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Ultrasonic Formation of a 3-D Droplet Array in Elastomer
超音波による弾性ポリマー中の 3 次元液滴配列の形成

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1. Introduction

Optics reseachers have been interested in microspheric resonators, since they provide efficient microlasers and biochemical sensors that are based on the whispering-gallery mode (WGM).[1–4] Arrayed spheres are also useful for creating photonic crystals.[5] Being compared to ordinary solid spheres, e.g., polystylene or glass spheres, droplets have the following advantages;[6,7] i.e., the surface tension helps self-formation of spheres with a smooth surface, deformability of droplets allows resonance wavelength tuning, and solubility of fluorescent or photochromic dyes provides droplets with various optical functions. Unfortunately, however, droplets are difficult to handle and lacks chemical and mechanical stabilities. These probelms can be solved if droplets are encapsulated in an elastomer, i.e., a deformable resin.[8] In this study, we created a 3-D droplet array in a polydimethyl siloxane (PDMS) resin by using the ultrasonic trapping method;[9–15] i.e., droplets were arranged by the 2- or 8-MHz standing waves in a PDMS resin during its solidification process.

2. Fabrication apparatus

The PDMS resin is a suitable solid matrix for suspending droplets, since it has the following advantages. 1) The starting material is a liquid (oil) that can suspend droplets as well as various particles. 2) The starting liquid solidifies at room temperature by adding a curing agent. 3) The chemical and thermal stabilities as well as nonwettability are suited for molding processes including nano-imprinting. 4) No toxic materials is contained in this polymer. 5) High transparency and uniformity render this polymer an excellent optical material. 6) The elasticity or deformability provides tunability of optical parameters such as a resonance wavelength.[8] 7) The sound velocity (980 m/s) is lower than those of any other solids or liquids, e.g., 5400 (glass) or 1500 m/s (water), which is advantageous for arranging droplets at a small interval (half wavelength).[14] 8) The sound velocity changes little during the solidification process, and hence, the ultrasonic standing wave for the droplet arrangement is not disturbed by solidification.

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A special sample cell was prepared by molding PDMS, since ordinary glass cells caused acoustic reflection at the glass-liquid boundary disturbing the standing-wave excitation. A rectangular cavity (2.5 mm square and 4 mm depth) was created in the cubic PDMS cell (6.5^3 mm^3). As Fig. 1(a) shows, three ultrasonic transducers were attached to the sides and the base of the cell. These transducers were made of lead titanate zirconate (PZT) films of 4×15 mm^2, whose oscillation frequency was 2 or 8 MHz. Glass plates and a glass prism were attached to the opposite sides and top of the cell to reflect the acoustic waves; i.e., the standing waves were excited in three directions between the transducer and the opposite glass surface. The prism realized sample observation from the cell side, as shown in Fig. 1(b).

3. Experiment

Hydrophilic solution had to be used for creating droplets in hydrophobic oil (PDMS). Small, stable droplets were obtained when the oil was mixed with methanol containing a surfactant (Merck, tween 20). Fluorescent dye (rhodamine 6G) was added to methanol for clear observation of the droplets; i.e., the droplets exhibited red color in transparent PDMS. The mixing ratio of the methanol solution to the PDMS was varied between 0.5 and 2.5 vol%. After adding the curing agent, the mixed solution was put into the sample cell. Then the sample cell was set in the apparatus shown in Fig. 1. The oscillation frequencies of the transducers were tuned suitably so that the standing waves were excited in the sample cell. The standing wave excitation was confirmed by observing

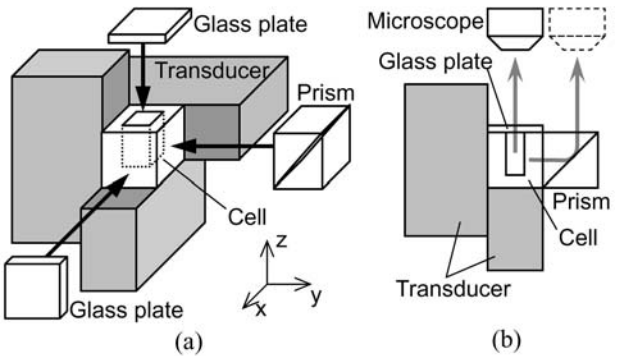

Fig. 1 (a) The structure and (b) the cross-section of the ultrasonic arrangement apparatus.

