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S. M. Frank
D. D. Keiser, Jr.
K. C. Marsden

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Immobilization of Technetium in a Metallic Waste Form

*S. M. Frank, D. D. Keiser, Jr., and K. C. Marsden

Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415-6150, USA

*Tel: 208-533-7391, Fax: 208-533-7471, E-mail: steven.frank@inl.gov

ABSTRACT

Fission-product technetium accumulated during treatment of spent nuclear fuel will ultimately be disposed of in a geological repository. The exact form of Tc for disposal has yet to be determined; however, a reasonable solution is to incorporate elemental Tc into a metallic waste form similar to the waste form produced during the pyrochemical treatment of spent, sodium-bonded fuel. This metal waste form, produced at the Idaho National Laboratory, has undergone extensive qualification examination and testing for acceptance to the Yucca Mountain geological repository. It is from this extensive qualification effort that the behavior of Tc and other fission products in the waste form has been elucidated, and that the metal waste form is extremely robust in the retention of fission products, such as Tc, in repository like conditions. This manuscript will describe the metal waste form, the behavior of Tc in the waste form; and current research aimed at determining the maximum possible loading of Tc into the metal waste and subsequent determination of the performance of high Tc loaded metal waste forms.

I. INTRODUCTION

A major goal of the Global Nuclear Energy Partnership Strategic Plan is to develop, demonstrate and deploy advanced technologies to recycle spent nuclear fuel [1]. Advanced methods of reprocessing of spent nuclear fuel for actinide recovery, such as the aqueous UREX+ separation or pyrochemical methods, also requires the immobilization of isolated fission products for disposal. The fission-product radionuclide technetium-99 is of particular concern because of its relatively high concentration among the fission products and technetium's long half-life of 2.1×10^5 years. Furthermore, technetium is highly soluble in geologic ground waters as the stable pertechnetate anion TcO_4^- and can diffuse rapidly through geological systems [2]. Thus it is essential to immobilize Tc to minimize release from the repository.

In the case of the UREX+ method [3], fission-product Tc remains in the uranium stream after spent-fuel processing and Tc must be removed from the uranium

product. Once the Tc is removed from the uranium product, a reasonable solution for Tc immobilization is to incorporate elemental Tc into a corrosion-resistant metallic waste form. This has been the approach of spent-fuel cladding consolidation and immobilization of various fission-product radionuclides during pyrochemical processing of spent Experimental Breeder Reactor-II (EBR-II) fuel at the Idaho National Laboratory (INL) [4]. This waste material, known as the metal waste form (MWF), has been well characterized and tested for acceptance to the Yucca Mountain geological repository for permanent disposal.

II. BASELINE METAL WASTE FORMS

Development of the metal waste form at Argonne National Laboratory and the INL has resulted in two baseline, multiphase alloys. The two alloys types are a stainless steel with 15 weight percent zirconium (designated SS-15Zr) and a zirconium with 8 weight percent stainless steel (Zr-8SS). Production of these alloys is based on the processing and consolidation of either stainless steel clad or zircaloy clad spent fuel. The high-level metal waste form currently being produced at the Idaho National Laboratory results from the treatment of stainless steel clad fuel and contains approximately 1 weight percent (wt%) fission products. This type of metallic waste form is very effective in the immobilization of second- and third-series transition metal fission products, such as Tc, in the elemental state. For example, developmental SS-15Zr metal waste form alloys have been produced containing up to 2 wt% technetium, and up to 5 wt% surrogate fission product elements (Re, Nb, Ru, Rh, and Pd), and as high as 12 wt% loadings of actinide elements and surrogate fission products without altering the structure or properties of the base alloy. It is therefore believed that Tc loading of 10 wt% or greater can be incorporated into the SS-15Zr alloy without changing the basic properties of the waste form. Furthermore, incorporation of elemental Tc into the stainless steel- zirconium multiphase alloy is readily accomplished at reasonable melt temperatures using standard furnaces and crucibles. The resulting SS-15Zr alloy is very durable, corrosion resistant, and has very low release rates of Tc.

