

## **Impact of Pretreatment and Aging on the Iodine Capture Performance of Silver-Exchanged Mordenite - 12314**

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### **ABSTRACT**

Volatile gas emissions from a nuclear fuel recycle facility in the United States are governed by several key regulations, including 10 CFR 20, 40 CFR 61, and 40 CFR 190. Under 40 CFR 190, the total quantity of iodine that may be released to the environment from the entire fuel cycle is limited to 5 millicuries of I-129 per gigawatt-year of electrical energy produced by the fuel cycle. With a reasonable engineering margin, an iodine decontamination factor (DF) of approximately 1000 will be required for the complete fuel cycle.

Off-gas treatment in a fuel reprocessing plant must address several gas streams containing iodine, among a number of volatile radionuclides. Past research and developmental activities identified silver-exchanged mordenite (AgZ) as a very promising sorbent based on its acid resistance, relatively high iodine and methyl iodide capacity, and high achievable DF. Recent studies at ORNL have focused on the impacts of long-term exposure to simulated off-gas streams (aging) and pretreatment on the iodine adsorption performance of hydrogen-reduced silver-exchanged mordenite (Ag<sup>0</sup>Z). Experiments were conducted to determine the effects of long-term exposure to both dry and moist air on the iodine sorption capacity of Ag<sup>0</sup>Z. The data indicates that aging reduces the capacity of Ag<sup>0</sup>Z, which must be accounted for to prevent degradation of DF.

### **INTRODUCTION**

A generic high-level fuel reprocessing flowsheet is shown in Figure 1. It illustrates the mass distribution of the key components from the processing of 1 metric ton (t) of light-water reactor (LWR) used nuclear fuel (UNF) with a burnup of 60 GWd/tIHM (metric ton initial heavy metal) and 5 years of cooling. Highlighted by the red circles are the masses and curies of the volatile components of interest for off-gas processing.

Off-gas treatment in a fuel reprocessing plant must address four primary gas streams containing iodine, among a number of volatile radionuclides. The first stream is the off-gas from the head-end operations, which include the shear, an optional voloxidizer, and the dissolver. This stream is typically referred to as the dissolver off-gas (DOG). The

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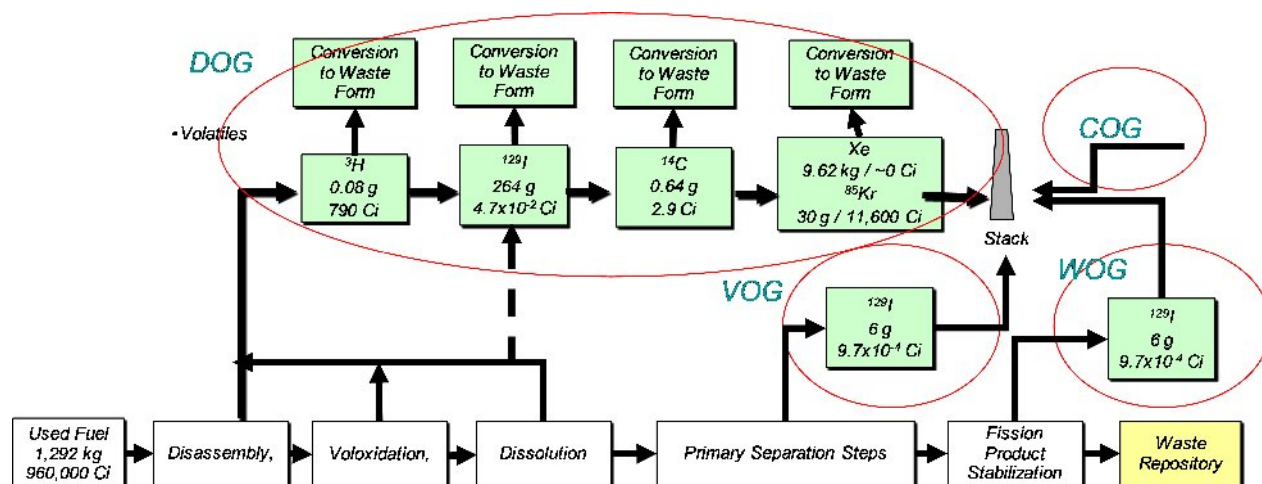


Fig. 1. Volatile fission / activation products from processing 1 t of spent nuclear fuel at 60 GWd/tIHM with 5 years of cooling.

second stream that contains iodine is called the vessel off-gas (VOG), which consists of process equipment off-gas (e.g., the instrument air used in bubblers, air sparge discharge, in-leakage, etc.) The third is the cell off-gas (COG), which provides confinement to the process cell. The fourth is the waste systems off-gas (WOG) that originates from the operations to produce/solidify waste forms. Each of these streams has unique characteristics and off-gas processing challenges.

Volatile gas emissions from a nuclear fuel recycle facility are addressed in several regulatory documents. The U.S. Environmental Protection Agency (EPA) has established through 40 CFR 190 annual dose limits resulting from nuclear fuel cycle facilities in the commercial sector [1]. The dose limits for specific organs and for the whole body are provided in 40 CFR 190.10. Specific release limits for Kr-85, I-129, and Pu-239 in terms of curies released per unit of power produced are also defined in 40 CFR 190 [1]. Under 40 CFR 190, the total quantity of iodine that may be released to the environment from the entire fuel cycle is limited to 5 millicuries of I-129 per gigawatt-year of electrical energy produced by the fuel cycle. For iodine the minimum required decontamination factor (DF) based on 40 CFR 190 is  $\sim 200$  (with no margin and complete allocation to the reprocess portion of the fuel cycle) [2]. The dose limits for both workers and individual members of the public are provided in 10 CFR 20 [3]; 40 CFR 61.92 provides additional limits of 10 mrem/year dose equivalent to the public [4]. Depending on the size and siting of the facility, the DF requirements for the four radionuclides of interest (H-3, C-14, Kr-85, and I-129) based on 10 CFR 20 or 40 CFR 61 may exceed those required by 40 CFR 190. With a reasonable design margin, the required DF for iodine is expected to be on the order of 1000.

The head-end voloxidizer off-gas (VoxOG) and DOG concentrations [5] shown in Table I were estimated using data from a large engineering-scale reprocessing equipment test facility located at ORNL by Birdwell [6] and reported by Jubin et al. [7]. In addition to tritium, minor but radiologically significant quantities of other fission products are

Table I. Source terms – High Burnup, Short-Cooled Fuel.

