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Authors

Choi, J.
Lee, C.
Farmer, J.
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Application of Neutron-Absorbing Structural-Amorphous metal (SAM) Coatings for Spent Nuclear Fuel (SNF) Container to Enhance Criticality Safety Controls

Jor-Shan Choi[1], Chuck Lee[1], Joseph Farmer[1], Dan Day[1], Mark Wall[1], Cheng Saw[1], Moe Boussoufi[2], Ben Liu[2], Halold Egbert[2], Dan Branagan[3], and Andy D'Amato[4]
[1]Lawrence Livermore National Laboratory, Livermore, CA, 94550
[2]McClellan Nuclear Radiation Center, McClellan, CA, 95652
[3]Nanosteel Company, Idaho Falls, ID, 83402
[4]Plasma Technology Incorporated, Torrance, CA, 90501

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ABSTRACT

Spent nuclear fuel contains fissionable materials (^{235}U, ^{239}Pu, ^{241}Pu, etc.). To prevent nuclear criticality in spent fuel storage, transportation, and during disposal, neutron-absorbing materials (or neutron poisons, such as borated stainless steel, Boral™, Metamic™, Ni-Gd, and others) would have to be applied. The success in demonstrating that the High-Performance Corrosion-Resistant Material (HPCRM)[1] can be thermally applied as coating onto base metal to provide for corrosion resistance for many naval applications raises the interest in applying the HPCRM to USDOE/OCRWM spent fuel management program. The fact that the HPCRM 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 HPCRM qualify the material to be a neutron poison.

INTRODUCTION

Corrosion-resistant, iron-based amorphous metals have been tested to determine their relative corrosion resistance. Many of these materials can be applied as coatings with advanced thermal spray technology. SAM2X5 is an amorphous-metal alloy composition that has been identified as having outstanding corrosion resistance. Because of the high boron content, SAM2X5 can be applied as the neutron-absorbing coatings to the metallic support structure to enhance criticality safety for spent nuclear fuel in baskets inside the storage containers, the transportation cask, and eventually the disposal container for repository disposal. Table I shows the composition of SAM2X5.

Table I Composition of SAM2X5 in Atomic Percent

	Fe	Cr	Mo	Ni	C[1]	W	Mn	Si	B[1]	Gd
SAM2X5	48.8	17.6	7.2	0	3.7	2.5	2.4	2.7	15	0

Note [1]: Weight percent for boron is 3.22, and for carbon is 0.83

To demonstrate the spray coating of the SAM2X5 powders onto a metallic substrate, two half-scale stainless steel 316L basket modules were fabricated. The SAM2X5 powder and the pre-coated basket module plates were sent to Plasma Technology Incorporated (PTI) located in Torrance, CA for thermal-spray by the high velocity oxy-fuel (HVOF) coating method. The HVOF used oxygen and a fuel gas as combustion media to accelerate the melted powder to high velocities through special designed nozzle and torch gun. For the basket module, about ½ mm of SAM2X5 coating was applied to both sides of the stainless steel 316L plates, achieving bond strengths approximately 8,000 psi.

CRITICALITY ANALYSIS

A disposal container designed to hold twenty-one spent PWR fuel assemblies was modeled for criticality analyses. Each of the 21 Westinghouse designed 17 x 17 assembly containing 264 pins of spent UO_2 fuel, and void spaces previously occupied by 24 guide thimbles and one instrumentation tube were modeled. The PWR fuel assembly was modeled at 35 GWd/tonne of burn-up and with 10 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[2]. The criticality analyses for a disposal container model were performed[3]. The results are shown in Table II.

Table II 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	¼" 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. It indicates that the borated stainless steel basket with 0.12 wt. % of boron would drop the k_{eff} to about 4% of that of the no boron 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 boron). The stainless steel basket contains either no or 0.12 wt. % boron. The results indicate that the 1mm SAM2X5 is 2 times more effective neutron poison than the borated stainless steel with 0.12 wt. % boron. 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. Gadolinium is a more effective neutron absorber than boron for low energy neutron (i.e., neutron energy < 0.025 eV). But its absorption capability drops very rapidly with higher neutron energy (starting from E > 0.1eV). The gadolinium cross sections also have a wide resonance region 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.

NEUTRON EFFECT MODELING

Several series of SAM melt-spun ribbons were prepared for irradiation experiment in the Neutron Irradiation Facility (NIF) at the McClellan Nuclear Radiation Center (MNRC). The MNRC, operated by the University of California at Davis (UCD), is a facility operating a 2 Megawatt TRIGA reactor. Table III shows the specific ingredients of the 2 SAM series of ribbons (SAM2X and SAM3X).

Table III Two Series of SAM Melt-Spun Ribbons Prepared for Irradiation Experiment

Mo Series DAR40 + Mo	+ 1% Mo (SAM2X1)	+ 3% Mo (SAM2X3)	+ 5% Mo (SAM2X5)	+ 7% Mo (SAM2X7)
Y Series DAR40 + Y	+ 1% Y (SAM3X1)	+ 3% Y (SAM3X3)	+ 5% Y (SAM3X5)	+ 7% Y (SAM3X7)

