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CHARACTERIZATION OF AN ADVANCED GADOLINIUM NEUTRON ABSORBER ALLOY BY MEANS OF NEUTRON TRANSMISSION

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ABSTRACT

Neutron transmission experiments were performed on samples of an advanced nickel-chromium-molybdenum-gadolinium (Ni-Cr-Mo-Gd) neutron absorber alloy. The primary purpose of the experiments was to demonstrate the thermal neutron absorbing capability of the alloy at specific gadolinium dopant levels. The new alloy is to be deployed for criticality control of highly enriched DOE SNF. For the transmission experiments, alloy test samples were fabricated with 0.0, 1.58 and 2.1 wt% natural gadolinium dispersed in a Ni-Cr-Mo base alloy. The transmission experiments were successfully carried out at the Los Alamos Neutron Science Center (LANSCE). Measured data from the neutron transmission experiments were compared to calculated results derived from a simple exponential transmission formula using only radiative capture cross sections. Excellent agreement between the measured and calculated results demonstrated the expected strong thermal absorption capability of the gadolinium poison and in addition, verified the measured elemental composition of the alloy test samples. The good agreement also indirectly confirmed that the gadolinium was dispersed fairly uniformly in the alloy and the ENDF VII radiative capture cross section data were accurate.

I. INTRODUCTION

The National Spent Nuclear Fuel Program (NSNFP), located at the Idaho National Laboratory (INL), coordinates, and integrates national efforts in managing the disposal of U.S. Department of Energy (DOE)-owned spent nuclear fuel (SNF). These functions include the development of a DOE standardized canister for packaging, storage, transport, and long-term disposal of DOE SNF. DOE SNF will be packaged in standardized canisters with internal baskets. Since some types of DOE SNF contain highly enriched uranium, an advanced canister neutron absorber material may be required for criticality control.

By deploying a neutron absorber alloy as a basket material, the number of DOE SNF packages can be reduced and handling of individual SNF elements will be eliminated at the repository. The internal basket material is envisioned to be: 1) highly corrosion resistant, such that under the projected storage, transportation, and disposal conditions the neutron-absorbing element will not leach out, 2) acceptable mechanical properties based on American Society of Mechanical Engineers (ASME) code approval, 3) material production using conventional ingot metallurgy techniques, and 4) be weldable.

Directed by the NSNFP, a research and development project has been implemented to develop an advanced nickel-chromium-molybdenum-gadolinium (Ni-Cr-Mo-Gd) neutron absorber alloy, or now more commonly referred to simply as the Advanced Neutron Absorber (ANA). The Idaho National Laboratory is leading all aspects of the development in conjunction with Sandia National Laboratories supporting melt refractory and production studies, with additional support from Lehigh University supporting mechanical property measurements. Natural gadolinium (Gd) was chosen as the neutron absorbing element based on its high thermal neutron absorption cross section and limited solubility in the disposal environment. The base metal was chosen for its high corrosion resistance and consists of nickel-chromium-molybdenum.

The alloy has been manufactured with conventional techniques involving a Vacuum Inductive Remelt process. The follow on conversion process from ingot form to plate involves standard hot working practices at commercial rolling mills. Mechanical, physical, and chemical results were submitted to the American Society for Testing and Materials (ASTM). The ASTM accepted the results and is now designated as UNS N06464, ASTM Specification B932-04 [1]. The alloy was also approved for use for ASME Boiler and Pressure Vessel Code construction per Section III, Division 3 Code Case N-728 in the non-welded condition [2].

Additional studies beyond mechanical, physical, and chemical property measurements have been performed that include neutron reactivity, corrosion and weldability. Reactivity experiments were carried out at the Los Alamos Critical Experiments Facility (LACEF) [3]. The average weight percent of gadolinium in the experiment prototype Ni-Cr-Mo-Gd alloy was approximately 2.34 wt%. The experiment had a measured k_{eff} of 1.002 verses a predicted value of 1.001, or a negative worth of the Gd alloy plates of about 8.8$ of reactivity. As a comparison in the same configuration, the calculated negative worth of an equivalent volume of borated stainless steel plate (1.7% natural boron) was approximately 6.4$ of reactivity. The results of these experiments are documented in the International Handbook of Evaluated Criticality Safety Benchmark Experiments by the Nuclear Energy Agency Nuclear Science Committee [4].

In addition, the INL has performed numerous corrosion tests. The results have shown, the Gd intersecting the surface along the grain boundaries is easily removed followed by repassivation of the Ni-Cr-Mo-Gd matrix, the corrosion rate will drop to an extremely low rate [5]. Welding studies continue to be performed to generate the required data for the extension of ASME Code Case N-728 to welded construction. Current results show that mechanical properties of weld joints have reduced ductility relative to the base metal. However, the properties can be recovered with the use of post weld heat treatment [6].

