexistence for uneven mobilities ($\Lambda_{S_2} \neq \Lambda_{S_1}$). For example, Fig. 1(b) shows that the composition of the dilute phase decreases with $\Delta\chi$. Similarly, $\Delta\chi$ has an influence on the onset of phase separation, quantified by modified binodal lines [Fig. 1(c)]. Moreover, the coexisting phases no longer correspond to parallel tangent lines of the equilibrium bulk free energy [Fig. 1(d)], which are all consequences of the system’s nonequilibrium nature. These observations suggest that active processes in the interfacial regions might also affect bulk coexistence.

At interfaces, active chemical reactions can generate circular fluxes [46], which can “pump” components across the interface. To include these fluxes, we need to investigate the full dynamics given by Eq. (2). Keeping the idea of an effective solvent described by $\phi_S = \phi_{S_1} + \phi_{S_2}$, we add the dynamic equations of both solvents to find $\partial_t \phi_S = \Lambda_{S_1} \nabla^2 \mu_{S_1} + \Lambda_{S_2} \nabla^2 \mu_{S_2}$. Introducing an effective mobility, $\Lambda_S = \Lambda_{S_1} + \Lambda_{S_2}$, and chemical potential, $\mu_S^{\text{eff}} = (\Lambda_{S_1} \mu_{S_1} + \Lambda_{S_2} \mu_{S_2}) / (\Lambda_{S_1} + \Lambda_{S_2})$, this becomes $\partial_t \phi_S = \Lambda_S \nabla^2 \mu_S^{\text{eff}}$. The solvent thus obeys a generalized diffusion equation governed by a mobility-weighted average of the chemical potentials of both solvents.

So far, we have unveiled two effects of the two solvent components: first, their chemical conversion controls their relative fractions, leading to the effective interaction parameter χ_{AS}^{react} between the solute A and the effective solvent S in the bulk. Second, the diffusive mobilities of S_1 and S_2 inform the dynamic behavior of S, which is particularly relevant at interfaces. To combine both effects, we next calculate the effective exchange chemical potential of the solute A with respect to the lumped solvent, $\hat{\mu}_A^{\text{eff}} = \mu_A - \mu_S^{\text{eff}} = \chi_{AS}^{\text{eff}} (\phi_S - \phi_A) + \log \phi_A - \log \phi_S$, where the constant term is omitted. Apparently, $\hat{\mu}_A^{\text{eff}}$ is equivalent to the exchange chemical potential of a passive binary

