Activation Assessment of Liquid Metal Corrosion-Resistant ODS Alloys

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Introduction

The potential for alloying the high chrome ferritic steels with aluminum to improve the liquid metal corrosion resistance is an important aspect for the dual-cooled lithium lead (DCLL) blanket – the preferred blanket concept in the US for future devices. Oxide dispersion strengthened (ODS) alloys are being investigated in the US at ORNL [1-3] for possible PbLi applications at 700-800°C. Also, there is work at Kyoto University [4,5] to develop "Super ODS steels" for Gen-IV fission reactors with improved corrosion resistance via further alloying with other elements, such as Zr and Hf.

The development of such new alloys with enhanced corrosion resistance in static and flowing PbLi is in an early proof-of-principle stage with a number of experimental alloys being studied at ORNL and Japan. This work is being funded both by the fusion and Gen IV fission programs with all studies focusing on the beneficial effects of aluminum additions to ODS alloys with Cr ranging from 11-20 wt%. The primary alloy composition under investigation at ORNL (ODS 125Y), given in Table I, is a good representative of a corrosion-resistant alloy with 4.8 wt% Al. This particular alloy showed promising behavior in static PbLi at 700°C [2,3,6]. Other candidate alloys contain additions of ~0.5 wt% of either Zr or Hf (alloying additions or as oxides), replacing 0.5 wt% of Fe, to enhance the creep strength and improve the stability of the microstructure and oxide precipitates [2,3,5].

It is essential to assess the activation implications of adding Al (and other Zr and Hf additives) to the ODS structure of the DCLL blanket. The main concern is that Al-27 generates a long-lived radioisotope (Al-26; 7.2×10^5 y half-life) through (n,2n) reactions with highly energetic neutrons, particularly in plasma surrounding components. We considered three corrosion-resistant ODS alloys for a DCLL blanket and compared their activation characteristics to that of F82H [7] – the first generation of reduced-activation ferritic/martensitic (RAFM) steel that limits the blanket operating temperature to less than 550°C. As a side point, all corrosion-resistant ODS alloys increment the tritium breeding ratio of the DCLL blanket by ~1%.

The ODS 125Y alloy consists of 11.4 wt% Cr, 4.8 wt% Al, 0.19 wt% Y, 0.05 wt% W, 0.02 wt% Si, 0.01 wt% Ti, 842 wppm O, 380 wppm C, 455 wppm N, 20 wppm S, and Fe making the balance. A comprehensive analysis of the impurity concentrations characteristic is not available for large-scale processing of ODS alloys via mechanical alloying. Therefore, for the purposes of assessing the activation characteristics of the experimental ODS alloys in Table I, we adopt the approach proposed by Klueh et al. [7]. In an effort to reduce the long-term radioactivity, Klueh et al. [7] provided a list of the lowest 17 impurities that have ever been achieved in large-scale melting and fabrication practices of various RAFMs and designated them as "present" impurities. In other words, these are the lowest concentrations that have ever been achieved in large-scale melting and fabrication practices for the RAFMs. They are not specific to any particular RAFM composition and should be achievable at *present* with a relatively modest effort and cost.

Management of Fusion Radioactive Waste

The strategy for handling fusion activated materials calls for three potential schemes: recycling within the nuclear industry, clearance or release to the commercial market if materials contain traces of radioactivity, and disposal in geologic repositories. Plasma facing components (such as FW, blanket, and divertor) normally contain high radioactivity and do not qualify for clearance. Recycling is the preferred option as it helps minimize the radwaste assigned for disposal through the reuse of the continuous stream of activated materials generated during operation and after decommissioning. Since there is design latitude in materials selection, fusion has been able to avoid creating high-level waste (HLW) that requires long-term storage in deep geologic repositories. However, the low-level waste (LLW) amounts are quite large in fusion [8], so efforts to recycle are essential to reclaim resources and support fusion deployment. For these reasons, the geologic disposal option should be avoided to promote fusion as an attractive source of nuclear energy with minimal environmental impact [8-12].

