Critical Mass Calculations for ^{241}Am, ^{242m}Am and ^{243}Am

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Critical mass calculations are reported for ^{241}Am, ^{242m}Am and ^{243}Am using the MONK and MCNP computer codes with the UKNDL, JEF-2.2, ENDF/B-VI and JENDL-3.2 nuclear data libraries. Results are reported for spheres of americium metal and dioxide in bare, water reflected and steel reflected systems. Comparison of results led to the identification of a serious inconsistency in the ^{241}Am ENDF/B-VI DICE library used by MONK - this demonstrates the importance of using different codes to verify critical mass calculations. The ^{241}Am critical mass estimates obtained using UKNDL and ENDF/B-VI show good agreement with experimentally inferred data, whilst both JEF-2.2 and JENDL-3.2 produce higher estimates of critical mass. The computed critical mass estimates for ^{242m}Am obtained using ENDF/B-VI are lower than the results produced using the other nuclear data libraries – the ENDF/B-VI fission cross-section for ^{242m}Am is significantly higher than the other evaluations in the fast region and is not supported by recent experimental data. There is wide variation in the computed ^{243}Am critical mass estimates suggesting that there is still considerable uncertainty in the ^{243}Am nuclear data.

KEYWORDS: critical mass, americium, MONK, MCNP, UKNDL, JEF-2.2, ENDF/B-VI, JENDL-3.2

1. Introduction

This paper reports critical mass calculations for ^{241}Am, ^{242m}Am and ^{243}Am using the MCNP and MONK computer codes with various nuclear data libraries.[1,2] MCNP was developed at Los Alamos National Laboratory, New Mexico (USA) – this code is widely used in the field of criticality safety. MONK is the acknowledged standard criticality code in the UK.

The latest version of MONK – MONK8b – is supplied with the UKNDL, JEF-2.2, ENDF/B-VI and JENDL-3.2 continuous energy libraries.[3-6] For comparison, calculations were performed with MCNP using the JEF-2.2, ENDF/B-VI and JENDL-3.2 point-wise nuclear data sets. The calculational methodology in both MONK and MCNP is based on the continuous energy approach. Results are reported for spheres of americium metal and dioxide in bare, water reflected and steel reflected systems. This work is part of the UK contribution to the Working Group for the revision of American National Standard 8.15 – "Nuclear Criticality Control of Special Actinide Elements".[7]

2. Historical Review of MONK

MONK replaced the GEM computer code, which was developed during the 1960's at the United Kingdom Atomic Energy Agency (UKAEA) Health and Safety Branch at Risley in support of the UK atomic weapons programme. MONK5, which was developed in the late 1970's, combined the best features of the previous Aldermaston and Risley versions of the code.[8] MONK6 was developed during the early 1980's – a key feature of MONK6 was the incorporation of the DICE collision processing package. The original version of DICE was developed at Aldermaston.[9] Historically, the DICE nuclear database was derived from the UKNDL and all the early versions of the MONK criticality code utilised only this nuclear data set.

More recently, new development work on MONK has been co-ordinated under the auspices of the NCD (Nuclear Code Development) collaboration – the formal members of this collaboration are Serco Assurance and British Nuclear Fuels plc.[10] MONK7 and MONK8 developments have been described elsewhere.[11,12] The NJOY nuclear data processing system was used, *inter alia*, in the generation of the additional JEF-2.2, ENDF/B-VI and JENDL-3.2 DICE libraries issued with MONK8.[13]

3. Nuclear Data Libraries

3.1 UKNDL

The first UKNDL collation of data was produced at Aldermaston in the early 1960's.[3] Subsequent evaluations and revisions were conducted within the UKAEA until the library was frozen in the early 1980's – no maintenance or updates to UKNDL have been made since then. In contrast to more modern

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data evaluations, the UKNDL is "adjusted"; i.e. certain nuclear data were adjusted, based on experimental evidence, to ensure that computer calculations show good agreement (or are pessimistic) when compared with experimental data.

