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Demonstrate Scale-up Procedure for Glass Composite Material (GCM) for Incorporation of Iodine Loaded AgZ

Fuel Cycle Research & Development

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Materials Recovery and Waste Form Development

Prepared for
U.S. Department of Energy
Office of Nuclear Energy –
Separations Working Group

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Sandia National Laboratories

U.S. DEPARTMENT OF
ENERGY

Fabrication of Large Size GCM Waste Forms Containing I_2 or CH_3I Loaded AgI-MOR

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SUMMARY

Two large size Glass Composite Material (GCM) waste forms containing AgI-MOR were fabricated. One contained methyl iodide-loaded AgI-MOR that was received from Idaho National Laboratory (INL, Test 5, Beds 1 – 3) and the other contained iodine vapor loaded AgI-MOR that was received from Oak Ridge National Laboratory (ORNL, SHB 2/9/15). The composition for each GCM was 20 wt% AgI-MOR and 80 wt% Ferro EG2922 low sintering temperature glass along with enough added silver flake to prevent any I_2 loss during the firing process. The silver flake amounts were 1.2 wt% for the GCM with the INL AgI-MOR and 3 wt% for the GCM contained the ORNL AgI-MOR. The GCMs, nominally 100 g, were first uniaxially pressed to 6.35 cm (2.5 inch) diameter disks then cold isostatically pressed, before firing in air to 550°C for 1hr. They were cooled slowly (1°C/min) from the firing temperature to avoid any cracking due to temperature gradients. The final GCMs were ~5 cm in diameter (~2 inches) and non-porous with densities of ~4.2 g/cm^3. X-ray diffraction indicated that they consisted of the amorphous glass phase with small amounts of mordenite and AgI. Furthermore, the presence of the AgI was confirmed by X-ray fluorescence. Methodology for the scaled up production of GCMs to 6 inch diameter or larger is also presented.

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1. ABBREVIATIONS AND ACRONYMS

Ag	Silver
Ag°	Silver metal
AgI	Silver Iodine
Bi	Bismuth
°C	Degrees Celsius
FCRD	Fuel Cycle Research and Development
FY	Fiscal Year
g	Gram
GCM	Glass Composite Materials (waste form)
hr	Hour
I	Iodine
I_2	Iodine (gas)
INL	Idaho National Laboratory
kg	kilogram
K_{sp}	Solubility Product
ksi	kilopounds per square inch
L	Liter
min	Minute
MOR	Mordenite Zeolite
MS	Mass Spectroscopy
ORNL	Oak Ridge National Laboratory
Si	Silicon
SNL	Sandia National Laboratories
TGA	Thermo Gravimetric Analysis
XRD	X-ray Diffraction
XRF	X-ray Fluorescence

2. INTRODUCTION

In the spent nuclear fuel reprocessing procedures under consideration by the US Department of Energy, off-gas containing $^{129}I_2$ and organic-^{129}I vapors is passed through a bed of a silver-exchanged Ag°-Mordenite (Ag°-MOR), that selectively captures the ^{129}I to form AgI–MOR.[1,2] The majority of the captured ^{129}I reacts with the silver in the Ag°-MOR to form AgI, while the rest is either chemically adsorbed, or passed downstream. Silver iodide (melting point = 558°C and density = 5.675 g/cm^3)[3] has a low solubility in water, 3 x 10^{-6} g/L at 20°C[3] (corresponding to a K_{sp} of $10^{-16.1}$), which makes silver a good candidate for the capture of ^{129}I. For safe storage, the AgI-MOR must be incorporated into a dense, durable solid waste form, so that the release of ^{129}I is inhibited with time or environmental contact. Because of their flexibility of synthesis and chemical durability, low temperature sintering glasses are being examined for the encapsulation of AgI-MOR iodine capture materials to form Glass Composite Materials (GCMs), for subsequent use as waste forms.[4-7] They are easier and less expensive to prepare than conventional ceramics but also offer high durability, as shown in our recent work.[8]

In our previous reports[5-8] on I_2-loaded Ag°-MOR, we identified a commercially available low-temperature sintering glass powder based on silicon and bismuth oxides that sinters to full density at 550°C and has excellent durability. To prevent surface/bulk chemisorbed I_2 vapor from escaping from the zeolite during GCM sintering, additional Ag flake was added to the GCM mixture to capture the desorbing I_2 vapor. The optimized amount of Ag flake necessary to prevent loss of I_2 during GCM thermal processing in air (of a GCM containing I_2 vapor loaded Ag°-MOR) was studied in detail in FY14[9] and of CH_3I loaded Ag°-MOR in FY15.[10] We have found that additional silver is necessary even when there is an excess of silver to iodine, on a molar basis. This process was highly successful as the excess Ag captured the adsorbed iodine and formed AgI. As a note, little to no Ag flake is necessary if the GCM is sintered in inert atmosphere,[10] an option for future scaleup studies.

The purpose of the present study is to demonstrate the fabrication of larger size GCM waste forms containing 20 wt% AgI-MOR loaded with both I_2 and CH_3I, respectively. Specifically, the goal is to scale up to 100 g with a 5 cm diameter as a first step towards scaling to a size optimal for production. The two AgI-MOR materials used included: (1) CH_3I loaded sample prepared at Idaho National Laboratories (Test 5), and (2) an I_2 loaded sample prepared at Oak Ridge National Laboratory (SHB 2/9/15).

3. EXPERIMENTAL METHODS

3.1 Starting Materials

Three samples of CH_3-I loaded AgI-MOR were received from INL, from which a portion was used in experiments in a previous study.[10] The samples were labeled Beds 1, 2 and 3, from Test 5, respectively. The silver mordenite was originally supplied by Molecular Products in an engineered pelletized form (Ionex-Type Ag 900 E16). It contains ~9.5% silver by weight and has a 1/16-in. pellet diameter. Before CH_3-I exposure, the Ag-MOR was heated at 150°C under H_2 flow (3% in N_2) for ~12 hours to reduce the occluded Ag^+ to the metallic state.[1] The remaining material from each test bed was combined to make a GCM of the desired size.

