A MODIFICATION TO THE ORIGEN2 CODE FOR CALCULATION OF NEUTRON EMISSION FROM (α, n) REACTIONS IN MIXTURES OF LIGHT ELEMENTS

S.Appleton, A.Johnstone, D.Vande Putte
Electrowatt Engineering Services (UK) Ltd
Grandford House, Carfax, Horsham
West Sussex, RH12 1UP, England

ABSTRACT

The radiological characteristics of high level waste glass blocks are becoming of increasing interest in view of the operation and pending completion of several plants for vitrification of such waste.

An important parameter for the storage and handling of the waste is its neutron emission. This is due to spontaneous fission of heavy elements (e.g curium) as well as the (α, n) nuclear reaction. The importance of the latter reaction derives from the presence of alpha emitting actinides and light element constituents of the glass; its relative importance increases with lower burn-up. For example, 90% of the neutron emission from vitrified high level waste from Magnox reactor fuel is due to the (α, n) reaction.

The actinide content of the wastes can be calculated using the ORIGEN2 code. Unfortunately, in versions of ORIGEN2 code currently available from international data banks, the only neutron source considered for the (α, n) reaction is that where a single element (oxygen) is the target. A formalism for calculating neutron yields from (α, n) reactions in mixtures of light elements was developed by West in the United Kingdom. This relies on the ratio of the specific ionization energy loss of alpha particles for two elements remaining more or less constant as the alpha particles slow down. This fact allows the use of energy dependent single element thick target yield measurements in calculating neutron yields from mixtures of elements. The ORIGEN2 code was modified to calculate the neutron yield from (α, n) reactions in mixtures of light elements using West's formalism. The output is in the form of a table showing the contribution to the neutron yield of each light element. A total source strength is also given. Typical results have been checked against hand calculations thus verifying the modification.

Work is also in hand to modify ORIGEN2 by incorporating a routine to calculate an energy spectrum for a mixture of spontaneous fission and (α, n) neutrons. The requirement for this stems from the fact that neutrons from the (α, n) reactions resulting from actinide alpha particles is harder than the standard Cf252 spontaneous fission spectrum.

INTRODUCTION

Radiological protection in installations handling actinides must take account of any resultant neutron emission. Such emission can be due to the spontaneous fission of actinides. It could also result from the nuclear reaction between the alpha particle emitted by an actinide and light elements mixed in with the actinides. Such an event is known as an (α, n) reaction.

The radiological importance of the (α, n) reaction has long been recognized, as is must be accounted for in plants which chemically process concentrated solution of actinides (eg reprocessing). It has become particularly relevant in the last decade with the advent of High Level Waste vitrification. Such vitrified products consist mainly of light elements, some of which yield relatively large numbers of neutrons from the (α, n) reaction eg boron.

Measurements of neutron yields from the (α, n) reaction in individual light elements are available (1,2,3,4). The calculation of the (α, n) yield in a mixture of light elements, using yield values for single light elements is not straightforward. This is explained in the next section of the paper, and a solution to this difficulty is described.

The ORIGEN2 code (5) provides a useful means of calculating the radionuclide content and chemical composition of irradiated fuel and of any waste resulting from its chemical processing. The output from the code includes tables giving the neutron emission from spontaneous fission, and from the (α, n) reaction, but in oxygen only. This paper explains how the ORIGEN2 code was modified so as to calculate and print out the values for neutron emission from the (α, n) reaction with all light elements specified in the input.

Finally, examples of results are given, demonstrating the importance of expanding the code's capabilities. Work in progress to exploit this modification is also described.