Although only the ^{241}Am evaluation was formally published, the last UKNDL americium evaluations were all produced at Harwell.[14] Details of the ^{243}Am neutron data evaluation, parts of which were adopted in JEF-2.2, can be downloaded from the Los Alamos T-2 Nuclear Information Service.[15]

3.2 JEF-2.2

The JEF-2.2 nuclear data library is described in JEFF Report 17.[4] The JEF-2.2 DICE library was frozen in 1996 to allow benchmarking studies. More specific details of the JEF-2.2 americium evaluations can be downloaded from the T-2 Nuclear Information Service.[15]

3.3 ENDF/B-VI

The ENDF/B-VI data evaluations have been periodically up-dated – the latest version available from the NEA is release 8. The T-2 Nuclear Information Service provides full details of the various evaluations in ENDF/B-VI.[15]

The CENDL-2 evaluation of the neutron nuclear data for ^{241}Am was adopted in release 2.[16] This evaluation was revised for issue as release 3 by Young and Madland.[15] The ^{241}Am evaluation is unchanged since ENDF/B-VIr3.

Apart from the addition of the delayed fission neutron spectrum in release 1, the ENDF/B-VI ^{242m}Am evaluation is still unchanged from ENDF/B-V.[15,17]

The ENDF/B-V ^{243}Am evaluation was due to Mann *et al.*[18] The cross section data were updated for ENDF/B-VIr1 by Weston.[15] In 1996 a new evaluation of the neutron data for ^{243}Am was issued as ENDF/B-VIr5 – this was prepared by Young and Weston.[15] The ^{243}Am evaluation is unchanged since ENDF/B-VIr5.

3.4 JENDL-3.2

The ^{241}Am and ^{243}Am data for JENDL-2 were evaluated by Kikuchi.[19] The ^{242m}Am evaluation for JENDL-2 was by Nakagawa and Igarasi.[20] These data were revised for JENDL-3.2 by Nakagawa.[21]

4. Results

Tables 1-3 give the computed minimum critical masses of ^{241}Am, ^{242m}Am and ^{243}Am, respectively. Results are reported for spheres of full density americium metal and dioxide in bare, water reflected (30cm) and 304 stainless steel reflected (20cm) systems. The densities of americium metal and dioxide are based on the compilation by Haire.[22]

Note that the ENDF/B-VI DICE library issued with MONK is release 4.[2] Consequently, the MONK ENDF/B-VI DICE library should contain the most recent evaluations for ^{241}Am and ^{242m}Am – but not the latest (release 5) evaluation for ^{243}Am (see Section 3.3).

The MCNP calculations for ^{241}Am and ^{242m}Am used the latest version of ENDF/B-VI available from the NEA – release 8. So as to facilitate comparison with the MONK results, the results presented in Table 3 were obtained using the ENDF/B-VIr1 ^{243}Am evaluation. The ^{243}Am results obtained using ENDF/B-VIr8 (i.e. including the latest evaluation for ^{243}Am – release 5) are shown in parentheses.

5. Discussion

The results presented in Table 1 show significant differences between the MONK and MCNP calculations for ^{241}Am using ENDF/B-VI. Other workers have reported minimum critical mass calculations for bare ^{241}Am metal - a value of 57.01kg is quoted using ENDF/B-VIr6.[23] This reported value of the critical mass compares favourably with the MCNP calculation, but is inconsistent with the MONK result given in Table 1. This suggests that the MONK results for ^{241}Am using ENDF/B-VI may be in error.

Table 1 Computed Minimum Critical Mass of ^{241}Am

Chemical form	Reflector	Critical mass (kg)						
		MONK				MCNP		
		UKNDL	JEF-2.2	ENDF/B-VI	JENDL-3.2	JEF-2.2	ENDF/B-VI	JENDL-3.2
Metal	Bare	56.4	75.7	88.0*	76.1	73.3	57.7	73.7
ρ = 13.66	Water	50.9	68.3	79.9*	69.2	65.8	52.0	66.7
g/cm³	Steel	33.6	42.4	51.8*	45.4	40.9	33.8	43.6
Dioxide	Bare	98.2	132	161*	135	129	94.6	131
ρ = 11.69	Water	92.0	124	151*	125	120	87.6	123
g/cm³	Steel	65.4	86.0	109*	91.4	83.9	62.4	89.0

*MONK results for ENDF/B-VI are subject to error – see text.