The I_2 loaded AgI-MOR (~20 g) was prepared at ORNL and was labeled “SHB 2/9/15”. It consisted of the same type of Ag-MOR sent to INL for CH_3-I loading. It was exposed to a dry air stream containing 50 ppm I_2 for 5 days, then purged with dry air for 2 days to remove any physisorbed iodine.

The glass powder used to make the GCMs was Ferro EG2922 (Ferro Electronic Packaging Materials, Mayfield Heights, OH), a lead-free bismuth zinc silicate composition. It has a 3 µm average particle size, a density of 5.8 g/cm^3 and a coefficient of thermal expansion of 7.8×10^{-6} / °C. The glass has a composition of: 7.8 wt% ZnO, 63.4 wt% Bi_2O_3, 5.4 wt% Al_2O_3, 23.4 wt% SiO_2. The glass sinters to essentially full density after only 15 min at 550°C and does not crystallize during the sintering process.

The silver flake (<10 µm, Aldrich, Milwaukee, WI) was added to the GCMs to react and fix with any evolved iodine vapor.

3.2 Characterization

Simultaneous thermogravimetric analysis and differential scanning calorimetry (TGA/DSC, SDTQ600, TA Instruments, Newcastle, DE) with mass spectroscopic (MS, Thermo Star™, Pheiffer Vacuum, Inc., Asslar, Germany) off-gas analysis was performed on powder samples of the as-received AgI-MOR materials, the powdered as-received AgI-MOR materials with added Ag flake and on small portions of the pressed GCM samples with various amounts of added Ag flake. The experiments designed to determine the minimum Ag additions were performed on small pieces of the preforms that were heated in air at 5°C/min heating rate up to 550°C for a 1 hr soak, since this is the thermal treatment used to densify the GCMs. The sintered GCMs were examined using powder X-ray diffraction (XRD, Siemens Kristalloflex D 500 diffractometer, Bruker-AXS Inc., Madison, WI) and X-ray fluorescence (XRF, Bruker M4 Tornado Micro-XRF, Bruker-AXS Inc., Madison, WI).

3.3 Determination of Silver Flake Addition

The first step in producing a GCM is to determine how much, if any, additional silver need to be added to the mixture of AgI-MOR and glass powder to prevent the release of any iodine vapor during the subsequent thermal processing. The added silver reacts *in situ* with the escaping iodine to form AgI. For the INL CH_3I-loaded AgI-MOR's (Test 5, Beds 1, 2, and 3), the amounts the silver flake needed were determined earlier.[10] These amounts were: for a 20 wt% AgI-MOR and 80 wt% glass composition, 1.10 wt% Ag (Bed 1), 1.15 wt% Ag (Bed 2) and 1.2 wt% Ag (Bed 3).

For example, figure 1 shows the results of thermogravimetric analysis (TGA) coupled with mass-spectroscopic off-gas analysis (MS) of ground Test 5 Bed 2 AgI-MOR. Iodine vapor started to evolve around 325°C and continued up to 600°C, the maximum temperature in this run. Complimentary data is shown in figure 2 where a piece of pressed GCM compact (20 wt% of Bed 2 AgI-MOR and 80 wt% glass with 1.1 wt% added Ag) is heated to 550°C in air and held for 1 hr, a minor (fraction of 0.1 wt%) small amount of I_2 is detected at the temperature hold start. Therefore, since all three beds were mixed together to have enough material to make the large size GCM, a compromise value of 1.3 wt% Ag flake was determined optimal and added to the mixture (relative to the total AgI-MOR plus glass powder mass).

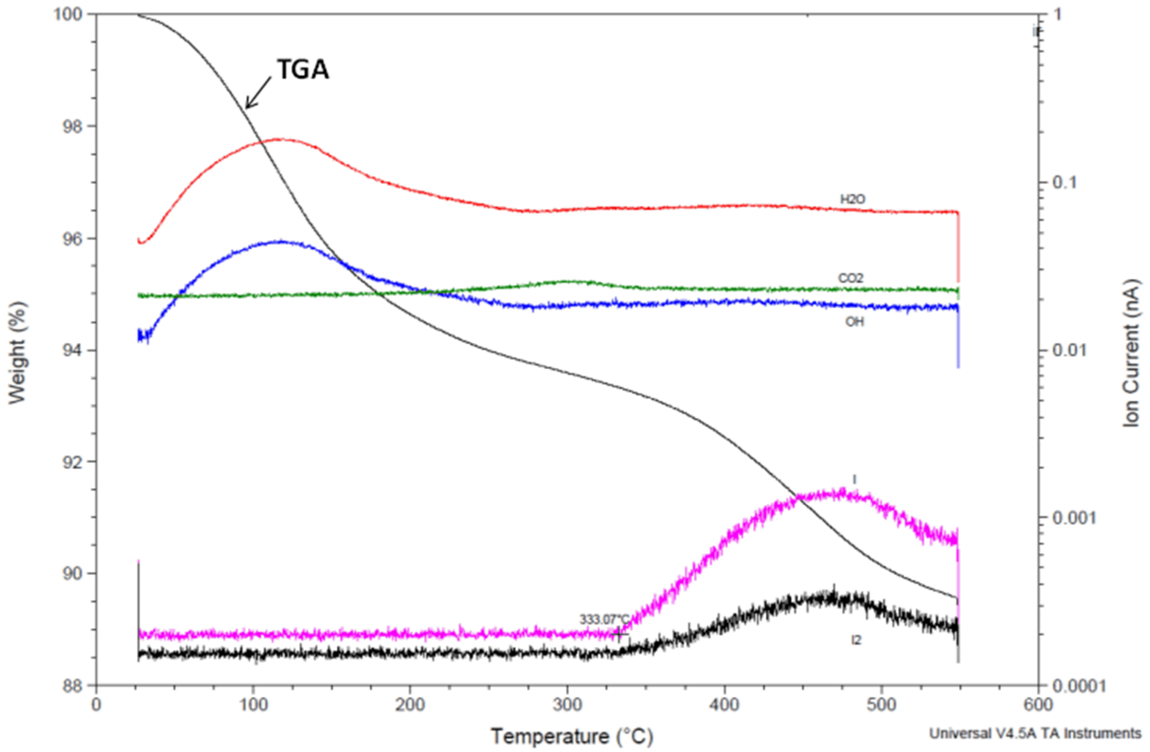

Figure 1. TGA-MS of INL AgI-MOR Test 5 Bed 2 powder heated in air at 5°C/min to 600°C for 1hr.

