Structure development in mixing flows

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1 Introduction

Different processes like phase separation, interfacial deformations, coalescence and break-up often occur simultaneously during industrial processing. It is clear that the influence of a final morphology on the mechanical properties is of great interest in many applications. Apart from the characteristics of phase separation, the influence of flow which is imposed to create a structure with desired properties during a processing step, such as mixing, injection moulding or extrusion on the final morphology, is a subject of study for quite some time now.

In this paper two approaches which can be applied to study structure development in mixing flows are discussed. The first model is based on the use of the mapping method and can be applied to study distributive mixing in complex mixers. The second approach is based on a coupling of hydrodynamic and thermodynamic conservation laws and makes it possible to study details of interfacial deformations, including coalescence and break-up in prototypical mixing flows.

2 Distributive Mixing: mapping method

The “mapping” method is proposed based on the original ideas of Spencer and Wiley (1951), and the main idea is not to track each material volume in the flow domain separately, but to create a discretized mapping from a reference grid to a deformed grid. Within the mapping method a flow domain \( \Omega \) is subdivided into \( N \) non-overlapping sub-domains \( \Omega_i \) with boundaries \( \partial \Omega_i \). The boundaries \( \partial \Omega_i \) of these sub-domains are represented by polygons and tracked from \( t = t_0 \) to \( t = t_0 + \Delta t \) using an adaptive front tracking model (Galaktionov et al., 2000), and, as a result, deformed polygons are obtained. Note that the subdivision of \( \Omega \) into \( \Omega_i \)'s is not related to any finite differences, or finite element discretization used to solve the velocity field in \( \Omega \). Two different mixers are addressed: the Kenics and SMX mixer.

Application of the mapping technique to Kenics mixer

One of the widely used static mixing devices is the Kenics mixer (see e.g. Middleman (1977)). It consists of a cylindrical pipe with twisted, perpendicularly placed, rectangular inserts fixed inside. The Kenics static mixer is not regarded as the most efficient device, but the simplicity of its design and the comparatively low pressure drop required for its operation, justifies its frequent use.

The increasing computational power allowed different researchers to perform direct simulations of the three-dimensional flow in Kenics mixers (Avalosse and Crochet, 1997; Hobbs and Muzzio, 1997, 1998a; Hobbs et al., 1998; Hobbs and Muzzio, 1998c; Byrde and Sawley, 1999; Fourcade et al., 2001). The last paper considers even flows with higher Reynolds numbers up to \( Re = 100 \). These studies analyzed only certain particular flows and unlike Khakhar et al. (1987) did not allow for the optimization of the mixer geometry, due to high cost of 3D simulations.
In this work we take into account the most important results of Hobbs and Muzzio (1998b), but we will use a more flexible approach. The mapping method is used to systematically study the performance of Kenics mixers of different geometries (twist direction and angle of the blades) and to find its optimal design.

A more precise evaluation was performed by investigating mixer configurations with a blade twist ranging from $90^\circ$ to $360^\circ$ with a step of $5^\circ$. The results are summarized in figure 2, where the logarithm of intensity of segregation is plotted as a function of pressure drop $\Delta P$ and the blade twist angle $\theta$. This three-dimensional plot exhibits a distinctive valley, the bottom of which, in the region of the larger pressure drops $\Delta P$, is located around the value of blade twist angle $\theta = 140^\circ$. The small “ripple” visible along the $\theta = 180^\circ$ is caused by the fact that for the configurations with larger $\theta$ every blade is modeled with the use of more then three mapping matrices (as for smaller values), causing a slight increase in the “numerical diffusion”, introduced by the mapping computations (more mapping operations mean more per-cell averaging). This, however, does not alter the general trend. Sub-figures a-f of the figure 2 illustrate the mixture patterns, created by mixers with different $\theta$ at roughly the same pressure drop. For illustration purposes the pressure drop chosen is relatively low. The mixer with $\theta = 90^\circ$ creates noticeable “irregularities” in large regions near the tube surface close to the blades. This effect is milder for $\theta = 120^\circ$ and, for the optimal configuration with $\theta = 140^\circ$, these badly mixed zones are small and packed closely to the channel corners. At this location their influence on mixer performance is minimal, since the flux through these zones is low. The mixer with the traditional value of $\theta = 180^\circ$ performs well, achieving good distributions, but due to increased pressure drop per blade, it is less energy-efficient. Finally, it is clear that the poor mixing at higher $\theta$ values, around $\theta = 270^\circ$ (see figure 2e), corresponds to systems with large regular, dead, zones. With further increase of the blade twist angle, the mixer seems to work again, but the high pressure drops required render it inefficient. From figure 2 it can be concluded that the preferable blade twist angle for a RL Kenics mixer with the pitch angle considered in this paper (the same as in Avalosse and Crochet (1997)) operated at close to zero Reynolds number, with Newtonian fluids, should be $\theta = 140^\circ$. The more traditional value of the blade twist, $\theta = 180^\circ$, corresponds to a sharp slope of the valley in figure 2 (line d) and small changes of parameters can be expected to have a strong influence on its performance, although not necessarily deteriorating it.
Figure 2: Optimization of the blade twist angle for RL mixer (Newtonian fluid): logarithm of the intensity of segregation is plotted as a function of the blade twist angle and total pressure drop.
SMX mixer geometry and operation principle

