DERIVED ERLs FOR INHALATION OF ALPHA-EMITTING AEROSOLS

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Emergency dose limits recommended recently by the Medical Research Council (MRC) and improved dose-intake data for transuranic radionuclides enable limits for accidental exposure of the public to alpha-emitting fuel aerosols to be evaluated.

1. INTRODUCTION

Current arrangements at CEGB nuclear power stations for the initial assessment of inhalation hazards following a reactor accident leading to a release of radioactivity to the environment are based on the total β/γ count rates of air samples collected by mobile emergency survey teams (1). These measurements are made in the field and reported back to the emergency control centre by radio; they are followed up rapidly by detailed γ-spectrometric analyses of the air samples at the station district survey laboratory. In addition, the total α count rates of the air samples are measured at this time in order to eliminate the remote possibility of a significant α-hazard associated with actinide isotopes in the absence of a greater hazard due to the β/γ emitting fission products. A derived α action level, expressed in dpm/m^3, is used and represents the airborne concentration of an assumed mixture of α-emitters which if inhaled for a period of four hours would result in a dose commitment to the critical group in the exposed population no greater than the Emergency Reference Levels (ERLs) of dose recommended by the MRC (2). The period of four hours is selected as a realistic time within which to assess the severity of a release and initiate appropriate countermeasures.

2. INHALATION DOSES FROM THE TRANSURANICS

Dose-intake data for transuranic radionuclides inhaled as 1 μm AMAD particles have been given in two recent reports (2,3); these considered doses to selected human body organs due to ingestion and inhalation of ^{239}Pu and ^{242}Cm, based upon the latest ICRP lung and GI tract models for the transuranics (4). The results of similar calculations of the adult inhalation doses to lung and bone for a more complete range of actinide isotopes are presented in Table I (5); these data also assume inhalation of a 1 μm AMAD aerosol and lung clearance parameters appropriate to Class W (soluble) and Class Y (insoluble) compounds.

Isotope	Adult Dose per μCi Inhaled (rems/μCi)			
	Soluble (Class W)		Insoluble (Class Y)	
	Lung	Bone	Lung	Bone
^{238}Pu	30	1400	300	510
^{239}Pu	28	1540	280	590
^{240}Pu	28	1540	280	590
^{241}Pu	0.012	31	0.89	13
^{241}Am	30	1600	300	600
^{242}Cm	25	31	80	4.5
^{244}Cm	31	806	284	280

TABLE I Dose Intake Data for Transuranic Radionuclides (5)

Although the dose distribution will vary with the particle size, the results for a 1 μm AMAD aerosol are considered appropriate here since the variations are relatively small, being less than a factor of 2 for particles in the range 0.5-2 μm AMAD in the worst case of insoluble Class Y compounds (3). The bone doses given are for a skeletal mass of 7000 g, with a Quality Factor of 10 and a Distribution Factor of 5 in accordance with current ICRP practice, and the data of Table I are in good agreement with other recently reported actinide dose-intake data (2,3,6).

As the transuranics are deposited primarily on bone surfaces, rather than throughout the volume of mineral bone, the relevant tissues are the endosteal surfaces for which the ERL of dose is 30 rem, the same as that for the lung (2). An estimate of the endosteal dose was obtained by increasing the bone doses of Table I by a factor of 3; this is equivalent to the use of a Distribution Factor of 15 and yields endosteal doses consistent with recent estimates by Thorne and Vennart (7). Further, as young children might form the critical group in the population, appropriate rems/μCi inhaled were estimated from Table I; for the lung and ^{242}Cm irradiation of bone, where the dose is delivered over a few hundred days, the adult rems/μCi values were increased by a factor of 10 for the reduced mass of the child's organs. Combined with a breathing rate approximately one-third of that of an adult, this leads to a nett increase in the dose commitment by a factor of 3 for exposure of young children in the first year of life.

In the case of bone irradiation by the long lived actinides, where the dose is delivered essentially uniformly over the assumed 50 year integration period, the adult rems/μCi inhaled values were increased by only a factor of 3 to allow for the reduced skeletal mass of children during the early years post-intake. With the reduced breathing rate mentioned above, this leads to similar dose commitments for exposure of adults and young children.

