Observing the Random Dispersion of Compartmentalized Droplets in an Oil-Water Emulsion in Microgravity

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***Abstract*— By injecting a droplet of oil into water, a contact surface forms between the two liquids, known as a liquid-fluid interface. The jamming of nanoparticle-ligands assemblies at this interface forms a permeable membrane. In microgravity, multiple droplets of a liquid can be suspended in another exterior liquid phase, forming a stable emulsion without coalescing of the interior droplets. The negligible force of gravity on the International Space Station (ISS) allows for the random distribution of these encapsulated droplets, whereas on Earth buoyant forces immediately affect the structures due to differences in density. On earth and the ISS, liquid-fluid interfaces were created by injecting a solution of POSS nanoparticles and silicon oil into aqueous polyacrylic acid, and their shapes were compared. In microgravity, the formation of fluid-liquid-fluid and fluid-liquid-gas structures further proved the stability of these systems in the absence of significant buoyant forces. The formation of stable, dispersed structures in liquid-fluid emulsions allows for a wide variety of applications including drug delivery, reagent encapsulation for on-demand reactive systems, and all-liquid batteries.**

I. INTRODUCTION

The contact surface between two different liquids constitutes a liquid-fluid interface. Differences in the interaction forces between the two liquids changes the interfacial potential energy as molecules are moved toward the border. Work must be done against the unbalanced interaction forces to bring the molecule to the interface. To reduce potential, liquid structures tend to minimize surface area by forming spherical domains, coalescing, or absorbing particles to the interface. In addition, surfactants, which reduce interfacial energy, are often used to stabilize smaller droplets, since the energy cost of the droplets is reduced.

Polyacrylic acid (PAA), which exhibits low binding energies and a slight negative charge, is dissolved in water and subsequently injected in a solution of silicone oil. The inherent negative charge of the water-oil interface hinders the polymer from binding to the interface; furthermore, the low-binding energy of the nanoparticles prevents the formation of an irreversible bond.

To anchor the charged PAA to the interface, positively charged POSS nanoparticles (NP) are mixed into the oil, forming NP-ligand assemblies at the interface by electrostatic attraction. The absorption of the nanoparticle-ligands at the interface, in addition to the droplet's tendency of lowering interfacial surface area, irreversibly jams the particles in place, forming a flexible, permeable structure with special chemical properties. This structure can then be modified by injecting or reducing the volume of the droplet, shining a light of a specific wavelength to weaken the membrane, or adjusting the pH level of the liquid to alter the characteristics of the structure.

II. METHODS

Experimental Setup:

A solution of PAA in water is initially injected into a sealed chamber. Silica capillary tubing connected to a bi-directional pump protrudes into the chamber, functioning as an injecttion needle. By turning on the pump, a solution of NP and silicone oil is injected into the PAA solution, forming a spherical droplet 2 millimeters in diameter. Pictures are taken between intervals.

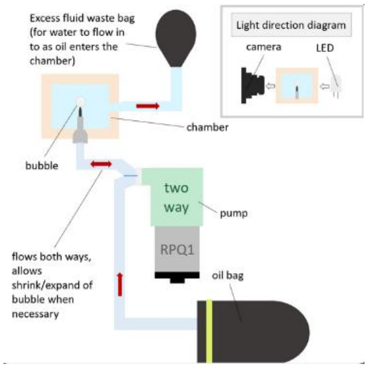

Figure 1: Experiment Block Diagram

Flight Test:

The experiment module was placed in a microgravity environment on the ISS and ran autonomously. The experiment loop consists of 7 one second injection intervals followed by 7 one second retraction intervals. Each injection interval increases by 30 milliseconds per loop, while retraction times stay constant. Photos are taken between intervals.

III. RESULTS

For the first fourteen cycles, the injected droplet can be seen oscillating on the needle, slowly increasing in size. However, after several cycles, in which we were able to observe a slight wrinkling effect of the membrane, the droplet volume continued increasing such that it adhered to the chamber by surface tension, and the unexpected formation of different structures was observed. When the needle detached from the oil droplet, it retracted PAA solution from the surrounding chamber and injected it into the larger droplet of oil. These smaller droplets dispersed into the surrounding oil, eventually stabilizing in individual locations. Coalescence was not observed, and the droplets of PAA with diameter around 0.08 mm remain in place inside the 3-layer emulsion, encapsulated inside the oil droplet even during subsequent cycles of injection. Thus, we were able to observe the formation of water-oil-water emulsions, and the stable, undisturbed distribution of all-aqueous structures separated only by a NP-surfactant membrane. Similarly, gaseous bubbles are

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injected into the oil droplet in a later phase, and the chambers of air remain encapsulated inside the oil. Some of the air bubbles do coalesce, but at a rate much slower than that on Earth, and the droplet grows as it envelopes gas. The formation of these fluid-liquid-fluid and fluid-liquid-gas structures proves the concept of stable encapsulation of both gaseous and liquid phases, which do not tend to coalesce in the surrounding liquid. The experiment demonstrates significant evidence that randomly dispersed, all-liquid structures can be stably formed microgravity due to the lack of significant buoyant forces arising from liquid density differences.

