ENHANCING HEAT TRANSFER RATES BY INDUCING LIQUID-LIQUID PHASE SEPARATION: APPLICATIONS AND MODELING

Amos Ullmann,1,* Pietro Poesio,2 & Neima Brauner1

1School of Mechanical Engineering, Tel-Aviv University, Tel-Aviv, Israel
2Department of Mechanical and Industrial Engineering, University of Brescia, Italy

*Address all correspondence to Amos Ullmann, E-mail: ullmann@eng.tau.ac.il

This paper focuses on heat transfer enhancement during spinodal decomposition, and it provides an updated review as well as a discussion of future developments. The analysis is mainly based on the work of two research groups at Tel-Aviv University (Israel) and at University of Brescia (Italy). We review the theory of spinodal decomposition of liquid–liquid binary mixtures and we discuss the diffuse interface (DI) approach. While mass and momentum equations in the DI approach have been developed and discussed in other works, we also look into the energy equation, which has been only recently investigated. Direct visualizations of both static and flowing mixture during decomposition are provided. Visualizations of the decomposition in a quiescent fluid have been previously reported, while flowing conditions have been analyzed only recently. Interestingly enough, the morphology is rather different during flowing conditions, where the decomposition exhibits a nucleationlike morphology and not the typical bicontinuous structure observed during spinodal decomposition of a quiescent fluid. Enhancement of heat transfer performances is shown in channels (sizes of 0.8 and 2 mm) using an upper critical solution temperature (UCST) mixture. Although different conditions are analyzed, the results show a consistent enhancement of the heat transfer. The paper reports also some new experimental work on the heat transfer for a lower critical solution temperature (LCST) mixture that can be actually used in cooling applications. A coarse-grained model that could be potentially used for the sizing of large-scale equipment is discussed in term of a possible future development that needs to be further investigated and validated.

KEY WORDS: spinodal decomposition, convective heat transfer, phase separation, enhancement

1. INTRODUCTION

Progress in the miniaturization of electronic chips is hampered by the requirement to remove a very large amount of heat per unit surface and/or per unit volume. To enable efficient cooling, the focus of the heat transfer community has shifted from air cooling to single- and two-phase (boiling) liquid cooling. Using liquid cooling in miniature heat sinks carries several key advantages over their large-scale counterparts—much higher heat transfer coefficients, significantly larger surface area, elimination of thermal interfaces, and a reduced thermal path from the heat sink to the electronic circuit. Very good performance was obtained by using various techniques such as microchannel flows, micropins, sprays, and jet impingement (e.g., Kandlikar et al., 2007; Ebadian and Lin, 2011; Sharma et al., 2012).

One of the emerging technologies for removal of a large amount of heat from a small area is subcooled forced-convection boiling in small (mini and micro) channels (e.g., Ghiaasiaan and Abdel-Khalik, 2001). The high heat transfer rates during boiling are commonly attributed to the pumping mechanism due to the bubble detachment from the heated surface (e.g., Bejan, 1995; Collier and Thome, 1994). Bubble detachment results in inflow of fresh, cold liquid into the thermal boundary layer, thus increasing the rate of heat removal from the surface. Water is the first choice as working fluid: it is cheap, harmless, and available in large amounts. The saturation temperature of water
can be brought to 50°C at a pressure of 0.13 bar. To avoid depressurized systems, other working fluids have been suggested and used.

The application of convective boiling for heat removal is constrained by the critical heat flux (CHF). At the CHF the surface is covered by vapor (dry-out), leading to a very large increase in the surface temperature that results in overheated and damaged equipment. Controlling the system to operate close to, yet below the CHF is one of the main concerns in employing convective boiling cooling, particularly under a variable heat loads. This problem is crucial in microchannels, when the size of the bubble reaches the channel diameter already before its detachment and earlier dry-out occurs. Other problems include nonuniformities in heat transfer and pressure fluctuations when operating in parallel channels due to the vapor compressibility and to uneven flow splitting in the channel array (e.g., Hapke et al., 2000). When considering the combined effects of heat removal, operating pressure losses, operational stability, and device fabrication efforts, single-phase flow appears to be the most promising concept for future development of a range of electronic systems (Kandlikar, 2004, 2012).

To overcome the above limitations of boiling in small-diameter pipes, while preserving the “pumping” effect, the possibility of using phase separation of liquid solutions, instead of evaporating systems, for enhancing heat transfer rates has been investigated independently by our research groups (e.g. Poesio et al., 2007; Gat et al., 2006, 2009, Di Fede et al. 2012, Ullmann et al. 2014a,b).

Temperature-induced phase separation is encountered in partially miscible solvent systems that possess a critical solution temperature (CST). Such systems can be altered from a state of a single liquid phase to a state of two separated liquid phases by a small change of temperature. In solvent systems that are characterized by an upper critical solution temperature (UCST), the transition from a single phase to two phases is brought about by reducing the temperature. In other solvent systems, which are characterized by lower CST (LCST), the phase separation occurs with increasing the temperature. For cooling purposes a heat sink is required, whereby an LCST system should be used. Lists of binary and multicomponent systems possessing a critical temperature and the equilibrium data can be found in Francis (1943) and Sorensen and Arlt (1980). It is worth noting that the use of multicomponent systems (e.g., three components, Gat et al., 2009) enables adjustment of the phase transition temperature to the desired operation range.

The boundary between the complete miscibility of the system and the region where the system separates into two phases is given by the coexistence (binodal) curve. This curve provides the equilibrium compositions of the two separated phases as a function of temperature (see, for example, Fig. 1 for a one-parameter Margules model for regular

![Diagram](Image)

**FIG. 1:** Description of the simulated quenching process and the equilibrium compositions using the one-parameter Margules equation for representing the coexistence and spinodal curves of a UCST system ($\Psi_{w} = 3$).
solution, Sec. 3 below). Within the binodal curve, the spinodal curve defines the boundary between the unstable and metastable regions.

When a partially miscible liquid–liquid mixture is quenched (or heated) from its single-phase region to a temperature below (above) the composition-dependent spinodal curve, it phase separates via the so-called spinodal decomposition, characterized by the spontaneous formation of domains, which then proceed to grow and coalesce. Unlike nucleation, which starts from a metastable state, where activation energy is required to initiate the separation, spinodal decomposition starts from an unstable state and involves the growth of any fluctuation whose wavelength exceeds a critical value. It was observed experimentally that the typical size of the domains forming during spinodal decomposition grows with time according to a power law $t^n$, with an exponent $n$ dependent on the dominant mechanism of phase separation. The scaling exponents are confirmed by theoretical studies [see Bray (1994) for a review] based on modeling the phase transition with one of the variations of the van der Waals square gradient model, known as Cahn-Hilliard theory, model H, the diffuse interface (DI) model, or Ginzburg–Landau phase-field theory. Reviews on spinodal decomposition can be found in the literature (e.g., Gunton et al., 1983; Ullmann et al., 2008).

