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Some aging aspects of DEB getters

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Disclaimer

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ABSTRACT:

The changes in uptake capacity of 1,4-bis(phenylethynyl)benzene (DEB) blended with carbon-supported Pd (DEB-Pd/C) in the form of pellets as a function of temperature and time were investigated. Experimental results revealed a segregation and crystallization of DEB molecules toward the geometrical surfaces of the getter pellets, but very little or no diffusion-aggregation of Pd nano-catalysts even after long term storage at 75 °C in nitrogen. Despite the observation of surface segregation and crystallization of DEB molecules with increasing temperature and time, statistically there was no reduction in uptake capacity for the getter pellets stored at higher temperature. However, significant reversible reductions in uptake capacity was found among getter pellets exposed to air for extended time. The possible causes for these observations and their respective roles in the aging of getters are discussed.

INTRODUCTION:

Organic getters such as 1,4-bis(phenylethynyl)benzene (DEB) blended with a carbon supported palladium catalyst have a rapid and irreversible hydrogen uptake capability as well as a substantial hydrogen uptake capacity. The proper use of such organic getters can prevent hydrogen gas accumulation in sealed containers, which may lead to hydrogen corrosion of nuclear materials, undesirable effects on electronic components, or an explosion hazard [1-3]. There are a number of published studies on the kinetics of the gettering or scavenging of hydrogen by organic based getters [4-8]. However, there is a lack of information in the current literature on the aging of such organic based getters and the associated negative effect on the hydrogen uptake capacity.

The intention of this study was to consider a number of possible aging mechanisms of DEB getters concentrating on:

- Surface segregation of the organic DEB
- Agglomeration of palladium particles
- Poisoning of the palladium catalyst

These phenomena were to be studied via accelerated aging of DEB blended with carbon-supported Pd (DEB-Pd/C) in the form of pellets and was carried out by storing batches of getter pellets at different temperatures and under different storage conditions for extended time. Every few months, some pellets were removed from storage for observation and hydrogen uptake measurements. High resolution scanning electron microscopy (HRSEM) and transmission electron microscopy (TEM) techniques were used to probe for any sign of agglomeration of the Pd nano-catalysts in a possible diffusion-aggregation process resulting from higher temperature storage. The uptake capacities of getter pellets stored at room temperature in laboratory atmosphere for extended amounts of time were also compared with those which had undergone a short elevated heating under dynamic vacuum prior to uptake measurements. Finally, the potential causes for the differences in uptake capacities measured for the getter pellets stored under different environments and temperatures are discussed in relation to getter aging.

EXPERIMENTS:

Getter Materials

Most of the work was done with DEB pellets manufactured by Kansas City Plant, Honeywell Federal Manufacturing & Technologies. These DEB pellets were obtained from Kansas City Plant around 2005 and stored at room temperature (~21 °C) in air until use in the experiments reported here. The chemical structure of a virgin (or unhydrogenated) DEB molecule is shown at the top of the cartoon in Fig. 1 while that of a fully hydrogenated DEB molecule is at the bottom of the same cartoon. These pressed DEB pellets have a composition of 75 wt.% DEB and 25 wt.% catalyst (5 wt.% Pd on activated carbon). The role of the nano particulate Pd catalyst is to disassociate molecular hydrogen into atomic hydrogen. This atomic hydrogen is then able to add across the unsaturated carbon-carbon bonds in the DEB molecule, and its intermediates until fully hydrogenated, irreversibly storing the hydrogen in an inert form. DEB itself will not react directly with molecular hydrogen. The activated carbon support has a very high specific surface area (1100 m^2/g) and serves to disperse the Pd catalyst throughout the DEB pellet structure. The DEB pellets are right circular cylinders with a diameter of approximately 2.85 mm and a height of 3.16 mm. Each pellet weights roughly 20.4 mg for a density of about 1.01 g/cm^3.

Oven aging experiment & hydrogen uptake measurement:

After 7 years of storage under laboratory atmospheric conditions and ambient temperature, DEB pellets were then packed in N_2-filled vacuum containers and stored in ovens set at 45 °C, 60 °C, and 75 °C. Every few months, some of the pellets were removed from storage for visual observation, optical imaging, and hydrogen uptake experiments.

The experimental setup to measure hydrogen uptake from the getter pellets is presented in Fig. 2. The setup consists of a ~ 600 cubic centimeter (cc) stainless steel vacuum chamber with copper-flange ports. The chamber is connected to a turbo molecular vacuum pump system through a right angle valve and to a research grade hydrogen lecture bottle, regulator set to 5 psi(g), through a high accuracy MKS mass flow controller (M-200S) capable of 0.002 standard cubic centimeter per minute (sccm) to 0.2 sccm constant input. The chamber is equipped with MKS Baratron (capacitance manometer) gauges and a K type thermocouple. For a typical hydrogen uptake experiment, 20 DEB pellets were counted, weighed on a microbalance, transferred to a glass boat, and then loaded inside the chamber which was evacuated to the high vacuum level (1.3×10^{-4} Pa) for a few hours prior to hydrogen input by the mass flow controller. During all hydrogen uptake experiments, the chamber was kept at 40 °C by external heating with a silicone heat tape connected to a temperature controller. In some experiments, virgin DEB getter pellets, which had been exposed to laboratory air at room temperature for years, were heated under dynamic vacuum to 75 °C for 2 days prior to hydrogen uptake at 40 °C. The cartoon on the right side of Fig. 2 illustrates the kind of information which could be obtained from this type of constant input experiment. When there was no getter in the chamber, such a constant input experiment (or blank experiment) yields a linear relationship between the pressure rise in the chamber and elapsed time. When there were getter pellets in the chamber, due to the of hydrogen uptake of the getter pellets, the same constant input experiment yields a much lower initial pressure rise in the chamber. However, as soon as the getter pellets run out of capacity, they no longer uptake hydrogen and the rate of pressure rise in the chamber from that moment on became the same as that obtained from the blank experiment. The difference between the linear pressure rise vs. time curve obtained from the blank experiment and the corresponding curve obtained from the experiment with getter pellets represents the actual hydrogen uptake by the getters. The moles of hydrogen absorbed by the getter can be calculated from the ideal gas law ($PV = n RT$). The uptake capacity of the getter pellets can then be deduced and compared with maximum theoretical uptake capacity of DEB getter (which is 4 H_2 molecules per DEB molecule).

SEM and TEM investigations:

For SEM, the sample preparation involved crushing and dusting on carbon padded SEM stubs while the measurement was with a Zeiss ultra 55 field emission electron microscope equipped with in-lens secondary electron and backscattered detectors. For TEM, the samples were ground between two glass slides and dusted onto a holey carbon coated Cu TEM grid. The samples were examined in a Tecnai F20 transmission electron microscope using the following instrumental conditions: 200 kV voltage, 30μm and

50μm C2 aperture, bright field mode. High angle annular dark field (HAADF) method and analytical method of energy dispersive X-ray spectroscopy (EDX) were also employed. All SEM and TEM works were performed by Johnson Matthey Co. in Sonning Common, UK.

