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Off-Gas System Development for Melt-Dilute Treatment of Aluminum Based SNF

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OFF-GAS SYSTEM DEVELOPMENT FOR THE MELT-DILUTE TREATMENT OF ALUMINUM-BASED SPENT NUCLEAR FUEL

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ABSTRACT

The Melt-Dilute treatment for aluminum-based spent nuclear fuel involves melting of the fuel to form a liquid solution of uranium-aluminum at a temperature of approximately 850°C. The containment and capture of the gaseous and volatile fission products potentially released at these temperatures is one of the key elements of the treatment process. Thermodynamic calculations have been performed to estimate the quantity and type of fission products released from a typical melt-dilute process. Cesium is established to be the primary fission product potentially released at these temperatures. A testing program has been systematically implemented studying off-gas releases with non-radioactive surrogates in bench-scale and small-scale tests. The results showed that zeolite 4A is effective at trapping cesium from off-gas streams. Tests on irradiated fuel were conducted to demonstrate the effectiveness of an off-gas system in capturing volatized fission products. The results confirm that zeolite 4A adsorber beds are effective at capturing cesium. The evolution of volatized species from melted fuel and the irradiated testing results of laboratory-scale off-gas capture testing are discussed in this paper.

I. INTRODUCTION

A key component of the melt-dilute process for the treatment of spent nuclear fuels is the off-gas system because of the volatilization of radioactive species during the melting stage. The volatized radionuclides must be removed from the off-gas stream and trapped in components of off-gas system. Although a suitable means of trapping or absorbing the volatile radioactive species, including particulates, has been developed (granular beds of activated alumina and molecular sieves), the efficiency of the trapping process varies with process flow, temperature and bed characteristics. A Optimization of the bed design has been performed using a combined experimental and fundamental modeling approach that predicts trapping and absorption from the off-gas stream. This effort will provide a technical basis to assure safe and efficient system operation of the melt-dilute process and have general applicability to off-gas streams.

The volatilization of fission products during the melting stage of the process (1hr at temperatures from 850°C) primarily constitutes the off-gas in this process. The volatilization and containment of some of the primary species that have previously been studied include krypton, iodine, and cesium [2-6]. All of these species were shown to be released over a wide range of conditions. The fraction released is highly dependent upon atmosphere, fuel burnup, temperature, and fuel composition. A key element in the technology development of the melt-dilute process is the development of the off-gas system. In order to address this element of the technology, an analytical and small-scale experimental program has been undertaken to assess the volatility and capture of species under melt-dilute operating conditions. This phased approach used both surrogate and irradiated melt testing to develop a technical basis for the technology. The portion of the program related to analytical and experimental studies on laboratory scale irradiated fuel are presented here. The results from these studies will be validated during testing of irradiated MTR (Materials Test Reactor) fuel assemblies in a pilot scale facility to be constructed in L-Area at SRS.

II. BACKGROUND - PREVIOUS STUDIES

Two melt-dilute tests using irradiated SNF coupons were performed. The temperature profiles for the first experiment are illustrated in Figure 2. The temperature gradient across the filter bed was extremely high (725 to 325°C) due to its proximity to the furnace. The temperature gradient across the bed was changed in the second test (not shown) to more accurately reflect conditions in the melt-dilute process by translating the furnace with respect to the test train. The temperature varied across the bed in the second test from ~475 to 275°C.

The sample produced from the first test was sectioned and its microstructure is shown in Figure 3. A mixture of aluminum and an Al + UAl_4 eutectic can be observed in this microstructure. The melt-dilute process maintains the composition of the melt at or near, the eutectic composition of Al-13.2 wt. % U in order to minimize the process temperature resulting in such a microstructure.