Since the early 1980s, reduced-activation materials have been developed for fusion based on waste disposal criterion and releases during accidents. Since then, many studies evaluated a wide variety of reduced-activation materials and identified areas in which these materials need to be improved to satisfy the pre-determined fusion safety and environmental criteria. This required careful choice of all materials from the outset, excluding specific alloying elements (such as Al, Mo, Ni, Mn, Cu, Re, and Ir) and controlling certain impurities (such as Nb, Mo, Ir, and Ag). Such alloying elements and impurities tend to generate HLW. However, more recent fusion designs [8,13] prefer to recycle all materials instead of burying tons of precious materials in repositories. This means past restrictions imposed on many alloying elements and impurities (like Al, Mo, Nb, Re, Ni, Cu, etc.) could be lifted out or relaxed considerably since the requirements for recycling are quite different from disposal [8-12]. In fact, long-lived radionuclides or impurities (that determine the waste classification) are of less importance to the recycling process while radioisotopes with intermediate half-lives are more important for advanced recycling equipment that handle the highly irradiated fusion components.

Activation Model

The selected design for such an activation analysis is ARIES-ACT-2 [8] – the most recent power plant design in the ARIES series. The design utilizes the DCLL blanket with helium-cooled F82H structure, PbLi breeder and SiC flow channel inserts (FCI). The PbLi allows not only breeding, but also self-cooling of the blanket. The final design has a 9.75 m major radius, 2.44 m minor radius, 2637.5 MW fusion power, and 1 GW net electric power.

The first wall and DCLL blanket layout is shown in Fig. 1. The 3.8 cm thick FW consists of \sim 34% F82H and 66% He coolant, by volume, while the blanket contains 14% F82H, 68% PbLi (with 40% enriched Li), 4% SiC, and 14% He coolant. The 14 MeV source neutrons will activate all components and generate radioactive materials at the end of the FW/blanket service lifetime (\sim 10 years). In the ARIES-ACT-2 assessment, it was assumed that an advanced RAFM steel would be capable of surviving a lifetime neutron dose of \sim 200 dpa. This dpa limit determined the \sim 10 year lifetime of the FW/blanket. It is

now recognized that this is an unrealistic expectation for RAFMs and that the only structural materials with potential neutron doses of that order are the ODS alloys with very high sink strengths for point defects and for the trapping of high concentrations of helium.



Fig. 1. Midplane cross section of outboard DCLL blanket of ARIES-ACT-2 showing the 40 cm thick inner blanket segment (that includes the 3.8 cm thick FW), 60 cm thick outer blanket segment, PbLi flow channels (in brown), cooling channels (in grey), and SiC FCI (in orange).

Table 1. Composition of Corrosion-Resistant Alloys (in weight %). F82H Composition Included for Comparison. Asterisk Indicates Weight Part per Million (wppm)

Alloy	F82H	F82H	ODS 125Y	ODS 125Y	ODS 125Y
	with Nominal	with Present	Alloy (Al)	Alloy (Al+Zr)	Alloy (Al+Hf)
	Impurities	Impurities	with Present	with Present	with Present
Density	7 89	7 89	7 799	7 799	7 799
(g/cm ³)	7.89	7.89	1.199	1.199	1.199
С	0.1	0.1	*380	*380	*380
Ν			*455	*455	*455
0			*842	*842	*842
Al	1.40E-03	*30	4.8	4.8	4.8
Si			0.02	0.02	0.02
S			*20	*20	*20
Ti			0.01	0.01	0.01
V	0.2	0.2			
Cr	7.5	7.5	11.4	11.4	11.4
Fe	90.11586	90.173301	83.356601	82.856601	82.856601
Со	*28	*8	*8	*8	*8
Ni	*474	*13	*13	*13	*13
Cu	*100	*10	*10	*10	*10
Y			0.19	0.19	0.19
Zr				0.5	
Nb	*3.3	*0.5	*0.5	*0.5	*0.5
Мо	*21.0	*5	*5	*5	*5
Pd	*0.05	*0.05	*0.05	*0.05	*0.05
Ag	*0.1	*0.05	*0.05	*0.05	*0.05
Cd	*0.4	*0.05	*0.05	*0.05	*0.05
Eu	*0.05	*0.02	*0.02	*0.02	*0.02
Tb	*0.02	*0.02	*0.02	*0.02	*0.02
Dy	*0.05	*0.05	*0.05	*0.05	*0.05
Но	*0.05	*0.05	*0.05	*0.05	*0.05
Er	*0.05	*0.05	*0.05	*0.05	*0.05
Hf					0.5
Та	0.02	0.02			
W	2	2	0.05	0.05	0.05
Os	*0.05	*0.05	*0.05	*0.05	*0.05
Ir	*0.05	*0.05	*0.05	*0.05	*0.05
Bi	*0.02	*0.05	*0.05	*0.05	*0.05
U	*0.05				