	Total released to off-gas streams (g/tIHM)	VoxOG (g/tIHM)	DOG (g/tIHM)	VOG (g/tIHM)	VoxOG (ppmv)	DOG (ppmv)	VOG (ppmv)
Tritiated Water as HTO (UNF)	0.545	0.545	--	--	0.79	Removed in VoxOG	
Water (UNF)	2.683	2.683			4.20	Removed in VoxOG	
H <sub>2</sub> O (process)		7.24	75205		12	32500	
CO <sub>2</sub> (UNF)	68	34	34		Combined with DOG	9.3	
CO <sub>2</sub> (process)		---	2206		Combined with DOG	390	
I (UNF)	358	---	347	10.7	Combined with DOG	8.2	0.16
Cl (from process - HNO <sub>3</sub> )	156		156		Combined with DOG	13.4	
Kr (UNF)	626	313	313	--	Combined with DOG	46	
Ar <sub>air</sub>	60924				Combined with DOG	9300	
Kr <sub>air</sub>	15.6				Combined with DOG	1.1	
Xe (UNF)	9616	4808	4808	--	Combined with DOG	450	

Basis: VoxOG rate of 270 L/m

DOG rate of 1000 L/m

VoxOG combined with DOG after <sup>3</sup>H removal

VOG rate of 2000 L/m

Gas to Voloxidizer has -60°C dew point

Air cell at 15°C dew point

DOG cooled to 25°C leaving dissolver

50% Kr/Xe release in voloxidizer to VoxOG – balance reports to DOG

50% CO<sub>2</sub> release in voloxidizer to VoxOG – balance reports to DOG

97% of iodine is released from dissolver into DOG – balance reports to VOG

Fuel Burnup of 60 GWd/tIHM, 5 years of cooling prior to processing

Assumed processing rate of 100 t fuel processed per year over 200 days

The concentrations of fluorine and bromine in HNO<sub>3</sub> are too low to report in this table.

The concentration of krypton in air is about 1 ppmv and xenon about 90 ppbv in air.

\*Chlorine will be trapped with I<sub>2</sub>, limiting AgZ sorbent bed capacity.

released during the standard voloxidation process. These include carbon (C-14), iodine (I-129), and krypton (Kr-85). The fraction released has been reported to be as high as ~50% of the carbon (as CO<sub>2</sub>), ~1% of the iodine, and ~5% of the krypton. Plant capacity and design of equipment can result in significant variations to the off-gas rates and the resulting radionuclide concentrations as well as the amount of nonradioactive constituents contained in the off-gas. The DOG rates for several facilities when normalized for throughput were ± a factor of 4 from the ORNL demonstration rates. Table I used radionuclide content from SCALE V6 [8] calculations for LWR fuel at a burnup of 60 GWd/tIHM and processing after a 5 year decay period following reactor discharge.

This assumed the implementation of voloxidation technology. These estimates assume an air atmosphere in the hot cell and a limited or controlled level of leakage into the process equipment. The oxidation gas for voloxidation is air with CO<sub>2</sub> removed. The VOG flow rate is assumed to be twice the total DOG flow.

Studies of the distribution of I-129 from UNF being processed into the gas and liquid process streams indicate that about 94% to 99% of the I-129 ends up in the DOG [9, 10]. As the DOG contains the highest fraction of the volatile iodine, the primary iodine recovery technology will be applied to this stream. Treatment of the VOG and other off-gas streams is also anticipated to recover the required amount of I-129.

A variety of technologies have been developed for the recovery of I-129 from the off-gas streams. These technologies include scrubbing with caustic or acidic solutions and chemisorption on silver-containing materials or adsorbents. Because of its high acid resistance, a Ag<sup>°</sup>Z sorbent has been developed specifically for application in treating DOG streams. While extensive tests were conducted in the United States on a form of this sorbent, the specific material previously tested is no longer commercially available, and similar materials are currently being evaluated.

Recent iodine adsorption studies at ORNL have focused on the impacts of aging on the Ag<sup>°</sup>Z with the ultimate goal of understanding the fundamental phenomena that control aging for this material and other zeolites that could be considered for use in off-gas treatment in the future. Since these materials must be in continuous service, it will be important to understand the impact of extended exposure to the off-gas streams. Iodine loading data were obtained for Ag<sup>°</sup>Z exposed to air for periods of up to 6 months, and the changes in the adsorbent material were characterized. The following sections of this paper summarize the tests performed to understand the aging of Ag<sup>°</sup>Z in dry and humid air and the impacts of aging on iodine capture.

## **TEST SYSTEMS**

### **Description of Ag<sup>°</sup>Z Aging System**

A cold test system was fabricated to simulate in-process aging of Ag<sup>°</sup>Z. This test rig is schematically represented by Figure 2a, and the equipment as installed in a hood is shown in Figure 2b. Major components of this system include two stainless steel aging columns and their associated tube furnaces, flow control valves, relative humidity probes, an air dryer system, and numerous isolation and pressure relief valves. Each column will hold a nominal 550 g charge of Ag<sup>°</sup>Z. The apparatus was designed to “age” Ag<sup>°</sup>Z under increasingly more challenging conditions than the iodine adsorbent would routinely experience in an actual nuclear fuel reprocessing plant, with air flowing into the bottom of the aging column in the lower furnace and exiting the top of the aging column in the upper furnace. Total exposure time (aging) with dry air was up to 6 months at various aging conditions. In situ hydrogen reduction of the AgZ was also achieved with this apparatus.

