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# Applications of Neutron-Absorbing Structural-Amorphous Metal (SAM) Coatings for Criticality Safety Controls of Used Fuel Storage, Transportation, and Disposal

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# Applications of Neutron-Absorbing Structural-Amorphous Metal (SAM) Coatings for Criticality Safety Controls of Used Fuel Storage, Transportation, and Disposal

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## ABSTRACT

Used nuclear fuel contains fissionable materials ($^{235}$U, $^{239}$Pu, $^{241}$Pu, etc.). To prevent nuclear criticality in used fuel storage, transportation, and disposal, neutron-absorbing materials (or neutron poisons, such as borated stainless steel, Boral™, Metamic™, Ni-Gd, etc.) would have to be applied. Corrosion-resistant, iron-based structural-amorphous metals (SAMs) have been tested to determine their relative corrosion resistance. Many of these materials can be applied as coatings with advanced High Velocity Oxy-Fuel (HVOF) thermal-spray technology. SAM2X5, SAM1651, and other SAMs are amorphous-metal composite alloys that have been identified as having outstanding corrosion resistance. Because of its high boron content, SAM2X5 can be applied as the neutron-absorbing coatings to the metallic support structure for criticality-safety controls of used fuel in racks in wet storage pool and baskets inside the storage containers, the transportation cask, and eventually the disposal containers in repository disposal.

Research and experiment conducted at Lawrence Livermore National Laboratory indicated that the high boron-containing SAM2X5 coating could be an effective criticality control material for used fuel management. The neutron irradiation experiments and the neutron transmission measurements conducted at McClellan Nuclear Radiation Center indicated that extensive fast neutron irradiation did not change the structure of the amorphous SAM2X5 melt-spun ribbons, and SAM2X5 exhibited effective neutron absorbing capability, similar to Boral™ and Metamic™.

## INTRODUCTION

Criticality prevention is an important regulatory requirement during all phases of managing used nuclear fuel. When used fuels are stored in wet pools at reactor sites, or in dry-cask containers on-site or at away-from-reactor sites, criticality safety controls must be certified under title 10 CFR Part 72[1]. When the loaded containers are transported to the geologic repository operations area (GROA), criticality controls must be certified under title 10 CFR Part 71[2]. At the GROA, a loaded container may be handled in a shielded transfer cask or aged in an over-pack, or disposed of in a waste package, criticality controls in these aging and disposal functions must be certified under title 10 CFR Part 63[3].

To prevent nuclear criticality in used fuel storage systems (water pools or dry containers), the nuclear industry uses aluminum-based boron-containing materials as neutron poisons, such as Boraflex™, Boral™, Metamic™. These neutron poisons are certified (or licensed) for a period less than fifty years. For longer time period, such as those required in long-term aging (storage) or disposal system, the use of non-aluminum based materials, such as borated stainless steel, Ni-Cr-Mo-Gd alloy [4] (or simply Ni-Gd), and others may be preferred.

A family of iron-based, amorphous metals with very good corrosion resistance has been developed that can be applied as a protective coating. The fact that these amorphous metals relies on the high content of boron to make the material amorphous – an essential property for corrosion resistance – and that the boron has to be homogenously distributed in the amorphous metals essentially makes the amorphous material a neutron poison. One of the formulations within this family is SAM2X5, which contains chromium (Cr), molybdenum (Mo), and tungsten (W) for enhanced corrosion resistance, and boron (B) to enable glass formation and neutron absorption [5-7]. Table 1 compares the compositions of SAM2X5, borated stainless steel, and Ni-Gd alloy, in atom percent.

