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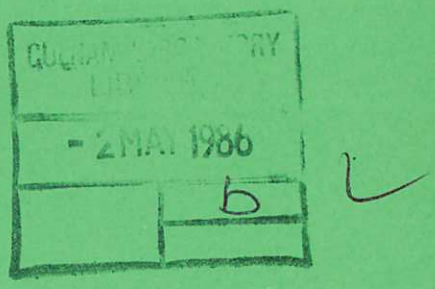

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IMPURITY ACTIVATION AND SURFACE γ-DOSE LEVELS IN FIRST WALL STRUCTURAL MATERIALS

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ABSTRACT

Research in the field of low-activity structural materials for the first wall and blanket of nuclear fusion reactors has assumed much importance in the last few years. In this report the effects of the presence of low concentrations of unintended impurities among the basic components of low-activation materials on their long term activation and dose rate levels have been investigated. A new library of neutron cross-sections has been generated with the THRES-F code for all those elements that are not traditional components of structural materials. The contribution of 1 ppm of each possible impurity element to the long term activity and γ-dose rate of a structural material has subsequently been determined with the aid of a modified version of the ORIGEN code.

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1. Introduction

In a D-T fusion reactor the main problems arising from induced radioactivity will probably be determined by the structural material chosen for the first wall and blanket. These components are likely to have service lives much shorter than the lifetime of the reactor and will therefore need to be replaced at regular intervals. Activated radiation-damaged material will thus accumulate over the life of the reactor and provision will need to be made for the storage and eventual treatment of such material.

In order to simplify the management of discarded structural material, research is in progress to develop materials having enhanced rates of decay of their induced activities, such that procedures for their reclamation or disposal following a suitable period of nuclear decay could be considered. Attention has largely been focused on low-activation versions of conventional stainless steels and on vanadium-based alloys. The activation and transmutation behaviour of the chemical elements forming the intended constituents of these materials are fairly well established and are described in the literature [1-5]. The activation properties of many of the less common elements are, however, subject to considerable uncertainty, owing to inaccurate or missing values of the cross-sections for relevant neutron activation reactions. There is a particular problem with some of the elements that are likely to be present only as trace impurities in highly-refined low activation materials but which, because of their long half-lives, give rise to a high long-term activity level in the composite material.

Efforts to determine the effect of impurities are strongly hindered by the lack of neutron activation cross-section data for those tramp elements that do not belong to the traditional range of structural materials, namely for nuclei with Z between about 52 and 70. First order estimates of the activation of such elements have been calculated by Jarvis [3], in which only primary 14 MeV neutron-induced reactions have been considered. Though only approximate, the results have served to identify potentially troublesome elements giving rise to high levels of long-lived activity even at very low concentration, for instance Eu and Tb, and to emphasise the need for more detailed evaluations.

The present study is intended to provide more accurate estimates of long-term activity and dose-rate for all possible impurities. The THRES-F code [6] has been utilised to generate neutron cross-sections for all the relevant isotopes and reactions of the elements concerned. The neutron induced radioactivity is then calculated by means of a modified version of the ORIGEN code [7] which represents all the possible routes for production of any particular radionuclide.

Although the results from the present work are still only approximations, owing to uncertainties in the computed cross-sections and to some conservative assumptions made in the calculations, they can be regarded as giving a 'second order' estimate of the activity and surface dose rate contributions to be expected from the elements studied.

2. Description of conditions

The starting point was the list of potentially troublesome elements identified by Jarvis [3], presented here with minor modifications as Table I, and for which no cross-section data are included in the UKCTR-III-A library [8]. The objective was to determine the neutron induced activity and the surface dose rate associated with the presence of 1 part per million by mass (hereinafter referred to as 1 ppm) of each of the elements listed in Table I in a matrix of medium Z material, such as steel or a vanadium alloy, after a cooling time of 100 years or longer.