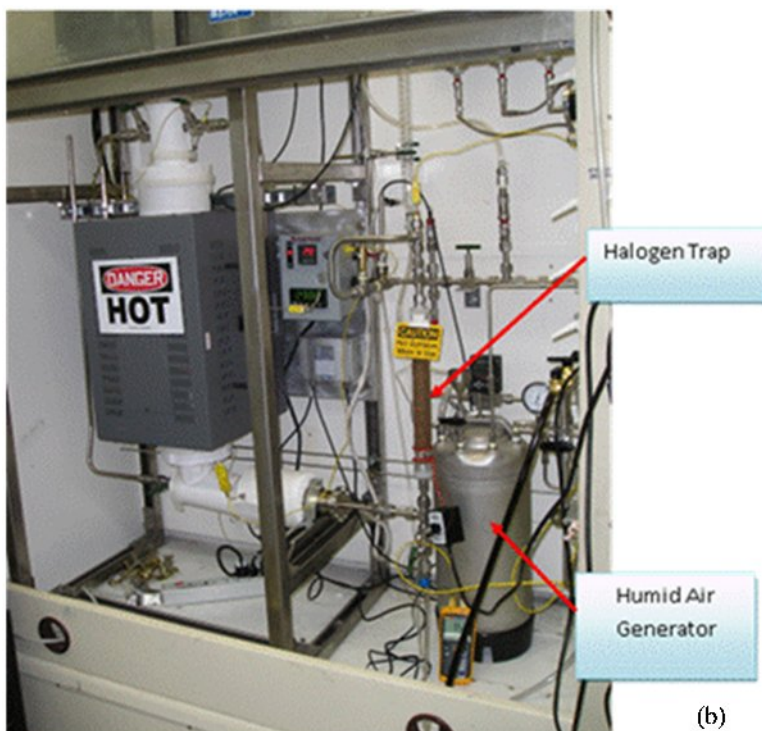
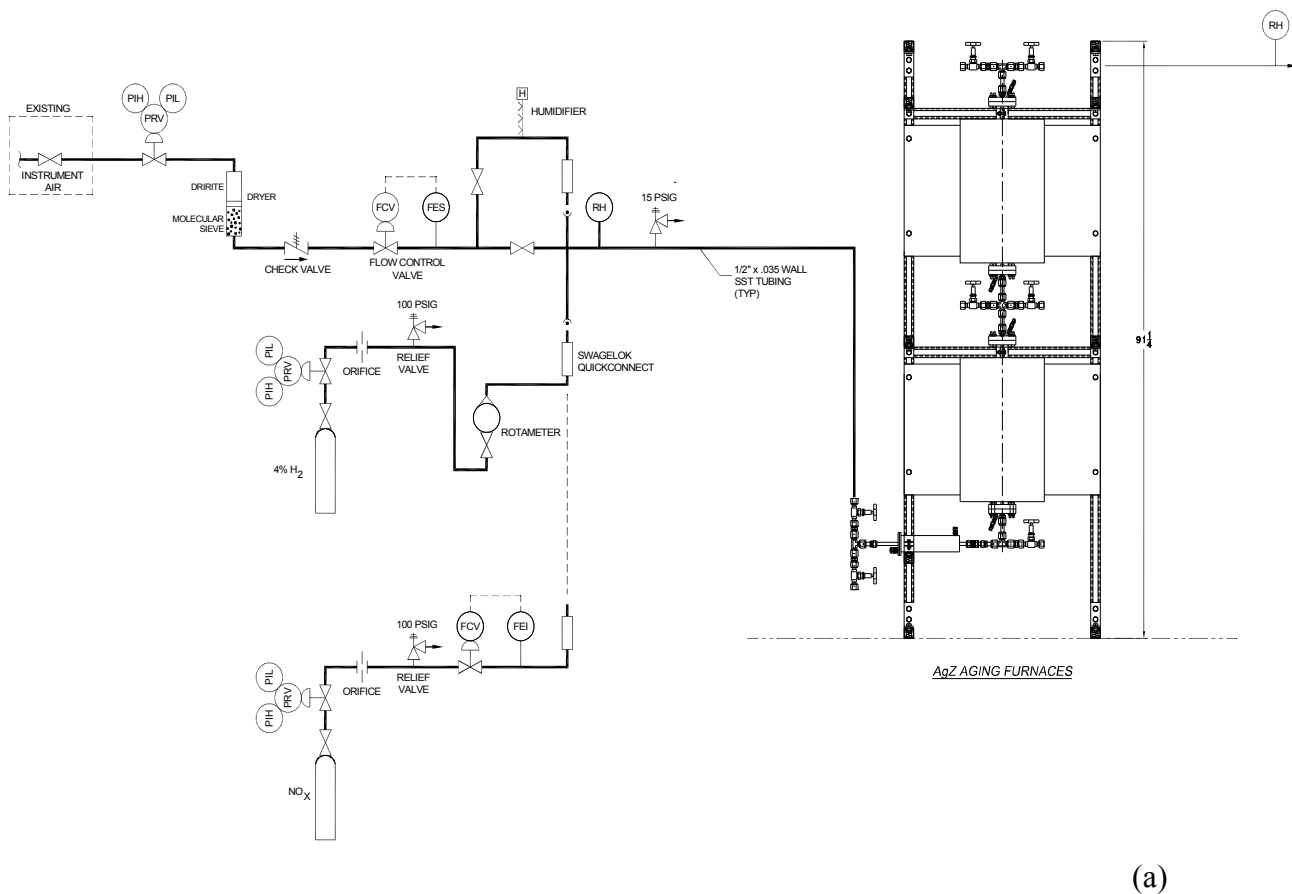


Fig. 2 a) Ag<sup>o</sup>Z aging equipment schematic; b) Ag<sup>o</sup>Z aging apparatus equipped with high-capacity humidifier and halogen trap.

The dry-air aging gas was furnished to the system by a regulated existing instrument air supply which flows through a combination canister dryer containing anhydrous calcium sulfate followed by a Type 3A molecular sieve. Under the flow conditions selected for aging, this dryer assembly would supply dry air with a dew point of a nominal  $-55^{\circ}\text{C}$ . The nominal superficial gas velocity is  $\sim 10$  m/min.

To facilitate humid-air aging of  $\text{Ag}^{\circ}\text{Z}$  for extended periods, a high-capacity humidifier was installed in the system. This 19 L humidifier can consistently supply  $15^{\circ}\text{C}$  dew point air at a rate of 17 L/min (nominal). This allowed the humid-air aging of  $\text{Ag}^{\circ}\text{Z}$  at  $150^{\circ}\text{C}$  for extended periods (6 months) with minimal attendance.

A sacrificial bed of heated  $\text{Ag}^{\circ}\text{Z}$  was added to the inlet of the aging system to “getter” potential halogen contaminants. The need for the sacrificial section was based on initial aging studies that are described by Jubin et al. [11] and summarized in the Results section below.