Activation calculations for this study made use of the PARTISN one-dimensional transport code [14] and ALARA activation code [15] with FENDL cross-section libraries [16,17]. We focused our attention on the outboard (OB) FW and blanket that were modeled in toroidal, cylindrical geometry with an average OB neutron wall loading of 1.5 MW/m². All external components were included in the model to provide the appropriate neutron reflection. The irradiation history took into account the 85% availability during the projected service lifetime of the FW/blanket (~10 years). In the transport and activation analyses, the alloying elements and complete set of impurities were included in the material definition as given in Table I.

Activation Results

The recycling Option: The technical feasibility of recycling is based on the dose rate to the remote handling (RH) equipment. Essentially, the dose determines the RH needs (hands-on, conventional, or advanced tools to handle the radioactive components) and the interim storage period necessary to meet the dose limit. Advanced RH equipment has been used in the nuclear industry, in hot cells and reprocessing plants, and in spent fuel facilities. While the fission processes may have no direct relevance to fusion, their success gives confidence that such advanced RH techniques could be further developed to handle higher doses (> 10,000 Sv/h) along with the necessary adaptation to the fusion needs (component size, weight, etc.). Beside the recycling dose, other important criteria include the decay heat level during reprocessing, recycling of tritium-containing materials, physical properties of recycled products, and economics of fabricating complex shapes remotely [12].

Figure 2 presents the recycling dose rate for the OB FW – the most radioactive component in the ARIES-ACT-2 design. As the figure indicates, all three corrosion-resistant alloys could potentially be recycled within one year of storage using advanced RH equipment. The main contributor to the dose at one year after shutdown is Mn-54 (97%) which is produced by iron - the main constituent of the ODS alloy. The less radioactive components outside the FW that contain PbLi and employ the corrosion-resistant ODS alloy (such as the blanket and PbLi manifolds) could be recycled with conventional equipment if longer storage period (10-100 y) is permitted. None of the invessel components could be recycled with hands-on operations.

The Disposal Option: In the US, the waste disposal rating (WDR) represents a metric for waste classification. It is the ratio of the specific activity (in Ci/m^3 at 100 y after component replacement) to the allowable limit summed over all radioisotopes. A WDR < 1 means LLW and a WDR > 1 means Greater than Class C waste (GTCC) or HLW.

The WDR was evaluated for fully compacted components using the waste disposal limits developed by Nuclear Regulatory Commission (NRC) 10CFR61 [18] and Fetter [19]. The NRC waste classification is largely based on radionuclides that are produced in fission reactors, hospitals, research laboratories, and food irradiation facilities. The NRC specific activity limits were developed for eight radionuclides only in addition to actinides. In the early 1990s, Fetter and others [19] expanded the NRC list and performed analyses to determine the Class C specific activity limits for all long-lived radionuclides

of interest to fusion using a methodology similar to that of 10CFR61 [18]. Although Fetter's calculations carry no regulatory acceptance, they are useful because they include fusion-specific radioisotopes, as illustrated in Fig. 3.



Figure 2. Time variation of recycling dose rate to the RH equipment for the OB FW of ARIES-ACT-2 design using corrosion-resistant ODS alloys. The F82H alloy is included for comparison.

Figure 4 displays the WDR of the FW and inner blanket segment. Since the FW is an integral part of the blanket, both components will be disposed of as a single, fully compacted unit.