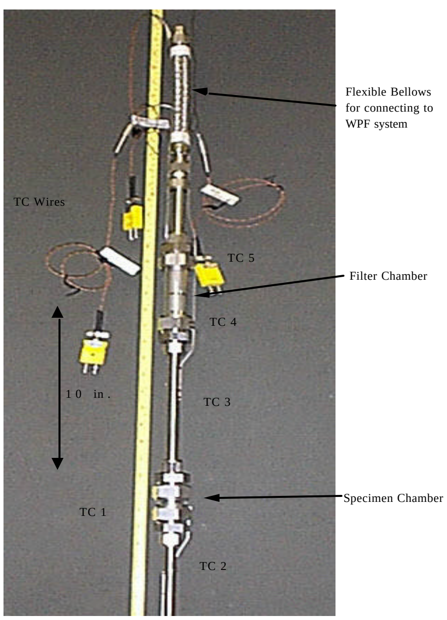

Figure 1: Hot cell set up for a laboratory-scale irradiated melt-dilute experiment

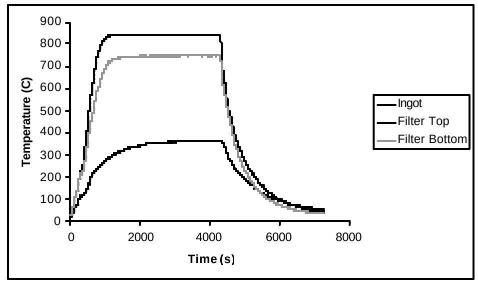

Figure 2: Temperature profile for the duration of the first irradiated melt-dilute experiment

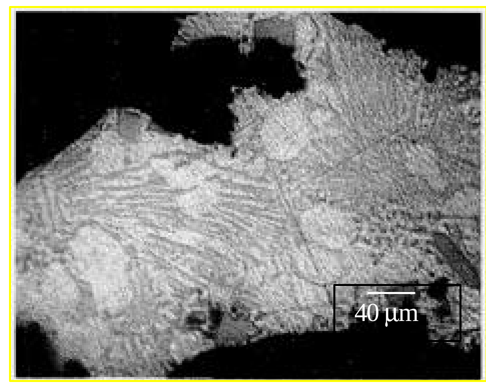

Figure 3: Near eutectic composition of the irradiated coupon from the first test.

A gamma activity scan of the crucible and absorber bed after the test is shown in Figure 4. Also shown in Figure 4 is the portion of the scan resulting from the activity of ^{137}Cs. When the hot cell background is subtracted from the gross activity, as shown by the lower curve in the figure, it was found that 78 % ^{137}Cs remained in the ingot/crucible region. Of the fraction released, ~16% was located at the bottom of the zeolite bed, and 6% plated out on the stainless steel tubing leading from the crucible to the sorption bed. An expanded gamma activity scan of the filter bed confirms the observation that the cesium is concentrated in the first inch of the adsorption bed. Figure 5 shows the energy spectrum of the adsorption bed's gamma scan. The presence of ^{60}Co can be explained from hot cell background and is consistently present in all gamma scans conducted. From this spectrum, it is evident that only ^{137}Cs is present in any reasonable quantity. A small amount of ^{134}Cs was also detected; its intensity is barely visible in this spectrum. No other fission products were detected on the bed and no activity was found in the wet scrubber solution; therefore, complete sorption of volatile cesium occurred on the bed.

The second test was conducted to validate these initial results. Figure 6 shows the gamma activity scan of the test train after the second test and appears entirely consistent with the first test. As is evident, all cesium appears to be trapped in the initial portion of the bed. In addition, the amount of ^{137}Cs released from the ingot is ~21%.

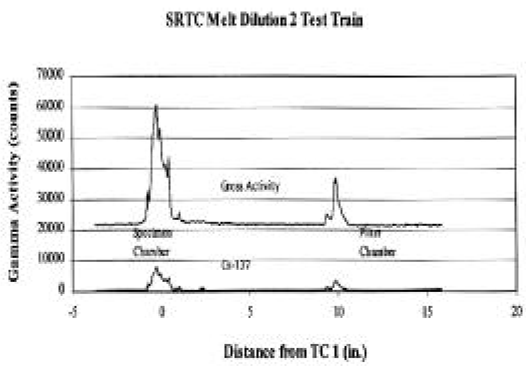

Figure 4: Gamma Radiation Scan of the test train after the first melt-dilute test.