Table 1 Compositions of SAM2X5, Borated Stainless Steel, and Ni-Gd Alloy (in Atom %)

| | Fe | Cr | Mo | Ni | C | W | Mn | Si | B | Gd |
|---|---|---|---|---|---|---|---|---|---|---|
| SAM2X5 MSR | 48.8 | 17.6 | 7.2 | 0 | 3.7* | 2.5 | 2.4 | 2.7 | 15* | 0 |
| Borated Stainless Steel | 59.1 | 21.5 | ND† | 10.7 | ND | ND | 3.1 | 0.4 | 5.2 | 0 |
| Ni-Cr-Mo-Gd Alloy | ND | 16.6 | 11.4 | 70.2 | ND | ND | ND | ND | 0 | 1.8 |

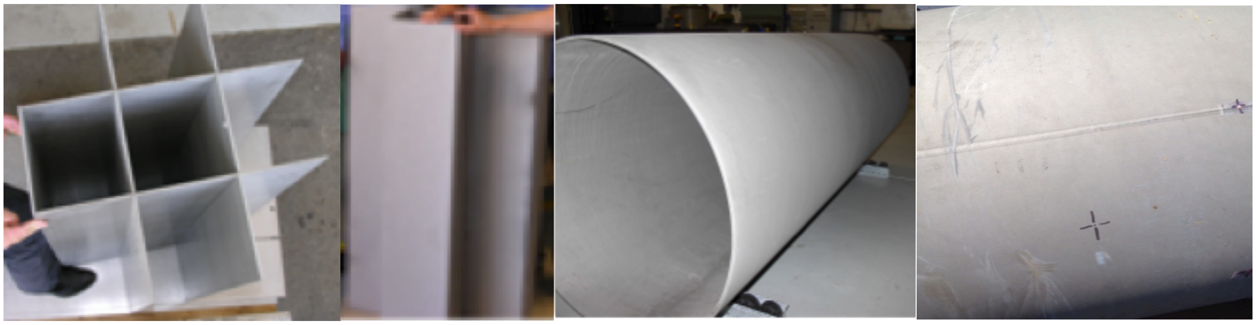
Note: * Weight percent for carbon and boron are 0.83 and 3.22, respectively,
† ND = Not detected

The applications of boron-containing iron-based amorphous metal coating to the metallic basket support structure inside the used-fuel containers can enhance criticality safety during the transportation, storage (aging), and disposal of used fuel. The outstanding corrosion-resistance of the amorphous coatings can protect the integrity of the basket support structure and help preserve the configuration control for used fuel assemblies placed inside the containers.

## APPLICATION OF CRITICALITY-CONTROL SAM2X5 COATINGS

A half-scale basket assembly, based on the criticality control modules included in the conceptual used fuel container designs was produced and assembled [8]. The half-scale basket assembly was coated with SAM2X5 by using a JK2000™ gun in a thermal spray process with a high-velocity oxy-fuel (HVOF) process. The HVOF involves a combination flame, and is characterized by gas and particle velocities that are three to four times the speed of sound. This process is ideal for depositing metal and cermet coatings, which have typical bond strength of 5,000 to 10,000 pounds per square inch (psi), porosity of less than one percent and extreme hardness. The cooling rate that can be achieved in a typical thermal spray process such as HVOF is on the order of 104 Kelvin per second (K/s), and is high enough to enable many alloy compositions to be deposited above their respective critical cooling rate, thereby maintaining the vitreous state.

A half-scale model used-fuel container made of Type 316L stainless steel was coated with SAM2X5 by using the same HVOF process. The container was fabricated from Schedule 10 pipe, had lengths of approximately 90 inches, diameters of approximately 30 inches, and wall thicknesses of approximately 0.3712 inches. The coating was nominally 17 ± 2 mils (~ 0.5 mm) thick. Figure 1 shows the half-scale basket assembly sized to fit inside a half-scale container, before and after coating with SAM2X5. It also shows the container coated with SAM2X5, and a section of the container body after 10 years stored out-door at the McClellan Nuclear Radiation Center (MNRC).



(a) (b) (c) (d)

Figure 1 – Half-scale basket assembly, (a) before and (b) after SAM2X5 coating, (c) a half-scale container coated with SAM2X5, and (d) coated container after 10 years stored out-door at MNRC.

