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Abstract—*The National Spent Nuclear Fuel Program, located at the Idaho National Engineering and Environmental Laboratory, coordinates and integrates national efforts in management and disposal of U.S. Department of Energy (DOE)-owned spent nuclear fuel. These management functions include using the DOE standardized canister for packaging, storage, treatment, transport, and long-term disposal. Nuclear criticality control measures are needed in these canisters because of the enrichment and total quantity of fissile material in some types of the DOE spent nuclear fuel.*

This paper will report the test results of one alloy heat from a metallurgical development program that is developing nickel-chromium-molybdenum-gadolinium alloys for nuclear criticality control in the DOE standardized canister. Gadolinium has been chosen as the neutron absorption alloying element due to its high thermal neutron absorption cross section. The microstructure, mechanical properties, and corrosion resistance of various alloys will be presented. These corrosion resistant, structural alloys can be used to fabricate components of spent nuclear fuel storage racks, storage canisters and internal structural baskets, and transportation cask internals.

The focus of this work is to qualify these materials for American Society of Mechanical Engineers code qualification and acceptance in the Yucca Mountain Repository.

I. INTRODUCTION

Safe, long-term storage and disposal of the U.S. Department of Energy (DOE)—owned spent nuclear fuel requires a corrosion resistant, long—lasting material that will absorb emitted neutrons for nuclear criticality control. DOE's National Spent Nuclear Fuel Program (NSNFP) at the INEEL is developing a corrosion-resistant, Nickel-Chromium-Molybdenum alloy containing gadolinium for criticality control in the DOE standardized spent nuclear fuel storage canister. These canisters will be stored in the waste package at the Yucca Mountain Repository. Gadolinium (Gd) is a potent neutron-absorbing element which has the highest available neutron absorption cross section. To meet the functional requirements for a structural material that will be used as an insert in the standardized canister, gadolinium must be alloyed into a corrosion resistant structural metal that will meet ASME code requirements. The criticality safety analysis results push the alloy development towards the highest obtainable gadolinium level.

The initial alloy development used 316L stainless steel as the base metal. It was found that there would be severe hot fabricability and localized corrosion problems with this approach.[1,2] It was also found that the gadolinium has no solubility in the matrix of a stainless steel or a nickel-based alloy and is present as a gadolinium rich second phase called a gadolinide. This has implications for the corrosion resistance and mechanical properties of the resulting alloys.

From earlier work with borated stainless steels,[3] it was found that the ductility of the alloy would decrease with increased volume fraction of the boron rich second phase. To balance the goals of the highest possible gadolinium level, adequate corrosion performance, and meeting the ASME code requirements; we are investigating control of the size, shape and distribution of the gadolinium rich second phase. The techniques being used are initial melt chemistry control, molten metal secondary refining techniques (Vacuum Arc Remelting) and thermo mechanical processing treatments.

II. PURPOSE AND NEED

Approximately 2,500 metric tones of spent nuclear fuel (SNF) managed by the Department of Energy (DOE) is planned for geologic disposal at Yucca Mountain. This SNF is much different from the commercial SNF produced by the power generation industry. DOE SNF has a variety of enrichments ranging from natural to fully enriched. The higher enrichments presented a significant challenge for disposal packaging to avoid a conservative fissile loading limit.

NRC regulations for the licensing of a geologic repository at Yucca Mountain require that criticality events be evaluated consistent with the risk-informed nature of 10 CRF 63. For preclosure, criticality must be prevented whereas for post closure, criticality must be evaluated in consideration of its probability and consequences as part of the overall performance assessment.

