Effects of Reversible Chemical Reaction on Morphology and Domain Growth of Phase Separating Binary Mixtures with Viscosity Difference

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I. Introduction

Phase separation of ordinary binary mixtures, including binary small molecules and polymer blends, has been intensively studied from both experimental and theoretical viewpoints over the past decades, since the resulting morphologies of phase separation play a key role in determining the properties of the mixtures. Usually, the two components of a binary mixture do not possess exactly equivalent characteristics, and there are some differences between them. The existing differences include the viscoelastic contrast between the two phases of polymer solutions and polymer blends with different glass transition temperature, elasticity difference of a binary alloy, and viscosity difference of a binary fluid mixture. Studies about above systems showed that the difference between two components could lead to some drastic changes of domain morphology. To get more interesting

Summary: The effects of a reversible chemical reaction on morphology and dynamics of phase separating binary mixtures with viscosity difference are studied by numerically solving modified time-dependent Ginzburg-Landau and Navier-Stokes equations. Much more interesting morphologies are observed in the system due to the coupling of reversible chemical reaction and viscosity difference between two components. When the chemical reaction rate is relatively low, the impact of viscosity difference on morphologies is prominent, so that the resulting patterns are affected by both reversible chemical reaction and viscosity difference. However, increasing the chemical reaction rate weakens the impact of viscosity difference on morphologies. Similarly, increasing the chemical reaction rate also suppresses the effects of viscosity difference on domain growth dynamics, which is prominent at the early stage of phase separation when the chemical reaction rate is relatively low. For both cases with relatively low and high chemical reaction rates, the average domain size eventually attains an equilibrium value due to the competition between the mixing of reversible chemical reaction and demixing of phase separation.
morphologies and further to provide necessary information for designing blend materials with good properties, the phase separation of binary mixtures with a difference between two components has been recently received more and more attention.

Actually, the phenomenon of viscosity difference between two components is common in the fluid-fluid blends. Even in the homogenous systems, when the two segments of the system fall into different temperature zones, viscosity contrast is created due to the temperature-dependence of viscosity.[6] The effects of viscosity difference between two components in a binary mixture on the morphology of phase separation was numerically studied, and this work showed that the viscosity difference made the morphology different from that of the general phase separation.[15] The morphology and rheology of a phase-separating system with viscosity difference were also studied from both experimental and numerical points of view in the recent years.[7–14] In these works some particular morphologies such as phase inversion of liquid-liquid dispersions under macroscopic shear flow were observed. The viscosity disparity between two components was thought to play an important role in the domain formation and rheological behavior of the blends.

When two nonequilibrium phenomena, such as phase separation and chemical reaction, simultaneously proceed in one system, the phase separation process becomes more complicated and the resulting domain patterns are quite different from those of the ordinary phase separating blends.[15] A wide range of mesoscopic structures such as spatially modulated, concentric (target) and/or labyrinthine patterns in the micrometer range were experimentally observed in the binary or ternary polymer mixtures coupled with photo-cross-linking reaction.[16–20] As a matter of fact, even in the binary polymer mixture coupled with a simple transesterification reaction, a new type of pattern was experimentally observed.[15] To understand the pattern formation and the phase separation dynamics of the binary systems coupled with simple reversible isomerization chemical reaction more clearly, the numerical simulations on this system were performed based on modified time-dependent Ginzburg-Landau (TDGL) equation.[21] It was found that the competing short-range (phase separation) and long-range (reversible chemical reaction) interactions gave rise to steady-state, spatially periodic structures, as those observed in the microphase separation of block copolymers. On the other hand, the formation of steady-state patterns suggests that a reversible chemical reaction can be used to control the final morphology in phase separating binary mixtures.[22]

Our objective in this paper is to numerically examine the effects of a simple reversible isomerization chemical reaction on the morphology and domain growth of phase separating binary blends with viscosity difference. The simulations are performed based on the modified time-dependent Ginzburg-Landau (TDGL) and Navier-Stokes equations. The rest of the paper is organized as follows. In Section II, the model equations and simulation algorithm are briefly introduced. In Section III, the simulation results are presented and discussed in detail, and in the last section, the concluding remarks are presented.

