(α,n) NEUTRON SOURCE LEVELS IN DHLW BOROSILICATE GLASS

B. H. Weren, N. L. Savin
WESTINGHOUSE ELECTRIC CORPORATION
Waste Technology Services Division
Madison, Pennsylvania 15663-0286

Abstract

This paper describes the methods used and results obtained in calculations of the neutron source levels in Defense High Level Waste (DHLW). It was found that the the potential contribution of (α,n) reactions to the total neutron source in borosilicate glass waste forms was significant enough to require explicit calculation and that this source needed to be accounted for in the design of radiation shielding for waste handling equipment.

Previous work, [1] presented preliminary estimates of (α,n) neutron source levels associated with high level waste in borosilicate glass. Based on these estimates made for Commercial High Level Waste (CHLW), three conclusions were drawn: 1. The potential contribution of the (α,n) reactions to the total neutron source in borosilicate glass waste forms is sufficiently significant to require explicit calculations; 2. Development is needed of techniques to perform, more rigorously, these non-trivial calculations; and 3. Experimental verification of the significance of the (α,n) neutron contribution and of the calculational techniques developed should be undertaken.

The purpose of this paper is twofold: 1. to report on the significant improvements that have been made in calculational techniques; and 2. to report the results of the application of these techniques to the calculation of neutron source levels associated with Defense High Level Waste (DHLW) borosilicate glass.

There are actually two types of reactions which contribute to the neutron source levels in borosilicate glass. These are spontaneous fission of certain actinides and (α,n) reactions with the light elements in the glass (most notably boron, sodium and oxygen). The alpha particles result from the decay of actinides in the waste. Methods for calculating the neutron source from spontaneous fission are well established. However, because there is a lack of historical data for comparison with the (α,n) calculations, two independent methods were used to determine this source, the first being use of the SOURCES computer code presently under development at Los Alamos National Laboratory, [2a] and the second, hand calculations as described by D. West in Reference 3.

Many of the actinides in DHLW decay by emission of alpha particles. These alpha particles are slowed down by various components of the glass/waste mixture. Some elements (typically light elements) give off neutrons when they absorb an alpha particle. There are significant amounts of these light elements in borosilicate glass; for example, boron, sodium and oxygen. These reactions are typically threshold reactions, that is, requiring alpha particles of a minimum energy level to initiate the reaction. The energy spectrum of the emitted neutrons depends on the particular light "target" element and the energy of the incident alpha particle. To obtain the correct proportion of the reactions which occur with each light target (or with elements which do not yield neutrons) it is necessary to account for the slowing down of the alpha particles in some realistic way. For these calculations, the DHLW glass was assumed to be a uniform mixture of the elements shown, by weight percent and atom fraction, in Table I. If the waste/glass mixture were not uniform and instead contained "large" particles of waste, then this model would overpredict the number of alpha particles which would interact with the light elements of the glass. However, it is expected that the waste particle sizes will be considerably less than the range of alpha particles in the heavy elements; thus the assumption of homogeneity should result in a realistic calculation of the magnitude of the neutron source.

Three sets of data are needed to input into the SOURCES code: 1. alpha sources, in this case, twenty-four actinides from U^{232} through Cm^{248}, as shown in Table II; 2. "stoppers", that is all of the elements in the mixture as shown in Table I; and 3. "targets" or light elements which undergo (α,n) reactions, in this case, the appropriate isotopes of

TABLE I

Elemental Composition of DHLW in SOURCES Code

Element	Weight Percent	Atom Fraction
Li	1.92	0.0546
B	3.29	0.0601
O	45.77	0.5652
Na	13.05	0.1121
Mg	0.96	0.0078
Al	1.77	0.0130
Si	21.08	0.1483
Ca	10.89*	0.0419*
Th	0.20	0.00017
U	1.06	0.00088
Pu	0.01	0.00001

[a]The invaluable and timely assistance of W. B. Wilson of Los Alamos National Laboratory in providing and updating the SOURCES code and its data libraries is gratefully acknowledged.

