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Activation of beryllium in a fusion power plant

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Abstract

The activation and transmutation behaviour of beryllium in the first wall of a fusion power plant has been studied using the up-to-date inventory code FISPACT-97. Assuming detritiation, the activity and dose rate will be determined by the impurities initially present in the material. The possible influences of sequential charged particle reactions and of trace amounts of Th and U as fissionable actinides are examined but found unlikely to be important. Beryllium appears to offer the prospect of reuse in further plant following minimal processing after decay storage for about 50 yr. © 1998 UKAEA. Published by Elsevier Science B.V. All rights reserved.

1. Introduction

Many conceptual designs for a fusion power plant involve the use of beryllium as a plasma-facing armour material or in substantial quantities as a neutron multiplier in the blanket. For a sustainable long-term fusion energy economy, it is desirable that used beryllium be recyclable, since global resources of this metal are limited. The present study examines the activation and transmutation behaviour of beryllium, with emphasis on the options for post-service management of beryllium components.

Since beryllium itself generates few radionuclides, the γ-dose rate from end-of-service beryllium will be dominated by the activation products of impurities. The influence of typical impurities, including some actinides, as well as possible contributions from secondary charged particle reactions, have been studied with the aid of the state-of-the-art activation code FISPACT-97.

2. Activation calculations

The results presented here assumed a continuous exposure for 5 yr in the outboard first wall position of the SEAFP-2 plant model PM-2 [1], which employs Be as plasma-facing armour, low activation martensitic steel as structural material, water as coolant and $Li_{17}Pb_{83}$ as tritium generator. The outboard first wall power loading is taken to be 2.4 MW m^{-2}, corresponding to a lifetime neutron fluence of 1.49×10^{27} n m^{-2} in the irradiated Be.

The calculations were performed with the European Activation System (EASY), which has been developed as a self-consistent set of data libraries and codes [2–4]. The newly updated inventory code FISPACT-97 used within EASY incorporates several facilities that enhance the reliability of activation calculations for beryllium. It includes the effects of sequential charged particle reactions and can treat modest concentrations of actinide impurities liable to fission. The latest version provides an improved estimate of the γ-dose by using new photon absorption data and a new calculational procedure, and calculations of the uncertainties in the output arising from uncertainties in both the cross-sections and half-lives of relevant nuclei. In addition it uses the EAF-97 data libraries which contain many improved cross-section data.

3. Pure beryllium

Natural beryllium consists entirely of the isotope ^9Be. At shutdown the dominant isotopes from irradiation of pure Be are as shown in Table 1. The He and Li isotopes are very short lived and within a few minutes the only

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PII: S 0 0 2 2 - 3 1 1 5 (9 8) 0 0 1 1 7 - 2

Table 1
Dominant nuclides at shutdown from pure beryllium

Isotope	Activity, %	γ-dose rate, %	Half-life, s
^{6}He	79.6	99.9	8.08×10^{-1}
^{3}H	20.4	0	3.89×10^{8}
^{8}Li	2.7×10^{-3}	2.9×10^{-2}	8.38×10^{-1}
^{11}Be	2.9×10^{-5}	1.7×10^{-2}	13.8
^{10}Be	3.6×10^{-6}	3.0×10^{-7}	5.05×10^{13}

Total activity = (7.9 ± 3.9) × 10^{14} Bq kg^{-1}, total activity excluding ^{3}H = 6.3 × 10^{14} Bq kg^{-1}, total γ-dose rate = (7.6 ± 4.6) × 10^{2} Sv h^{-1}.

significant remaining radionuclei are ^{3}H and ^{10}Be. Almost all of the ^{3}H is generated through the two reactions:

$$^{9}\mathrm{Be}(\mathrm{n},\alpha)^{6}\mathrm{He}(\beta^{-})^{6}\mathrm{Li}(\mathrm{n},\alpha)^{3}\mathrm{H} \quad (58\%),$$

