

Transition in the pattern of cracks resulting from memory effects in paste

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Experiments involving vibrating pastes before drying were performed for the purposes of controlling the crack patterns that appear in the drying process. These experiments revealed a transition in the direction of lamellar cracks from perpendicular to parallel when compared with the direction of the initial external vibration as the solid volume fraction of the paste is decreased. This result suggests a transition in the memory in paste, which is visually represented as morphological changes in the crack pattern. Given that the memory in paste represents the flow pattern induced by the initial external vibration, it should then be possible to control and design various crack patterns, such as cellular, lamellar, radial, ring, spiral, and others.

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It is very important to find methods to control crack patterns in various fields of science and technology. So far, few successful examples of such control have been reported, one such success being the case of quasistatic fractures in thin glass plate, in which control of the morphology of crack patterns was proven to be possible by changing the cooling rate during quenching [1–4]. Recently, we have developed a method for controlling the morphology of the crack patterns that appear during the drying of pastes [5–7]. A paste is a concentrated colloidal suspension with plasticity. When a mechanical force is applied to a paste before drying, the paste “remembers” the force in the form of microscopic structural changes within the suspension. This memory is represented visually by the crack pattern that later appears in the drying process. In this Rapid Communication, anisotropic colloidal particles were used to produce a transition in the crack patterns of pastes, which suggests the existence of various memory structures in the suspension.

A paste, as defined in this study, consists of a powder mixed with water. This mixture is poured into a container and kept in an air-conditioned room at a fixed temperature of 25 °C and a humidity of 30%. As the mixture dries, the mixture shrinks due to the evaporation of the water. When adhesion between the mixture and the container is strong, the mixture sticks to the bottom of the container, prohibiting contraction of the mixture in the horizontal direction unless the mixture splits into smaller pieces through the formation of cracks. The formation of cracks continues until the sizes of the fragments are approximately the same as the thickness of the mixture [8–11]. Usually, crack patterns form isotropic cellular structures, in that the sizes of these fragments are almost the same, the shape of each fragment is isotropic, and they are distributed randomly in space.

Recent work has indicated that the morphology of crack patterns can be controlled by applying an external horizontal vibration to a paste before it has dried [5–7,12,13]. In these experiments, we used calcium carbonate particles CaCO_3 as the powder to make pastes. Applying an initial vibration to the paste with a strength just above that of the yield stress of the paste led to the formation of lamellar cracks during the

drying process. The orientation of these lamellar cracks was perpendicular to the direction of the initial vibration.

In this Rapid Communication, the powder was changed from calcium carbonate to magnesium carbonate hydroxide (Kanto Chemical, Tokyo, Japan), and the same experiments were performed as those described above. The density of magnesium carbonate hydroxide is 2.0 g/cm^3 . Microscopic observations via a scanning electron microscope (Hitachi, Tokyo, Japan) indicated that the shapes of dry magnesium carbonate hydroxide particles are disk-like, with a diameter of $1.5 \mu\text{m}$ and a thickness of $0.2 \mu\text{m}$. In these experiments, square containers were used with the length of each side being 200 mm. The mass of the powder in the mixture was fixed at 100 g in each container, such that the final thickness of the mixtures with different solid volume fractions was equal when they dried, thus equalizing the characteristic sizes of the final crack patterns. The pastes were vibrated horizontally for 60 s immediately after pouring the paste into the container. The formation of cracks was then observed as the pastes dried.

Figure 1 indicates that a decrease in the solid volume fraction ρ of the paste results in a transition in the direction of the lamellar crack patterns from perpendicular [Fig. 1(a)] to parallel [Fig. 1(b)] when compared with the direction of the initial vibration. Systematic experiments were performed

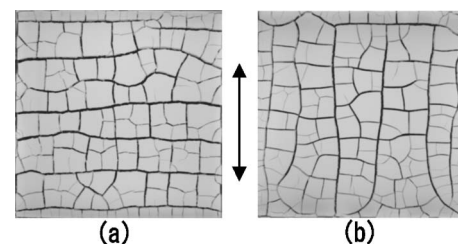


FIG. 1. Transition in the direction of lamellar crack patterns. The arrow between (a) and (b) indicates the direction of the initial vibration, where the amplitude r and the frequency f of the vibration are 15 mm and 2 Hz, respectively, i.e., the strength $4\pi^2rf^2$ is 2.4 m/s^2 . The lengths of the sides of both square containers are 200 mm. (a) The solid volume fraction ρ is 12.5%. The direction of the lamellar cracks is perpendicular to the direction of the initial vibration. (b) The solid volume fraction ρ is 6.7%. The direction of the lamellar cracks is parallel to the direction of the initial vibration.

