Americium 241
Criticality Safety Studies

Work Package 5,
October 2014 Progress Report
(for EGIEMAM-II)

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A Reminder of the Am241 Problem

Outline Solution

and

Future works

Industry, Stakeholder engagement and funding to October 2014 and beyond

THE Am241 PROBLEM

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NNL to separate kg quantities of Am241 from Magnox PuO_2

NNL Application:

heat/energy source as a Radioisotope Thermoelectric Generator (RTG) for space application (Space Battery)

Am241 market (other)

THE Am241 PROBLEM

Where present IN ISOLATION in sufficient quantity and specific conditions Am241 (metal or oxide) will theoretically support criticality

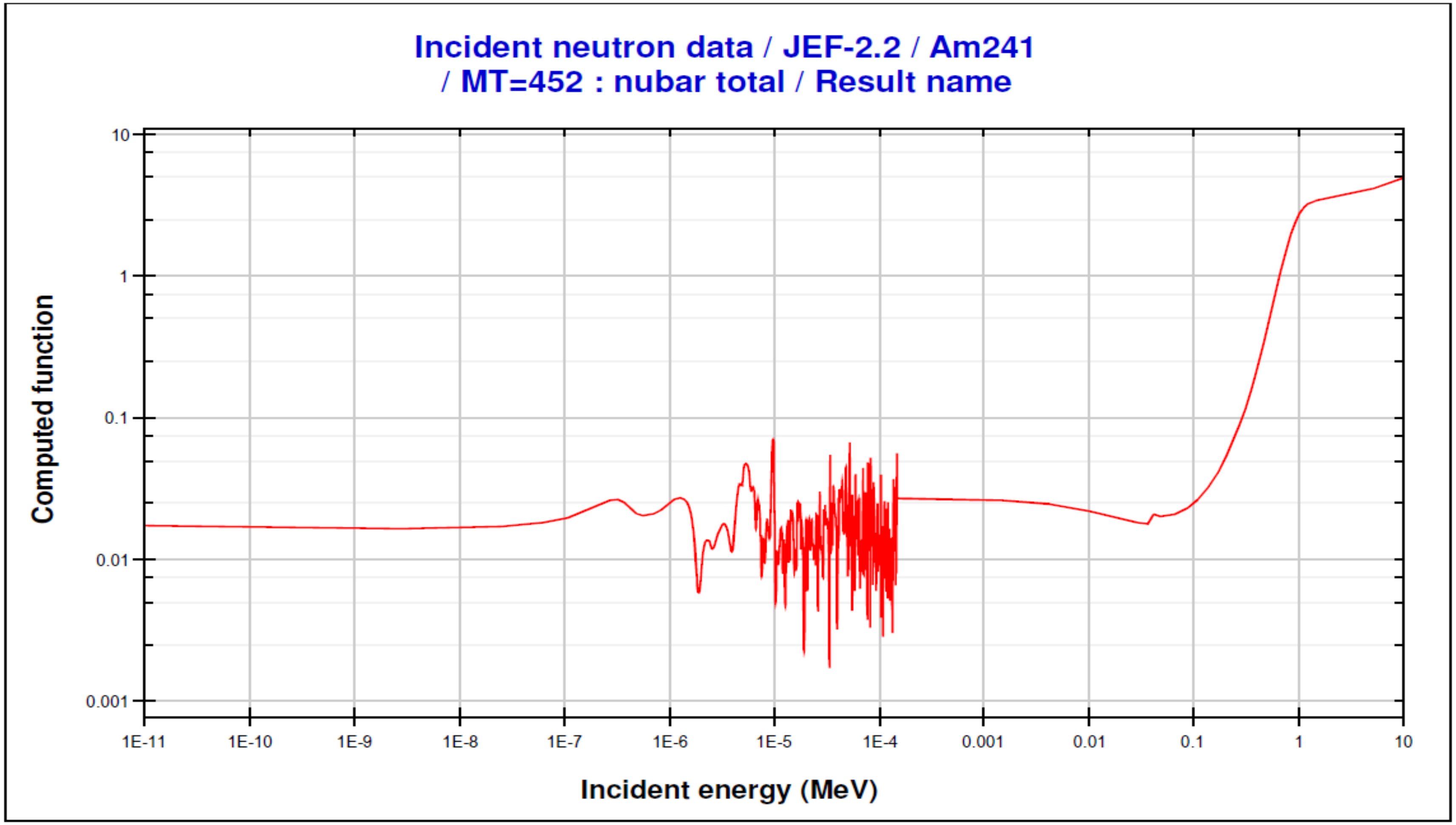
It is therefore a legal requirement in the UK that Am241 be subject to CRITICALITY CONTROL

In most (but not all) cases if <15g then an exemption from criticality control can be granted

