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Control of Both Particle and Pore Size in Nanoporous Palladium Alloy Powders

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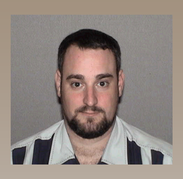

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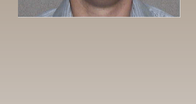
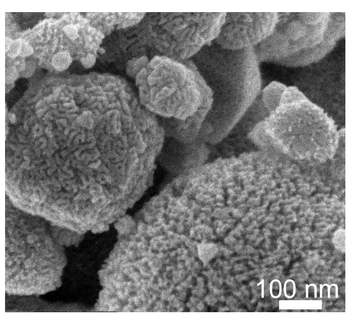


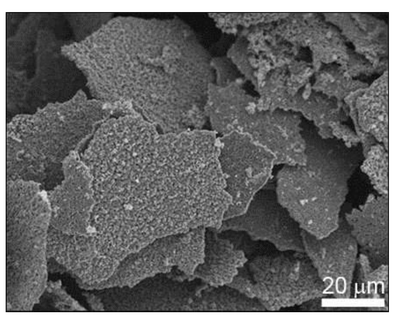
Motivation

Energy storage materials often involve chemical reactions with bulk solids or their surfaces. Porosity within the solids can enhance reaction rates. The porosity can be either within or between individual particles of the material. Surfactant templating is a known method to create well defined nanoscale pores. We seek methods to create materials that also have well defined microscale structure.

Problem

Nanoporous metals are typically prepared by chemical reduction. Particles nucleate in random locations and at random times, and grow without dimensional constraints.





The material shown above has nanopores of well defined length scale, but it forms random, uncontrolled flakes on the micrometer scale.

Proposed solution

We wish to constrain the location, and local amount, of the reducing agent so that particles nucleate there and grow to a defined size. Well defined particles of less noble metals can be easily obtained by methods without constraints on reaction conditions required for formation of nanopores. These can then be used as reductants for nanoporous noble metals.

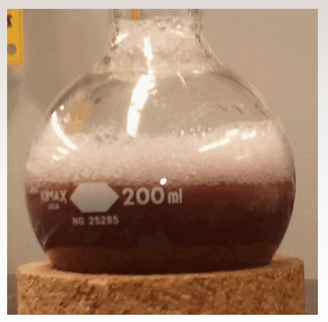
Uniform copper particles

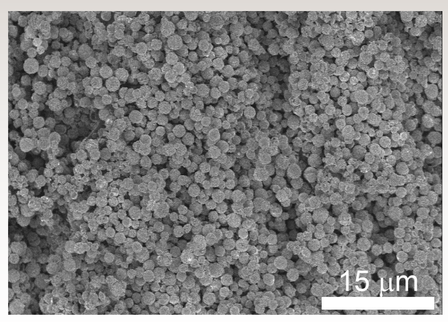
Hydrogen-storing palladium particles

Nanopores in palladium particles

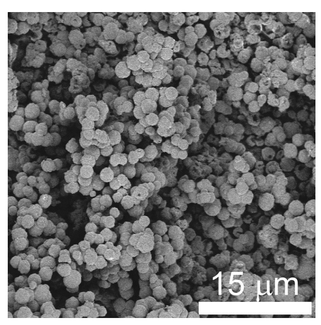
Methods

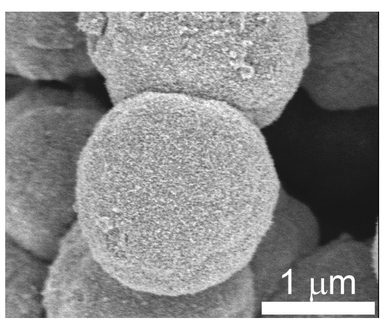
Micrometer-scale copper particles are made by reduction of copper sulfate by sodium ascorbate in the presence of a block copolymer surfactant (Pluronic F127).