### **Description of Iodine Loading System**

“Single-pellet” or monolayer iodine loading tests of the aged  $\text{Ag}^{\circ}\text{Z}$  were conducted to determine the impacts on the adsorption rate and iodine capacity. An in-house-designed, large-scale thermal gravimetric analyzer (TGA) was constructed, as shown in Figure 3a. The principal components of this system included a small laboratory oven, a microbalance, and a three-legged weighing basket with legs long enough to reach out of the heat zone to a microbalance located under the oven. The weighing pan held from 2 to 3 g of  $\text{Ag}^{\circ}\text{Z}$ .

An in-house iodine generator was constructed and installed upstream of the TGA (schematically represented in Figure 3b). A saturated stream of elemental iodine was generated by passing dry air through a bed of iodine crystals. This saturated stream of iodine was injected directly into a diluent air stream to produce the desired iodine concentration. Typical iodine concentrations used in the tests described in this paper were on the order of 150 to  $400\text{ mg/m}^3$  based on the source term estimates shown in Table I.

Water vapor is a major constituent in the off-gas stream. The in-house design of a system that will produce controlled and predictable humid air was installed in the experimental iodine loading system. It is a system similar in design to the one installed in the experimental aging system shown schematically in Figure 2a. The principal components of the humid air generator pictured on the cart in the foreground of Figure 3a are a water bubbler, flow control valves, reflux condenser, constant-temperature circulating water bath, and an in-line humidity probe.

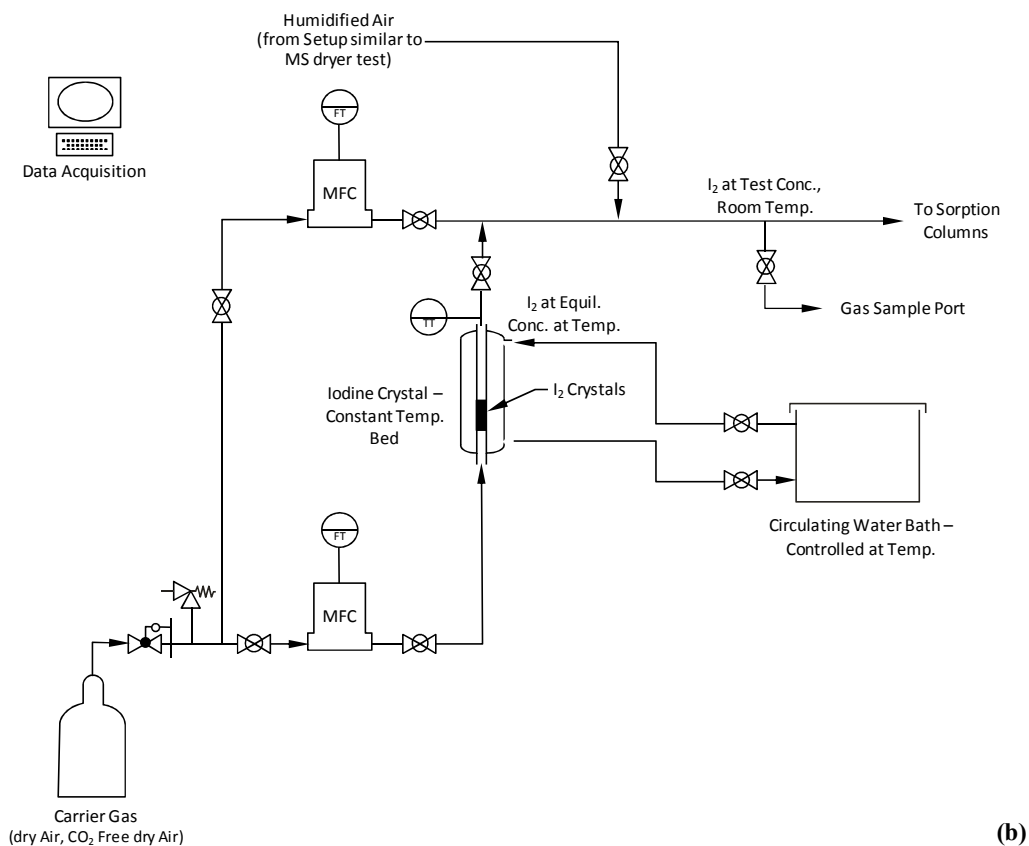
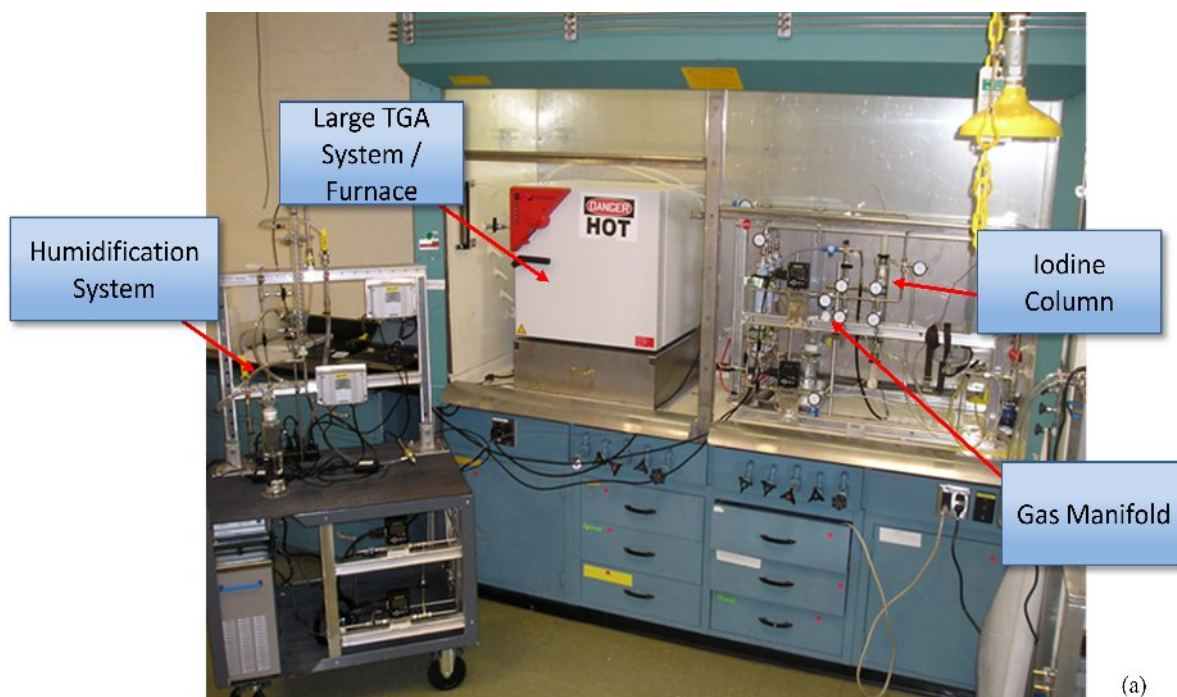


Fig. 3. a) Binder TGA Oven and companion gas delivery and humid air generator systems have doubled single-pellet-analysis capabilities; b) iodine generator flowsheet.