DOE SNF is expected to be co-disposed of in waste packages with vitrified high-level waste (HLW). This minimizes the amount of fissile material in a single waste package and takes advantage of a formally open space in the center of the HLW canisters. DOE SNF will be packaged such that single canisters are critically safe for all storage and transportation events. However, when DOE SNF is evaluated in degraded conditions as is expected in geologic timescales, an insoluble and long-life poison was needed to avoid an extremely conservative fissile loading limit. Other poisons available such as boron compounds are highly soluble in the expected Yucca Mountain underground environment and were postulated to be easily separated from the fissile material. An extensive review of options identified gadolinium compounds as a potential solution. As the gadolinium compound in the alloy degrades, the gadolinium will chemically combine with phosphate compounds in the waste package producing an essentially insoluble neutron poison that will be stable over geologic timescales

The deployment of gadolinium into the DOE SNF canisters considered a range of options. These included, gadolinium basalt compounds, gadolinium spray coatings, loose fill gadolinium compounds, and gadolinium alloys. The option that best met packaging and long-term disposal needs was gadolinium deployment in a highly corrosion resistant alloy. The alloy will then be fabricated into fuel basket assemblies as shown in Figure 1. This deployment method supports interim storage and transportation if needed, allows for easy retrieval, and introduces a highly insoluble gadolinium compound into the waste package.

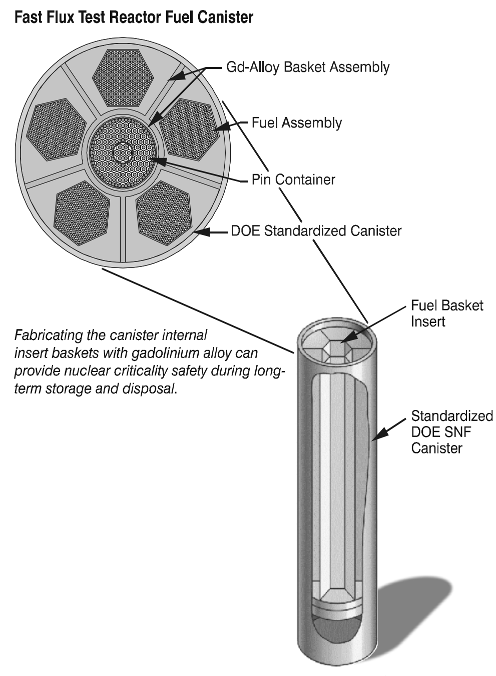

Figure 1 DOE Standardized Canister

III. WORK DESCRIPTION

The tasks required for the development of these new alloys are shown in Figure 2. We formulate and fabricate the alloys through the complete processing cycle from initial melting charge makeup, primary melting in a vacuum induction furnace, secondary refining in a Vacuum Arc Remelt (VAR) process, and thermo-mechanical processing which includes all steps necessary (hot working, heat treatment) to turn the cast ingot into a wrought plate. The testing program is presently concentrating on the measurement of the mechanical properties, corrosion resistance, neutron absorption capability and weldability of the alloys. The mechanical properties samples (tensile and Charpy Impact) and corrosion test specimens are machined from plates fabricated in the test program. This paper describes results from Heat HV 9810A that has the following chemistry in weight percent; Gd-1.58, Mo-14.16, Cr-16.21, Fe-0.147, C-0.011 and Ni-68.5.

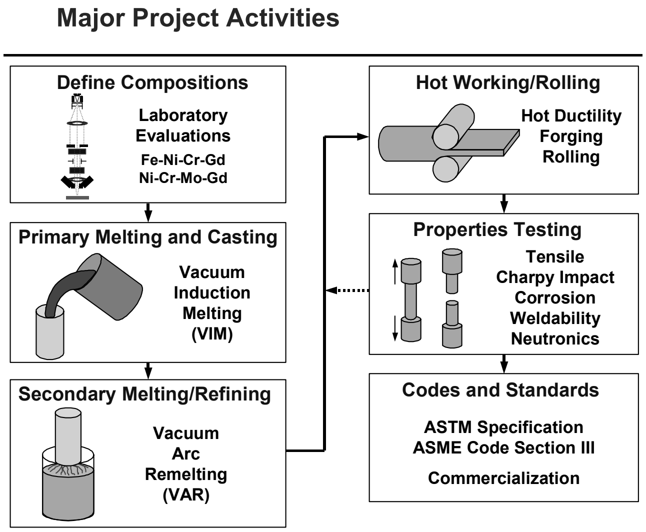

Figure 2. Project workflow.