The primary question currently under investigation with regard to incorporation of Tc into a metallic waste form is: what is the greatest amount of elemental Tc that can be incorporated into both the SS-15Zr and Zr-8SS metallic waste forms without altering the basic structure, phase composition or behavior of the waste form? Initial investigation, presented here, has involved producing highly loaded MWF containing rhenium acting as a surrogate for Tc. From these efforts, future investigations will involve producing targeted, high-loaded technetium metal waste forms with subsequent determination of the physical, chemical, corrosion and radionuclide release properties of the Tc waste form.

II.A. Production of the Metal Waste Form

Metal waste forms have been produced on three scales: developmental-scale MWFs; demonstration-scale MWFs; and large-scale MWFs. Developmental-scale MWFs have been produced in both resistive and induction heated furnaces at approximately 15 g charges and larger sizes of approximately 1 to 3 kg. Developmental-scale MWF alloys of SS-5Zr, SS-15Zr and SS-20Zr (5, 15 and 20 wt% Zr in stainless steel, respectively) were produced by placing zirconium metal in the bottom of a cylindrical yttria (Y_2O_3) crucible, and pieces of stainless steel on top of the Zr. As the stainless steel melts, it wets the Zr metal and induces melting of the Zr at a temperature of approximately 1600°C, much lower than the normal melting temperature of Zr metal at 1855°C. Loading and melting of the crucible charge was performed under an inert Ar atmosphere. After cooling, the ~15 mm in diameter by ~35 mm height alloy ingots were removed from the crucibles and sectioned for metallurgical examination and corrosion testing. Developmental-scale waste forms were also produced with surrogate fission products (Nb, Ru, Rh, Pd and Re) and/or actinide elements, and Tc. With regard to ingot production incorporating Tc, chemical analysis of the alloys indicated that no measurable Tc was lost during melting due to volatilization. The larger sized developmental-scale MWF ingots were used for mechanical, corrosion and thermophysical characterization.

Demonstration-scale ingots were prepared using EBR-II spent-fuel cladding after pyrochemical treatment in a electrorefiner in support of the Spent-Fuel Treatment Demonstration Project for the U.S. Department of Energy. Residual electrorefiner salt was removed by volatilization from the cladding hull segments at 1100°C under reduced pressure. After the salt was removed, the cladding hulls and additional Zr metal were placed in a yttria crucible for consolidation in a casting furnace. Typical processing conditions were approximately 1620°C with hold times of up to three hours at temperature.

From experience gained during the MWF development work and Demonstration Project, a new metal waste furnace has been designed, fabricated, tested and qualified for the production of large-scale MWF ingots [5]. The metal waste furnace incorporates salt distillation and cladding hull melting/consolidation into one furnace and produces ingots in the 75 kg range. The furnace utilizes a composite crucible of graphite coated with an aluminum refractory to resist chemical attack. The furnace also has a condenser assembly, located above the crucible, for the condensation of electrorefiner-salt vapor produced during the salt distillation step. This furnace is currently being installed into an inert-atmosphere, hot-cell facility.

II.B. Qualification of the Metal Waste Form

Waste form qualification for acceptance to a geological repository must meet minimum guidelines with respect to physical characteristics and consistency, and must have acceptable corrosion behavior. The waste form must also undergo process qualification to verify product consistency. To acquire this information, a test matrix, shown in Table I, was generated to qualify the MWF for repository acceptance. This test matrix was based on methodology outlined in the American Society for Testing and Materials document C 1174-91 [6]. The data obtained from this testing scheme provided information on the properties of the MWF, supported development of a MWF corrosion model, and address U.S. Department of Energy – Civilian Office of Radioactive Waste Management requirements, which include data needs for repository performance assessment and repository required waste form documentation. For process qualification, 5 MWF ingots were produced weighing from approximately 60 kg. These ingots were sampled followed by chemical analysis and metallurgical examination to confirm product consistency.