The geometry of the SMX static mixer is illustrated in figure 3, where two mixing elements are shown. Systems of obstacles, forming the mixing elements are fixed inside a cylindrical pipe (shown partially cut open in figure 3) and the flow is induced by an applied pressure difference. Each mixing element is formed by straight rigid strips of rectangular cross-section, which are fixed in alternating pattern inclined with respect to the pipe axis. The mixing elements are fixed tightly inside the pipe so that each next element is oriented perpendicularly with respect to a previous one. The velocity field used in this work was provided by Liu Shiping from the group of Prof. Dr. A. N. Hrymak (McMaster University, Hamilton, Canada). Figure 3 showing two mixing elements of SMX mixer actually depicts the computational domain used in their work.

The results of the performed mapping simulations are shown in figure 4. The initial pattern is composed of a 50-50 black and white distribution and is shown in figure 4a. Figures 4b-f show the concentration distributions after 1 – 5 periods respectively. The resulting patterns are similar to those computed by Zalc et al. (2002) for low Reynolds number $Re = 1$.

The results presented in figure 4 indicate that the mapping approach can be successfully used to simulate the distributive mixing in static mixers with the mixing elements of rather complex shape. The images in figure 4b-f reveal also another characteristic feature of the SMX mixer, which can be typical, however, to this particular design of the mixing elements. Although the mixer quickly creates a large number of material striations, a large scale non-uniformity persist. After five mixing elements it is still visible that one half of the cross-section contains more black material and another – more white. Similar effect can be noticed in the mixing results computed in Zalc et al. (2002). This large-scale distribution irregularity is gradually rotating. The overall rotation effect is caused by the symmetry properties of the mixing elements: the lateral velocity field possesses a rotational symmetry.
Figure 4: The concentration evolution along the SMX mixer. Initial state (a) is specified in the middle of the mixing element (cross section A in figure 3). The mapping one spatial period, that includes half of the mixing element plus half of the next element: for example between cross-sections A and B, according to figure 3. The concentration patterns are shown after 1 – 5 steps in sub-figures (b)-(f) respectively.

3 Structure Development during flow

The mapping method is an efficient way to study distributive mixing processes. It has been applied to prototype mixing flows, static and dynamic mixers. In some cases the method can be applied to optimize the geometry of the mixer. The original mapping method describes the mixture only on macro-level: the smallest unit is a cell and only structures larger than the typical cell size are resolved. The extended mapping technique, introduced in Anderson et al. (2001); Galaktionov et al. (2002), uses a multi-scale approach.

The extended mapping technique describes statistically the interfaces contained in each sub-domain of the mapping grid. The microstructure within each cell is characterized using the second-order area tensor (Wetzel and Tucker, 1999), which is defined for each cell.

The extended mapping technique is a multiscale method in the sense that it treats simultaneously the macroscopic transport, using coarse grain concentration, and describes statistically the inter-material interface patterns finer than the grid cell size using the coarse grain area tensor. The term “multiscale” is used in a sense that the variation (i.e. large striation in a mixture) larger than the grid cell size are resolved due to concentration differences – typical coarse grain density approach, while at the same time the structure (amount and orientation of interfaces) on a sub-grid level is also described.
using the area tensor. These features allow the technique to work for both initial and advanced stages of mixing processes, when macroscopic concentration variations disappear. Muzzio et al. (2000) observed that "for time-periodic chaotic flows, the mixing process is controlled by a stationary multiplicative operator that generates structures that are self-similar with respect to time", although this operator was not obtained. The extended mapping technique actually provides this operator.

If we wish to study details of initial stages of structure development during flow and include hydrodynamic interactions a different approach has to be used. Some results are presented in the following section.

3.1 Diffuse interface method

In the classical approach to multiphase flow, the interface is tracked and appropriate boundary conditions are applied to describe interfacial tension. In the diffuse interface theories first described by van der Waals (1979) and later more specific by Cahn and Hilliard (1958) the interface also has a non-zero thickness, and is no longer arbitrary but now a result of the governing equations. A distinction can be made between immiscible and partially miscible fluids in which phase separation can occur. Phase separation is governed by the free energy of a system. A diffusion-convection equation is introduced to model the topological changes and is driven by a chemical potential which is a result of the free energy description. Frequently, flow is introduced directly in the convective term of the diffusion-convection equation to introduce a simple shear (Berthier, 2001) or the coupled system can be solved to include hydrodynamic interactions.