## RESULTS

### Materials Tested

The AgZ used in these studies was purchased from Ionex Corporation (Ionex-Type Ag 900 E16). The silver content from this particular lot of material (Lot # 0911314-1) was 9.5% by weight. Physical properties include a density of 0.93 g/mL with a moisture content of <2%. The pelletized material was a 1.6 mm (1/16 inch) diameter pellet with a mesh size distribution of 0.3% >10, 19.1% > 12, 79.8% >14, 0.2% >16, and 0.6% <16. The batch of IONEX Type Ag-900 procured for this study was predominately gray in color but had a few (<1%) white pellets intermixed.

### Ag°Z Aging

A series of tests are being performed to evaluate the impact of aging under increasingly more challenging conditions which include aging in 1) dry air, 2) moist air, 3) dry air and NO<sub>2</sub>, and 4) wet air and NO<sub>2</sub>. The results of the first two tests with dry and moist air are summarized below.

The first step in the aging study was to reduce the silver compound(s) in the AgZ to elemental silver with a flow of N<sub>2</sub>-4%H<sub>2</sub> at a temperature in the range of 200°C. The aging process parameters were set at 150°C with a dry airflow rate of 17 L/min (9.59 m/minute superficial velocity). Aged samples of Ag°Z were removed after 1, 2, 4, and 6 months of processing for physical properties testing and iodine loading studies.

Beginning with the 2 month aging samples, a distinct color change was noted in the Ag°Z material when comparing the change from the top tube furnace with that from the bottom tube furnace. The preliminary analysis of the halogen loading on the material from the large aging apparatus seemed to indicate that the discoloration may have been caused by chlorine with concentrations on the AgZ ranging between 27 and 376 ppm and fluorine in the concentration range of 17 to 26 ppm [11]. A smaller aging test was then conducted to replicate the behavior. In this test, the fluorine concentration on the AgZ was on the order of 700–900 ppm, and no chlorine was detected. At halogen loadings of ~800 ppm, less than 0.8% of the silver would be consumed. In either case, the quantity of halogen (presumed to be reacting with the available elemental silver) is not of sufficient quantity to have a serious impact on iodine loading. The exact source of the halogen was not pinpointed, but the plant air was the prime suspect. It should be noted that the contamination may not have been from a consistent source and may be random in time as well as concentration. Later aging tests employed a sacrificial bed of AgZ to trap, or “getter” any tramp halogens in the air supply.

The next most challenging condition was believed to be moist air, and Ag°Z was similarly aged with humid air over a 6 month period. The aging process parameters were set at 150°C with a total flow of humid air at 17 L/min (9.57 m/min superficial velocity) with a dew point of a nominal 15°C. Samples from 1, 2, 4, and 6 month aging intervals were extracted for examination and testing. Contrary to the observed color changes noted



in the dry-air aged samples, none of the humid-air aged samples showed a distinct color change as a function of aging. In addition, the Ag<sup>0</sup>Z-filled halogen trap that treated the humid air before it entered the aging furnace showed no signs of color change.

### **Iodine Loading**

Single-pellet or monolayer iodine loading tests of the aged Ag<sup>0</sup>Z were conducted to determine the impacts on the adsorption rate and iodine capacity. Weight gain with time under known, constant iodine concentration, temperature, and gas mixture rate of flow over the sample are also key factors in determining the mass transfer rate controlling steps of the iodine loading. Five experimental runs with the Ag<sup>0</sup>Z aged in dry air were completed using the large TGA system.

As shown in Figure 4a, a benchmark was created with the first run using Ag<sup>0</sup>Z that had not been aged. If all of the silver contained in the AgZ reacts to form AgI, the maximum weight gain would be 11.1%. After approximately 90 hours of exposure to a dry gas stream containing 250 mg I<sub>2</sub>/m<sup>3</sup>, the weight gain for the un-aged Ag<sup>0</sup>Z was ~9%. This represented an 81% silver utilization.

The remaining curves in Figure 4a show the effects of aging on Ag<sup>0</sup>Z from 1 to 6 months. Comparing the no-aging data with the 6 month aged material shows about a 40% reduction in iodine loading capacity.

The impact of moist-air aging was determined in a similar manner by loading the aged material with elemental iodine and evaluating the maximum loading achieved and the rate of iodine uptake. Samples of the 1, 2, and 4 month humid-air aged Ag<sup>0</sup>Z were tested in the single-pellet TGA system. An iodine loading curve was generated for each of the three humid-air agings of Ag<sup>0</sup>Z samples. The curves indicate that humid-air aging has a drastic impact on iodine loading. Each sample was loaded under an iodine concentration of 200 mg/m<sup>3</sup> with a superficial velocity of 9.73 m/min at 150°C. As seen in Figure 4b, a significant decrease in the rate and total loading of iodine uptake can be observed beginning with the shortest aging period (i.e., after 1 month) when compared with the loading curve using Ag<sup>0</sup>Z with no aging. Increasing exposure time to the humid air used to age the Ag<sup>0</sup>Z beyond 1 month resulted in a slight additional reduction in capacity at 2 months. Virtually identical capacity was observed with 4 months of aging. Based on the 1, 2, and 4 month iodine loading results and in the interest of time, iodine loading of the 6 month humid-air aged sample was not performed.

In comparison with dry-air aging study (Figure 4a), the 1 month humid-air aged total loading (~5 wt%) is equivalent to 6 months of dry-air aged Ag<sup>0</sup>Z. In both cases, a significant reduction in iodine loading capacity is seen when compared to over 11 wt% loading of the freshly H<sub>2</sub>-reduced Ag<sup>0</sup>Z (i.e., no aging) sample.