Most experimental studies show that right after it is quenched below the miscibility curve, the mixture starts to separate by diffusion and coalescence, leading to the formation of well-defined patches whose bulk concentrations approach one of the two mutual equilibrium values. The morphology of these patches depends on the initial composition: for critical mixtures (when the mixture is brought directly to the unstable region), it is dendritic (bicontinuous), while for off-critical mixtures when the mixture is brought to (or paths through) the metastable region, it is of spherical drops. Eventually, these patches become large enough (the size of the capillary length) that buoyancy dominates surface tension and the mixture further separates by gravity (sedimentation). For a liquid mixture with a density difference of about 20%, the capillary length is of O(1 mm). If diffusion was the only driving force (i.e., domain growth process scales as $t^{1/3}$), it would take hours for a nucleus to reach the millimeter size and sediment. Consequently, diffusion and buoyancy alone cannot explain the observed phase separation with patches growing linearly with time and yielding demixing times of O(10 s). The dominant mechanism in this stage is spinodal-decomposition-driven coalescence, whereby the excess free energy associated with the phase change results in a nonequilibrium driving force (also known as Korteweg capillary force), causing fast coalescence and mixing. Korteweg stresses are distributed across the interface and they are null in the fluid bulk [see Eq. (7)] since they are proportional to the concentration gradient. At equilibrium (late stage of phase separation), the integral value of the Korteweg stresses across the interface will lead to force whose normal component is the surface tension, while the tangential component is Marangoni force. In other words, we can interpret the Korteweg stresses as a generalization to nonequilibrium states of the well-known surface tension and Marangoni forces.

Most of the published works on the modeling and numerical simulations of spinodal decomposition assume an instantaneous quench to a final (constant) temperature within the unstable or metastable regions, whereby an isothermal separation process is assumed (e.g., Vladimirova et al., 1999, 2000). The results of numerical simulations show that due to the strong coupling between concentration and velocity fields, spinodal decomposition of fluid mixtures strongly depends on the relative importance of diffusion and convection due to the Korteweg force. An attempt to simulate the heat transfer rates during spinodal decomposition of a critical solution was presented in Molin and Mauri (2007, 2008) and Lamorgese and Mauri (2011). The mass and momentum conservation equations were coupled with an energy conservation equation, represented by the transient heat conduction equation. However, in those works the energy source term due to the phase separation was either neglected or introduced in a simplistic form.

The self-propulsion of separated droplets (or patches) [see Poesio et al. (2009)] and the enhanced coalescence among droplets that result from Korteweg capillary forces during the intermediate, nonequilibrium stages of spinodal phase separation produce self-induced agitation and, therefore, effective fluctuations of the velocity and the scalar fields (temperature and composition). Similarly to turbulent fluctuations that produce enhanced (Reynolds) stresses, heat transport, and species diffusivities, these nonequilibrium fluctuations (induced by mass transfer) should also enhance transport in the form of enhanced (Korteweg) stresses, thermal transport, and species diffusivities.

The potential for employing those effects in heat transfer applications has been recognized and demonstrated by our research groups. In this paper the experimental results obtained with application of UCST solvent systems for enhancing heat transfer rates are summarized and the modeling approaches undertaken are outlined. Those results showed the potential for large improvements of heat transfer rates and encouraged further research with LCST solvent
systems. Such systems can serve as a heat sink that is required in cooling applications. The experimental results obtained very recently with an LCST system are also presented, which further substantiate the feasibility of enhancing single-phase convective cooling rates via liquid–liquid phase separation.

2. VISUALIZATION OF PHASE SEPARATION IN A CLOSED CELL

The morphology during phase separation of a deeply quenched critical mixture has been investigated by several authors—Guenoun et al. (1987), Tanaka (1995), and Wagner (1997)—and it involves three stages:

1. Diffusion stage: As soon as some minute disturbances kick a fluid element off the unstable equilibrium state, the mixture starts separating by diffusion and the domain size $R$ grows as $t^{1/3}$. This stage lasts as long as $\text{Pe}_a = V L / D < 1$, where $V$ is a characteristic velocity $V = \sigma / \eta$ and $L = a$ is the characteristic length (which represents an estimate of the interface width).

2. Convective stage: During this stage the domain growth is convection driven and the domain size grows linearly with time, $R(t) \sim t^1$; during this stage, domains increase their size as a consequence of coalescence driven by Korteweg forces;

3. Gravity stage: During this stage phase segregation is induced by gravity force as a consequence of (possible) density differences between the phases.

Poesio et al. (2006) used direct visualization to investigate the convective stage growth dynamics using a pseudo-binary mixture (62 wt % water, 35 wt % acetonitrile, and 1 wt % toluene) that possesses a UCST of 35°C. The setup, shown in Fig. 2, consists of a thermostatted quartz cell (8 × 45 mm sides, 1 mm thick). The cell is inserted within a square channel and cooled by a water stream. A high-speed camera, in a backlight configuration and equipped with a macro lens, allows direct visualization of phase separation. The field of view is 1.6 × 1.6 mm with a spatial resolution down to 5 µm. Illumination is performed using red light and, to enhance contrast, red violet dye is added to the mixture. The temperatures within the cooling bath and inside the cell are monitored by T-type thermocouples.

FIG. 2: Experimental setup used for phase separation in a static cell. The camera is placed below the cell, and a red light is used to enhance contrast.
In all the experiments, the mixture starts in its phase-separated state at a constant and uniform temperature of 20°C. The solution is first heated to 38°C, then mixed thoroughly, and eventually quenched back below its critical point ($T_{\text{crit}} = 35°C$). Mixing the solution before quenching ensures that the mixture is initially homogeneous.

Figure 3 shows a selection of photograms from a typical sequence obtained for the phase separation of a critical composition mixture. As expected, separation occurs simultaneously over the entire field of view and the mixture exhibits a bicontinuous structure during the entire period.

**FIG. 3:** Snapshots at different instants of time of phase separation of critical composition mixture. Field of view is 1.6 × 1.6 mm, and the optical resolution is down to 5 µm.
We see that single-phase domains grow very rapidly, as a result of the coalescence induced by their motion, showing that the process is dominated by convection. In fact, when domains reach a typical size, \( R \sim 35 \mu m \), we measure speeds in the order of \( V \sim 1 \text{ mm/s} \). Since molecular diffusivity is \( D \approx 5 \times 10^{-10} \text{ m}^2/\text{s} \), the Péclet number is \( \text{Pe}_R = V R / D \sim 70 \). When the gravitational crossover is reached (i.e., \( R \approx R_g \sim 1 \text{ mm} \)), the domain size is so large that gravitational effects start playing a role; the motion is not isotropic anymore and the vertical component becomes dominant.