The irradiation conditions were assumed to be those appropriate to the Culham DEMO reactor [9] which has a helium-cooled blanket with martensitic stainless steel as the structural material and a mixture of beryllium and lithium ceramic as breeder. The neutron wall loading was scaled up to 5 MWm^{-2} to approach the conditions of more advanced designs and spectra at both the first wall and the rear part of the blanket (Fig.1) have been used in order to assess the dependence of the results on the neutron energy spectrum. The neutron fluxes were determined with the aid of the one-dimensional code ANISN [11] for a cylindrical-geometry model of the reactor.

The irradiation time was fixed at 2.5 years with 100% load factor whilst decay calculations have been performed for cooling times ranging between 100 and 2500 years. Illustrative data are presented for 100 and 250 years cooling time.

TABLE I: Elements considered in the present study and their main long-lived products ($5y \lesssim T_{\frac{1}{2}} \lesssim 10^{12}$ y)

Z	ELEMENT	Long-lived products
31	Ga	-
32	Ge	-
33	As	-
34	Se	^{79}Se*
35	Br	^{79}Se*, ^{81}Kr
36	Kr	^{79}Se*, ^{81}Kr, ^{85}Kr
37	Rb	^{85}Kr
38	Sr	^{85}Kr
44	Ru	^{97}Tc*, ^{98}Tc, ^{99}Tc*
45	Rh	^{99}Tc*
46	Pd	^{107}Pd*, ^{108m}Ag
51	Sb	-
52	Te	^{129}I*
53	I	-
54	Xe	^{135}Cs*, ^{137}Cs
55	Cs	^{135}Cs*, ^{137}Cs
56	Ba	^{137}Cs, ^{133}Ba
57	La	^{137}Cs, ^{137}La*
58	Ce	^{137}La*
59	Pr	^{137}La*
60	Nd	^{145}Pm*, ^{146}Pm, ^{151}Sm* ^{150m}Eu, ^{152}Eu
62	Sm	^{145}Pm*, ^{146}Sm*, ^{151}Sm*, ^{150m}Eu, ^{152}Eu, ^{154}Eu
63	Eu	^{151}Sm*, ^{150m}Eu, ^{152}Eu ^{154}Eu
64	Gd	^{151}Sm*, ^{150m}Eu, ^{152}Eu ^{154}Eu
65	Tb	^{157}Tb*, ^{158}Tb
66	Dy	^{157}Tb*, ^{158}Tb
67	Ho	^{166m}Ho
68	Er	^{163}Ho*, ^{166m}Ho
69	Tm	^{166m}Ho
70	Yb	-
71	Lu	-
72	Hf	-
75	Re	^{186m}Re
76	Os	^{186m}Re
77	Ir	$^{192m_2}Ir$
78	Pt	$^{192m_2}Ir$, ^{193}Pt*
79	Au	-
80	Hg	-
83	Bi	^{207}Bi, ^{208}Bi, ^{209}Po