*Approximately 10 weight percent (or 0.034 atom fraction) of the glass is iron and a few other similar elements which are represented as calcium since iron and the other trace elements are not included in the SOURCES data library.

boron, lithium, oxygen, sodium, magnesium, aluminum and silicon, also shown in Table II. The code contains four data libraries which it uses in conjunction with the input data to calculate the magnitude and in some cases the energy spectrum of the resulting neutron source. The four libraries include: 1. a decay scheme library which defines the spontaneous fission and alpha particle yields of the various actinides; 2. a stopping cross section library used to calculate the slowing down of the alpha particles; 3. a library of (α,n) reaction cross sections for the light element targets; and 4. a library of neutron energy spectrum data.

Using the input data previously described, an (α,n) neutron source of 198 neutrons/cc/sec in DHLW was calculated using the SOURCES code. Of these, 178 resulted from interactions with alpha particles resulting from decay of Pu^{238}, 17 from decay of Cm^{244} and the remaining three from the other alpha emitters. According to these calculations, approximately 49 percent of the neutrons resulted from reactions with sodium, 42 percent from reactions with boron and the remaining 9 percent from the other target elements.

To confirm these results, the calculation of the (α,n) neutron source was repeated using the method of D. West in Reference 3 for calculating neutron yields in mixtures from the thick target (α,n) yields of the separate constituents. Briefly, this method involves four steps: 1. choosing a reference target element and alpha energy; 2. calculating, for each element in the mixture, the relative stopping power compared to the reference target using one of the several tabulations of stopping power by element; 3. weighting the measured thick target yield for each element by both the relative stopping power and the ratio of the density of the element in the mixture to the density in the thick target; and 4. summing the yields over all targets.

TABLE II

Actinide and Light Element Input to SOURCES Code

Actinide	Source Density (atoms/cc)	Light Element	Target Density (atoms/cc)
U^{232}	3.226+13*	Li^{7}	3.817+21
U^{233}	8.620+12	B^{10}	9.158+20
U^{234}	3.496+17	B^{11}	3.663+21
U^{235}	3.335+18	O^{17}	1.679+19
U^{236}	2.442+18	O^{18}	8.825+19
U^{238}	1.422+20	Na^{23}	8.543+21
Np^{236}	1.345+02	Mg(nat)	5.937+20
Np^{237}	5.798+16	Al^{27}	9.890+20
Pu^{236}	1.044+12	Si^{29}	5.306+20
Pu^{238}	3.964+17	Si^{30}	3.500+20
Pu^{239}	7.207+17		
Pu^{240}	1.762+17		
Pu^{241}	7.342+16		
Pu^{242}	1.393+16		
Am^{241}	1.434+16		
Am^{242}	8.016+07		
Am^{242m}	6.667+12		
Am^{243}	1.347+14		
Cm^{242}	4.800+10		
Cm^{243}	5.459+11		
Cm^{244}	5.866+15		
Cm^{245}	1.684+11		
Cm^{246}	7.668+09		
Cm^{248}	7.335+05		

*Read as 3.226×10^{13}

In applying the West method to our calculation, aluminum was assumed to be the reference target element (because tabulated data were not available for boron or sodium). Relative stopping powers were calculated assuming an alpha particle energy of 5 Mev (representative of Pu^{238} alphas). The relative stopping powers were calculated from Northcliffe and Schilling [4]. Measured thick target yields were obtained from various sources for boron, sodium, lithium, oxygen, silicon, magnesium and aluminum. The magnitude of the alpha source, approximately 1.07×10^{8} alphas/cc/sec, was calculated using the ORIGEN computer code [5].

Using this method, a neutron source of 124 neutrons/cc/sec in the DHLW was calculated. Of this, 89 neutrons/cc/sec were from (α,n) reactions with boron (compared to 84 calculated with the SOURCES code), 20 neutrons/cc/sec from sodium (compared to 97 from SOURCES), and 15 neutrons/cc/sec from the other targets (compared to 17 predicted by the SOURCES code). With the exception of the sodium contribution, the results of the two methods showed excellent agreement. Upon further investigation, it was found that the sodium reaction data used in the SOURCES code are preliminary and that in fact calculations [6] performed using those data overpredicted measured sodium thick target yield values by approximately a factor of four. A factor of four is approximately the difference between the SOURCES and "West method" results.