THE YIELD FROM (α ,n) REACTIONS IN MIXTURES

Consider an alpha particle of energy Eα emitted within one cc of a pure light element sample with an atom density of n_j atoms/cm^3 . The number of neutrons produced, also known as the thick target yield is:

$$Y_j = n_j \int_{}^{R_j} \sigma_j(E)dx \qquad \text{(Eq. 1)}$$

where Rj is the range of the α particle in the substance and σj(E) is the cross section from the (α, n) reaction. The variable in the integral can be changed and the expression for the thick target yield re-written as:

$$Y_j = n_j \int_{Ea}^{0} \sigma_j(E)\cdot[dx/dE]_j \; dE \qquad \text{(Eq.2)}$$

where [dx/dE]j is the reciprocal of the stopping power of the material for an α particle. For a mixture, the neutron

emission will be:

$$Y_M = \sum_J n'_j \int_{Ea}^{0} \sigma_J(E)\,[dx/dE]_M\,dE \qquad \text{(Eq.3)}$$

where n'_j is the atom number density of element j in the mixture and $[dx/dE]_M$ is the reciprocal of the stopping power of the mixture for an α particle. It is known that:

$$(dx/dE)_M = \left[\sum_J (dE/dx)_j\, n'_{j/n_j}\right]^{-1} \qquad \text{(Eq.4)}$$

Thus Eq. 3 can be re-written as:

$$Y_M = \sum_J n'_j \int_{Ea}^{0} j(E) \times \left[\sum_J (dE/dx)_j\, n'_j/n_j\right]^{-1} dE \qquad \text{(Eq.5)}$$

The measured values for thick target yields in light elements, represented by Eq. 2, are not sufficient in solving Eq. 5. This is because in developing the summation in Eq. 5, mixed terms of the type

$$\int_{Ea}^{0} \sigma_J(E)\,(dE/dx)_k^{-1}\,dE;\ k \neq j \qquad \text{(Eq.6)}$$

will appear, for which no experimental data are available, in contrast to terms where k = j which are related to the thick target yields.

West (6) proposed an ingenious solution to this problem. Having observed that there is a wide range of atomic numbers and energy for which the variation of the stopping power with energy is similar, he expressed the stopping power of a substance j as:

$$(dE/dx)_j\ (dE/dx)_1 \cdot K_j l \qquad \text{(Eq. 7)}$$

Where Kjl is the ratio of stopping powers in materials l and j, and element l is a reference element.

Hence for a mixture:

$$(dE/dx)_M = (dE/dx)_1 \sum_J kjl\ n'_j/n \qquad \text{(Eq.8)}$$

By substituting Eq. 8 into Eq. 5, one obtains

$$Y_M = \sum_J n'_j \int_{Ea}^{0} \sigma_j(E)\,(dE/dx)_1^{-1} \left[\sum_J n'_j\, Kjl/n_j\right]^{-1} dE$$

Since Kjl ≈ constant:

$$Y_M = \frac{\sum_j n'_j \int_{Ea}^{0} \sigma_j(E)\,(dE/dx)_1^{-1}\,dE}{\sum_j n'_j\, Kjl/n_j} \qquad \text{(Eq.9)}$$

Using Equations 1 and 7, Equation 9 reduces to:

$$Y_M = \frac{\sum_j n'_j\, Y_j\, Kjl/n_j}{\sum_j n'_j\, Kjl/n_j} \qquad \text{(Eq.10)}$$

Equation 10 therefore allows the calculation of the (α, n) yield of a mixture using the thick target yields of its constituents. West (6) also showed that by careful choice of the energy at which to evaluate Kjl, the method he proposes leads to an error of less than a few percent.

ACTINIDE AND LIGHT ELEMENT DATA.

The actinides considered in ORIGEN2 and of relevance to the (α, n) reaction are: Th-230; U-234; U-235; U-236; U-238; Np-237; Pu-238; Pu-239; Pu-240; PU-241; Pu-242; Am-241; Am-242m; Am-243; Cm-242; Cm-243; Cm-244; Cm-245.

The light elements considered for inclusion in the modification are: Li; B; Na; Mg; Al, Si; O.

Thick target yields for these elements and each of the a emitters listed above were derived from Refs. 1-4.

The constants Kjl expressing the ratios of stopping powers were derived from data in Ref. 7. In this respect, elements which do not contribute to the (a n) reaction also need to be accounted for, as they appear in the denominator of Eq. 9. These unproductive elements are grouped into 3 classes: fission and activation products ($Z<80$), heavy elements ($Z>80$), and iron. For each class a single, effective stopping power was selected. Oxygen was chosen as the reference element to which stopping powers are related Eq (7).

