Microgamma Scan System for analyzing radial isotopic profiles of irradiated transmutation fuels

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Abstract *– The U. S. Global Nuclear Energy Partnership / Advanced Fuel Cycle Initiative (GNEP/AFCI) is developing metallic transmutation alloys as a fuel form to transmute the long-lived transuranic actinide isotopes contained in spent nuclear fuel into shorter-lived fission products. A microgamma scan system is being developed to analyze the radial distribution of fission products, such as Cs-137, Cs-134, Ru-106, and Zr-95, in irradiated fuel cross-sections. The microgamma scan system consists of a precision linear stage with integrated sample holder and a tungsten alloy collimator, which interfaces with the Idaho National Laboratory (INL) Analytical Laboratory Hot Cell (ALHC) Gamma Scan System high purity germanium detector, multichannel analyzer, and removable collimators. A simplified model of the microgamma scan system was developed in MCNP and used to investigate the system performance and to interpret data from the scoping studies. Preliminary measurements of the microgamma scan system are discussed.*

INTRODUCTION

The U. S. Global Nuclear Energy Partnership / Advanced Fuel Cycle Initiative (GNEP/AFCI) is developing metallic and oxide transmutation alloys as a fuel form to transmute the long-lived transuranic actinide isotopes contained in spent nuclear fuel into shorter-lived fission products. The AFCI program has irradiated and examined eleven metallic alloy transmutation fuel specimens to evaluate the feasibility of actinide transmutation in advanced sodium-cooled fast reactors and thermal reactor implementation. Initial results of postirradiation examinations indicated the irradiation performance of the actinide-bearing compositions is similar to uranium-plutonium-zirconium ternary metallic alloy fuels (U-xPu-10Zr) [1]. Further studies to characterize radial burnup profile, constituent migration, and fuel cladding chemical interaction (FCCI) are in progress.

A microgamma scan (MCGS) system is being developed to evaluate radial burnup profile, cesium migration to the sodium bond and constituent migration within the fuel. These data will further clarify the comparative irradiation performance of actinide-bearing metallic transmutation fuel forms and uranium-plutonium-zirconium alloys. MCGS systems have been developed as a complementary technique to electron probe microanalysis (EPMA) [2]. The MCGS analysis is non-destructive and does not need special fuel preparation techniques to minimize surface effects. MCGS can distinguish between isotopes of an element.

The major components of the MCGS system are described. The final design of the MCGS system was based on results from initial measurements and a simplified model of the microgamma scan system developed in MCNP used to investigate the system performance and to interpret data from the scoping studies. Results from the scoping studies and MCNP modeling are summarized.

EXPERIMENT APPARATUS

MCGS System Description

A microgamma scan system is being developed to analyze the radial distribution of fission products, such as Cs-137, Cs-134, Ru-106, and Zr-95, in irradiated fuel cross-sections. A description of the microgamma scan system is provided. The microgamma scan system interfaces with the INL Analytical Laboratory Hot Cell (ALHC) Gamma Scan System, which includes a high purity germanium detector, a multichannel analyzer, removable collimators with diameters of 3.2, 12.7 and 25.4 mm (0.125, 0.5, and 1.0 in.) and a sample positioning track and trolley. The additional components of the MCGS consist of precision linear stage with integrated sample holder, tungsten alloy collimator, collimator saddle and mounting plate. Fig. 1 shows the microgamma scan apparatus in perspective and a plan view interfacing with the ALHC Gamma Scan System hardware.

The linear stage and integral sample holder is manually operated; one wheel rotation equates to 500 µm (0.021-in) of linear travel of the sample (a 1.5 mm (0.063-in) lead on the linear stage and a gear reduction with a 3:1 ratio). Linear travel is displayed by a ruled scale that is visible to the hot cell operators. Fig. 2 shows the sample holder and Octagon Sample Holder, which can be rotated one flat (45 degrees) to provide the capability of scanning multiple radial profiles at 45 degree increments (0, 45, 90, 135˚).

The collimator is fabricated from tungsten alloy with density of 17.5 g/cm^3 and has length of 101.6 mm and collimator diameter of 0.51 mm. An optional collimator with length of 50.8 mm and collimator diameter of 1.02 mm can be used. The outside diameter of both collimators is 50.8 mm, which accommodates a range of fuel samples sizes up to 25.4 mm diameter.

