

TYPE A PACKAGE LIMITS OF SPONTANEOUS FISSION RADIONUCLIDES

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TYPE A PACKAGE LIMITS OF SPONTANEOUS FISSION RADIONUCLIDES

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INTRODUCTION

Limits on the activity allowed in Type A packages in transport are prescribed in the International Atomic Energy Agency (IAEA) "Regulations for the Safe Transport of Radioactive Material," Safety Standards Series No. TS-R-1 (IAEA 2000a). Type A packages are designed to withstand normal conditions of transport (including minor mishaps) and are intended to provide economical transport for limited-activity packages while achieving an appropriate level of safety. Limits on the content of the packages ensure that the radiological consequences of severely damaged packages are not unacceptable. The numerical value of the content limits are based on the Q-System initially developed by Macdonald and Goldfinch (1980) and further developed by Bologna, Eckerman, and Hughes (IAEA 2000b).

The Q-System consists of a series of scenarios postulating pathways leading to radiation exposure, either external or internal, to persons in the vicinity of a damaged Type A package. The system leads to five constraints— Q_A , Q_B , Q_C , Q_D , and Q_E —on the package contents to limit potential doses. These constraints are based on doses resulting from

- irradiation by photon emissions from the package contents— Q_A ,
- irradiation by beta emissions from the package contents— Q_B ,
- inhalation of dispersed contents in the air— Q_C ,
- contamination of the person by the dispersed contents— Q_D , and
- submersion within the dispersed airborne activity of noble gases— Q_E .

In addition, the system uses pragmatic constraints for alpha emitters and to account for bremsstrahlung radiation. Only a few radionuclides undergo spontaneous fission, and they are subject to special considerations; however, the Q_A scenario is applicable to their neutron and photon emissions.

The Q-System addresses potential exposures to the contents of a damaged Type A package. In the current regulations, the reference effective dose used to constrain the exposure is 50 mSv. The duration of the exposure time is taken as 0.5 h at a distance of 1 m from the damaged package. The activity content for special form (non-dispersible) material, the A_1 value, is the lesser value of Q_A and Q_B , while the A_2 values for non-special form (dispersible) material is the least of the five Q values. For further details see IAEA 2000b.

DOSIMETRIC DATA FOR SPONTANEOUS FISSION RADIONUCLIDES

The three nuclides in the Q-System for which spontaneous fission is of significance are ^{248}Cm , ^{252}Cf , and ^{254}Cf . The characteristics of these radionuclides are summarized in Table 1. In setting the A_1 values for these nuclides in the 1996 edition of the IAEA transport regulations, their earlier values were decreased by about a factor of two based on the ICRP's recommended increase in the radiation weighting factor for neutrons (ICRP 1991). The earlier value was based on an external dose rate coefficient for ^{252}Cf given in ICRP Publication 21 (ICRP 1973). That coefficient represented the maximum in the depth-dose curve for normally incident neutrons on a cylindrical phantom, and it was applied to ^{248}Cm and ^{254}Cf with allowance for their respective neutron emissions. For all radionuclides other than the three neutron emitters, the derivation of Q_A was based on the effective dose as defined in the 1990 recommendation of the ICRP (1991); that is, the effective dose based on organ doses evaluated using an anthropomorphic phantom.

Publication 74 (ICRP 1996) provides data on the effective dose per unit neutron fluence for monoenergetic neutrons incident on an adult anthropomorphic phantom for various exposure geometries. Figure 1 compares the maxima neutron dose per unit fluence of ICRP 21 with the effective dose per unit fluence tabulated in ICRP 74 (1996). In Fig. 1, it can be seen that at most neutron energies, the maxima dose coefficient overestimates the effective dose coefficient without any adjustment for an increase in the neutron radiation weighting factor.

Effective dose rate coefficients are derived here for ^{248}Cm , ^{252}Cf and ^{254}Cf using information on the ^{252}Cf neutron spectra in ENDF/B-VI files (Rose and Dunford 1991) and the effective dose per unit neutron fluence for rotational exposure geometry tabulated in ICRP Publication 74 (1996). The effective dose rate coefficient at a distance r , \dot{e}_n , from a bare neutron source is given by

$$\dot{e}_n = \frac{\nu f}{4\pi r^2} \int_0^{\infty} Y_N(E) r(E) dE \quad (1)$$

where ν is the number of neutrons emitted per fission, f the fission per nuclear transformation, $Y_N(E)$ the probability of a fission neutron with energy between E and $E+dE$ (see Fig. 1), and $r(E)$ the effective dose to fluence coefficient as a function of energy from ICRP Publication 74 (1996). The calculations yield a value for fission neutrons of 1.54, 0.686, and 23.1 Sv TBq⁻¹ h⁻¹ for ^{248}Cm , ^{252}Cf , and ^{254}Cf , respectively, as listed in Table 2. The values for ^{248}Cm and ^{254}Cf assume the ^{252}Cf neutron spectrum $Y_N(E)$.