ORIGEN2 MODIFICATION

The reader is assumed to be familiar with the general concepts of the ORIGEN2 code, although a full description may be found in (8).

The modification has been incorporated such that it works in addition to the existing (α, n) calculations rather than replacing them. One additional page of results is produced at the end of the output file. This tabulates for each of the output vectors the calculated (α, n) production rate (with units of neutrons per second per basis unit), together with a list of fractional contributions from the seven light elements which are considered as neutron producing target materials. The identity of each nuclide in a vector is examined, and the quantity of material (in moles) is accumulated in the appropriate category of the group of ten target materials discussed above. The relative stopping power data for these categories are coded within the routine, and thus the denominator term in Eq. 10 may be calculated. The nuclides in the vector which are the alpha emitting actinides of interest are also identified.

The neutron yields from each actinide-light element interaction are calculated in turn. The thick target yield and specific activity (in Bq mol^{-1}) data are coded within the routine. These are used to calculate a quantity which is one term in the numerator of Eq.10 (ie a single actinide-light element interaction) multiplied by the alpha activity of the actinide under consideration. When divided by the previously calculated denominator term this becomes the neutron production rate from a single actinide light element interaction. These individual neutron production rates are summed to give a sub- total for each target material, and the subtotals are summed to give the overall total neutron production rate for the mixture of materials in the vector. Once the total has been calculated the subtotals are converted into fraction contributions. Two actinides, Pu-241 and Am- 242m, form special cases in that they emit significant amounts of alpha particles in only fractions of their (0.00245% and 0.475% respectively) of their decays; to accommodate this the numerator terms of Eq. 10 are multiplied by these factors in all interactions involving these nuclides.

The routine is self contained, and does not alter the values of any existing variables used outside of the routine in the ORIGEN2 code. Values are read from the existing arrays XNEW and NUCL, which are respectively the quantities and identifiers of the nuclides in the output vectors.

RESULTS, DISCUSSION AND VERIFICATION

Figure 1 shows the (α, n) yield in typical vitrified High Level Waste products for two types of reactor, assuming identical glass compositions and a 150 litre container. The first is a natural uranium gas cooled reactor (Magnox), the second a Light Water Reactor. The spontaneous fission yield is also indicated. These results, obtained using the modified ORIGEN2 code show that it is important to extend the ORIGEN2 list of (α, n) reactions beyond that in oxygen. Figure 1 also demonstrates that in some cases, these additional light elements contribute a substantial fraction of the total neutron emission.

The (α, n) yields for several ORIGEN2 runs were verified by hand calculation. In addition, the oxygen (α, n) yield was checked against the existing version of ORIGEN2. Satisfactory agreement to better than 2% was found in all cases.

FUTURE DEVELOPMENTS

The ORIGEN2 output includes a tabulation of an effective photon spectrum. Work is in hand to extend the output by tabulating an effective neutron spectrum. This will allow direct transfer of ORIGEN2 output to shielding code inputs.

The best place to calculate such a spectrum is within ORIGEN2, as the spectrum's hardness depends very much on the relative amounts of (α, n) and spontaneous fission neutrons. The difference in hardness between these two sources of neutrons is shown in Fig.2.

REFERENCES

1. H.Liskien and A.Paulsen. Neutron yields of light elements under α bombardment. Atomkernenergie, p 59, Vol 30, 1977.
2. J.K.Bair and J.Gomez del Campo. Neutron yields from alpha particle bombardment. Nuclear Science and Engineering. p 18, Vol 71, 1979.
3. D.West and A.C.Sherwood. Measurement of thick target (α,n) yields from light elements. Report AERE-R-10502, 1982.
4. B.H.Weren and J.E.Faulkner. (α,n) Neutron source in commercial high-level waste. Transactions of the American Nuclear Society. p 608, Vol 45, 1983.
5. M.J.Bell. ORIGEN-The ORNL isotope generation and depletion code. Report ORNL-4628, Oak Ridge National Laboratory, 1973.
6. D.West. The calculation of neutron yields in mixtures and compounds from the thick target (α,n) yields in the separate constituents. Annals of Nuclear Energy, p549, Vol 6, 1979.
7. J.F.Ziegler. Helium: stopping power and ranges in all elemental matter (The stopping and ranges of ions in matter). Pergamon Press, Oxford, 1977.
8. A.G.Croff. A user's manual for the ORIGEN2 computer code. Report ORNL-TM-7175, Oak Ridge National Laboratory, 1980.