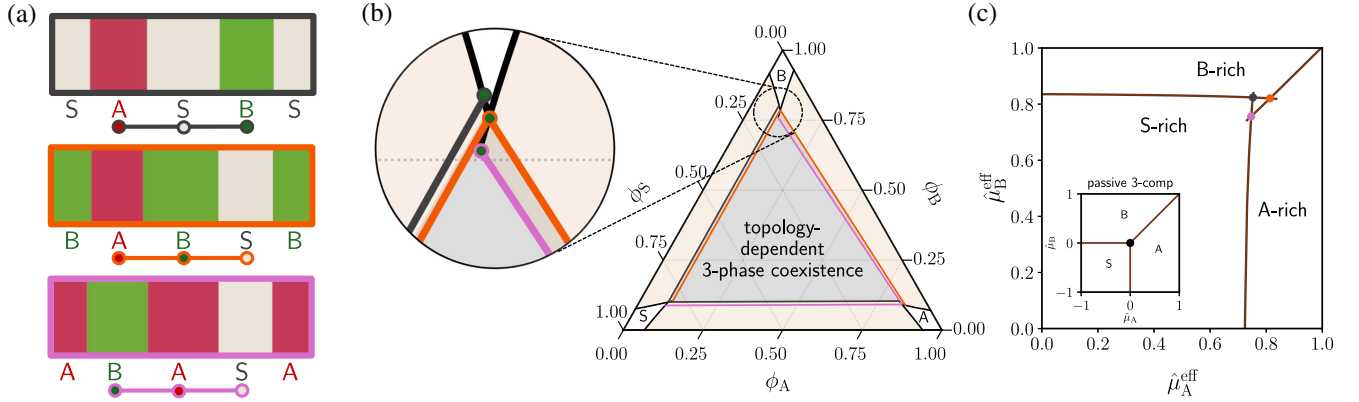


FIG. 2. Phase topology affects coexistence. (a) Three topologies of coexisting A-rich phase (red), B-rich phase (green), and dilute phase (solvent-rich, white). (b) Canonical phase diagrams of the three topologies (colors correspond to outline in panel a), distinguishing one-phase (white), two-phase (light orange), and three-phase (gray) regions. Inset magnifies the upper region around the compositions of B-rich phases. Coexisting compositions were extracted from numerical steady solutions of Eq. (2). (c) Grand canonical phase diagram as a function of the effective exchange chemical potential of the solutes with respect to the lumped solvent, $\hat{\mu}_A^{\text{eff}}$ and $\hat{\mu}_B^{\text{eff}}$, mapped from compositions in (b). Dots mark the chemical potential balance for the three topologies in panel a. Inset shows the grand canonical phase diagram of a passive three-component mixture. (a)–(c) Model parameters are $\chi = 3$, $\Delta\chi = 0.3$, and given in Fig. 1.

mixture of A and S with effective interaction parameter (Supplemental Material [43])

$$\chi_{AS}^{\text{eff}} = \chi + \frac{\delta r + \delta \Lambda}{2} \Delta\chi, \quad \delta \Lambda = \frac{\Lambda_{S_2} - \Lambda_{S_1}}{\Lambda_{S_2} + \Lambda_{S_1}}. \quad (4)$$

Consequently, the interaction between the solute A and the effective solvent S is governed by the average effect of reaction asymmetry δr and the mobility asymmetry $\delta \Lambda$.

The parameter χ_{AS}^{eff} allows us to apply coexistence conditions derived for equilibrium systems to our active system. Since $\hat{\mu}_A^{\text{eff}}$ has the form of an exchange chemical potential of an equilibrium binary mixture, the compositions of coexisting phases can still be determined from a parallel-tangent construction of an effective bulk free energy, $f_{\text{eff}} = \chi_{AS}^{\text{eff}} \phi_A \phi_S + \phi_A \log \phi_A + \phi_S \log \phi_S$. This is confirmed in Fig. 1(e), allowing us to use the effective solvent to predict the phase behavior of the active system. For instance, in the case of $r_{12} = r_{21}$ and $\Lambda_{S_2} > \Lambda_{S_1}$, the effective interaction χ_{AS}^{eff} is larger than the original interaction χ when $\Delta\chi > 0$, explaining the enhancement of phase separation [Fig. 1(c)]. When $r_{12}/r_{21} = \Lambda_{S_2}/\Lambda_{S_1}$, the two contributions coincide ($\delta r = \delta \Lambda$) and the static view given by Eq. (3) gives the correct prediction for chemical potential balance, suggesting a possible mapping to a passive binary mixture. These examples show that the effective solvent with interaction parameter χ_{AS}^{eff} can be used to analyze chemical potential balances between phases. However, the discrepancy between the two tangents in Fig. 1(e) [and the asymmetry of the binodals around $\phi_A = \frac{1}{2}$ in Fig. 1(c)] suggests that there is a pressure imbalance, which cannot be explained with equilibrium concepts.

The pressure imbalance $\Delta p = p(\Phi^{(1)}) - p(\Phi^{(2)})$ observed in Fig. 1(e) can be determined by quantifying the osmotic pressure $p = m_A + f - \sum_{\alpha} \phi_{\alpha} m_{\alpha}$ in the two bulk phases characterized by compositions $\Phi = \{\phi_{\alpha}\}$. In passive systems, Δp can generally be obtained by integrating chemical potentials across the interface [47–49]. In our cases, the active reactions can be recast as a non-local interaction to arrive at a passive surrogate system [21,32,49]. In particular, we introduce the reaction potentials ψ_{α} , which obey $\nabla^2 \psi_{\alpha} = R_{\alpha}$ to define the augmented chemical potentials,

$$\tilde{\mu}_{\alpha} = m_{\alpha} - \kappa \nabla^2 \phi_{\alpha} - \xi + \Lambda_{\alpha}^{-1} \psi_{\alpha}, \quad (5)$$

which allow us to write Eq. (2) as $\partial_t \phi_{\alpha} = \Lambda_{\alpha} \nabla^2 \tilde{\mu}_{\alpha}$, implying that stationary state exhibits constant $\tilde{\mu}_{\alpha}$. Integrating $\tilde{\mu}_{\alpha}$ across the interface, we find