II. Model Equations and Simulation Algorithm

In this paper, the model H is adopted to describe the phase separation process:[23]

$$\frac{\partial \phi}{\partial t} = -(\mathbf{v} \cdot \nabla)\phi + M \nabla^2 \delta H(\phi)$$

$$\rho \left[ \frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla)\mathbf{v} \right] = -\nabla P - \phi \nabla \frac{\delta F(\phi)}{\delta \phi} + \nabla \cdot \mathbf{\sigma}$$

where $\phi$ is the order parameter, defined as $\phi = \phi_A - \phi_B$, with $\phi_A$ and $\phi_B$ being the volume fractions of two components of the binary mixture, respectively. The volume fractions satisfy the conversation law of $\phi_A + \phi_B = 1$. $M$ is the mobility coefficient, and we set $M = 1$ in all the simulations. $H(\phi)$ is the modified Ginzburg-Landau-type free energy functional of a binary mixture with reversible chemical reaction, the same as that of a diblock copolymer,[24,25]

$$H(\phi) = F(\phi) + \frac{\alpha}{2} \int d\mathbf{r} d\mathbf{r}' G(\mathbf{r} - \mathbf{r}') [\phi(\mathbf{r}) - \bar{\phi}] [\phi(\mathbf{r'}) - \bar{\phi}]$$

here the positive constants $\tau$, $\varphi$, and $K$ are phenomenological parameters.

Equation (2) is the modified Navier-Stokes equation, which is used to describe the hydrodynamic flow. Here, $\mathbf{v}$ is the local velocity field, $P$ is the pressure, $\rho$ is the density, and $\mathbf{\sigma}$ is the stress tensor. For the sake of simplicity, we consider the binary blend as a Newtonian fluid mixture, and the stress tensor can be given as

$$\mathbf{\sigma} = \eta(\mathbf{r}) \left[ \nabla \mathbf{v} + (\nabla \mathbf{v})^T \right]$$

where $\eta(\mathbf{r})$ is the local viscosity of the system, which is assumed to be the linear combination of that of the
individual component,\(^5,12\)

\[
\eta(\mathbf{r}) = \phi_A \eta_A + \phi_B \eta_B = \frac{1 + \phi}{2} \eta_A + \frac{1 - \phi}{2} \eta_B
\]  

(6)

where \(\eta_A\) and \(\eta_B\) are the local viscosity of the two components, respectively. Thus the local viscosity \(\eta(\mathbf{r})\) is order parameter dependent.

The divergence of stress tensor is derived from the following equation:

\[
\nabla \cdot \sigma = \nabla \cdot \{ \eta(\mathbf{r})[\nabla \mathbf{v} + (\nabla \mathbf{v})^T] \} \\
= \eta(\mathbf{r}) \nabla^2 \mathbf{v} + \nabla \eta(\mathbf{r}) \cdot [\nabla \mathbf{v} + (\nabla \mathbf{v})^T] \\
\]  

(7)

When the local viscosity of the two components is uniform at all the spatial positions, i.e., \(\eta_A = \eta_B\), and the incompressibility condition of \(\nabla \cdot \mathbf{v} = 0\) is taken into account, Equation (7) becomes the following form:

\[
\nabla \cdot \sigma = \eta \nabla^2 \mathbf{v} \\
\]  

(8)

Equation (1) and (2) can be rewritten using rescaled variables,\(^5\) and the final forms are as follows:

\[
\frac{\partial \phi}{\partial t} = -(\nabla \cdot \mathbf{v}) \phi + [\nabla^2 (-\phi + \phi^3 - \nabla^2 \phi) - \alpha(\phi - \phi^2)] \\
\]  

(9)

\[
\rho \frac{\partial \mathbf{v}}{\partial t} = T_\perp \cdot \{ -\phi \nabla (-\phi + \phi^3 - \nabla^2 \phi) + \eta(\mathbf{r}) \nabla^2 \mathbf{v} \\
+ \nabla \eta(\mathbf{r}) \cdot [\nabla \mathbf{v} + (\nabla \mathbf{v})^T] \} \\
\]  

(10)