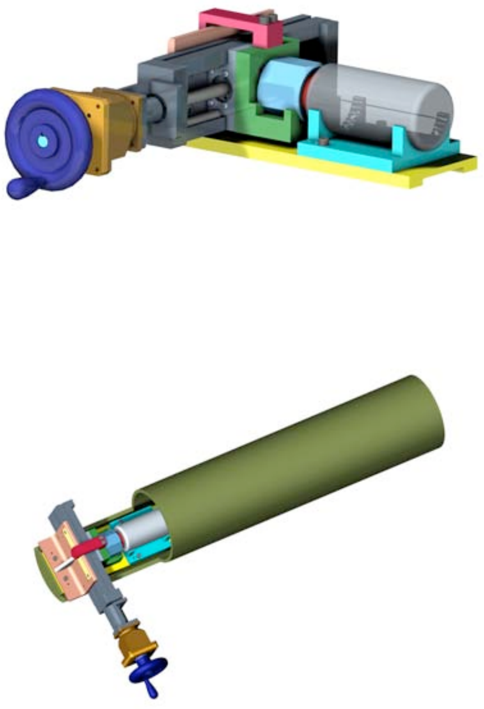


Fig. 1. Schematic of microgamma scan (MCGS) system a) perspective showing linear positioning stage, sample holder and collimator and b) Plan view of MCGS showing interface with ALHC Gamma Scan System hardware

Samples

The microgamma scan system is designed to interface with standard metallography mounts and can accommodate a range of sample sizes within this construct. Analyses are planned for metallic alloy, oxide and nitride actinide transmutation fuel samples from the AFC-1 and LWR-1a irradiation tests [1,3]. The microgamma scan fuel samples are transverse cross-sectional metallography mounts with standard diameter of 31.75 mm (1.25-in.) and 25.4 mm (1.0-in.) in height. The nominal fuel thickness is 2.54 mm (0.10 in.). Figure 2 shows a micrograph of an AFC-1 metallic alloy fuel metallography cross section with the collimated analyzed regions overlaid.

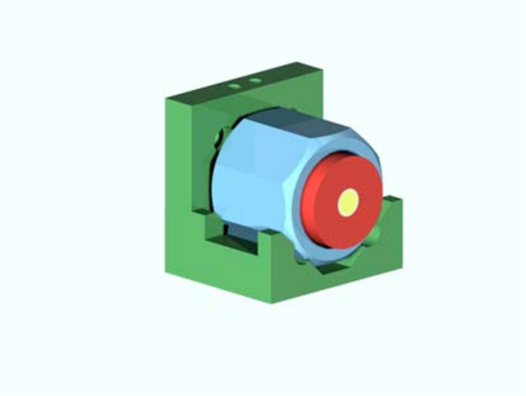


Fig. 2. Microgamma scan system sample holder with transverse fuel rod cross-section sample in metallography mount

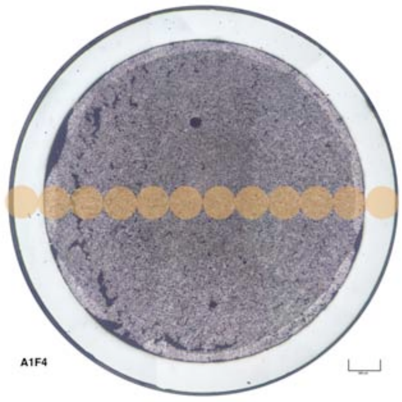


Fig. 3. AFC-1 Metallic Fuel Sample with Microgamma Scan locations overlaid.

RESULTS

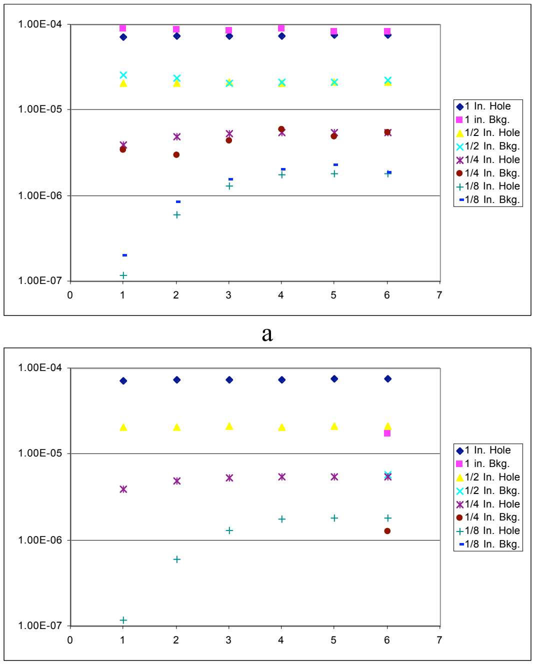
Scoping Studies

Scoping studies with irradiated fuel samples were performed with a preliminary design of the microgamma scan (MCGS) system. The preliminary design of the apparatus consisted of ten interchangeable collimator blocks. The collimator blocks were fabricated from 38.1 mm (1.5-in.) of lead with a 0.953 mm (0.0375-in.) collimator hole indexed at 0.953 mm intervals across the sample's diametral line. A "trolley" and track system was used to position the sample for counting. For the irradiated fuel samples, the MCGS measurements varied between 30-40% due to variability in the "trolley" positioning, which significantly limited the resolution of the MCGS analyses to an order of magnitude. These results indicated the high radiation fields from metallography mount sized irradiated fuel samples resulted in systematic errors in the data due to 1. very high count rates to the detector, 2. insufficient shielding of the collimator blocks and 3. inadequate repeatability in sample positioning.