$$\Delta p = - \int_{\Phi^{(1)}}^{\Phi^{(2)}} \sum_{\alpha} \frac{\phi_{\alpha}}{\Lambda_{\alpha}} d\psi_{\alpha}, \quad (6)$$

where we used incompressibility and that reaction fluxes vanish in the bulk [43,49]. For our example of linear chemical reactions, Eq. (6) reduces to

$$\Delta p = \left(\frac{r_{21}}{\Lambda_{S_1}} - \frac{r_{12}}{\Lambda_{S_2}} \right) \int_{-\infty}^{\infty} \phi_S \frac{\nabla \psi_{S_2}}{r_{12} + r_{21}} dx, \quad (7)$$

where $\nabla^2 \psi_{S_2} = r_{12} \phi_{S_1} - r_{21} \phi_{S_2}$ and x is the coordinate across the interface between two thermodynamically large phases (Supplemental Material [43]). The potential ψ_{S_2} captures reaction imbalances and cyclic fluxes of solvent components at interfaces. The integral in Eq. (7) then effectively couples these fluxes to the lumped solvent ϕ_S .

The prefactor in Eq. (7) shows that pressure imbalances vanish if $\Lambda_{S_2}/\Lambda_{S_1} = r_{12}/r_{21}$, consistent with the case where the effective chemical potential $\hat{\mu}_A^{\text{eff}}$ coincides with the one obtained from the static perspective. In this case, the last term in Eq. (5) becomes effectively reciprocal electrostatic interactions. In all other cases, the system cannot be mapped to equilibrium, suggesting interesting behavior.

Equation (7) shows that pressure is generally not a state function in our system. In particular, details of the concentration profiles matter at the interface [38,50,51], suggesting that the behavior of the system might depend on what phases are in contact with each other. To study such topology-dependent multiphase coexistence, we extend the ternary mixtures discussed so far by an additional solute component B. For simplicity, we set the additional interaction parameters to $\chi_{AB} = \chi$, $\chi_{BS_1} = \chi + \Delta\chi$ and $\chi_{BS_2} = \chi - \Delta\chi$ to create an A-rich, a B-rich, and a solvent-rich phase. Note that solute B repels S_1 more strongly than S_2 , in contrast to solute A, which had opposite interactions with the solvents. Chemical conversions still only take place between S_1 and S_2 , so the dynamics of B are described by Eq. (2) with $R_B = 0$. We thus end up with a system that generally forms three phases, separated by chemically active interfaces. The different possible topologies can be described by graphs, where nodes represent unique phases, which are joined by edges if phases are in contact. For three coexisting phases there are thus four possibilities: either all phases touch each other (discussed later), or one pair of phases is not in contact, leading to three distinct noncyclic topologies [Fig. 2(a)].

For the three noncyclic topologies, we numerically find steady states of Eq. (2), and extract the compositions of the coexisting phases to construct canonical phase diagrams at fixed χ [Fig. 2(b)]. Surprisingly, we find that these compositions depend on the topology, which is highlighted for the B-rich phase in the inset of Fig. 2(b). The topology-dependent coexistence is also visible in the grand-canonical phase diagram [Fig. 2(c)], which we determine by converting compositions to chemical potentials $\hat{\mu}_A^{\text{eff}}$ and $\hat{\mu}_B^{\text{eff}}$ to capture those aspects of solvent dynamics that cannot be absorbed into an equilibrium picture (Supplemental Material [43]). The grand-canonical phase diagram shows balances between pairs of phases, but the corresponding coexistence curves (brown curves) do not meet at a single point, which is required for passive systems (inset). Instead, there are three intersections, corresponding to the three topologies shown in Fig. 2(a). Our analysis generally suggests that three phases can be balanced if there are only two types of interfaces, but transitivity is violated, suggesting that the case with all three phases touching is more complicated.