II.C. Metallurgy and Microstructure of the Metal Waste Form

The baseline SS-15Zr MWF is a multiphase alloy comprised of two Fe rich solid-solution phases and several $ZrFe_2$ -type intermetallic phases. The thermodynamic behavior of the Fe-Zr binary system is expressed in the binary phase diagram in Figure 1 where these phase regions are identified. The two solid-solution phases are ferrite, a body-centered cubic (bcc) phase also referred to as α -Fe, and austenite a face-centered cubic (fcc) phase referred to as γ -Fe. The formation of one Fe solid-solution phase over the other depends on the Ni and Zr content of the alloy. Cladding stainless steel of 304 and 316 are austenitic in structure due to the relatively high levels of Ni that stabilizes the austenite phase in these steels; whereas, HT9 is ferritic/martensitic in

structure due to lower levels of Ni. Nickel, however, prefers the $ZrFe_2$ intermetallic phase in the MWF. Therefore, alloys produced with 304 or 316 stainless steel cladding, and with lower levels of Zr (less than 20wt%) will predominate in the austenitic solid solution phase; whereas, MWFs produced with HT9 cladding and/or with high levels of Zr will predominate in the ferrite solid-solution phase. The $ZrFe_2$ -type intermetallic phases observed in the MWF are actually $Zr(Fe,Cr,Ni)_{2+x}$ Laves polytypes C15, C14, C36 and 9H of varying concentrations and a $Zr_6(Fe,Cr,Ni)_{23}$ phase [7, 8].

The typical microstructure of the baseline SS-15Zr MWF is illustrated in Figure 2 showing a mixture of fine and coarse eutectic morphology. The $ZrFe_2$ -type intermetallic phases are white or light gray in color and the darker areas are the iron rich, solid-solution phases. No discreet actinide or NMFP phases have been observed in any MWF containing actinide elements, or noble-metal fission products. Actinide elements are preferentially incorporated into the $ZrFe_2$ intermetallic phases, while the NMFPs are distributed in either one or both of the main solid-solution and intermetallic phases. Table II shows the distribution of noble-metal fission products and activation products in the MWF.

Table I. Metal Waste Form Test Matrix

Test Category	Sample Type			
	Non-Radioactive, Variable Composition	Non-Radioactive, Variable Processing	Radioactive (Tc, U, Np, Pu) Surrogate NMFP Variable Composition	Radioactive Waste Ingots Produced from EBR-II Spent Fuel
Attribute:				
Scanning Electron Microscopy	P	P	P	P
Transmission Electron Microscopy	P		P	
X-ray Diffraction	P	P	P	
Neutron Diffraction	P		P	
Thermal Expansion	P	P		
Thermal Conductivity	P	P		
Thermal Diffusivity	P	P		
Specific Heat	P	P		
Density	P	P	P	P
Tensile Strength	P			
Compressive Strength	P	P		
Impact Strength	P			
Chemical Analysis	P		P	P
Characterization:				
Immersion Test	P		P	
Pulsed Flow Test	P		P	
pH Excursion	P		P	
Chloride Excursion	P		P	
Conc. J-13 Excursion	P		P	
Temperature Excursion	P			
Accelerated:				
Electrochemical	P	P	P	
Vapor Hydration	P		P	
High-Temp Immersion	P	P		
Chloride Excursion	P		P	
Conc. J-13 Excursion	P		P	
Service Condition:				
Thermal Aging	P			
Drip Tests	P		P	
Materials Interaction Tests	P			
Galvanic Interaction Tests	P			
Crevice Corrosion Tests	P			
Pitting Susceptibility	P			
TCLP				
TCLP for RCRA Metals	P		P	P
Mechanistic Study:				
Transmission Electron Microscopy	P		P	

P: Performed measurement

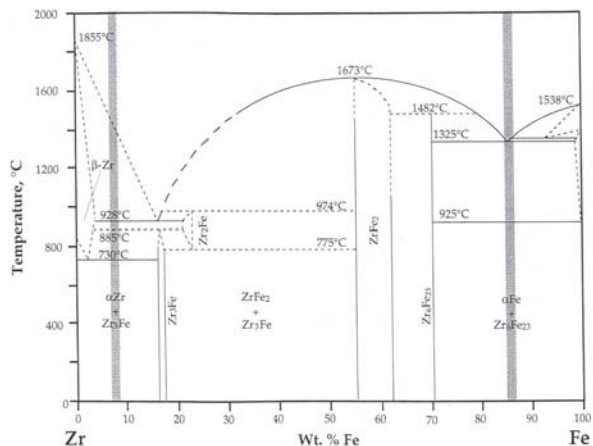


Figure 1. Fe-Zr phase diagram with the baseline SS-15Zr alloy phase composition region outlined in gray on the right-hand side of diagram and the high-zirconium alloy region outlined in gray shown on the left side of the diagram. Figure based on reference [9].