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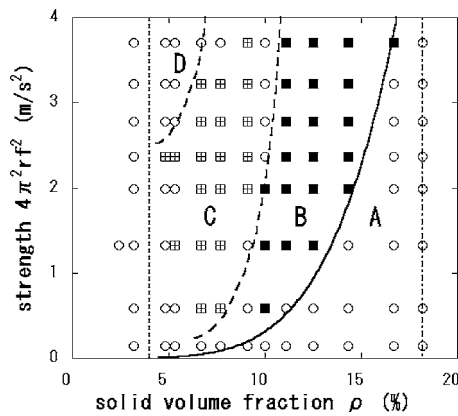


FIG. 2. Morphological phase diagram of the crack patterns that appear in the drying process of pastes made of magnesium carbonate hydroxide, shown as a function of the solid volume fraction ρ and the strength $4\pi^2 r f^2$ of the initial vibration. Open circles denote isotropic cellular crack patterns, solid squares denote lamellar crack patterns, the direction of which is perpendicular to the direction of the initial vibration, and open squares with a plus inside denote lamellar crack patterns, the direction of which is parallel to the direction of the initial vibration.

to obtain a morphological phase diagram of the crack patterns, shown in Fig. 2, as a function of both the solid volume fraction ρ and the strength $4\pi^2 r f^2$ of the initial vibration. We also performed rheological measurements of the yield stress of the pastes via a dynamic stress rheometer (Rheometrics, Piscataway, NJ). When the value of ρ is smaller than the liquid limit $\rho=4.0\%$, which is denoted by the vertical dotted line, only isotropic cellular crack patterns appear. When the value of ρ is larger than the plastic limit $\rho=18.2\%$, which is denoted by the vertical dash-dotted line, a morphological phase diagram is not obtained, as mixing powder with water homogeneously is not possible due to the lack of a sufficient amount of water. The region between these two vertical lines is divided by the solid and the dashed curves into four regions A, B, C, and D. The solid curve corresponds to the yield stress line, on which the strength of the initial vibration is equal to the strength of the yield stress of the paste. Isotropic cellular crack patterns appear in regions A and D. In region A, the value of the strength $4\pi^2 r f^2$ of the initial vibration is smaller than that of the yield stress, while in region D, turbulent flow induced by the initial vibration destroys the memory. In regions B and C, lamellar crack patterns are obtained that indicate a memory of the initial vibration, but the directions of the lamellar cracks are perpendicular in region B and parallel in region C when compared with the direction of the initial vibration.

We previously reported that in the drying process of a paste made of calcium carbonate, lamellar cracks emerged when the paste was vibrated at the strength just above the yield stress, and the directions of the lamellar cracks were all perpendicular to the direction of the initial external vibration [5–7]. This corresponds to the formation of lamellar cracks in region B in Fig. 2. It is postulated that, when the densely packed colloidal particles are vibrated, longitudinal density fluctuations emerge inside the suspension due to the inelastic collisions between particles. This is similar to the formation

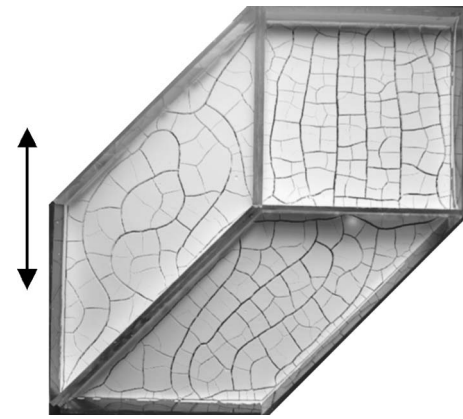


FIG. 3. Memory of flow patterns in region C. Here, the solid volume fraction ρ is 7.7%. The length of each side of the square container is 200 mm and the lengths of the neighboring sides of the two parallelogram-shaped containers are 200 and $200\sqrt{2}$ mm. The arrow indicates the direction of the initial external vibration, where the amplitude r and the frequency f of the vibration are 15 mm and 1 Hz, respectively, i.e., the strength $4\pi^2 r f^2$ is 0.6 m/s². The flow patterns caused by the initial external vibration are memorized and are exhibited by the crack patterns.

of density waves or jammed structures in dense granular flows [14]. The polydispersity of the particles also helps the shear-induced segregation caused by horizontal oscillations to form a lamellar pattern perpendicular to the direction of the oscillation [15]. Due to its plasticity, the network of colloidal particles maintains the microscopic anisotropic structure induced by the initial external vibration even after the vibration is removed [16].