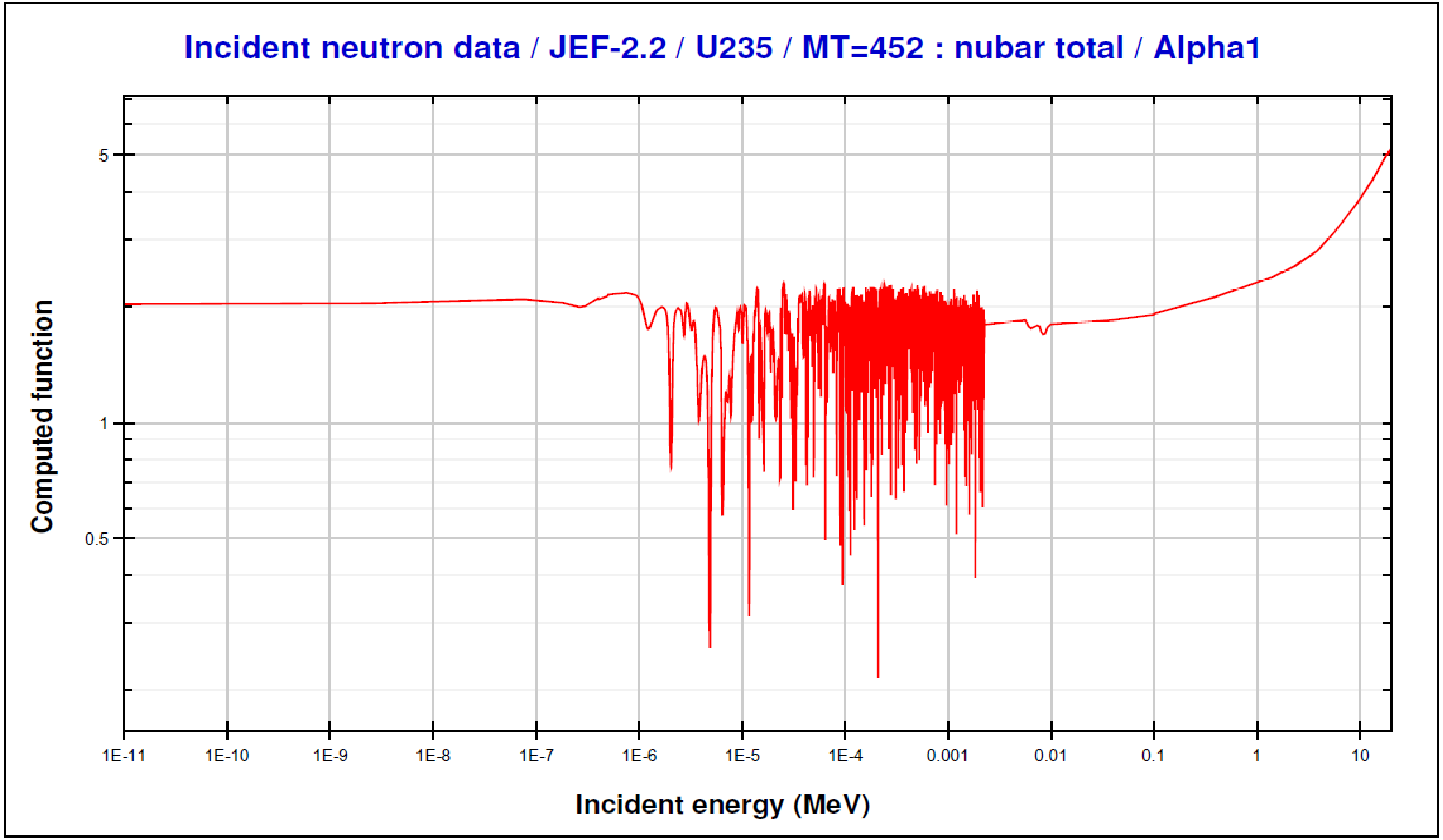
IAEA Regulations for transport contain no Am241 criticality limits yet

UK Regulator (ONR) has taken an Action (August 2014) to advise NNL/ESA as to how Am241 will be dealt with under IAEA Regulations