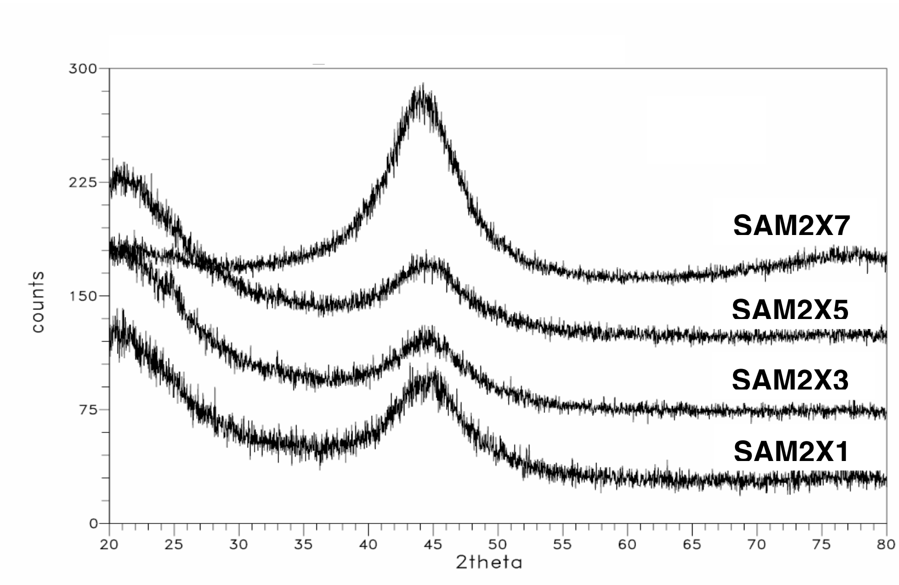
X-ray diffraction is used to examine the melt-spun ribbons after a total cumulative time exposure of 263 minutes at a fast neutron flux of 1.5 x 10^{10} n/cm^2-sec (or an equivalent fast neutron radiation exposure time inside the spent fuel containers in the Yucca Mountain repository environment of about 4000 years). The use of XRD is to identify the presence of crystalline phases in these ribbons. The XRD spectra of an amorphous material does 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 1 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 and SAM3X melt-spun ribbons.

THERMAL NEUTRON TRANSMISSION MEASUREMENT

Thermal neutron transmission measurements were performed at MNRC for a variety of metal plates with or without neutron absorbers. Preliminary results of transmission measurements of various neutron absorbing materials are obtained. The results are shown in Table IV 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 Σ_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 Gadolinium's large absorption cross section). The results indicate the low Σ_t for the borated stainless steel plates, due perhaps to the low boron content in these plates.

XRD of (Molybdenum)
SAM2X Series After 3rd Irradiation



XRD of (Yttrium)
SAM3X Series After 3rd Irradiation

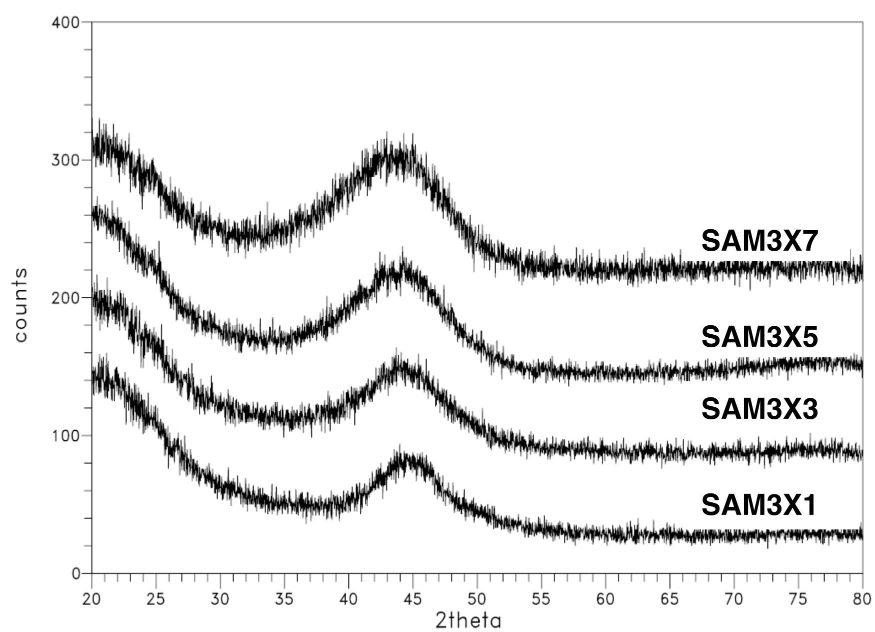


Figure 1 XRD Results of Neutron Irradiation of SAM Ribbons

Table IV Preliminary 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, Σ_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

SUMMARY

1. The high boron-containing SAM2X5 coating can be an effective criticality control material for the spent fuel containers. The HVOF thermal-spray process is a demonstrated technology to apply SAM coating onto alloy substrates.

2. The neutron irradiation experiments indicate that extensive fast neutron irradiation does not change the structure of the amorphous SAM2X5 melt-spun ribbons.

3. The neutron transmission measurements indicate that SAM2X5 exhibit effective neutron absorbing capability, similar to Boral™ and Metamic™.

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2. MCNP5, developed by the Los Alamos National Laboratory, is a general **M**onte **C**arlo **N**-**P**article code. MCNP5 was released to ORNL/RSICC and made available to people within the US in April 2003. This release is identified as MCNP5_RSICC_1.14.

3. J. S. Choi, J. C Farmer, C. K. Lee, "Applications of Neutron-Absorbing Amorphous Metal Coatings for Spent Nuclear Fuel (SNF) Containers: Use of Novel Coating Materials to Enhance Criticality Safety Controls," UCRL-MI-220385, Lawrence Livermore National Laboratory, Livermore, CA, June 16 (2006).