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Effect of Salt Concentration on the Pattern Formation of Colloidal Suspension

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Abstract

We study the effect of salt concentration on the drying process and pattern of thin liquid layer colloidal suspension. Panasonic camera is used to capture the drying process and macroscopic pattern. Microscopic patterns are analyzed by optical microscopy. It is shown that broad-ring pattern is avoided by adding little amount of sodium chloide into colloidal suspension. with the increase of salt concentraion, convection strength and interface instability are weakened, thus the edge of film becomes smooth and more homogeneous film forms. Beautiful microscopic patterns demonstrate that the cooperative interaction between sodium chloide and silica spheres has important influence on the pattern formation.

*Keywords:*colloidal suspension; interface instability; cooperative interaction

1. Introduction

Solids dispered in a drying drop will migrate to the edge of the drop and form a solid ring. This phenomenon produces ring-like stains and occurs for a wide range of surfaces, solvents, and solutes. Ring-like stains are a general phenomenon such as the residue left when coffee drying on the countertop and mineral rings left on washed glass ware. Controlling the distribution of solute during drying is vital in industrial and scientific processes. For example, paint manufactures use a variety of additives to ensure that the pigment is evenly dispersed during drying[1]. Generally speaking, most drying patterns in nature are formed by self-organization accompanied by the dissipation of free energy and in the nonequilibrium state[2]. Since patterns themselves in nature are complex to study, great attention has been devoted to the structural pattern formation in the drying process of colloidal suspension. The drying process of colloidal suspension can also be used in fabrication of various nanostructured materials and devices. Several papers on pattern formation in the drying process of suspensions and solutions on a cover glass have been reported

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hitherto. Reference [3-6] reported that an obvious feature of the drying pattern was the appearance of similar macroscopic broad-ring patterns. They clarified that the broad-ring patterns were formed unexpected fast in the suspension state. Electrostatic interparticle interactions have been pointed out as one of the important factors for the dissipative structures. Hydrophobic and hydrophilic interactions were also demonstrated to be important during the drying process. In addition, the role of capillary force and convectional flow in the dissipative pattern formation were also emphasized. Since a large-scale nanostructured film with a flat and uniform surface is anticipated for applications in photonic crystal engineering, it is necessary to avoid broad-ring formation.

In this study, the drying processes and patterns of colloidal suspensions containing different amount of sodium chloride were studied in open container. The main purpose of this study is to investigatd the influence of salt concentration on the dissipative structural patterns.

2. Experimental

The diameter of silica colloidal particles is 450nm, and the relative deviation in size is less than 5%. The silica spheres were dispersed into the deionized water containing sodium chloride with the assistance of an ultrasound bath forming a 1wt% suspension. Concentration of NaCl ranged from 0.006M to 0.06M. The glass substrates were rinsed with deionized water and dried under a flow of nitrogen gas. For each sample, a circular silica glue mat with 1cm inner diameter was set onto a glass substrate, and 180μl colloidal suspension was dropped onto the glass substrate. The samples were placed on a desk until dried up completely in a room air-conditioned at 22℃. Drying process and macroscopic patterns of colloidal silica spheres were observed with a Panasonic digital camera. Microscopic drying patterns were analyzed by optical microscopy(OM).

3. Results and discussion

Fig. 1 shows the macroscopic sedimentation and drying patterns of silica colloidal suspensions in the presence of salt. Distinct difference is observed among the drying patterns of the suspensions containing different amount of sodium chloride. We found that with the increase of salt concentration, the outer edge of film become more smooth. More homogeneous film was obtained at higher salt concentration. It is worth noting that we find that adding little amount of sodium chloide into the colloidal suspension can avoid broad-ring formation. As is shown in the blue rectangle of Fig. 1(b) and 1(c), when the salt concentration is relatively low, phenomenon of component separation between silica sphere and sodium chloide was observed. Sodium chloide distribute mainly in the thicker film region.