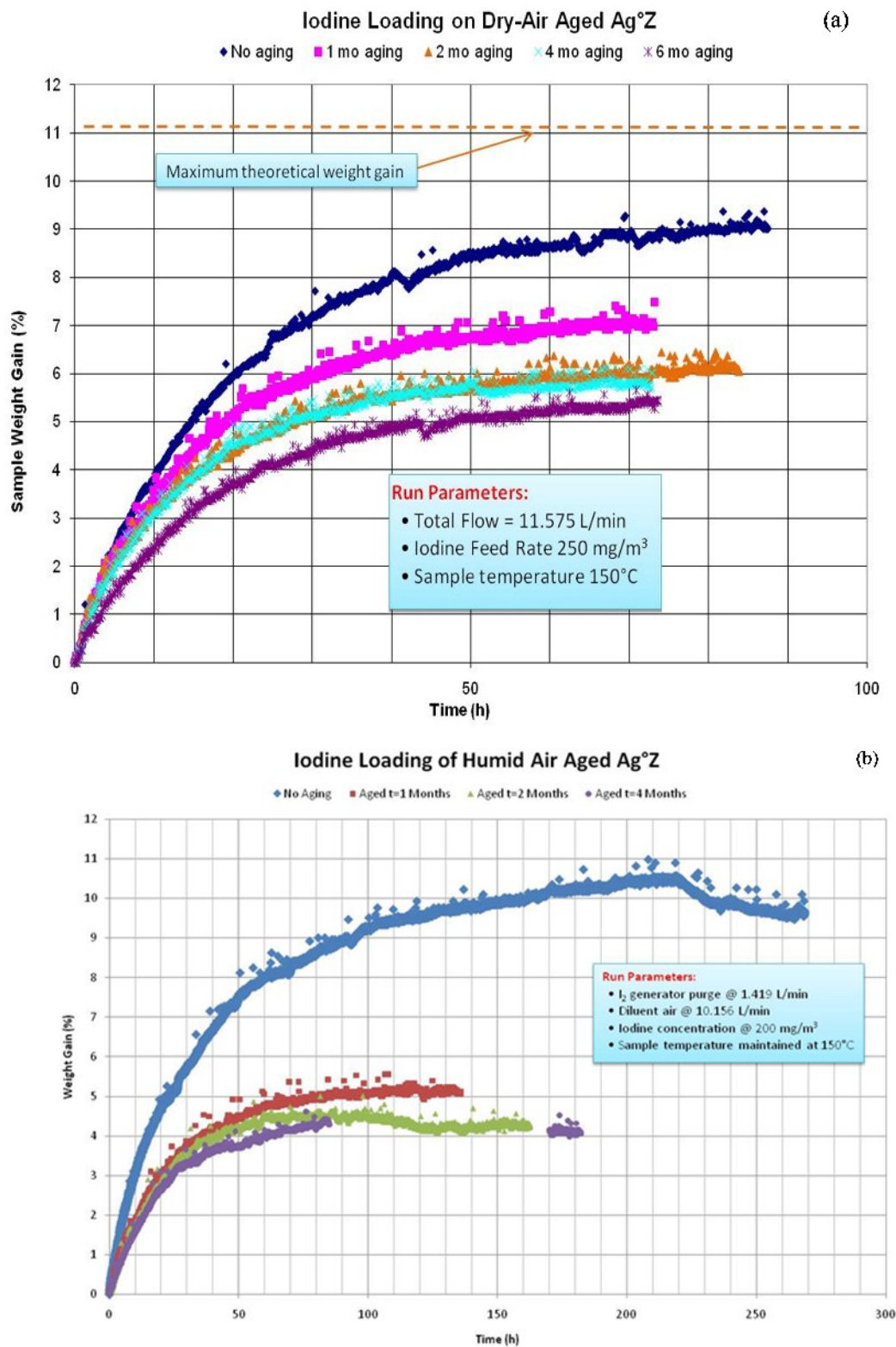


Fig. 4. Iodine loading curves for a) dry-air aged Ag<sup>o</sup>Z; b) humid-air aged Ag<sup>o</sup>Z.

## Storage of Ag<sup>0</sup>Z

During the numerous initial iodine loading runs, there appeared to be a possibility of a slight reduction in iodine loading performance with a lengthy storage time (i.e., the time between Ag<sup>0</sup>Z reduction and the time that the Ag<sup>0</sup>Z was loaded with iodine). The storage method for Ag<sup>0</sup>Z during the first 9 months (approximate) of iodine loading runs was to seal the hydrogen-reduced AgZ into a closed plastic container. Initially no effort was made to provide a dry or inert storage atmosphere for the Ag<sup>0</sup>Z. The Ag<sup>0</sup>Z was simply stored under whatever laboratory ambient atmospheric conditions existed at the time the container was sealed. The storage containers were opened numerous times to extract samples for analytical analysis, iodine loading runs, and other miscellaneous tests. The slight reduction in performance was first noted when an effort was made to repeat previous iodine loading runs using the same Ag<sup>0</sup>Z used in earlier runs.

A screening experiment was then performed to show the impact of storing Ag<sup>0</sup>Z in air on iodine loading and to explore the impact of re-reducing aged stored Ag<sup>0</sup>Z. The results, shown in Figure 5, indicate a slight, but notable, reduction in performance in iodine loading as a result of storage of Ag<sup>0</sup>Z for 3 months and a modest improvement in iodine loading from re-reducing the Ag<sup>0</sup>Z prior to use. All Ag<sup>0</sup>Z samples for subsequent tests were stored under an argon blanket.