$$^{9}\mathrm{Be}(\mathrm{n},^{7}\mathrm{Li})^{3}\mathrm{H} \quad (33\%),$$

the ^{10}Be is formed entirely by the reaction

$$^{9}\mathrm{Be}(\mathrm{n},\gamma)^{10}\mathrm{Be}.$$

Following initial decay, the composition of the irradiated material changes only gradually and the elemental constituents after 1 day are indicated in Table 2. Some 2.2% of the Be is burned up in the irradiation, the main products being He (1.8%) and Li (0.08%), with smaller quantities of H (0.047%). If present at the concentration predicted by FISPACT, tritium would dominate the activity and the ingestion and inhalation doses of the material. Hydrogen, however, is mobile in metals and tends to diffuse out, especially at elevated temperatures. Moreover, the tritium inventory in armour beryllium would probably be dominated by interchange with the plasma. However, the bulk of any tritium retained in the material at the end of its service life would be extracted in a detritiation operation soon after its removal from the plant. In the absence of tritium, the used material (after 1 day) would have a specific activity of 2.84(±0.70) × 10^{7} Bq kg^{-1} and a contact γ-dose rate (from bremsstrahlung) of 2.32 μSv h^{-1}, due entirely to the nuclide ^{10}Be. Since this isotope has a very long half-life of about 10^{6} yr, these values effectively remain constant over all timescales of practical interest. The low external γ-dose rate should, however, permit 'hands-on' recycling of the used material, with suitable precautions against internal exposure from ingestion or inhalation. Since the scrap material would need to be remelted, the more volatile transmutation products He and Li could readily be removed in such an operation. Although the reclaimed beryllium would be too active for unrestricted reuse, it should be suitable for further use within the fusion energy industry.

4. Impure beryllium

In order to explore the influence of potential impurities, activation calculations were performed for impure beryllium having the nominal composition given in Table 3 (column 2). It was found that the specific activity and the ingestion and inhalation doses for this composition were, within the uncertainties of the calculation, identical to those of ideally-pure beryllium. As illustrated by Fig. 1, the γ-dose rate, on the other hand, was dominated at all post-shutdown times by the activation products of impurities. Reference to Fig. 1 shows that the dose rate decreases to 10 mSv h^{-1} at about 50 yr and continues to fall until about 100 yr, after which its decline is very slow. In the region 10–100 yr the γ-dose is dominated by ^{60}Co ($T_{1/2} = 5.3$ yr) and beyond 100 yr by a number of long-lived isotopes such as ^{26}Al. With the assumed impurity content the dose rate would not reach a level, around 10 μSv h^{-1}, at which 'hands-on' recla-

Table 3
Elemental compositions in mass percent of impure beryllium prior to irradiation, following 50 yr decay storage after first service irradiation, and after 50 yr decay storage following second service irradiation

Element	Starting composition	After first service cycle	After second cycle
H	–	4.44×10^{-3}	2.88×10^{-2}
He	–	1.80	3.60
Li	–	8.01×10^{-2}	1.22×10^{-1}
Be	99.415	97.2	95.2
B	–	2.76×10^{-7}	9.47×10^{-7}
C	–	2.34×10^{-3}	4.66×10^{-3}
O	4.50×10^{-1}	4.47×10^{-1}	4.44×10^{-1}
Mg	5.00×10^{-3}	5.36×10^{-3}	5.71×10^{-3}
Al	2.00×10^{-2}	1.98×10^{-2}	1.96×10^{-2}
Si	3.00×10^{-2}	2.98×10^{-2}	2.95×10^{-2}
Ti	6.00×10^{-3}	6.01×10^{-3}	6.00×10^{-3}
Cr	5.00×10^{-3}	5.07×10^{-3}	5.16×10^{-3}
Mn	–	7.85×10^{-4}	1.48×10^{-3}
Fe	6.00×10^{-2}	5.93×10^{-2}	5.87×10^{-2}
Co	1.00×10^{-4}	1.12×10^{-4}	1.20×10^{-4}
Ni	9.00×10^{-3}	8.80×10^{-3}	8.61×10^{-3}
Mo	1.00×10^{-4}	9.83×10^{-5}	9.66×10^{-5}

Table 2
Elemental composition of irradiated pure Be (5 yr @ 2.4 MW m^{-2} wall loading) 1 day after shutdown

Element	H	He	Li	Be	B
g kg^{-1}	4.67×10^{-1}	17.6	8.06×10^{-1}	978	3.25×10^{-7}

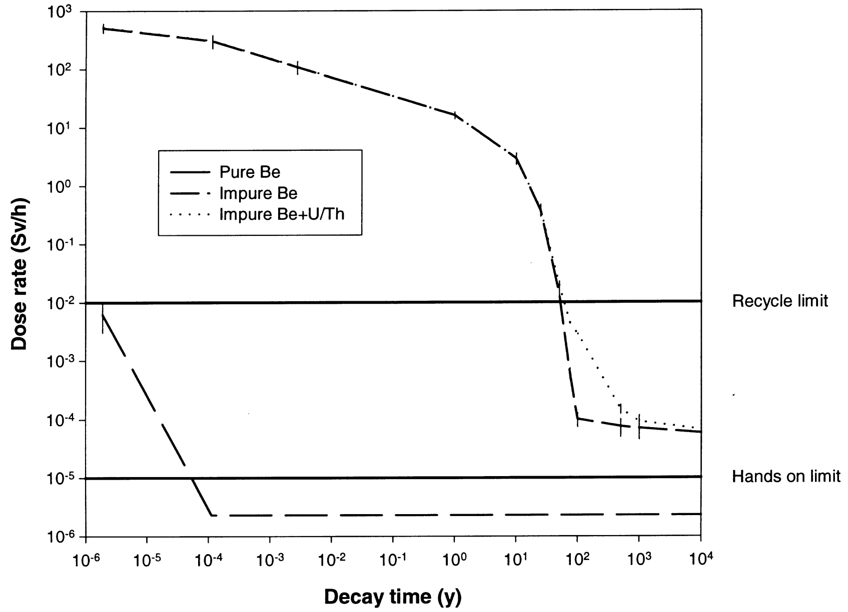

Fig. 1. Dose rate of pure, impure and impure with actinides beryllium at times following irradiation. The error bars show uncertainties in the dose rate due to cross-section and half-life uncertainties.