## CRITICALITY ANALYSIS

A disposal container designed to hold twenty-one used PWR fuel assemblies was modeled for criticality analyses. Each of the 21 Westinghouse designed 17 x 17 assembly containing 264 pins of used $UO_2$ fuel, and void spaces previously occupied by 24 guide thimbles and one instrumentation tube were modeled. The burn-up of the PWR fuel assemblies was 35 GWd/t with 4-year decay. Several fission product isotopes (e.g., $^{149}Sm$, $^{103}Rh$, $^{143}Nd$, $^{155}Gd$, and $^{83}Kr$, etc.) were also included in the evaluation model.

MCNP Version 5, a three dimensional (3-D) Monte-Carlo transport code with continuous energy groups of neutron cross-sections was used to calculate the multiplication eigenvalue ($k_{eff}$) of the critical configurations [9]. The criticality analyses for a disposal container model were performed. The results are shown in Table 2.

Table 2 Results of Criticality Analysis for a Disposal Container Model

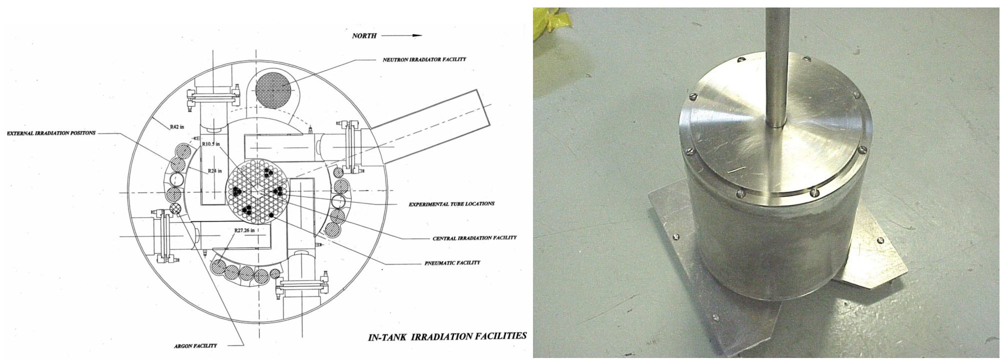
| | ¼" (6.4 mm) stainless steel basket | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | No Boron | 0.12 wt.% B | 1 wt.% B | 2 wt.% B | No Boron 1mm SAM2X5 | 0.12 wt.% B & 1mm SAM2X5 | No Boron 1mm SAM1651 | 0.12 wt. % B & 1mm SAM1651 | ½" Ni-Gd basket material |
| $k_{eff}$ | 1.00 | 0.96 | 0.90 | 0.88 | 0.92 | 0.91 | 0.95 | 0.94 | 0.93 |
| del $k_{eff}$ | 0.0 | 0.04 | 0.10 | 0.12 | 0.08 | 0.09 | 0.05 | 0.06 | 0.07 |

The first set of calculations consists of borated stainless steel basket with various concentration of natural boron (B). It indicates that the borated stainless steel basket with 0.12 wt. % of B would drop the $k_{eff}$ to about 4% of that of the no B case. The second set of calculations consists of stainless steel basket coated with 1mm of SAM2X5 and another SAM material SAM1651 (containing 1.24 wt % natural B). The stainless steel basket contains either no or 0.12 wt % B. The results indicate that the 1mm SAM2X5 is 2 times more effective neutron poison than the borated stainless steel with 0.12 wt % B. The SAM1651 has less boron its neutron-absorbing effectiveness is comparable to the borated stainless steel.

For comparison, the $k_{eff}$ of a Ni-Gd basket was also calculated. The Ni-Gd basket (0.635 cm thick) contains 2 wt % gadolinium (Gd). Gd is a more effective neutron absorber than B at low neutron energy (i.e., < 0.025 eV). But its absorption capability drops very rapidly between neutron energies from 0.1eV to 1 eV. The Gd cross sections also have a wide resonance region at epi-thermal neutron energy where the prediction of absorption capability varies widely. The calculation result for the Ni-Gd basket indicates that the neutron absorbing effectiveness of Ni-Gd is in between of the 1mm thick SAM2X5 and SAM1651.