Table 2 Computed Minimum Critical Mass of ^{242m}Am

Chemical form	Reflector	Critical mass (kg)						
		MONK				MCNP		
		UKNDL	JEF-2.2	ENDF/B-VI	JENDL-3.2	JEF-2.2	ENDF/B-VI	JENDL-3.2
Metal	Bare	13.0	13.9	8.96	12.3	14.2	9.06	12.5
ρ = 13.72	Water	4.59	4.69	3.23	4.38	4.78	3.25	4.45
g/cm^3	Steel	5.21	5.11	3.73	4.97	5.21	3.74	5.02
Dioxide	Bare	14.1	14.8	9.96	13.4	15.0	10.0	13.6
ρ = 11.73	Water	5.23	5.34	3.74	5.04	5.39	3.74	5.08
g/cm^3	Steel	6.04	5.86	4.42	5.79	6.03	4.44	5.85

Table 3 Computed Minimum Critical Mass of ^{243}Am

Chemical form	Reflector	Critical mass (kg)						
		MONK				MCNP		
		UKNDL	JEF-2.2	ENDF/B-VI	JENDL-3.2	JEF-2.2	ENDF/B-VI	JENDL-3.2
Metal	Bare	181	217	222	296	206	211 (143)*	284
ρ = 13.77	Water	165	200	205	277	189	194 (132)*	262
g/cm^3	Steel	111	133	144	193	127	135 (89.0)*	181
Dioxide	Bare	473	616	618	926	578	572 (300)*	864
ρ = 11.77	Water	450	593	599	876	558	553 (282)*	822
g/cm^3	Steel	342	456	467	698	429	438 (215)*	662

*Results in parentheses are for the ENDF/B-VIr8 evaluation – see text.

Following discussions with Serco Assurance, it is apparent that difficulties were encountered during the creation of the ENDF/B-VI DICE library for ^{241}Am.[24] The ENDF/B-VIr3 ^{241}Am evaluation utilised the Madland-Nix fission spectrum representation.[25] The version of NJOY then available at Serco Assurance was unable to deal with this representation of the fission spectrum. [The Madland-Nix fission spectrum representation is not normalised to unity – the spectrum has to be re-normalised prior to processing into a form suitable for Monte Carlo analysis.[24]]

Nuclear data sets may be readily compared using the JANIS display program.[26] The ^{241}Am fission neutron spectrum was abstracted from the ENDF/B-VI DICE library in a form compatible with JANIS.[24] Note that the fission spectrum held in DICE is simulated by a number of equiprobable ranges i.e. the energy distribution is approximated by a histogram with intervals chosen so that there is an equal area under the histogram in each interval.[9]

The ^{241}Am fission neutron spectra from ENDF/B-VI DICE, ENDF/B-VIr2, ENDF/B-VIr3 and JEF-2.2 were compared using JANIS - see Fig. 1. The ENDF/B-VI DICE fission spectrum for ^{241}Am is a good representation of the JEF-2.2 evaluation, but is significantly different from the ENDF/B-VIr2 and ENDF/B-VIr3 evaluations – see Fig. 1. Based upon the evidence presented in Fig. 1, it is clear that the JEF-2.2 fission spectrum for ^{241}Am has been substituted for the ENDF/B-VIr3 evaluation in the ENDF/B-VI DICE library. The ENDF/B-VI DICE library for ^{241}Am is subject to error and is therefore unsuitable for critical mass calculations – the MONK results for ENDF/B-VI presented in Table 1 will not be considered further in this analysis.