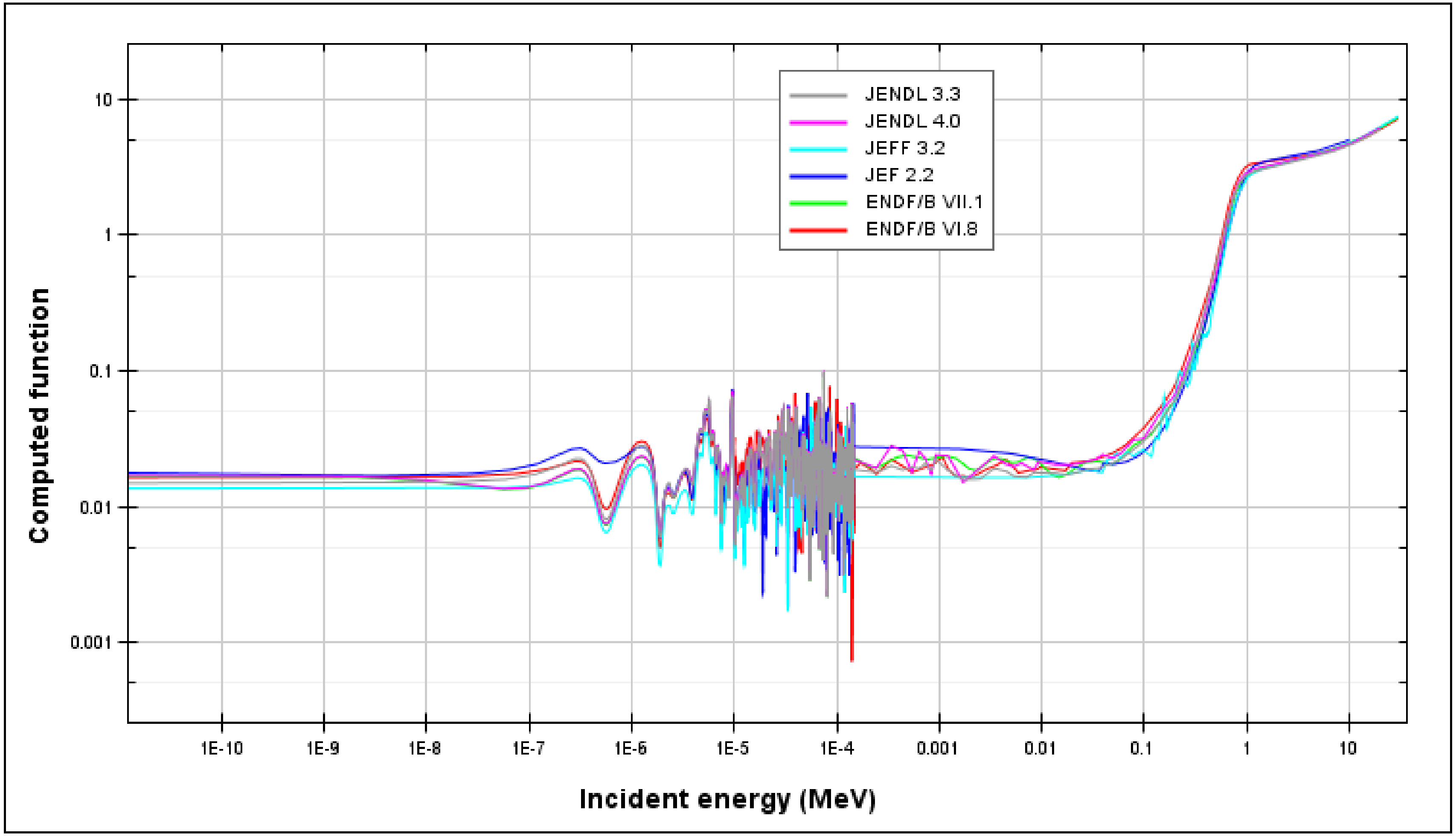
Am241 vs U235



Am241 vs U235



Am241 – various nuclear data



More Am241 specifics

The absence of any significant hydrogenous moderator is necessary for Am241 criticality

(H/Am241<1.25 approx)

HOWEVER

Carbon-12 “moderated” and reflected Am241 matrix (e.g. Space Battery) would support criticality if large enough

MASS CONTROL

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MASS control is the only really practical Criticality Control for Am241 for the systems and processes of interest

So we need to establish reliable safe (and operational) mass limits for Am241

Am241 – current mass limits

	Mass limit for safe operation	Moderator, Reflector or geometry limits	Status
Current limit for operations at NNL	**Up to 100g Am241 in any form**	**None**	**Currently approved criticality limit for use in NNL Central Laboratory**
Current limit for transport (ONR, IAEA Regulations)	**Up to 15g Am241 in any form**	**None**	**The 15g exemption embedded in IAEA Regulations as adopted by ONR**
Current (criticality) limit for Am241 in PCM wastes	**"Natural" Am241 levels only**	**None**	**Wastes artificially enriched in Am241 will require case by case consideration as they may affect Pu assay**

Am241 - target mass limits

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	Indicated Mass limit for safe operation	Moderator, Reflector or geometry limits	Status
Single Batch limit for normal operations	**1.0kg** For Am241 in any metal or oxide form.	**None, although heterogeneous systems need to be investigated to confirm simple sphere is worst case**	**Mass limit is indicative not proven for operations yet but likely to be supportable**
Single package limit for transport	**1.0kg** for Am241 in any metal or oxide form. **.**	**As above**	**As above**
Bulk storage limits	**10.0kg** for Am241 in any metal or oxide form**.**	**Some engineered control may be required (storage racks) and some limitations on reflectors (by design)**	**As above**