As for the lamellar cracks in region C in Fig. 2, however, it is believed that there is a relation between the direction of the lamellar cracks and the direction of the flow induced by the initial external vibration. To test this theory, experiments were performed in different container shapes. Figure 3 shows that, in parallelogram-shaped containers, the direction of lamellar cracks is parallel to the direction of the flow along the oblique direction and not to the direction of the external vibration. That is, the memory in region C represents the flow pattern.

Figure 4 shows the results of experiments designed to check the effects of surface waves induced by the initial vibration. Here, the solid volume fraction ρ is 7.7%, which corresponds to the paste in region C. We used a circular container with a diameter of 500 mm, and the mass of the powder in the mixture was 491 g in order to equal the final thickness of the mixtures in Figs. 1–3. Before drying, the paste was covered to suppress the formation of surface waves. The cover was then rotated counterclockwise, and then removed for drying. Figure 4 shows that, even with no surface waves, a ring pattern forms, which is the flow pattern produced by the rotation of the cover. It also shows that vibration is not necessary for the formation of a ring pattern. Note that vibration is necessary for the formation of lamellar cracks in region B. The transition of the direction of the lamellar cracks from region B to region C is a visual representation of the transition of the memory in the paste from “memory of vibration” to “memory of flow.”

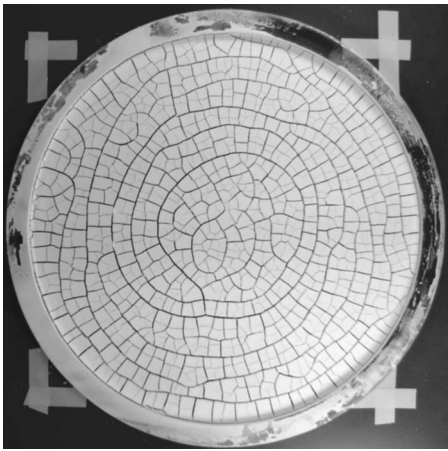


FIG. 4. A ring pattern. Here, the solid volume fraction ρ is 7.7%. The diameter of the circular container is 500 mm. Before drying, we put a cover on the paste, rotated the cover counterclockwise at a period of 1 s for 10 s, and then removed the cover for drying. It can be seen that no surface wave or vibration is necessary for the formation of a flow pattern.

Given the above results, one question that can be asked is, why does the paste made of magnesium carbonate hydroxide retain the flow pattern, while that of calcium carbonate does not? To answer this, we must consider how the particles interact with each other in each of the suspension. In both pastes, the colloidal particles interact via van der Waals attractive forces. Additionally, CaCO_3 particles also interact with each other through Coulombic repulsive forces [7]. Given these Coulombic repulsive forces, only at high solid volume fractions at which particles are densely packed do CaCO_3 particles attract each other to form a network structure. A mixture of powder and water with such a high solid fraction has plasticity with a nonzero yield stress. In comparison, the colloidal suspension of magnesium carbonate hydroxide can form a network structure even at low solid volume fractions, because the particles interact via attractive forces only. The shapes of the particles also play a role. It is easy to pack CaCO_3 particles densely because the shape of the particles is isotropic, while the disklike particles of magnesium carbonate hydroxide stick together easily and form a structure similar to a card house with many voids in the interior, resulting in a low solid volume fraction. This rationale is supported by the experimental results in that the liquid limit and the plastic limit of the colloidal suspensions of calcium carbonate were as high as 25.0% and 54.0%, respectively, while those of magnesium carbonate hydroxide were as low as 4.0% and 18.2%, respectively. Since a colloidal suspension of magnesium carbonate hydroxide has plasticity even at low solid volume fractions, the flow that emerges at low solid volume fractions can be retained. It is believed that the flow induced by the initial external vibration elongates a dilute network structure to produce a lamellar microstructure, the direction of which is parallel to the direction of the flow, with the normals of the disklike particles perpendicular to the direction of the flow [17,18]. In fact, when we prepared two pastes with the same solid volume fraction and vibrated only one of them before drying, the lamellar cracks