mation might be considered, within an acceptable decay time. It should, however, be feasible to process such scrap material under remote handling conditions, once the contact dose rate falls to about 10 mSv h^{-1}, namely after about 50 yr storage.The dominant nuclides after a decay time of 50 yr are indicated in Table 4. Since ^{60}Co governs the γ-dose rate up to about 100 yr, reductions in the concentrations of impurities leading to ^{60}Co production, notably Co, Ni and Fe, would reduce the storage time needed to attain the dose rate level at which recycling could be undertaken. The elemental composition of the irradiated impure material is indicated in Table 3 (column 3). Except for He, H and Li, the elements introduced by transmutation are present at very low levels, comparable with the concentrations of the impurities in the material prior to service. Since the impurities in the original material may be assumed to have already been reduced to the lowest practicable levels, it follows that it would be difficult to remove the transmutation products in any ordinary, non-specific purification process. Moreover, there appears to be no clear need to remove the majority of the transmutation products from material destined for reuse. As noted earlier, vacuum remelting for the production of new components would incidentally allow a high degree of separation of volatile impurities such as hydrogen, he-

Table 4
Dominant nuclides in impure Be at 50 yr after shutdown

Nuclide	Activity, %	Nuclide	γ-dose rate, %
^{3}H	100.0	^{60}Co	99.4
^{63}Ni	1.9×10^{-3}	^{26}Al	2.6×10^{-1}
^{10}Be	2.9×10^{-4}	^{59}Ni	1.6×10^{-1}
^{60}Co	1.9×10^{-4}	^{91}Nb	1.2×10^{-1}
^{14}C	2.6×10^{-5}	^{55}Fe	5.6×10^{-2}
^{59}Ni	2.1×10^{-5}	^{93}Mo	2.8×10^{-2}
^{55}Fe	1.1×10^{-5}	^{93m}Nb	4.8×10^{-3}
^{91}Nb	8.0×10^{-6}	^{94}Nb	3.2×10^{-3}
^{93}Mo	1.8×10^{-6}	^{42}K	2.3×10^{-3}
^{93m}Nb	1.6×10^{-6}	^{53}Mn	1.7×10^{-3}
^{26}Al	4.7×10^{-7}	^{98}Tc	9.9×10^{-6}
^{53}Mn	3.9×10^{-7}	^{22}Na	5.6×10^{-6}
Total activity	$(9.6 \pm 5.1) \times 10^{12}$ Bq kg^{-1}	Total γ-dose rate	$(1.4 \pm 0.3) \times 10^{-2}$ Sv h^{-1}

lium, lithium and manganese, without leading to the generation of significant quantities of secondary wastes.

The potential for a faster-than-linear accumulation of transmutation products and post-service activity and dose rate was investigated by simulating a second service irradiation cycle of the impure material following 50 yr of decay storage. The elemental composition at 50 yr after the second period of service is presented in Table 3 (column 4). It appears that most of the transmutations proceed in a more or less linear fashion and do not lead to any rapid increases in the concentrations of detrimental species.

5. Effect of actinide impurities

Whilst there appears to be little reason to expect significant concentrations of actinide impurities in beryllium, some trace element analyses have indicated the presence of uranium at around 1 ppm. Such results may, however, be unrepresentative and arise from contamination of the analytical equipment or other causes. Nevertheless, in order to examine the possible influence of traces of actinides on the activation characteristics of beryllium, the activation calculation was repeated for the impure beryllium composition with the addition of thorium and uranium at levels of 1 ppm by mass.

For this case the inventory included a large number of low-level fission products and actinides generated from fission reactions. The total activity was not appreciably affected but significant increases in the γ-dose rate and the ingestion and inhalation doses were apparent after decay times of about 100 yr, as can be seen from Figs. 1 and 2 for the γ-dose rate and the inhalation dose. The isotopes dominant at 100 yr are indicated in Table 5. The pathways responsible for producing some of the dominant nuclides can be complicated as can be seen below for ^{241}Am, but these pathways are routinely produced by FISPACT-97.

$$^{238}\mathrm{U}(\mathrm{n},\gamma)^{239}\mathrm{U}(\beta^-)^{239}\mathrm{Np}(\beta^-)^{239}\mathrm{Pu}(\mathrm{n},\gamma)^{240}\mathrm{Pu}(\mathrm{n},\gamma)^{241}\mathrm{Pu}(\beta^-)^{241}\mathrm{Am}\ (98\%)$$

It is clear that Th and U can act as potent impurities, with greatest effects at long decay times in excess of 100 yr, though they are unlikely to exert much influence at the low concentrations, well below 1 ppm, expected in beryllium.