Here, \(T_\perp\) denotes the transverse operator. For the case of simplicity, \(\rho\) is assumed to be 1. Thus, we numerically solve the above two equations to study the ordering dynamics of the system. In our simulations, integration are performed using the cell dynamics scheme (CDS) proposed by Onoo and Puri,\(^26\) where the Landau form of \(-\phi + \phi^3 - \nabla^2 \phi\) is replaced by the form of \(-A \tanh(\phi + \phi - D \nabla^2 \phi).\) In the above polynomial \(A\) is a phenomenological parameter greater than 1, characterizing the quench depth, and \(D\) is an interfacial parameter. In our case, \(A\) and \(D\) are set to be 1.3 and 0.5, respectively. All simulations are performed on a two-dimensional \(256 \times 256\) square lattice, in which the lattice size \(\Delta x = \Delta y = 1,\) and the time step \(\Delta t = 0.2.\) The initial order parameter, \(\phi_{ini},\) is artificially set at the beginning of phase separation, and its fluctuation is randomly within \(\pm 0.01.\) The measurement of the domain size \(R(t)\) is given by an interface statistical method, \(R(t) = L_x \cdot L_y/L_0,\) where \(L_x\) and \(L_y\) are the lattice points along the \(x\) and \(y\) axis directions, respectively. The interface length, \(L_0,\) is given by counting lattice points of the discrete space, where \(\phi(\mathbf{r}) \phi(\mathbf{r}') < 0\) with \(\mathbf{r}\) and \(\mathbf{r}'\) being the nearest-neighbor lattice points.

III. Results and Discussions

In the phase separating system coupled with a simple reversible chemical reaction, various interesting morphologies could be obtained. In our previous work, we specially investigated the effect of hydrodynamic effects on the binary mixture coupled with a simple reversible isomerization chemical reaction between the two components.\(^27\) It was found that hydrodynamics played a key role in the formation of special spiral domain patterns in this system when the chemical reaction rate was relatively high. Here, we further examine the coupling effects of viscosity difference and simple reversible chemical reaction between two components in the binary system to understand the evolution behavior of domain pattern and domain size under both critical and off-critical quenches.

A. Critical Case

Figure 1 shows the pattern evolution of the system coupled with reversible chemical reaction and with or without viscosity difference between two components under critical quench. From both Figure 1(a) and Figure 1(b), it can be seen that with the proceeding of phase separation and chemical reaction, new small domains emerge in the initially established domains and the final pattern looks like a perforated sheet. The resulting patterns are quite different from those obtained in the ordinary macrophase separation of binary systems, since in the system coupled with a reversible chemical reaction, the competition between the mixing induced by reversible chemical reaction and the demixing of the phase separation suppresses the concentration fluctuation. Therefore, the simple reversible chemical reaction has strong effects on the pattern formation of phase separating systems.\(^5\)

Although the reversible chemical reaction has same effects on the pattern formation in the phase separation of Figure 1(a) and Figure 1(b), the difference of the pattern evolution and the final pattern between them can be seen clearly. Obviously, this difference is induced by the viscosity difference between two components. As shown in Figure 1(b), when the viscosity difference between the two components is taken into account, the \(A\)-rich phase with low viscosity tends to form isolated domains, and the more viscous \(B\)-rich phase forms connected domains. Our study shows that the more different the viscosity between two components is, the more isolated domains of less viscous phases are observed. It closely agrees with the phenomenon observed in the usual macrophase separation with viscosity difference but without reversible chemical reaction.\(^5\) Indeed, in our case, due to the reversible chemical reaction, in some isolated domains of less viscous \(A\)-rich phase, the secondary droplet domains of more viscous \(B\)-rich phase are created, but the essential characteristic that the less viscous phase forms isolated domains and the more viscous
phase forms connected domains is not altered. Therefore, in this case, the reversible chemical reaction does not severely change the intrinsic behavior of phase separation with viscosity difference. However, if the chemical reaction rate is increased to $G_1 = G_2 = 0.01$, for both the cases with and without viscosity difference between two components, the resulting patterns are quite different from those shown in Figure 1. Figure 2(a) shows the pattern with reversible chemical reaction but without viscosity difference. First, it can be seen that compared with that of the case with relatively low reaction rate, as shown in Figure 1(a), the domain thickness decreases. Second, the special spiral domain patterns are observed when increasing the chemical reaction rate. With the same high chemical reaction rate, when the viscosity difference between two components is taken into account, bicontinuous domain patterns with less domain thickness and the special spiral domain patterns are also observed, as shown in Figure 2(b). Obviously, the patterns shown in Figure 2(a) and Figure 2(b) are similar, which reveals that in this case the effects of viscosity difference between two components on the morphology are not prominent. As it is well known, the chemical reaction continues reducing the driving force of phase separation and a large concentration fluctuation is never established.\(^{[15]}\) Therefore, increasing the chemical reaction rate not only decreases the domain thickness but also weakens the effect of viscosity contrast between two components on morphology. Bu and He\(^{[28]}\) experimentally examined the effect of mixing time on the morphology of immiscible polymer blends and found that at a short mixing time, the morphology of each phase depended on not only the composition, but also the viscosity difference of the two components, whereas as the mixing time increases, the effect of viscosity ratios became less, and the morphology was no longer affected by the viscosity difference. There are some similarities between their experiments and our simulations, since the reversible chemical reaction leads to the mixing between two components, which competes with the coarsening process of macrophase separation in the binary mixtures.\(^{[22]}\) Therefore, the mixing derived from reversible chemical reaction weakens the effect of viscosity difference on the morphology of phase separating system.