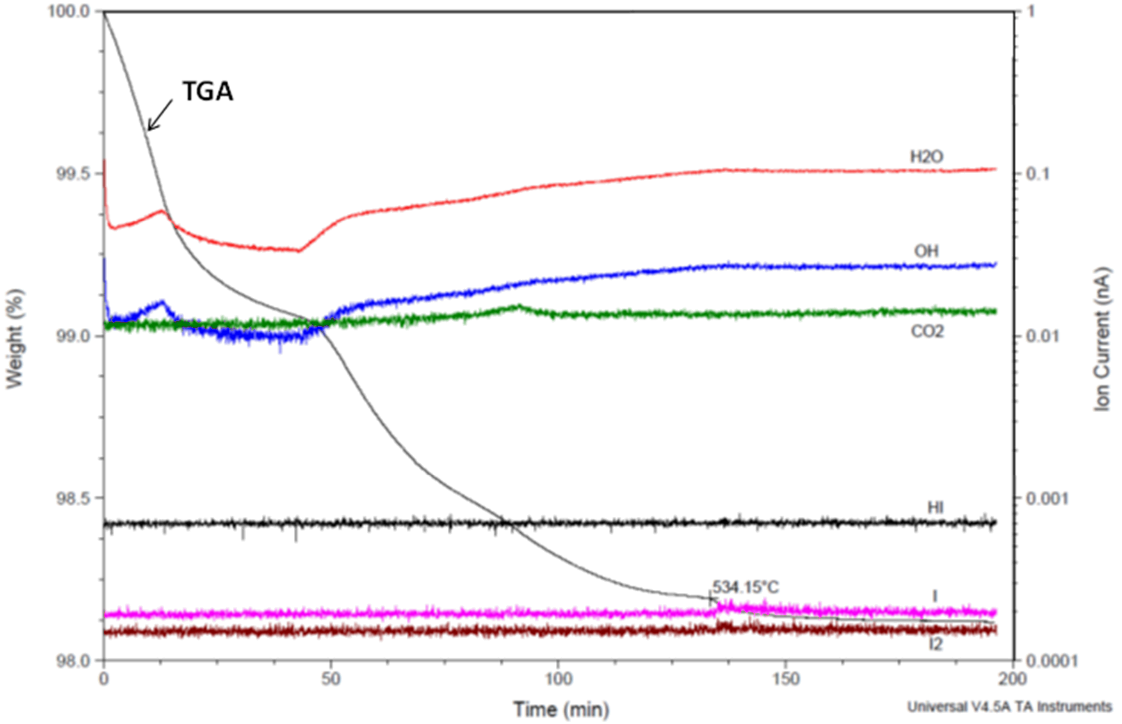

Figure 2. TGA-MS of GCM pellet with 20 wt% INL AgI-MOR powder (Test 5, Bed 2) and 80 wt% glass powder with 1.1 wt% added Ag flake heated in air at 5°C/min to 550°C for 1 hr.

A similar procedure was followed for the I_2 vapor loaded AgI-MOR that was received from ORNL. Figure 3 shows the TGA-MS results for the as-received ground material during heating in air at 5°C/min to 600°C. Iodine vapor was first detected at ~ 200°C and it continued to evolve off up to 600°C. The TGA-MS data in figure 4 shows that when piece of a pressed GCM compact (with 20 wt% of this ORNL AgI-MOR and 80 wt% glass with 2.8 wt% added Ag) is heated to 550°C in air and held for 1 hr, a fraction of 0.1 wt % of iodine is lost, and detected near the start of the temperature hold. Therefore, an optimized addition of 3 wt% silver flake was determined, and added to this scaled up GCM of AgI-MOR and the glass powder.

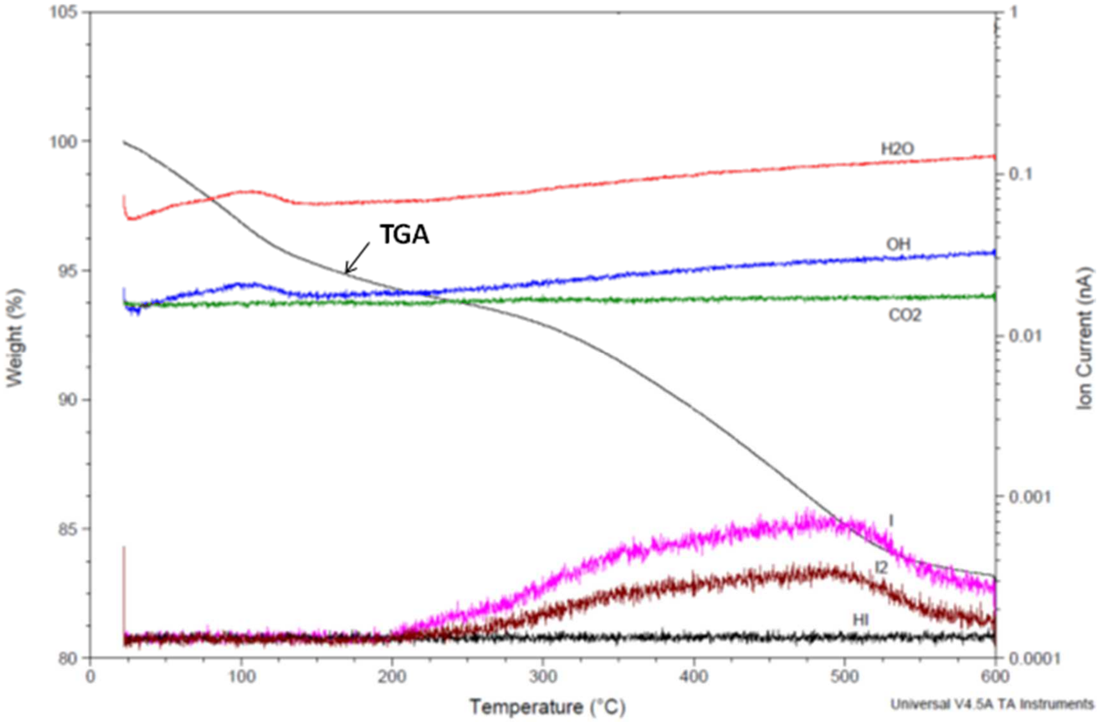

Figure 3. TGA-MS of ORNL AgI-MOR powder heated in air at 5°C/min to 600°C for 1 hr.