For the Ginzburg-Landau approximation the following can be derived:

\[
\frac{dc}{dt} = \frac{1}{Pe} \nabla^2 \mu,
\]

\[
\mu = c^3 - c - C^2 \nabla^2 c,
\]

\[
\nabla^4 \psi = \frac{1}{Ca} \frac{1}{C} \nabla \times \mu \nabla c,
\]

The dimensionless groups in the equation are defined by:

\[
P_e = \frac{\rho \xi^2 LV}{M \epsilon}, \quad C = \frac{\xi}{L}, \quad Ca = \frac{\xi \eta V}{\rho c_B^2} = \frac{\eta V}{\gamma},
\]

where the Peclet number $Pe$ is a ratio of the convective and diffusive term, Cahn $C$ a measure of the interface relative to the domain $L$ that is considered and the capillary number a ratio of the viscous forces over the interfacial forces. These three dimensionless groups characterize the set of equations (1 - 3) and define the physical problem to be solved.

3.1.1 Phase separation in shear flow

In this section we present the diffuse interface results for phase separation in a homogeneous shear flow. Two different approaches were taken to apply shear. Shear was applied either via the boundary conditions or prescribed in every nodal point. The concentration, chemical potential and velocity was made periodic.

To solve the Cahn-Hilliard equations spatial and temporal discretisation are required. Model simulations were performed using different time steps and different spectral orders to make sure that
sufficient accuracy is reached. To illustrate the influence of the chosen time step, the maximum concentration in a mesh was followed as a function of time. For the set of dimensionless equations described in the previous section the concentration can vary between -1 and 1 and an initial concentration of the homogeneous mixture can be chosen in between these two values. The simulations described in figure 5 use an initial concentration of 0.3 and a higher order spectral grid \((p = 4)\) with \(20 \times 20\) nodal points. The time steps equal 0.001.

![Figure 5: Phase separation in shear flow, \(C = 0.02\), \(Pe = 1/C\) and \(v_{max} = 0.4\). The initial composition is 65/35 of PMMA and SAN.](image)

The results obtained via a direct implementation of the velocity field in the local composition equation are visualized in the top row of figure 5. The Cahn-Hilliard equations are solved simultaneously:

\[
\frac{\partial c}{\partial t} + v \cdot \nabla c = \frac{1}{Pe} \nabla^2 \mu, \quad Pe = \frac{\beta \xi^2 L V}{M \epsilon},
\]

\[
\mu = c^3 - c - C^2 \nabla^2 c, \quad C = \frac{\xi}{L},
\]
in which the chemical potential \( \mu \) is the driving force for the topological changes \((\partial c/\partial t)\). Besides \( \mu \) and the concentration \( c \) the velocity \( v \) is unknown in these two equations.

For an imposed velocity flow, the velocity is known in every nodal point of the mesh and is therefore unaffected by local concentrations, chemical potentials or gradients of the concentration. In this case only two degrees of freedom remain and two equations to resolve the system. In the top row of figure 5 can be observed how the droplets deform and coalesce in the direction of flow. The droplets tend to align under a certain angle with respect to the flow direction.

If, however, the velocity is an unknown, additionally also the momentum balance has to be solved to resolve the velocity field which is required as input for equations 5 and 6. In the momentum balance interfacial tension is incorporated as a body force to manipulate the velocity field. The velocity is only prescribed on the boundaries such that homogeneous shear can be applied, the interfacial tension \((\mu \nabla c)\) can now be introduced and has, based on its ratio relative to the viscosity, a larger influence on the velocity field.

A smaller capillary number gives a larger contribution when the unknown scalar \( \psi \) and consequently \( v \) \((v = (\partial \psi / \partial y, -\partial \psi / \partial x))\) is solved. The results for different ratios of the interfacial forces relative to the viscous forces are shown in the second, third and fourth row in figure 5 corresponding to a capillary number of 100, 1, 0.1 respectively. Clearly, it is observed how the low-capillary-number simulations show a faster increasing growth of the droplet-size as a function of time. The droplet size in the morphology obtained via the inserted velocity field (top row) closely resembles that of the coupled solution with high capillary number (second row, \( Ca = 100 \)).

4 Conclusions

In the current work the possibility to quickly analyze a wide range of twist angles of the Kenics static mixer with smaller increments \((\Delta \theta = 5^\circ)\) yielded a distinct optimal twist angle equal to \( \theta = 140^\circ \). The criterion used to find this optimum (the volume-flux weighed, slice-averaged, discrete intensity of segregation) seems superior (and of more direct nature) compared to the one used by Hobbs and Muzzio (1998b). Moreover, the mapping method reveals the distinct material striations at more advanced mixing stages then it is typically achievable with marker tracking (Avalosse and Crochet, 1997; Hobbs and Muzzio, 1998a,b).

The diffuse interface calculations were performed using high order spectral elements. Using these special type Galerkin based finite elements, it was possible to obtain a higher degree of accuracy. The result predict in a short time a fair indication of the final morphology that develops given a certain set of dimensionless parameters.

Introducing interfacial tension via the momentum balance increases the range of possibilities to describe phase separation in the presence of a flow field. Not only the limit situation where viscous forces are far more dominant over the interfacial forces can be realistically modeled. Finally, the influence of interfacial tension on the final morphology was clearly demonstrated.
References