**Fast Neutron Radiation Effect**

Several series of SAM melt-spun ribbons were prepared for irradiation experiment in the Neutron Irradiation Container (NIC) and in the core of the TRIGA reactor at MNRC (Figure 2).




(a) (b)

Figure 2 – (a) MNRC Reactor Internal, (b) Container for the Neutron Irradiation Experiment

Table 3 shows the specific ingredients of the SAM2X5 series of ribbons used in the irradiation experiment.

Table 3 – Two series of SAM melt-spun ribbons prepared for irradiation experiment

| | | | | |
|---|---|---|---|---|
| Mo Series<br>DAR40* + Mo | + 1% Mo<br>(SAM2X1) | + 3% Mo<br>(SAM2X3) | + 5% Mo<br>(SAM2X5) | + 7% Mo<br>(SAM2X7) |

Note: * DAR40 – $Fe_{49.7}Cr_{17.7}Mn_{1.9}Mo_{2.4}W_{1.6}B_{15.2}C_{3.8}Si_{2.4}$

The melt-spun ribbons, after X-ray diffraction (XRD) were inserted into the NIF for three irradiation cycles. Table 4 shows the irradiation cycles; time of exposure in reactor; total exposure fluence (defined as the flux multiplied by time) and the equivalent time, in years of radiation exposure of material inside the spent fuel containers in a repository environment.

Table 4 – Two series of SAM melt-spun ribbons prepared for irradiation experiment

| Irradiation cycle (fast flux = $1.5 \times 10^{10}$ (n/cm2-sec) | 1st | 2nd | 3rd |
|---|---|---|---|
| Total time exposed in reactor, min | 44 | 132 | 263 |
| Total exposure fluence, flux x time | $4.3 \times 10^{13}$ | $1.3 \times 10^{14}$ | $2.6 \times 10^{14}$ |
| Equivalent years in a repository | ~670 | ~2000 | ~4000 |

X-ray diffraction (XRD) was used to examine the melt-spun ribbons at each irradiation cycle and after a total cumulative time exposure of 263 minutes at a fast neutron flux of $1.5 \times 10^{10}$ n/cm$^2$-sec. This exposure equates to a total neutron fluence of $2.6 \times 10^{14}$ n/cm$^2$, or an equivalent time in a repository of ~4000 years. A ten-fold increase in fluence was achieved in subsequent irradiation of these ribbons by inserting them into the central location of the reactor core.

The use of XRD is to identify the presence of crystalline phases in these ribbons. The XRD spectra of an amorphous material do not have sharp peaks; whereas, the XRD spectra of a crystalline material or a material that is a mixture of amorphous and crystalline material will have sharp peaks.

Figure 3 shows the post-irradiation XDR results of the two series of SAM melt-spun ribbons. It indicates that the extensive fast neutron irradiation does not change the structure of the amorphous SAM2X melt-spun ribbons.