RESULTS & DISCUSSION:

An optical picture and a series of three SEM images in the order of increasing resolution of a typical virgin DEB pellet are presented in Fig. 3 (a), (b), (c), and (d), respectively. Both virgin and fully hydrogenated DEB crystallites have a white color. The dark color of the DEB pellet, as seen in Fig. 3, is simply the result of the presence of activated carbon in the catalyst. The pressed DEB getter pellet's morphology as observed from Fig. 3 is somewhat porous (as expected for pressed material) and thus provides easier pathways for gas diffusion.

In Fig. 4, optical images of DEB getter pellets after 7 months and 12 months of oven aging are presented. The getter pellets stored at 75 °C clearly look much lighter grey than the remaining samples displayed in Fig. 4. The lightening in color between pellets kept at 45 °C and 60 °C is not easily observable in the optical images presented in Fig. 4. However, when observed with the naked eye, all pellets lighten up in color upon storage in nitrogen-filled containers at elevated temperatures in comparison with those kept at room temperature. Since the only material in the virgin DEB getter pellet that is white is virgin DEB molecules, the lightening observed on the oven aged pellets corresponds to the amount of DEB molecules diffusing to the surfaces of the pellets. In Fig. 5, higher magnification microscope images of the pellets stored at room temperature, 45 °C, 60 °C, and 75 °C after 12 months of aging are presented. It possible to suggest that the DEB molecules not only diffused to the surface of the pellets, they also formed larger crystallites at higher temperature. After 12 months of aging, the differences in crystallite sizes look subtle between 45 °C and 60 °C but clearly more pronounced at 75 °C. After 20.5 months of aging, the lightening, or equivalent density of DEB crystallites, on the surfaces of DEB pellets visibly increases when compared with those after 12 months of aging. However, the crystal size does not appear to have increased further. Note that there was evidence of DEB crystallites on the pellets stored at room temperature as well. This is expected since the blending of 75 wt.% of DEB with 25 wt.% catalyst (5 wt.% Pd on activated carbon) should leave some DEB on the surface of the pellets.

It has been reported that, experimentally, after hydrogen molecules are disassociated into hydrogen atoms at catalyst sites, these hydrogen atoms can diffuse away from catalyst sites on graphitic carbon substrates over a distance of many micrometers and still remain as atomic hydrogen [9]. The maximum path length during which hydrogen atoms can diffuse on activated carbon or through hydrogenated DEB without recombination into H_2 molecules is currently not known. However, in general, the further the DEB molecules are from Pd catalyst sites, the less chance they have in reacting with hydrogen. A practical estimation on the effect of DEB surface segregation on the uptake capacity of the getter pellets can be based on actual hydrogen uptake experiments to be presented later in this report.

Three batches of DEB getter from Kansas City Plant have been supplied to AWE over the years, one batch in 1992, one in 1998 and most recently in 2008. A comparison of the hydrogen uptake of these materials demonstrated a drop off in the concentrations of hydrogen absorbed with age. The 1992, 1998, and 2008 materials achieved approximately 60%, 65%, and >90% uptakes, respectively. High resolution transition electron micrographs (HRTEM) of these materials allowed for a determination of the palladium particle size to be made (Fig 6). The DEB getter supplied in 1992 was found to have a significant number of larger palladium particles, while the remainder had an average particle size of 4.41 nm. By comparison the 1998 and 2008 DEB getters displayed none of the large palladium particles found in the 1992 material and had average palladium particle sizes of 2.85 nm and 4.04 nm respectively. The palladium particle size at manufacture was unknown and therefore it was not possible to determine if the resultant Pd particle sizes recorded were a result of palladium particle agglomeration or a manufacturing artifact. However, it was believed that these larger palladium particles were potentially responsible for the drop off in reactivity observed.

This diffusion-aggregation of catalysts may bring about a drop in the hydrogen uptake capacity and is illustrated in the cartoon shown in Fig. 7. In Fig. 8, HRTEM images of Pd distributions in virgin DEB pellets kept at room temperature and at 75 °C are shown. The darker nanometer-scale spots are Pd sites which are distributed from a few nanometers to less than 100 nm away from each other. To further investigate the feasibility of this aging mechanism, HRTEM images of Pd sites and size distributions similar to those seen in Fig. 8 were obtained for more regions from the different DEB pellets stored at different temperatures. The statistics on Pd size distributions were extracted and are presented in Fig. 9 for a virgin DEB pellet sample kept at 21 °C and 2 virgin DEB pellet samples stored at 75 °C for one year. Taking into account large sample

to sample variation and limited statistics, the mean Pd size and the major parts of the distributions (2 nm to 9 nm range) are not that different for the samples under investigation (see the statistics summarized in table I). Therefore, a temperature increase from room temperature up to 75 °C does not appear to have a significant effect on the average Pd particle size. With an average particle size of
~ 3.5 nm, the average weight of each Pd nano-catalyst is $\sim 8.6 \times 10^{-23}$ kg. The weight of a DEB molecule is $\sim 4.6 \times 10^{-25}$ kg. So, the mobility of Pd nano-catalysts is supposed to be ~200 times slower than that of DEB molecule. In other words, the activation energy barrier for the diffusion-aggregation of Pd nano-catalysts should be much greater than that for the surface segregation of DEB molecules. As a result, the surface segregation effect of DEB molecules should be much more dominant than the diffusion aggregation of Pd nano-catalysts in the aging of DEB pellets.

The plots of hydrogen uptake vs. time for experiments carried out at 40 °C are presented in Fig. 10 (a) and (b) for 20 virgin DEB pellets stored at 45 °C, 60 °C, and 75 °C in dry nitrogen for 127 days of aging prior to the uptake experiment. The rate of hydrogen input during the uptake experiments was 0.095 sccm. The uptake rates for all three sets of data up to 75% theoretical uptake capacity were pretty much the same as the H_2 input rate. Thereafter, the uptake rates for all three samples dropped toward zero and the differences in the uptake rates for the individual samples became clearer as can be seen in Fig. 10 (b) which is a zoomed portion of Fig. 10 (a) between 65% and 95% theoretical uptake capacity. However, the differences in uptake rates near uptake saturation as well as the ultimate uptake capacities varied pretty much randomly, with storage temperature, from one set of experiments to the next. The experimentally measured uptake capacities for virgin DEB pellets kept at room temperature in air, and the 45 °C, 60 °C, and 75 °C virgin DEB pellets kept in nitrogen filled containers are shown in Fig 11. Surprisingly, there is no measureable decrease in the total uptake capacity for the pellets kept at higher temperature, albeit the scatter in the uptake measurement is rather large at each experimental condition. The random differences in the uptake rates after reaching 75% getter capacity, as well as the ultimate uptake capacities among the pellets stored under different conditions are probably the result of some slight level of non-uniformity in the mixing of the pellets' constituents during synthesis at Kansas City Plant. This slight non-uniform mixing can take the form of non-homogeneous distribution of Pd catalysts and/or deviation from the nominal composition of 75 wt.% DEB and 25 wt.% catalyst (5 wt.% Pd on activated carbon). Overall, the above results indicate that the visible DEB surface segregation for samples stored at higher temperature does not negatively impact the uptake capacity of the DEB getter pellets. Such an observed phenomenon implies that atomic hydrogen, after being disassociated from molecular hydrogen at Pd surfaces, can diffuse away from Pd catalyst sites over distances of many micrometers or more on activated carbon surfaces and even through micrometer thick hydrogenated DEB crystallite regions to reach the DEB getter molecules that have segregated to the pellets' surfaces.