According to the NRC 10CFR61 [18], the OB FW/blanket qualifies as Class C LLW under ARIES-ACT-2 operating conditions for any alloy with "Present" impurities. The only restriction is that the Nb impurity should be strictly controlled to a very low level (< 0.5 wppm). The main contributors to the WDR of ODS alloys are C-14 (~80%) and Nb-94 (~20%). As noticed, the well-developed F82H RAFM steel (with 3.3 wppm Nb and 21 wppm Mo) still needs impurity control as it generates GTCC waste.

According to Fetter [19], the OB FW/blanket qualifies as GTCC waste for all corrosionresistant ODS alloys with "Present" impurities. Nb and Mo should be strictly controlled below 0.5 wppm and 5 wppm, respectively. The main contributors to the WDR of the corrosion-resistant ODS alloys are Al-26 (~85%), Nb-94 (~8%), and C-14 (~4%). Adding 0.5 wt% Zr or Hf doesn't impact the WDR significantly.



Figure 3. List of nuclides that NRC (left) and Fetter's (right) used to determine the specific activity limits for low-level waste.



Figure 4. Waste disposal rating of fully compacted FW/blanket after ~ 13 MWy/m² of irradiation. Red bars for WDR using Fetter's limits. Green bars for WDR using NRC limits.

As mentioned earlier, the adopted atomic displacement limit for the RAFM structure is 200 dpa for future ARIES power plants that could be built in 2050 or beyond. This relatively high dpa limit allowed the FW/blanket to operate for ~10 years before requiring replacement due to radiation damage considerations. If we consider a more conservative lifetime dose (on the order of ~100 dpa or less), the FW/blanket lifetime will be shortened accordingly. Since the WDR is fluence-dependent, there will be a notable impact on the WDR and waste classification shown in Fig. 4. For instance, if the service lifetime of the FW/blanket that employs the ODS 125Y alloy is shortened from ~10 y to 2 y, the WDR will drop below one, meeting the Class C waste classification using Fetter's limits, as illustrated in Fig. 5. The corresponding dose would be 40 dpa at the end of 2-y operation. Note that the impact of the irradiation time on the flux-dependent recycling dose of Fig. 2 is insignificant.



Figure 5. Reduction of WDR with OB FW/blanket service lifetime for ODS 125Y structure.

Conclusions

An important aspect to the high Cr ODS steel is the potential for alloying with Al to improve the liquid metal corrosion resistance. At present, ODS FeCrAl alloys are being investigated at ORNL in the US and at Kyoto University in Japan for possible PbLi applications at 700-800°C. Some alloys also have additions of ~0.5 wt% of either Zr or Hf that have been shown to further improve the stability of alloys.

Our activation results showed that the three candidate corrosion-resistant ODS alloys could potentially be recycled with advanced remote handling equipment shortly after replacing the DCLL blanket of the ARIES-ACT-2 design.

According to the NRC, a FW/blanket employing the ODS FeCrAl alloy could qualify as low-level waste at the end of 10 y operation (with a corresponding dpa dose of 200 dpa) if the Nb impurity is kept below 0.5 wppm. According to Fetter, the FW/blanket could qualify as GTCC waste if the Nb and Mo impurities are controlled below 0.5 wppm and 5 wppm, respectively. Adding 0.5 wt% Zr (or Hf) to the ODS FeCrAl alloy has insignificant impact on the waste disposal rating and will not alter the waste classification of the FW/blanket. If it is essential to qualify the FW/blanket as Class C LLW based on Fetter's limits, the service lifetime of the blankets should be limited to 2 years with an attainable dpa dose of 40 dpa.

What has once been a fairly routine geologic disposal process for fusion radwaste in the 1980s and 1990s is now becoming unattractive option because of the large amount of radioactive materials that fusion generates compared to other nuclear sources. Recent fusion designs strongly support the recycling process and avoid the disposal option in order to reclaim valuable resources and promote fusion as an attractive source of nuclear energy with minimal environmental impact. Adopting this strategy, research related to the further development of the three candidate corrosion-resistant ODS alloys should continue to improve the stability of these alloys in flowing PbLi, support operating the DCLL blanket at higher temperature, and ultimately enhance the thermal conversion efficiency of the overall design.

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