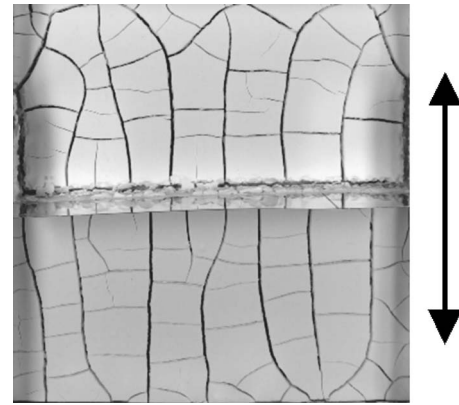


FIG. 5. Experimental result to check the position of the lamellar cracks in region *C*. Here, the solid volume fraction ρ is 6.7%, and the lengths of the sides of the square container are 200 mm. The arrow indicates the direction of the initial vibration, where the amplitude r and the frequency f of the vibration are 15 mm and 1.5 Hz, respectively, i.e., the strength $4\pi^2rf^2$ is 1.3 m/s^2 . After vibrating the paste, we cut the paste into halves and let them dry. The positions of the lamellar cracks are discontinuous at the boundary between the halves.

that appeared in the vibrated paste always emerged earlier than the isotropic cellular cracks in the paste that was not vibrated. This result indicates that the network of colloidal particles is weakened by the formation of an anisotropic microstructure.

It is believed that the order parameter that quantifies the transition between phases *B* and *C* is anisotropy in the microstructure, which is visualized by the macroscopic crack pattern. Only one shake is enough to create anisotropy in a paste, but to obtain a beautiful lamellar crack pattern, a paste should be vibrated more than ten times. Note that, near the transition line between phases *B* and *C*, a combined lamellar and cellular pattern is observed. Since the level of anisotropy is low near the transition line, dynamic instability results in

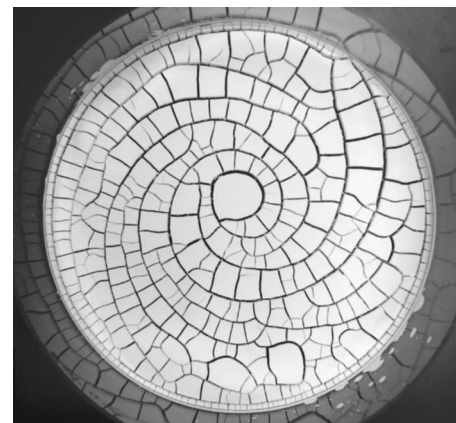


FIG. 6. A spiral crack pattern. Here, the solid volume fraction ρ is 7.7%. The diameter of the circular container is 500 mm. Before drying, the container was translated along a circuit with a radius of 15 mm clockwise at a period of 1 s for 60 s, so that the container pushes the paste inward and clockwise simultaneously to produce a spiral flow. Note that the container was not rotated around its center.

unstable straight crack propagation, which leads to a cellular pattern in crack formation [7].

The positions of the lamellar cracks in region *C* were checked to explore the possibility that the lamellar cracks were induced by macroscopic hydrodynamic instability. If the positions of the lamellar cracks were determined by the initial vibration, macroscopic hydrodynamic instability, such as convection or surface waves, should play a role in memory formation in pastes. Any effects from surface waves can be eliminated based on the results in Fig. 4. Figure 5 shows that even if the directions of the lamellar cracks are determined by the direction of the initial vibration, the positions of the lamellar cracks are random. These experimental results support our interpretation that the memory in pastes is retained by a microscopic anisotropic structure of an orientational order, the scale of which is extremely small compared with the characteristic size of the macroscopic crack patterns.

To summarize, we performed drying experiments and revealed a transition in the memory in pastes that is visually represented as morphological changes in the crack pattern. The ability to mechanically imprint a flow pattern into a paste by changing the shape of container and/or the mechanical force allows for design of various crack patterns, including cellular, lamellar, radial [5–7], ring, and spiral (shown in Fig. 6) patterns.

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