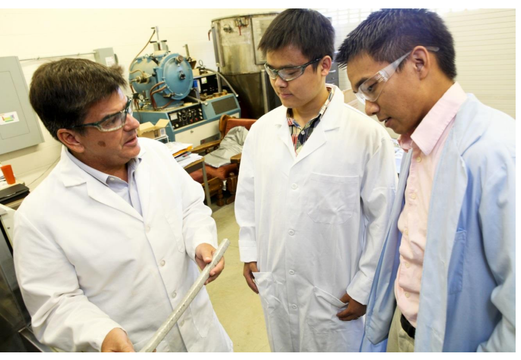


Sandia National Laboratories

Exceptional service in the national interest

SAND2014-16585D

Investigation of H Sorption Properties of Cryomilled Pd-10%Rh

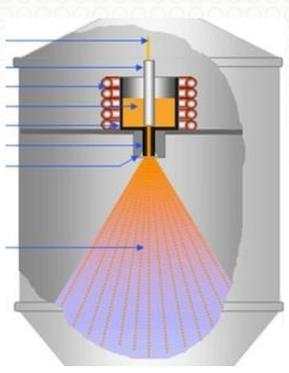
Josh Yee (University of California, Davis)

Mentors: Patrick Cappillino (8651), Chris San Marchi (8252), and Nancy Yang (8651)

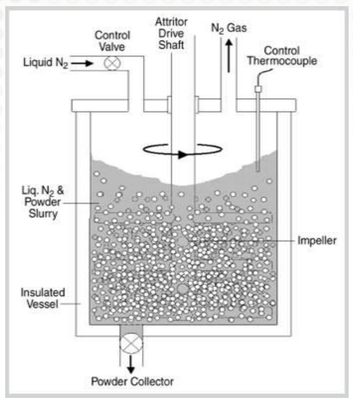
Abstract

Cryomilling was used to modify the microstructure (e.g., grain size, dislocation density) of Pd-10%Rh alloy to study the role of defects on hydrogen sorption in metals. The nanoscale characteristics of the cryomilled microstructures of the Pd-10%Rh alloy were characterized and correlated with the measured hydrogen sorption characteristics. Greater surface areas, higher dislocation densities, and smaller grain sizes resulted in greater hydrogen solubility and greater rates of hydrogen uptake.

Processing and materials



Spray atomization



Cryomilling

Final resulting powder

- High temperature spray atomization to form spherical particles
- Cryogenic milling or "cryomilling"
- Grain growth suppression due to cryogenic temperatures
- Particle morphology modification

Results

Microstructural Characterization

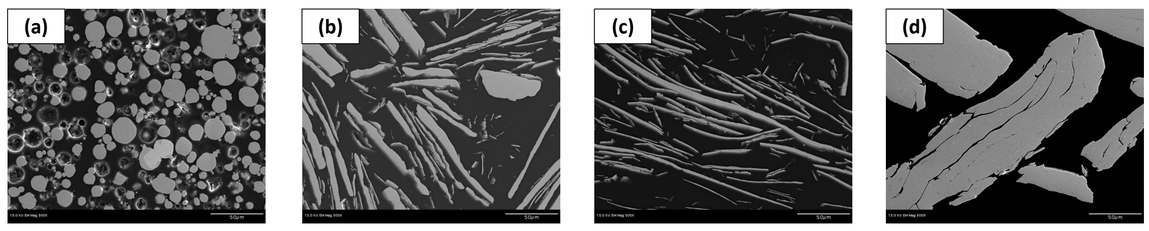


Figure 1. Cross section SEM micrographs of **(a)** atomized, **(b)** 2h, **(c)** 8h, and **(d)** 16h cryomilled Pd-10%Rh.

Sample	Surface Area (m^2/g)	Grain Size (nm)	Dislocation Density (m^{-2})
Atomized	0.037	--	1.22×10^{14}
2h CM	0.13	129.3	1.02×10^{15}
8h CM	0.44	77.03	2.35×10^{15}
16h CM	0.045	29.53	1.10×10^{15}

- Spherical particles flattened into flake-like morphology (Fig. 1)
- Surface area increases with more cryomilling; occurrence of cold welding causes decrease in 16h sample
- Increasing dislocation density and decrease to sub micron grain sizes (Fig. 2)

