Catalyst support structures facilitate high-temperature fuel reforming

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The catalytic reforming of liquid fuels offers an attractive solution to supplying hydrogen to fuel cells while avoiding the safety and storage issues related to gaseous hydrogen. Existing catalytic support structures, however, tend to break down at the high temperatures needed to prevent fouling of the catalytic surface by soot.

Now, researchers at the University of Illinois at Urbana-Champaign have developed porous support materials that can withstand the rigors of high-temperature reforming of hydrocarbon fuels.

“These novel materials show great promise for the on-demand reforming of hydrocarbons such as diesel fuel into hydrogen for portable power sources,” said Paul Kenis, a professor of chemical and biomolecular engineering at Illinois and a corresponding author of a paper to appear in the August issue of the journal Advanced Functional Materials.

To be useful for hydrocarbon fuel reforming, a catalyst support must have a high surface area, be stable at high temperatures, and possess a low pressure drop.

“Our new materials satisfy all three key requirements,” said Kenis, who also is a researcher at the Beckman Institute for Advanced Science and Technology. “They have a large surface area created by a network of interconnected pores. They can operate at temperatures above 800 degrees Celsius, which prevents the formation of soot on the catalytic surfaces. And they have a low pressure drop, which means it takes less

pressure to push the fuel through the catalyst."

To fabricate the supports, the researchers begin by placing a polydimethylsiloxane (PDMS) mold onto a flat surface, forming a channel about 500 microns wide that is open at both ends. A slurry containing polystyrene spheres 50 nanometers to 10 microns in diameter is then allowed to flow into the channel from one end by capillary action.

"Once the slurry reaches the other end of the channel, the spheres begin to pack together as a result of solvent evaporation, and the packing process continues toward the inlet end," Kenis said. "After the packing process is completed, we remove any remaining solvent, which leaves a sacrificial template consisting of a bed of closely packed spheres."

Next, the researchers fill the spaces between the spheres with a low-viscosity, preceramic polymer-based liquid. After low-temperature curing, the mold is removed, leaving a stable, freestanding structure.

Lastly, the cured ceramic precursor is pyrolyzed at 1,200 degrees Celsius for two hours in an inert atmosphere. "The polystyrene spheres decompose during the pyrolysis process," Kenis said. "The end result is a silicon carbide or silicon carbonitride replica with a tailored structure of interconnected pores."

The overall size of the replica can be precisely tailored through the dimensions of the mold, Kenis said, while the pore size can be tailored independently by the size of spheres used in the sacrificial template.

To demonstrate the use of these materials as catalyst supports, the researchers coated samples of the porous structure with ruthenium. The structure was then incorporated within a stainless steel housing, where it successfully stripped hydrogen from ammonia at temperatures up to 500 degrees Celsius. In work not yet published, Kenis and his colleagues

incorporated the structure in a ceramic housing, which enabled the successful decomposition of ammonia at operating temperatures up to 1,000 degrees Celsius.

The researchers also showed that the silicon carbide and silicon carbonitride structures are stable at temperatures as high as 1,200 degrees Celsius in air, thus showing their promise to perform fuel reforming at temperatures where fouling of the catalyst by soot does not occur.

While the demonstration was performed on a microscale reformer, the material could be used for large-scale reformers, Kenis said, with improvements in the fabrication processes.

Source: University of Illinois at Urbana-Champaign