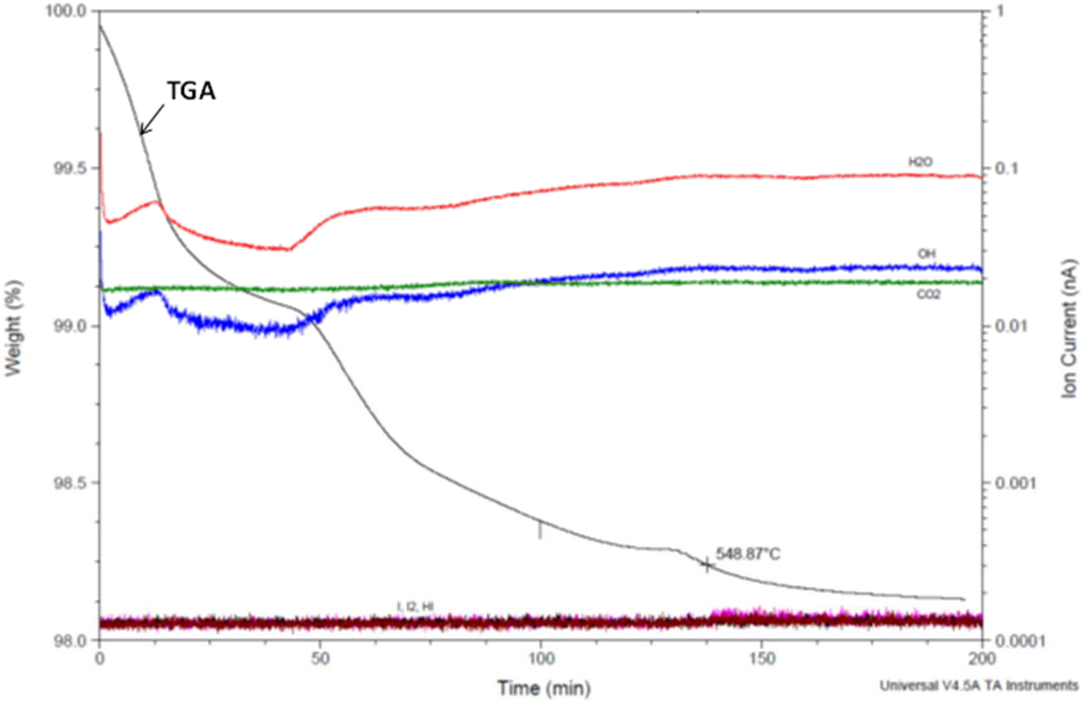

Figure 4. TGA-MS of GCM pellet with 20 wt% ORNL AgI-MOR powder and 80 wt% glass powder with 2.8 wt% added Ag flake heated in air at 5°C/min to 550°C for 1 hr.

3.4 GCM Compact Fabrication

The process used to fabricate the large size GCMs was a scaled up version of the optimized process of making smaller experimental GCMs. The AgI-MOR was first ground to less than 150 µm using an aluminum oxide mortar and pestle to grind and a stainless steel sieve to separate the oversize material for further grinding. After grinding, there was about 18 g of the INL AgI-MOR and 19.5 g of the ORNL AgI-MOR, respectively, to use in large scale GCM fabrication. The ground AgI-MORs were then mixed with the Ferro EG2922 glass powder using four times the mass of the AgI-MOR to give a composition of 20 wt% AgI-MOR and 80 wt% glass. An additional amount of silver flake determined earlier and listed in the previous section (1.2 wt% for the INL AgI-MOR and 3.0 wt% for the ORNL AgI-MOR, respectively) was added to the mixtures along with enough deionized water to facilitate mixing. The mixing was done by hand using a metal spatula. After thorough mixing, the water was allowed to evaporate and the dried mixture was lightly crushed to less than 600 µm to facilitate die filling. A 6.35 cm (2.5 inch) steel die was then used to press the mixtures into compacts. No binder was added. A pressure of ~7 MPa (~1000 psi) was applied. After ejection from the die, each GCH powder compact was sealed in a plastic bag and then cold isostatically pressed at 280 MPa (40 ksi). Optical images of the pressed powder GCM compacts are presented in figure 5. As the figure indicates, both were just over 1 cm in thickness.