A quantitative analysis of the separation can be done by image postprocessing. The structure factor of the concentration field is defined as

\[
S(k, t) = \langle \hat{\phi}(k, t) \ast \hat{\phi}(k, t) \rangle
\]

Here \( \hat{\phi}(k, t) \) is the two-dimensional discrete Fourier transform of the concentration field, \( k \) is the wavelength vector, and \( k \) is the wavelength. The structure factor is the Fourier transform of the real-space correlation function (see Dattagupta and Puri, 2004). The structure factor of the concentration field is used since it is the direct output of most experimental techniques (neutron or light scattering) employed in spinodal decomposition experiments. As can be seen in Fig. 4, the data for \( \hat{\phi}(k, t) \) are axisymmetric, confirming that phase separation occurs isotropically and the cell boundaries did not influence phase separation.

The structure factor, plotted in a dimensionless form, is shown for different instants of time in Fig. 5. The typical domain size is defined as the first moment of the structure factor (see Tanaka, 1995):

\[
R^{(1)}(t) = \frac{2\pi}{\sum_{k=0}^{k_{\text{max}}} k S(k, t)}
\]

where \( k_{\text{max}} \) is the upper cutoff. It is evident from Fig. 6 that during the observation time \( R^{(1)}(t) \) grows linearly with time. Additionally, it can be shown that the structure factors, at different instants of time, collapse on the same master curve when properly scaled (see Fig. 5). Visual observation experiments confirm the existence of the Porod tail.

**FIG. 4:** Fourier transform of the concentration field at two different instants of time. It is evident from the picture that the system is isotropic and boundaries do not influence the phase distribution (see Poesio et al., 2006).
FIG. 5: Dimensionless structure factor plotted at four different instants of time as a function of the dimensionless wavelength $k$.

FIG. 6: Linear growth of the domain typical size $R^{(1)}(t)$ for a critical composition mixture.

large $k$ [i.e., at large $k$ the structure factor decreases as $S(k, t) \sim k^{-4}$], as also shown by light-scattering experiments (Guenoun et al., 1987). The presence of very thin interfaces (defects) in phase ordering systems results in power-law or Porod decay of the structure factor tail, $\sim k^{-(d+1)}$ where $d = 3$ is the dimension. The exact result of this behavior can be found in Tomita (1984, 1986).

During the linear, convection-driven stage, the structure factor is single peaked (which suggests the presence of one characteristic dimension), and therefore during this stage double-phase transition is not observed. At later stages, however, we notice the appearance of a secondary nucleation structure (Fig. 7), characterized by droplets of one phase within the other. Unfortunately, it is not possible to quantify the evolution of this secondary structure, since when it appears gravity has already begun to influence the process and the primary structure starts regimenting.

The presence of double-phase separation raises questions regarding the hypothesis of local equilibrium (Tanaka, 1995), as all the coarsening mechanisms that have been proposed to explain the late-stage phase separation are based
on the local equilibrium assumption (i.e., the compositions of both phases coincide with their final equilibrium values). Departure from the local equilibrium assumption may affect their growth rate and even the scaling, and needs to be further investigated. Despite the fact that the scaling relation is not as sensitive to a slight deviation of the concentration from the final equilibrium value (see Tanaka, 1995), the effect of this hypothesis on the heat transfer needs to be investigated and is still an open question.

3. MODELING OF PHASE SEPARATION AND TRANSPORT ENHANCEMENT

As the interface between the separating phases is initially nonexistent and gradually evolves in time, the simulation of the phase separation process can be carried out via the diffuse interface approach, where all properties are considered to vary continuously over the diffuse interface.

Following Cahn and Hilliard (1958, 1959), the phase separation is driven by the system tendency to minimize the generalized free energy \( \tilde{G} \), which is represented by the coarse-grained Landau-Ginzburg functional:

\[
\tilde{G} = \frac{1}{V} \int \left[ g + \frac{1}{2} RTa^2 (\nabla \phi)^2 \right] dV
\]

The generalized free energy (averaged over the volume \( V \)) is composed of the thermodynamic molar Gibbs free energy of the mixture \( g \) and a nonlocal term that penalizes sharp gradients in the concentration \( \phi \) (i.e., molar/mass fraction, for a binary mixture \( \phi = \phi_A, \phi_B = 1 - \phi \)), \( R \) is the universal gas constant, and \( T \) is the temperature. The parameter \( a \) represents a typical length scale of the spatial inhomogeneity in the composition field (i.e., of the order of the interface thickness). As shown by van der Waals (1894), \( a \) is proportional to the surface tension between the two phases at equilibrium and is typically of the order of 0.01 ÷ 0.1 \( \mu \)m. Equilibrium is achieved when \( \tilde{G} \) is minimized subject to the mass conservation constraint, namely,

\[
\bar{\phi} = \frac{1}{V} \int \phi dV = \phi_0
\]

When a system (of a uniform concentration, \( \phi_0 \)) is quenched to the two-phase region (i.e., below the binodal curve, see Fig. 1), phase separation may lower the free energy; however, the associated concentration gradients affect an
increase of the system energy. At equilibrium, when a sharp interface is formed, the nonlocal term represents the system surface energy stored in the interface between the separated phases and is manifested by the surface tension.

The solution of the constrained minimization problem [Eqs. (3) and (4)] is carried out by calculus of variation and yields the generalized mixture molar chemical potential:

\[
\hat{\mu} = \mu - RTa^2 \nabla^2 \varphi; \quad \mu = \mu_A - \mu_B
\]  

(5)

where \( \mu_{A,B} \) are the chemical potentials of species \( A, B \). The generalized mixture molar chemical potential is used to derive a closure for diffusive mass flux (see Ullmann et al., 2008; Segal et al., 2012),

\[
j_\varphi = -\rho D\varphi (1 - \varphi) \left[ \frac{d^2(g/RT)}{d\varphi^2} \nabla \varphi - a^2 \nabla^3 \varphi \right]
\]  

(6)

and for the Korteweg force,

\[
F_\varphi = \frac{\rho}{M} \mu_{NL} \nabla \varphi
\]  

(7)

where \( \mu_{NL} = -RTa^2 \nabla^2 \varphi \) is the nonlocal part of the generalized chemical potential and \( D \) is a molecular diffusion coefficient.