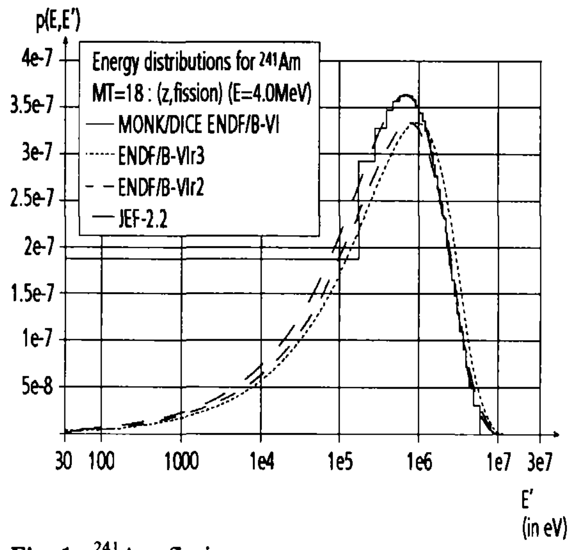

Fig. 1 ^{241}Am fission neutron spectra

The ^{241}Am minimum critical mass calculations fall into two groups – the higher critical mass estimates are obtained using JEF-2.2 and JENDL-3.2, whilst UKNDL and ENDF/B-VI yield lower critical mass estimates (Table 1). The JEF-2.2 results for ^{241}Am metal in bare, water reflected and steel reflected systems show good agreement with calculations reported by Duluc and Anno.[27] The JENDL-3.2 results for ^{241}Am metal and dioxide in bare and water reflected systems compare favourably with the data reported by Nojiri and Fukasaku.[28]

Critical masses of ^{241}Am have been inferred from reactivity coefficient measurements in fast critical assemblies.[29,30] The inferred critical mass estimates are as follows: 58kg – bare system; 51kg – water reflected; and 34kg – steel reflected. The computed results obtained using UKNDL and ENDF/B-VI are in excellent agreement with the experimentally inferred data – Table 1. Note that the UKNDL nuclear data may have been "adjusted" to ensure agreement with these experimental data (see Section 3.1).

There are differences between the results presented in Table 1 and ^{241}Am bare critical mass calculations reported by Brewer *et al.*[31] However, these workers assumed a density of 11.7g/cm^3 in their study which seems to correspond to that of AmO_2, rather than americium metal.

The ^{242m}Am calculations with the UKNDL, JEF-2.2 and JENDL-3.2 nuclear data libraries compare reasonably well, although the JEF-2.2 and JENDL-3.2 critical mass estimates obtained using MONK are slightly lower than those computed using MCNP – Table 2. The JENDL-3.2 results for ^{242m}Am metal in bare, water reflected and steel reflected systems show good agreement with the data reported by Okuno and Kawasaki.[32] There are discrepancies between the results given in Table 2 and the data reported by Duluc and Anno.[27] In particular, these authors claim that the critical mass of water reflected ^{242m}Am metal obtained using JEF-2.2 is ~6.5kg – this critical mass estimate is inconsistent with the results presented in Table 2 and should be treated with caution.

The ENDF/B-VI ^{242m}Am critical mass calculations are significantly lower than the results obtained using the other nuclear data libraries – Table 2. For example, calculations using UKNDL, JEF-2.2 and JENDL-3.2 give bare ^{242m}Am minimum critical mass estimates in the range 12.3 – 14.2kg, whereas the bare ^{242m}Am critical mass obtained using ENDF/B-VI is ~ 9kg. The ENDF/B-VI ^{242m}Am fission cross section is significantly higher than the UKNDL, JEF-2.2 and JENDL-3.2 evaluations in the fast energy region (by ~ 50% at 1MeV) and is not supported by recent experimental data.[33,34] The ^{242m}Am critical mass estimates calculated using ENDF/B-VI should therefore be considered spurious.

There is wide variation in the computed minimum critical mass estimates for ^{243}Am – Table 3. Calculations using the JEF-2.2 and ENDF/B-VIr1 data libraries compare favourably, whilst the UKNDL gives lower estimates of the ^{243}Am critical mass. The largest estimates of ^{243}Am critical mass are obtained using JENDL-3.2. The JEF-2.2 results for ^{243}Am metal in bare, water reflected and steel reflected systems compare favourably with computations reported by Duluc and Anno.[27] The JENDL-3.2 results for ^{243}Am metal and dioxide in bare and water reflected systems are in reasonable agreement with the data reported by Nojiri and Fukasaku.[28]

There are large differences between the ^{243}Am critical mass results obtained using the ENDF/B-VIr1 evaluation and those obtained using ENDF/B-VIr8 – Table 3. For ^{243}Am metal the ENDF/B-VIr8 results are ~33% lower than the ENDF/B-VIr1 calculations. For ^{243}Am dioxide the ENDF/B-VIr8 results are ~50% lower than those obtained using ENDF/B-VIr1. The results presented in Table 3 suggest that the nuclear data for ^{243}Am are still subject to considerable uncertainty.