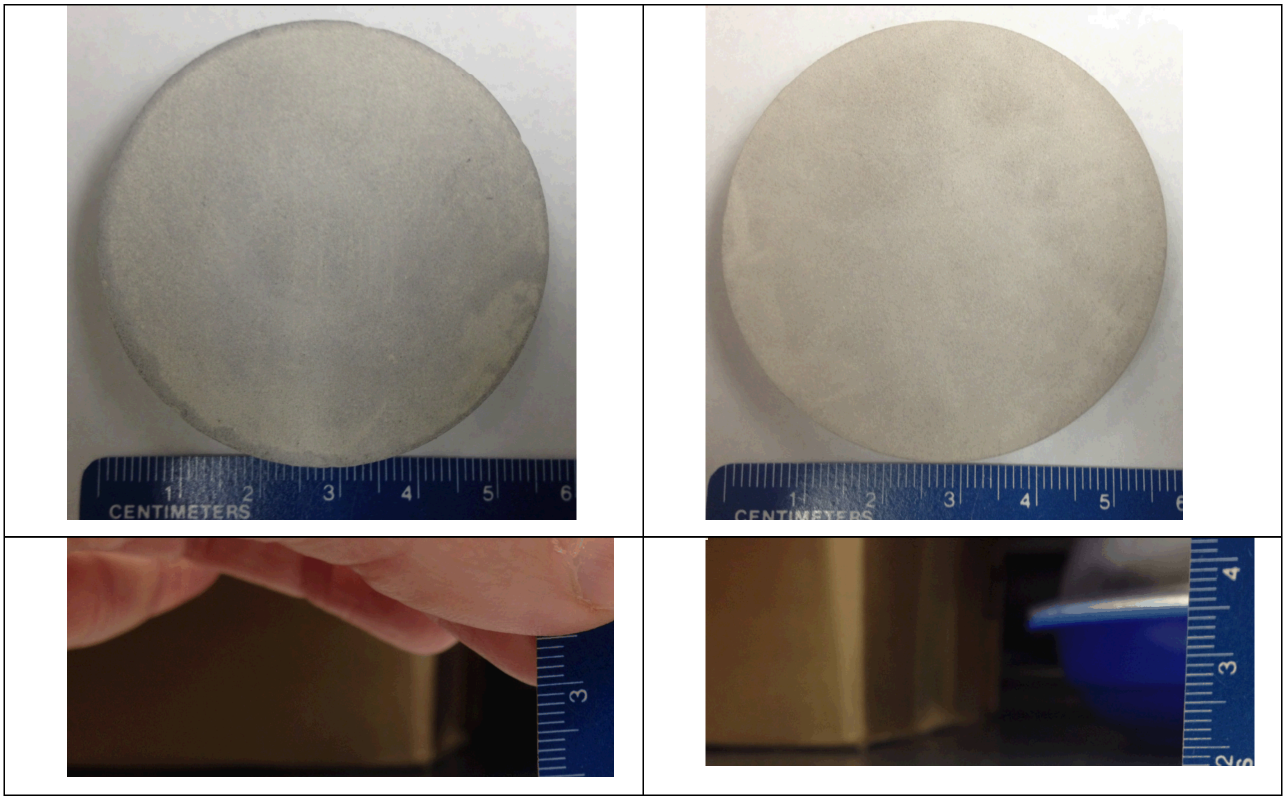

Figure 5. Optical images of the pressed, unfired GCMs. The GCM containing 20 wt% INL CH_3I-loaded AgI-MOR is on the left and the GCM containing 20 wt% ORNL I_2-loaded AgI-MOR is on the right.

The GCM pressed compacts were then placed on a boron nitride (BN) coated alumina plate for sintering; BN was present to prevent adhesion between the GCM and the alumina setter plate. The sintering was done in air with a heating rate of 5°C/min to 550°C with a 1 hr hold before cooling at 1°C/min. A slower than usual cooling rate was used to ensure that no GCM cracking would occur due to the formation of temperature gradients in the GCMs.

4. CHARACTERIZATION OF FIRED GCM COMPACTS

Post sintering, the resultant GCMs were 4.9 cm in diameter for a linear shrinkage of 22.8%. The GCM made using the INL AgI-MOR was 89.4 g in mass, 1.1 cm thick, with a density of 4.23 g/cm^3. The GCM made using the ORNL AgI-MOR was 96.8 g in mass, 1.21 cm thick for a density of 4.28 g/cm^3. Optical images of the final GCMs are presented in figure 6. As can be seen, both scaled up GCMs were flat and uniform, without cracking or warping. Surface water drop tests further indicated that the sintered GCMs do not have any open porosity, see bottom images of figure 6.