Am241 - target mass limits

	Indicated Mass limit for safe operation	Moderator, Reflector or geometry limits	Status
Final RTG assembly (assuming generic modular nominal 200g Am241/pellet design)	**Up to 10.0kg Am241 in oxide form only based upon a worst case.** **Equivalent to a 50We RTG.**	**None other than the final assembled geometry will be credited to some extent in the justification and may allow >10.0kg**	**Mass limit is indicative not proven for operations yet but likely to be supportable**

MONK9/JEF2.2 CRITICAL MASS

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System modelled	Computer Code and Nuclear Data Library	Reflector	Critical Mass in kg
Am241 metal sphere	**MONK9/JEF2.2**	**None**	**74**
Am241 metal sphere	**MONK9/JEF2.2**	**Full Water**	**67**
Am241 metal sphere	**MONK9/JEF2.2**	**Steel**	**44**
Am241 metal sphere	**MONK9/JEF2.2**	**30cm Carbon-12 (graphite)**	**44**
Am241 metal sphere (13.0g/cm^3 density)	**KENO ENDFB/VII.0 33 Energy Group**	**Carbon-12 (graphite)**	**53.5**

UK SAFETY CRITERION

$$\text{k-effective} + 3\sigma \leqslant X - E_{PD} - E_{SM} - E_{R}$$

X is the sub-critical limit

For simplicity 0.95 will be adopted.

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SUB CRITICAL LIMITS

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E_{SM} is the allowance for error due to specification and modelling of the system being addressed. Since the basis of the Am241 reference values will be a worst case spherical geometry (or a specific detailed model) an E_{SM} of 0 can be applied.

E_R is the allowance for the susceptibility of the system to increase in reactivity – an E_R of 0 is applied for the same reasons as above, a 'worst case' will be used to determine Am241 parameters.

E_{PD} is the allowance for error due to the MONK program and the nuclear data used.

Currently there is no means of setting this value with any confidence for Am241 and again here lies the problem and the driver for remedial works.

SUB CRITICAL LIMITS

This leads to a MONK criticality safety criterion for Am241 systems of:

$$\textbf{k-effective} + 3\sigma \leqslant 0.95 - E_{PD}$$

where E_{PD} is simply unknown.

SOURCES OF UNCERTAINTY

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Many factors contribute to the uncertainty in Am241 critical mass is wide range of results but some key ones are:

- **Lack of critical experiment**
- **Uncertainty in nuclear data**
- **Variations in nuclear data processing**
- **Uncertainty in Am241 density**
- **Variations in code computational techniques**

THE Am241 PROBLEM AGAIN

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Critical experiments feed into nuclear data and give significant confidence in computer codes for U, Pu

No such confidence in Am241, no critical experiments, large uncertainties in nuclear data at the 'fast' end > 1.0Mev, lead to large (kg) uncertainties in critical mass

Critical experiments with Am241 not feasible

Nuclear Data Error sources

- **Fission cross section**
- **Capture cross section**
- **Nubar**
- **Scattering**
- **Fission energy spectrum**
- **Nuclear Data Processing**