* = associated radiations fall below 200

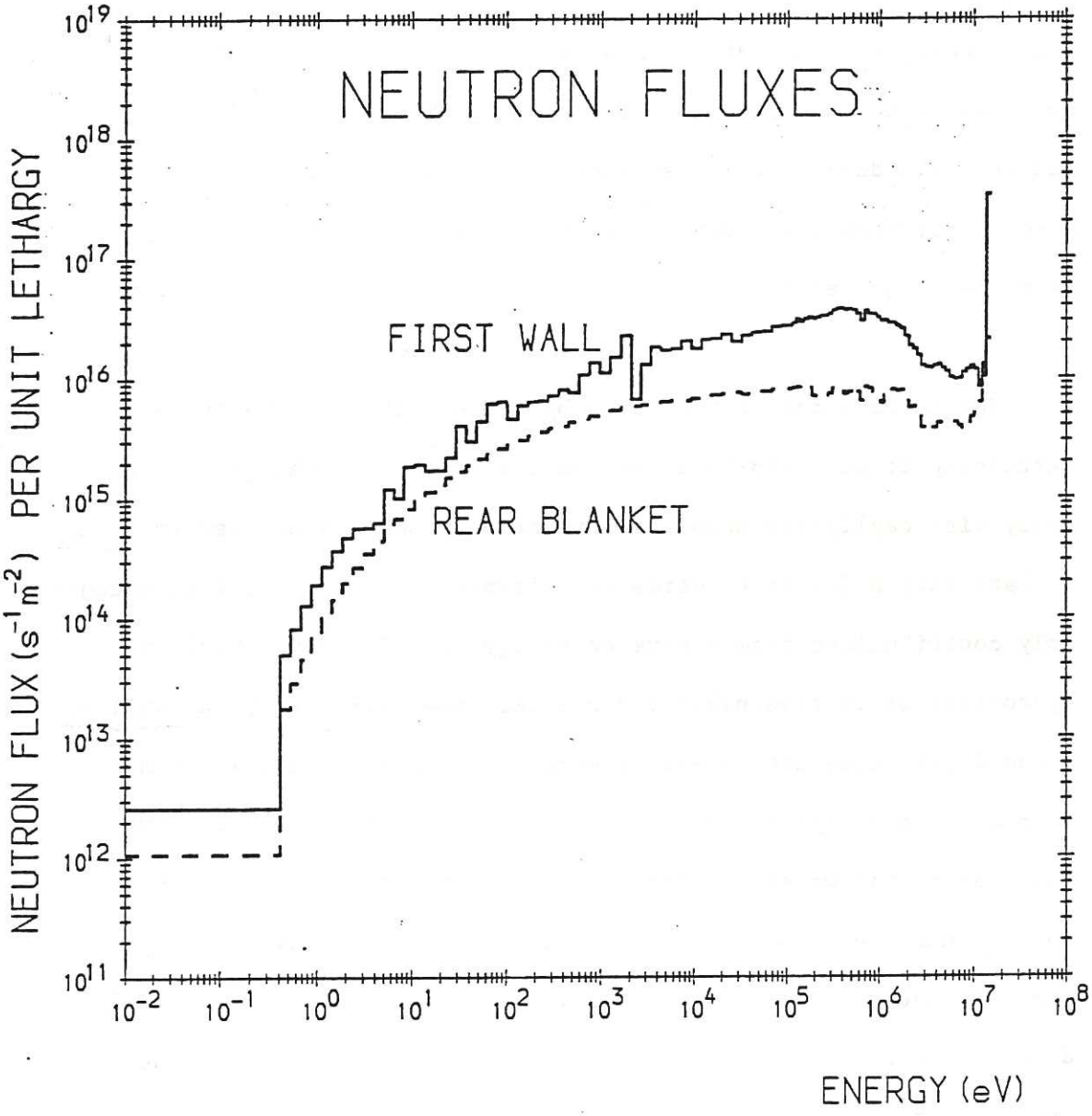

Fig.1 Neutron fluxes considered in the present study.

3. Method of calculation

The procedure consists essentially of two steps; the generation of a set of neutron reaction cross-sections for the elements not represented in the UKCTR-III-A activation library, and the use of the ORIGEN code, with the above data as input, to calculate the activity and dose rate as functions of the cooling time. Recent data on the isotopic abundances, decay schemes and emission energy for most of the elements studied have been taken from Ref.10 and defined in the input libraries required by the code.

The surface dose rates given by the ORIGEN code are those pertaining to an infinite thick sheet of material, though they also apply with negligible error to a relatively small area, say 1m x 1m, of a plate only a few centimetres in thickness. They take into account only contributions from γ-rays of energy over 200 keV. Such an approximation is reasonable because less energetic γ-rays as well as α and β particles can be easily screened with a relatively thin shield, whereas hard γ-rays would require such a thick shield that direct handling of the waste material would be impractical. In the present work it has been assumed that the elements under study are dispersed in a host material consisting of iron. Thus the results should apply directly to ferrous alloys and also to vanadium, which has a density and nuclear charge similar to that of iron.

