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NICKEL-BASED GADOLINIUM ALLOY FOR NEUTRON ADSORPTION APPLICATION IN RAM PACKAGES

G. W. Wachs. J. W. Sterbentz, W. L. Hurt
Idaho National Laboratory

P. E. McConnell, C. V. Robino
Sandia National Laboratories

F. Tovesson, T. S. Hill
Los Alamos National Laboratory

ABSTRACT

Neutron transmission experiments were performed on samples of an advanced nickel-chromium-molybdenum-gadolinium (Ni-Cr-Mo-Gd) neutron absorber alloy and chromium-nickel (Cr-Ni) stainless steel, modified by the addition of boron. The primary purpose of the experiments was to demonstrate the thermal neutron absorbing capability of the materials at specific gadolinium and boron dopant levels. The Ni-Cr-Mo-Gd alloy is envisioned to be deployed for criticality control of highly enriched U.S. Department of Energy (DOE)-owned spent nuclear fuel (SNF). For these transmission experiments, test samples were fabricated with 0.0, 1.58 and 2.1 wt% natural gadolinium dispersed in a Ni-Cr-Mo base alloy and 1.16 wt% boron in stainless steel. 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 total neutron cross sections. Excellent agreement between the measured and calculated results demonstrated the expected strong thermal absorption capability of the gadolinium and boron elements and in addition, verified the measured elemental composition of the Ni-Cr-Mo-Gd alloy and borated stainless steel test samples. The good agreement also indirectly confirmed that the size and distribution of the gadolinium in both the hot-top (as-cast) and Ni-Cr-Mo-Gd converted to plate was not a discriminator related to neutron absorption. Moreover, the Evaluated Nuclear Data File (ENDF VII) total neutron cross section data were accurate.

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 neutron absorber material may be required for criticality control. By deploying a neutron absorbing 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.

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 absorbing alloy. The alloy is now commonly referred to simply as the Advanced Neutron Absorber (ANA) alloy. The Idaho National Laboratory is leading all aspects of the development supported by Sandia National Laboratories in melt refractory and production studies and with additional support from Lehigh University in 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 compatibility with the gadolinium containing phase.

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

Preliminary results from neutron transmission measurements 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 total neutron cross sections for the major isotopic constituents. Agreement between measured and calculated transmission results provides verification of wet chemical analysis of the materials isotopic constituents, but more importantly, the verification of the thermal neutron absorption capability of the natural gadolinium in the ANA alloy. In addition, samples of borated stainless steel were measured as a direct comparison.

ANA SAMPLE CHARACTERIZATION

Although the full scale production process for the fabrication of ANA alloys has not been finalized, ANA alloy samples were selected that were available and provide a representation of the manufactured product currently envisioned. The investigated ANA alloy samples were not randomly distributed samples of the finished plate but in contrast were located adjacent to the chemical analysis samples.

Several samples of borated stainless steel were provided by Sandia National Laboratories. These borated stainless steel samples contained different wt% boron concentrations, 1.16, 1.47, and 1.70. The borated stainless steel sample with the lowest wt% boron (1.16 wt% B) was chosen as a comparison with different gadolinium wt% ANA alloys.

ANA Samples

Samples were obtained from two ANA alloy plates designated as D5-8302 with 1.58 wt% Gd and plate E2-14827(C) with 2.1 wt% Gd. In addition, an ANA alloy sample was removed from the “hot-top” in the as-cast condition of ingot D5-8302. While the chemical composition of the Hot-Top and converted plate is identical, the microstructure features are quite different. Metallographic sections of a similar ANA alloy plate containing approximately 1.90 wt% Gd were performed at Lehigh University [3]. Metallographic samples were prepared in thermosetting epoxy mounts and were ground and polished using standard metallographic procedures, with the final step consisting of vibratory polishing. The micrographs clearly show a

significant improvement in gadolinium grain structure following conversion. Little difference is noted between transverse and longitudinal views. All the sample plates were approximately 0.9525 cm (3/8-inch) or 0.317 cm (0.125-inch) thick and 5.08 cm (2-inches) square. To confirm the isotopic compositions of gadolinium added to the ANA alloy plates, INL performed an isotopic analysis of the gadolinium feed stock samples. The results clearly indicated that the gadolinium isotopic ratios were consistent with "naturally occurring" gadolinium [4]. The main chemical analysis of the two ANA alloy plates, at varies locations, were taken from finished plates. The chemical analysis results are provided in Table 1.

Table 1. Chemical Analysis of Measured ANA Alloy Plates

Element	D5-8302, 02-S2 * (wt%)	E2-14827(C), CS-1 (wt%)
Mo	14.50	14.63
Cr	16.65	16.18
Gd	1.58	2.1
Ni	Balance (~67)	Balance (~67)
N	0.000	<0.001

*average of triplicate sample measurements

Non-Gadolinium Samples

To contrast the gadolinium affects on neutron transmission measurements; alloys with similar chemistries were measured that did not contain gadolinium. Plate D5-7689 (M-319) was chosen due to similar chemistries as the ANA alloy. The main chemical analyses of plate M-319 are provided in Table 2.