The governing set of equations for a nonisothermal, incompressible mixture of two species (of the same density and molecular weight) includes conservation of mass, momentum, and energy:

\[
\nabla \cdot \nu = 0
\]  

(8)

\[
\frac{D\varphi}{Dt} = \frac{\partial \varphi}{\partial t} + \nu \cdot \nabla \varphi = -\frac{1}{\rho} \nabla \cdot j_\varphi
\]  

(9)

\[
\rho \frac{D\nu}{Dt} = -\nabla p + \eta \nabla^2 \nu + F_\varphi
\]  

(10)

\[
\rho c DT \frac{Dt}{Dt} = -\nabla \cdot q + \sum \bar{H}_i (\nabla \cdot j_i)
\]  

(11)

where \( \nu \) is the mixture velocity, \( \rho \) is the mixture density and, \( p \) is the pressure. In the energy Eq. (11), \( c \) is the specific heat of the mixture, \( q = q^c + q^d \) is the sum of conductive (\( q^c = -k \nabla T \)) and interdiffusion energy fluxes (\( q^d = \sum \bar{H}_i j_i \)) and \( \bar{H}_i, j_i \) are the partial enthalpy and diffusive mass flux of component \( i \), respectively (see Bird et al., 1960).

Using a “regular solution” as a model for the thermodynamic Gibbs free energy of a nonideal two-component system, with the one-parameter Margules model to represent the excess part, the system molar Gibbs energy is given by (Lupis, 1983):

\[
g = g_{id} + g_{ex}; \quad g_{ex} = RT \Psi \varphi (1 - \varphi); \quad g_{id} = g_A \varphi + g_B (1 - \varphi) + RT [(1 - \varphi) \ln(1 - \varphi) + \varphi \ln \varphi]
\]  

(12)

(13)

where \( g_A \) and \( g_B \) are the molar free energies of the pure species \( A \) and \( B \), respectively, at temperature \( T \) and pressure \( p \). \( \Psi = 2Tc_c/T \) and \( R \) is the universal gas constant. With this thermodynamic model, the critical composition is at \( \varphi = 0.5 \) and \( \Psi = 2 \), and the energy equation can be manipulated to the following form (Segal et al., 2012):

\[
\rho c DT \frac{Dt}{Dt} = k \nabla^2 T + 4RTc \nabla \varphi \cdot j_\varphi
\]  

(14)

The last term on the r.h.s. represents the heat source due to heat of solution in a nonideal mixture when using the regular solution model (see Segal et al., 2012, for more details).

The above conservation equations [(8)–(10), (14)] were used to simulate the phase separation process of a nonflowing UCST system (\( \nabla p = 0 \)) of a critical composition, which is confined between two infinite plates. The system temperature is initially above the CST (\( T > T_c, \Psi < 2 \)) and the plates are quenched to a constant temperature.
\( T_w < T_c, \Psi_w = 3 \) (see Fig. 1). Convective terms are retained to account for the Korteweg force-driven velocity. The boundary conditions for the 2D conservation equations are shown in Table 1.

Figures 8 and 9 show examples of the 2D simulation results for two values of the dimensionless specific heat of the mixture. The mixture overall composition is 0.5, which is the initial condition for the concentration in the computational domain. The initial temperature is above the UCST with \( \Psi = 1.9 \). The sidewalls are instantaneously quenched to the final temperature below the UCST, corresponding to \( \Psi_w = 3 \). On the other two boundaries, periodic boundary conditions are maintained. No initial fluctuations were imposed; numerical “perturbations” are sufficient to initiate phase separation in the unstable spinodal region. The dimensionless parameters of the solution includes \( N_{Pe} = \frac{a^2 \rho RT}{D \eta} \), which is the Péclet number, (with \( a = \hat{a} \sqrt{\Psi} \)), which is the ratio between convective and diffusive mass fluxes, \( N_{Le} = \alpha / D \), which is the Lewis number, and \( \hat{c} = c / R \) is the dimensionless heat capacity. Note that the definition of \( N_{Pe} \) is consistent with that of Pe\(_a\) (see Sec. 2) since \( a \propto \sigma M / \rho RT \) (van der Waals, 1894).

As expected, phase separation starts at the cold walls and with time proceeds to the center. The shape of the

<p>| TABLE 1: Boundary conditions for phase-separation simulation (2D model) |</p>
<table>
<thead>
<tr>
<th>Other surfaces</th>
<th>Quenched walls</th>
<th>Variables</th>
</tr>
</thead>
<tbody>
<tr>
<td>Periodic</td>
<td>No flux</td>
<td>Concentration</td>
</tr>
<tr>
<td>Periodic</td>
<td>Constant value, ( T_w )</td>
<td>Temperature</td>
</tr>
<tr>
<td>Periodic</td>
<td>No-slip</td>
<td>Velocity</td>
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</table>

**FIG. 8:** The concentration and velocity fields at times \( \tilde{t} = D / \hat{a}^2 \) = 100 and 150 for \( \Psi_w = 3 \) (at the upper and lower walls), \( N_{Pe} = 100, N_{Le} = 10, \hat{c} = 5 \). The domain is \( 100 \times 100 \) cells, \( \Delta \tilde{x} = 0.75 \) and \( \Delta \tilde{t} = 10^{-2} \) were used.

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FIG. 9: The concentration and velocity fields at times \( \tilde{t} = 100, 150 \) and \( 1000 \) for \((N_{Pe} = 100, N_{Le} = 10, \tilde{c} = 10, \Psi_w = 3, \) see definitions in the caption of Fig. 8). The domain is \( 100 \times 100 \) cells, \( \Delta \tilde{x} = 0.75 \) and \( \Delta \tilde{t} = 10^{-2} \) were used.

domains is similar to that observed experimentally during spinodal decomposition. They grow in time and their boundaries become sharper. Inspection of the velocity vectors (see enlarged regions in the figures) reveals the convection driven by the Korteweg force, which is pronounced across the diffuse interface where the concentration gradients are higher. This convective motion induces mixing and enhances the mass transfer, thereby accelerating the phase transition and thus the heat release. Comparison of Figs. 8 and 9 shows that as expected, increasing the specific heat enhances the separation process (as it attenuates the effect of heat of mixing released during the phase separation).
The propagation of the phase separation is shown in Fig. 10, where the average deviation of the concentrations from the equilibrium values at the (final) wall temperature are shown vs. time. This is obtained by averaging the local deviations of the concentration from the equilibrium value approached (i.e., \( \phi_H = 0.92928, \phi_L = 0.07072 \), corresponding to \( \Psi = 3 \), see Fig. 1), whereby for a local concentration of \( \phi > 0.5, \phi < 0.5 \) the deviation is \((\phi_H - \phi), (\phi - \phi_L)\), respectively.