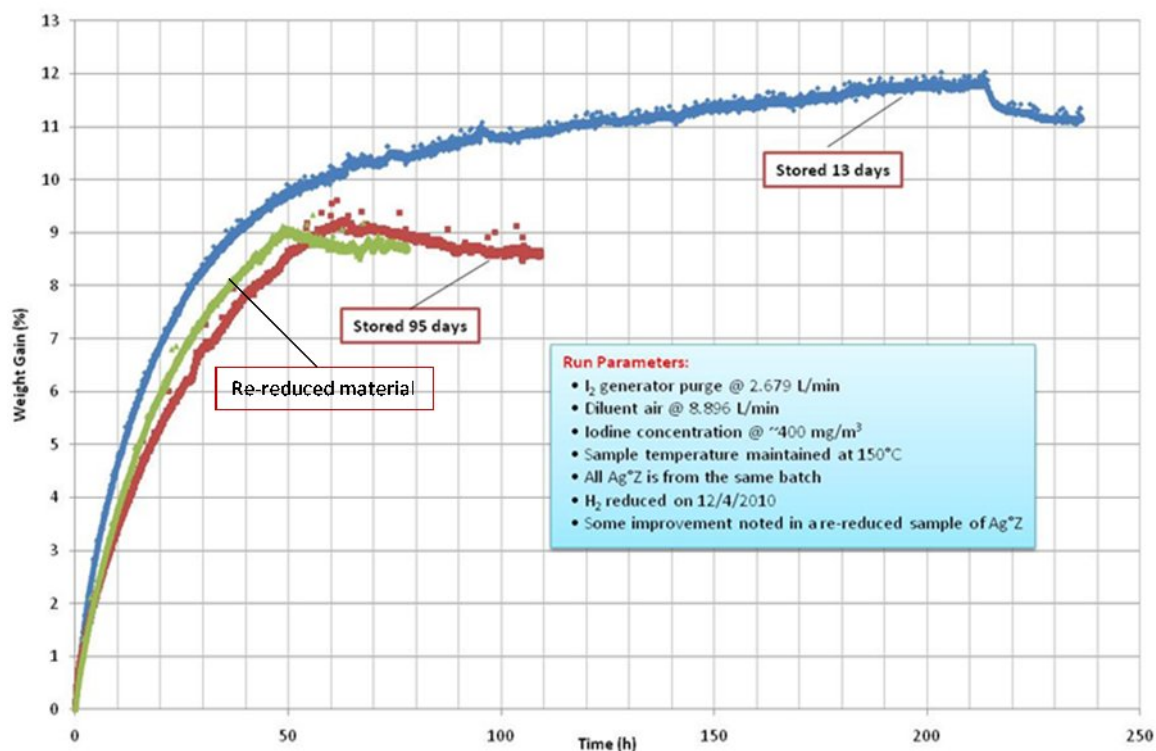


Fig. 5. Effect of storage time on reduced Ag<sup>0</sup>Z and re-reducing Ag<sup>0</sup>Z prior to iodine loading.

## Materials Analyses

Analyses by x-ray diffraction (XRD) and scanning electron microscope (SEM) techniques were performed on the AgZ as it was received from the vendor, as well as the freshly reduced material and aged material. Very little elemental silver ( $\text{Ag}^0$ ) was found in the as-received AgZ (0.3 wt%). After hydrogen reduction, the AgZ contained approximately 5 wt% elemental Ag, i.e. over 50% of the available 9.5 wt% Ag was estimated to have been converted to  $\text{Ag}^0$ . Because of the complex matrix of the zeolites, only semi-quantitative analysis was possible.

After 2 months of aging in dry air, the  $\text{Ag}^0$  content was reduced from 5 wt% to about 1.3 wt%. The quantity of  $\text{Ag}^0$  in the yellow-green samples was noted to be somewhat less than the  $\text{Ag}^0$  in the charcoal colored sample but was within experimental variations of the analyses. The form into which the  $\text{Ag}^0$  was converted was not determined.

A plot of the XRD spectra obtained for reduced material and material aged for 1, 2, and 4 months is shown in Figure 6. It illustrates that the freshly reduced natural  $\text{Ag}^0\text{Z}$  contains the largest quantity of elemental silver and that aging decreases the quantity of elemental silver. Calculated concentrations of elemental silver based on semi-quantitative analysis appear to have been reduced by a factor of 2 to 3 for 4 month aged  $\text{Ag}^0\text{Z}$  compared to the freshly reduced  $\text{Ag}^0\text{Z}$ . Photomicrographs in Figure 7 show clear changes in the general morphology as the materials ages. The impacts of these changes on the iodine capture performance are currently unknown.

## CONCLUSIONS

Because of its high acid resistance, a AgZ sorbent has been selected specifically for application in treating off-gas streams containing iodine. While extensive tests have been conducted in the United States on a form of this sorbent, the specific material previously tested is no longer commercially available and similar materials are currently being evaluated. As part of this evaluation, tests were conducted to determine the iodine sorption properties of this replacement media and the effects of long-term (up to 6 months) exposure to simulated off-gas streams. The ultimate goal is to develop an understanding of the fundamental phenomena that controls aging for this material and other zeolites that could be considered for use in off-gas treatment in the future.

The trends in the study results indicate that the amount of elemental silver observed by XRD increases from 0.3 wt% in vendor-supplied AgZ to approximately 5 wt% by reducing the material with hydrogen. The study also concluded that aging decreases the quantity of elemental silver in the material. After 2 months of aging, the  $\text{Ag}^0$  content of an experimental sample was reduced from 5 wt% to about 1.3 wt%. The form into which the elemental silver is converted during aging was not determined.

Experimental tests have been initiated to study how aging of  $\text{Ag}^0\text{Z}$  impacts iodine loading on the zeolite. Loading tests with unaged  $\text{Ag}^0\text{Z}$  resulted in an 81% silver utilization. The loading capacity of iodine on  $\text{Ag}^0\text{Z}$  was reduced with aging in dry air.

Material aged for 6 months in dry air had a 40% reduction in iodine loading capacity.

Under moist-air aging conditions, a significant decrease in the rate and total loading (~45% of theoretical) of iodine uptake can be observed beginning with the shortest aging period (i.e., after 1 month) when compared with the loading curve using  $\text{Ag}^\circ\text{Z}$  with no aging. Increasing exposure time to the humid air used to age the  $\text{Ag}^\circ\text{Z}$  beyond 1 month resulted in a slight additional reduction in capacity to about 35% of theoretical at 2 months. Virtually identical capacity was observed with 4 months of aging. Compared to the non-aged material, the 1 month dry-air aged  $\text{Ag}^\circ\text{Z}$  shows about a 35% reduction (approximate) in iodine loading capacity and the 6 month dry-air aged  $\text{Ag}^\circ\text{Z}$  shows about a 50% reduction.