IV. RESULTS

IV.A. Microstructure

The microstructural features of the HV9810A plate are described in this section. Figure 3 shows light optical micrographs of the HV9810A plate in the three principal orientations; rolling plane, transverse to the rolling direction, and longitudinal to the rolling direction. The gadolinium has no solubility in the austenite phase of the Ni-Cr-Mo base alloy structure and solidifies as a gadolium rich second phase. The composition of the second phase (light gray) of the rolled plate was measured with electron probe microanalysis and electron backscattered diffraction (EBSD) which identified the second phase as a Ni5Gd intermetallic mixed with other dissolved elements such as chromium, molybdenum, and iron. Gadolinium oxides are also apparent in all three orientations and are visible as small black specs distributed throughout the matrix. It should be noted that this particular heat did not undergo the VAR treatment.

Additional work to be performed includes detailed microstructural characterization of a number of alloy heats using a variety of electron microscopy techniques. This data will be used be to understand the influence of thermo-mechanical processing on microstructure. The microstructure will then be related to the measured mechanical properties.

IV.B Corrosion Testing

Electrochemical (Cyclic Polarization) corrosion testing methods were used to test samples of alloy HV9810 A in the following environments and a simulated Yucca Mountain Project chemistry: (a) 0.1 M HCl, 30°C, (b) 0.1 M HCl, 60°C, (c) 0.028 M NaCl, 30°C, (d) 0.028 M NaCl, 60°C, (e) Yucca Mountain J-13 solution, 30°C. The acidic chloride solutions were chosen for known propensity to initiate localized corrosion and are a bounding case for early waste package failure. The J-13 solution is considered to be the Yucca Mountain, in-drift chemistry at the end of the regulatory period (10,000 years) in a fully flooded condition.

The corrosion test results show that the alloy will be subjected to initial loss of gadolinium through localized attack of the gadolinides that intersect the exposed metal surface. Figure 4 shows an unexposed corrosion test sample. Figure 5 shows the sample after exposure to the J-13 solution with partial removal of the gadolinides. The testing has shown that the corrosion rate will then drop off to very low levels as the newly exposed metal surface (Ni-Cr-Mo matrix) passivates after removal of the second phase. Additional electrochemical and long term immersion corrosion testing is scheduled to be performed.

IV.C Mechanical Properties Testing

The strength values for the HV9810A alloy are similar to those expected for commercial Ni-Cr-Mo corrosion resistant alloys, and will be suitable for repository applications (Table I). In general, the alloys exhibit slightly higher strength properties with slightly reduced ductility. The Charpy Impact testing data is shown in Table II. The ASME Boiler and Pressure Vessel Code requirements[4] for impact toughness in nuclear applications call for minimums of 20 J (15 ft-lb) impact energy and 0.38 mm (0.015 inch) lateral expansion, and it is clear that the longitudinal orientation for the HV9810A alloys easily meets this requirement. Impact toughness in the transverse orientation is marginal in this respect. For these alloys impact toughness is controlled by the particle shape and spatial distribution, as well as volume fraction. Additional measurements will be made on alloys with differing gadolinium contents and the results correlated with the microstructural characterization studies.

IV.D Welding

Initial welding tests were performed with the electron beam (EB) and gas tungsten arc (GTAW) processes. Figure 5 shows light optical photomicrographs of the autogeneous electron beam weld made on the 9810A alloy in the as-polished and etched conditions. The weld exhibits columnar grains, which grow epitaxially from the base metal, and this grain morphology is typically observed in fusion welds. No solidification cracks or other defects were observed in this weld. Additional work with these alloys will involve Varestraint solidification cracking tests in order to determine the weldability of these alloys. The results obtained will be compared with previous results generated on commercial stainless steels, Ni base alloys, and Gd-containing alloys. In conjunction with the microstructural work described earlier, additional Differential Thermal Analysis and various microscopy techniques will be used for microstructural characterization in order to understand the influence of composition on the resultant weld metal microstructure and weldability.