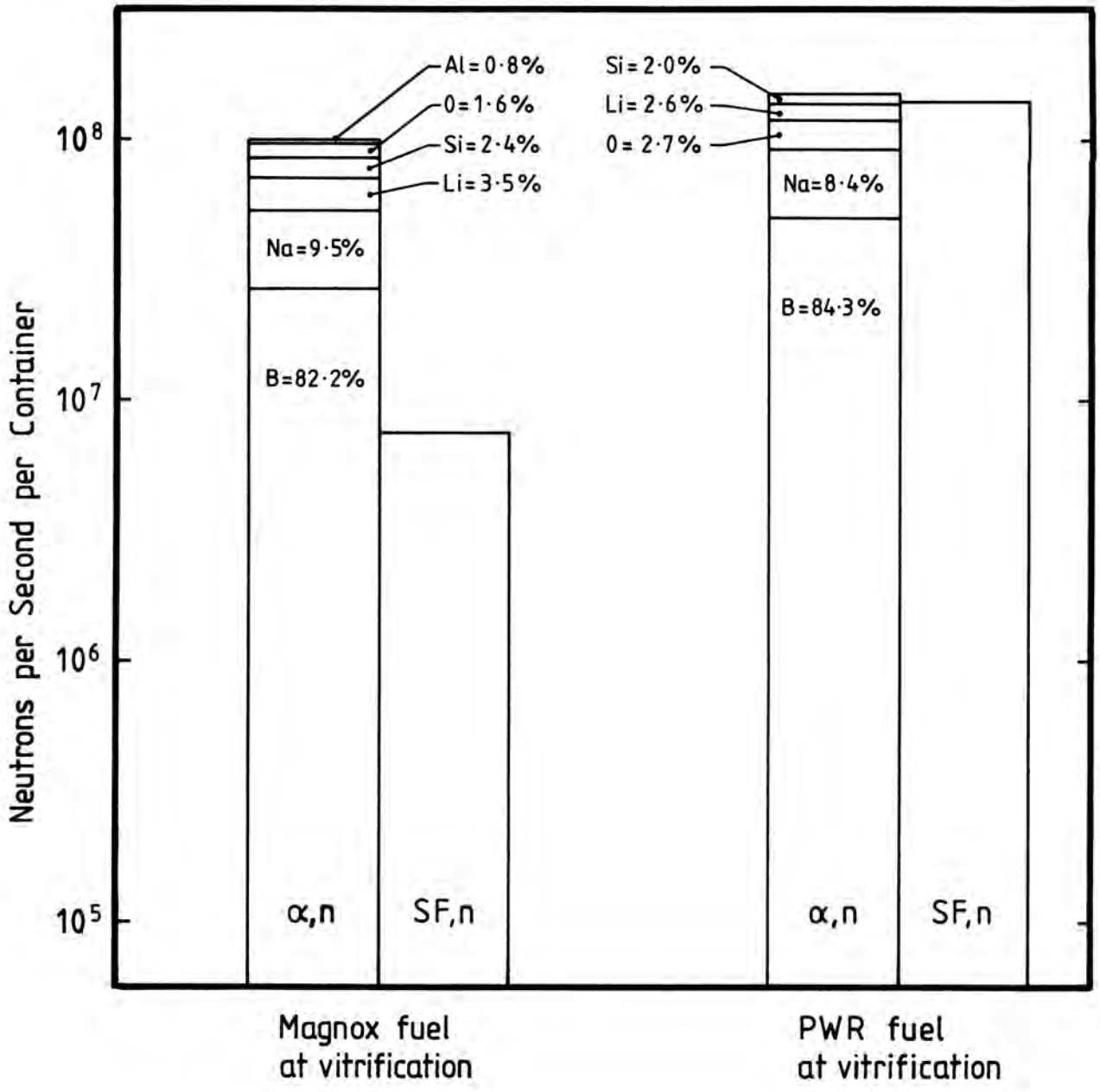

Fig. 1. Neutron Production in Vitrified High Level Waste Containers.