Some Nuclear Data variation

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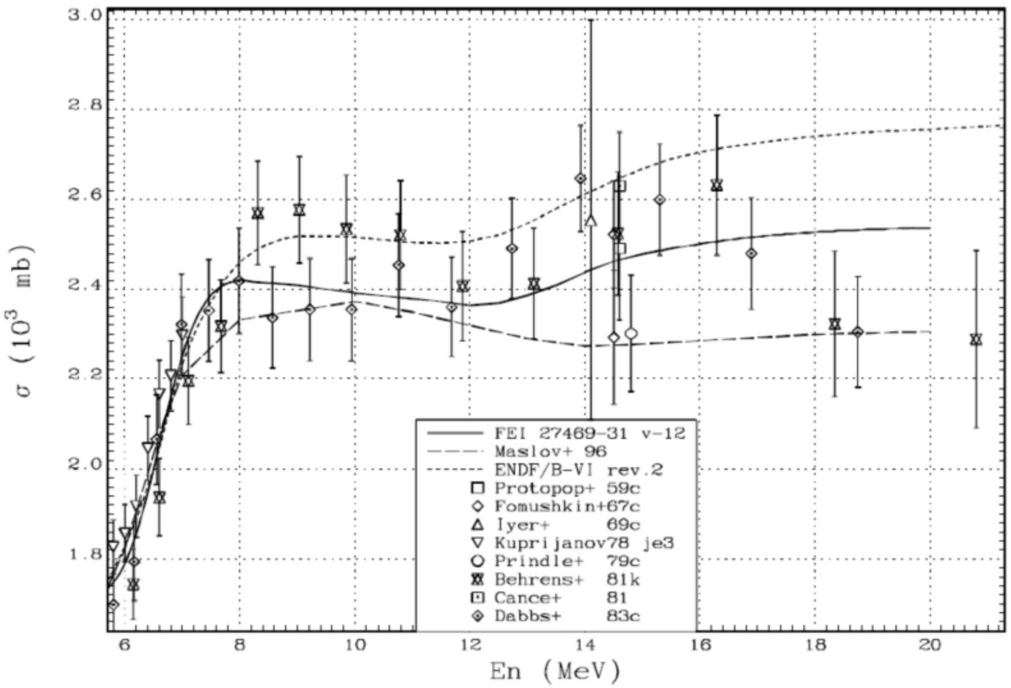
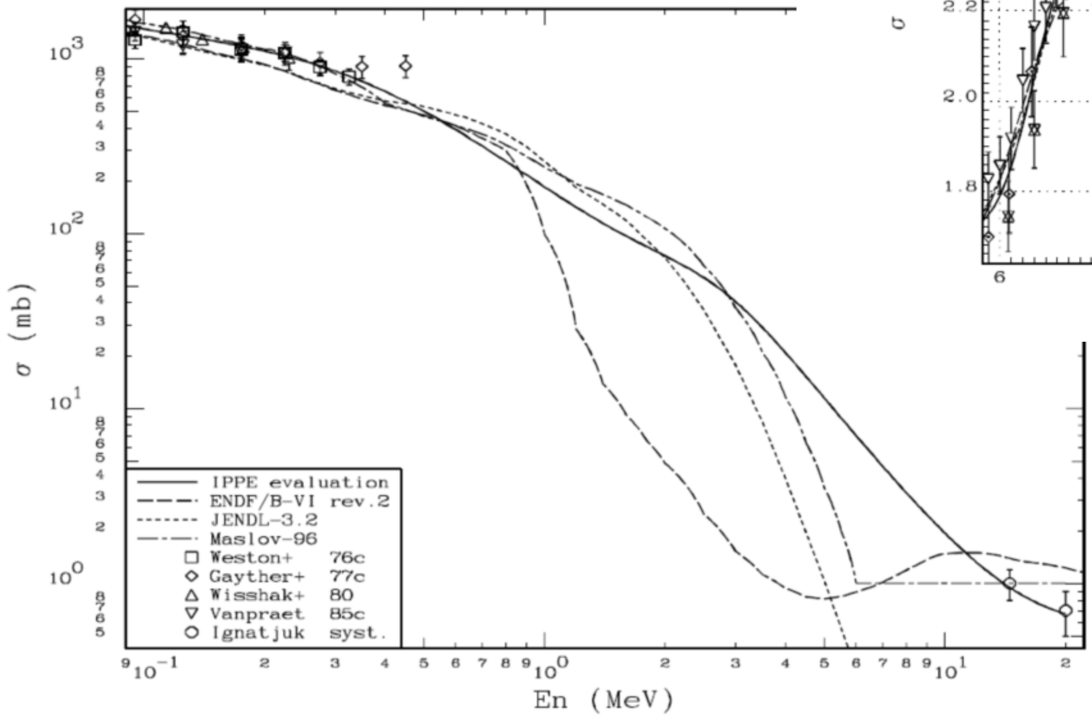
Fission cross section





Capture cross section

Figures from Ignatyuk et al

Outline Solution and Future Works

Most simplistically:

Justify a value for E_{pd}!

INITIAL BASIS

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MONK9A with JEF 2.2 Nuclear Data is currently the UK combination of choice for new assessment.

Validated and Verified for U and Pu and mixed U/Pu systems

Am241 – not validated

SIMPLE BENCHMARK

- **Am241 metal**
- **13.67g/cm^3 density**
- **Spherical geometry**
- **Various reflectors including steel and carbon-12**
- **MONK9/JEF2.2**

SUMMARY FORWARD PLAN

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- **Understand, attribute and quantify the nuclear data uncertainties (or worst case)**
- **Identify the best available nuclear data set for criticality safety assessment in light of these uncertainties**
- **Understand potential improvements to the nuclear data, including new integral experiments (via NEA Expert Group or otherwise)**
- **Specify and run (theoretical) Am241 benchmarks in support of improvement process.**