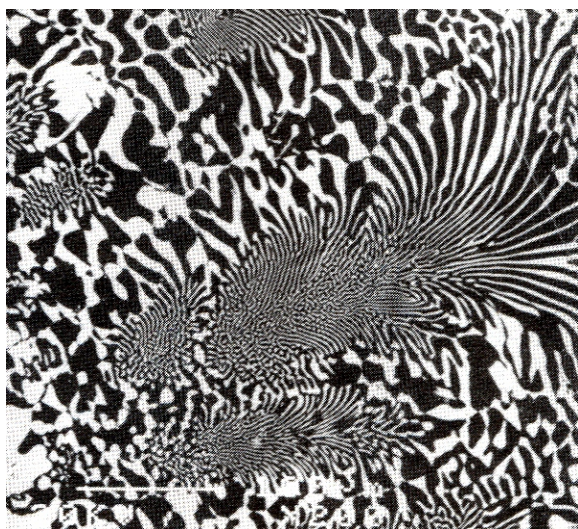


Figure 2. Representative backscatter electron (BSE) image (200x) of baseline SS-15Zr alloy showing eutectic morphology of iron solid-solution phases (black) and the $ZrFe_2$ intermetallic phases (white).

II.D. Corrosion Behavior of the Metal Waste Form

Radionuclide release rates from a waste form are dependent on the environmental conditions of the repository, the metallurgy of the waste form and surface corrosion layer, the corrosion and degradation mechanisms of waste form, and the mechanisms of radionuclide release from the corrosion layer. For the Yucca Mountain Project Viability Assessment and licensing requirements, the waste form generator must provide to the repository a model that predicts the corrosion behavior and long-term release of radionuclides

Table II. Phase Distribution of Activation Products and Noble Metal Fission Products in the Baseline SS-15Zr MWF

Element	Preferred Phase	Approximate Percent Distribution	
		Intermetallic	Fe Solid Solution
Co	Both Phases Equally		
Mn	Both Phases Equally		
Mo	Both Phases Equally		
Nb	Intermetallic	90%	10%
Tc	Fe Solid Solution	30%	70%
Ru	Intermetallic	90%	10%
Rh	Intermetallic	90%	10%
Pd	Intermetallic	90%	10%
Ag	Intermetallic	80%	20%
Sn	Intermetallic	90%	10%
Ta	Intermetallic	90%	10%
W	Both Phases Equally		
Re	Both Phases Equally		

from the waste form to the repository environment [10]; with the model containing aspects of the waste forms growth of a passive oxide layer, long-term uniform aqueous corrosion, release of actinides and fission products from the oxide layer under long-term uniform aqueous corrosion, passivity breakdown via crevice corrosion, and release under conditions of passivity breakdown. Thus, a major aspect of MWF qualification for repository acceptance has been to generate a mechanistic model for the waste form's long-term performance. Currently, the body of experimental data required to develop a model that fully encompasses all the mechanisms of corrosion and release is incomplete, particularly with regard to crevice corrosion and behavior of the passive oxide layer. The current empirical model that predicts the overall corrosion behavior of the MWF is based on: (1) the uniform aqueous corrosion rate of the waste form determined by a linear-polarization electrochemical method [11], and (2) the release rate of radionuclides from the waste form obtained by immersion methods either in demineralized water, solutions of simulated ground water (J13), water of specific pH or ion concentration; and all of these solution types at various temperatures. For the metal waste form in Yucca Mountain repository-type conditions, it has been concluded that uniform aqueous corrosion is the main degradation mechanism for radionuclide release.