3. EVALUATION OF DERIVED ERLs

Total α count rates equivalent to an ERL for Magnox and advanced gas-cooled reactor (AGR) fuels at zero and 1000 days cooling evaluated using the dose-intake data discussed above are presented in Table II. The fuel irradiation histories assumed were 4 GWd/T at 2.7 MW/T, and 18 MWd/T at 13 MW/T for Magnox and AGR respectively and the values quoted assume a four hour exposure as noted earlier. The total α values equivalent to an ERL tend to increase with fuel burn-up, or on progressing from Magnox to AGR, but exhibit a reverse trend with increasing cooling time of the irradiated fuels. These effects arise from the increased build-up and subsequent decay of the relatively less radiotoxic, shorter lived isotopes of curium.

Cooling Period (days)		Total α Count ≡ ERL (4 hour) dpm/m^3			
		Magnox		AGR	
		Adult	Child	Adult	Child
Bone Class W	0	1.4×10^4	1.3×10^4	2.1×10^4	1.7×10^4
	10^3	2.9×10^3	2.9×10^3	3.0×10^3	3.0×10^3
Lung Class Y	0	1.7×10^5	5.0×10^4	1.9×10^5	5.6×10^4
	10^3	6.6×10^4	2.0×10^4	6.5×10^4	2.0×10^4
Bone Class Y	0	3.7×10^4	3.6×10^4	5.6×10^4	5.2×10^4
	10^3	7.1×10^3	7.1×10^3	7.6×10^3	7.6×10^3

TABLE II Total α Count Rates Equivalent to an ERL for Irradiated Reactor Fuels

For completeness, total α count rates equivalent to an ERL for unirradiated natural and enriched uranium fuels were evaluated; here the situation is complicated by the chemical toxicity of uranium which limits its intake in a soluble form by inhalation to 2.5 mg/day(8). Assuming a four hour exposure, this gives total α ERLS of 1.2×10^3 and 2.7×10^3 dpm/m^3 for natural uranium and 2.5% ^{235}U enriched fuels respectively. For inhalation of insoluble material the lung dose is limiting, due to the much shorter biological half-life of uranium in bone compared with the higher actinides, and the corresponding α ERL for a four hour exposure is 2.4×10^4 dpm/m^3.

The data of Table II show that the limiting α ERLs for irradiated fuels are set by the inhalation doses to bone from long-cooled fuel particulates for both soluble (Class W) and insoluble (Class Y) materials. In the latter case, the ERL based on the dose to a child's lung is a factor of 2-3 times less restrictive than that set by the bone dose; this somewhat unusual result for insoluble material arises from the extremely long radioactive and biological half lives of the plutonium isotopes involved. These are principally ^{241}Pu and its ^{241}Am daughter, which account for $\sim$ 50% of the bone dose for the fuels considered here; ^{239}, ^{240}Pu, and for the higher burn-up AGR fuel the ^{238}Pu daughter of ^{242}Cm, are the other main contributors to the bone doses.

In contrast, for the unirradiated fuels there is more than an order of magnitude difference between the derived ERLs for material inhaled in soluble and insoluble forms. This is due to the relatively low specific activity of unirradiated low enrichment uranium fuels and the consequent limitations imposed by chemical toxicity.

Although any release of actinides in a reactor accident is likely to occur in an insoluble form in the chemical sense, this does not necessarily imply that they will behave as insolubles in the ICRP sense. Current radiobiological evidence on the transportability of inhaled transuranics, notably isotopes of plutonium, suggests that in some situations they can exhibit anomalously high solubility, particularly if inhaled as micron or sub-micron aerosols (9). In view of this uncertainty it is suggested that an emergency action level should be based on the soluble bone dose data of Table II; bearing in mind the possible equal importance of the dose to liver noted by MRC (2,3), and that the ERLs of dose refer to individual organs irradiated separately, this leads to a derived α ERL of $\sim 1.5 \times 10^3$ dpm/m^3 for a four hour exposure. This is only marginally greater than the value of 1.2×10^3 dpm/m^3 for unirradiated natural uranium fuel discussed earlier.

4. CONCLUSION

The occurrence of a significant airborne α-hazard due to the actinide isotopes, in the absence of a more limiting hazard associated with the β/γ emitting fission products, is considered unlikely in most postulated reactor accident situations. The effects of the actinide content of fuel are only likely to be important in specialised circumstances, such as incidents resulting in the generation of fuel aerosols without preferential release of the more volatile fission product isotopes. Based upon the total α levels equivalent to an ERL presented here, an appropriate derived α ERL is 10^3 dpm/m^3, (170 Bq m^{-3}) for a four hour exposure period; this value is equally applicable to incidents involving both unirradiated and irradiated reactor fuels.

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