SPECIFIC Work Programme

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Task 1: understand, attribute and quantify the current Am241 nuclear data uncertainties (JEF2.2 and beyond, to date)
Task 2: understand/propose/justify the best available Am241 nuclear data set for criticality safety assessment in light of these uncertainties
Task 3: review/understand potential improvements to the proposed Am241 nuclear data set(s), including new integral experiments (via NEA Expert Group or otherwise)
Task 4: Develop, specify and run (theoretical) Am241 benchmarks in support of the improvement process (see Figure 1) - engage with NEA

Work Programme

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Task 5: Based upon output of Tasks 1-4 propose suitable integral experiment programme through NEA/EGIEMAM to further improve nuclear data, if appropriate. Loop back.
Task 6: Based upon studies undertaken propose a nuclear data set suitable for use in a criticality safety case, quantifying its uncertainties, to feed into setting of suitable sub-critical margins
Task 7: Justify applicability of safe parameters (operations) to stakeholder groups (SL, ONR)
Task 8: Justify applicability of safe parameters (transport) to stakeholder groups (SL, IAEA, Dept of Transport)

The Nuclear Data improvement Cycle

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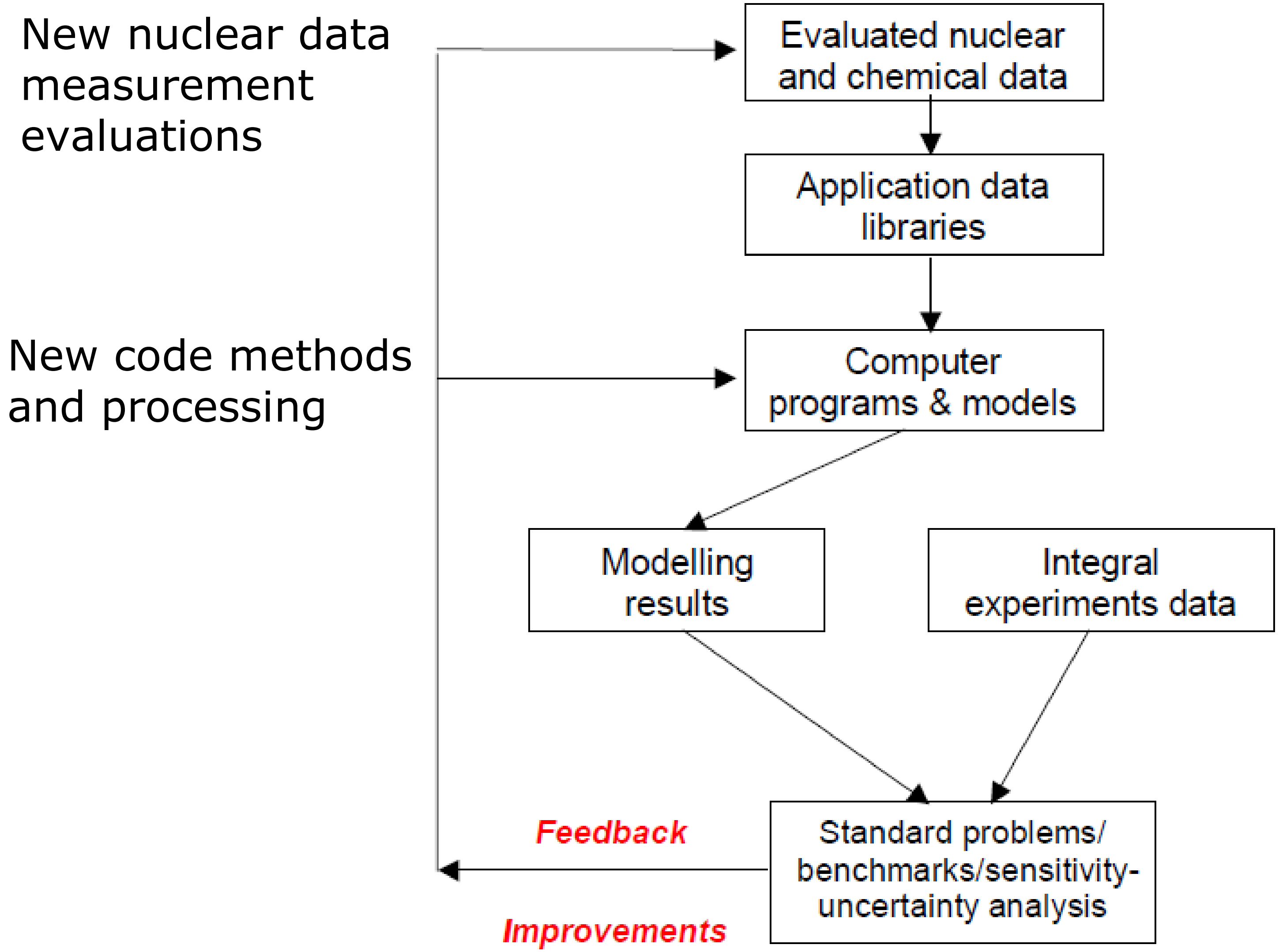