Spontaneous fission is accompanied by the emission of photons. Photons emitted within the first 60 ns after the fission are referred to as prompt-fission photons, and photons associated with the de-excitation of the fission products as the delayed-fission photons. Dillman and Jones (1975) adopted the prompt photon spectral measurements of Peelle and Maienschein (1970) for ^{235}U as representative of fissioning radionuclides. Prompt photons range in energy from 0.1 to about 10 MeV with the total energy carried by these photons being about 7.6 MeV per fission. Similarly, Dillman and Jones adopted a delayed photon spectrum based on the work of Zigman and Mackin (1961) and Bunney and Sam (1970). Dillman and Jones, however, noted that the total energy of

the delayed photons varies from one nuclide to another, and they provide an empirical factor to modify the delayed photon dose based on the ^{236}U spectral measurements. This factor is tabulated in Table 1. The delayed photon emission of ^{236}U results in a total energy release of 7.2 MeV. Dillman and Jones approximate the prompt and delayed photon emissions by a series of discrete lines, and thus the effective dose rate coefficient at 1 m for the prompt gamma emission, $\dot{e}_{p,\gamma}$, is given by

$$\dot{e}_{p,\gamma} = \frac{1}{4\pi r^2} \sum_{i=1}^n y_p(E_i) r(E_i) \quad (2)$$

where $y_p(E)$ is the prompt photon spectrum and $r(E_i)$ is the effective dose per unit fluence for monoenergetic photons incident on an adult anthropomorphic phantom. These values are based on ICRP Publication 74. The corresponding equation applicable to the delayed photon emission is

$$\dot{e}_{d,\gamma} = \frac{R_d}{4\pi r^2} \sum_{i=1}^n y_d(E_i) r(E_i) \quad (3)$$

where R_d is the adjustment for the total photon energy associated with the delayed photon emissions (see Table 1) and $y_d(E)$ is the prompt gamma photon spectrum. The effective dose rate coefficients at 1 m for the fission gamma photons are given in Table 2.

The spontaneous fission nuclides considered here also undergo alpha decay as indicated in Table 1. The energy and intensity of discrete photon emission associated with the alpha decay are tabulated in ICRP Publication 38 (1983). The effective dose coefficients associated with the alpha decay are given in Table 2 as derived in the manner of Eq. 2.

Note that the derivation of dose rate coefficients does not include attenuation and buildup of the fluence because, at a distance of 1 m, these processes have a negligible influence on the dose rate (i.e., less than 10%). The total effective dose rate coefficient due to the neutron and photon emissions is 1.68, 0.747, and 24.8 Sv TBq⁻¹ h⁻¹ for ^{248}Cm , ^{252}Cf , and ^{254}Cf , respectively, as listed in the bottom row of Table 2.

Updated A_1 Value

The value of Q_A and hence A_1 constrained by the reference effective dose E_R acquired during the exposure period of length t is

$$A_1 = Q_A = \frac{E_R}{\dot{e} t} \quad (4)$$

where \dot{e} is the effective dose rate coefficient applicable to the Q_A scenario. Table 3 lists the A_1

values derived in this work and the corresponding earlier values. The A_2 values for these radionuclides remain unchanged as they are based on the potential inhalation of airborne materials. The A_1 values derived here are 3, 2, and 4 times the values in the 1996 edition of the IAEA transport regulations for ^{248}Cm , ^{252}Cf , and ^{254}Cf , respectively.

CONCLUSIONS

The maxima value of the depth dose coefficient for fission neutrons in ICRP Publication 21 was a reasonable estimator of the effective dose coefficient recently tabulated in ICRP Publication 74. Thus the inflation of the coefficient in the 1996 Q-System analysis (IAEA 2000b) for the purpose of being consistent with respect to ICRP guidance on the neutron weighting factor was unnecessary from the standpoint of the effective dose. The consequence resulted in an unnecessarily restrictive value of A_1 for ^{248}Cm , ^{252}Cf , and ^{254}Cf . The calculations presented here support a relaxation of the A_1 limits for these radionuclides.