The heat released (enthalpy of mixing) during the phase separation slows down the cooling of the mixture compared to the case of cooling without phase separation [i.e., 1D transient heat conduction model, Carslaw and Jaeger (1959)]. The convective motion brings the hotter mixture to the vicinity of the cold wall. These effects result in higher temperature gradients at the wall, which leads to augmented heat flux at the wall compared to transient heat conduction without phase separation. The augmentation factor presented in Fig. 11 is the instantaneous ratio of the

**FIG. 10:** Average separation in the computational domain vs. the dimensionless time, \( Pe \equiv N_{Pe}, Le \equiv N_{Le}, c \equiv \tilde{c} \). See definitions of dimensionless numbers in the caption of Fig. 8.

**FIG. 11:** Augmentation of the heat transfer at the cooled wall with phase separation compared to the values obtained without phase separation vs. time. It is the ratio of the instantaneous average value of the heat flux along the cooling surface during phase separation, and that predicted by the 1D transient conduction model for a liquid confined between two infinite plates.
heat flux at the wall obtained with phase separation and that obtained at the same time without phase separation. Note that the former represents the instantaneous averaged value along the cooling surface.

4. A COARSE-GRAINED MODEL

The microscale modeling approach described above (Sec. 3) entails detailed analysis of liquid–liquid interface structures (due the presence of nonlocal terms in the Cahn-Hilliard and Navier-Stokes equation) and, consequently, the mesh size has to be as small as 0.1 µm (or finer), preventing the use of the model for practical applications.

Because of the extremely high computational effort, it is evident that such equations cannot be directly employed to design industrial equipment. An attempt (see Molin et al., 2012) to establish an effective macroscale model has been developed that is based on synthetically generated concentration and velocity fields.

In the synthetic concentration-fluctuation (SCF) approach, instead of solving Eq. (9) to get the concentration field, an artificially fluctuating concentration field is generated so that it respects some statistical properties (such as the shape of the concentration structure factor). The procedure for generating the SCF field is similar to the artificial turbulence method used in Rosales and Meneveau (2006) to study the mixing of a passive scalar in a random turbulent field. Rosales and Meneveau (2006) generate a velocity field and solve the species equation to detect the concentration field of the passive scalar. In our case, the concentration field is imposed to provide a structure factor:

\[
S(k, t) = R^{(1)}(t) f(k R) = A_0(R^{(1)})^n \frac{(CR^{(1)}k)^2}{\left[1 + (CR^{(1)}k)^2\right]^3}
\]

where \( n \) is the number of dimensions \( (n = 3 \text{ for 3D}) \), and the dimensionless self-similarity function features (1) a peak at \( k \sim 1/R^{(1)} \); (2) a \( k^{-4} \) Porod tail as observed in numerical studies (Kendon et al., 1999, 2000); and (3) a \( k^2 \) power law at small \( k \), as prescribed by Kendon et al. (1999, 2000) and Sain and Grant (2005). As discussed in Sec. 1, this behavior has also been observed experimentally.

This approach allows a significant simplification of the problem, since it removes the strongly nonlinear Cahn-Hilliard term in the species concentration equation, Eq. (9), and effectively the Navier-Stokes equation, Eqs. (10) and (9) are decoupled, since the Korteweg body force originates only from the steep interfacial gradients of the fluctuation field (that can be computed from the concentration field). Hence, the Korteweg force is now a known source term in the momentum equation. This approach, therefore, has two main effects: (i) removal of a strongly nonlinear equation (9) and (ii) decoupling of the equations. On the other hand, this procedure does not provide temporal correlation of the concentration fluctuation. We are not aware of any investigation we can use to enforce such a correlation. Our procedure, however, enforces a constant dimensionless structure factor, thus imposing self-similarity over time whereby the concentration field respects some sort of a (weakly) temporal constrain.

The constant \( C = 2 \) [see Eq. (15)] is chosen so that \( 2\pi n \) times the inverse of the first moment of the structure factor provides the average domain size \( R^{(1)} \) [Eq. (2)]. The prefactor \( A_0 \) provides the right amplitude to the concentration fluctuations and, together with the constant rate of growth of \( R^{(1)} \), it must be modeled in terms of the quenching rate and the overall mixture composition, based on relatively abundant data in literature.

In this coarse-grained approach, at every time step a random concentration field is generated in the Fourier space and it is multiplied by \( \sqrt{(R^{(1)})^n f(k R^{(1)})} \) so as to impose the desired statistical behavior [Eq. (15)]. \( \phi(k) \) is transformed into real space to obtain the new concentration field \( \phi(x) \). This procedure substitutes the exact solution of Eq. (9), which is now, as already mentioned, decoupled from Eqs. (10) and (11).

The comparison between the exact solution (by Cahn-Hilliard equation) and the coarse-grained approach is carried out in terms of the structure factors of the velocity and temperature fields, defined as

\[
S_{vv}(k, t) = \langle \hat{v}(k, t) * \hat{v}(-k, t) \rangle \quad S_{TT}(k, t) = \hat{T}(k, t) * \hat{T}(-k, t)
\]

Figure 12(top) shows the structure factor of the velocity field \( S_{vv}(k, t) \), obtained from the full Cahn-Hilliard Navier-Stokes equation (CHNSE) simulation, while Fig. 12(bottom) shows the results of the SCF model. As can be seen,
they compare reasonably well in that at large $k$ they both exhibit a $k^{-4}$ Porod tail, like the well-known feature of the structure factor of the concentration field.

Figure 13(top) shows the structure factor of the temperature field $S_{TT}(k, t)$, obtained from the full CHNSE simulation, and Fig. 13(bottom) shows the result of the SCF model. These also compare reasonably well, in that at large $k$ they both exhibit a $k^{-1}$ tail, as expected in the absence of excess enthalpy contribution in the energy equation.

It is worth mentioning, however, that this coarse-grained approach is still under development, and even if it has shown some potential, more and deeper investigations are needed.

FIG. 12: Comparison between the velocity structure factor $S_{vv}(k, t)$ obtained by the CHNSE (numerical experiments) and by the SCF model (lower lines) at various times.

FIG. 13: Comparison between the temperature structure factor $S_{TT}(k, t)$ obtained by the CHNSE and by the SCF model.
5. VISUALIZATION OF PHASE SEPARATION IN MICROCHANNELS

Keeping in mind that our main interest is the application of phase separation for heat transfer rate enhancement in flowing systems, particularly in small-diameter channels, flow visualization in such systems is of importance. Therefore, in addition to the visualization of the phenomena in a closed cell (see Sec. 2), visualization experiments in a flowing cell have been recently carried out using a binary mixture, acetone and hexadecane at critical composition (Farisé, 2012). In Fig. 14, a sketch of the experimental setup used for the visualization is shown. The channel (800 µm × 700 µm) is made by a square groove in a microscope glass slide with particular attention given to the surface roughness. A complete description of the setup can be found in Farisé et al. (2012).