As a first step in the calculations a new neutron activation cross-section library has been assembled, taking into account all the reactions that could lead to the production of a long-lived isotope from the initial stable element. All the cross-sections except those of the (n,γ) reactions have been calculated using the THRES-F code [6],

with normalisation of the results to experimental data where these are available. In constructing this library the format and the 100-group structure of the UKCTR-III-A library has been utilised.

The most important neutron absorption cross-sections have been added to the library, using the data from the literature [12,13] and, following any required interpolation or extrapolation, transforming them into the same 100-energy-group format of the other cross-sections. The resulting data are least accurate in the low-energy region below 100 keV, where resonances begin to be important, though the inaccuracies are not expected to affect the results appreciably since the fusion spectrum is dominated by the high-energy component.

In the cases where isomeric nuclei are formed and where the branching ratio is unknown the reactions have been assumed to lead to the metastable state if this is long-lived, which is a pessimistic approximation.

4. Activity calculations

For all the elements listed in Table I the long-term activity has been calculated for a concentration of 1 ppm in a ferrous matrix and for two neutron fluxes, namely the first-wall flux and the flux in the last 2 centimetres of the blanket region, referred to as the rear blanket flux.

The results of the calculations are presented in Table II for a cooling time of 100 years. The activity of newly refined uranium metal, namely $2.04 \cdot 10^7$ Bq kg^{-1}, has been adopted as a reference level and all the activities below one millionth of this value have been excluded.

TABLE II: Activity (Bq kg^{-1}) for 1 ppm of impurity element.
First wall and rear blanket fluxes (5 MWm^{-2} wall loading)
• 2.5 y operating time; 100 y cooling time •

Chemical Symbol	First wall Activity	Rear blanket Activity
Se	5.08E+04*	1.47E+04
Br	1.32E+03	1.61E+02
Kr	4.31E+05	1.74E+05
Rb	3.28E+03	5.20E+03
Sr	3.25E+04	2.91E+03
Ru	1.98E+02	1.97E+01
Rh	4.84E+01	-
Pd	1.26E+04	3.54E+02
Sb	2.19E+01	-
Te	9.09E+01	-
I	2.10E+01	-
Xe	2.33E+04	8.78E+03
Cs	3.16E+04	2.59E+03
Ba	2.25E+04	2.61E+03
La	2.22E+03	4.20E+01

Chemical Symbol	First wall Activity	Rear Blanket Activity
Ce	1.60E+02	-
Pr	3.79E+01	-
Nd	2.57E+06	1.14E+06
Sm	2.20E+07	1.08E+07
Eu	1.99E+07	1.53E+07
Gd	7.36E+03	1.76E+03
Tb	2.28E+07	2.84E+06
Dy	1.39E+05	7.03E+04
Ho	2.81E+06	1.92E+06
Er	3.72E+05	1.02E+05
Tm	1.33E+03	1.52E+02
Re	1.04E+05	1.02E+05
Ir	1.54E+07	2.13E+06
Pt	6.88E+06	9.01E+05
Bi	5.73E+05	1.13E+04

* Read $5.08 \cdot 10^4$

Since even for pure low-activation steels the total activity after 100 y cooling time is still greater than 10^8 Bqkg^{-1}, the results show that at concentrations of the order 1 ppm the elements considered do not contribute significantly to the activity of the host material. The specific activity of a material is not, however, an accurate indication of its biological hazard, which depends on the nature and energy of the particles released rather than simply on the number of disintegrations per unit time. For this purpose the surface γ dose rate, taking into account the energy of the emitted γ-rays, is a more representative parameter, as will be discussed in the following section.

