PRESENTATION

by

Dr. R.W. MILLS

Dr. Mills presented twopapers, ofwhich extracts areattached. Full versions of both papers are available on application to:

NEA Data Bank
Bâtiment 45
91191 Gif-sur-Yvette Cedex
France

Title:

Review of fission product yields and delayed neutron data for the actinides Np-237, Pu-242, Am-242m, Am-243, Cm-243 and Cm-245.

Author:

R W Mills, School of Physics and Space Research, The University of Birmingham, B15 2TT. England. With the advice and assistance of D R Weaver of the University of Birmingham.

Abstract:

A review of fission product yields and delayed neutron data for Np-237, Pu-242, Am-242m, Am-243, Cm-243 and Cm-245 has been undertaken. Gaps in understanding and inconsistencies in existing data were identified and priority areas for further experimental, theoretical and evaluation investigation detailed.

Introduction:

Since 1982 it has been recognised that long term basic research into utilisation of high level nuclear waste by separation into individual actinides and fission products could produce many benefits. It is currently planned that the actinides be disposed of in geological formations, unseparated from the fiiion products. However, they could, if separated, yield a more efficient approach to the utilization of the limited world nuclear resources and the reduction of long-lived nuclear waste. Fission products from the uranium and plutonium isotopes conventionally present in nuclear fuel could be a source of noble metals to be used as catalysts in industrial chemistry, and of radionuclides for use in medicine and industry. The higher actinides, on the other hand, could be introduced into reactors as fuel. This could have dual benefits; firstly by contributing to the generation of heat, and secondly by reducing the quantities of the-se materials through their being fissioned. Thus they would be present in lower concentrations in the high-level nuclear waste and, as the actinides are longer lived than most of the fission products, it would reduce the time high level waste must be monitored.

The Japanese. government has decided. to fund research and development in this field, through a project entitled 'OMEGA': 'Options Making Extra Gains of **Actinides** and fiiion products generated in the nuclear fuel cycle-.

This report was funded by the OMEGA project through the Nuclear Energy Agency **(NEA).** It represents a critical review of fission product yields and delayed neutron data for **actinides** of interest to the OMEGA project. **The nuclides considered** were the most important higher **actinides** Np237, **Pu-242, Am-242m,** Am-243, Cm-243 and Cm-245. **The** objects of this study were the identification of **gaps** in understanding and inconsistencies in **existing data** and also to identify priority areas for further experimental, theoretical and evaluation efforts

For fiiion products the open literature and available computer readable **databases (CINDA** and **EXFOR)** were scanned for **chain,** cumulative, independent and ternary **yields.** For data on delayed neutrons the quantities of interest were: total delayed neutron yields ν_d, the time dependence of the neutron activity, and the delayed neutron spectra. The same **sources** of data were searched as for fission yields.

The search for data was restricted to neutron induced and spontaneous fission, with the energy beiig **specified** as one of the three following classes

(1) Thermal neutrons **Maxwellian** distributed neutrons with a mean energy of **1/40** eV.

(2) Fast neutrons here the definition is **less** precise as a fast reactor can have a wide range of average energies from a few hundred **keV** to several MeV, depending on the **composition of** the reactor. In practice, most **fast** reactor designs have a mean energy of about 400 keV.

(3) **High** energy neutrons: these are formed around 14-16 **MeV** by accelerator induced **reactions,** and if commercial fusion becomes viable would be produced by fusion reactors.

At the moment it is felt that charged particle and photon induced fission would not produce sufficiently great reaction rates both from consideration of the appropriate cross-sections and due to the low fluxes currently obtainable from accelerator. Also it is noted that within the data extracted there were no results from the use of monoenergetic neutron beams, except for those for the third class mentioned above "High Energy Neutrons-.

In the medium term only thermal and fast reactors are available, however it should be noted that, if fusion becomes commonly available, the mass and charge distribution from fission can be greatly changed by changing the energy of the neutrons causing fissions from thermal or fast to 14MeV. On one hand this may enable, by variation of neutron energy, the minimisation of fission product activity in waste or the maximisation of the production of rare radionuclides or of highly valuable stable nuclides. Several papers on actinide burning reactors were presented at the PHYSOR conference at Marseilles, France during May 1990, proceedings of which are to be published.