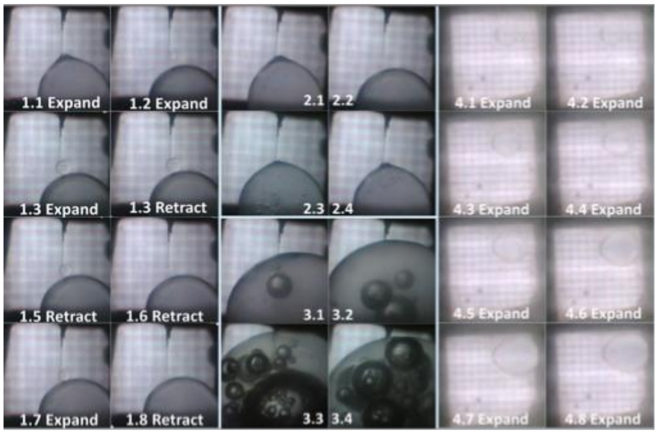

Figure 2: Pictures of the chamber from the camera.

On the ground, we were able to obtain valuable data through testing as well. Due to gravity and density differences, buoyant force causes the water droplets to accumulate at the top of the larger host droplet and individual droplets to coalesce before a membrane is stabilized from the assembly of POSS nanoparticles and PAA at the interface. As seen from our ground tests, the silicone oil droplet, denser than the aqueous PAA solution, sagged downward, falling off the injection needle regardless of chamber orientation. The formed droplet could not reach the size of the droplet in the flight experiment, and all droplets that fell coalesced into a single oil bubble, adhering to the bottom chamber panel. Thus, due to gravity, it is extremely difficult to form the compartmentalized, all-liquid systems on the ground, or observe the characteristics of interior liquid-fluid interfaces.

We were able to evaluate our hypothesis with the data collected, and used ImageJ, an open source image processing package, to identify trends, take precise measurements, compile animations, and calculate area of the droplet, in order to convert the 1271 photos collected into a visual form.

IV. CONCLUSION

Originally, we wanted to observe the effect of the reduction in surface area of the droplet on the buckling of the nanoparticle surfactant assemblies. As observed in the injection/reduction phase, there were no significant wrinkles on the droplet. However, a permeable, irregular membrane can be seen in Figure 2 images 2.-2.4, suggesting that the membrane undergoes wrinkling, yet gravity causes sagging and vertical wrinkling due to the constant downward pull. As the liquid-fluid interface minimizes its free energy by decreasing total surface area and coalescing into spherical shapes, the presence of an uneven interface indicates the existence of a semi-permeable membrane.

While we had originally expected the formation of a singular liquid-fluid-liquid emulsion, we instead observed the formation of multiple layers of encapsulated liquid-fluid interfaces (figure 2, images 3.1-3.4). On Earth, the interior droplets would concentrate at the top of the host liquid, forming gradient structures based on fluid densities. On the International Space Station, the droplets stay randomly dispersed, generating a randomly mixed droplet configuration that can only be achieved in microgravity.

Nanoparticle systems at liquid-fluid interfaces cover a vast field of technological applications, including the stabilization of dispersion devices, flotation systems, encapsulation, and pharmaceutical formulations. The stability and random, undisturbed emulsion of gas and liquid droplets encased by a nanoparticle surfactant membrane demonstrates the potential of reagent encapsulation, opening a wide window of potential applications in biomedical engineering, drug delivery, and selective diffusion and reactions. The all-liquid structures could be configured to be sensitive to a certain stimulant, which breaks down the nanoparticle monolayer weakening it to allow for the selective or partial diffusion of the enclosed liquid, or the coalescing of multiple droplets into one structure. For example, the physical and chemical properties of the liquid membrane could be altered as the surrounding solution reaches a certain pH threshold, or after being exposed to certain wavelengths of light, changing the properties of interaction between structures as well as the surrounding environment. We would recommend future microgravity experiments to explore these characteristics.

The modifiable, permeable characteristics of liquid-fluid interfaces and all-liquid structures can be applied in the cosmetics industry, with compartmentalized reagents interacting only after a certain amount of time, or in the medical industry, as a chemical transport vessel breaking down under certain conditions. Studying liquid-fluid interfaces in microgravity, where the distribution of liquids is unaffected by density, allows for the observation of new characteristics of these unique structures.

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