5. Surface γ-dose rate calculations

The calculated surface dose rates associated with the presence of 1 ppm of the selected elements in the base material are given in Table III for a cooling time of 100 years. The results can also be expressed in another convenient form; taking the limit of $2.5 \cdot 10^{-5}$ Svh^{-1} as an acceptable surface dose rate, which is the maximum permissible dose for a radiation worker present for a 40-hour working week, the maximum allowable concentration has been determined for each element, in units of ppm of the base material, such that the dose rate from the element does not exceed this value after the illustrative cooling times of 100 and 250 years. The results of these calculations are presented in Table IV, in which only the elements restricted to less than 100% of the material are included.

It is seen that five of the elements listed, namely Eu, Tb, Ho, Ir and Bi, satisfy the above dose rate criterion only if present at concentrations well below 1 ppm, with only a slight relaxation at the

TABLE III: Surface dose rate (Sv h^{-1}) for 1 ppm of impurity element
First wall and rear blanket fluxes (5 MWm^{-2} wall loading)
• 2.5 y operation time; 100 y cooling time •

Chemical Symbol	Surface dose rate first wall	Surface dose rate rear blanket
Br	8.92E-10*	4.52E-11
Kr	2.58E-07	1.05E-07
Rb	1.91E-09	3.04E-10
Sr	1.91E-09	1.71E-09
Ru	1.64E-09	1.39E-10
Rh	2.84E-11	-
Pd	4.11E-06	6.74E-08
Xe	2.70E-06	1.02E-06
Cs	1.20E-06	7.22E-08
Ba	1.55E-06	1.52E-07
La	1.18E-10	-
Ce	1.28E-10	-

Chemical Symbol	Surface dose rate first wall	Surface dos rate rear blanke
Nd	4.08E-07	1.05E-07
Sm	4.33E-06	1.09E-06
Eu	5.28E-03	4.13E-03
Gd	1.31E-06	3.32E-07
Tb	4.96E-03	6.36E-04
Dy	3.54E-06	1.27E-07
Ho	8.21E-04	5.63E-04
Er	7.88E-07	6.36E-08
Tm	3.86E-07	4.42E-08
Re	2.13E-09	2.09E-09
Ir	9.87E-04	1.44E-04
Pt	3.13E-07	7.17E-09
Bi	2.30E-04	4.91E-06

* Read $8.92 \cdot 10^{-10}$

TABLE IV: Number of ppm (by weight) permitted for troublesome impurity elements

ELEMENT	First wall flux neutron wall load: 5 MW m^{-2}		Rear blanket flux neutron wall load: 5 MW m^{-2}	
	100 y surface dose criterion	250 y surface dose criterion	100 y surface dose criterion	250 y surface dose criterion
35-Br	$2.8 \cdot 10^4$(2.8%)	$2.8 \cdot 10^4$(2.8%)	$5.53 \cdot 10^5$(55.3%)	$5.53 \cdot 10^5$(55.3%)
36-Kr	96.9	3440.	238.	6650.
37-Rb	$1.31 \cdot 10^4$(1.3%)	no limit	$8.22 \cdot 10^4$(8.22%)	no limit
38-Sr	1310.	no limit	$1.46 \cdot 10^4$(1.46%)	no limit
44-Ru	$1.52 \cdot 10^4$(1.5%)	$1.52 \cdot 10^4$(1.5%)	$1.80 \cdot 10^5$(18%)	$1.80 \cdot 10^5$(18%)
45-Rh	$8.80 \cdot 10^5$(88%)	no limit	no limit	no limit
46-Pd	6.08	13.8	371.	845.
54-Xe	9.26	295.	24.5	767.
55-Cs	20.8	$4.16 \cdot 10^5$(41.6%)	346.	no limit
56-Ba	16.1	828.	164.	$1.18 \cdot 10^4$(1.18%)
57-La	$2.12 \cdot 10^5$(21.2%)	no limit	no limit	no limit
58-Ce	$1.95 \cdot 10^5$(19.5%)	no limit	no limit	no limit
60-Nd	61.3	6980.	238.	$8.31 \cdot 10^4$(8.31%)
62-Sm	5.77	772.	22.9	8220.
63-Eu	$4.73 \cdot 10^{-3}$	0.56	$6.05 \cdot 10^{-3}$	2.15
64-Gd	19.1	$1.63 \cdot 10^4$(1.63%)	75.3	$3.02 \cdot 10^5$(30.2%)
65-Tb	$5.04 \cdot 10^{-3}$	0.01	0.04	0.08
66-Dy	7.06	14.12	197.	393.
67-Ho	0.03	0.033	0.044	0.05
68-Er	31.7	34.6	393.	429.
69-Tm	64.8	70.6	566.	617.
75-Re	$1.17 \cdot 10^4$(1.17%)	$1.17 \cdot 10^4$(1.17%)	$1.20 \cdot 10^4$(1.2%)	$1.20 \cdot 10^4$(1.2%)
77-Ir	0.025	0.039	0.17	0.27
78-Pt	79.9	123.	3490.	5360.
83-Bi	0.11	1.32	5.09	16.9