For thermal and fast reactors the delayed neutron component of the reactor flux is of great importance in the control and design of the reactor. As, in the future, greater concentrations of the higher actinides will be generated both through usc of recycled fuels and as reactor burnups are increased, there will be increasing need for their delayed neutron data in order to predict reactor kinetics, a procedure that had not, until recently, been attempted without the use of greatly simplified models (private communication, M.Brady, ORNL). In fusion reactors, on the other hand, the difference in the typical energies of delayed neutrons and those from fusion, plus the need to place the actinides outside the reactor core make it unlikely that delayed neutrons from actinides would have any major effects.

<u>Fission Yields:</u>

Fission yields are reported in 3 types

(i) The independent yield, Yi(A,Z,I), is the yield of a particular nuclide of mass A, charge Z and isomeric state I produced directly from fission.

(ii) The cumulative yield Yc is the yield of a nuclide (A,Z,I) over all time, i.e. Yc is the independent yield for (A,Z,I) plus all the contributions from (A', Z',I') decaying into (A,Z,I) when

Conclusion on data nuclides of interest:

Fission yields.

The datasets for Np-237F, Pu-242F, Am-242mT, Cm-243T and Cm-245T have a good enough coverage of chain yields that the predictive models, interpolations and extrapolations described above wilt give a good estimate of values in the remaining gaps in the total chain yield distribution. Of the remaining reactions understudy, some have no experimental results at all, while the rest are only poorly defined. Appendix A lists cases where there are at Least some data, but systems which have no published chain yield data are not mentioned in the tables. Clearly filling of gaps or estimating of the whole chain yield distribution is much less certain than if there is good coverage from experimental results.

There is need for new measurements of chain yields both to confirm previous results and to fill gaps in the unmeasured regions. New work should concentrate on those nuclides whine fission rates are highest for what are considered reasonable estimates of typical fuel composition in reactors of interest

There is little data available for fractional independent yields of the nuclides under study. However, if it is required that complete yield sets be produced, then it is desirable to have some experimental values so as to be able to determine the appropriate model parameters. Thus, if large enough samples of these materials can be produced, the relatively new mass separators would be advantageous for new measurements because they allow the measurement of many independent yields without complicated and time consuming chemical separations Examples of these mass separators are Lohengrin and Hiawatha.

Until now, only empirical models have been used for the modelling of fission yields; it would be useful to investigate whether nuclear physics theory of fission can provide a deeper insight into the processes involved and hence the form of equations which might be used to represent fission product emission.

Delayed neutron data.

Delayed neutron measurements are difficult to make accurately, and considering the small amount of data currently available, more measurements are required both of ν_d, the time dependence (allowing all parameters to vary and using a full covariance analysis), and measurement of spectra.

However, in the short and medium term, improvements in fission yields and decay data for the summation methods would in my opinion produce a greater improvement in the data. This would have to be followed up by experimental measurements however. to confirm the results, because at present, the charge distributions for fission in these nuclides are produced by extrapolation from the charge distribution model parameters for better characterised systems. Thus the predicted charge distributions are uncertain and hence, as these distributions are very important in determining the delayed neutron precursor formation, so are the delayed neutron emission characteristics

Acknowledgements.

I would like to thank the following for helping me in this work, Dr C Nordborg, Dr S Webster, Dr M Lammer, Dr V McLane, Dr M Brady, Mr M F James and the staff of the NEA Data Bank.

References:

1. R W Mills, M F James and D R Weaver, United Kingdom Atomic Energy Establishment (Winfrith) Report. "A new evaluation of fission product yields and the production of a new library (UKFY2) of independent and cumulative yields", this will be published during 1990 in 4 parts.

2. A Wahl, Atomic and Nuclear Data Tables 39, 1 (1988).

3. A Wahl, Roe. of the '50 Years with Nuclear Fission" meeting at NIST Washington DC, page 525. (1989).

4. A R Musgrove, J L Cook and G D Trimble, Roe. Panel Fission Product Nuclear Data, Bologna, IAEA-169, 2, 163-200 (1973).

5. R W Mills et al, Proc. of the ● 5O Years with Nuclear Fission meeting at NIST Washington DC, page 471. (1989).

6. M C Brady "Evalation and application of delayed neutron precursor data", Thesis published as Los Alamos report LA-11534-T.

7. R J Tuttle "Delayed neutron yields in nuclear fission" Roe. consultants' meeting on delayed neutrons properties, Vienna, 26-30 March 1979.

8. R J Tuttle, Nucl. Sci. Eng. 56, 37 (1975).