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Complete Iodine Loading of NO-Aged Ag^0-Functionalized Silica Aerogel

Fuel Cycle Research & Development

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SUMMARY

In off-gas treatment systems within a nuclear fuel reprocessing plant, capture materials will be exposed to a gas stream for extended periods during their lifetime. This gas stream may be at elevated temperature and could contain water, NO_x gas, or a variety of other constituents. For this reason, it is important to understand the effects of long-term exposure, or aging, on proposed capture materials. One material under consideration for iodine sequestration is silver-functionalized silica aerogel (Ag^0-aerogel). The aim of this study was to determine the effect of extended exposure at 150°C to an air stream containing NO on the iodine capture capacity of Ag^0-aerogel.

Ag^0-aerogel was provided by the Pacific Northwest National Laboratory (PNNL), which manufactures the material at a lab scale. Prior to aging, the material has an iodine loading capacity of approximately 290 mg I/g Ag^0-aerogel. Previous studies have aged the material in a dry air stream or in a moist air stream for up to 6 months. Both tests resulted in a 22% loss in iodine capacity.[1,2] Aging the material in a static 2% NO_2 environment for up to 2 months results in a 15% loss of iodine capacity.[3] In this study, exposure of Ag^0-aerogel to 1% NO at 150°C for 2 months produced a loss of 43% in iodine loading capacity. This is largest loss observed for aerogel aging studies to date.

The performance of Ag^0-aerogel in this study was compared to the performance of reduced silver mordenite (Ag^0Z) in similar studies. Ag^0Z is a zeolite mineral considered to be the current standard technology for iodine removal from off-gas streams of a potential US used fuel processing plant. In an aging study exposing Ag^0Z to 1% NO for 2 months, an iodine capacity loss of over 80% was observed.[3] This corresponds to a silver utilization of 13.5% for 2 month NO-aged Ag^0Z, compared to 57% silver utilization for 2 month NO-aged aerogel. While iodine loading capacity and silver utilization are critical parameters in evaluating these materials, other properties must also be considered when selecting the appropriate material (e.g., relative material densities and potential waste form production technology). The resistance of Ag^0-aerogel to NO is promising, and investigations of this material for use in iodine capture should continue to be pursued.

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ACRONYMS

Ag^0-aerogel	Silver-functionalized silica aerogel
Ag^0Z	Reduced silver-exchanged mordenite
PNNL	Pacific Northwest National Laboratory

MATERIAL RECOVERY AND WASTE FORM DEVELOPMENT CAMPAIGN

COMPLETE IODINE LOADING OF NO-AGED Ag^0-FUNCTIONALIZED SILICA AEROGEL

1. INTRODUCTION

In an off-gas system within a nuclear fuel reprocessing plant, any capture material will be exposed to a gas stream for months at a time. This gas stream may be at elevated temperature and could contain water, NO_x gas, or a variety of other constituents. For this reason, it is important to understand the effects of long-term exposure, or aging, on proposed capture materials. One material under consideration is silver-functionalized silica aerogel (Ag^0-aerogel), currently being produced in bench-scale quantities by collaborators at Pacific Northwest National Laboratory (PNNL).

Previous studies have examined the effects of extended aging at 150°C under a dry air stream, a humid air stream, and a static NO_2 environment on the iodine capture capacity of Ag^0-aerogel.[1,2] The loss in iodine capacity due to aging in each environment is shown in Table 1. The aim of this study was to determine the effects of extended exposure at 150°C to an air stream containing NO on the iodine capture capacity of Ag^0-aerogel. The results of this aging study in concert with other previously completed studies will provide insight into the ability of Ag^0-aerogel to withstand exposure to corrosive off-gas streams.

Table 1. Effects of aging environment on the iodine capacity of Ag^0-aerogel.[1,2]

Aging environment	Aging time (months)	Capacity loss (%)
Flowing dry air; 150°C	6	22
Flowing humid air; 150°C	6	22
Static 2% NO_2; 150°C	2	15

2. MATERIALS AND METHODS

Ag^0-aerogel was provided by PNNL in FY 2015. It is a granular, friable material with a particle size of <0.85 μm. The particles are black with brown and tan areas (Figure 1).

The aging study was conducted in a deep-bed test system. A 20.4 g charge of Ag^0-aerogel was contained in a stainless steel tube (I.D. 1.87 in.) located in a tube furnace. The furnace was held at 150°C for the duration of the study. A dry air stream (dew point < -70°C) was mixed with 100% NO gas in ¼ in. stainless steel tubing approximately 12 in. prior to entering the tube furnace. There was a second gap of approximately 12 in. in length between the bottom of the tube furnace and the sample bed; this was to ensure that the gas was fully heated prior to encountering Ag^0-aerogel and that the sample charge was fully located in the heating zone of the furnace. Thermocouples were placed in this heating zone, in the sample bed, and at the tube exit to monitor the gas and sample temperatures. The gas velocity through the tube was 3.5 m/min.

At selected intervals of 1 week, 1 month, and 2 months, the flow was suspended and the oven cooled. The material was removed from the tube and gently mixed to ensure homogeneity; a ≈ 5 g sample was subsequently removed, and the remainder of the material was returned to the tube for continued aging. After the aging test was complete, the 1 and 2 month samples were loaded with 50 ppm iodine from a dry feed stream (< -70°C dew point) in a thermogravimetric analyzer, described in detail in a previous report.[4] The gas velocity for iodine loading was 10 m/min, and the temperature was held at 150°C. The iodine loading for each sample was determined by sample weight upon conclusion of iodine loading.