As noted in Section 2, the DICE nuclear database was originally derived from the UKNDL. Secondary neutron energy data were defined for different incident neutron energy ranges. In more modern nuclear data libraries, the secondary data are commonly defined at a series of incident energy points – data for intermediate incident neutron energies are then obtained by linear interpolation. Interestingly, although MONK is now issued with JEF-2.2, ENDF/B-VI and JENDL-3.2 DICE libraries, DICE has not yet been updated to perform this interpolation.[24] For most major actinides any effect will be insignificant because the secondary data are well characterised. For some minor actinides the secondary data are occasionally more coarsely defined and errors may be introduced if the current DICE representation of the secondary data is used.

For example, the ENDF/B-VI ^{242m}Am fission spectrum is defined at four incident energies: 1×10^{-5}eV, 4MeV, 7MeV and 20MeV. The current version of DICE uses the thermal fission spectrum up to 4MeV, the 4MeV spectrum up to 7MeV, and the 7MeV spectrum up to 15MeV. [The DICE energy grid does not extend beyond 15MeV.[9]] The ^{242m}Am fission spectra were abstracted from the ENDF/B-VI DICE library in a form compatible with JANIS.[24] The JANIS display program was then used to compare the fission spectra at these incident neutron energies – see Fig. 2. Close examination reveals only barely discernible variations in the fission spectra (Fig. 2). All the other americium evaluations considered herein are either well characterised or show barely discernible variations in the fission spectra. It follows that the DICE

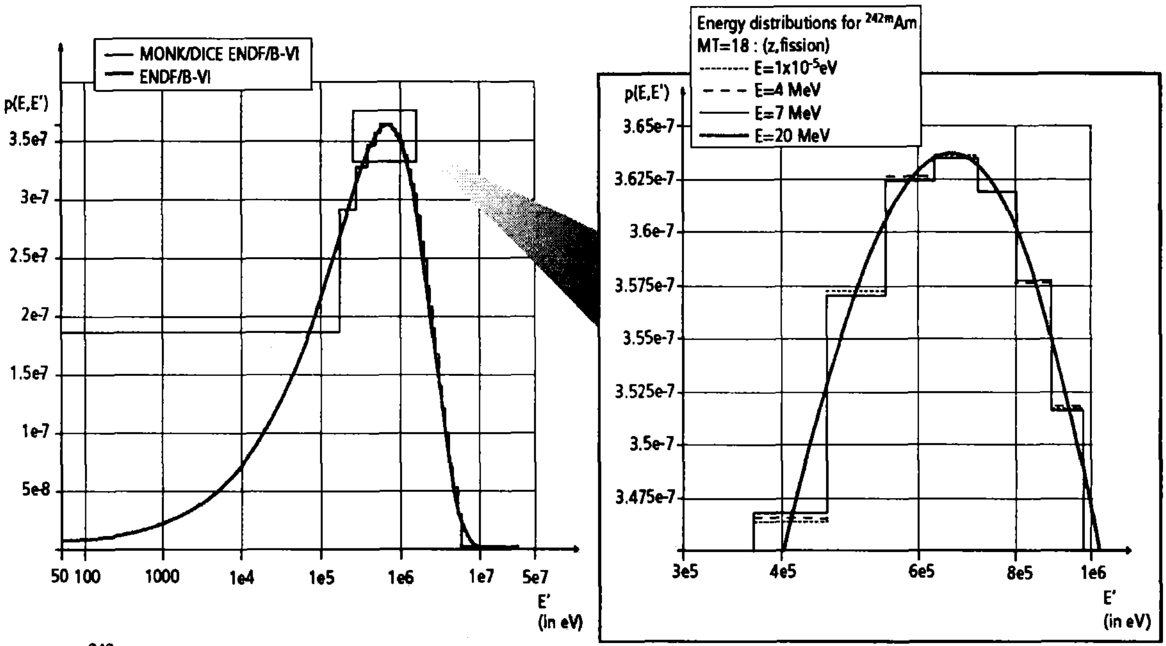

Fig. 2 ^{242m}Am fission neutron spectra

representation of secondary data is unlikely to introduce any major source of error in the critical mass calculations reported herein.

6. Conclusion

Minimum critical mass calculations are reported for ^{241}Am, ^{242m}Am and ^{243}Am using the MONK and MCNP computer codes with the UKNDL, JEF-2.2, ENDF/B-VI and JENDL-3.2 nuclear data libraries. Results are reported for spheres of americium metal and dioxide in bare, water reflected and stainless steel reflected systems.

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TOPIC 7

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