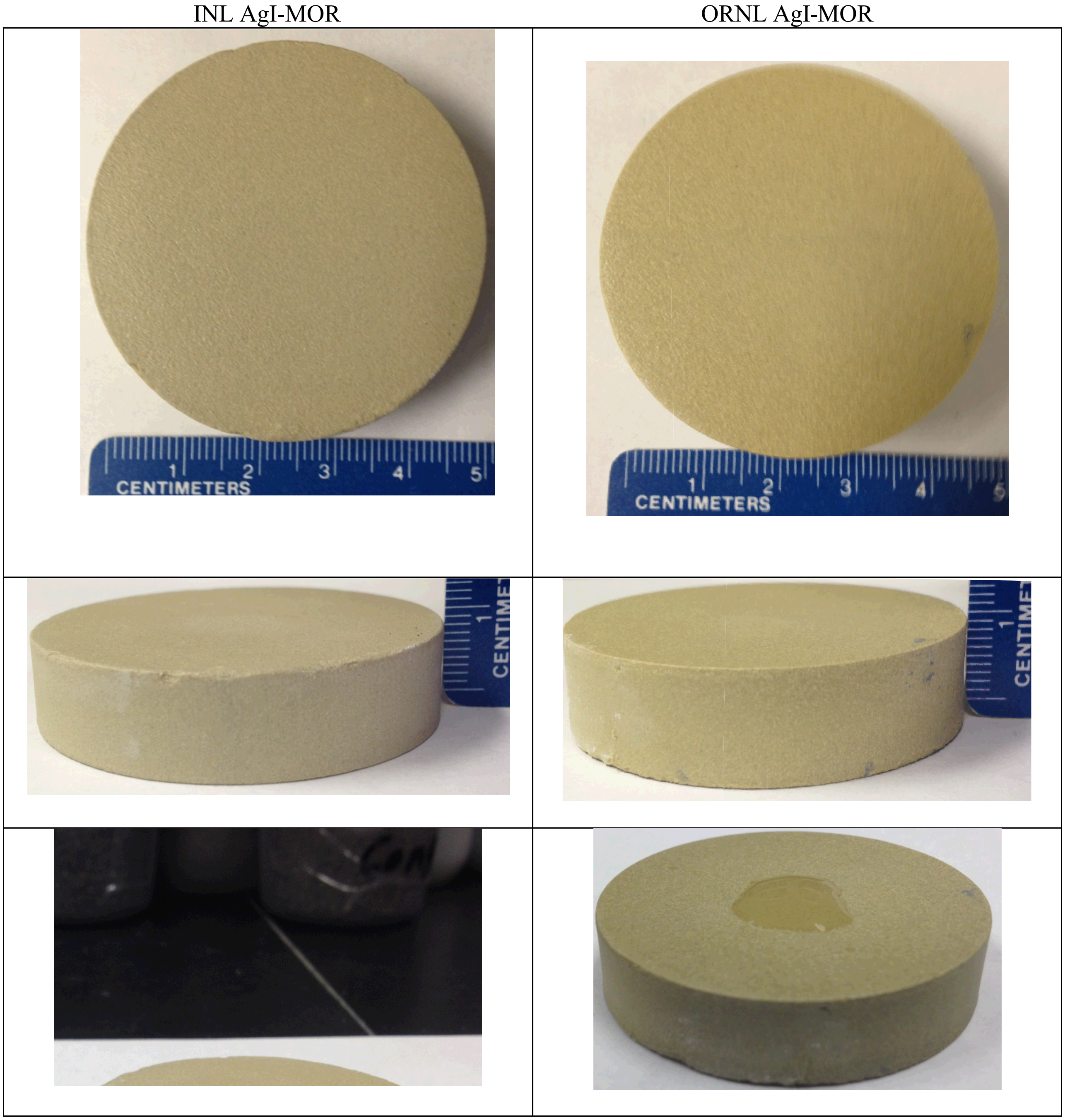

Figure 6. Optical images of the fired GCMs. (Left Column) GCM containing 20 wt% INL CH_3I-loaded AgI-MOR; (Right Column) GCM containing 20 wt% ORNL I_2-loaded AgI-MOR.

4.1 XRD and XRF

A small portion of the surface of each GCM was analyzed using XRD. The results are presented in figure 7; they indicate that the glass is essentially amorphous after the sintering process as evidenced by the broad peak between 20° and 35° 2*Θ*. Small crystalline peaks for AgI (at ~22, 24 and 75° 2*Θ*), mordenite (at ~10° 2*Θ*) and possibly eulytite (at ~32° 2*Θ*) are present. The XRF spectra for each GCM ("A" for INL AgI-MOR, "B" for ORNL AgI-MOR) are seen in figure 8. Peaks are present as expected for the elements in the glass (Bi, Zn, Si, Al) and the mordenite (Si, Al, Fe) as well as for Ag and I. The iodine peaks are slightly higher for the ORNL AgI-MOR containing GCM, consistent with its higher initial iodine loading level that required a larger silver addition.

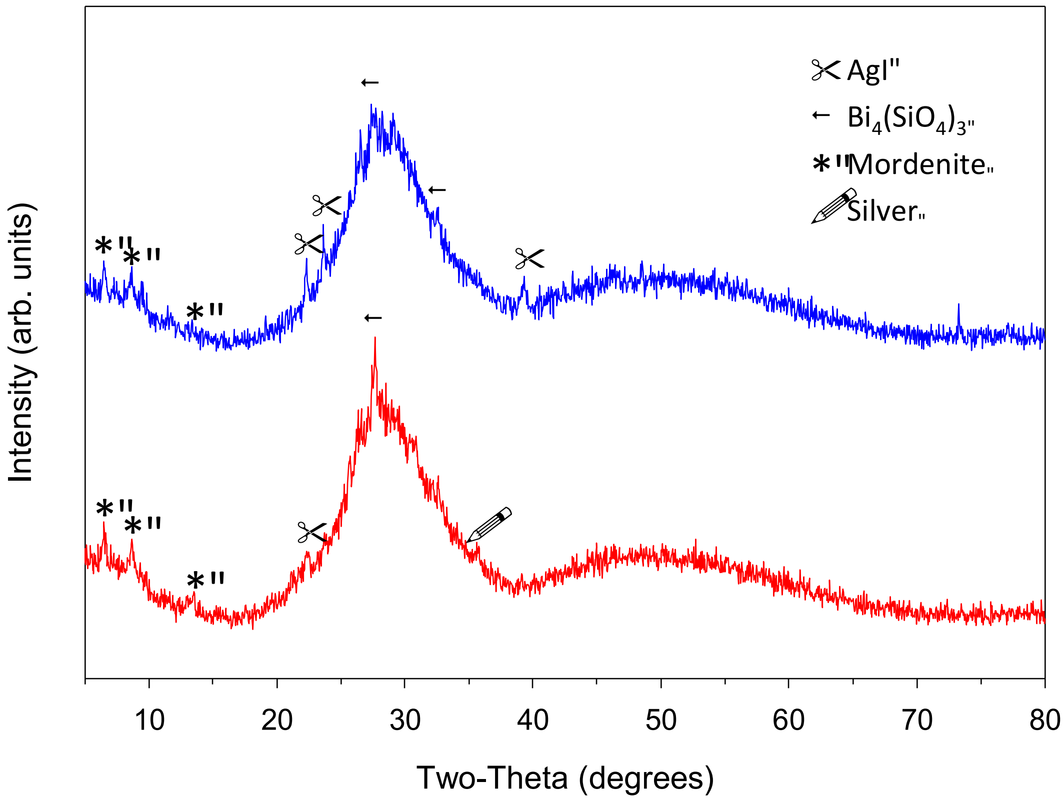

Figure 7. XRD patterns for the sintered GCM compacts.
(Top, blue data) INL AgI-MOR GCM; (Bottom, red data) ORNL AgI-MOR GCM.