These studies generated several questions that will be addressed in future tests. They include the following: Is there indeed degradation over time (in storage) in the iodine adsorption performance of  $\text{Ag}^\circ\text{Z}$ ? Once reduced, how should the  $\text{Ag}^\circ\text{Z}$  be stored—under a hydrogen atmosphere, an inert atmosphere, a desiccant, or some other method or combination of methods? Does  $\text{Ag}^\circ\text{Z}$  have a “shelf life” that must be considered after receipt from a vendor and before use in a plant? Also, how should a column of  $\text{Ag}^\circ\text{Z}$  be stored offline in a processing plant (parallel column) arrangement? Future tests will also include increasingly challenging aging environments such as acid vapor conditions.

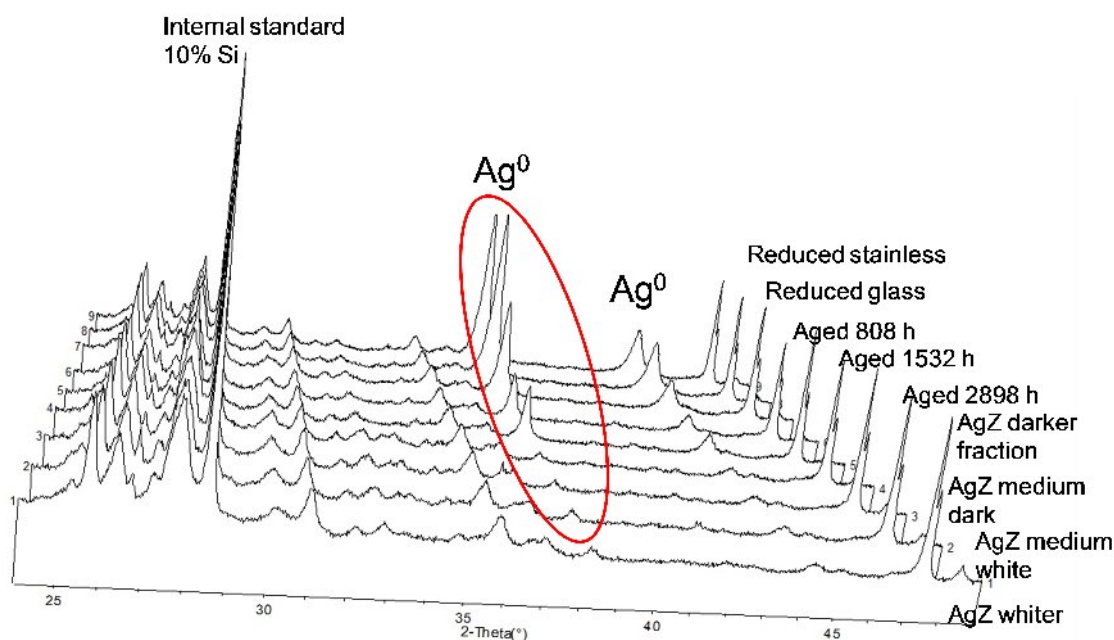


Fig. 6. XRD spectra obtained on different mordenite specimens.



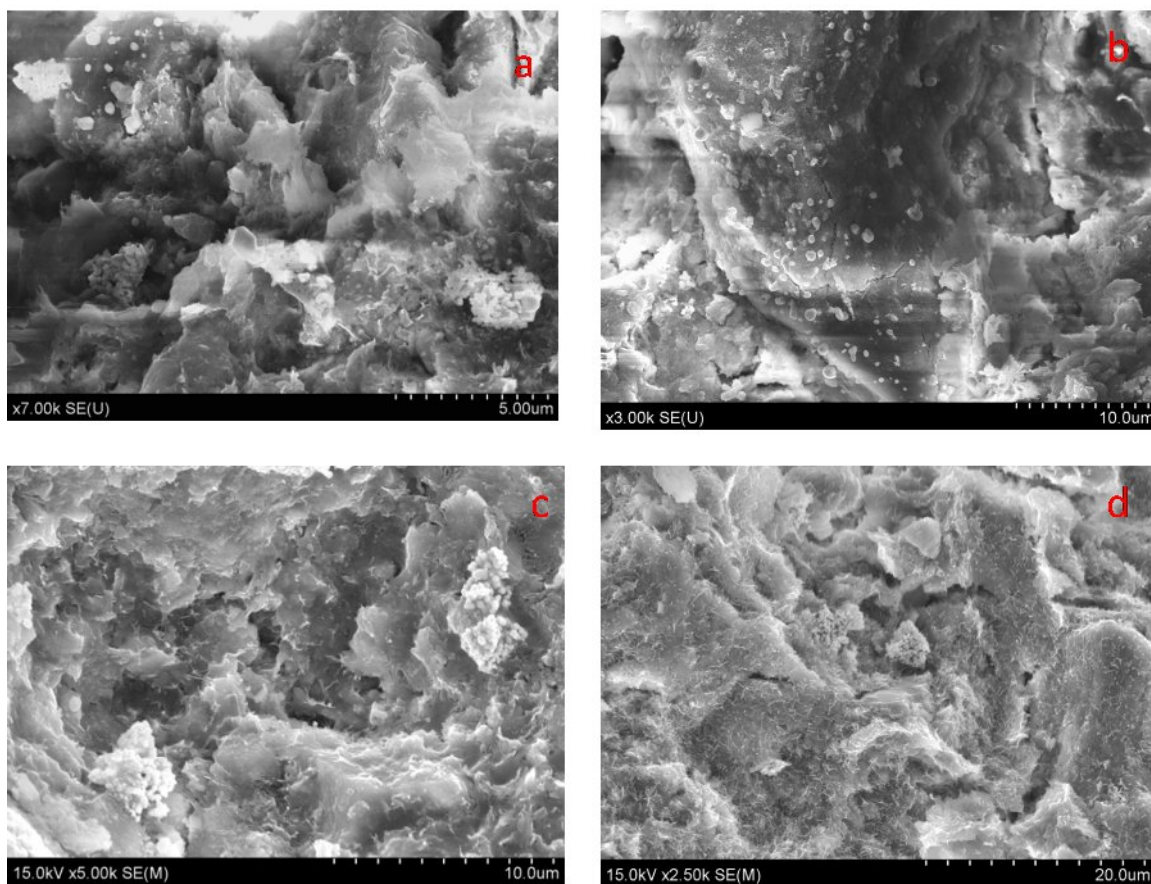


Fig. 7. Photomicrographs of aging Ag°Z: a) no aging; b) 1 month of aging; c) 2 months of aging; d) 4 months of aging.

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