Team devises easier way to make 'bijels,' a complex new form of liquid matter

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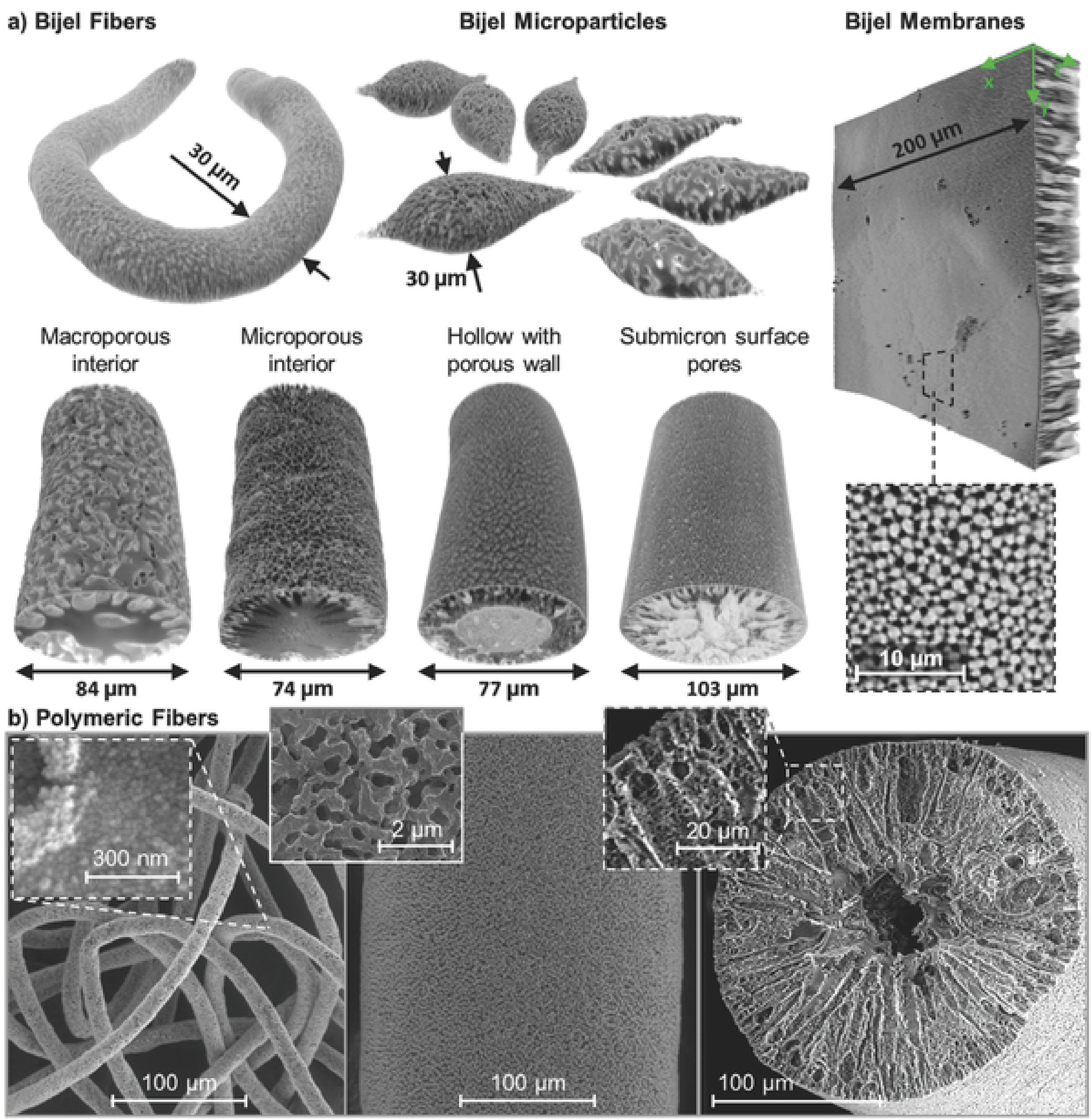

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Oil and water famously don't mix, but finely dispersing one in the other produces a liquid mixture with many useful properties. An emulsion consisting of tiny droplets of one of those liquids immersed in the other is the most common form, found in everything from salad dressings, to cosmetics to industrial lubricants.