It is suggested that the surfaces of Pd catalysts inside porous DEB pellets stored at room temperature in containers filled with laboratory air may be poisoned by CO_2, CO, and other contaminants. As a result, the dirty Pd catalyst surface loses its effectiveness [10-14] and the overall uptake capacity suffers. In that sense, the Pd catalysts in the aged getter pellets stored at higher temperature in nitrogen filled containers might have a better chance to desorb some contaminants and therefore have greater efficiency than those in getter pellets stored at room temperature in laboratory air. In order to test this theory, a set of experiments involving virgin DEB pellets stored at room temperature for over 8 years has been performed in the following way. In three of the experiments, the virgin DEB pellets were simply taken out of the air-filled storage container at room temperature and transferred to the hydrogen uptake chamber where the pellets were pumped out by a turbo pump (10^{-4} Pa) for a day prior to hydrogen exposure at 40 °C. In the other three experiments, after being taken out from the storage container, the virgin DEB pellets were heated under dynamic vacuum at 75 °C for 48 hours, then cooled down to 40 °C prior to hydrogen exposure. The results from this set of experiments are presented in Fig. 12. Clearly, all the samples that have been vacuum-heated at 75 °C for 48 hours showed superior total uptake capacities, confirming the correlation between cleaner Pd surfaces (through desorption of surface contaminant gases) and higher ultimate uptake capacities. Mass-spectra of the desorbed species from DEB getter pellets at room temperature and 75 °C (after many hours of vacuum pumping at room temperature) are shown, Fig 13. Masses 2 (H_2) and 4 (He) were solely a residue from the mass spectrometer chamber in use for this experiment. Mass 2 (H_2) was partially from the mass spectrometer vacuum background and partially from the breaking up of H_2O by the ionizer in the mass spectrometer. The gaseous desorption is composed of mass 18 (H_2O), mass 28 (CO and/or N_2), trace amount of mass 32 (O_2), and mass 44 (CO_2). The majority of CO_2, which could also originate from the reaction of desorbed CO and O molecules from Pd surfaces, was greatly removed at 75 °C, in qualitative agreement with a previous report [14]. Hence, it can be postulated with a reasonable degree of certainty that absorption of gaseous contaminants from laboratory air do have a negative effect on hydrogen uptake when present at sufficient quantities on the Pd surfaces [10-14]. Fortunately, such a poisoning effect from air exposure is reversible with vacuum heating.

SUMMARY & DISCUSSION

The changes in uptake capacity of DEB getter pellets were investigated as a function of storage temperature. There was not any observable agglomeration of Pd nano-catalyst particles even at 75 °C after 12 months of aging. However, the surface segregation followed by crystallization of DEB getter molecules is clearly observed after only a few months of aging. The DEB crystallite sizes at the surfaces of the pellets seem to be observably larger with higher storage temperature. Interestingly, within the scatter of results of hydrogen uptake measurements, there is, statistically, no reduction of uptake capacity with increasing level of DEB surface segregation and crystallization. This is probably a result of inhomogeneous mixing of the constituents in the DEB getter pellets. The results of the experiments reported here are consistent with the hypothesis that hydrogen atoms can diffuse large distances away from Pd catalyst sites on activated carbon surfaces and have the ability to diffuse through micrometer thick hydrogenated DEB regions without recombination to reach the unreacted DEB getter molecules that have segregated to the pellets' surfaces. However, gaseous contamination of the surfaces of Pd nano-particles overtime by storage in laboratory air has been confirmed to detrimentally reduce the uptake capacities. As a result of this report, long-term storage of DEB getter pellets is recommended to be carried out in an inert gas atmosphere and the getter pellets should also undergo a vacuum-heat treatment (for example at 75 °C) so as to remove gaseous contaminants such as CO_2, O_2, CO, and H_2O from the Pd surfaces prior to use.

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FIGURE CAPTIONS:

Fig. 1: The chemical structures of virgin and fully hydrogenated DEB.

Fig. 2: The experimental setup to measure hydrogen uptake from the getter pellets (left photo) and the type of information that can be obtained from such a setup (right cartoon).

Fig. 3: An optical picture (a) and a series of three SEM images (b, c, and d) in the order of increasing resolution of a typical virgin DEB pellet.

Fig. 4: Optical images of DEB getter pellets at 7 months and 12 months of oven aging.

Fig. 5: Higher magnification microscope images of the pellets stored at room temperature, 45 °C, 60 °C, and 75 °C after 12 months of aging.

Fig. 6: HRTEM images of Pd distributions for 1992, 1998 and 2008 DEB getters.

Fig. 7: An illustration of the possible diffusion-aggregation of catalysts in DEB getter pellets.

Fig 8: HRTEM images of Pd distributions in virgin DEB pellets kept at room temperature and at 75 °C.

Fig. 9: Statistical size distributions for a virgin DEB pellet sample kept at 21 °C and 2 virgin DEB pellet samples stored at 75 °C for one year.

Table I: Statistical comparisons on the size distributions for a virgin DEB pellet sample kept at 21 °C and 2 virgin DEB pellet samples stored at 75 °C for one year.

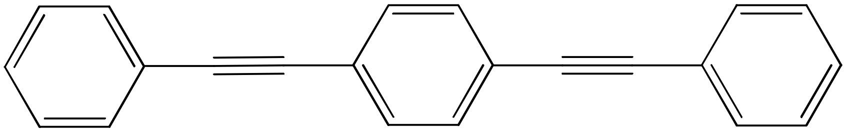
Fig. 10: Plots of hydrogen uptake vs. time for uptake experiments carried out at 40 °C for 20 virgin DEB pellets previously stored at 45 °C, 60 °C, and 75 °C in dry nitrogen for 127 days prior to the experiment.

Fig. 11: A plot of the experimentally measured uptake capacity vs. temperature for virgin DEB pellets kept at room temperature in air, and the 45 °C, 60 °C, and 75 °C virgin DEB pellets kept in nitrogen filled containers.

Fig. 12: A comparison of the uptake capacities for virgin DEB getters exposed to air for many years without a 75 °C vacuum heat treatment (the first three columns) and with a 2 day vacuum heat treatment at 75 °C (the last three columns).

Fig. 13: Mass-spectra of desorbed species from DEB getter pellets at room temperature and of those heated to 75 °C (after many hours of vacuum pump at room temperature).