V Neutron Absorption Measurement

Neutronic evaluation of 2% gadolinium alloyed samples has been completed at the Los Alamos National Laboratory Planet Critical Facility. As expected, with a massive thermal neutron cross-section, the test specimens performed well. A full report of these tests will be issued in late spring 2003.

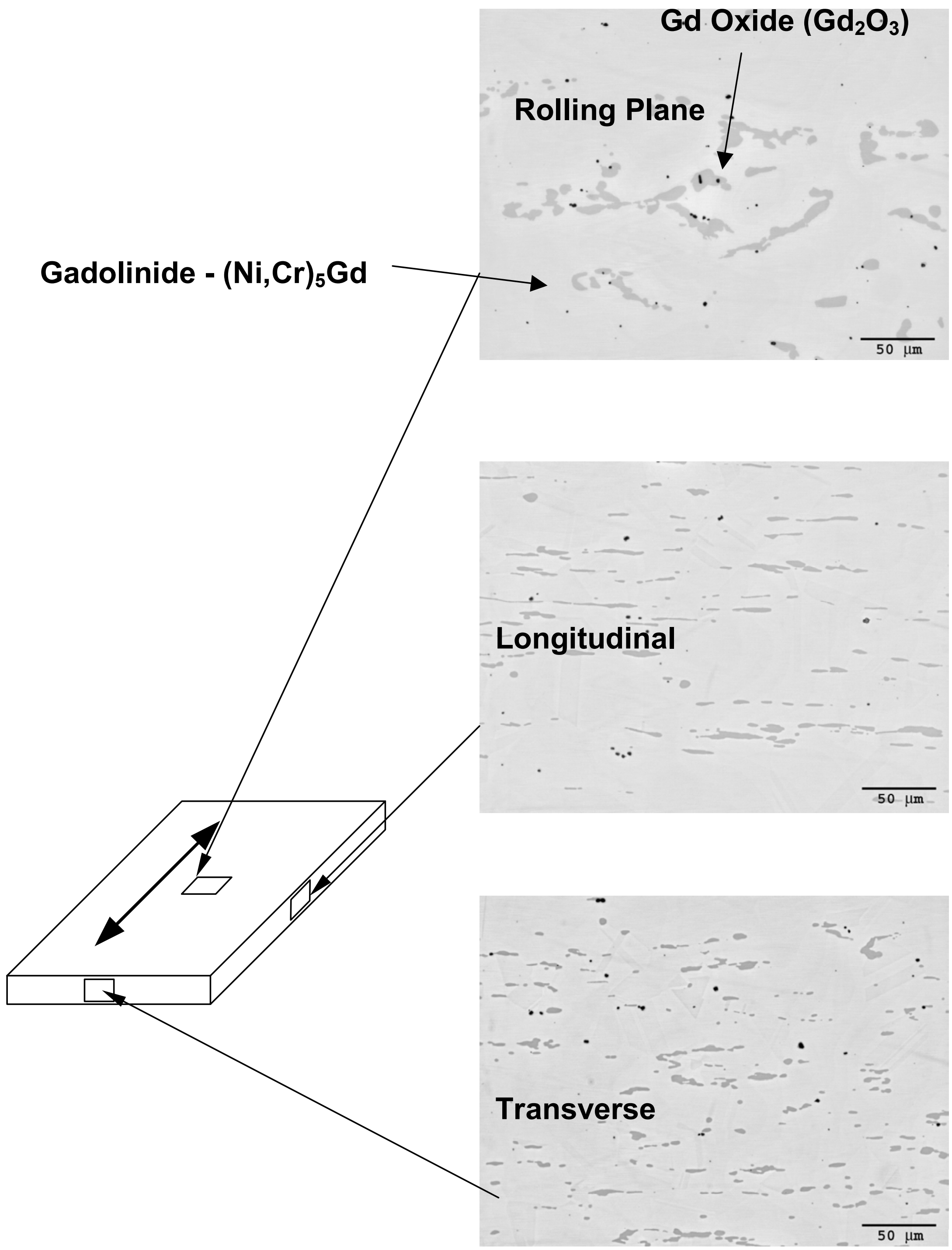

Figure 3 Light optical micrographs of HV9810A plate in the three principal orientations.