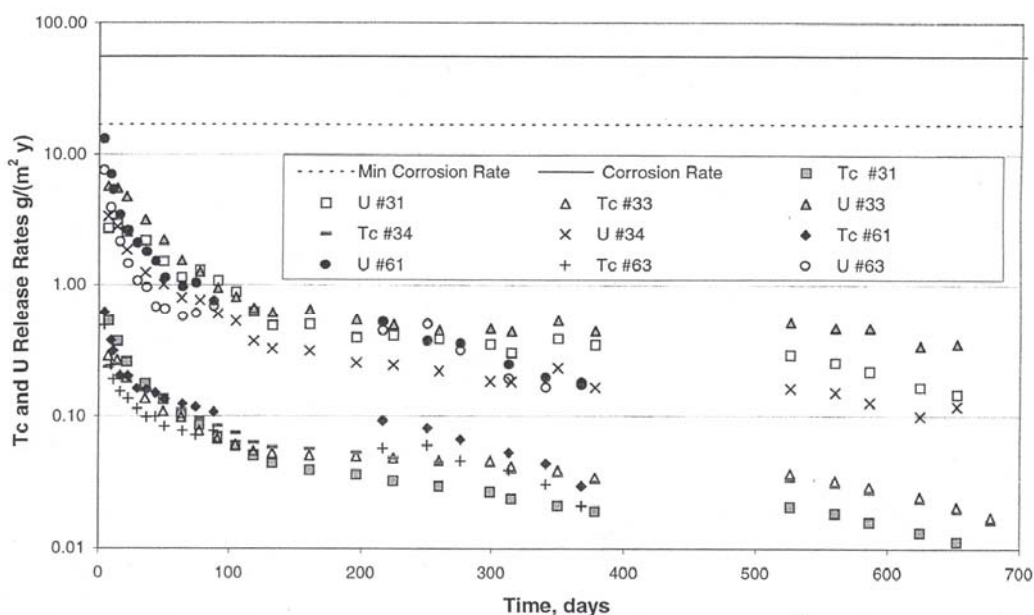


Figure 3. Average release rates of Tc and U in simulated J13 ground water at 90°C, as a function of time, from various SS-15Zr MWF test samples containing Tc or U. Total release rates ($\text{g}/\text{m}^2\text{y}$) were obtained from measured normalized mass loss (g/m^2) over the test duration [12]. The solid line is the calculated corrosion rate from equation 1, and the dashed line is the minimum electrochemical corrosion rate measured in J13 water at 90°C.

The electrochemical uniform corrosion rate, determined as $\mu\text{m yr}^{-1}$, of the metal waste form with no passive oxide layer was measured over a temperature range from 20°C, to 90°C, a pH range from 2 to 12 and chloride concentrations from 4 to 10,000 ppm. The measured corrosion rate for all MWF test specimens under all experimental conditions was typically less than $1 \mu\text{m yr}^{-1}$. By applying the density of the waste form, the corrosion rate can also be expressed as $\text{g m}^{-2} \text{yr}^{-1}$, and for the test conducted this corrosion rate was approximately $10 \text{ g m}^{-2} \text{yr}^{-1}$. The measured radionuclide release of Tc and U from MWF test samples in simulated J13 ground water at 90°C is shown in Figure 3 giving an overall release rate estimate for Tc from the MWF of $0.83 \text{ g m}^{-2} \text{y}^{-1}$. Other surrogate noble metal fission product (Nb, Ru, Rh and Pd) release rates were also determined in concentrated J13 water (100 to 150 times the chloride and bicarbonate content of J13 water) at 90°C. These results of corrosion and release rates of the baseline SS-15Zr alloy show that the waste form is very corrosion resistant and has very low release rates for Tc, U as well as the other base components. These results are very desirable from a repository performance perspective, but make the actual measurement of corrosion and release rates from the waste form challenging.