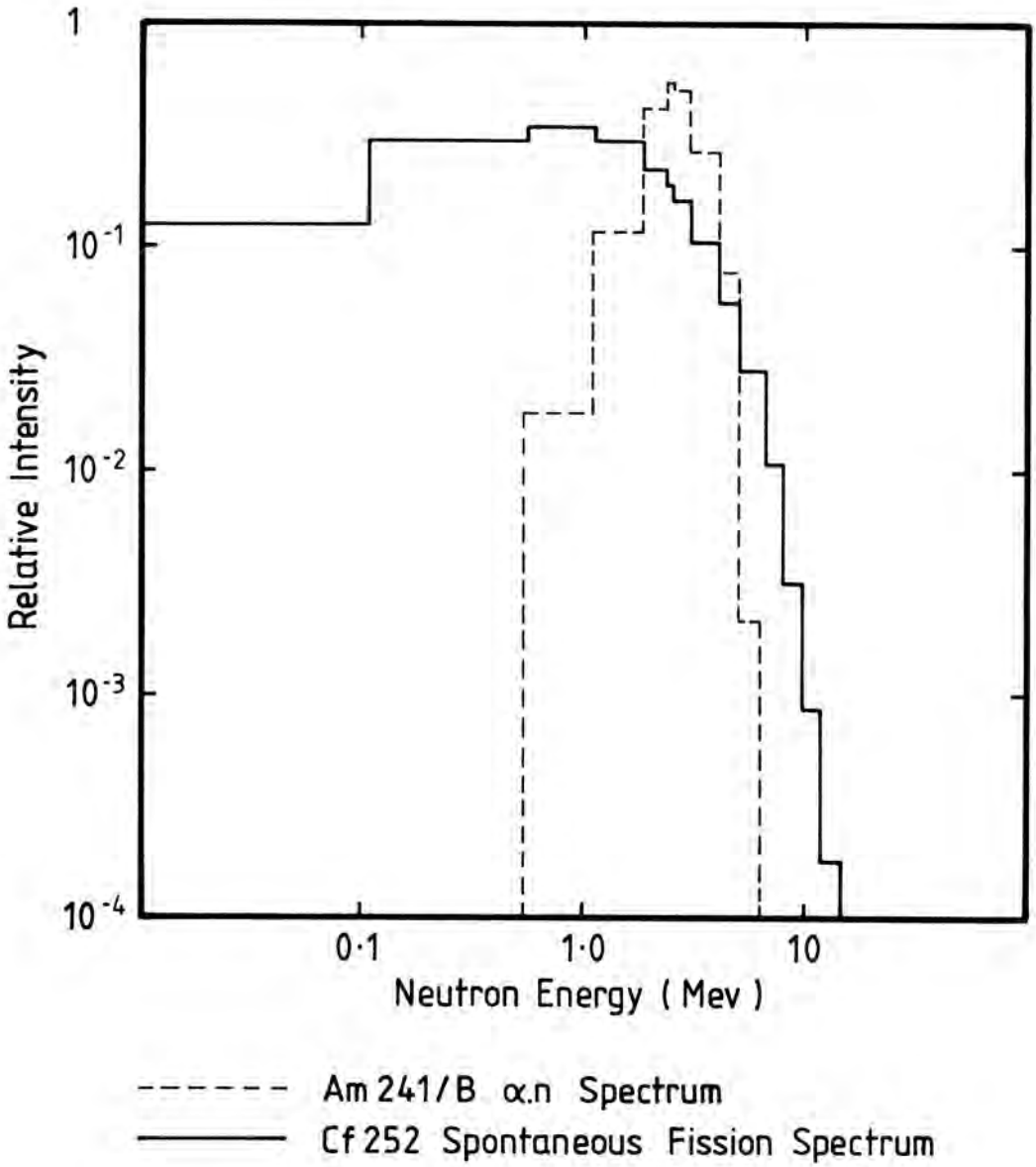

Fig. 2. Comparison of Neutron Spectra.