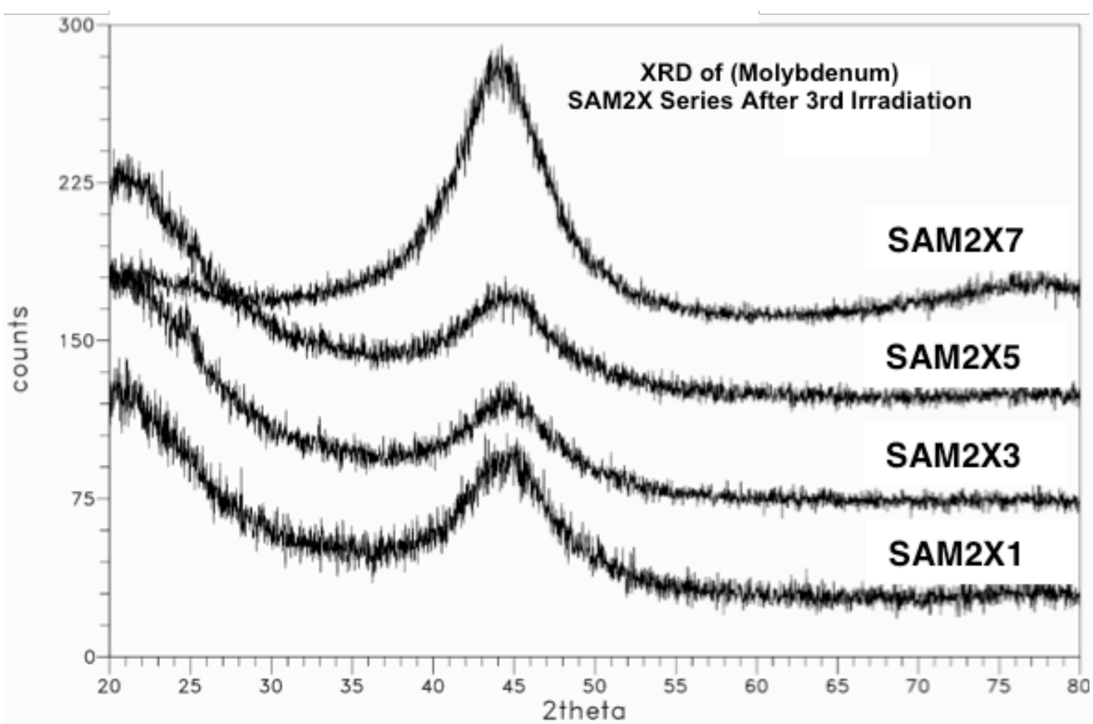



Figure 3 – XRD results of neutron irradiation of SAM2X5 ribbons.

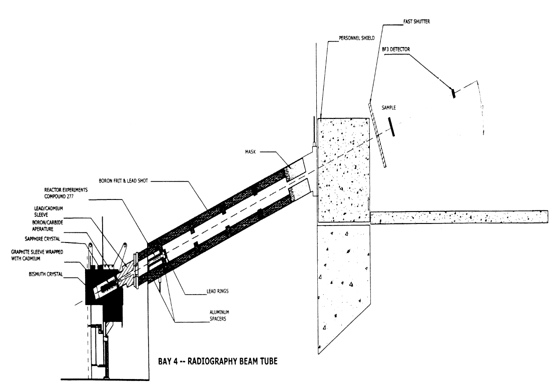
## NEUTRON-ABSORBING EFFECTIVENESS MEASUREMENTS

When thermal neutron passes through an absorbing material, it can be absorbed, scattered away, or transmitted through. The ability of the neutron absorbing material to capture the neutron can be estimated by the transmission measurement. For strong thermal neutron absorbers (e.g., $^{157}$Gd, $^{155}$Gd, and $^{10}$B, etc) the thermal neutron incident intensity is reduced mainly by the absorption of neutron, and minimally by the scattering effect. The transmission intensity is hence, estimated based on the following relationship:

$$
\begin{aligned}
I_t &= I_o * e^{(-\sigma * n * x)} \\
&= I_o * e^{(-\Sigma * x)}
\end{aligned}
$$

where $I_t$ is the thermal neutron transmission intensity
$I_o$ is the incident intensity of the thermal neutron
σ is the microscopic transmission cross section
n is the atom density of the neutron absorbing material (e.g., $^{10}$B, etc.)
x is the thickness of the neutron absorbing material
Σ is the macroscopic cross section, defined as the probability per unit path length that a neutron will interact as it moves about in a medium

Both the transmission and incident thermal neutron intensity can be measured by experiment. These transmission measurements were performed for various neutron absorbers including Boral™, Metamic™, Ni-Gd, and stainless steel substrates coated with SAM2X5 at MNRC). The schematic and experimental apparatus are shown in Figure 4.