longer cooling time. Furthermore, this conclusion is not very sensitive to the change in neutron flux in passing from the first wall to the rear blanket, although for some other elements this results in an increase of about one order of magnitude in the permitted concentration.

6. Bremsstrahlung dose rate correction

The γ-dose rate estimates given in the previous section do not take into account the bremsstrahlung radiation accompanying high-energy β-emissions. In the present study an estimate has been made of the bremsstrahlung contribution to the surface γ-dose rate from the impurity elements. Such a contribution has been determined using the equations given in Appendix 1 and compared with the results given in section 5.

In general the corrections to the direct γ-dose values at 100 y cooling time are found to be less than 1% and can thus be considered negligible, except for the case of rhenium where the dose rate due to bremsstrahlung radiation is dominant. In this element the radiation arises from β-decay of the isotope ^{186}Re, a daughter product of the long-lived isomer ^{186m}Re, with the emission of electrons having two different end-point energies, namely 0.9 MeV (21% intensity) and 1.05 MeV (74% intensity). For 1 ppm of Re the surface dose rate is $3.65 \cdot 10^{-8}$ Svh^{-1} after 100 years cooling time and this corrected value leads to the more restrictive permitted concentration of 680 ppm, with no relaxation for longer cooling time because the half-life of ^{186m}Re is greater than 10^7 years. This result is pessimistic, however, since all the production routes of the isotope ^{186}Re have been assumed to lead to its long-lived metastable state.

In conclusion, whilst the calculations indicate that the bremsstrahlung contribution is negligible in the case of many of the elements, the result for rhenium highlights the importance of considering this component in dose rate calculations for elements besides those examined here.

7. Residual impurity content of low-activation steels

Efforts to develop low-activation materials have so far concentrated on the selection of elements for the intended alloying constituents. There is currently much uncertainty as to the likely concentrations of the unintended elements that will inevitably be present to some degree as impurities, depending on the basic composition of the alloy, the sources of the starting materials, the fabrication processes adopted and so on.

Nevertheless, a tentative list of impurity elements likely to be present in any low-activation steels produced by current steelmaking technology has been drawn up by the British Steel Corporation [14] and this is shown in Table V. It must be emphasized that the concentrations indicated in Table V are only estimates and are based on existing steelmaking practices, with careful selection of the starting materials. In the table the number of ppm for each element is given and is compared with the corresponding permitted level for the 100-y surface dose rate criterion in the case of first wall irradiation conditions. In the case of elements not investigated in the present study the permitted level values have been taken from Ref.3. From a comparison between the two sets of values it emerges that for 6 elements the estimated level in low activation steels is much higher than the permitted level. Consequently for such elements further

TABLE V: Present estimates of the impurity levels in low-activation steel compared with the permitted levels (100 y cooling time).