Virgin DEB



Fully hydrogenated DEB

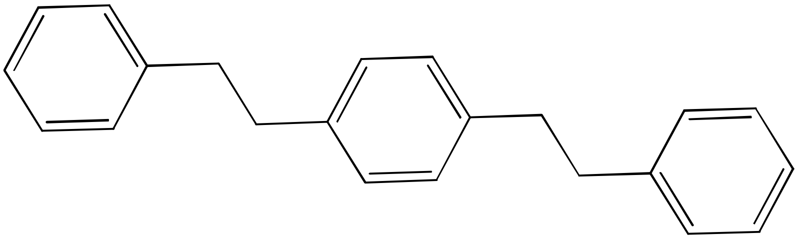


Fig. 1

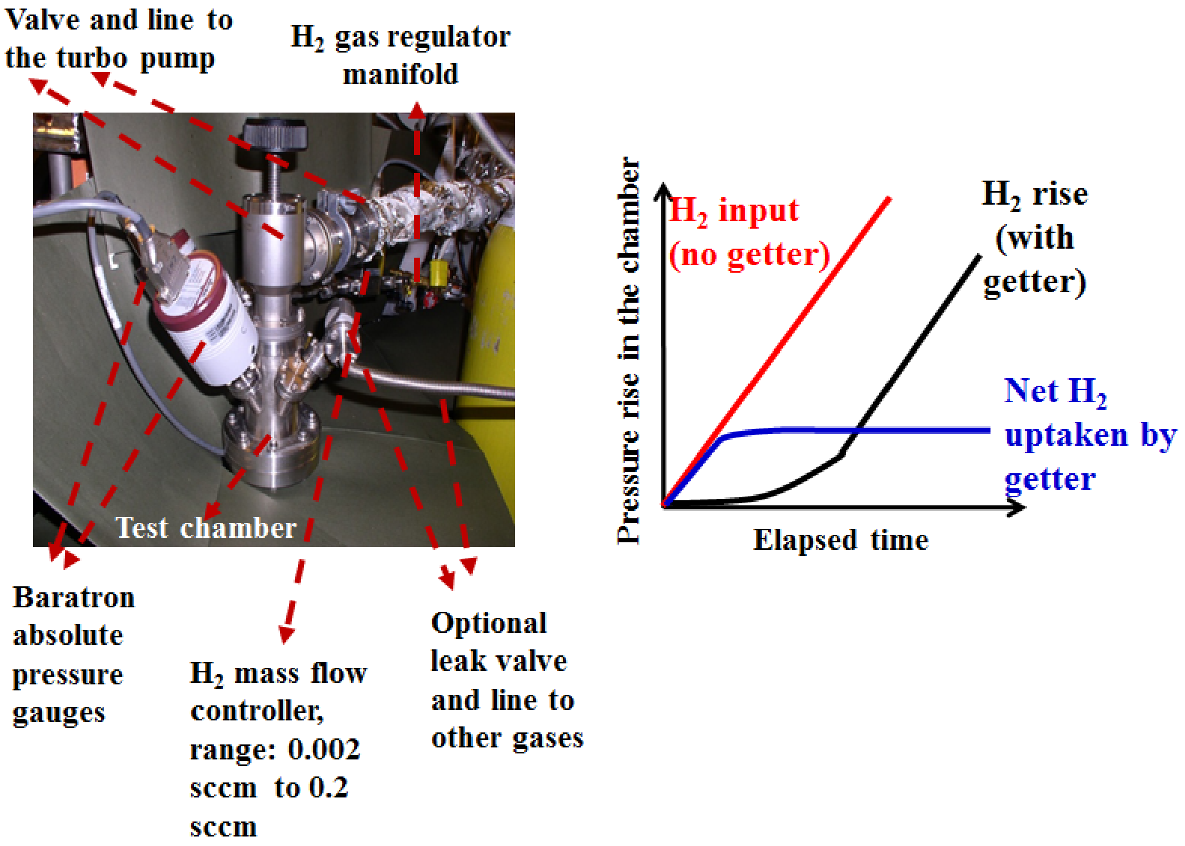

Valve and line to the turbo pump
H_2 gas regulator manifold
Test chamber
Baratron absolute pressure gauges
H_2 mass flow controller, range: 0.002 sccm to 0.2 sccm
Optional leak valve and line to other gases
H_2 input (no getter)
H_2 rise (with getter)
Net H_2 uptaken by getter
Pressure rise in the chamber
Elapsed time

Fig. 2

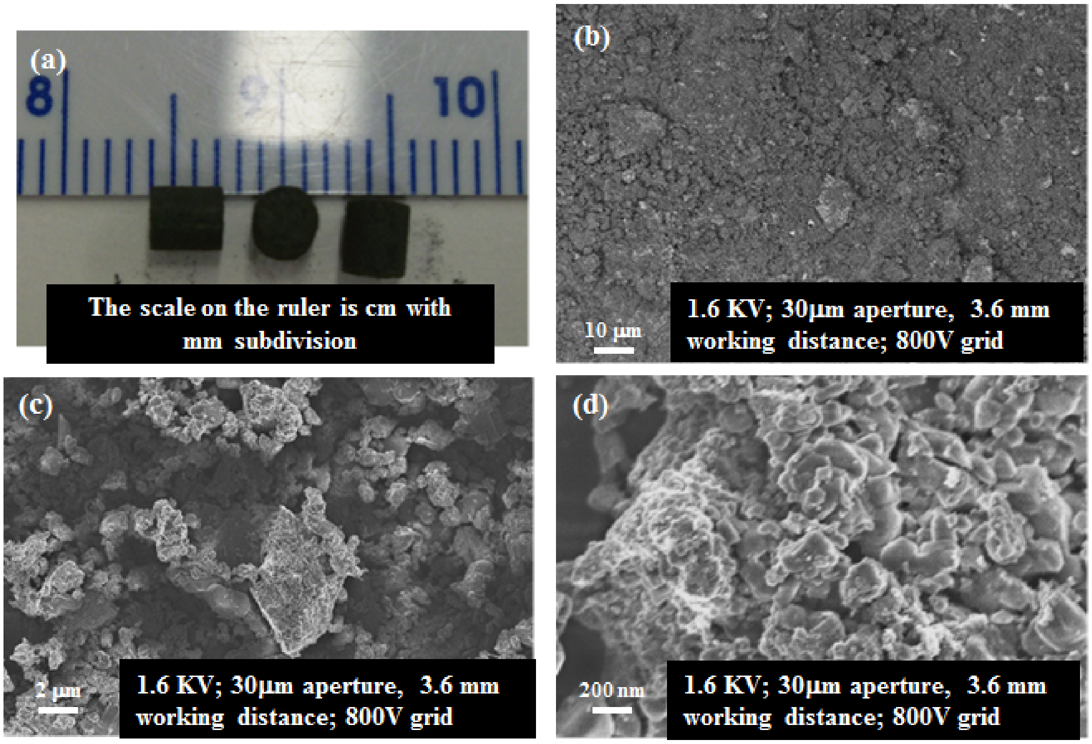
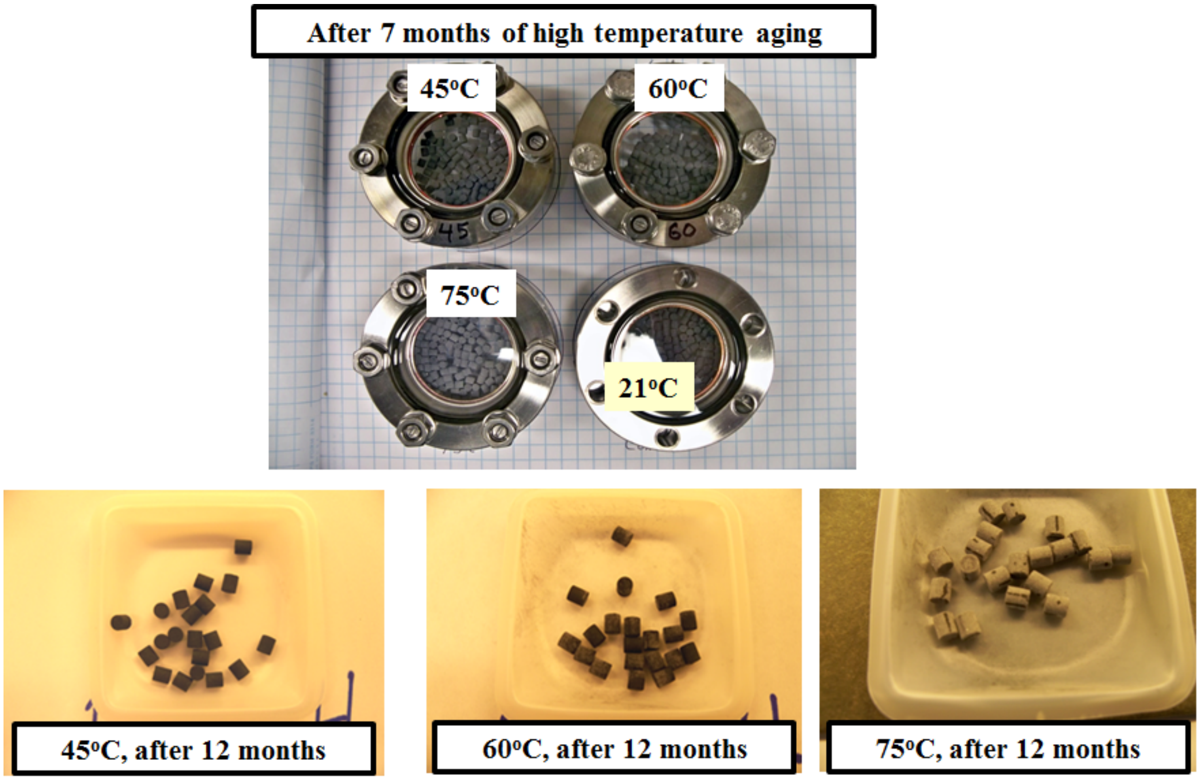