(a)

(b)

Figure 4 – (a) Schematic of the Transmission Measurement Apparatus, (b) Transmission Measurement Experiment Set-Up for a Ni-Gd Plate

The results are shown in Table 5 and compared with non-neutron absorbing substrates. It is noted that the SAM2X5 absorbs thermal neutron with an average neutron transmission cross section of about 7.1 cm$^{-1}$. The average $\Sigma_t$ for the Ni-Gd plates from measurements is about 2.3 (and about 5.9 after adjusted for the "flux suppression" effect due to Gd's large absorption cross section). The results indicate the low $\Sigma_t$ for the borated stainless steel plates, due to the low boron content in these plates.

Table 5 - Results of Transmission Measurement of Various Neutron Absorbing Plates

| Plate # | Plate ID | Description | Transmission Count Rate (cpm) | Bare Beam Count Rate (cpm) | Ratio | Transmission Cross Section, $\Sigma_t$, $cm^{-1}$ |
|---|---|---|---|---|---|---|
| 1 | MNRC | Boral 40 mil or 0.1 cm thick | 7550 | 73017 | 0.103 | 22.7 |
| 2* | 316L | Base plate, 1/4” or 0.635 cm thick | 39309 | 77478 | 0.507 | 1.07 |
| 3* | C22 | Base plate, 0.28” or 0.711 cm thick | 31033 | 77478 | 0.401 | 1.29 |
| 4* | SAM2X5 on C-22 (M18W3) | C22 1/8” or 0.317 cm thick with coating by TNC | 26831 | 77478 | 0.346 | 6.52 |
| 5 | SAM2X5 on C-22 (M10S14) | C22 1/4” or 0.635 cm thick with coating by TNC | 14482 | 70644 | 0.205 | 7.65 |
| 6 (1) | NiGd | Labeled “Extra”, 3/8” or 0.952 cm thick | 1948 | 70644 | 0.0276 | 3.77 |
| 6 (2) | NiGd | Labeled “Extra”, 3/8” or 0.952 cm thick | 1897 | 70095 | 0.0271 | 3.79 |
| 7 | Metamic | $B_4C$/ Al, 1/16” or 0.158 cm thick | 4891 | 70644 | 0.0692 | 16.9 |
| 8 | NiGd | Labeled (1), 3/8” or 0.952 cm thick | 1637 | 67700 | 0.0242 | 3.91 |
| 9 | NiGd | Labeled (2), 3/8” or 0.952 cm thick | 1672 | 67700 | 0.0247 | 3.89 |
| 10 | SAM2X5 on 316L-C1 | 316L 1/4" or 0.635 cm thick with coating by PTI | 26037 | 68622 | 0.379 | 5.82 |
| 11 | SAM2X5 on 316L- C2 | 316L 1/4" or 0.635 cm thick with coating by PTI | 24875 | 68622 | 0.362 | 6.73 |
| 12 | SAM2X5 on 316L- W1 | 316L 1/4" or 0.635 cm thick with coating by PTI | 24026 | 67928 | 0.354 | 7.18 |
| 13 | SAM2X5 on 316L- W2 | 316L 1/4" or 0.635 cm thick with coating by PTI | 24263 | 67928 | 0.357 | 7.01 |
| 14 | SAM2X5 on C22- C15 | C22 1/4" or 0.635 cm thick with coating by PTI | 21555 | 67062 | 0.321 | 6.34 |
| 15 | SAM2X5 on C22- C16 | C22 1/4" or 0.635 cm thick with coating by PTI | 19500 | 67062 | 0.291 | 8.30 |
| 16 | SAM2X5 on C22- W15 | C22 1/4" or 0.635 cm thick with coating by PTI | 19876 | 68606 | 0.290 | 8.37 |
| 17 | SAM2X5 on C22- W16 | C22 1/4" or 0.635 cm thick with coating by PTI | 20857 | 68606 | 0.304 | 7.43 |
| 18 | Borated S.S. (182193) | Borated S.S. 5/8” or 1.587 cm thick | 4438 | 63011 | 0.0704 | 1.67 |
| 19 | Borated S. S. (182194) | Borated S.S. 5/8” or 1.587 cm thick | 1904 | 63011 | 0.0302 | 2.21 |
| 20 | Borated S. S. (182196) | Borated S.S. 5/8” or 1.587 cm thick | 1014 | 63011 | 0.0161 | 2.60 |
| 21 | Borated S. S. (03180) | Borated S.S. 5/8” or 1.587 cm thick | 941 | 63011 | 0.0149 | 2.65 |