TECHNICAL NEEDS

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Within these tasks specific technical consideration needs to be given to:

- **Feeding in any improved data now available for Am241 fission and capture cross sections**
- **Focussing on improved scattering data (or understanding worst case)**
- **Better understanding energy spectra of prompt and delayed fission neutrons (or setting worst case)**

TECHNICAL NEEDS

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Carbon-12 aeroshell (graphite) specification will significantly affect scatter

The effect of temperature needs to be investigated (Doppler effect)

Effect of other shielding materials (as reflectors) may need to be investigated

Understanding whether alpha/n reactions in AmO_2 are of significance

TECHNICAL NEEDS

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Separate out nuclear data uncertainties from uncertainties that are due entirely to different processing of the same nuclear data by different criticality codes

The assumption that unmoderated spheres represent worst case needs to be further supported

TECHNICAL AIMS

Recommend a nuclear data set for use in an operational safety case based upon the above and/or justify a value for E_{PD}.

Stakeholder and Industry engagement

European Space Agency

Sellafield Ltd

ONR

OECD / NEA

AWE Aldermaston

IAEA

National and International Experts on nuclear data and uncertainty / covariance analysis

Acknowledgements

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esa space for europe European Space Agency

SEVENTH FRAMEWORK PROGRAMME

END

SUPPORTING INFO ONLY FOLLOWS
AND IN RANDOM ORDER

Existing literature and standards

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The American National Standard for Special Actinides ANS8.15 gives some guide limits for Am241

Not robust enough to use in modern safety case (and our US colleagues and UK Regulator concur)

Various computer calculations undertaken by many with wide range of results

Critical Masses for Bare ^{241}Am Sphere in Literature

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Density in g/cm3	Code	Nuclear Data Library	Critical Mass in kg
Metal, 13.6	MCNP4A	ENDL-83	54.4
Metal, 11.7	MONK	UKNDL	56.4
Metal, 13.66	MONK 8	UKNDL	56.4
Metal, 13.66	EMS?	ENDF/B-VI.2 DN	57.6
Metal, 13.66	MCNP	ENDF/B-VI	57.7
Metal, 11.7	XSDRNPM	238 group; ENDF/B-VI	59.5
Metal, 11.7	XSDRNPM	199 group; ENDF/B-VI	60.0
Metal, 13.67	TRIPOLI 4.1	JEF 2.2	72.695
Metal, 13.66	MCNP	JEF 2.2	73.3
Metal, 13.66	MCNP	JENDL 3.2	73.7
Metal, 13.6	MCNP4A	JENDL-3.2	74.8
Metal, 13.67	APOLLO2	172 group, JEF 2.2	75.614
Metal, 13.66	MONK 8	JEF 2.2	75.7
Metal, 11.7	MONK	JEF 2.2	75.9
Metal, 13.66	MONK 8	JENDL 3.2	76.1
Metal, 11.7	MCNP	ENDF/B-VI.2	78.5+/-0.9

Density in g/cm3	Code	Nuclear Data Library	Critical Mass in kg
Metal, 11.7	Dantsys	30 group; ENDF/B-VI	82.8
Metal, 11.7	MCNP	JENDL 3.2	101.6+/-1.5
Metal, 13.6	SCALE 4.3	ENDF/B-IV	102
Metal, 13.6	MCNP4A	ENDF/B-V	108.3
Metal, 11.7	Dantsys	ABBN-93	122.5
Metal, 11.7	Dantsys	Scale 4.4 44 group; ENDF/B-V	139.9
Metal, 11.7	Dantsys	30 group; ENDF/B-V	145.3
Metal, 11.7	MCNP	ENDF/B-V	147.9+/-2.0
Metal, 13.6	SCALE 4.3	ENDF/B-IV	197
Oxide, 11.68	MCNP4A	ENDL-83	91.6
Oxide, 11.69	MCNP	ENDF/B-VI	94.6
Oxide, 11.69	MCNP	JEF 2.2	129
Oxide, 11.69	MCNP	JENDL 3.2	131
Oxide, 11.68	MCNP4A	JENDL-3.2	135
Oxide, 11.68	SCALE 4.3	ENDF/B-IV	199
Oxide, 11.68	MCNP4A	ENDF/B-V	207
Oxide, 11.68	SCALE 4.3	ENDF/B-IV	313

From Noijiri & Fukasaku; Brewer, Westfall and Clayton; Dias & Tancock; Lavarenne et al