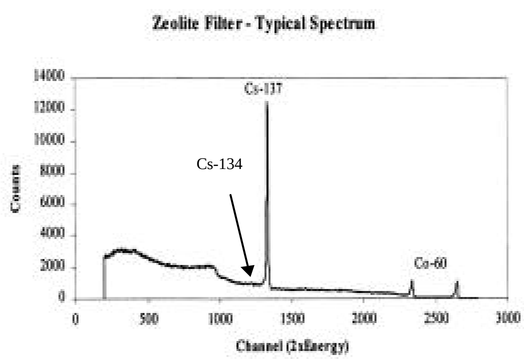

Figure 5: The Gamma Spectrum from the zeolite filter bed after the first melt dilute test.

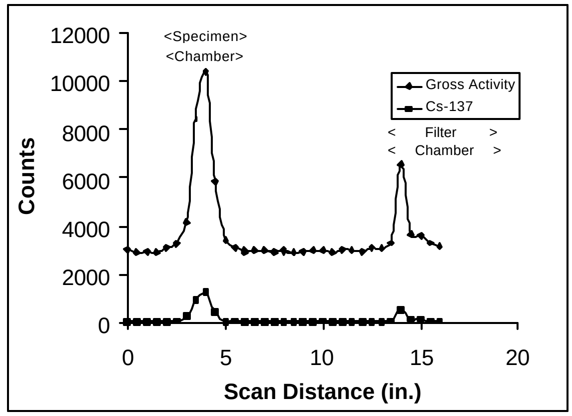

Figure 6: Gamma Radiation Scan of the test train after the second melt-dilute test

III. DESCRIPTION OF THE WORK

L-Area Experimental Facility Development: In support of the L-Area Experimental Facility development at SRS an LEF scale off-gas system was fabricated and installed in the Alpha-Gamma Hot Cell facility at ANL(Figure XX). The intended focus of the experiments for this apparatus was to demonstrate the LEF off-gas design using irradiated fuel coupons with ^{137}Cs loadings equal to or greater than those expected in the LEF melts. A preliminary melt test was conducted at ANL using the SRTC Off-gas apparatus to check out operation and performance parameters. This test utilized 1/16" zeolite-4A beads in the primary off-gas bed which was backed with a secondary zeolite bed and a HEPA filter. The graphite crucible and primary zeolite bed were both preheated to approximately 400°C. The heating cycle for the test was designed to simulate the proposed LEF cycle of heating to 400°C for 30 minutes for drying the fuel followed by heating to 850°C for 1 hour for the treatment step.

Gamma scanning results form this start-up test indicated that some ^{137}Cs in the off-gas stream had made it through the primary and secondary zeolite beds and was captured by the in-line HEP filter. Based on these results and with the LEF goal of allowing no cesium past the primary bed, a new set of experiments was undertaken to determine why the LEF Off-gas bed design had not performed as well as the previous ANL irradiated coupon test which showed all the cesium being trapped in the first inch of zeolite bed. The zeolite bed used in the previous two lab-scale SRTC-ANL irradiated tests was comprised of 1/8 inch diameter beads that were crushed in order to yield a variable particle sizes between #18 and #50 mesh. This was in comparison to the 1/16" zeolite-4A beads used in the LEF Prototype off-gas bed start-up testing at ANL.

As a result of this difference, a series of laboratory tests were developed in SRTC to examine the influence of the following: zeolite particle size, system flow rate, and off-gas bed temperature. These test were conducted using the full-scale LEF Simulator in the Materials Laboratory at SRTC. The LEF Simulator is a 1-to-1 mock-up of the LEF system used to test and optimize system design, prototypic equipment, and operation parameters. In addition, to the experimental activities related to

optimizing the LEF off-gas bed design, fundamental modeling of the off-gas flow, pressure drop, and fluidization were preformed.