(a)
8
9
10
The scale on the ruler is cm with mm subdivision
(b)
10 µm
1.6 KV; 30µm aperture, 3.6 mm working distance; 800V grid
(c)
2 µm
1.6 KV; 30µm aperture, 3.6 mm working distance; 800V grid
(d)
200 nm
1.6 KV; 30µm aperture, 3.6 mm working distance; 800V grid

Fig. 3


After 7 months of high temperature aging

45°C, after 12 months

60°C, after 12 months

75°C, after 12 months

Fig. 4

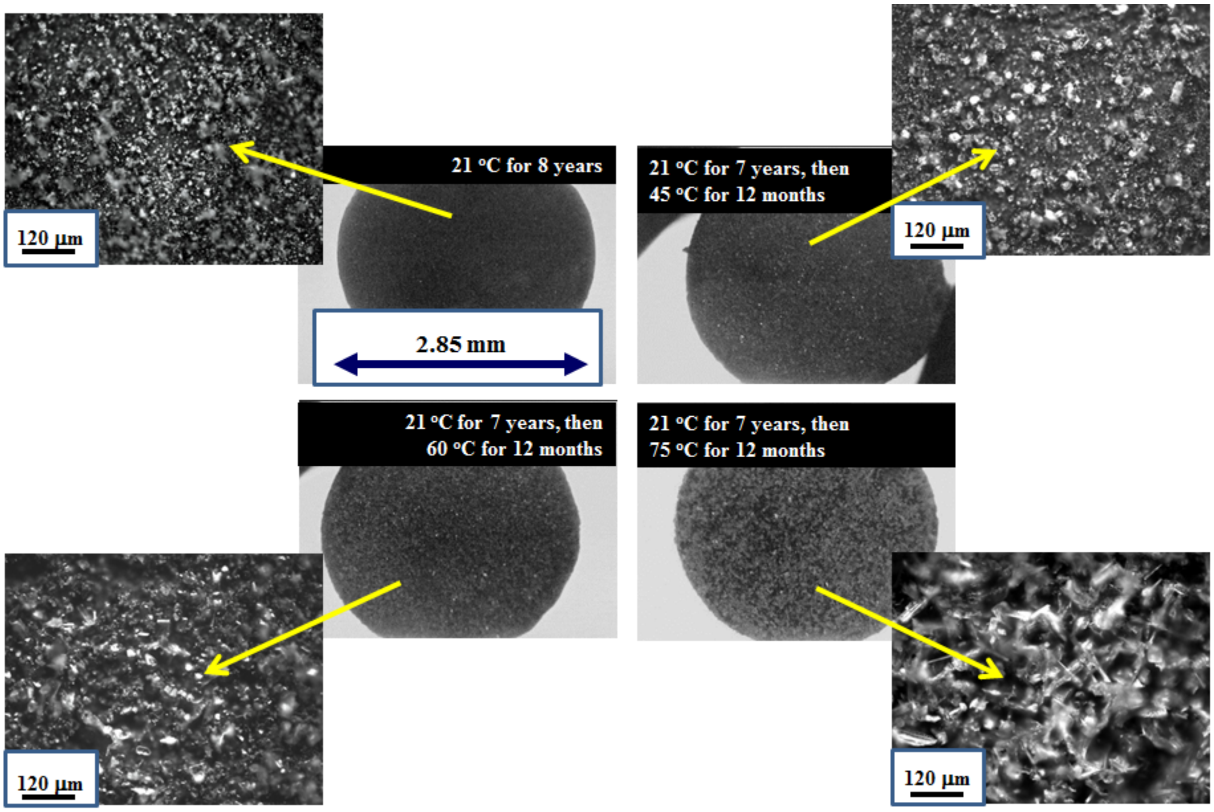

21 °C for 8 years
21 °C for 7 years, then 45 °C for 12 months
21 °C for 7 years, then 60 °C for 12 months
21 °C for 7 years, then 75 °C for 12 months
2.85 mm
120 µm
120 µm
120 µm
120 µm