6. Effect of sequential charged particle reactions

Although the FISPACT calculations included any products arising through sequential charged particle activation reactions, the latter were found not to exert

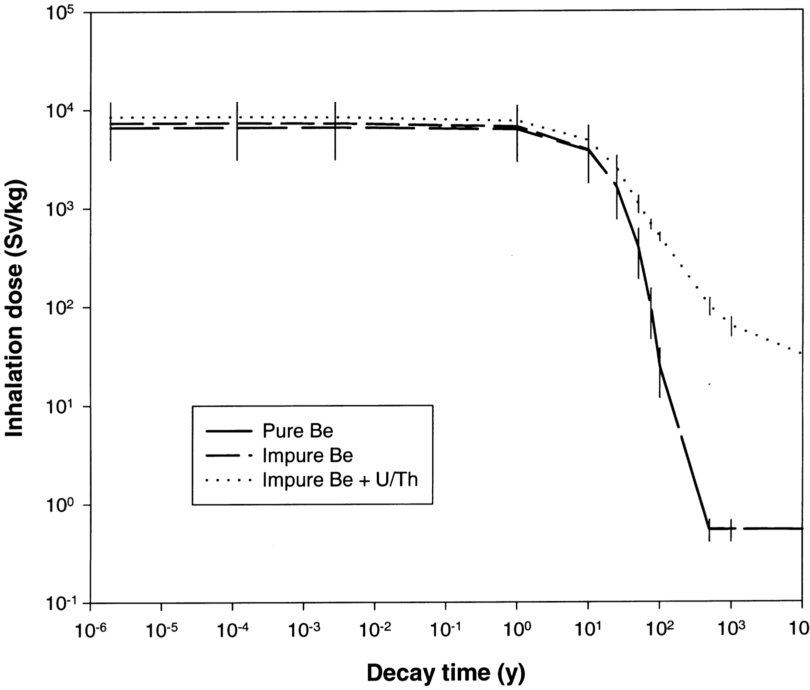

Fig. 2. Inhalation dose of pure, impure and impure with actinides beryllium at times following irradiation. The error bars show uncertainties in the inhalation dose due to cross-section and half-life uncertainties.

Table 5
Dominant nuclides at 100 yr for impure Be containing Th and U

Nuclide	Activity, %	Nuclide	Dose rate, %	Nuclide	Ingestion dose, %	Nuclide	Inhalation dose, %
^{3}H	100.0	^{208}Tl	61.3	^{3}H	86.6	^{228}Th	26.9
^{63}Ni	2.2×10^{-2}	^{137m}Ba	22.9	^{232}U	4.8	^{232}U	21.2
^{10}Be	4.9×10^{-3}	^{212}Bi	4.7	^{241}Am	2.5	^{241}Am	19.2
^{137}Cs	7.8×10^{-4}	^{212}Pb	3.8	^{238}Pu	2.4	^{238}Pu	17.3
^{137m}Ba	7.4×10^{-4}	^{241}Am	2.0	^{228}Th	1.1	^{3}H	4.7
^{212}Pb	7.3×10^{-4}	^{26}Al	1.3	^{224}Ra	1.0	^{227}Ac	4.2
Totals	5.8×10^{11} Bq kg^{-1}		2.8×10^{-3} Sv h^{-1}		2.8×10^{1} Sv kg^{-1}		5.0×10^{2} Sv kg^{-1}

any appreciable effect in any of the radiological responses of either pure or impure Be.

7. Conclusion

Fusion neutron irradiation of pure Be yields only ^{3}H and ^{10}Be as significant activation products and H, He and Li as transmutants. The dose rate associated with bremsstrahlung from the ^{10}Be is low enough to permit 'hands-on' reclamation and reuse of end-of-service beryllium in future plant. In practice, however, the dose rate of Be will be determined by activation products from impurities such as Co, Ni, Fe and Si. It should be possible to recycle used material following decay storage for about 50 yr, using remote handling and without the need to remove the bulk of the activation products. Sequential charged particle reactions make no appreciable contribution to the activation. At initial trace concentrations of 1 ppm, the actinides Th and U would significantly affect the γ-dose rate and the ingestion and inhalation doses of irradiated Be at decay times greater than 100 yr. Actinide elements are not expected to occur as significant impurities in nuclear-grade beryllium, however, and in practice should not pose any problems.

Acknowledgements

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