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Journal of Nuclear Materials 329–333 (2004) 1643–1647
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Transmutation and activation of reduced activation ferritic martensitic steel in molten salt cooled fusion power plants

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Abstract

Neutron activation analysis was conducted for the reduced activation ferritic/martensitic (RAFM) steel used in flibe molten-salt cooled fusion blankets. After 22.4 MW yr/m^2 of neutron exposure, the RAFM steel first wall in a molten salt blanket with 40% lithium-6 enrichment in lithium was found to be within 1 mSv/h in contact dose rate after 100 yr of cooling. The contact dose rate drops to 30 and 20 μSv/h or less, respectively, when the cooling times are 300 and 500 yr after discharge. The RAFM steel discharged from the high-temperature shield component would be allowed for hands-on recycling after 100 yr of cooling, when the contact dose rate is 10 μSv/h or less. The most significant changes found in the RAFM steel first wall due to nuclear transmutation, are 10% decrease in W and 10% increase in Ti. Additionally, there are minor elements produced: Mn – <1.2%, V – <0.26%, Re – <0.2%, Ta – <0.08%, and Os – <0.1%, all in weight percent. The gaseous elements generated are H and He, and the, respectively, accumulated quantities are about 260 and 190 wppm.

1. Introduction

Reduced activation ferritic martensitic (RAFM) steel is a candidate structural material for the first generation fusion power plants [1]. Characteristics of transmutation and activation of ferritic steels due to neutrons in the fusion power reactor environment are needed to facilitate the development of these alloys. Transmutation of main alloying elements in the RAFM steel may change the structural properties of the alloy and thus may cause to limit the component lifetime in the power reactor. Activation of fusion materials is an important consideration in selecting reactor materials for future fusion power plants, particularly when long-term environmental issues associated with management and disposal of discharged components are concerned. Contact dose rate from materials in a discharged power plant component is one of useful factors as to whether deep geologic disposal, materials recycling or clearance is to be selected for handling the discharged materials.

Neutronics studies were performed recently for several molten salt fusion blankets made of RAFM steel structure [2]. Flibe molten salt, which is a mixture of LiF and BeF2, has been found to be attractive as a coolant for fusion power plants. This paper discusses results of a study pertaining to effects of neutrons on the RAFM steel in the flibe cooled power plants.

2. Methods of study

The tokamak power plant, ARIES-RS [3], was used as a reference for the study. In this study, a toroidal geometry reactor model was used in all neutronic calculations. The power extraction components in the outboard of the power plant include a 3 mm first wall made of RAFM steel, a 0.2 m thick first tritium breeding zone composed of 60% molten salt, 30% beryllium (95% dense), and 10% RAFM, if beryllium is employed as a neutron multiplier, otherwise the compositions of this breeding zone would be 90% molten salt and 10% RAFM steel. Behind the first breeding zone, there is a 0.3 m second breeding zone made of 90% molten salt and 10% RAFM steel. It is then followed by a 70 mm

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doi:10.1016/j.jnucmat.2004.04.148

replaceable shield made of 10% molten salt and 90% RAFM steel, and a 0.26 m high-temperature shield zone made of 5% molten salt, 4% W, 76% SS304, and 15% RAFM steel. Note that all percentages used for describing the compositions are all by volume.

The power extraction components in the inboard include a 3 mm first wall made of RAFM steel, a 0.2 m thick tritium breeding zone composed of 60% molten salt, 30% beryllium (95% dense), and 10% RAFM steel, if beryllium is employed as a neutron multiplier, otherwise the compositions in this breeding zone would be 90% molten salt and 10% RAFM steel. Behind the breeding zone, there is a 0.2 m replaceable shield made of 10% molten salt and 90% RAFM steel. It is then also followed by a 0.26 m high-temperature shield zone made of 5% molten salt, 4% W, 76% SS304, and 15% RAFM steel.

Neutronic calculations performed for these flibe/RAFM blankets considered various cases of lithium-6 enrichment in lithium. The lithium-6 enrichment factors include natural (7.42% lithium-6), 20%, and 40%.

Neutron fluxes were calculated with the one-dimensional particle transport code ANISN [4] and FENDL nuclear data libraries [5]. Activation calculations were performed with the radioactivity calculation code REAC3 [6], and FENDL/A-2 activation data library [7].