Note: *Runs at 1.8 MW operating power. Other measurements were obtained when reactor was run at 1.5 MW

Average measured values of the transmission neutron absorption cross section (Σ) for Type 316L stainless steel, Alloy C-22, borated stainless steel, Ni-Gd alloy, and SAM2X5 were determined to be approximately 1.1, 1.3, 2.3, 3.8, and 7.1 $cm^{-1}$, respectively[9]. Data are shown in Figure 5.

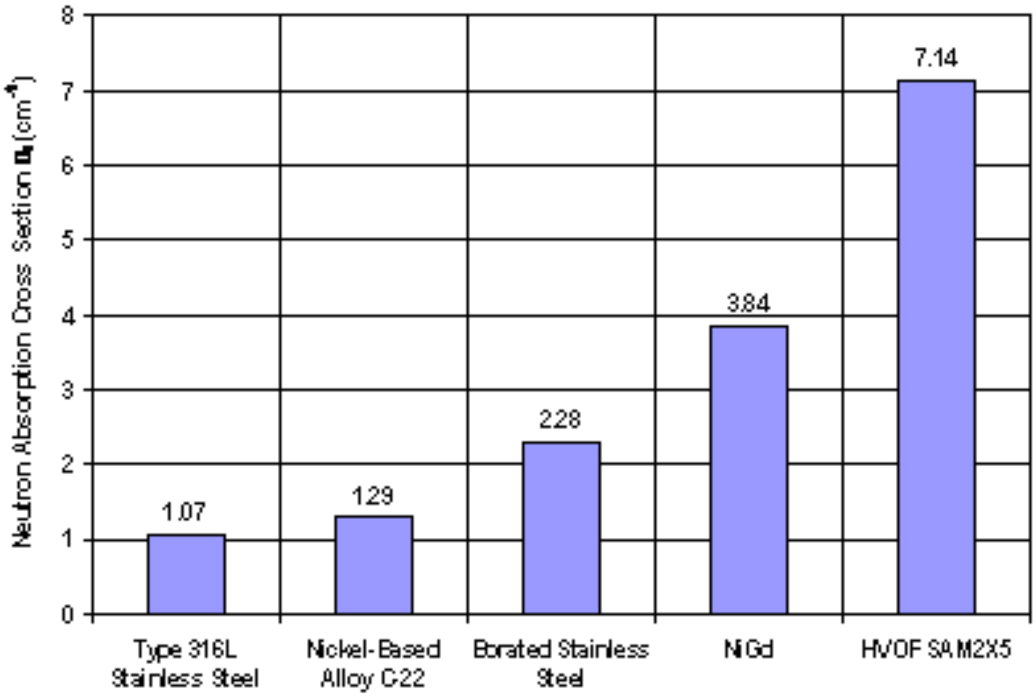



Figure 5 – Average measured values of thermal neutron absorption cross section in various alloys

## CONCLUSIONS

This study concludes that Fe-based structural amorphous metal SAM2X5:

1. The high boron-containing SAM2X5 coating can be an effective criticality control material for the used fuel containers [10,11].
2. The neutron irradiation experiment indicates that the amorphous alloys are stable and their amorphous structures are not changed after exposure to high doses of fast neutron irradiation.
3. The neutron transmission measurements indicate that SAM2X5-costed substrates exhibit effective thermal neutron absorbing capability. The transmission cross-sections of SAM2X5 coatings are three-to-four times higher than that of borated stainless steel, and twice that of the nickel-gadolinium alloy.


## ACKNOWLEDGEMENTS

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