Fig. 5

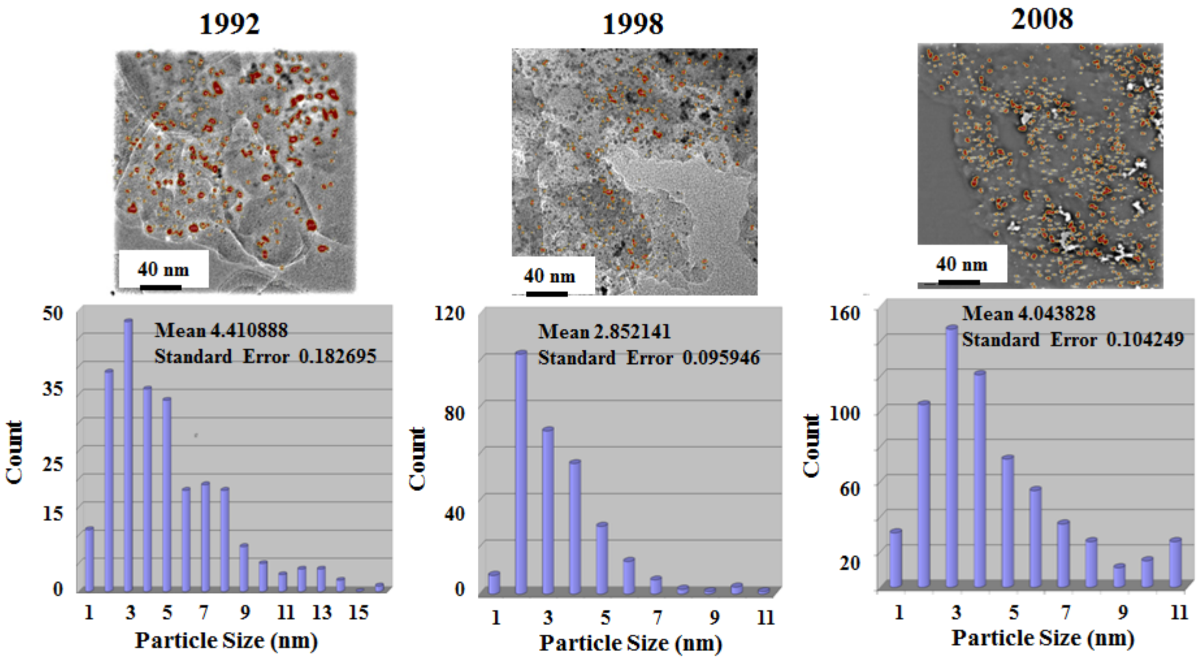
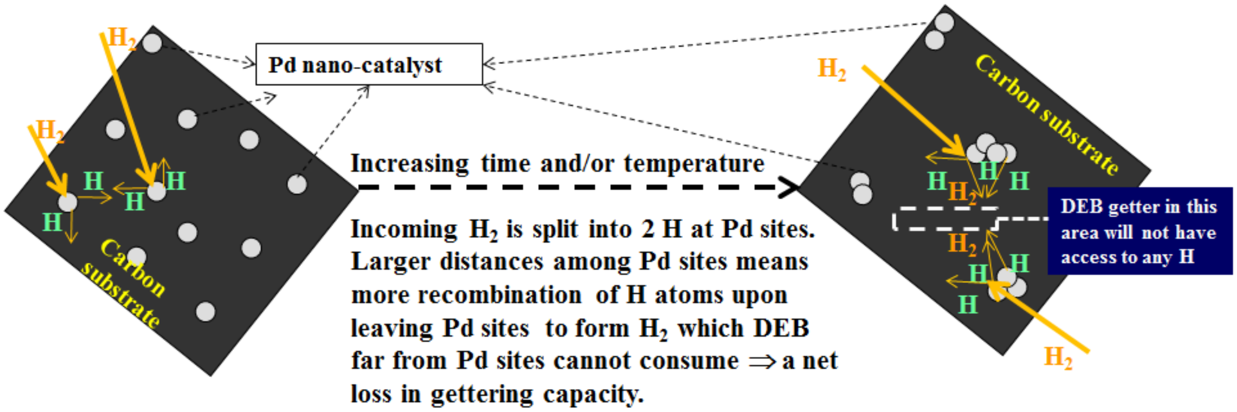

1992
40 nm
Mean 4.410888
Standard Error 0.182695
Count
50
35
25
15
0
1
3
5
7
9
11
13
15
Particle Size (nm)
1998
40 nm
Mean 2.852141
Standard Error 0.095946
Count
120
80
40
0
1
3
5
7
9
11
Particle Size (nm)
2008
40 nm
Mean 4.043828
Standard Error 0.104249
Count
160
100
60
20
1
3
5
7
9
11
Particle Size (nm)

Fig. 6



Increasing time and/or temperature

Incoming H_2 is split into 2 H at Pd sites. Larger distances among Pd sites means more recombination of H atoms upon leaving Pd sites to form H_2 which DEB far from Pd sites cannot consume ⇒ a net loss in gettering capacity.

Fig. 7

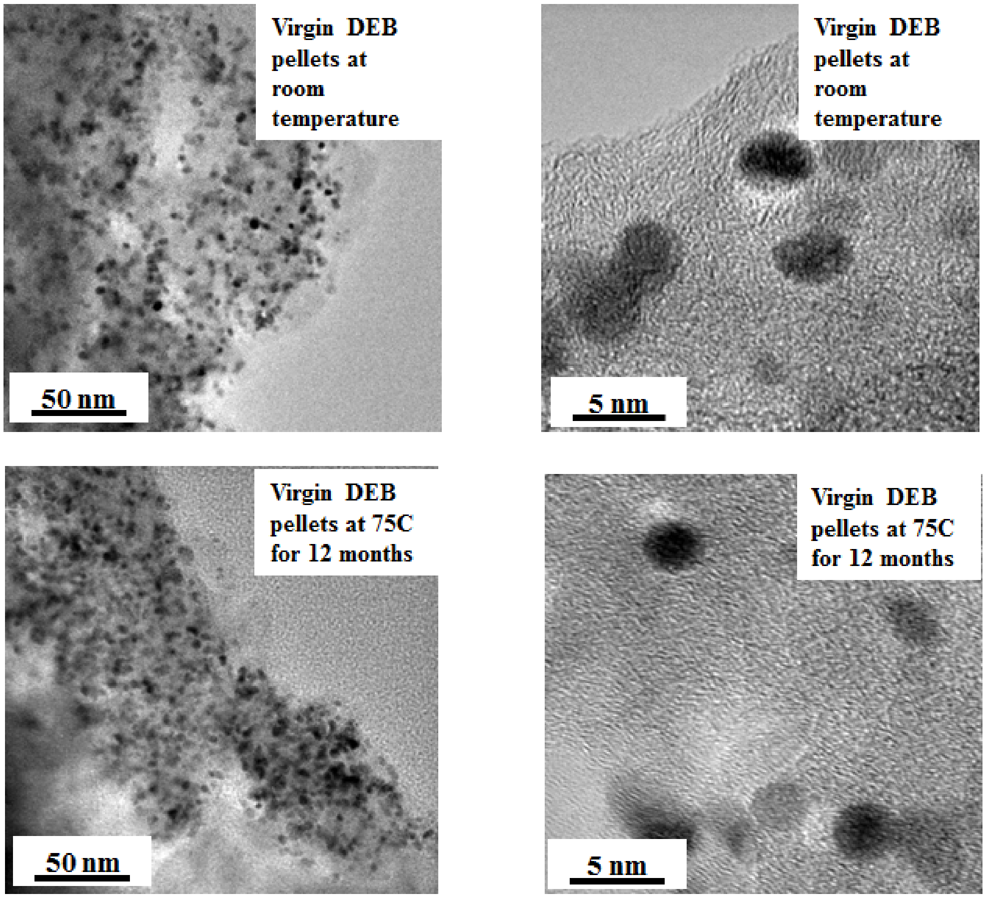

Virgin DEB pellets at room temperature
50 nm
Virgin DEB pellets at room temperature
5 nm
Virgin DEB pellets at 75C for 12 months
50 nm
Virgin DEB pellets at 75C for 12 months
5 nm