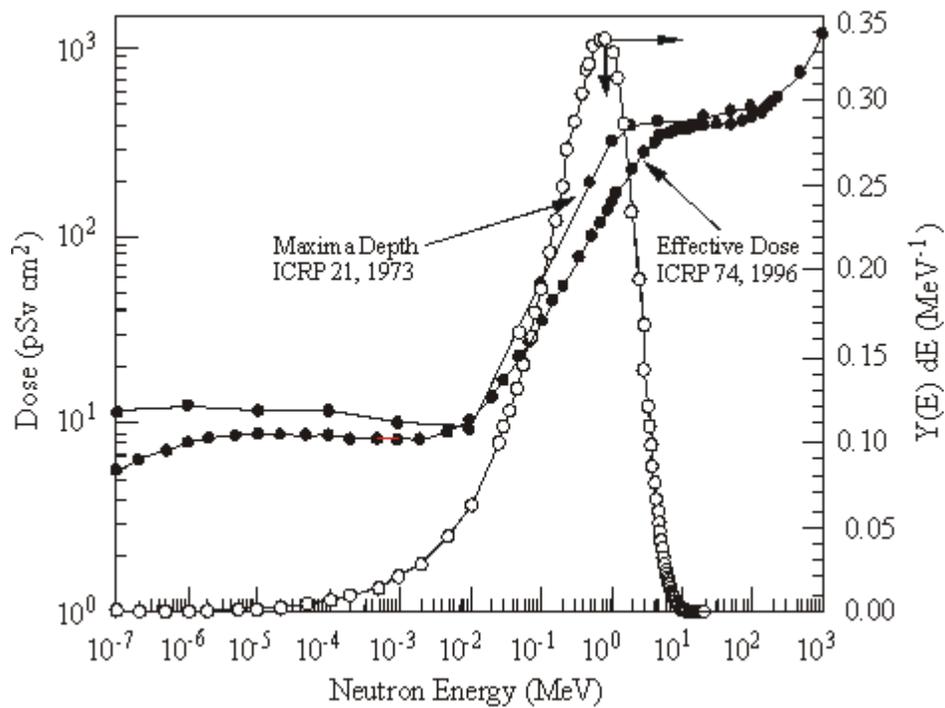


Fig. 1. The maxima neutron dose to fluence conversion of ICRP Publication 21 (1973), which was used in the 1996 edition of the Q-System, and the effective dose to fluence conversion from ICRP Publication 74 (1996.) The Publication 21 data are for normally incident neutrons on a cylindrical phantom, and Publication 74 data are for a rotational exposure geometry of an anthropomorphic phantom. The normalized distribution in energy of neutrons emitted by ^{252}Cf (from ENDF/B) is shown by the open circle data points.

Table 1. Physical Properties of Spontaneous Fission Radionuclides^{a)}

Property	²⁴⁸ Cm	²⁵² Cf	²⁵⁴ Cf
Half-life	3.39 10 ⁵ y	2.638 y	60.5 d
Specific activity (TBq g ⁻¹)	1.58 10 ⁻⁴	19.9	314
Fission per decay (%)	8.26	3.09	99.7
Neutron per fission	3.14	3.73	3.89
Number of α per fission	11	31	0.0031
Neutron per (g s)	4.1 10 ⁷	2.3 10 ¹²	1.2 10 ¹⁵
Relative energy of delayed photons ^{b)}	1.23	1.08	1.36

^{a)} Based on Shultis and Faw (1996).

^{b)} Relative to delayed gamma photon emissions of ²³⁶U (total 7.2 MeV per fission), based on Eq. 15 of Dillman and Jones (1975).

Table 2. Effective Dose Rate Coefficient at 1 m From a Bare Source

Radiations	Effective Dose Rate (Sv h ⁻¹ TBq ⁻¹)		
	²⁴⁸ Cm	²⁵² Cf	²⁵⁴ Cf
Fission neutrons	1.54	0.686	23.1
Fission prompt photons	0.0627	0.0235	0.757
Fission delayed photons	0.0726	0.0237	0.968
α -decay photons	0.00398	0.00341	< 0.0001
Total	1.68	0.747	24.8

Table 3. Type A Package Content Limits: Values for special form (A_1) and non-special form (A_2)

	1996 edition ^{a)}		This work ^{b)}	
	A_1 (TBq)	A_2 (TBq)	A_1 (TBq)	A_2 (TBq)
^{248}Cm	0.02	$3 \cdot 10^{-4}$	0.06	$3 \cdot 10^{-4}$
^{252}Cf	0.05	0.003	0.1	0.003
^{254}Cf	0.001	0.001	0.004	0.001

^{a)} IAEA Safety Standards Series No. TS-R-1 (IAEA 2000a).

^{b)} A_2 values are based on Q_C (inhalation pathway) and thus remained unchanged.

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