In the case where all three phases are in contact, the system has a cyclic adjacency topology, containing three types of interfaces [Fig. 3(a)]. For this system to obey a

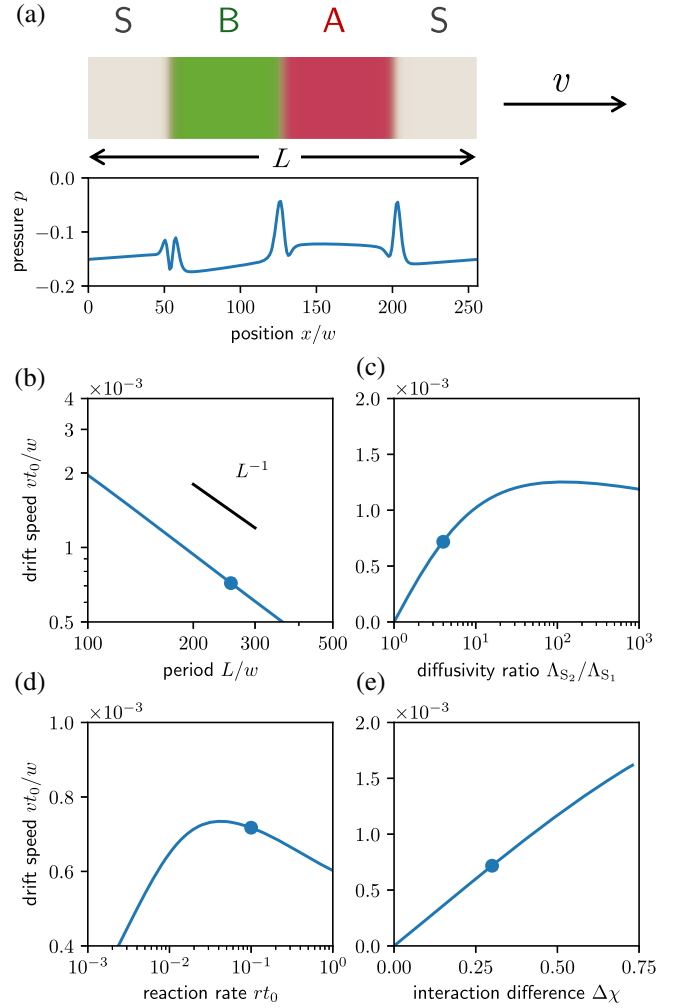


FIG. 3. Cyclic topology causes drift. (a) Visualization of volume fraction profiles that lead to drift, and corresponding pressure profile. The drift speed v depends on the domain period L (b), the diffusivity ratio $\Lambda_{S_2}/\Lambda_{S_1}$ (c), reaction rate r (d), and the interaction difference $\Delta\chi$ (e). (a)–(e) Other parameters are $L = 256w$, $\Lambda_{S_2}/\Lambda_{S_1} = 4$, $r = 0.1t_0^{-1}$, and $\Delta\chi = 0.3$ [blue dots in (b)–(e)] for $r_{12} = r_{21} = r$, $\Lambda_{S_1}\Lambda_{S_2} = \Lambda^2$, and $t_0 = w^2/\Lambda$.

stationary state, each phase would need to be homogeneous and the pressure differences across all interfaces would need to balance ($\Delta p_{S \rightarrow B} + \Delta p_{B \rightarrow A} + \Delta p_{A \rightarrow S} = 0$) to obey periodicity. However, the pressure differences depend on the detailed reaction rates and mobilities [Eq. (7)] and thus generally do not add to zero, suggesting that cyclic systems do not exhibit stationary states. To test this, we solved Eq. (2) in one dimension, which indeed showed a drift towards one direction [Fig. 3(a)]. Qualitatively, this drift results from the total pressure imbalance, which also implies pressure gradients in each phase [Fig. 3(a)]. These gradients drive diffusive fluxes, leading to an overall drift. Since pressure imbalances are dictated by interfacial details, the pressure gradient in a domain should scale inversely with its length, predicting that the drift speed v also scales inversely