III. HIGH LOADINGS OF RHENIUM IN THE METAL WASTE FORM

Two experimental goals were desired in the investigation of high fission product loaded MWFs. The first goal was to produce a waste form with, what is believed - based on limited thermodynamic data [13] and previously produced surrogate MWFs, a maximum Tc loading of approximately 20 atom% and without significantly changing the basic phase composition, physical properties, and the corrosion and release rates of the MWF. The second experimental goal was to produce a high-loaded waste form that would clearly demonstrate by physical and chemical examination that the loading limits of the baseline MWF had been exceeded. To this end, surrogate MWF samples were prepared using Re as a surrogate for Tc. Baseline SS-15Zr alloys with 20 and 30 wt% Re loadings (designated SS-15Zr-20Re and SS-15Zr-30Re, respectively), and a Zr-8SS waste form with 20 wt% Re loading (Zr-8SS-20Re) were prepared in a manner similar to the small, developmental-scale waste forms. Cut zirconium wire and Re metal powder were placed in the bottom of a yttria coated crucible and a 304 stainless-steel disk placed on top of the Zr and Re. Ingots were produced by melting at 1600°C for 2 hours while under vacuum in a graphite resistive furnace. The ingots were then sectioned for electron microscopy, x-ray diffraction, destructive chemical analysis and corrosion testing. At the time of manuscript submission only

microcopy had been performed on the SS-15Zr-20Re and -30Re specimens.

III.A. Electron Microscopy of the SS-15Zr-20Re Alloy

Figure 4 shows the phase morphology and elemental composition, as determined by x-ray fluorescence spectroscopy analysis, of SS-15Zr-20Re surrogate MWF. Table III shows the elemental distribution for locations indicated in Figure 4. The elemental composition of the -20Re specimen indicates a phase composition similar to the baseline SS-15Zr waste form consisting of Zr-Fe type intermetallic and Fe solid solution phases. However, it appears that two intermetallic phases are present, intermetallic I and intermetallic II as indicated in Table III. The difference between the two intermetallic regions is a greater Re atomic concentration in the intermetallic I region and lower Re concentrations but higher Ni atomic concentrations in the intermetallic II region. Additionally, two Fe solid solution phase may exist - one rich in Re the other with less Re, with Zr absent in both solid solution phases. These phase compositions are consistent throughout the examined SS-15Zr-20Re specimen. Overall, initial SEM observations indicate the SS-15Zr-20Re specimen is very similar to the baseline SS-15Zr alloy.

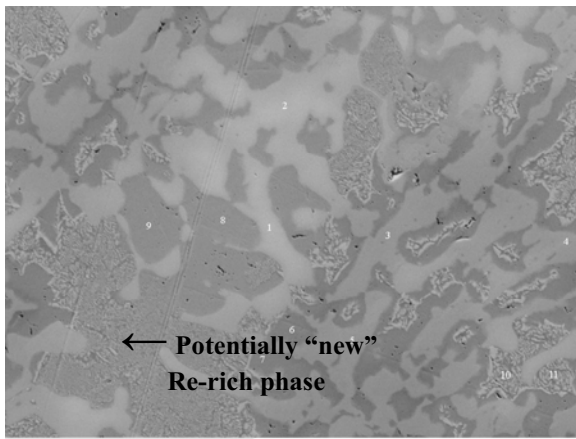


Figure 4. Electron Micrograph of the SS-15Zr-20Re Alloy. The light colored and gray areas correspond to the intermetallic I and II phases. The dark areas is the Fe rich solid solution phase with the potentially new Re-rich phase indicated.

III.B. Electron Microscopy of the SS-15Zr-30Re Alloy

Phase morphology for the SS-15Zr-30Re alloy is shown in Figure 5. The phase elemental composition determined for the SS-15Zr-30Re alloy (Table IV) clearly shows two discreet, high-Re regions separate from the Zr-Fe intermetallic or Fe solid solution phases of the baseline MWF. Also observed in the phase elemental

distribution, and similar to the SS-15Zr-20Re alloy, are the two Fe-Zr intermetallic phases containing high and low concentrations of Re and Ni; and two Fe solid solution regions containing Re and Re-rich compositions, but no Zr.

