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Journal of Nuclear Materials 307–311 (2002) 1047–1051

journal of nuclear materials

www.elsevier.com/locate/jnucmat

The zero waste option: clearance of activated and first wall/blanket materials

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Abstract

Management of activated waste from fusion power reactors is part of the long-term action of the European Fusion Programme. The options of *recycling*, the reuse in novel reactors for in-vessel materials, and *clearance*, the declassification to non-active waste, for ex-vessel materials, were analysed with the aim to reduce the amount of radioactive waste. A further step is the attempt to reduce the long-term radioactivity of selected materials to levels allowing clearance also of in-vessel structures. Clearance can be an alternative option also for in-vessel materials. Vanadium-based alloys belong at the moment to the materials with sufficiently low-activation constituting elements. Attainment of clearance conditions in activated in-vessel V–4Cr–4Ti structures would require very low concentrations of impurities, not achievable with the current purification methods and in some cases below the present detection limits, and the development of methods to reprocess the activated alloy for extracting radiotoxic nuclides.

1. Recycling and clearance

Management of activated waste from fusion power reactors is part of the long-term action of the European Fusion Programme. The options of *recycling*, the reuse in novel reactors for in-vessel materials, and *clearance*, the declassification to non-active waste, for ex-vessel materials, were analysed with the aim to reduce the amount of radioactive waste. A further step, explored here, is the attempt to reduce the long-term radioactivity of selected materials to levels allowing clearance also of in-vessel structures. The radioactive conditions for recycling within the nuclear industry and clearance are given in Refs. [1–3].

1.1. Recycling

Recycling depends on contact dose rates after a 50–100 years decay. During this time the material would have to be kept in a secure interim store. Contact dose rates D_c of less than 2 and 20 mSv/h define respectively the SRM and CRM levels, i.e., simple- and complex-recycle material, according to the degree of radioactive protection required. $D_c > 20$ mSv/h defines PRM, permanent disposal waste which is unfit for recycling.

1.2. Clearance

Clearance is assessed in compliance with limits of the specific activities of radiotoxic nuclides, which are weighted according to their hazard [3]. Materials can be cleared either by disposal as non-active waste (NAW), i.e., clearance (D) or by recycling outside the nuclear industry, i.e., clearance (R). Feasibility of clearance (D) is assessed from the clearance levels L_c of radionuclides contained in the material. L_c is the specific activity of the radionuclide that would allow the declassification to non-active waste of the material containing this radionuclide as only contaminant. Clearance (D) levels are taken from an IAEA proposal [4] where they are derived from categorisation of safety analyses of waste disposal sites producing a maximum individual dose of 10 μSv/a.

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0022-3115/02/$ - see front matter
PII: S0022-3115(02)01280-1

These clearance levels vary from 300 Bq/kg (^{60}Co, ^{94}Nb) to 3×10^6 Bq/kg (T). Contributions of each radionuclide present in the material are weighted in a clearance index I_c(D). If A_i and L_i are the specific activity and clearance level of the ith nuclide, it is

$$I_c(\mathrm{D}) = \sum_{i=1}^{z} \frac{A_i}{L_i}. \quad (1)$$

The material can be cleared to NAW if: $I_c < 1$. Concerning clearance (R), recent EC recommendations [5] propose specific activity levels for recycling radioactive materials outside the nuclear industry (unconditional recycling). Clearance (D) levels are generally less (up to more than a factor of 10) than those for clearance (R).

1.3. Sorting of fusion waste

Assessments on power reactor designs having FW/blanket systems in V–4Cr–4Ti/Li_2O or reduced activation ferritic steel/Pb–17Li subjected to a 50 years decay have shown the following [2]: (a) clearance (D) conditions are achieved in ex-vessel structures, typically 30–40% of the total waste amount, (b) the adoption of clearance (R) instead of clearance (D) would not increase significantly the amount of declassified material, (c) radioactive conditions for recycling are attained in practically all other waste.

2. Clearance potential for blanket materials and elements

Recycling as proposed in Section 1.1 is 'conditional', linked to the reuse of material in the nuclear industry.