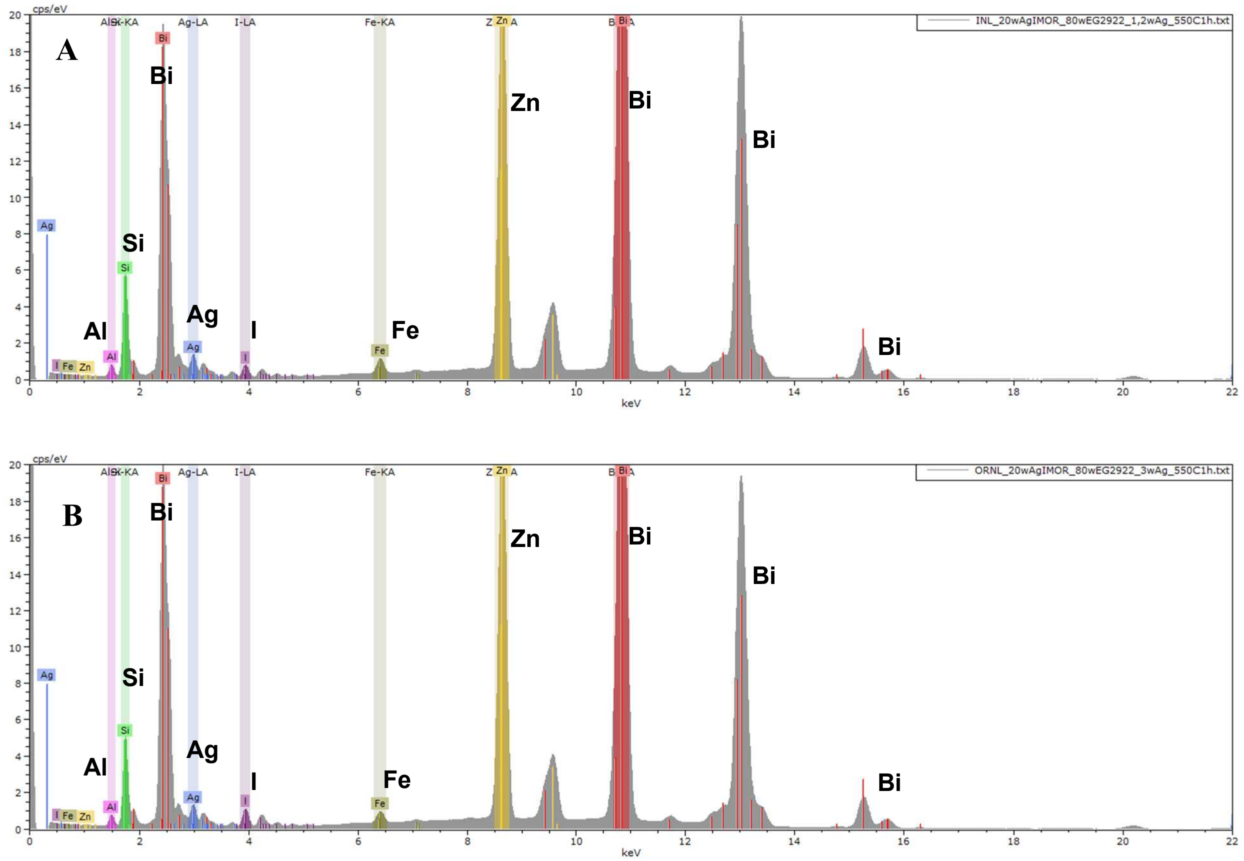

Figure 8. XRF results on the sintered GCM compacts. The top spectrum (A) is for the INL AgI-MOR containing GCM and the bottom (B) is for the ORNL AgI-MOR containing GCM.

4.2 Durability Studies

Originally, durability studies such as PCT-B, MCC-1 and Single Pass Flow Through (SPFT) tests were used to study the durability of the GCM. These studies uncovered a variety of mechanisms at play in the degradation of the GCM and its components. This resulted in a re-evaluation of the durability testing and methodologies for composite waste forms in this campaign. With the approval and guidance of DOE/NE, we have suspended durability studies on the GCM until new guidance is determined. It is for this reason we do not have any durability studies on the scaled up GCMs in this milestone report.

As a replacement for the durability studies in this milestone, we have explored methodologies and costs associated with scaling up the GCM to much larger, industrial sizes. Initial information from this analysis is presented in section 5, below.

5. FUTURE PLANS FOR LARGE SCALE GCM FABRICATION

While the results of the present study have demonstrated the ability to scale up the size the GCM to ~5 cm (~2 inch) in diameter, further scale-up will be required to make this approach viable in a production environment. Currently, the scaled up GCMs were approximately 100 g in mass. It is envisioned that for this approach to be viable, the mass of the GCM should be at least 1 kg and ideally up to 10 kg. The graph in figure 9 shows the predicted GCM mass as a function of diameter, assuming the same cylindrical shape and a ratio of diameter to thickness of 1:4. The graph indicates that a diameter of 12.5 cm (~5 inches) will result in a waste form with a mass ~1.6 kg, whereas a 15 cm (~6 inch) GCM will be over 2.8 kg. A 20 cm (~8 inch) diameter would weigh ~ 7 kg and a 25 cm (~10 inch) diameter will be > 13 kg. Therefore, we envision that a GCM with a diameter in the 15 to 25 cm (6 to 10 inch) range will be required.