Cooling by cold-air forced convection is performed to facilitate the visualization, since it solves some practical problems, allowing for a better optical setup (see Fig. 14). A high-speed camera, combined with an inverted microscope, allows direct visualization. All the experiments are conducted in backlight conditions.

The visualization procedure is quite simple and it consists of a flowing mixture within the channel (using a syringe pump in a suction mode). At the beginning the mixture is in single-phase conditions (i.e., above the coexistence curve). At that moment the image acquisition is started. Thereafter, compressed air is turned on and the mixture undergoes a drastic cooling.

In Fig. 15 the quenching process of a critical composition mixture flowing within the channel is shown at different quenching depths. Even from those pictures it can be noticed that phase decomposition occurs quite differently when compared to the decomposition in a closed cell (Sec. 2). With higher magnification (Fig. 16), the phase morphology can be better observed. Even for a critical composition, the morphology is not bicontinuous, but it looks more like a nucleation process. Nuclei form at the walls and then separation continues toward the center of the channel. As can be observed, as the separation penetrates deeper into the channel, the domains at the wall keep on growing.

The domain size grows while the droplets of the dispersed phase flow along the channel. By tracking each droplet individually (like in the particle tracking velocimetry, or PTV technique) we can measure their velocity, which is in the range 0.5–1.5 mm/s, in close agreement with the values observed in the closed-cell configuration.

6. APPLICATION OF PHASE SEPARATION FOR CONVECTIVE HEAT TRANSFER ENHANCEMENT

6.1 Selection of an Appropriate Partially Miscible Liquid System

Even if there are several liquid–liquid mixtures that exhibit spinodal decomposition, the choice of the most suitable one is not straightforward. Figure 17 shows the bimodal curve of two partially miscible solvent systems. The UCST binary system (left) consists of acetone and hexadecane. A UCST system is in fact relevant for enhancing the heat transfer rate in heating applications, since phase separation is induced by cooling (whereby the cooling media is heated). For cooling applications, an LCST system should be used (e.g., triethylamine-water system, Fig. 17, right), which undergoes phase separation upon being heated and can thus serve as a heat sink. In addition, the selected
FIG. 15: Morphology of phase decomposition under flowing condition at different instants of time, i.e., at different quenching depths.

FIG. 16: Phase decomposition of acetone–hexadecane in flowing conditions (field of view corresponds to 0.8 × 0.8 mm); flow direction is indicated by the arrow. Mean velocities are in the range of 0.5–1.5 mm/s.
FIG. 17: Binodal curves of UCST and LCST partially miscible systems: (a) acetone–hexadecane UCST system for enhancing heat transfer in heating applications; (b) TEA–water LCST system for enhancing heat transfer in cooling applications.

system should meet technological requirements, such as the correct temperature range. Price and environmental aspects should be considered as well.

6.2 Experiments with UCST Systems

The solvent system of acetone–hexadecane possesses a UCST at 27°C. This system is of particular interest as both components have practically the same density (0.7876 and 0.7714 g/cm³ at 25°C for acetone and hexadecane, respectively, and the density difference between the separating phases is even smaller). Therefore gravity effects during phase separation are minor and the results obtained can be considered relevant also to microgravity conditions.

The convective heat transfer rates obtained using this solvent system were studied in a series of experiments conducted in a 2 mm I.D. stainless steel pipe, \( L = 180 \text{ mm} \). The pipe was immersed in a cold-water tank to maintain a constant wall temperature below the CST. Under these conditions phase separation takes place in the pipe, as shown in Fig. 18. T-type thermocouples were used to measure the inlet and outlet temperatures and the tube inner surface temperature at several locations (spaced evenly, the deviations between the thermocouples’ reading was about 0.1°C). All the experiments were conducted with laminar flow in the pipe, with Reynolds number varying in the range of \( \text{Re} = 130–750 \). The Pr number of the solvent mixture is typically in the range of 10–15. The reported results correspond to

FIG. 18: Schematic description of the flow loop and test section pipe diameter 2 mm (or 4 mm I.D. in tests with LCST) and pipe length 180 mm (or 130 mm in tests with LCST).
steady-state conditions, which are typically achieved after at least five liquid volume replacements in the pipe. More details on the experimental setup and the experimental procedure can be found at Ullmann et al. (2014a).

In order to calculate the total heat transfer in the pipe test section, the nonideality of a partial miscible solvent system and the associated heat of mixing should be taken into account. An energy balance on a control volume, which represents the pipe test section, considers the enthalpies of the mixture at the inlet and the separated heavy and light phases at the outlet \( h_{\text{mix}}, h_{h,l} \), respectively:

\[
Q = \dot{m}_h (m_h h_h + m_l h_l) - h_{\text{mix}}; \quad \dot{m} = \rho_{\text{mix}} \dot{V}
\]

(17a)

\[
m_h = \frac{\dot{m}_h}{\dot{m}} = \frac{\omega_{\text{mix}} - \omega_l}{\omega_h - \omega_l}; \quad m_l = 1 - m_h
\]

(17b)

The density of the solvent mixture \( \rho_{\text{mix}} \) was calculated based on the overall composition and the temperature at the volumetric flow rate (\( \dot{V} \)) measurement point. The mass fraction of the heavy and light outlet streams, \( m_h \) and \( m_l \), (in terms of the corresponding the mass fraction of acetone \( \omega \) in each of the phases, see Fig. 19). The heat of mixing \( (\Delta h_{\text{mix}}) \) and the mixture heat capacity for the enthalpy calculation were taken from the literature. [According to Shen and Negata (1995), \( \Delta h_{\text{mix}} \) is about 10%–20% of the sensible heat]. Using Eq. (17a) for obtaining the total heat transferred during phase separation, the average heat transfer coefficient \( h \) is defined by

\[
h = \frac{Q}{A_{\text{in}}} \Delta T_{lm}; \quad \Delta T_{lm} = \frac{(T_{\text{out}} - T_w) - (T_{\text{in}} - T_w)}{\ln \left( \frac{T_{\text{out}} - T_w}{T_{\text{in}} - T_w} \right)}
\]

(18)

where \( T_w \) is the tube inner (constant) surface temperature, \( \Delta T_{lm} \) is the log-mean temperature difference, and \( A_{\text{in}} \) is the internal surface area of the test section. The results were compared to the values predicted by the well-established correlation proposed by Hausen (1943) for the convective heat transfer in the thermal developing region (the flow in the test section is assumed to be fully developed laminar pipe flow) of pipe with constant wall temperature:

\[
\text{Nu}_D = 3.66 + \frac{0.0668 Gz}{1 + 0.04 Gz^{2/3}}
\]

(19)

FIG. 19: Binodal curve in terms of the mass fraction \( \omega \) of the separated phases of the UCST acetone–hexadecane system. Determination of the exit flow rate fractions of the heavy and light phases by the lever rule.
where $Gz = (D/L)Pe$, $(D$ is the tube inner diameter, $L$ is the length of the test section, $Gz$ is the Graetz number, and $Pe$ is the Péclet number). The experimental setup and calculation procedure for obtaining the convective heat transfer coefficient were tested and validated by conducting control experiments using water flow and single-phase flow of the CST mixtures (where the wall, inlet, and outlet temperatures are above the UCST), and the validity of this correlation to predict the single-phase heat transfer coefficient was confirmed. Therefore, for all practical purposes, this correlation can be used as a reference to evaluate the extent of heat transfer enhancement induced by phase separation.