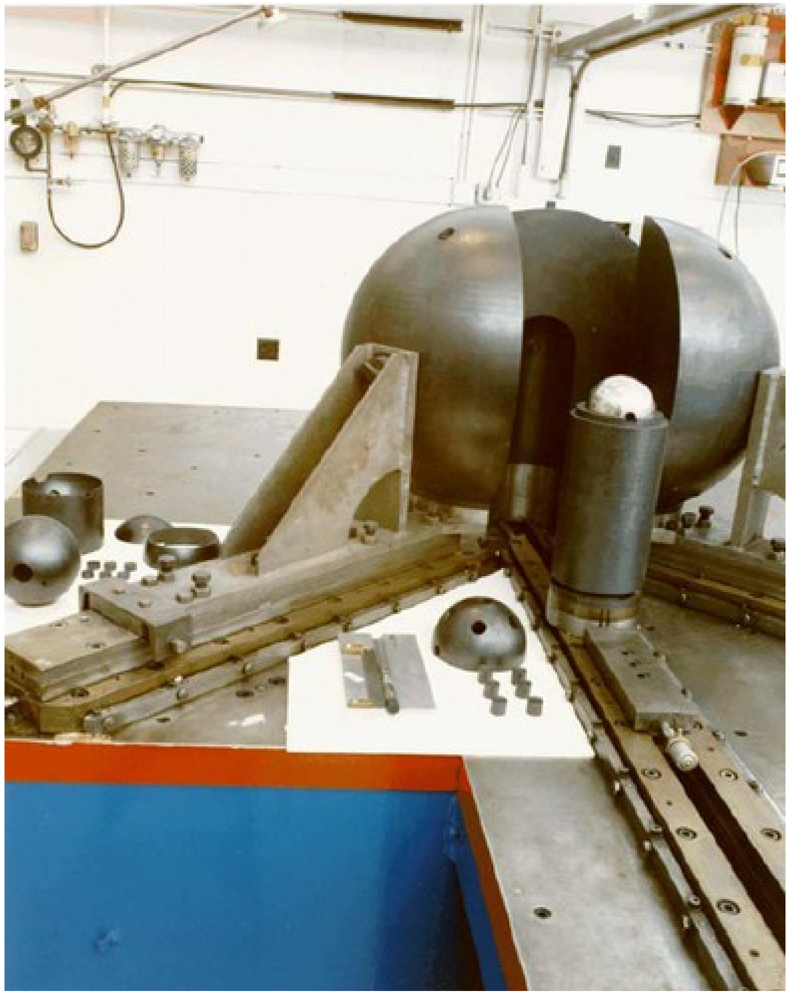
ANS 8.15 Standard Values Based on FLATTOP Experiment

Table I
Subcritical Mass Limits for Single Units
Reflected by Infinitely Thick Water or Steel

Non-Fissile Nuclide	Chemical Form	Subcritical Mass Limit (kg) Water Reflector[a]	Steel Reflector[b]
^{237}Np	Np	30	20
	NpO_2	140	90
^{238}Pu	Pu	4	3
	PuO_2	11	7
^{240}Pu	Pu	20	15
	PuO_2	70	45
^{242}Pu	Pu	60	40
^{241}Am	Am	24	16
	AmO_2	40	32
^{243}Am	Am	35	25
	Am_2O_3	50	37
	AmO_2	60	45
^{244}Cm	Cm	5	3
	Cm_2O_3	7	5
	CmO_2	7	5

[a] The mass limits for water reflectors may also be applied to combinations of steel and water (steel backed by water) for a steel thickness ≤ 1 cm.

[b] The steel reflector thickness is 20 cm, i.e., effectively infinite.



NNL new calculations: current nuclear data comparison

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Evaluation	Bare Sphere		Water Reflector	
	Radius (cm)	Mass (kg)	Radius (cm)	Mass (kg)
UKNDL	9.94554 ± 0.00711	56.330 ± 0.120	9.59357 ± 0.007231	50.559 ± 0.114
JEF 2.2	10.9555 ± 0.00573	75.292 ± 0.118	10.5734 ± 0.005615	67.686 ± 0.107
JENDL 3.2	10.9859 ± 0.00644	75.921 ± 0.133	10.6248 ± 0.005112	68.678 ± 0.099
ENDF B/VI	11.5385 ± 0.01181	87.964 ± 0.270	11.1654 ± 0.008645	79.703 ± 0.185

Table 1: Critical radius and critical mass values for bare and water reflected Am^{241} spheres.

Various Nuclear Data with KENO
Nuclear Data Uncertainty

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Preliminary results calculated using KENO with 33 energy group (53.5kg sphere 13g/cm3 density – INL)

Library	ENDF/B-VII.0	JEFF 2.2	ENDF/B-VI.8	JEFF 3.1
K_{eff}	1.00035	1.00868	1.04679	1.04566

Library effect: Case of graphite reflector (9.94 cm Am-241 radius and graphite in outward region up to 30 cm radius).

Thanks are due to G.Palmiotti (INL) for these calculations