From Figure 1 it can be seen that the reversible chemical reaction has effects on the pattern evolution of the phase separation. In order to quantitatively describe the domain pattern evolution during the process of phase separation coupled with the competition of reversible chemical reaction, we study the average domain growth process of the phase separation system with and without viscosity difference between the two components, which is shown in Figure 3. It is seen that for the case of relatively low reaction rate, as shown in Figure 3(a), at the early stage the average domain growth exponent is different in different cases. When the viscosities of both components are identical, the domains of the less viscous system grow faster than those of the more viscous system since in the less viscous system the hydrodynamic effects are more prominent. When the viscosity difference between two components exists, the domain growth exponent locates between the less viscous and the more viscous systems, and nearer to

![Figure 1. Evolution of domain patterns of a critical system with $\phi_0 = 0$, and $\Gamma_1 = \Gamma_2 = 0.001$. Chemical reaction and phase separation take place simultaneously. A-rich regions are shown by white areas whereas B-rich regions are shown by black areas.](image)
the viscous one. It reveals that the domain growth process of phase separation in fluid mixtures is affected by both diffusion and hydrodynamic flow. When any one of the two components has a higher viscosity, it will impede the flow of another component with lower viscosity, and the total fluidity of the system will be reduced. This may interpret the experimental phenomena observed in blends of small molecules and polymer\cite{29} and the system of polymer solutions\cite{30}, where it was found that the growth exponent of domains was around 1/3 although the hydrodynamic flow was relatively prominent in these systems. In our cases, however, after the early stage, the average domain size attains a maximum, then begins to reduce and finally attains a steady value regardless of the viscosity value. This reveals that with the proceeding of both reversible chemical reaction and phase separation, the viscosity difference no longer impacts the domain growth and the average domain size. The competing mixing derived from reversible chemical reaction makes the final average domain size uniform no matter whether there is a viscosity difference between two components or not. It can be confirmed further by increasing the chemical reaction rate. As shown in Figure 3(b), when the chemical reaction rate increases to $\Gamma_1 = \Gamma_2 = 0.01$, the competition of mixing from reversible chemical reaction with demixing of phase separation is so strong that even at the early stage of phase separation, the domain growth is not any longer affected by the viscosity difference between the two components. On the other hand, similar to the result shown in Figure 3(a), the average domain size finally attains a steady value regardless of the viscosity value, but it appears earlier than that of the low reaction rate case shown in Figure 3(a). Therefore, it is confirmed that due to the strong mixing derived from the reversible chemical reaction, the domain growth of phase separation is not impacted by the viscosity difference any longer.

![Figure 2](image.png)

**Figure 2.** Domain patterns of a critical system with $\phi_{ini} = 0$, and $\Gamma_1 = \Gamma_2 = 0.01$ at $t = 10000$. Chemical reaction and phase separation take place simultaneously. A-rich regions are shown by white areas whereas B-rich regions are shown by black areas.

![Figure 3](image.png)

**Figure 3.** Time evolution of the domain size with $\phi_{ini} = 0$. (a) $\Gamma_1 = \Gamma_2 = 0.001$, (b) $\Gamma_1 = \Gamma_2 = 0.01$. Chemical reaction and phase separation take place simultaneously.
To further understand the mixing derived from the reversible chemical reaction, and to study the effects of viscosity difference between two components on domain pattern and domain growth, we numerically examine the phase separating system where the reversible chemical reaction lags behind the phase separation process. The pattern evolutions with relatively low reaction rate and with or without viscosity difference between the two components are shown in Figure 4. It can be seen that before the reversible chemical reaction is turned on, the effects of viscosity difference on morphology are prominent. Although the system is symmetric, the viscosity contrast between two phases leads to the formation of isolated domains of a less viscous phase and continuous structures of a more viscous phase. Once the reversible chemical reaction takes place, however, inside the first established domains new secondary domains are created, and the resulting morphologies are quite different from those obtained before the chemical reaction occurs. Compared with the morphologies observed in Figure 1, it can be seen that for the case without viscosity difference, the morphology observed in Figure 4(a) is similar to that in Figure 1(a), whereas for the case with viscosity difference, as shown in Figure 4(b), more concentric domains are obtained due to the lag of a chemical reaction. Indeed, for this case although the reversible chemical reaction \( \Gamma_1 = \Gamma_2 = 0.001 \) leads to the change of morphology, the effects of viscosity difference on morphology can still be observed clearly.