Neutron activation analysis was performed taking into account important impurity elements because they are the principal precursors of long-lived radioactive nuclides induced in the RAFM steel. Concentrations of these impurities were taken from Kleuh et al. [8] assuming ultra purification process is possible in the future. The neutron wall loading for the ARIES-RS was also used for the molten salt/RAFM power plant. It is 5.6 MW/m^2 peaked at the outboard first wall location. The respective lifetimes for the various power extracting components are similar to those originally assumed for the ARIES-RS components, except for the first wall and the first breeding zone, which are assumed to be four full-power years (FPYs). The lifetimes for the other components include 7.5 FPYs for the second blanket and the replaceable shield components, and 40 FPYs for the high-temperature shield.

3. Calculational results and discussions

Tritium breeding characteristics for the flibe/RAFM blankets modeled above have been reported in Ref. [2] for the cases with and without the beryllium neutron multiplier, and for 7.4%, 20%, and 40% lithium-6 enrichment in lithium. Studies concluded that a high lithium-6 enrichment, such as 40% lithium-6 in lithium, would not only maintain an attractive tritium breeding ratio, but could also achieve the maximum advantage in reducing the dose release during a unexpected power plant accident which results in the rupture of the vacuum vessel and release of mobilized molten salt into atmosphere [2].

In the following the results of transmutation and activation calculations for the RAFM steel structure in the flibe cooled power extracting components are reported.

3.1. Activation

The principal task in this study is to examine the gamma contact dose rate from RAFM steel structure in power plant components after reaching the anticipated lifetime. First wall and the high-temperature shield components represent the two most important components that cover the entire range of the contact dose rate that would occur to the RAFM steel structure in the power plant. Contact dose rate profiles from the RAFM steel first wall after four full-power years of operation in the power plant, without and with a beryllium neutron multiplier, respectively, were obtained for comparison. Several lithium-6 enrichment factors in lithium, namely 7.4% (natural lithium), 20%, and 40% lithium-6 in lithium were considered, and their respective contact dose rate profiles compared as well.

The results of the comparison revealed that lower induced activity and thus lower attendant contact dose rate in the structural alloys are generally obtainable when higher value of lithium-6 enrichment factor is employed. Because of this and the fact that attractive tritium breeding ratios are attainable at this enrichment factor, as concluded in Ref. [2], the cases with 40% lithium-6 in lithium were selected for more detailed analyses.

Contact dose rate profiles from the main alloying elements of the RAFM steel and several relatively more important deleterious impurities include Ag, Al, Co, Eu, Ho, Mo, and Nb, were summarized and analyzed. Tables 1 and 2 then show the sums of the contact dose rates from main alloying elements and the deleterious impurities with ultimate concentrations as outlined in Ref. [8].

Discussions concerning the significance of the results are given below.

The waste disposal relevant radionuclides induced in power plant components due to fusion neutrons are generally divided into two categories. One is medium-lived. The other is long-lived. Medium-lived radionuclides are mainly those whose half-lives are 30 yr or shorter, and the activity pertaining to these radionuclides could generally decay away in 500 yr. (At least 4 orders of magnitude reduction in activity is expected in a cooling time of more than 16 half-lives.) The radionuclides with half-lives longer than 30 yr are generally considered long-lived, and activities due to these radionuclides would not decay away in 500 yr.

Table 1
Contact dose rates (μSv/h) from reduced activation ferritic martensitic (RAFM) steels including the best educated guessed ultimate impurities (Ref. [8]): first wall in a flibe cooled power plant; four full-power years of operation with 40% Li-6 in lithium (numbers in parentheses are percentages contributing to the respective sums)