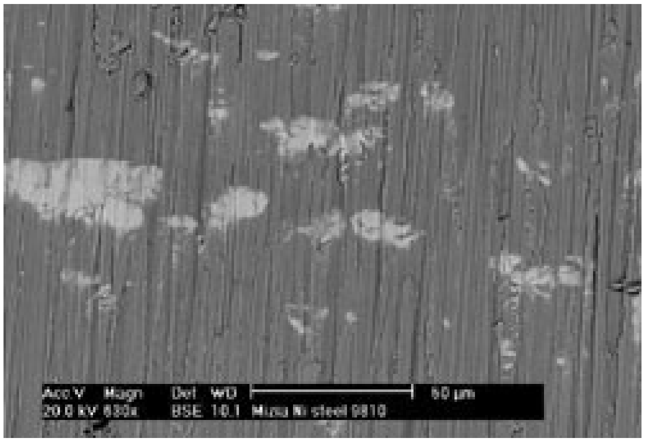

Figure 4. Unexposed corrosion sample, SEM- BSE, gray particles are $(Ni,Cr)_5Gd$, black particles are Gd_2O_3.

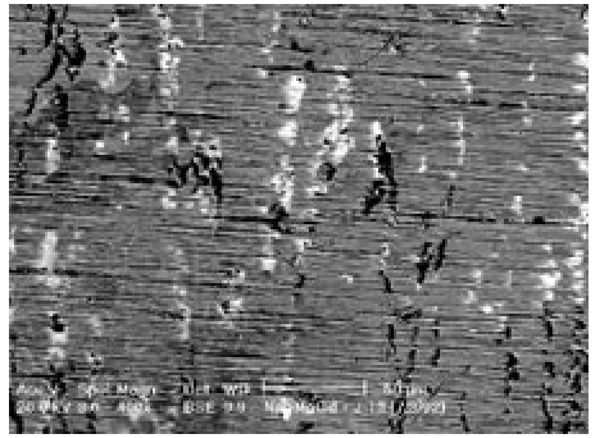

Figure 5 Sample after exposure to J-13 water at 30°C, SEM-BSE, $(Ni,Cr)_5Gd$ and Gd_2O_3 removed from surface.

Table I Summary of tensile properties for HV9810A

			0.2% Offset				
Alloy	Orientation	Temperature °C (°F)	Yield Strength[a] Mpa (ksi)	Tensile Strength[a] Mpa (ksi)	Uniform Strain[a] (%)	Total Strain[a] (%)	RA[a] (%)
HV9810A	Long	23 (75)	372 (54.0)	814 (118.1)	41.7	44.4	38.4
HV9810A	Trans	23 (75)	380 (55.1)	789 (114.5)	32.6	33.3	28.8
HV9810A	Long	350 (660)	276 (40.0)	709 (102.9)	36.9	39.7	41.2
HV9810A	Trans	350 (660)	289 (41.9)	703 (102.0)	33.3	35.1	31.4

a. Average of three tests per condition.

b. All failures outside of gauge length.

Table II. Summary of Charpy Impact values for heat 9810A.