An interesting alternative is to attempt to reduce the long-term radioactivity of in-vessel materials so to reach clearance conditions. The clearance option for blanket materials has been explored, determining a 'clearance potential' for each chemical element. The procedure has involved a complete survey of the activation properties of each of the elements of the blanket materials as follows: the irradiation in the SEAFP Plant Model 2 blanket of an ideal component was simulated, assuming it was made of a single element (say, for instance, 100% Fe) and the clearance index after 100 years of cooling was determined, assuming clearance with disposal of the material. The simulation was carried out by means of the EASY-2001 package [6]. From the clearance index, the maximum allowable concentration of each element in a blanket structural material was determined, in order to have a clearance index equal to 1. The highest this allowable concentration is, the highest is the clearance potential of that element.

Results are reported in Fig. 1. The following conclusions can be made:

- The maximum allowable concentration for Fe (less than 1%) shows that no clearance could be obtained for any iron-based blanket material (all types of steels). The conclusion is the same for SiC (max. Si concentration less than 2%).
- Many elements have low-activation properties allowing clearance: among the relevant ones are H, Li, V, Cr and Mn.
- V and Cr, in particular, have a 100% maximum allowable concentration. This indicates that V-alloys blanket materials could be a good candidate for clearance. However Ti has a concentration limit of less than 1%. This could be a critical value.
- There are many elements which have very low allowable concentrations, and some of them (Co, Ag, Eu, Nb, Gd, Tb, Dy, Ho, Er, Lu, Hf and others) can be found as impurities in steels and vanadium alloys.
- Critical elements, with maximum allowable concentrations between 1 ppb and 1 ppm are the following: N, Ca, Co, Nb, Mo, Pd, Cs, Ba, Nd, Sm, Gd, Dy, Er, Tm, Yb, Lu, Hf, Os, Ir, Pt, Bi.
- There are also some elements (Ag, Eu, Tb, Ho, Th, U) with very critical limits, below 1 ppb. Detection of these impurities at these levels could be a very difficult issue for blanket materials.

3. Clearance of V–4Cr–4Ti in-vessel structures

The clearance option was analysed on V–4Cr–4Ti in-vessel structures, after a 100 years decay [7], a time period, which would still allow an institutional control in interim storage. Clearance (D) levels are taken from [4], or evaluated with fitting formulae, and reduction factors up to one order of magnitude have been applied.

Table 1 shows the method adopted to reduce the activity levels of the alloy. The 20 dominant radionuclides, i.e., those with the highest activities, see Ref. [8], are examined. Assuming that these nuclides represent the total specific activity of the alloy, disposal as non-active waste should be possible if the related clearance index, i.e., the sum of the A/L_c should be less than unity. Conversely it may be seen that all single fractions are greater than the unity by orders of magnitude. The concentrations of the parent elements are reduced to very low levels. This method was used to specify an ideal composition for the alloy, with very low impurity concentrations.

Purification factors F_p are then defined, equal to the fractions of the original concentrations on the reduced concentrations. Thus, reduced specific activities and new fractions $(A/L_c)_m$ are evaluated accordingly. This method was adopted for all radionuclides deriving from impurities. ^{42}K and ^{42}Ar specific activities cannot