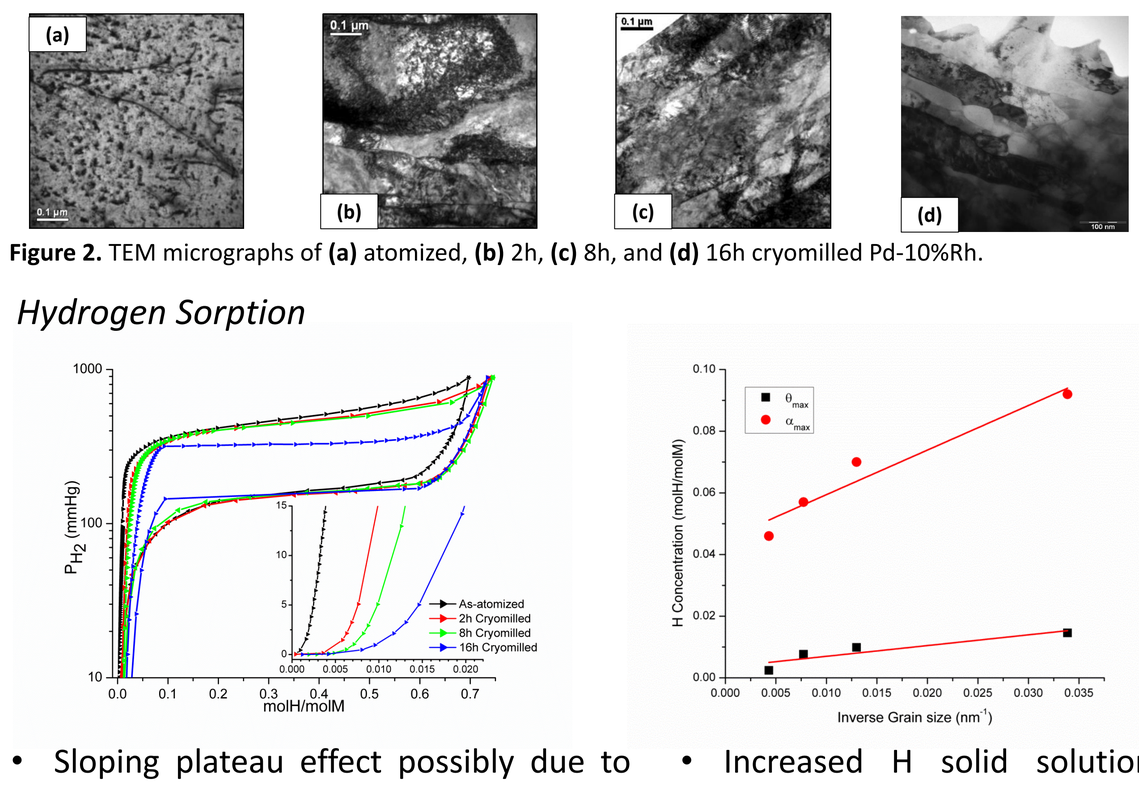
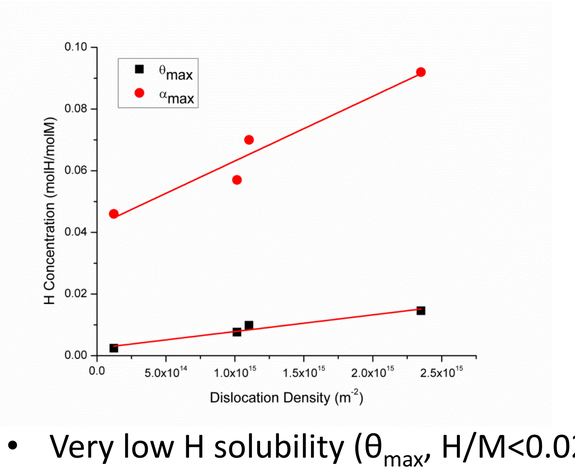


Figure 2. TEM micrographs of **(a)** atomized, **(b)** 2h, **(c)** 8h, and **(d)** 16h cryomilled Pd-10%Rh.

Hydrogen Sorption

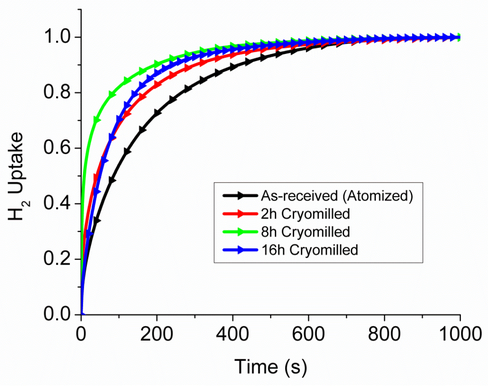
- Sloping plateau effect possibly due to inhomogeneous Rh distribution in powder
- No significant decrease in miscibility gap width observed

- Increased H solid solution solubility (α_{max}) due to higher defect concentration
- α_{max} trends in good agreement with the literature



- Very low H solubility (θ_{max}, H/M<0.02) due to surface defect concentration increase
- Reasonable to expect θ_{max} influenced by both grain size and dislocation density

Rate of Hydrogen Uptake



Sample	Absorption Half Life (s)
Atomized	88
2h CM	42
8h CM	8
16h CM	44

- Absorption half life correlates to decrease in surface area

Conclusions

- Cryomilling is an effective powder metallurgy process for modifying physical properties of Pd-10%Rh
- Microstructural changes caused by cryomilling affected H sorption properties due to the increased concentration of mechanically induced defects (e.g., dislocations, grain boundaries)

Acknowledgements

The authors wish to thank Prof. Enrique J. Lavernia (University of California, Davis) for discussions and technical input on this project. JKY gratefully acknowledges the support of the Sandia National Laboratories Campus Executive Fellowship.

Publications and Conference Presentations

Cappillino, P. J.; Stavila, V.; Yang, N.Y.; Zhang, Z.; Wolfer, W.; Yee, J.; Ong, M.; Lavernia, E., "Microstructural Evolution of Cryomilled Pd/Rh Alloy Powder and Its Correlation to Hydrogen Storage" Poster Presentation given at the 2012 International Hydrogen Conference, Jackson Hole, WY. September, 2012.

Yee, J. K.; Cappillino, P. J.; Stavila, V.; Yang, N.Y.; Lavernia, E. J., "Processing of nanostructured Pd for H storage" Oral presentation given at MS&T 12, Pittsburgh, PA. October 2012.

Yee, J. K.; Kuramanaeva, L.; Cappillino, P. J.; Stavila, V.; Yang, N.Y.; Lavernia, E. J., "Nanostructured Pd for H storage" Oral presentation given at TMS 2014, San Diego, CA. February 2014.

Submitted to Acta Materialia: Yang, N.Y.; Yee, J.K.; Zhang, Z.H.; Kurmanaeva, L.; Cappillino, P.J.; Stavila, V.; Lavernia, E.J., San Marchi, C.W. "Hydrogen sorption characteristics of nanostructured Pd-10Rh processed by cryomilling.

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.
SAND No. 2011-XXXXP

U.S. DEPARTMENT OF ENERGY

National Nuclear Security Administration