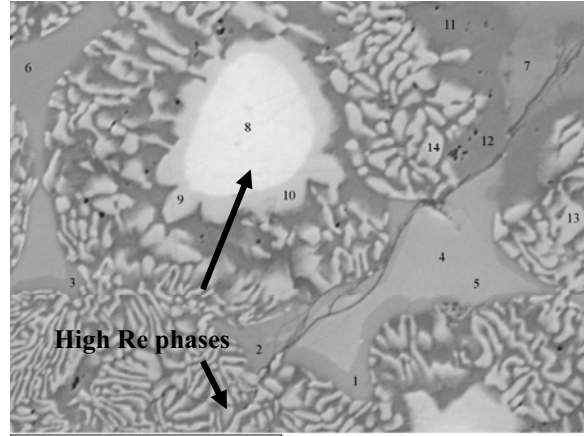


Figure 5. Electron Micrograph of the SS-15Zr-30Re Alloy. The white and light colored areas are the high Re concentration areas not observed in previous baseline alloys containing fission products or surrogates. The gray areas are the intermetallic I and II phases, and the darkest area is the Fe rich solid solution phase.

IV. CONCLUSIONS

Zirconium and stainless steel metallic waste forms have been developed for the consolidation of spent-fuel cladding and the immobilization of technetium and other second- and third-series transition metal fission products in the elemental state. These metallic waste forms have also undergone extensive testing and characterization for acceptance to the Yucca Mountain Geological Repository. From the repository qualification testing and characterization program, it has been determined that the stainless steel-Zr waste forms are extremely durable, corrosion resistant and have very low radionuclide release rates compared to other waste form types engineered for geological disposal.

Recently, the stainless steel-Zr waste forms have been identified as potential waste form candidates for the immobilization of Tc from advanced separation processes in support of GNEP. Current investigations involve determination of the maximum Tc loading possible in these waste forms. Results from initial rhenium surrogate studies indicate that loadings as high as 20 weight percent produce waste forms similar to the baseline alloys. Investigations are continuing to better characterize the surrogate materials and to produce waste forms with comparable loadings of Tc with follow-up studies to determine corrosion and radionuclide release characteristics of high Tc loaded waste forms.

Table III. SS-15Zr-20Re Elemental Composition (Atomic %) of Phase Regions Indicated in Figure 4.

Point	Cr	Fe	Ni	Zr	Mo	Re	
1	5.1	44.6	11.8	27.2	0.7	10.7	Fe-Zr intermetallic I
2	5.6	45.4	10.6	26.9	1.0	10.5	
3	4.4	44.0	21.7	27.0	0.6	2.2	Fe-Zr intermetallic II
4	4.2	44.4	21.3	26.4	0.5	3.2	
5	4.3	43.9	23.1	25.4	1.0	2.4	
6	15.6	70.2	8.7	0	0.9	4.8	Fe solid solution
7	15.5	70.7	8.5	0	0.8	4.7	
8	20.0	66.5	3.9	0	1.4	8.5	Re-rich phase
9	19.4	67.0	4.9	0	1.2	7.8	
10	22.2	63.3	3.8	0	1.8	9.2	
11	17.9	69.8	5.5	0	1.3	5.5	

Table IV. SS-15Zr-30Re Elemental Composition (Atomic %) of Phase Regions Indicated in Figure 5.

Point	Cr	Fe	Ni	Zr	Mo	Re	
1	5.8	48.4	14.0	22.4	0	9.4	Fe-Zr intermetallic I
2	6.1	49.7	15.5	22.5	0	6.3	
3	5.6	48.9	16.1	22.6	0.3	6.6	Fe-Zr intermetallic II
4	4.4	41.4	15.5	27.5	0.8	10.5	
5	4.3	42.3	14.9	26.8	0.8	10.9	
6	4.4	42.6	20.0	26.7	0.5	5.9	Fe solid solution
7	3.8	41.6	23.3	26.2	0.7	4.5	
8	20.3	41.5	1.7	0	2.0	35.1	High Re phase I
9	21.3	50.8	3.8	0	2.4	22.1	
10	21.6	50.4	4.0	0	2.2	19.3	High Re phase II
11	14.1	67.2	10.0	0	0.7	8.3	Re-rich phase
12	13.6	67.7	10.2	0	0.6	8.3	
13	22.8	52.3	3.6	0	2.2	19.3	
14	22.2	52.3	3.2	0	2.2	20.6	

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