Element (impurity level wppm)	No beryllium multiplier			With beryllium multiplier		
	Cooling time (yr)			Cooling time (yr)		
	100	300	500	100	300	500
RAFM: main alloying elements (Ref. [9])	530 (94%)	9.2 (49%)	3.4 (31%)	510 (94%)	15 (52%)	10 (49%)
Ag (<0.005)	<3.6	<2.6 (14%)	<1.9 (17%)	<2.9	<2.1	<1.5 (7%)
Al (0.5)	0.11	0.11	0.11	0.1	0.1	0.1
Co (<0.02)	<0.012	2.2×10^{-6}	2.2×10^{-6}	<0.15	2.7×10^{-5}	2.7×10^{-5}
Eu (0.01)	25	0.19	0.0022	17	0.08	0.0014
Ho (0.01)	3.4 (18%)	3.4 (25%)	2.7	6.8	6.0 (21%)	5.4 (25%)
Mo (1)	1	0.92	0.82	0.96	0.86	0.77 (3.5%)
Nb (<0.02)	<2.3	<2.2 (12%)	<2.2 (20%)	<4.6	<4.6 (16%)	<4.5 (20%)
Sum	<570	<19	<11	<540	<29	<22

Table 2
Contact dose rates (μSv/h) from reduced activation ferritic martensitic (RAFM) steels including the best educated guessed ultimate impurities (Ref. [8]): high-temperature shield in a flibe cooled power plant; 40 full-power years of operation with 40% Li-6 in lithium (numbers in parentheses are percentages contributing to the respective sums)

Element (impurity level wppm)	No beryllium multiplier			With beryllium multiplier		
	Cooling time (yr)			Cooling time (yr)		
	100	300	500	100	300	500
RAFM: main alloying elements (Ref. [9])	0.9 (20%)	0.021 (1.6%)	0.011 (0.9%)	0.69 (7.6%)	0.028 (0.8%)	0.025 (0.8%)
Ag (<0.005)	<0.073 (1.6%)	<0.053 (4%)	<0.038 (3%)	<0.14	<0.098	<0.07
Al (0.5)	0.0002	0.0002	0.0002	0.00006	0.00006	0.00006
Co (<0.02)	<0.001	1.9×10^{-9}	1.9×10^{-9}	<0.0036	1.5×10^{-9}	1.5×10^{-9}
Eu (0.01)	2.2 (49%)	0.0013	2.2×10^{-5}	4.7 (52%)	0.0004	4.6×10^{-6}
Ho (0.01)	0.9 (20%)	0.8 (61%)	0.71 (59%)	2.4 (26%)	2.1 (62%)	1.9 (61%)
Mo (1)	0.0065	0.0057	0.0052	0.0035	0.0033	0.0031
Nb (<0.02)	<0.43 (10%)	<0.43 (33%)	<0.43 (36%)	<1.2 (13%)	<1.2 (35%)	<1.1 (35%)
Sum	<4.5	<1.3	<1.2	<9.1	<3.4	<3.1

Activation characteristics of the induced radionuclides have two aspects. One of them is half-life, as described above. The other is the magnitude of induced activity, and the attendant gamma dose rate. A comparison of contact dose rate is also to be made against dose rate levels meaningful for hands-on handling ($\leqslant 10$ μSv/h) and remote handling ($\leqslant 1$ mSv/h) to conduct materials recycling. Specific contact dose rate characteristic are discussed below.

(a) *Main alloying elements of the RAFM steel*. 12 w/o Cr, 3 w/o W, 0.4 w/o Ti, 0.25 w/o Y_2O_3, and balance of Fe [9]. Medium-lived radionuclides are mainly Co60 (half-life 5.27 yr) which is generated from Fe58(n,γ)Fe59 → Co59(n,γ) reactions, and Hf178n (31 yr) which is primarily due to the W182(n,n′α) reaction. Note that W is a substitute for Mo in the RAFM steel. The long-lived radionuclides are Re186m (2×10^5 yr) which is due to W186(n,γ)W187 → Re187(n,2n) and W184(n,γ)W185 → Re185(n,γ) reactions, and Mn53 (3.7×10^6 yr) from Fe54(n,2n)Fe53 → Mn53 reaction. It takes about 500 yr for medium-lived radionuclides to diminish until the long-lived radionuclides to become the dominating activity level. The first wall activity could be about 10 μSv/h after 500 yr of cooling. For the high-temperature shield component, however, it would take only 100 yr or less for the contact dose rate to drop below the 10 μSv/h level. The power plant with a beryllium neutron multiplier shows an advantage in shortening the needed cooling times to allow the activity to decay below 10 μSv/h dose rate level. These cooling times are 300 and 70 yr, respectively, for the first wall and the high-temperature shield components compared

to 500 and 100 yr for the power plant without a beryllium neutron multiplier, as previously mentioned.