3. TESTING RESULTS

Exposure to 1% NO gas significantly affected the structure and appearance of the aerogel. As shown in Figure 1, an initial change in material color was observed after 1 week of aging. The black granules did not change color, but the lighter colored granules turned bright orange and red. After 1 month of aging, a substantial amount of fines had developed and the material was mostly dark brown with orange, white, yellow, and red granules dispersed throughout. The material was sieved through a 30×30mesh, and the fines were found to constitute 30% by weight of the material remaining in the tube after the 1 week sample was removed. The fines were not returned to the tube furnace for continued aging. No additional change in color was observed upon 2 months of aging, and no significant amount of additional fines was created during the second month of aging.

Figure 1. Effects of NO aging on Ag^0-aerogel (L to R: 0 month, 1 week, and 1 month aged Ag^0-aerogel).

Iodine loading curves for 0, 1, and 2 month aged samples are shown in Figure 2, and the total iodine capacity for each is shown in Table 2. A loss of 24% iodine capacity was observed within the first month of aging, and after 2 months, the total capacity loss increases to 43%.

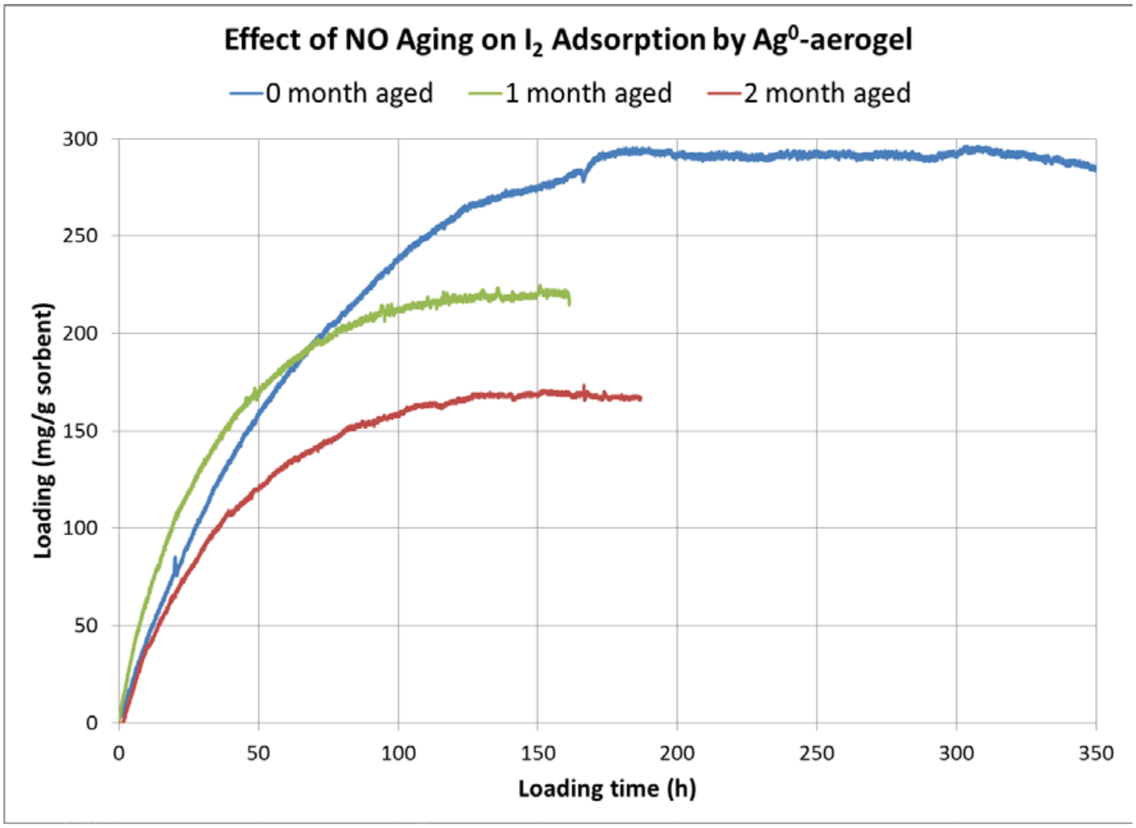

Figure 2. Effect of NO aging on I_2 adsorption by Ag^0-aerogel.

Table 2. Iodine loading capacities for NO-aged Ag^0-aerogel.

Aging time (months)	**Iodine loading capacity (mg I/g Ag^0-aerogel)**	**Decrease in Iodine Capacity (%)**
0	290	-
1	220	24
2	165	43

4. DISCUSSION

In this study, exposure of Ag^0-aerogel to 1% NO at 150°C for 2 months produced a loss of 43% in iodine loading capacity. This is largest loss observed for aerogel aging studies to date. The performance of Ag^0-aerogel in this study was compared to the performance of reduced silver mordenite (Ag^0Z) in similar studies.[5] Ag^0Z is a zeolite mineral considered to be the current standard technology for iodine removal from the off-gas streams of a potential US used fuel processing plant. In an aging study exposing Ag^0Z to 1% NO for 2 months, an iodine capacity loss of over 80% was observed. This corresponds to a silver utilization of 13.5% for 2 month NO-aged Ag^0Z, compared to 57% silver utilization for 2 month NO-aged aerogel. While iodine loading capacity and silver utilization are critical parameters in evaluating these materials, other properties must also be considered when selecting the appropriate material. The density of these two materials is significantly different (Ag^0Z, 0.77 g/cm^3; Ag^0-aerogel, 0.35 g/cm^3) and is an important factor in plant design. Additionally, the potential waste forms that might be appropriate for final disposition of either material are still under investigation and will guide the selection of a sorbent that is ultimately required to be treated as high-level waste. However, the results of this study demonstrate that the resistance of Ag^0-aerogel to NO is promising, and both fundamental and applied investigations of this material for use in iodine capture should continue to be pursued.

5. REFERENCES

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