Future research and development is directed at alloy scale-up to 1000-3000 pound slabs and continued weld process development. One of the primary issues during scale-up will be to focus on the loss of gadolinium when introduced during the melt process. Gadolinium has a very high attraction for oxygen. Techniques will need to be developed to account for these gadolinium losses. Welding process development continues at the INL studying process parameters and different weld wire. These efforts will result in future revision to the ASME Code Case to included welded conditions once the process is finalized.

Preliminary results from neutron transmission measurements on the ANA alloy are described herein. The neutron transmission measurements are intended to provide data that can be compared to calculated transmission results using a simple exponential transmission formula with microscopic radiative capture cross sections for all ANA isotopic constituents. Agreement between measured and calculated transmission results provides verification of wet chemical analysis of the ANA materials isotopic constituents, but more importantly, the verification of the thermal neutron absorption capability of the natural gadolinium in the ANA alloy.

II. ANA SAMPLE CHARACTERIZATION

Although the final process for the fabrication of ANA materials has not been finalized, ANA samples were selected that were available and provide a representation of the manufactured product currently envisioned. Plate D5-8302 and Plate E2-14827(C) were chosen since these plates have been characterized by wet chemistry analysis at various locations across the finished plate. The investigated samples were not randomly distributed samples of the finished plate but in contrast were located adjacent to the chemical analysis samples.

II.A Gadolinium Samples

Samples from two ANA plates manufactured during FY-06 were chosen for these measurements. ANA plate designation is D5-8302 with 1.58 wt% Gd and plate E2-14827(C) with 2.1 wt % Gd. To assess the isotopic compositions of gadolinium added to the ANA plates, INL performed an isotopic analysis of the gadolinium feed stock samples. The gadolinium (Gd) neutron poison in the ANA plates was natural Gd with the following isotopic atom fractions: Gd-152 (0.20%), Gd-154 (2.18%), Gd-155 (14.8%), Gd-156 (20.47%), Gd-157 (15.65%), Gd-158 (24.84%), and Gd-160 (21.86%). The results clearly indicated that the gadolinium has not been enriched from “naturally occurring” gadolinium [7].

The largest thermal neutron absorber is the Gd-157 with an estimated thermal neutron radiative capture cross section of 255,000 barns [7]. All the plates measured were approximately 0.9525 cm (3/8-inch) thick and 5.08 cm (2-inches) square. Chemical analysis of the two ANA plates, at varies locations, were taken from finished plates. The chemical analysis results are provided in Table I.

Table I. Chemical Analysis of Measured Plates

Element	D5-8302, 02-S2 * (wt %)	E2-14827(C), SC-1 (wt %)
Mo	14.50	14.63
Cr	16.65	16.18
Gd	1.58	2.1
Mn	<0.01	<0.001
Mg	<0.005	0.001
Ni	Balance (~67.24)	Balance (~67.04)
Fe	0.02	0.021
Co	<0.01	0.014
C	0.010	0.0081
Si	<0.01	0.006
S	<0.001	<0.001
N	0.000	<0.001

*average of triplicate sample measurements

II.B Non-Gadolinium Samples

To evaluate the gadolinium affects on neutron transmission measurements samples with similar chemistries were measured that did not contain gadolinium. Plate D5-7689 (M-319) was chosen due to similar chemistries as the ANA materials. The chemical analyses of the non-Gd plate are provided in Table II.

Table II. Non-Gadolinium Chemical Analysis

Element	D5-7689, M-319 (wt %)
Mo	15.3
Cr	16.32
Gd	0
O	0.0069
Mn	0.01
Mg	0.0023
Ni	Balance (~68.34)
Fe	0.01
Co	0.01
C	0.003
Si	0.001
S	0.0006
N	0.0004

III. EXPERIMENTAL AND FACILITY

LANSCE requires experiment requesters to register their experiments. This registration process is the primary mechanism at LANSCE for counting users. An INL experiment proposal was submitted (S083) and a proposal number (0062551) assigned. Neutron transmission experiments used standard LANSCE experimental techniques.

Figure 1 depicts the layout of Lujan Centers' large array of capabilities. Each of the experimental ports is labeled with a flight path (FP) number. Short, intense proton pulses are directed at a tungsten target. The tungsten target is coupled to two different neutron moderators for the production of cold (<0.025 eV), thermal, and epithermal neutrons. The neutrons are collimated to form beams in each FP. FP-5 was chosen for these experiments as shown in Figure 1. This flight path is commonly used for fission cross section measurements, and it has a parallel-plate fission ionization chamber placed about 8 meters from the tungsten target. A 200 µg/cm^2 thick U-235 fission foil was loaded in the chamber, and neutron induced fission from the foil was measured. The short flight path, the large fission cross section of U-235, as well as the high efficiency of the parallel-plate chamber all contribute to high count rates which allow reasonable sort irradiation times to be used to collect sufficient statistics. Figure 2 shows the LANSCE measured detector counts as a function of neutron energy at FP-5, without a sample present.