When the wall temperature was set below the UCST, phase separation took place in the pipe test section. The results obtained for the Nu number are shown in Fig. 20(a). It can be seen that while the value of the heat transfer

![FIG. 20: (a) Experimental results for forced-convection Nu number obtained with phase separation for various $T_w$ and $T_{in} = 28^\circ$C ($T_{out} < T_{cp}$ see Fig. 8). Comparison with the Nu number obtained without phase separation. The data of $T_{in} = 35^\circ$C and $T_w = 22^\circ$C are associated with $T_{out} > T_{cp}$. (b) Augmentation factor as a function of the flow rate for $T_{in} = 28^\circ$C.](image-url)
The coefficient depends on the wall temperature, it is significantly higher compared to single-phase flow (without phase separation). The augmentation factor, \( AF = \frac{Nu(\text{with phase separation})}{Nu(\text{single phase})} \), of the heat transfer coefficient [Fig. 20(b)] is by a factor of more than 2 for the lower values of the pipe wall temperature, which corresponds to a deeper quench into the unstable region. The decline of the \( Nu \) number and the corresponding \( AF \) at the range of high flow rate is attributed to the decrease of the residence time of the mixture in the cooled test section, whereby the potential augmentation due to phase separation may not have been fully exploited.

The same mixture was also used in Farisé et al. (2012) to assess the heat transfer performances in small-scale heat exchangers. To measure the heat transfer enhancement effect due to spinodal decomposition, a closed-loop experimental setup has been developed (Fig. 21) to pump the mixture from a hot thermostatic bath to the experimental test section, where it is quenched by the cold channel surface. The test section consists of a micro heat-exchanger. A Peltier cell is used to set and maintain the temperature of the cold side of the exchanger throughout the test. The temperature of the cold side of the Peltier cell is kept constant by a PID controller that regulates the duty cycle of the electric power supplied to the cell to keep the temperature constant at \( T_{Cu} \). A controlled temperature of the (single-phase) mixture at the inlet of the heat exchanger is imposed to be above the UCST, 27\( ^\circ C \). A heating thermostat is employed to control the mixture temperature in the bath \( T_{bath} \) by a PID controller. The mixture in the bath is agitated with a magnetic stirrer to keep the temperature as uniform as possible and to facilitate the mixing.

By reading the value of the duty cycle set by the feedback control, an estimate could be obtained on the rate of heat provided by the hot thermostatic bath to the mixture reservoir in order to keep the temperature constant for the imposed conditions of the cooling wall temperature and the flow rate. Experiments have been done on two different types of copper heat exchangers.

The first and most simple heat exchanger is made of nine parallel channels [0.7 mm wide, 1.5 mm tall, and 72 mm long, Fig. 22(a)]. The second heat exchanger, Fig. 22(b), is very similar to actual heat sinks used to cool CPUs on PC motherboards. It is a compact multichannel array (14 U-shaped channels). The two exchangers were sealed with a glass and cooled with the Peltier cell attached to the multichannel array surface.

The exchangers were tested with two quench temperatures of 25\( ^\circ C \) and 20\( ^\circ C \). With 25\( ^\circ C \), visualizations show mild spinodal decomposition, whereas with 20\( ^\circ C \), the quench is deep enough and vigorous spinodal decomposition was observed within the test section. Figure 23 demonstrates results obtained for the heat exchanged by using pure

![FIG. 21: Sketch of the experimental setup used to measure the heat exchange enhancement in the heat exchangers.](image-url)
FIG. 22: The two heat exchangers tested in the experimental setup of Fig. 21: (a) 9 parallel channels and (b) 14 U-shaped parallel channels.

FIG. 23: Electric power absorbed by the 9-parallel-channel heat exchanger with $T_{\text{bath}} = 35^\circ \text{C}$ and $T_{\text{Cu}} = 20^\circ \text{C}$ (i.e., 7°C quench into the bimodal curve). “Mixture theoretical” represents the average of the power absorbed with single-phase flow of acetone and hexadecane.

Fluids (acetone or hexadecane) compared to the heat exchanged by using a critical mixture that separates in the channels.

To evaluate the enhancement effect, the augmentation factor is defined as

$$AF = \frac{\dot{Q}_{\text{Joule}}}{\dot{Q}_{\text{Joule}}^{\text{no decomposition}}}$$

(20)

where $\dot{Q}_{\text{Joule}}$ is the measured electrical power absorbed by the hot-reservoir resistor for the actual flow conditions, and $\dot{Q}_{\text{Joule}}^{\text{no decomposition}}$ is assumed to be equal to the average of the measured electrical power absorbed when using either one of the pure fluids. Further details can be found in Farisé et al. (2012).

Figure 24 shows the heat transfer enhancement vs. the flow rate in each of the multichannels of the heat exchanger. The values of AF are in the range of 1.2–3 and are in reasonable agreement with the values obtained in the single-pipe experiments and in the different experimental setup described above. The AF values are larger at lower flow rates.
Similarly to the trends observed in the pipe flow experiments [Fig. 20(b)], the decreasing trend at high flow rates is attributed to the decrease of the residence time in the test section. This effect becomes pronounced already at lower flow rates in the heat exchanger experiments where the inlet temperatures were higher, well above the UCST (see also Ullmann et al., 2014a).

6.3 Experiments with LCST Systems

The feasibility of using phase separation of LCST systems for enhancing cooling rates was demonstrated in a series of experiments conducted in a 4 mm I.D., $L = 131$ mm stainless steel pipe. A water-triethylamine (TEA) mixture with a LCST of $18.2^\circ$C was used [see Fig. 17(b)]. The experimental setup is similar to that used with USCT solvents, except that in this case the pipe was immersed in a hot-water tank to maintain a constant wall temperature above the CST, while the inlet temperature was below the CST. All the experiments were conducted with laminar flow in the pipe, with Reynolds number varying in the range of $Re = 120–1100$. A mixture with critical composition was used in the experiments. The $Pr$ number of the solvent mixture is in the range of $12–17$. The heat capacity of the solvent mixture at various compositions was measured by an in-house calorimeter and compared to available data from the literature (Flewelling et al., 1996). The heat of mixing data was taken from Bertrand et al. (1968), who measured the heat of mixing at $15^\circ$C. This temperature was selected as a reference temperature in the energy balance [Eq. (17a)]. Other physical properties are available in Yaws (2003). For this LCST mixture the heat of mixing is much higher than that of the UCST system and is up to 80% of the total enthalpy change of the mixture in the test section. Obviously, higher heat of mixing is beneficial, as it reduces the temperature variation of the cooling liquid while flowing in the heat exchanger. Consequently, it enables maintaining the surface at a lower temperature under constant heat flux conditions.