Fig. 8

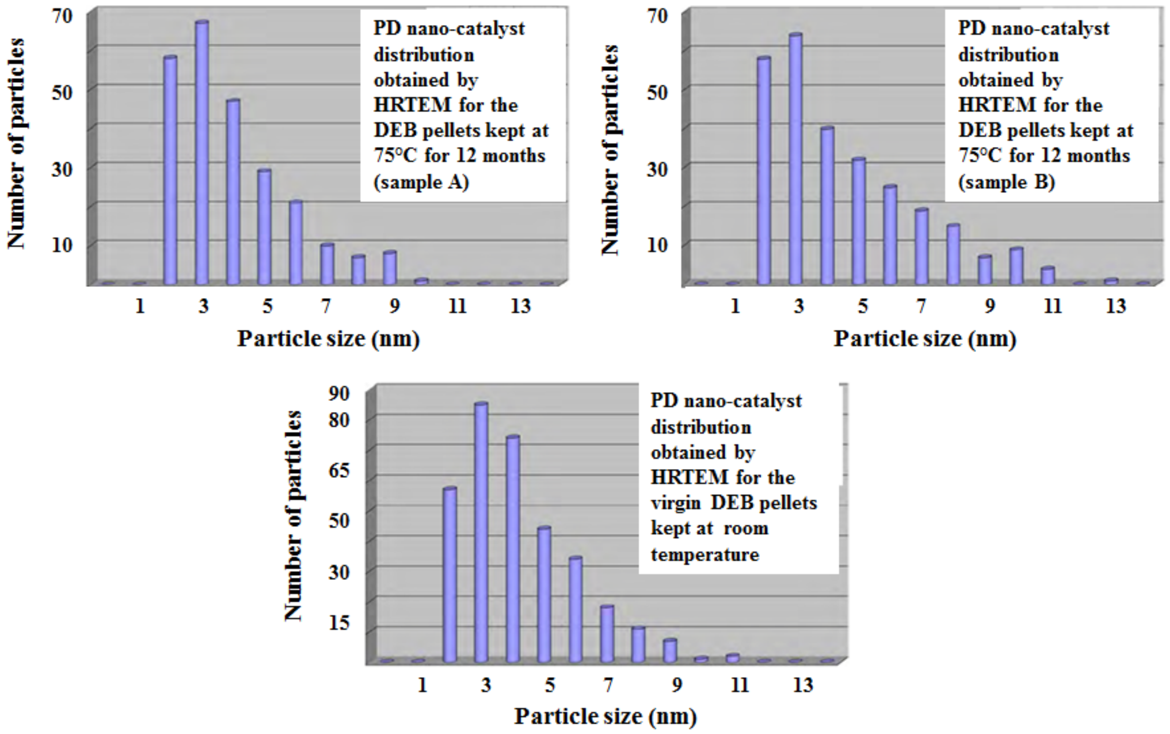
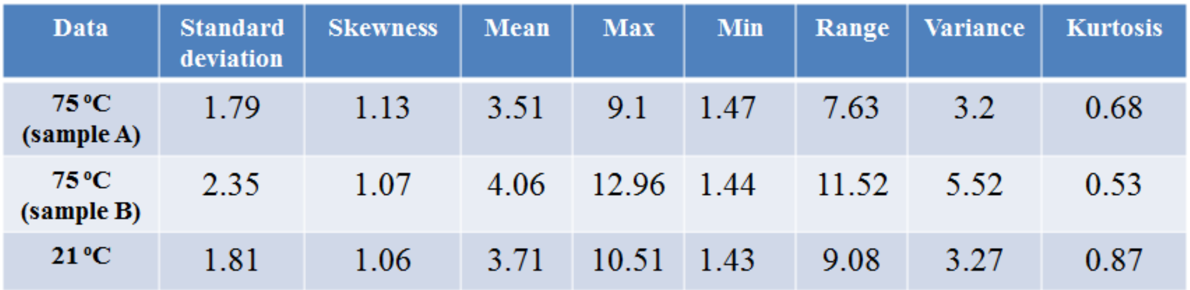

Number of particles
70
50
30
10
1
3
5
7
9
11
13
Particle size (nm)
PD nano-catalyst distribution obtained by HRTEM for the DEB pellets kept at 75°C for 12 months (sample A)
Number of particles
70
50
30
10
1
3
5
7
9
11
13
Particle size (nm)
PD nano-catalyst distribution obtained by HRTEM for the DEB pellets kept at 75°C for 12 months (sample B)
Number of particles
90
80
65
50
30
15
1
3
5
7
9
11
13
Particle size (nm)
PD nano-catalyst distribution obtained by HRTEM for the virgin DEB pellets kept at room temperature

Fig. 9



Data	Standard deviation	Skewness	Mean	Max	Min	Range	Variance	Kurtosis
75 ºC (sample A)	1.79	1.13	3.51	9.1	1.47	7.63	3.2	0.68
75 ºC (sample B)	2.35	1.07	4.06	12.96	1.44	11.52	5.52	0.53
21 ºC	1.81	1.06	3.71	10.51	1.43	9.08	3.27	0.87