MCNP Modeling

A simplified model of the microgamma scan system was developed in Monte Carlo N-Particle Transport Code (MCNP) and used to investigate the system performance and to interpret the data from the scoping studies [4]. The effectiveness of the indexed sample collimator blocks as configured with lead and with a higher density material (tungsten) was modeled. The MCNP model was also used to investigate the MCGS system's dependence on removable collimator diameters. The relative counts from a Cs-137 point source with shielding and without shielding were calculated using MCNP. The count rate without shielding represents the signal measurement and the count rate with shielding represents the background counts. In the preliminary design, the sample collimator were fabricated from lead; the performance benefits of tungsten were characterized with the MCNP model. Figure 4 shows the results predicted for lead and tungsten sample collimator blocks as a function of radial position.

The MCNP modeling results show the signal to background ratio is a little less than one for lead sample collimator blocks. The S/B ratio is around four for tungsten (the background was only modeled for position no. 6) indicating the significant benefit of replacing the sample collimator material. The plots show the effect of the different sizes of the removable collimator. Total counts decrease with removable collimator size, which informs us that background counts (from the non-collimated part of the sample) are significant. The radial profile of the background counts is dependent on the removable collimator diameter and sample size due to the high activity of the fuel samples. For removable collimator diameters of 25.4 (1 in.) diameter and 12.7 mm (0.5 in.), the radial profile is constant. However, the radial profile measured for collimator diameters of 6.4 mm (0.25 in.) and particularly for 3.2 mm (1/8 in.) show a decreasing trend in counts for radial positions near the edge of the sample (position nos. 3, 2, and 1). This was observed for both lead (the current material) and tungsten collimators.



a

b

Fig. 4. MCNP model of relative counts from collimated signal and background as a function of radial position for a) 38.1 mm (1.5 in.) thick lead collimator and b) 38.1 mm (1.5 in.) thick tungsten collimator.

DISCUSSION

The MCGS scoping measurements and MCNP modeling resulted in a number of design improvements, 1. replace the set of collimator blocks and track and trolley positioning with a single collimator and an high precision linear stage and integrated sample holder, 2. increase collimator efficiency by using tungsten alloy in lieu of lead, 3. reduce sample radioactivity by decreasing sample thickness from 5.08 mm to 2.54 mm.

First, the MCGS system trolley and indexed collimators were replaced with a stationary, high lineal density (e.g., tungsten-alloy) collimator and a precision linear stage to move the metallography mount fuel sample. This type of "collimator – sample holder" arrangement alleviated the positioning and repeatability uncertainty.

Second, the collimator was fabricated from a tungsten alloy (density of 17.5 g/cm^3) instead of lead (11.3 g/cm^3). This design change increased shielding efficiency by 50%. The better machinability of tungsten allowed for a smaller collimator, which achieves a 'micrometer' scale measurements.

Third, non-standard thickness metallography mount samples are specified for microgamma scan analyses. Fuel cross-sections with a 2.54 mm thickness will be prepared instead of the nominal 5.08 mm thickness as a compromise between in-cell handling and reduction in sample radioactivity. Further reduction in sample thickness could be accomplished, if desired, by a combination of sectioning and grinding.

Further improvements being considered are replacing the ALHC Gamma Scan System detector with lower-efficiency non-germanium detectors. Two non-germanium detectors that could be used are a cadmium-zinc-telluride (CZT) detector and a lanthanum-bromide (LaBr3) detector.

SUMMARY

A microgamma scan system was developed to evaluate radial burnup profile, cesium migration to the sodium bond and constituent migration within the fuel. MCGS scoping measurements on irradiated fuel samples and MCNP modeling were performed to investigate the system performance and resulted in a number of design improvements, 1. replace the set of collimator blocks and track and trolley positioning with a single collimator and an high precision linear stage and integrated sample holder, 2. increase collimator efficiency by using tungsten alloy in lieu of lead, 3. reduce sample radioactivity by decreasing sample thickness from 5.08 mm to 2.54 mm.

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