Other, more complex emulsions are possible; getting the interfaces between the two liquids into different shapes unlocks new kinds of behaviors and applications. And thanks to new research from University of Pennsylvania School of Engineering and Applied Science, one special kind of emulsion is becoming easier to make.

Known as a bicontinuous interfacially jammed emulsion gel, or bijel, this type of emulsion is eyed as a kind of liquid conveyor belt for continuous chemical reactions. This is because, rather than isolated droplets, both the oil and water phases in these materials consist of densely intertwined but fully connected networks that other molecules can flow through. A layer of particles "jammed" at the interface between the oil and water networks prevents them from dispersing further, but could also serve as a catalyst for those reactions.

Such a complex structure has only recently been made possible through advances in soft matter physics, and has until now been limited to a small number of compatible oils. The Penn team's method for making bijels, which uses ethanol instead of precise temperature changes to drive the formation of the networks, works with a much wider array of oils, opening the door to new applications.

The method also makes it easier to control the overall shape of the bijel, enabling it to take the form of blobs, membranes or fibers. They can also control the dimensions of the internal networks, potentially fine-tuning it to a given application.

The team consists of Daeyeon Lee, an associate professor in Penn Engineering's Department of Chemical and Biomolecular Engineering, Kathleen Stebe, the Richer & Elizabeth Goodwin Professor of Chemical and Biomolecular Engineering and Martin Haase, a postdoctoral researcher who works in both of their labs.

They published their results in the journal *Advanced Materials*.

Originally devised by a team of researchers at the University of Edinburgh, bijels were first made using a process known as thermally induced spinodal decomposition. There, oil and water are forced to mix by heat, then to separate by dropping the temperature of the mixture. An injection of particles that go to the intertwined interfaces between the liquids stops that process before the oil and water separate into discrete droplets.

"The problem with this process is that it's very delicate," Stebe said. "While there are a handful of special sets of oil and water that will do the temperature trick, there are hundreds of sets that work with our method. We went from a finicky, delectate space to a workhorse space that we can use again and again."

When Hasse, an expert in three-component chemical systems and nanoparticles at interfaces, joined Penn, he, Lee and Stebe brainstormed potential research projects that involved these elements. Bijel manufacture jumped out to them in what they call an "aha" moment.

The method they devised, known as solvent transfer-induced phase separation, or STRIPS, uses ethanol instead of heat to mix and un-mix the oil and water.

In addition to the wider range of materials that is compatible with STRIPS over the heat-based method, the Penn method allows for greater control of the shape that the final bijel takes.

"Because we pull ethanol out directionally," Haase said, "the rate of bijel formation decreases from the outside to the inside of the material. That gives us structures with different length scales, such as intertwined fingers that are really fine on the surface but really open in the center. The multi-step nature of the phase separation additionally gives us nano-sized features with high surface areas. All of this helps us make what we call 'asymmetric hierarchical structure' which is helpful for size selective separation processes, amongst other applications."

With their complicated internal structures, bijels could solve a problem intrinsic to chemical reactions that take place in water and have products that are soluble in oil. So-called "emulsion microreactors" involve packing the reactants into water droplets with catalyzing particles on their surfaces, then immersing them all in an oil bath.

"The problem with this is that once you use up the reactants in your droplets, you're done," said Lee. "All the droplets are isolated, so there's no easy way to "restock" them with new reactants. That's where bijels come in. You could keep on feeding the water phase with reactants and keep on pulling out product from the oil phase. We call that a 'continuous reactor,' and they'd be very useful for things like refining biofuels."

The team's next steps involve looking for applications to which STRIPS-formed bijels are particularly suited.

More information: Martin F. Haase et al. Continuous Fabrication of Hierarchical and Asymmetric Bijel Microparticles, Fibers, and Membranes by Solvent Transfer-Induced Phase Separation (STRIPS), *Advanced Materials* (2015). DOI: 10.1002/adma.201503509

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