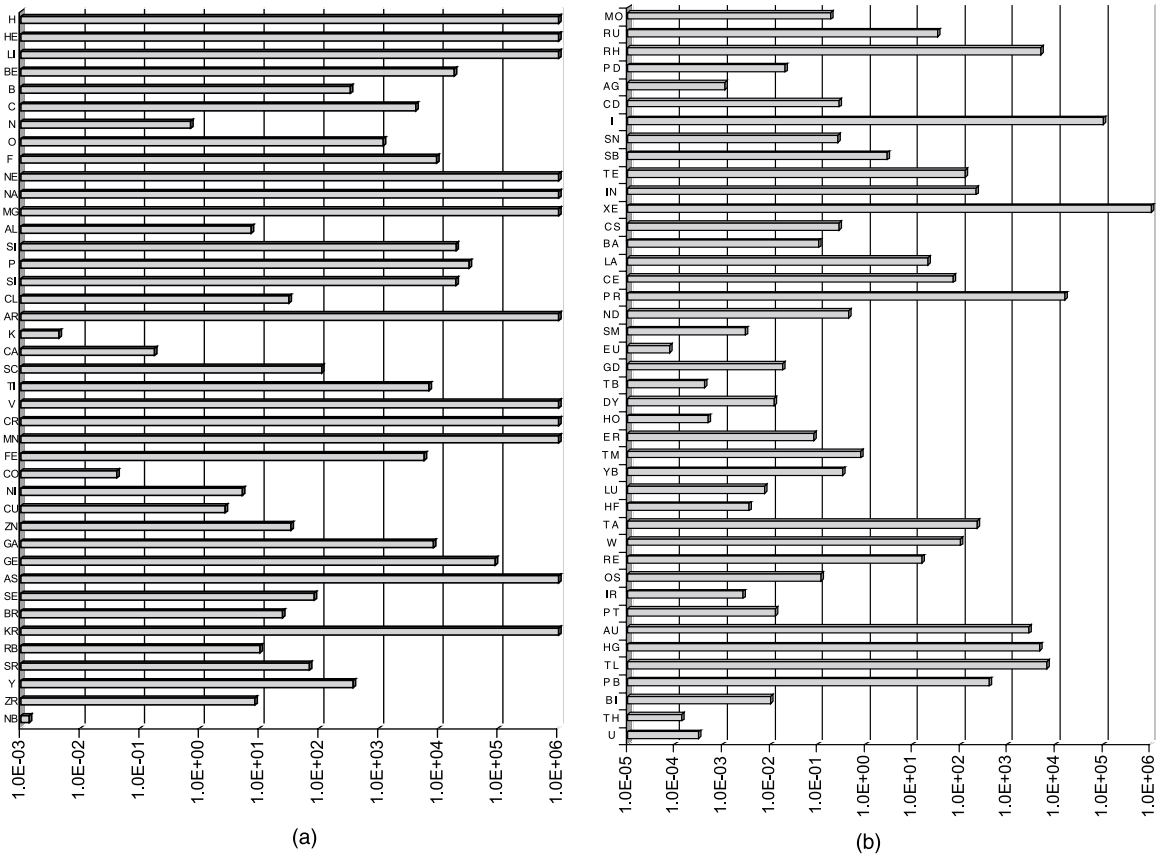

Fig. 1. Maximum allowable concentration (weight ppm) of elements in order to permit clearance of blanket materials in SEAFP Plant Model 2, after 100 years of cooling. (a) Elements from H to Nb, (b) elements from Mo to U.

be reduced, as they derive from Ti, which is a component of the alloy. The sum of $(A/L_c)_m$, in the last column of the table is still higher than unity, especially for the contributions arising from ^{42}K and ^{42}Ar daughters.

Reprocessing of the activated alloy should be considered. It could be based on elemental dilution: potassium, argon, strontium and other natural elements could be added to the alloy in such concentration to strongly dilute their radioactive isotopes. It may be observed that none of the radiotoxic nuclides is an isotope of the alloying elements. Therefore, the proposed method of reprocessing by elemental dilution could be applied to all of them. Radioactive and natural isotopes could be extracted together as high-concentration impurity; this secondary waste stream is expected to be small. Note that the above procedure has been applied only to the 20 dominant nuclides. It needs to be repeated for other radionuclides, i.e., the 21st, 22nd, ..., etc., if they make significant contributions to the clearance index.

Management options proposed here have been based on activation levels only: however, the influence of tritium permeated in the activated materials must be taken into account. Results of experiments performed at CEA [9] show that, after detritiation procedures, structural materials (such as steels) reprocessed from in-vessel zones have residual tritium outgassing rates such that the dose rate due to inhalation is well below 0.15 μSv/h, a negligible value compared to the hands-on recycling limit (10 μSv/h). Tritium detritiation, followed with a sufficiently long intermediate decay period, can also reduce tritium concentration well below the clearance levels for tritium (3×10^6 Bq/kg, see for instance Ref. [10]), then reducing its contribution to clearance indices to a negligible value.