If the reversible chemical reaction still lags behind the phase separation process, but the chemical reaction rate increases to \( \Gamma_1 = \Gamma_2 = 0.01 \), the resulting morphologies are quite different. As shown in Figure 5, once the reversible chemical reaction takes place, domains with narrow thickness are created. At the beginning of the chemical reaction \( (t = 6000) \), the difference of the domain patterns between the two cases, with and without viscosity difference, is clear, since more concentric domains are obtained in Figure 5(b) than in Figure 5(a) due to the impacts of viscosity difference. As the chemical reaction proceeds, however, the difference between the two domain patterns becomes not obvious as before. Therefore, it reveals again that the mixing derived from the reversible chemical reaction weakens the effects of viscosity difference on morphologies no matter whether the reversible chemical reaction occurs with the phase separation simultaneously or lags behind it.

Figure 6 shows the evolution of the average domain size of the phase separating system where the reversible chemical reaction lags behind the phase separation. The low reaction rate \( \Gamma_1 = \Gamma_2 = 0.001 \) and the high reaction rate \( \Gamma_1 = \Gamma_2 = 0.01 \) cases are shown in Figure 6(a) and Figure 6(b), respectively. It can be seen that for both cases, before the chemical reaction begins, the average domain size grows quickly when the viscosities of two components are low, \( \eta_A = \eta_B = 1 \), since the hydrodynamic effect is prominent in the less viscous system. Once the viscosity of any of the two components increases, for instance, \( \eta_A = 1 \) and \( \eta_B = 5 \), the domain growth exponent decreases, and it is similar to that of the case where both viscosities of two components are high, \( \eta_A = \eta_B = 5 \). It further confirms that the more viscous component dominates the domain growth.

![Figure 4](image-url)
in the system with viscosity difference. However, in our case, once the reversible chemical reaction takes place, the domain size decreases sharply and eventually attains a steady equilibrium value whether or not the viscosity difference exists between the two components. The equilibrium domain size of the case with low reaction rate is higher than that of the case with relatively high reaction rate, and they are almost of the same magnitude as those shown in Figure 3(a) and 3(b), respectively. Therefore, the mixing of the reversible chemical reaction eliminates the difference of domain growth due to viscosity contrast and suppresses the macrophase separation no matter when the chemical reaction is turned on.

To understand the suppression of the reversible chemical reaction on concentration fluctuation of the phase separating system more quantitatively, we examine the temporal change in the normalized concentration distribution function $Q(f)$. Figure 7(a) and 7(b) show the temporal change of $Q(f)$ in the two cases where a chemical reaction takes place with phase separation simultaneously and where it lags behind it, respectively. For the former, the concentration fluctuation is retarded during the whole phase separation process due to the suppression of the reversible chemical reaction throughout. Thus the concentration distribution function in this case presents peaks with broad distribution during the whole phase separation process. For the latter where the reversible chemical reaction takes place lagging behind the phase separation, before the chemical reaction occurs, the concentration fluctuation has reached a relatively large value, and the concentration distribution function has two sharp peaks ($t = 5000$). Once the reversible chemical reaction starts, the distribution of $Q(f)$ is broaden and presents two multi-peaks ($t = 5000$). Although it reverts to double peaks as the chemical reaction proceeds, the distribution of $Q(f)$ still has a broad distribution.
Therefore, the reversible chemical reaction, which competes with the macrophase separation, suppresses the concentration fluctuation during the phase separation process.