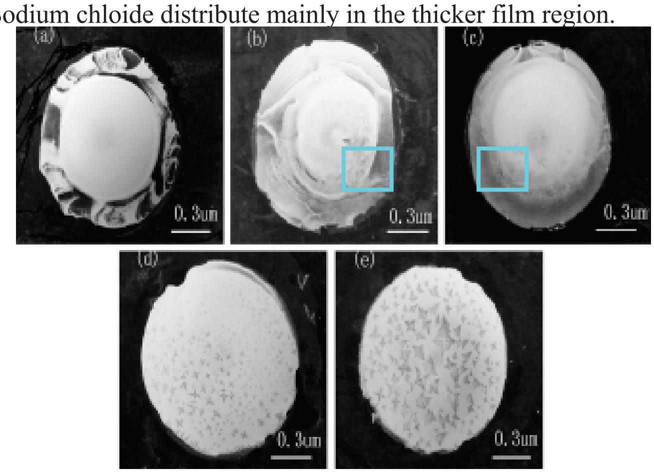

Figure 1. Drying patterns formed for colloidal silica spheres in open containers at 22 ℃
(a)[NaCl]= 0M (b) 0.006M (c) 0.01M (d) 0.02M (e) 0.06M

Results of optical micrographs prove the phenomenon of component seperation, in Fig. 2. Black region indicate thiner film composed of silica spheres in the outer edge, spot-like and branch-like pattern formed by sodium chloide distribute non-uniformly in the white region, which indicate the thicker film in the center. This is due to the different diffusion coefficient of the sodium chloide and silica sphere in fluid, that is $D_{NaCl}=1.5\times10^{-9}m^2/s$, $D_{SiO2}=2\times10^{-12}$ m^2/s. According to a Peclet number, *Pe* proposed by Routh and Russel, defined as

$$Pe=HE/D \tag{1}$$

Where H is the initial thickness of the film, E the rate of evaporation, D the diffusion coefficient. The Peclet number is used to denote relative proportion of convection and diffusion. From (1), it is easily to be understood that convection strength has a inverse proportional variation with the diffusion coefficient. In this letter, the higher diffusion coefficient of sodium chloide is, the weaker convection strength is. Convection strength plays an important role in drying pattern formation, as in [7]. Formation of broad-ring pattern during drying colloidal suspension is mainly caused by the Marangoni convection. The rate of evaporation at the edge is greater than that at the center. To compensate for water lose at the edge, the capillary flow occurs from the center to the edge, enabling colloidal particles to flow and accumulate near the round edge, as in [1]. For our expeiment, when adding little amount of sodium chloide into silica colloidal suspension, sodium chloide and silica sphere will flow to the edge at different convectional rate, which lead to the phenomenon of component seperation.

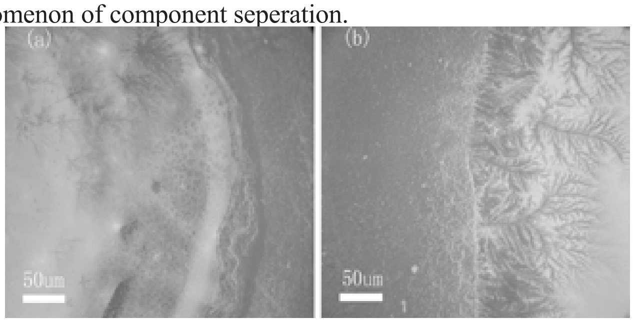

Figure 2. Optical micrographs of colloidal suspensions in blue retangle
(a)[NaCl]= 0.006M (b) 0.01M

Fig. 3 shows microscopic drying patterns of colloidal suspensions containing different amounts of sodium chloide ranging from 0.006M to 0.06M. Black spot-like microscopic patterns appear for the suspension containing sodium chloide of 0.006M, as shown in Fig. 3(a). Fig. 3(b) shows beautiful branch-like patterns and a few amount of spot-like patterns are formed for the suspension containing 0.01M NaCl. Fig. 3(c) and 3(d) show that star-like patterns are formed in the microscopic patterns of the suspension containing sodium chloide of 0.02M and 0.06M. Different shape and size of final condensate appear in the microscopic drying patterns of silica colloidal suspensions containing different concentration of sodium chloide. These beautiful micoscopic patterns demonstrate that the cooperative interaction between sodium chloide and silica spheres has important influence on the pattern formation in the processes of solidification of colloidal spheres and sodium chloride.

Fig. 4 shows comparison of the evaporation and drying processes for colloidal suspensions containing different amount of sodium chloide ranging from 0.006M to 0.06M. When the salt concentration is 0.006M (Fig. 4 (a-c)) , ring-like liquid layer occurs rupturing suddenly during drying process, which has great influence on the final drying pattern. This phenomenon is related to interface instability. When liquid layer is very thin(10-100nm), even in static liquid layer, rupture occurs because of long-range molecular force. The rupture phenomenon during drying process of thin layer colloidal suspension containing 0.006M sodium chloide was never reported so far. With the increase of salt concentration, interface instability is weakened and more homogeneous and flat film forms. A main reason is that the electrical double layers around the spheres are thin when the ionic concentration is high. The diffusion of spheres will be more vigorous because the effective size of spheres including the double layers decreases. According to (1), convection strength is weakened, and sedimentation plays an important role on self-organization of spheres and pattern formation. Thus, more flat film formed.