4. Conclusions

Clearance can be considered as an alternative option for in-vessel materials. Vanadium-based alloys are at the moment the only materials with sufficiently low-activation constituting elements. Attainment of clearance conditions in activated in-vessel V–4Cr–4Ti structures would require:

Table 1
Reduction of impurities in V–4Cr–4Ti in-vessel structures to reduce the activities of the 20 dominant nuclides after 100 years of decay. Fluence: average blanket conditions

Nuclide	A (Bq/kg)	PE	Percentage of formation	A/L_c	$C_a \Rightarrow C_m$ (wppm)	F_p	$(A/L_c)_m$
Ar-39	5.6E+08	Ar	95.27	9.3E+4	129 ⇒ 1E−4	1.29E6	6.9E−2
		K	3.88		2.6 ⇒ 1E−4	2.6E4	1.4E−1
H-3	4.2E+08	–	–	–[a]			
Nb-91	7.8E+07	Mo	97	2.1E+4	180 ⇒ 1E−4	1.8E6	LIE−2
		Nb	3		31 ⇒ 1E−4	3.1E5	2.0E−3
C-14	2.1E+07	N	100	5.7E+2	100 ⇒ 1E−3	1.0E5	5.7E−3
Nb-93m	1.7E+07	Nb	100	5.5E+2	31 ⇒ 1E−4	3.1E5	1.8E−3
Mo-93	1.1E+07	Mo	100	6.8E+2	180 ⇒ 1E−4	1.8E6	3.8E−4
Ni-63	6.1E+06	Ni	66.78	8.6E+0	22 ⇒ 4E−4	55E4	l.OE−4
		Cu	11.94		0.6 ⇒ 4E−4	1.5E3	6.8E−4
		Zn	19.89		12 ⇒ 4E−4	3.0E4	5.7E−5
Nb-94	2.6E+06		97.4	5.3E+3	31 ⇒ 1E−04	3.1E5	1.6E−2
		Mo	2		80 ⇒ 1E−4	1.8E6	5.9E−4
Tc-99	1.6E+06	Mo	100	5.3E+1	180 ⇒ 1E−4	1.8E6	2.9E−5
Cs-137	1.6E+05	Xe-136	100	1.5E+2	50 ⇒ 2E−3	2.5E4	6.0E−3
Ba-137m	1.5E+05	Ba	49.15	1.4E+2	1.2 ⇒ 1E−4	1.2E4	5.7E−3
		Xe	49.8		50 ⇒ 2E−3	2.5E4	2.7E−3
Zr-93	1.4E+05	Zr	100	2.4E+0	920 ⇒ 1E−3	9.2E5	2.6E−6
Kr-85	1.2E+05	Kr	100	1.5E+1	2 ⇒ 4E−4	5E3	3.0E−3
Ni-59	6.0E+04	Ni	100	1.9E+0	22 ⇒ 4E−4	5.5E4	3.4E−5
Y-90	5.6E+04	Zr	100	1.4E+0	920 ⇒ 1E−3	9.2E5	1.5E−6
Sr-90	4.1E+04	Sr	100	1.4E+1	4E−2 ⇒ 4E−4	1E2	1.4E−1
		Y			2E−2 ⇒ 2E−4	1E2	(approx.)
Sm-151	4.1E+04	Sm	100	5.0E−1	l.2E−2 ⇒ 1E−4	1.2E2	4.1E−3
Al-26	2.5E+04	Al	99.956	6.2E+1	920 ⇒ 1E−2	9.2E4	6.7E−4
K-42	2.5E+04	Ti	100[b]	2.2E+1	Ti no reduction	1	2.2E+1
Ar-42	2.2E+04	Ti	100[b]	1.4E+1	Ti no reduction	1	1.4E+1
Rest	2.2E+04						
Total act.	3.1E+05						

A – activity, PE – parent element, C_a, C_m (wppm) – actual and modified concentration of parent element, A/L_c, $(A/L_c)_m$ – ratio of activity to clearance level, due to C_a and C_m respectively, F_p – purification factor. It is: $(A/L_c)_m = (A/L_c)/F_p$.

[a] Tritium content will be negligible, see Section 3 and Ref. [9].

[b] Approximate evaluation.

(a) very low concentrations of impurities, not attainable with the current purification methods and, in some cases, below the present detection limits and
(b) the development of methods to reprocess the activated alloy to extract radiotoxic nuclides. However, since V alloys are envisaged as structural materials for power fusion reactors, the time schedule should be sufficient to develop purification processes to reduce the levels of impurities.

Acknowledgement

It is a pleasure to thank ASP (Associazione per lo Sviluppo Scientifico e Tecnologico del Piemonte) for the financial support given to this research activity.

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