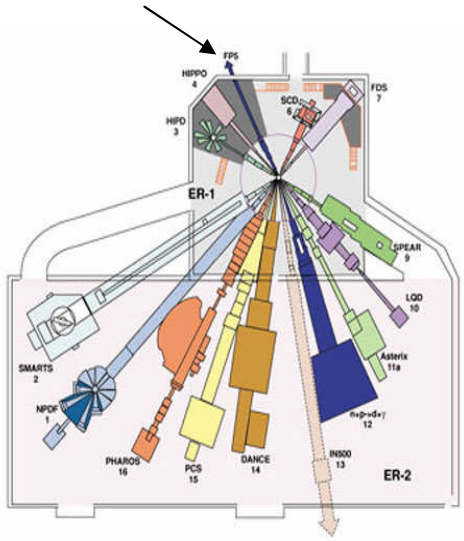

Figure 1. Layout of the Lujan Center

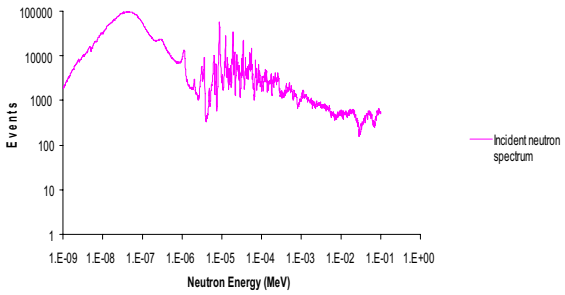

Figure 2. LANSCE Measured results without sample

IV. RESULTS TO DATE

Three neutron transmission experiments on three different material samples have been completed at the LANSCE facility. The three samples include the M-319 (no Gd), 02-S2 (1.58 wt% Gd), and E2-14827(C) (2.1 wt% Gd) samples. A fourth test measured the neutron spectrum and intensity without a sample present in the beamline.

IV.A. Calculated Macroscopic Cross Sections

Evaluated Nuclear Data File (ENDF) microscopic cross sections were provided for the four main elements in the ANA material, mainly nickel, molybdenum, chrome, and gadolinium [8]. The total cross sections for were imported into an Excel spreadsheet. The energy spectrum given for each microscopic cross section was cross referenced to the measured energy spectrum provided by LANSCE for each sample measured. The data was further "normalized" based on a LANSCE calculated charge (coulombs) delivered to the neutron-producing tungsten target for each of the sample runs. The number of events is proportional to the incident number of neutrons on each transmission sample.

Macroscopic cross sections as a function of energy were calculated for the three samples tested: (1) D5-8302 (1.58 wt% Gd), (2) E2-14827 (2.1 wt% Gd), and (3) M-310 (0 wt% Gd). The calculated macroscopic cross sections are based solely on ENDF VII cross section data and the measured isotopic concentrations of constituents in each test sample. The macroscopic cross sections ($\sum$) were calculated using the following formula:

$$\sum = \sigma * N$$

Where:

σ = microscopic radiative capture cross section (cm^2)
N = atomic number density ($atoms/cm^3$)

and

$$N = (\rho * A * W_j * AP_i / M_i)$$

ρ = Alloy density ($8.76 g/cm^3$)
A = Avogadro's number ($6.023\ 10^{23}$ atoms/mole)
W_j = weight percent of alloy element j (based on chemical analysis)
AP_i = atom percent of isotope i in natural occurring element
M_i = atomic weight of isotope i (g/mole)

Figure 3 shows the calculated total (N,TOT) microscopic cross section for sample 02-S2 (1.58 wt% Gd) by element.

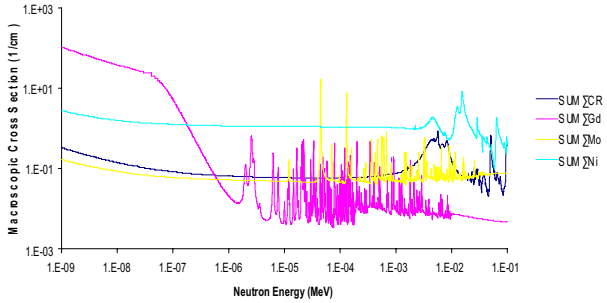

Figure 3: Calculated total macroscopic cross sections for ANA sample 02-S2 (1.58 wt% Gd) by element.