Alloy	Orientation	Temperature °C (°F)	Impact Energy[a] J (ft-lb)	Lateral Expansion[a] mm (inch)
HV9810A	Long	-40 (-40)	34.4 (25.4)	0.66 (0.026)
HV9810A	Trans	-40 (-40)	18.4 (13.6)	0.39 (0.016)
HV9810A	Long	23 (73)	33.8 (24.9)	0.68 (0.027)
HV9810A	Trans	23 (73)	19.8 (14.6)	0.45 (0.018)
HV9810A	Long	150 (302)	38.9 (28.7)	0.73 (0.029)
HV9810A	Trans	150 (302)	21.0 (15.5)	0.50 (0.020)
HV9810A	Long	300 (572)	44.9 (33.1)	0.70 (0.028)
HV9810A	Trans	300 (572)	28.9 (21.3)	0.53 (0.021)
HV9810A	Long	450 (842)	50.6 (37.3)	0.67 (0.027)
HV9810A	Trans	450 (842)	32.4 (23.9)	0.57 (0.023)

a. Average of two tests per condition.

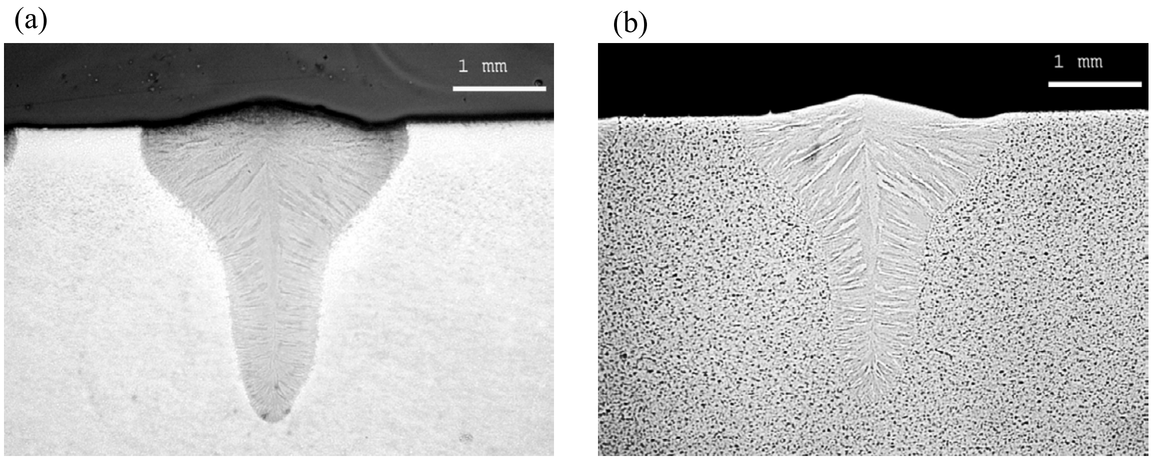

Figure 6 Light optical photomicrographs of an autogeneous electron beam weld made on the 9810A alloy in the as-polished (a) and etched (b) conditions.

IV. CONCLUSIONS AND DISCUSSION

The corrosion resistance of these alloys will be dependent on the amount of gadolinium addition, which will determine the amount of the gadolinide that will be present in the alloy matrix. The gadolinides that intersects the surface exposed to aqueous solutions simulating the Yucca Mountain J-13 solution will be preferentially attacked and removed but the underlying Ni-Cr-Mo matrix will then repassivate and the corrosion rate will drop off to an extremely low rate.

The mechanical property performance (Charpy Impact) may be affected by our simulating the standard commercial practices used for Ni-Cr-Mo alloys. We are constrained by the fact that our ingots (6 inch diameter) do not receive the same amount of hot work that a common commercial size ingot (18” to 24” diameter) would receive in being reduced to a plate product of 3/8” thickness. As a result, the amount of secondary phase refinement due to hot working may be less in our pilot size ingots than in full-scale commercial ingots. Thus, we may be in the unusual situation of having our small-scale results being overly conservative as compared to what may be obtainable with large-scale commercial practice. We are therefore evaluating methods to replicate large-scale hot working procedures in smaller ingots, while at the same time maximizing the alloy mechanical properties.

The initial welding studies gave positive results, which will be confirmed with further testing.

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