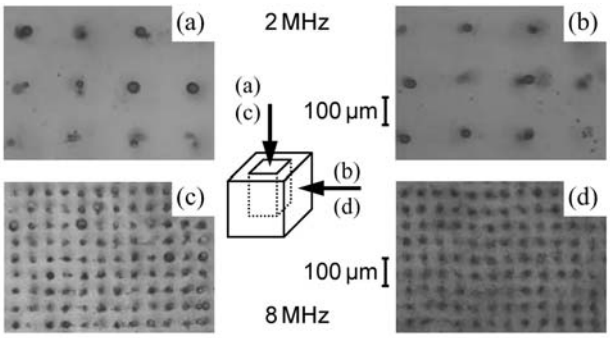

Fig. 2 Micrographs of the 3-D droplet arrays in elastomer. The ultrasonic frequency was (a), (b) 2 or (c), (d) 8 MHz. The samples were observed from (a), (c) the top or (b), (d) side.

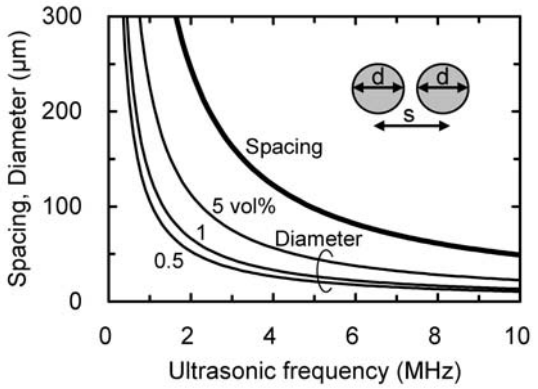

Fig. 3 Frequency dependence of the array parameters (theoretical values for PDMS). The thick line shows the droplet spacing. The thin lines show the droplet diameters for the concentrations of 0.5, 1, and 5 vol%.

droplets from both the cell top and side. The ultrasonic oscillation continued for ~2 h until the solution became viscous enough. Solidification was complete in ~8 h.

Figures 2(a) and 2(b) show photographs of a sample that was fabricated with 2-MHz ultrasonic waves. The droplet concentration was 1.5 vol%. A 3-D droplet array is visible. The droplet diameter varies between 10 and 30 μm. The spacing is ~240 μm, corresponding to the half wavelength, i.e., the wavelength of the 2-MHz ultrasonic wave is 490 μm. Figures 2(c) and 2(d) show the sample that was fabricated with 8-MHz ultrasonic waves. The droplet concentration was 0.5 vol%. The droplets of ~20 μm diameter are arranged at 60 μm spacing, i.e., half of the ultrasonic wavelength (120 μm) at this frequency.

4. Discussion

As mentioned above, the droplet spacing s is determined by the sound velocity v(=980 m/s) and the ultrasonic frequency f; i.e., $s = v/(2f)$. If the droplet diameter is d, the droplet concentration (the ratio of the droplet volume to the sample volume) is $c=(\pi d^3/6)/s^3$, and hence, $d=(6c/\pi)^{1/3}s=(6c/\pi)^{1/3}v/(2f)$. Figure 3 shows the spacing s and diameter d as functions of the ultrasonic frequency f. Both the droplet diameter and spacing decrease as the ultrasonic frequency increases. If the concentration is raised to 5 vol%, the droplet diameter will be half of the spacing.

5. Conclusion

The ultrasonic trapping method is useful to create a 3-D droplet array in a deformable PDMS resin. Unique optical functions are expected with this droplet-elastomer compound, since interactions between these droplets will affect the absorption or fluorescence characteristics of dye molecules in them.

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