Figure 25 shows the experimental dimensionless heat transfer coefficient ($Nu = hD/k$) obtained in laminar flow with phase separation, in comparison to the values obtained with the same flow rate (same $Gz$ number) without phase separation (depicted in the figure by the single-phase curve). The advantage of inducing phase separation is clearly demonstrated in this figure.
Enhancing Heat Transfer Rates by Inducing Liquid-Liquid Phase Separation

FIG. 25: Experimental results for forced-convection Nu number obtained with phase separation of the LCST water–TEA system for various \( T_w \) and \( T_{cp} \) at \( T_{out} > T_{cp} \), see Fig. 17(b)]. Comparison with the Nu number obtained without phase separation.

The heat transfer rates obtained with laminar flow indicate that phase separation can enhance the convective heat transfer coefficients by 200%–700% (augmentation factor, \( AF = 3–8 \)). It is important to note that the augmentation factor represents the ratio of the Nusselt numbers obtained with phase separation to those obtained with the single phase of the tested liquid system. Therefore it also represents the augmentation relative to heat transfer rates obtained with any other laminar single-phase flow with the same Graetz number.

As water is commonly considered to be the preferred choice due to its superior heat transport properties, the enhancement of the heat transfer coefficients obtained with the LCST water–TEA mixture compared to that of water is of particular interest. Although the thermal conductivity of water is about twice that of the solvent system, the heat transfer coefficients obtained with the phase-transition-induced heat transfer of the LCST system is from 1.5 to 3.5 times that of water for the same \( Gz \) number. When compared to the same volumetric flow rate in the same pipe, the enhancement is even higher—up to 6 times larger (Fig. 26). Preliminary tests were also conducted to examine the effect of phase separation on the pressure drop. The results obtained indicate that augmentation of the heat transfer does not involve a noticeable penalty of increased pressure drop. In LCST systems the pressure drop may even decrease upon inducing phase separation due to the decrease of the fluid viscosity with temperature (Lipstein, 2015).

To estimate the potential enhancement of the heat transfer coefficients, a unified dimensionless correlation to represent the augmentation factor (compared to the single-phase flow value, \( h_{sp} \)) obtained both with the UCST system and with the LCST system was derived:

\[
AF = \frac{h}{h_{sp}} = 0.964Gz^{-0.36}Ja^{-0.46} \left( \frac{T_{out} - T_{in}}{T_w - T_{cp}} \right)^{0.14} \left( \frac{T_{out} - T_{cp}}{T_w - T_{cp}} \right)^{0.55}
\]

(21)

\( T_{in}, T_{out}, T_w, \) and \( T_{cp} \) are the liquid inlet, liquid outlet, wall, and the cloud point (phase transition) temperatures, respectively; \( Ja = (C_p)_{mix}(T_{cp} - T_w)/\Delta h_{mix}/T_w \) and \( Gz = Pe (D/L) \). Ullmann et al. (2009, 2014a,b) reasoned that the last two terms on the right-hand side of Eq. (21) represent the heating rate and the depth (in terms of temperature) of the penetration into the unstable region of the coexistence curve. This agrees with previous findings that increasing these two parameters results in a higher growth rate and movement of the separating domains/droplets during the phase transition (Ullmann et al., 2008).

It is of interest to examine whether the heat transfer augmentation factor \( AF \) can be attributed to the lateral velocity \( v \) of the separating droplet/domain. Assuming the \( AF \) is related to the ratio of the lateral heat convection and the
conduction at the wall, \( AF \approx \rho C_p v/h_{sp} \), the experimental values of \( AF \) and the corresponding \( h_{sp} \) values can be used to estimate the lateral velocity. The values obtained are in the range of \( v = 0.1\text{–}1 \text{ mm/s} \), which are in agreement with measured droplet velocities during phase separation in the closed-cell experiments (see Sec. 2 and Gat et al., 2009).

7. CONCLUSION

In this paper we have reviewed most of the work on heat transfer enhancement by spinodal decomposition. Two research groups (at Tel-Aviv University and at the University of Brescia) have focused on this topic and they have shown by different, but complementary experiments, that phase-separation-induced motion can be effectively used to increase heat transfer performances, especially in small and micro scale devices. Experiments have been conducted with an upper critical solution temperature (UCST) mixture, which can be used for heating applications, and with a lower CST mixture, which can be used for cooling applications. The actual Nusselt number is increased by several times compared to a nonseparating mixture case. Experiments have been performed both in a single-channel configuration as well as in realistic heat exchangers used to cool down electronic devices, such as CPUs. In all cases, increased performances are recorded.

In addition to heat transfer experiments, visualizations have been reported and commented. Spinodal decomposition in static conditions (closed cell) shows a bicontinuous structure (as already reported by previous works), while spinodal decomposition in flowing conditions exhibits morphology closer to nucleation. By image analysis techniques, statistical properties of the concentration field have been obtained.

From a modeling viewpoint, the diffuse interface model was briefly reviewed. Compared to a few other review works on this model, we have stressed the importance of the energy equation, which has been written in term of temperature. This equation includes a source term, which represents the heat of solution in a nonideal mixture. Those simulations, applied to a small domain, show that the convective motion occurring during spinodal decomposition significantly increases the heat transfer at the wall. Even though the diffuse interface model can be solved numerically, as shown in this paper, this method calls for a mesh size so fine that it prevents its use for any practical applications, such as sizing of actual devices. For this reason, we have proposed a coarse-grain approach. In this approach, a
concentration field, which possesses the desired statistical features, is artificially generated. The concentration field is fed into the momentum and energy equations, which are solved independently. By this approach, the solution of the (strongly nonlinear) concentration equation is avoided and equations are decoupled. Such an approach, which is still under development, calls for a much smaller computational effort and therefore can be used, at least in prospective, as a way to design actual devices. Although the models have not yet been developed to the stage where they can be applied to quantitatively predict the very complex multiscale phenomena involved in phase separation of nonideal (real) solution and the associated heat transfer augmentation, the understanding gained enables the identification of controlling dimensionless parameters that were used to develop a semiempirical correlation for the heat transfer enhancement.

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