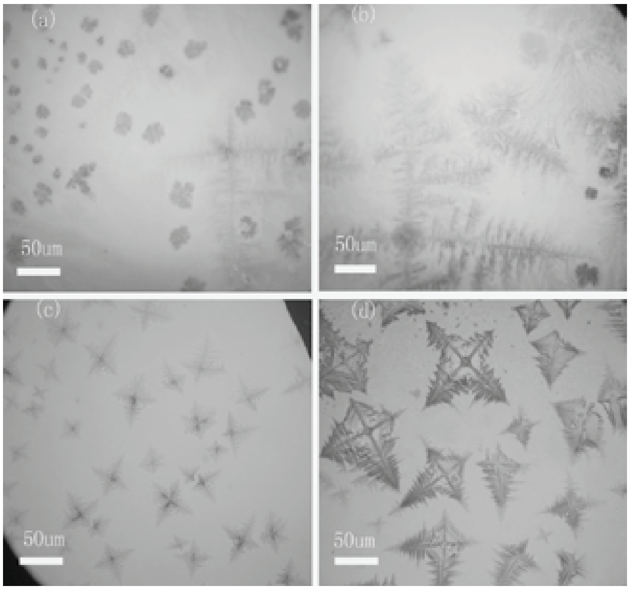

Figure 3. Microscopic drying patterns of colloidal suspensions containing different amount of salt
(a) [NaCl]= 0.006M (b) 0.01M (c) 0.02M (d) 0.06M

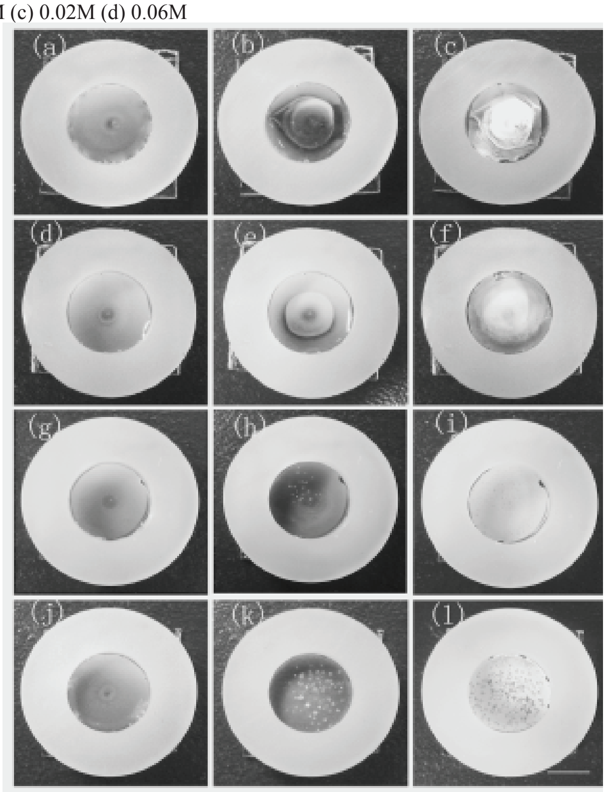

Figure 4. Drying process of colloidal suspensions of different salt concentration
(a-c) [NaCl]=0.006M (d-f) 0.01M (g-i) 0.02M (j-l) 0.06M
(a)After 8h (b) 11.5h (c) 12.5h (d)7.5h (e) 11h (f) 11.5h (g)7.25h (h) 10.5 h (i) 11h (j) 7h (k) 10.25h (l)10.5h

4. Conclusions

In summary, our experiments show that salt concentration has great influence on the macroscopic and microscopic pattern of colloidal suspension. Adding little amount of sodium chloide into the colloidal suspension can avoid broad-ring formation. Component separation phnomenon is observed because different diffusion coefficient of sodium chloide and silica sphere, which affects the convection strength. When the salt concentration is low, rupture because of interface instability is observed. Interface stability and convection strength are weakened, and more flat film can be produced at higher salt concentration. Spot-like, branch-like, and star-like microscopic patterns are observed, which demonstrate that the cooperative interaction between sodium chloide and silica spheres has important influence on the pattern formation.

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