The elemental macroscopic cross sections were totaled and are shown in Figure 4.

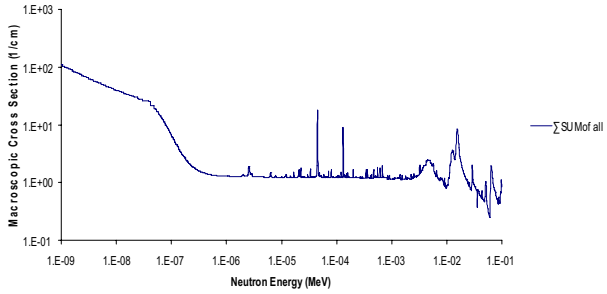

Figure 4: Calculated total macroscopic cross section for Sample D5-8302 (1.58 wt% Gd).

IV.B. Results of Calculated Versus Measured

The transmission intensities (events) were calculated based on the given normalized events from the sample out (I_o) measured values by using the following relationship:

$$I_t = I_o * e^{-\sum x}$$

Where:

I_t = number of events (calculated),
I_o = number of measured events (sample out),
$\sum$ = macroscopic (N,G) or radiative cross section for a given energy (cm^2), and
x = thickness of the neutron absorbing material (0.952 cm or 3/8-inch)

The results are shown in Figures 5, 6 and 7 as comparison plots between calculated and measured events.

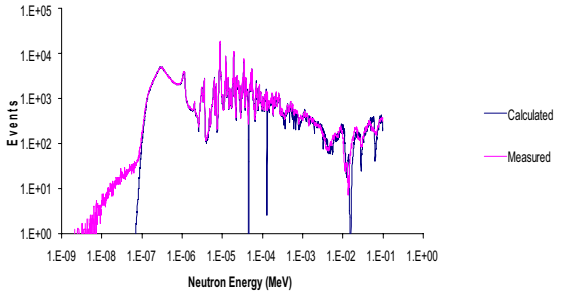

Figure 5: Calculated versus measured events based on total macroscopic cross sections for ANA sample D5-8302 (1.58 wt% Gd).

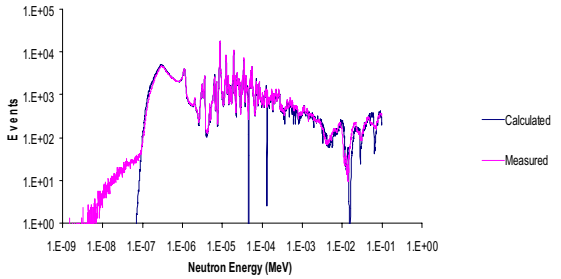

Figure 6: Calculated versus measured events based on total macroscopic cross sections for ANA sample E2-14827 (2.1 wt% Gd).

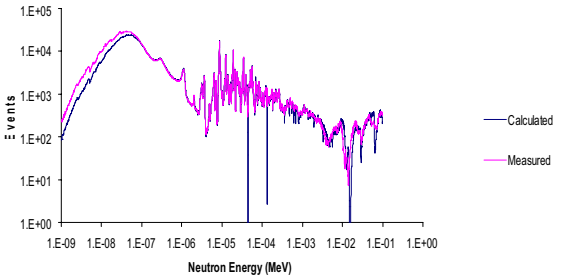

Figure 7: Calculated versus measured events based on total e macroscopic cross sections for sample M-319 (0 wt% Gd).

V. DISCUSSION OF RESULTS

The calculated results are in very good agreement with the measured results as shown in Figures 5, 6 and 7. The agreement is best over the thermal neutron energy range, but good agreement extends up to approximately 3.0E-3 MeV. The thermal range (0.01eV to 1eV) is the range where the total macroscopic cross section is dominated by the Gd-157 cross section.

V.B. Conclusions

It is clear that the addition of natural gadolinium in the ANA samples absorbs thermal neutrons. The severely depressed thermal flux (events) in Figures 5 and 6 is due primarily to the large thermal cross section of the Gd-157 isotope.

The very good agreement between the measured data and the calculated results (Figures 5, 6 and 7) shows that the simple exponential transmission formula combined with ENDF energy-dependent total macroscopic cross sections for the ANA sample are sufficient to predict the physical phenomena. The good agreement also demonstrates indirectly the integrity of the ENDF cross section used in the exponential transmission formula and the relative uniformity of the gadolinium distribution in the ANA sample, since the formula implies a homogenized or uniform gadolinium distribution in the 3/8-inch thick samples.

Additional transmission experiments with as-melted ANA material without rolling and heat treatment are planned in the near future at LANSCE. These experiments will be based on thinner plate.

ACKNOWLEDGMENTS

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