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UCRL-TR-208096

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L. N. Dinh, M. A. Schildbach, R. S. Maxwell, B. Balazs, W. McLean II

November 18, 2004

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This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

DPB progress report:
Hydrogen uptake capacity for a particular sample of DPB

L. N. Dinh, M. A. Schildbach, R. S. Maxwell, B. Balazs, W. McLean II

Chemistry & Materials Science Department
Lawrence Livermore National Laboratory

Experiments:

We have performed hydrogen uptake to measure the remaining capacity for two DPB pellet samples. The measurements were made with a Cahn microbalance operating at room temperature. The DPB samples were placed inside a quartz boat and the microbalance was then evacuated to a base pressure of 10^{-4} Torr overnight. This overnight pumping removed volatile contaminants from the DPB pellets prior to the introduction of 600 Torr of N_2 into the microbalance chamber. Next, a leak valve connecting the microbalance with a research grade source of H_2 was opened to add 100 Torr of H_2 on top of the 600 Torr of N_2 already in the microbalance. When the total pressure in the microbalance reached 700 Torr, the H_2 leak valve was not completely shut off but reduced to a very small positive rate. An automatic gate valve connecting the microbalance to a turbo molecular pump was programmed by a controller to keep the total pressure in the microbalance at 700 ± 3 Torr. The balance between H_2 consumption due to DPB uptake and H_2 leak rate into the microbalance was such that the gate valve to the turbo molecular pump was only slightly opened by the controller once in a while (a few seconds per few hours) to keep the total pressure constant at 700 Torr.

Results:

Fig. 1(a) and (b) show the weight gain of sample 1 and sample 2 as a function of time in 600 Torr N_2 + 100 Torr H_2. The curve in each plot is drawn to help the readers to easily follow the trend of the noisy data. After ~ 6 days of hydrogen exposure, the masses of both samples 1 and 2 leveled off and started to decrease due to the much higher volatility of heavily hydrogenated DPB. At this point, the samples were removed from the microbalance and ground into powder in ambient air. The powder was then reintroduced to the microbalance for further hydrogen uptake at 600 Torr N_2 + 100 Torr H_2. The weight gain curves obtained from the powder samples were normalized to account for material loss during the grinding procedure. The weight gain curve of sample 2 in Fig. 1(b) is smoother than that of sample 1 in Fig. 1(a). The reason for this difference in behavior between sample 1 and sample 2 is not understood at the moment, but should be resolved with additional experience with both the microbalance and DPB.

The remaining capacity for hydrogen uptake can be approximated from Fig. 1 by eq. (1):

$$\%\ of\ remaining\ capacity \approx 100 \times \frac{(final\ wt. - initial\ wt.)/(0.75 \times initial\ wt).}{(8/202)} \qquad (1)$$

In the above formula, 202 is the molecular weight (m.w.) of DPB. The m.w. of fully hydrogenated DPB is 210. The 0.75 factor used in the above formula is employed to

account for the fact that the DPB pellets were actually composed of 75% DPB and 25% Pd-C. An implicit but reasonable approximation used in the above formula is that the amount of weight gain due to hydrogenation that took place prior to the hydrogen microbalance uptake experiment described here was much less than the weight of the virgin DPB pellet. This results in a maximum underestimation error of $100 - \frac{8/210}{8/202} \times 100 = 3.8\%$ in the worst case scenario. Using eq. (1), the remaining hydrogen uptake capacity for sample 1 is estimated to be ~ 74% while that of sample 2 is ~ 100%. The error associated with these results estimated from the noisiness of the data is ~ 15% for sample 1 and only a few percent for sample 2 if the noisiest portions of the weight gain curves corresponding to hydrogen uptake into ground heavily hydrogenated powder are ignored. The average of the results from sample 1 and sample 2 corresponds to an estimated remaining hydrogen uptake capacity in the range of 80% to more than 90%.

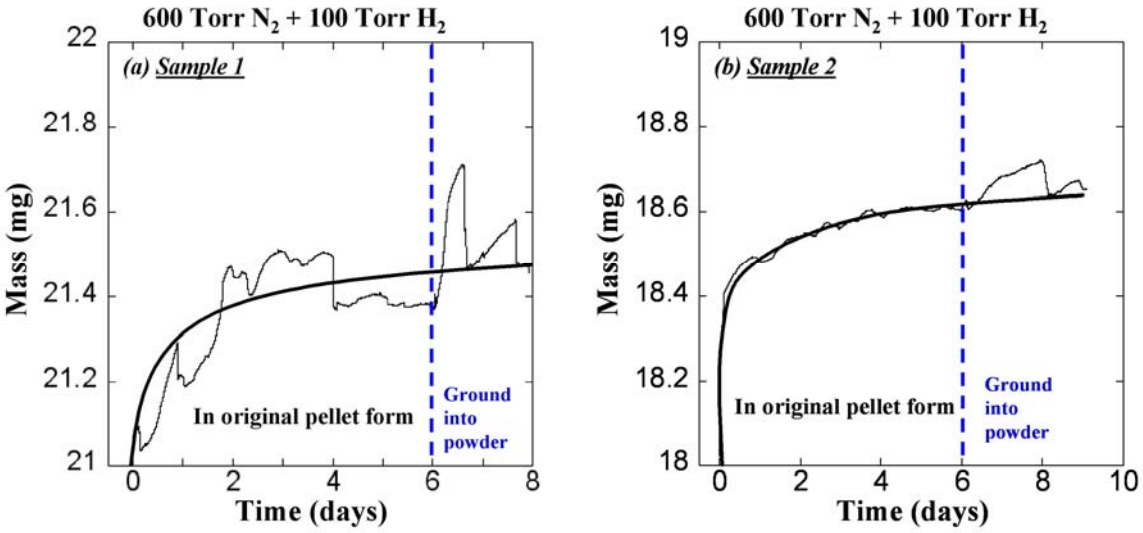

Fig. 1

Discussions:

1. We observed that upon hydrogenation, DPB pellets became much more powdery and readily fragmented at/near the surface. This also suggests that the hydrogenation proceeded from the surface inward. This explains why experiments on powder and flakes originating from the same batch of DPB pellets showed much lower remaining capacity for hydrogen uptake (50 to 67%).
2. We performed the hydrogen uptake at 100 Torr of H_2 to reduce the experiment duration to fewer than 10 days. In order to obtain the rate limiting mechanism and associated kinetics for the hydrogen uptake, we need to perform additional experiments with different H_2 pressures at a few different temperatures of interest. Reasonable extrapolation of the uptake rates to relevant pressures and temperatures will then be possible. We have purchased the necessary gas mixtures and will begin the kinetic measurements soon.

3. We have only recently acquired the Cahn microbalance used in these experiments and believe we can fine-tune it for better performance with time. At the moment, we are not sure what caused the roughness on the hydrogen uptake curve in Fig. 1. This kind of noise is not seen with other materials running in air or under vacuum. The roughness in the hydrogen uptake curve for DPB in 100 Torr of H_2 might have resulted from the volatility of heavily hydrogenated DPB. We will have an update on this issue in a future report.

Future work:

We plan to identify the volatile species and measure their associated equilibrium vapor pressures in virgin and hydrogenated DPB by mass spectrometry in FY2004-2005. Isothermal hydrogen uptake experiments at different temperatures and H_2 partial pressures, which are necessary to develop hydrogen uptake kinetics in DPB, are being carried out in our laboratory and will continue in the coming years.