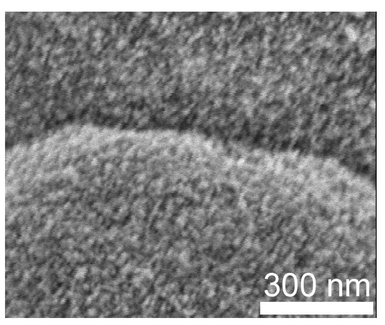




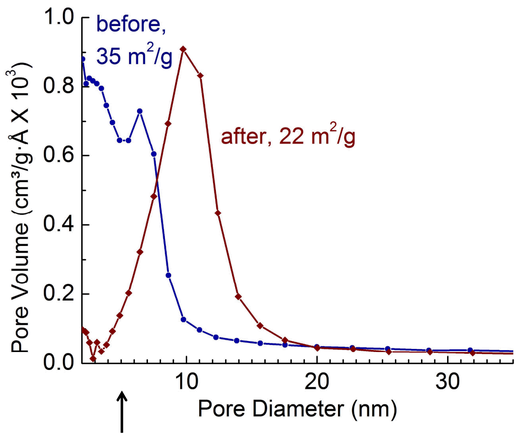
Palladium particles are made by mixing copper particles with a solution of tetrachloropalladate, excess chloride, and more surfactant. Subsequent treatment with palladium nitrate helps remove residual copper that has alloyed with the palladium.



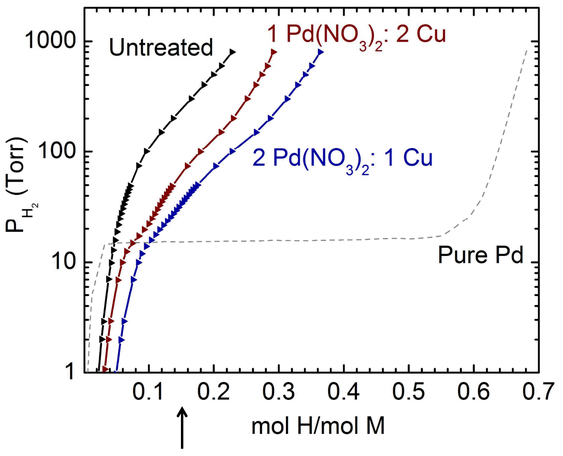




Properties



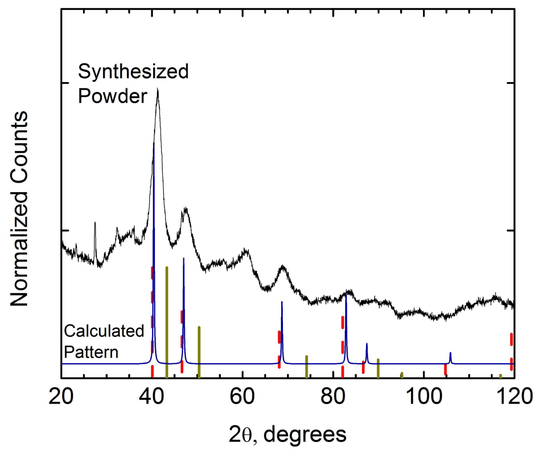
The pore size distribution from nitrogen porosimetry on a 100 mg scale sample is consistent with pore diameter and spacing on the 10 nm scale. Hydrogen treatment removes sub-10nm porosity.



Residual copper reduces the hydrogen storage capacity, but the nitrate treatment restores capacity to a useful range.

Broad peaks in powder x-ray diffraction suggest small grain size and/or Pd:Cu alloy nonuniformity, but not a mix of bulk Pd and bulk Cu.

Red dash: Pd; yellow line: Cu; blue curve: predicted 30% Cu alloy



References

This work: *Powder Technology* 267 (2014) 95-102
Prior work (flaky particles):
Cappillino et al., *J. Mater. Chem. A* 1 602 (2013)

Conclusions

Use of well defined reductant particles is an effective route to well defined nanoporous particles. Copper leads to products that are potentially useful, but a reductant that leads to purer products is desirable.

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