(b) *Ag*. There is no medium-lived radionuclide observed. The long-lived radionuclide is Ag108m (481 yr), which is generated from Ag107(n,γ) and Ag109(n,2n) reactions. The first wall contact dose rate due to 1 wppm of Ag is about 1 mSv/h, while the high-temperature shield contact dose rate is a few tens μSv/h for several hundred years after discharge. The power plant with a beryllium neutron multiplier shows a slightly lower dose rate for both components.

(c) *Al*. There is no medium-lived radionuclide observed. The long-lived radionuclide is Al26, which is generated from the Al27(n,2n) reaction. The contact dose rates due to 1 wppm Al in the first wall component is about 0.2 μSv/h for both power plants. Since the induced reaction for Al26 is a threshold reaction, the 14 MeV neutron flux is greatly attenuated in the high-temperature shield component, and hence the Al26 activity is also drastically diminished. The contact dose rate from 1 wppm of Al in this component is about 0.5 pSv/h or lower, a level much lower than the natural radiation background.

(d) *Co*. The medium-lived radionuclide is Co60 (5.27 yr), generated via the Co59(n,γ) reaction. The long-lived, but very low activity, radionuclide is Fe60 (1.5×10^6 yr), generated due to the (n,p) reaction with the radioactive Co60. The dominance of Co60 is about 100 yr for the first wall component and 50 yr for the high-temperature shield component before the contact dose rates due to 1 wppm of Co drop below 10 μSv/h. The power plant without the beryllium neutron multiplier has a slight advantage in having a lower Co60 activity due to the reduced neutron population in the blanket components. The dose rate due to Fe60 (main gamma rays are emitted from its decay daughter radionuclide Co60) at 300 yr after discharge and thereafter is well below the natural background level.

(e) *Eu*. There is no long-lived radionuclide observed, but are quite a few medium-lived radionuclides generated. These include Eu152 (13.5 y), via the Eu151(n,γ) reaction, Eu154 (8.59 yr), via the Eu153(n,γ) reaction, and Eu150 (36.9 yr), via the Eu151(n,2n) reaction. The 1 wppm of Eu shows a contact dose rate of 10 μSv/h in the first wall component at about 300 yr after discharge for both power plants. In the high-temperature shield, the dose rate could drop below 10 μSv/h at 100 yr or less after discharge.

(f) *Ho*. The only significant radionuclide generated is the long-lived Ho166m (1200 yr), which is generated via the Ho165(n,γ) reaction. The contact dose rate levels due to 1 wppm of Ho are about 0.4 and 0.1 mSv/h, respectively, at the first wall and high-temperature shield components of the power plant with no beryllium neutron multiplier. The contact dose rates due to Ho in the power plant with beryllium neutron multiplier are about a factor of 2 higher than that without a neutron multiplier, and it is primarily because of the increased neutron population in the blanket components.

(g) *Mo*. There is one, but not significant, medium-lived radionuclide generated in Mo: namely Nb93m (16.1 yr), induced due to the Mo93(n,p) reaction. The long-lived radionuclides include Nb91 (680 yr),due to the Mo92(n,2n)Mo91Nb91 reactions, Mo93 (4000 yr), due to Mo94(n,2n) and Mo92(n,γ) reactions, and Nb94 (20 000 yr), due to Mo94(n,p), and Mo95(n,n′p) reactions. The contact dose rate levels due to 1 wppm Mo are 1 μSv/h and 10 pSv/h, respectively, at the first wall and high-temperature shield components. The power plant with a beryllium neutron multiplier has a slight advantage in generating a lower long-lived radioactivity in the high-temperature shield component.

(h) *Nb*. There is no medium-lived radionuclide observed. The only long-lived radionuclide generated is Nb94 (20 000 yr), and it is via the Nb93(n,γ) reaction. The contact dose rate levels due to 1 wppm of Nb are 0.1 mSv and 20 μSv/h, respectively, in the first wall and high-temperature shield components of the power plant without the beryllium neutron multiplier. The contact dose rates in the power plant with the beryllium neutron multiplier are higher by a factor of 2, and it is primarily due to the higher neutron population in the blanket components.