Element	Estimated level (ppm)	Permitted level (ppm)
13-Al	10	30
19-K	2	$4 \cdot 10^5$
20-Ca	2	$5 \cdot 10^4$
27-Co	10	10^3
28-Ni	100	10^4
29-Cu	50	10^4
40-Zr	30	$3 \cdot 10^{-3}$
41-Nb	20*	1
42-Mo	50	100
47-Ag	0.5*	0.01

Element	Estimated level (ppm)	Permitted level (ppm)
50-Sn	50	10^4
56-Ba	2	16
58-Ce	4	$2 \cdot 10^5$
60-Nd	2	61
62-Sm	0.5	6
63-Eu	0.2*	$5 \cdot 10^{-3}$
65-Tb	0.5*	$5 \cdot 10^{-3}$
77-Ir	0.1*	0.025
83-Bi	5*	0.11

* Greater than the permitted level.

investigations are required and the next section provides a more detailed specification of the problem.

8. Detailed discussion for critical impurities

The data presented in sections 5 and 7 show that certain elements could cause difficulties in the production of low-activation structural materials. In particular niobium, silver, europium, terbium, holmium, iridium and bismuth appear to be critical impurities that could be tolerated only at extremely low levels if the possibility of hands-on or semi-remote handling of scrap material is to be contemplated after a reasonable storage period. In the case of these elements it is worthwhile to consider in a little more detail the accuracy of the activation data presented here, in order to gain some appreciation for the reliability of the results and their sensitivity to the pertinent reaction cross-sections, not all of which are known with a high accuracy.

A list of the isotopes responsible for the long-term surface dose rate and of the main reactions which lead to their production is presented in Table VI. Although not listed in the table, all the burn-up reactions of such isotopes could be important to the total balance of the final concentration and for this reason they should be considered with high priority in the cross-section data evaluation programme. The accuracy of available data for the production reactions listed in Table VI is not the same for all the reactions. In the case of Nb and Ag the data are those contained in the UKCTR-III-A library. These are derived from experimental evaluations and it can be assumed that their values are well established, to an accuracy of about 20%. For the other elements listed in Table VI it is seen that all the main

TABLE VI: Detailed data for particularly troublesome elements

Element	Isotopes produced	Half-life (y)	Gamma energy (MeV) $E\gamma>0.2$ MeV	Main production reactions
Nb	^{94}Nb	$2.4 \cdot 10^4$	1.56	$^{93}Nb(n,\gamma)$
Ag	^{108m}Ag	$1.3 \cdot 10^2$	1.60	$^{107}Ag(n,\gamma)$;f = 0.0081 $^{108}Ag(n,2n)$;f = 0.39
Eu	^{150m}Eu	36.	1.28	$^{151}Eu(n,2n)$;f = 1.00
	^{152}Eu	13.4	0.99	$^{151}Eu(n,\gamma)$ ^{153}Eu (n,2n)
	^{154}Eu	8.5	0.81	$^{153}Eu(n,\gamma)$
Tb	^{158}Tb	150.	0.71	$^{159}Tb(n,2n)$
Ho	^{166m}Ho	$1.2 \cdot 10^3$	1.51	$^{165}Ho(n,\gamma)$;f = 0.06
Ir	$^{192m_2}Ir$ (^{192}Ir)	241. (73.8d)	- (0.76)	$^{191}Ir(n,\gamma)$;f = 0.001 $^{193}Ir(n,2n)$;f = 1.00
Bi	^{207}Bi	32.2	1.47	$^{208}Bi(n,2n)$
	^{208}Bi	$3.68 \cdot 10^5$	2.59	$^{209}Bi(n,2n)$

* f = fraction leading to metastable state (utilized value).

production reactions involve (n,2n) or (n,γ) cross-sections. The former have been calculated by means of the THRES-F code, the results from which have shown agreement within 30% with experimental measurements for this particular reaction, so that this order of accuracy can be assumed. Data for (n,γ) cross-sections have been taken from Ref.12 and, although the corresponding accuracies are not specified, they are expected to be correct within a factor 2. Since such reactions are the ones mainly responsible for the calculated dose-rate levels, the accuracy of the results follows directly from the accuracy of the cross-section value concerned, although no data are available for taking into account spectrum effects.