Table 2. Non-Gadolinium Chemical Analysis

Element	D5-7689, M-319 (wt%)
Mo	15.3
Cr	16.32
Ni	Balance (~68)

Borated Stainless Steel Samples

Borated stainless steels are defined in ASTM A887 [5], where there are eight types (304B through 304B7) which define boron concentrations from 0.2 to 2.25 wt% B. There are two grades (A & B) defined where requirements are controlled by mechanical properties (Charpy impact energy). The grade of the material is defined by the uniformity of the dispersion of the boron within the melt. Grade A corresponds to the near-optimal dispersion, while Grade B corresponds to a less-than-optimal dispersion of the boron. Conventional wrought metallurgical practice conforms to Grade B properties. The quality of the boron dispersion is measured indirectly through the ductility requirements [5]. Each of the types is further defined into sixteen discrete alloys. Borated stainless steel UNS Designation S30464, Type 304B4 Grade A was measured containing 1.16 wt% boron. Table 3 provides the chemical compositions of the borated stainless steel plate reported by Carpenter Technology Corporation (Heat Number 182194). The plate was machined into 0.9525 cm (3/8-inch) thick and 5.08 cm (2-inches) square. The outer layer of inert stainless steel, generated by the Hot Isostatically Pressed (HIP) fabrication process, was removed prior to the machining. The chemical analysis of the borated stainless steel is provided in Table 3.

Table 3: Chemical Analysis of Borated Stainless Steel 304B4 Grade A Sample

Element	Heat Number -182194 Neutrosorb Plus© (wt%)
Cr	19.70
Mn	1.78
Ni	13.79
B	1.16
Fe	Balance (~64)

EXPERIMENTAL FACILITY

The transmission experiments were performed at the LANSCE facility, more specifically the Lujan Center. 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 from the accelerator 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. This flight path is commonly used for time-of-flight fission cross section measurements, and it has a parallel-plate fission ionization chamber placed about 8 meters from the tungsten target. The samples were placed directly in the collimated bean. A shield wall made of a polymer plate isolates the sample by approximately 2 meters from the 200 µg/cm^2 thick U-235 fission foil loaded in a chamber, where neutron induced fission (events) from the foil are 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 short irradiation times to be used to collect sufficient statistics. Figure 2 shows typical LANSCE neutron spectrum (measured events) as a function of neutron energy that would be incident on target samples in FP-5.

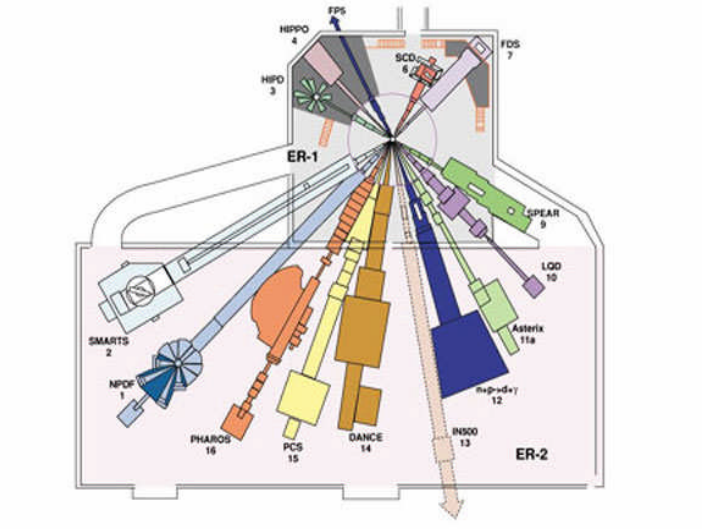

Figure 1. Layout of the Lujan Center

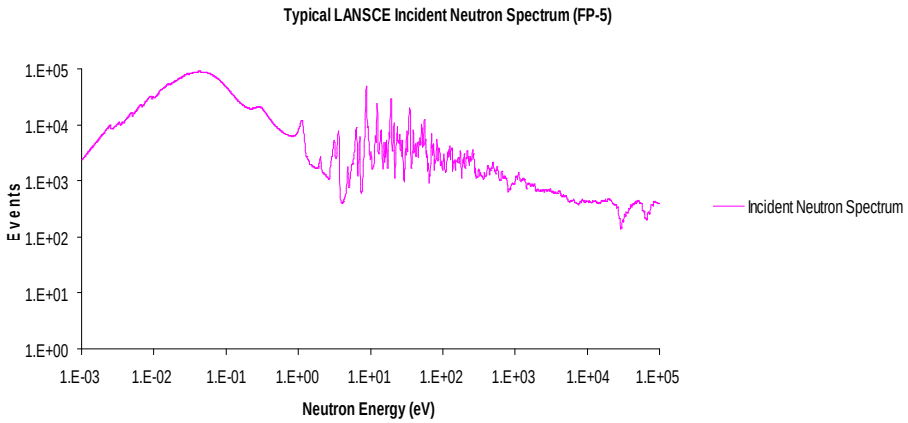

Figure 2. Typical LANSCE incident neutron spectrum for FP-5

RESULTS

Neutron transmission experiments on five different material sample types have been completed at the LANSCE facility. The five types of samples include:

- one sample from plate M-319 (ANA alloy with no Gd),
- two from ANA alloy plate 02-S2 (1.58 wt% Gd) in as-cast and converted conditions,
- one from borated stainless steel plate heat number 182194 (1.16 wt% B), and
- one from ANA alloy E2-14827(C) (2.1 wt% Gd) samples.

A sixth test measured the neutron spectrum and intensity without a sample present in the beamline.

Calculated Total Macroscopic Cross Sections

Evaluated Nuclear Data File (ENDF) microscopic neutron total cross sections [6] were obtained for the main elements in the ANA alloy and borated stainless steel. The total neutron cross sections (N, TOT) were imported into an Excel spreadsheet. The energy spectrum given for each total 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 measured 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 samples tested. The calculated macroscopic cross sections are based on the total ENDF VII cross section data and the chemically 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 = total neutron microscopic cross section (cm^2)
N = atomic number density ($atoms/cm^3$)

and;

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

ρ = Alloy density (8.76g/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)

Comparison 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,TOT) or total neutron cross section (cm^{-1}) for a given energy, and
x = thickness of the neutron absorbing material (0.952 cm or 0.317 cm)

DISCUSSION OF RESULTS

The calculated results are in very good agreement with the measured results, based on the simple exponential transmission formula used in the prediction of the calculated transmission events using total cross section (N,TOT) data of all the applicable sample isotopes.

Figure 3 provides the as-measured values for each experiment with the same sample thicknesses (0.952-cm, 3/8-in). Figure 3 clearly shows the significance boron and gadolinium have on neutron spectrums below the 1.0 eV neutron energy ranges. The effect from the alloy base materials above approximately 3.0 eV neutron energy range also shows an effect as well, as compared to sample out measurements.

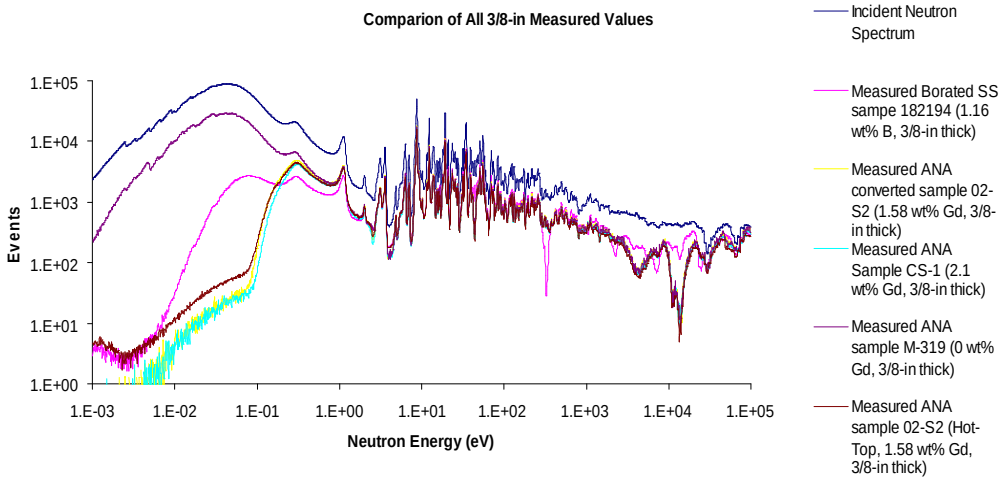

Figure 3. Comparison between all 0.952 cm (3/8-in) thick samples

Figure 4 shows that the as-cast (Hot-Top) microstructural differences have little or no effect on neutron transmission measurements, above the 0.1 eV neutron energy range in comparison to the same ANA material converted (1.58 wt% Gd). The differences below the 0.1 eV neutron energy range have not been determined at this time. However, in Figure 4, similar differences below the 0.1 eV energy range were noted between ANA alloy with 1.58 wt% gad and ANA alloy with 2.1 wt% gad.