The anticipated ultimate level of deleterious impurities, as presented in Ref. [8], are Ag – <0.005, Al – 0.5, Co – <0.02, Eu – 0.01, Ho – 0.01, Mo – 1, and Nb – <0.02, all in units of wppm. Using the contact dose rates from the impurities and the main alloying elements, a contact dose rate from the RAFM structure can then be readily derived. Tables 1 and 2 report such integrated dose rate results. As shown in Table 1, the first wall contact dose rates from the RAFM steel structure including impurities are about 20 and 30 μSv/h at 300 yr after discharge, respectively, for the power plant without and with a beryllium neutron multiplier. At 500 yr after discharge, these dose rates are reduced to 10 and 20 μSv/h, respectively. Out of the overall dose rates, impurities contributed about 50–70%. Important elements are Ag, Ho, and Nb.

On the other hand, the contact dose rates from the high-temperature shield component in these two power plants are 10 μSv/h or less at 100 yr after discharge, as shown in Table 2. This would allow for hands-on recycling,. Note that more than 80% of the contact dose rates are due to impurities when the cooling time is more than 100 yr. These contributing radionuclides are mainly Eu, Ho, and Nb. However, when the cooling time is 300 yr and longer, then the contact dose rate is almost completely due to the impurities. The dominating elements at cooling times longer than 300 yr are Ho and Nb.

3.2. Transmutation

Transmutation of alloying elements in the structural material is one of the important factors to determine the service lifetime of such a structural material in a neutron field. The most important location of the structural material is perhaps the first wall facing the plasma because of the highest neutron flux level.

The major composition changes in the first wall RAFM steel after lifetime irradiation were calculated to be 8–39% decrease in W and 10% increase in Ti, for the power plants without and with a beryllium neutron multiplier. The large range of reduction in W is because transmutation of W is mainly due to neutron absorption in W and which is highly affected by the low energy neutron flux whose population depends on the lithium-6 content. Additionally, there are minor elements produced in the alloy after lifetime irradiation. These include Mn: 1.2%, V: 0.26%, Re: 0.08–0.2%, Ta: 0.012–0.13%, and Os: 0.2–1%, all in weight percent. Note that among the above minor elements produced due to transmutation, Re and Ta are primarily due to the transmutation of W, and are subject to the effect of low energy neutron spectrum. The gaseous elements generated are hydrogen and helium, and their lifetime accumulated levels are about 260 and 190 wppm, respectively, for hydrogen and helium.

4. Conclusions

One of the mechanisms to lower the Induced long-term radioactivity in fusion power plants is by enriching lithium-6 in lithium to suppress the low energy neutron fluxes. The other is using the reduced activation reactor materials, as generally accepted and is a basis for developing the reduced activation fusion material. Waiting for a longer cooling time than normally considered, such as waiting from 50 to 100 yr or longer after discharge, before taking action to handle the discharged reactor components is also found to be effective in reducing the contact dose rate from the discharged components, because it allows the medium-lived radionuclides generated in the reactor materials to decay away.

The RAFM steel from the first wall component of flibe cooled power plants with 40% lithium-6 enrichment in lithium was found to be within 1 mSv/h in contact dose rate after 100 yr of cooling. The contact dose rate could be further reduced by longer cooling. It drops to 30 μSv/h or less when the cooling time is 300 yr after discharge.

The RAFM steel discharged from the high-temperature shield component would be allowed for hands-on recycling after 100 yr of cooling. The contact dose rate at that time after discharge and thereafter is 10 μSv/h or less.

The most significant changes found in the RAFM steel as the first wall in the flibe cooled power plants with 40% lithium-6 enrichment in lithium, are 10–40% decrease in W and 10% increase in Ti, after 22.4 MW yr/m^2 of neutron exposure (four full-power years at 5.6 MW/m^2 neutron wall loading). Additionally, there are minor elements produced in the alloy after irradiated with the same amount of neutron fluence. These elements and the corresponding amounts are Mn – <1.2%, V – <0.26%, Re – <0.2%, Ta – <0.08%, and Os – <0.1%, all in weight percent. The gaseous elements generated are H and He, and the, respectively, accumulated quantities are about 260 and 190 wppm.

Acknowledgements

This research was supported by the US Department of Energy, Office of Fusion Energy Sciences, under Research Grant No.: DE-FG03-92ER54137.

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