In the case of metastable isotopes the branching ratio, f, has been indicated for each reaction concerned. The ORIGEN code requires the definition of an energy-independent value, so such an approximation has been made. In those cases where a value of f smaller than unity is shown the data come from experimental values and here a reasonable accuracy could be expected. In the remaining cases, where f is not known, the assumption that all the production routes lead to the metastable isomer has been made. This assumption results in an over-pessimistic estimation by at least a factor 2.

In general a relaxation of the allowable concentration of an element will be obtained only if cooling times comparable with the half life of the controlling radionuclide are considered. Thus, of the particularly troublesome elements listed in Table VI, the permitted concentrations for Nb and He, both depending on very long-lived isotopes, remain the same whether the cooling time criterion of 100 y or 250 y is chosen. The allowable concentrations of the other elements, depending totally or, as in the case of Bi, partially on

radioisotopes with half lives shorter than 250 y, show some relaxation for the longest cooling time criterion. This is particularly important in the case of Eu for which the permitted concentration becomes greater than the estimated one given in Table V.

9. Conclusions

The present study has underlined the importance of taking into account the presence of low concentrations of impurities in the composition of low-activity materials when calculating the long term activation and dose rate. If, in particular, the ability to handle the material with minimal precautions after a storage time no longer than 100 years is placed as a requirement on candidate low-activity compositions then the presence of certain tramp elements, especially Ag, Nb, Eu, Tb, Ho, Ir and Bi, could be very deleterious and their permissible concentrations have been determined to be well below 1 ppm. For these seven elements further studies are required, firstly to establish a more exact and complete activation data base and secondly to examine the potential for their elimination from proposed low-activity alloy compositions.

In the present calculations the bremsstrahlung radiation associated with β-emission has been found to have a negligible effect on the total dose rate except in the case of ^{186}Re, where it is dominant. This result shows, however, that this contribution could be important in γ-dose rate calculations for basic components of structural materials.

Appendix 1: Contribution to surface γ-dose rate from bremsstralung radiation accompanying β-emissions.

In general the surface dose rate from a thick slab of material is approximately represented by the equation [3]:

$$D = 5.76 \cdot 10^{-10} \mu_a S_v \frac{B}{2\mu_M} \left[Gy\ h^{-1}\right], \qquad (A.1)$$

where μ_a = γ-energy absorption coefficient of the atmosphere,

μ_m = linear attenuation coefficient of the material,

B = build-up factor (approximately 2),

S_v = rate of gamma emission $\left[MeV\ kg^{-1}\ s^{-1}\right]$.

If E_γ is the energy of the γ-radiation and N_γ the number of quanta per decay, then:

$$S_v(t) = E_\gamma N_\gamma A(t), \qquad (A.2)$$

where A(t) = activity of the material $\left[Bq\ kg^{-1}\right]$

In order to determine the value of N_γ associated with bremsstrahlung the following formula, valid for continuous radiation from monoenergetic electrons and for $E_\gamma \gtrsim E_m$ = 200 keV (consistent with the assumption made in previous calculations), has been used [3]:

$$N_\gamma = 6 \cdot 10^{-4}\ Z\ (U\ \ell nU - (U - 1)), \qquad (A.3)$$

where $U = \frac{E}{E_m}$, Z = charge of host material, and

E = average electron energy

The average electron energy in β-emissions is about half the end-point energy. The latter has been taken from Ref.10 for the long-lived isotopes and the corresponding values of N_γ computed, assuming the nuclear charge of Z=26 of iron. From Eq (A.1), setting $\mu_a = 3 \cdot 10^{-3}$ (m^2 kg^{-1}) for the relevant energy range and μ_m between 0.014 and $6 \cdot 10^{-3}$ (m^2 kg^{-1}) for E_γ between 0.2 and 1.0 MeV, the surface dose rate contribution from bremsstrahlung can be calculated, using the activity values given by ORIGEN and the same γ-energy group structure of the main dose-rate calculations.

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