Nanoscale self-assembling salt-crystal 'origami' balls envelop liquids

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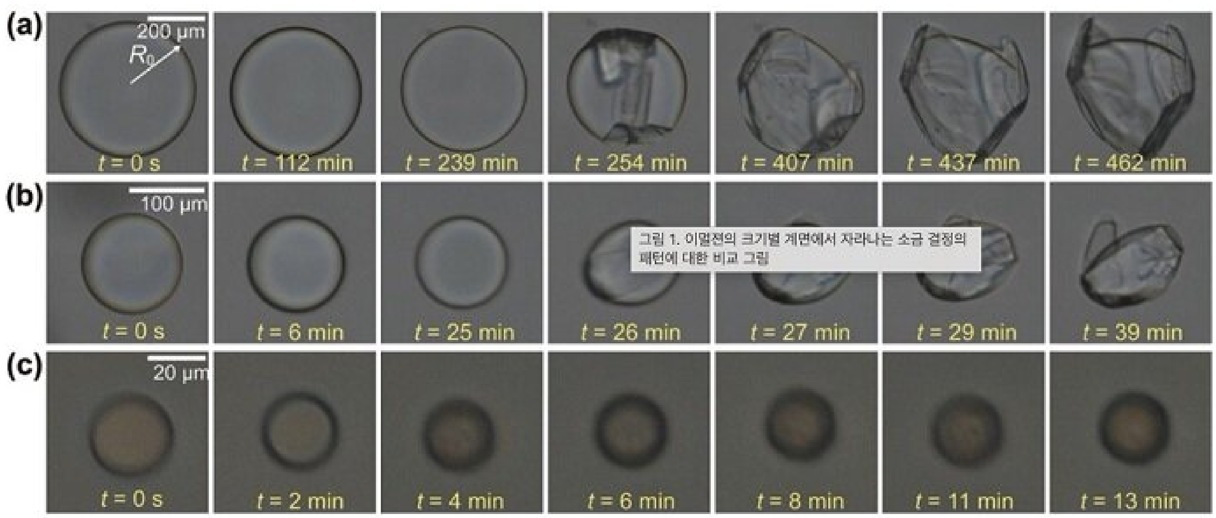

Crystal growth at the interface of calcium propionate-saturated water–oil emulsions depending on the initial emulsion size. The initial diameter of the emulsion in oil is (a) 496 µm, (b) 135 µm, and (c) 34 µm. Credit: The Korea Advanced Institute of Science and Technology (KAIST)

Researchers have developed a technique whereby they can spontaneously encapsulate microscopic droplets of water and oil emulsion in a tiny sphere made of salt crystals—sort of like a minute, self-constructing origami soccer ball filled with liquid. The process, which they are calling 'crystal capillary origami,' could be used in a range of fields from more precise drug delivery to nanoscale medical devices. The technique is described in a paper appearing in the journal *Nanoscale* on September 21.

Capillary action, or 'capillarity,' will be familiar to most people as the way that water or other liquids can move up narrow tubes or other porous materials seemingly in defiance of gravity (for example within the vascular systems of plants, or even more simply, the drawing up of paint between the hairs of a paintbrush). This effect is due to the forces of cohesion (the tendency of a liquid's molecules to stick together), which results in surface tension, and adhesion (their tendency to stick to the surface of other substances). The strength of the capillarity depends on the chemistry of the liquid, the chemistry of the porous material, and on the other forces acting on them both. For example, a liquid with lower surface tension than water would not be able to hold up a water strider insect.

Less well known is a related phenomenon, elasto-capillarity, that takes advantage of the relationship between capillarity and the elasticity of a very tiny flat sheet of a solid material. In certain circumstances, the capillary forces can overcome the elastic bending resistance of the sheet.

This relationship can be exploited to create 'capillary origami,' or three-dimensional structures. When a liquid droplet is placed on the flat sheet, the latter can spontaneously encapsulate the former due to surface tension. Capillary origami can take on other forms including wrinkling, buckling, or self-folding into other shapes. The specific geometrical shape that the 3D capillary origami structure ends up taking is determined by both the chemistry of the flat sheet and that of the liquid, and by carefully designing the shape and size of the sheet.

There is one big problem with these small devices, however. "These conventional self-assembled origami structures cannot be completely spherical and will always have discontinuous boundaries, or what you might call 'edges,' as a result of the original two-dimensional shape of the sheet," said Kwangseok Park, a lead researcher on the project. He added, "These edges could turn out to be future defects with the potential for failure in the face of increased stress." Non-spherical particles are also known to be more disadvantageous than spherical particles in terms of cellular uptake.

Professor Hyoungsoo Kim from the Department of Mechanical Engineering explained, "This is why researchers have long been on the hunt for substances that could produce a fully spherical capillary origami structure."

The authors of the study have demonstrated such an origami sphere for the first time. They showed how instead of a flat sheet, the growth of salt-crystals can perform capillary origami action in a similar manner. What they call 'crystal capillary origami' spontaneously constructs a smooth spherical shell capsule from these same surface tension effects, but now the spontaneous encapsulation of a liquid is determined by the elasto-capillary conditions of growing crystals.

Here, the term 'salt' refers to a compound of one positively charged ion and another negatively charged. Table salt, or sodium chloride, is just one example of a salt. The researchers used four other salts: calcium propionate, sodium salicylate, calcium nitrate tetrahydrate, and sodium bicarbonate to envelop a water-oil emulsion. Normally, a salt such as sodium chloride has a cubical crystal structure, but these four salts form plate-like structures as crystallites or 'grains' (the microscopic shape that forms when a crystal first starts to grow) instead. These plates then self-assemble into perfect spheres.

Using scanning electron microscopy and X-ray diffraction analysis, they investigated the mechanism of such formation and concluded that it was 'Laplace pressure' that drives the crystallite plates to cover the emulsion surface. Laplace pressure describes the pressure difference between the interior and exterior of a curved surface caused by the surface tension at the interface between the two substances, in this case between the salt water and the oil.

The researchers hope that these self-assembling nanostructures can be used for encapsulation applications in a range of sectors, from the food industry and cosmetics to drug delivery and even tiny medical devices.

More information: Kwangseok Park et al, Crystal capillary origami capsule with self-assembled nanostructures, *Nanoscale* (2021). DOI: 10.1039/d1nr02456f

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