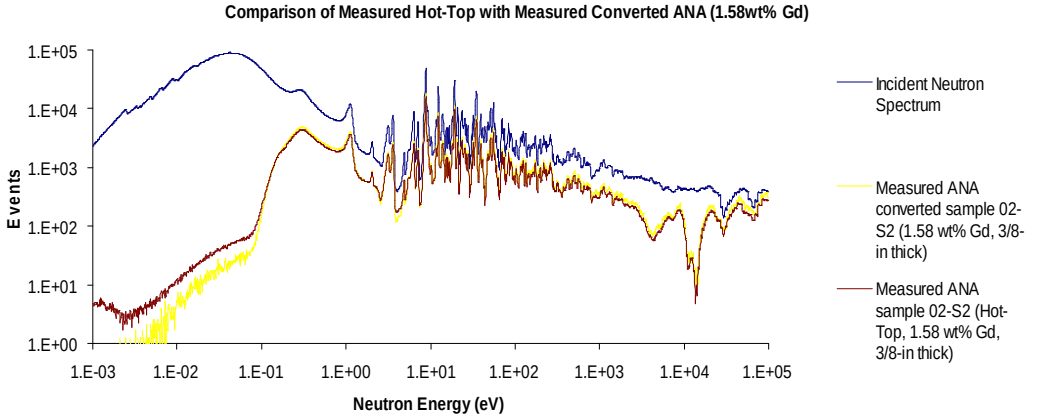

Figure 4. Comparison between Hot-Top and converted 02-S2 (1.58 wt% Gad) samples

Figure 5 clearly demonstrates gadolinium in the ANA alloy has a significant advantage even at 1.58 wt% over Borated SS with 1.16 wt% B below approximately the 0.3 eV neutron energy range. However, Borated SS has only a slight advantage over the ANA alloy between 0.3 eV to 1.0 eV neutron energy ranges, even when compared to ANA alloy with 2.1 wt% Gd.

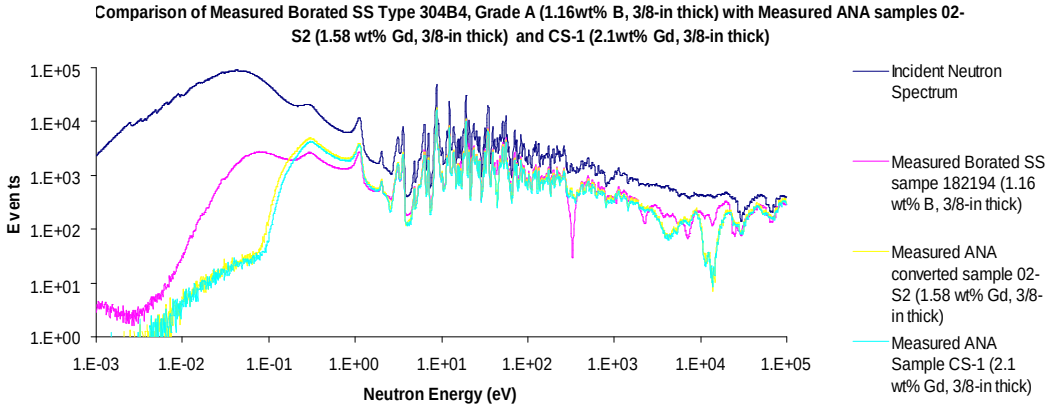

Figure 5. Comparison between Borated SS (1.16 wt% boron) and 02-S2 (1.58 wt% Gad) samples

CONCLUSIONS

Very good agreement between the measured data and the calculated results demonstrates that the simple exponential transmission formula combined with ENDF [6] energy-dependent macroscopic total neutron cross sections for the ANA alloy and Borated SS are sufficient to predict the physical phenomena. In summary, the neutron transmission results clearly demonstrate:

- strongly depressed thermal neutron flux (events) below 1.0 eV is due primarily to the large thermal neutron cross section of the Gd-157 and B-10 isotopes,
- alloy base materials have an effect on neutron flux above approximately 3.0 eV neutron energy ranges,
- ANA alloy (1.58 wt%) has a significant neutron advantage over borated SS (1.16 wt% B) below approximately the 0.3 eV neutron energy range,
- borated SS (1.16 wt% B) has only a slight advantage over the ANA (1.58 wt%) alloy between 0.3 eV to 1.0 eV neutron energy ranges,
- indirectly, the integrity of the ENDF [6] total cross sections used in the simple exponential transmission formula,
- since the exponential transmission formula implies a homogenized or uniform elemental distribution, the relative uniformity of the gadolinium and boron distribution in the samples has little or no influence on transmission performance for thicknesses above 3/8-inch,
- current chemical analysis techniques for element measurements in these alloys are adequate,
- virtually no thermal neutron absorption gain using 2.1 wt% Gd versus 1.58 wt% Gd, and
- conversion of as-cast material appears to have little or no effect on the neutron transmission properties.

ACKNOWLEDGMENTS

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