Table I

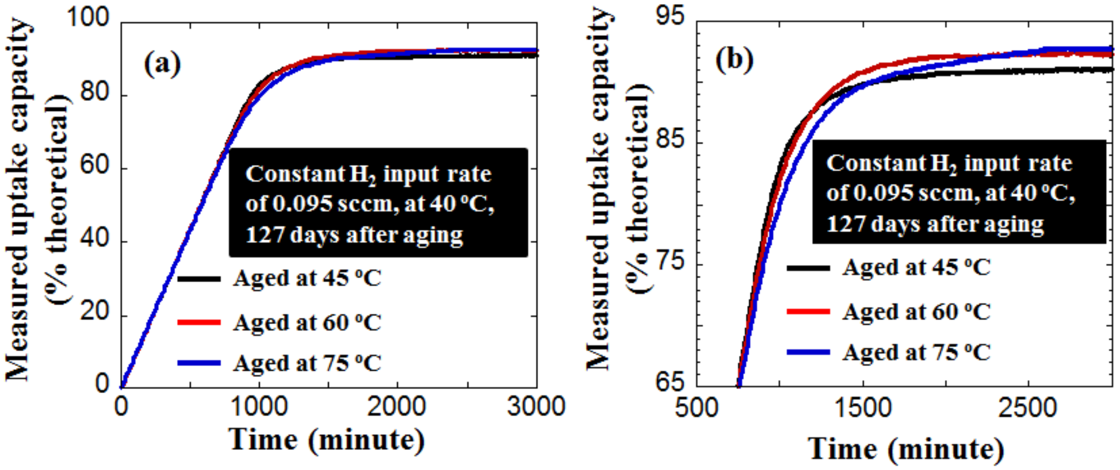
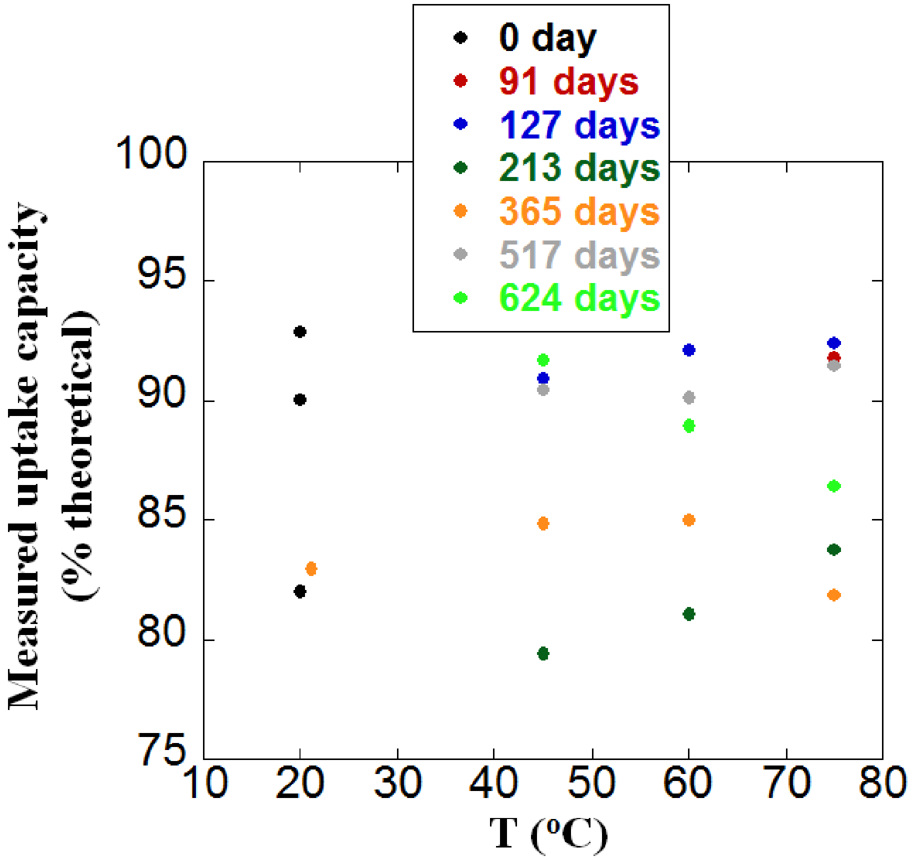
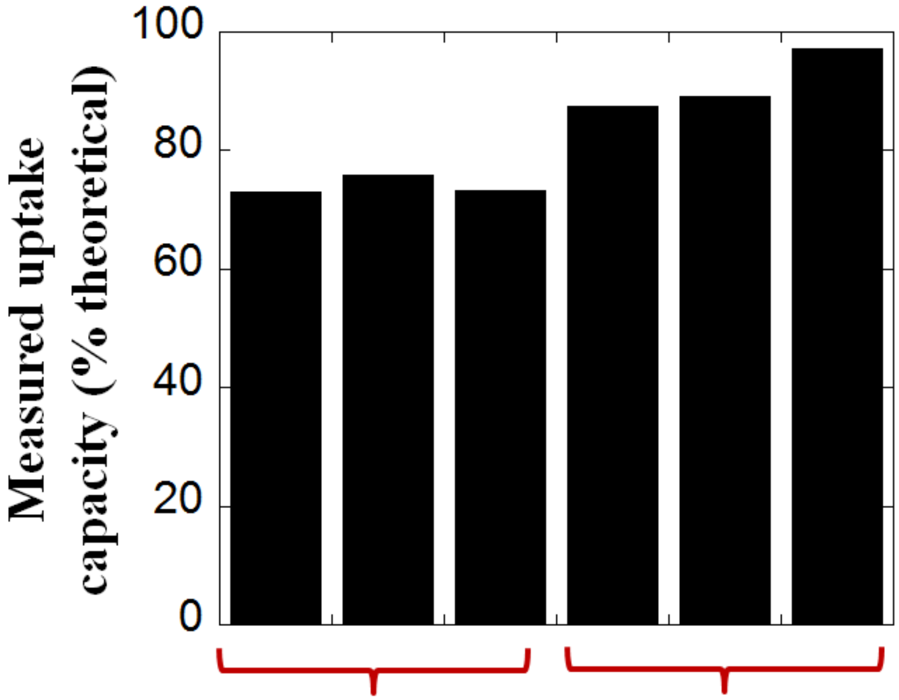

(a)
Measured uptake capacity (% theoretical)
100
80
60
40
20
0
0
1000
2000
3000
Time (minute)
Constant H_2 input rate of 0.095 sccm, at 40 °C, 127 days after aging
Aged at 45 °C
Aged at 60 °C
Aged at 75 °C
(b)
Measured uptake capacity (% theoretical)
95
85
75
65
500
1500
2500
Time (minute)
Constant H_2 input rate of 0.095 sccm, at 40 °C, 127 days after aging
Aged at 45 °C
Aged at 60 °C
Aged at 75 °C

Fig. 10



● 0 day
● 91 days
● 127 days
● 213 days
● 365 days
● 517 days
● 624 days
Measured uptake capacity (% theoretical)
100
95
90
85
80
75
10
20
30
40
50
60
70
80
T (ºC)

Fig. 11



These long-term air exposed getter pellets did not undergo 75 °C vacuum heating prior to uptake.

These long-term air exposed getter pellets underwent 75 °C vacuum heating for 2 days prior to uptake.

Fig. 12

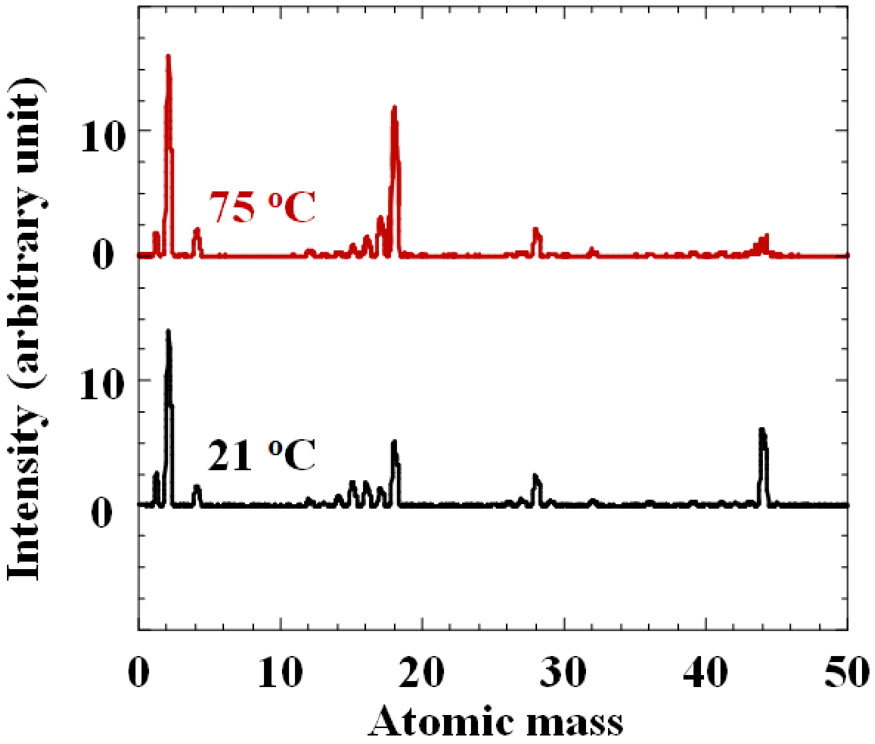

Intensity (arbitrary unit)
10
0
75 °C
10
0
21 °C
0
10
20
30
40
50
Atomic mass

Fig. 13