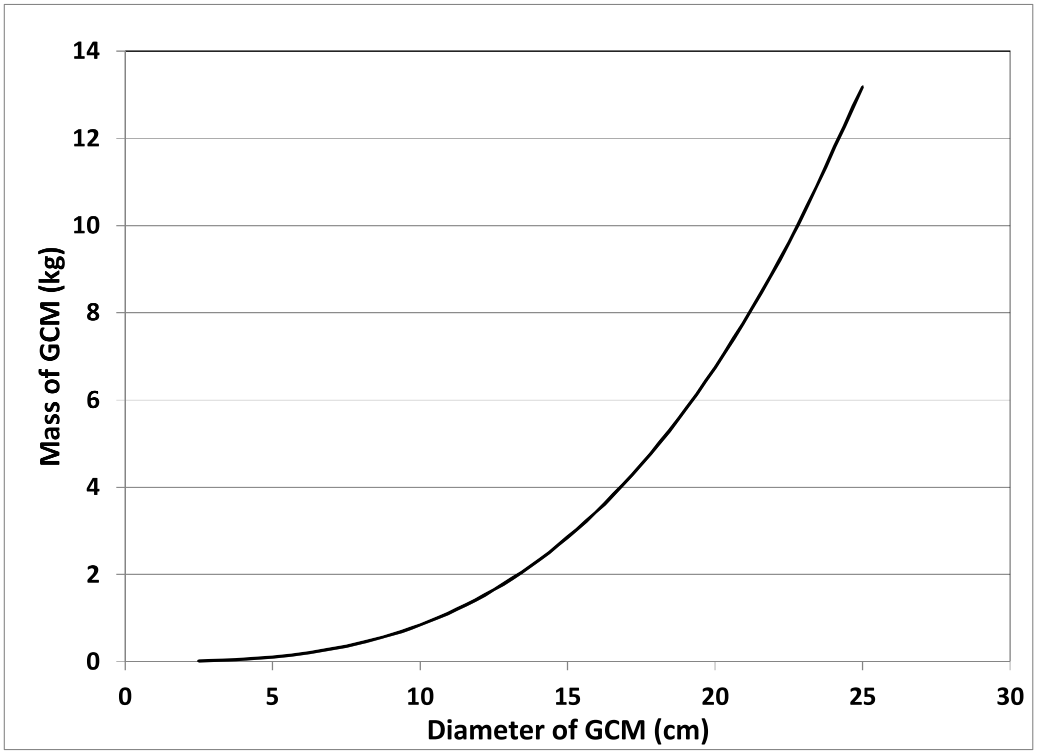

Figure 9. Scale up predictions: GCM mass as a function of diameter for 20 wt% AgI-MOR loading and a constant ratio of thickness to diameter of 1:4.

5.1 Cold Uniaxial Pressing

One approach to further scale-up is to use the same fabrication process, cold uniaxial pressing of powder in a steel die set (die and rams), followed by cold isostatic pressing and then pressure-less sintering, only utilizing a larger die set to press the powder compact. Because ~

23% linear shrinkage occurs during sintering, the powder compact will have to be pressed to an initial diameter of ~25% larger than the desired final diameter. Thus 25 cm (~10 inch) pressed powder diameter would yield a ~19 cm (~7.5 inch) final sintered diameter GCM. As the diameter of the die set increases, so do its cost, its mass and the load required during pressing of the powder to achieve the same pressure. For example, to press a 20 cm (~8 inch) diameter GCM, a die set weighing over 100 kg (220 lbs) would be required, assuming only a 2.5 cm (1 inch) die wall thickness. This is more of an issue for lab-scale production where pressing is done by hand than in an automated production environment.

Pressure pressing has similar load issues. In the present work, a load of 22.2 kN (5000 lbs) was used to press a (2.5 inch) diameter compact to a pressure of ~7 MPa (~1000 psi). To press a 20 cm (~8 inch) diameter powder compact at the same pressure, a ten times greater load (222 kN or 50,000 lbs) will be required. This is higher than the capability of most lab-scale presses but is easily achievable using industrial hydraulic presses which can go to 200 tons (400,000 lbs) or higher. Producing the powder performs using another technique such as filter pressing of a slurry could also be a possibility. Also, large size cold isostatic presses are commercially available whereas lab scale ones, such as the one used in this work which has a 20 cm (8 inch) chamber, may not be able to accommodate large diameter compacts.

Another concern with scaling up to larger sizes is the increased potential for temperature gradients in the GCM during cooling from the sintering temperature to generate stresses that could cause the GCM to crack due to its brittle nature. To avoid cracking, cooling would have to be very slow. Models exist that would be helpful in predicting the maximum cooling rate for a given size GCM. Obviously, having to cool slowly will impact the total production time. A cooling rate of 1°C/min was used in this work, which translates to a cooling time on the order of 10 hr. This allows one furnace run per day in our lab. If a rate of 0.1°C/min is necessary, then only one run per week will be possible. Therefore, using a large furnace that could sinter many GCMs simultaneously would be advantageous.

5.2 Hot Pressing

Another approach will be to hot-press the GCMs. There are two types of hot-pressing: uniaxial and isostatic. An advantage to the uniaxial approach is not needing to press a powder compact preform. This is in contrast to the methods for pressure-less sintering and hot-isostatic pressing; powder is placed directly in the hot-pressing die with no metal can required as is needed for hot-isostatic pressing. Uniaxial hot-presses with 25 cm (10 inch) capability are commercially available. If larger GCMs are required, hot isostatic pressing could be used.

5.3 Costs

Both types of hot presses can be expensive. The costs originate from their components allowing for operations at much higher temperatures (2000°C) and pressures than would be required for this application. One disadvantage to uniaxial hot-pressing is that only one GCM could be processed at a time in a press, as opposed to being able to pressure-less sinter multiple GCMs in a large furnace.

6. CONCLUSIONS

Two scaled up Glass Composite Material (GCM) waste forms containing AgI-MOR were fabricated. One contained methyl iodide-loaded AgI-MOR that was received from Idaho National Laboratory (INL, Test 5, Beds 1 – 3) and the other contained iodine vapor loaded AgI-MOR that was received from Oak Ridge National Laboratory (ORNL, SHB 2/9/15). The composition for each GCM was 20 wt% AgI-MOR and 80 wt% Ferro EG2922 low sintering temperature glass along with enough added silver flake to prevent any I_2 loss during the firing process. Those silver amounts were 1.2 wt% for the GCM with the INL AgI-MOR and 3 wt% for the GCM contained the ORNL AgI-MOR. The GCMs, nominally 100 g, were first uniaxially pressed to 6.35 cm (2.5 inch) diameter disks then cold isostatically pressed before firing in air to 550°C for 1hr. They were cooled slowly (1°C/min) from the firing temperature to avoid any cracking due to temperature gradients. The final GCMs were ~5 cm in diameter (~2 inches) and were non-porous with densities of ~4.2 g/cm^3. X-ray diffraction indicated that they consisted of the amorphous glass phase with mordenite and AgI, whose presence was confirmed also by x-ray fluorescence. Next steps include further scale up of the GCM to industrial sizes, durability studies on the GCM, optimization of composition and sintering conditions based on variables such as N_2 or He versus Air sintering atmosphere (in N_2 or He environments, additional Ag flake is not as necessary as sintering in air).[10]

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