IV. RESULTS

SRTC LEF Simulator Testing: The focus of the testing at SRTC using the LEF Simulator was to optimize the performance of the LEF Off-gas bed design by evaluating the impact of flow rate, particle size, and bed temperature on cesium trapping. These test were conducted using stable cesium metal added directly to eutectic uranium-aluminum alloy melts. Off-gas bed efficiency was determined by using a wet scrubber on the outlet of the primary off-gas bed. The data from this series of test is shown in Figure 7. The results in Figure 7 indicate that a mixture or distribution of particles sizes for the zeolite bed media similar to that used in the first two ANL lab-scale irradiated coupon tests (Avg. Bead size 0.037254 in.) is most effective for cesium trapping. This particle distribution was shown to be relatively insensitive to bed temperature and flow rate. Single particle size beds and other particle size beds were outperformed by the ANL distribution. Test with the 1/16" beads confirmed the results from the SRTC-ANL LEF Apparatus start-up test, which showed 1/16" indeed do allow for some cesium to pass through the primary off-gas bed.

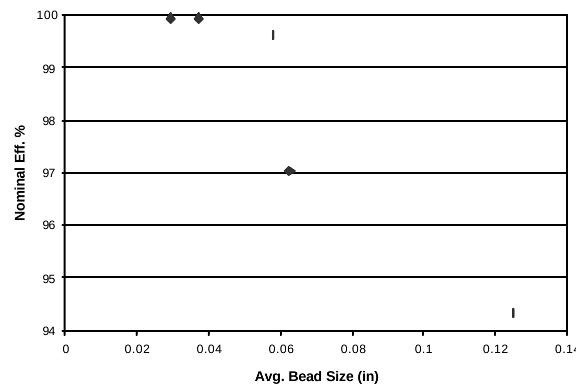

Figure 7. Nominal Efficiency for Off-gas Bed Particle Size

Fundamental Modeling: In order to aid in the optimization of the off-gas bed design, two computational models to determine operating conditions to avoid fluidization of the bed and to predict pressure drop through a porous bed under proposed flow conditions have been developed using empirical correlations for the friction factor found in the literature.(1)

Primary design parameters of the off-gas bed are bed size or volume, gas flowrate, particle size, and bed porosity. Minimum particle size is determined by the flow regime condition of the off-gas bed. Gas flowrate is determined by the operational requirement such as bed loading and minimum pressure drop for a given particle size. As discussed in the earlier section, the fixed bed is desirable because Zeolite particle becomes radioactive as a result of cesium absorption from the gas mixture inside the bed. In this case, maximum allowable limit of gas inlet flow into the off-gas bed is required to avoid fluidizing the Zeolite bed for given particle size and operating conditions.

Steady-state flow conditions were assumed to perform the calculations of the model for the design and operating conditions shown in Table 1. Mean particle size for the ANL test bed was found to be about 0.037. Maximum allowable flowrates required for prevention of the fluidized bed regime are shown in Fig. 8. The results show the maximum allowable airflow is not sensitive to the change of operation temperature for a wide range of Zeolite particle sizes when bed temperature changes from 20°C to 600°C. In the calculations, the particle surface is assumed to be smooth and spherical.

A hydraulic model was used to compute the pressure drop through the 3.068-in absorber bed as shown in Fig. 9. The porous bed is actually composed of three different layers of packed beds with upward flow direction. Bed temperature was assumed to be isothermal and to be room temperature for pressure calculations. Figure 9 schematically shows the detailed geometrical dimensions and bulk flow direction for each of the three packed beds as modeled in the present calculations. Non-uniform porosity effect due to the wall boundary was assumed to be negligible in predicting the pressure loss. In addition, average value of theoretically maximum and minimum porosity was used, that is, Ý = 0.37.