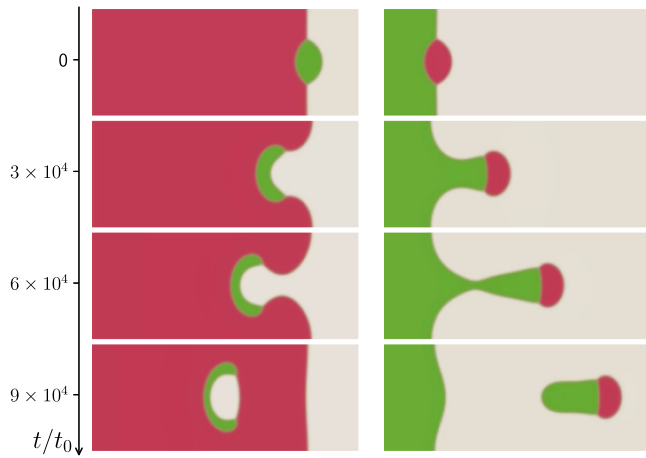


FIG. 4. Cross section of 3D simulations showing the engulfment of a small B-rich droplet (left) and the budding of a B-rich bulk phase (right), starting from equilibrium steady state without reactions. Model parameters are given in Fig. 3.

with the period L , consistent with numerical data [Fig. 3(b)]. Moreover, Eq. (7) predicts that the pressure difference Δp , and thus v , depends on the reaction rates and mobilities. To analyze these dependencies, we next consider the simple case of equal reaction rates ($r_{12} = r_{21} = r$). In this case, Δp and v vanish for $r = 0$ or $\Lambda_{S_1} = \Lambda_{S_2}$, as expected. Figures 3(c) and 3(d) show that v first increases with r and $\Lambda_{S_1}/\Lambda_{S_2}$, but eventually reaches a maximum and decreases for very large values. This nonmonotonous effect results from nonlinear dependencies of the volume fraction profiles on these parameters, which influence ψ_{S_2} in Eq. (7). Similarly, we find that v depends on the interaction difference $\Delta\chi$ [Fig. 3(e)], which also affects the pressure imbalance Δp through the volume fraction profiles. For $\Delta\chi = 0$, the two solvents S_1 and S_2 are equivalent, such that the chemical reaction fluxes vanish everywhere. Taken together, the chemical conversion ($r > 0$) of the asymmetric solvents ($\Lambda_{S_1} \neq \Lambda_{S_2}$ and $\Delta\chi \neq 0$) sets up a pressure difference driving diffusive fluxes, which leads to a self-propulsion of the domains.

So far, we have analyzed 1D systems, but cyclic topology are even more common in higher dimensions. Exemplary 3D simulations show that multiple phenomena such as running multicomponent droplet, engulfment, and budding, can emerge (Fig. 4). For example, when a small B-rich droplet sits on the surface of an A-rich bulk, the interfaces drift in direction of $S \rightarrow B \rightarrow A$, similar to the 1D case. Therefore, the B-rich droplet will gradually intrude the A bulk, along with part of the solvent (left panel of Fig. 4). The same mechanism leads to the budding of a B-rich bulk phase induced by a small A-rich droplet, which finally leaves as a self-propelled two-phase droplet (right panel of Fig. 4). Taken together, complex dynamical patterns emerge from broken transitivity of multiphase coexistence.

We have derived our results using several simplifications, but many of these can be relaxed. For instance, allowing interactions between the two solvents ($\chi_{S_1 S_2} \neq 0$), still allows constructing effective interaction parameters, and compositions of coexisting phases follow from two parallel tangent lines at an effective free energy (Supplemental Material Sec. S5 and Fig. S1 [43]). We can also generalize the mobilities Λ_α to depend on composition. The natural choice $\Lambda_\alpha = D_\alpha \phi_\alpha$ still leads to nontransitive phase coexistence and self-propulsion (Supplemental Material Sec. S6 and Fig. S2 [43]). Note that while the construction of effective chemical potential balance is only exact for constant mobilities, the effective interactions given by Eq. (4) can be viewed as an approximation near the critical point.

In summary, our results show that the effective interaction between components depends on dynamic properties, such as mobilities, when active reactions are present. Surprisingly, the composition of phases does not only depend on the model parameters, but also on which phases are in contact. This topology-dependency implies broken transitivity, defying the construction of a pseudopressure, which has been reported in other models of active matter [48,49,52–56]. Moreover, cyclic topologies then lead to self-propulsion of multiphase droplets. Since isotropy is already broken in those topologies, self-propulsion happens for arbitrarily small activities, in contrast to other chemically active droplets [20]. In principle, this propulsion mechanism could also work for size-controlled droplets [45], which could form mobile emulsions.

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Data availability—Data supporting our findings are openly available [57].

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