**B. Off-Critical Case**

For simplicity, only the system slightly far from the critical point is taken into account here, where the initial volume fraction of the A component is set at 0.55. The corresponding patterns of off-critical systems with and without viscosity difference at $t = 10000$ are shown in Figure 8. In this case, the phase separating systems are also coupled with a reversible chemical reaction, where the reaction rate, $\Gamma_1 = \Gamma_2 = 0.001$. It should be noted that the order parameter in equilibrium would be zero due to the equal forward and backward reaction rates although the initial asymmetry of the volume fractions (It will be confirmed latter by showing the evolution of the average volume fraction of component A, $\phi_A$). However, the morphologies in equilibrium do not present the expected bicontinuous structures in both the cases with and without viscosity contrast between the two components, as shown in Figure 8. The likely interpretation is that the macrophase separation proceeds faster than the change of the average order parameter since the chemical reaction rate is not large enough.\[20\] For all the cases shown in Figure 8, the initial major A-rich phase forms a network pattern, and droplet domains of the opposite B-rich phase disperse in the A-rich phase. Although more or less secondary droplet domains of A-rich phase are created inside the B-rich domains due to the reversible chemical reaction, the effects of viscosity difference on morphologies can be observed clearly. Obviously, the morphology of $\eta_A = 1$ and $\eta_B = 5$ is similar to that of the case $\eta_A = \eta_B = 1$, and the morphologies of $\eta_A = 5$ and $\eta_B = 1$ and $\eta_A = \eta_B = 5$ are similar to each other. It reveals that the domain patterns are dependent on the viscosity of the initial major phase. However, if we increase the chemical reaction rate to $\Gamma_1 = \Gamma_2 = 0.01$, it is found that due to the strong mixing derived from the reversible chemical reaction, in all the cases with and without viscosity difference between the two components, the final patterns present the random bicontinuous structure with some screw dislocations or target domain patterns, which are similar to those observed in the critical system shown in Figure 2. Therefore, increasing the chemical reaction rate would not only make the change of

![Figure 7](image_url)

Figure 7. Temporal change of $Q(\phi)$ with $\eta_A = 1$, $\eta_B = 5$, $\phi_{ini} = 0$, and $\Gamma_1 = \Gamma_2 = 0.001$. (a) Chemical reaction and phase separation take place simultaneously, (b) Chemical reaction occurs after $t = 5000$.

![Figure 8](image_url)

Figure 8. Domain patterns of an off-critical system at $t = 10000$, with $\phi_{ini} = 0.1$, and $\Gamma_1 = \Gamma_2 = 0.001$. Chemical reaction and phase separation take place simultaneously. A-rich regions are shown by white areas whereas B-rich regions are shown by black areas.
average order parameter match the macrophase separation process, but also weaken the impact of viscosity contrast on morphologies.

For the phase separating systems with reversible chemical reaction, the order parameter in equilibrium can be not consistent with the initial one through adjusting the reaction rate constants. In our case, the forward and backward reaction rates are equivalent, so that the volume fractions of the two components should be equivalent if the chemical reaction is allowed to proceed for enough time. Figure 9 shows the time evolution of the volume fraction of the A-rich phase in the off-critical system with and without viscosity difference between the two components. Both the cases where the reversible chemical reaction takes place with the phase separation simultaneously or occurs lagging behind the phase separation are taken into account, and the corresponding results are shown in Figure 9(a) and Figure 9(b), respectively. It can be seen that for both cases no matter whether there is a viscosity contrast or not in the system, the volume fraction of component A evolves from 0.55 to 0.5 as the chemical reaction proceeds, and all evolution curves overlap together. It reveals that the evolution of the volume fraction of different cases with and without viscosity difference follows the same behavior, which is dominated by the reversible chemical reaction.

IV. Conclusions

In this work, the effects of a reversible chemical reaction on the phase separation of a binary blend with viscosity difference were studied with modified time-dependent Ginzburg-Landau and Navier-Stokes equations. Compared with the usual binary mixture, the morphologies of the system are much more multifarious due to the coupling of reversible chemical reaction and viscosity contrast. For the critical system, when the chemical reaction rate is relatively low, the impact of viscosity difference between the two components on morphologies is prominent. The viscosity difference also has effects on the domain growth dynamics, but the impacts become not prominent after the early stage of phase separation since the domain size attains an equilibrium value due to the mixing derived from the reversible chemical reaction. If the chemical reaction rate increases, the impacts of viscosity difference between the two components on morphologies and domain growth become not prominent due to the strong mixing by the reversible chemical reaction. In the off-critical system, due to the equivalent forward and backward reaction rates, the order parameter in equilibrium is zero regardless of the initial asymmetry of the volume fraction. When the chemical reaction rate is relatively low, the final domain patterns are dependent on the viscosity of the initial major phase, and the viscosity difference between two components has effects on the domain patterns. However, when we increase the chemical reaction rate, the impacts of the viscosity difference between two components on domain patterns become not prominent any longer. In addition, in spite of the diversity of morphologies and the distribution of order parameter in different systems with or without viscosity contrast, the evolution of the volume fraction follows the same behavior.

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