In the calculations of the pressure drop across the bed, three different bed porosities were used to examine the sensitivities of the pressure drop with respect to the change of the porosities from theoretical minimum porosity (0.26) to the average value (0.37). The results for the bed with spherical particles are shown in Fig. 10. The effect of geometrical shape of particle on pressure drop across the bed was evaluated for typical cube-shaped and spherical particles and was shown to be minimal.

Table 1. Off-gas System Design and Operating Conditions

Parameters	Conditions
Flow regime	Fixed bed
Bed temperature	20°C to 600°C
Bed diameter	3" Sch40 (3.068")
Particle diameter	0.004" to 0.125"
Sphericity (see Table 1)	1.0 (spherical)
Particle density	1197.84 (kg/m^3)
Fluid in bed	Air

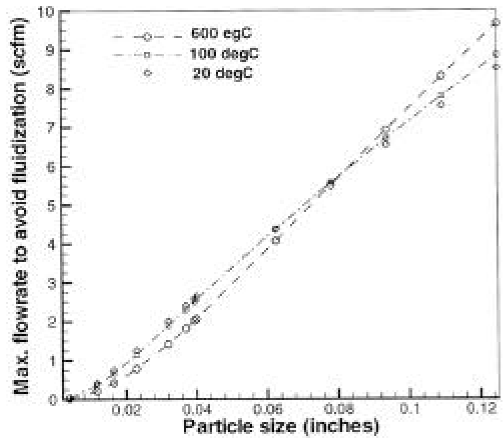

Figure 8. Maximum allowable flowrates required for prevention of the fluidized bed regime

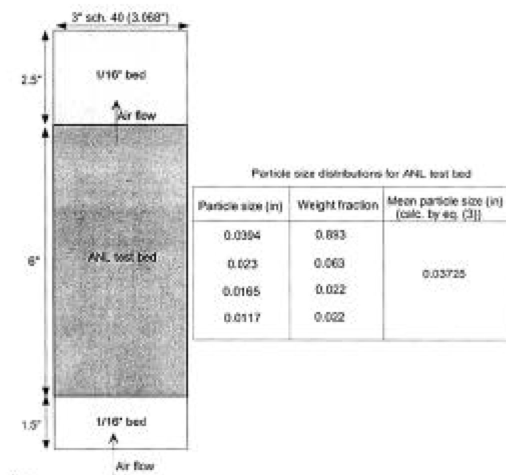

Figure 9. Pressure drop through the 3.068-in absorber bed

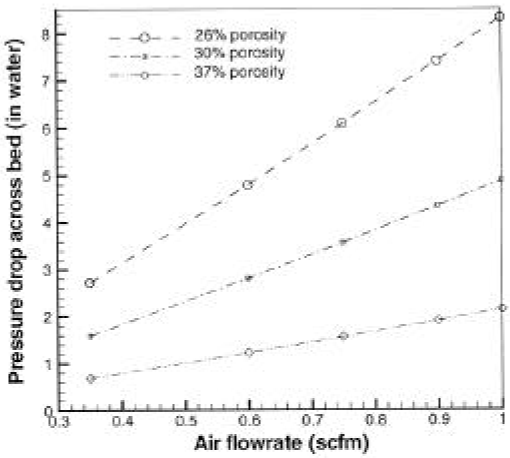

Figure 10. Effect of geometrical shape of particle on pressure drop across the bed

V. CONCLUSIONS

From the SRTC LEF Simulator experiments and the fundamental flow modeling the following observations can be made:

- the experimental tests have identified a mixed particle size bed (#18-#50 mesh) approach as most efficient for trapping cesium
- as the flow rate is decreased the efficiency increases and the role of bed temperature is decreased
- fundamental off-gas flow modeling correlates well with the experimental testing performance.

Based on the results from these studies an off-gas system concept employing dry zeolite 4A adsorber beds containing a distribution of particle sizes in the range of #18-#50 mesh was recommended as the primary cesium-trapping medium for LEF. Validation of this off-gas concept was be conducted during full-scale irradiated testing in the L-Area Experimental Facility, however, due to suspension of the program no experimental data from the LEF is available at this time.

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