The spontaneous fission neutron source levels were calculated using both the SOURCES code and the ORIGEN code, with excellent agreement between the two methods. The ORIGEN code predicted approximately 27.5 neutrons/cc/sec and SOURCES predicted 26.3 neutrons/cc/sec from spontaneous fission reactions, almost entirely from Cm^{244}. The total neutron source in DHLW (spontaneous fission plus (α,n) neutrons) is then estimated to be in the range of 152 neutrons/cc/sec, based on the "West method", to 225 neutrons/cc/sec using the SOURCES code.

To determine the significance of this source on radiation shielding requirements, neutron radiation levels were calculated for both an unshielded DHLW canister and for a conceptual design of a handling cask for DHLW. The one dimensional transport code, ANISN-W, [7] was used to perform the radiation shielding calculations. The geometries evaluated are shown in Fig. 1. The BUGLE-80 47 neutron energy group cross section library was used [8]. These calculations utilized a P_3 scattering cross section approximation, an S_6 order of angular quadrature and ANSI N666 dose conversion factors. The resulting neutron dose rates were then multiplied by a factor of two to account for an anticipated increase in fast neutron quality factors. One advantage of the SOURCES code is that it permits editing of the energy spectra of the calculated neutron source according to a user specified energy structure. At the time these calculations were performed, no data were included in the SOURCES library for energy spectra of neutrons emitted from (α,n) reactions with sodium. Based on Ref. 9, it is believed that the sodium (α,n) neutron energy spectrum is similar to lithium, although, probably harder than lithium. Therefore, two cases were considered, one in which the "sodium neutrons" were assumed to have a hard spectrum like boron, and a second in which the lithium spectrum was used. It is believed that use of these two spectra will bound the actual value of the sodium contribution.

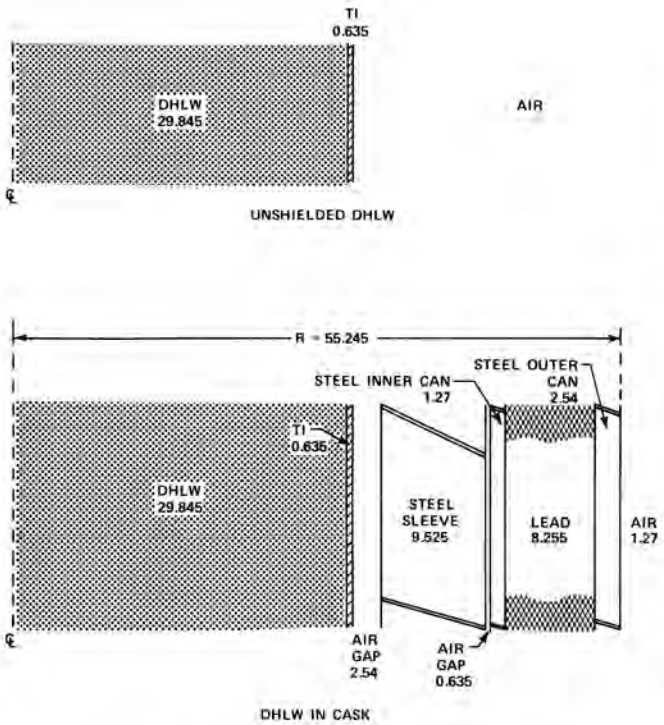

Figure 1. ANISN Geometries (All Dimensions In Centimeters).

The ANISN calculations were actually repeated four times, two cases with the different spectra using the SOURCES calculated neutron source levels, and two cases with the different spectra using the "West method" source levels. The calculated neutron radiation levels at the surface of the unshielded DHLW canister were: 760 mRem/hr using the SOURCES values and a boron spectrum for the "sodium neutrons"; 553 mRem/hr using the SOURCES data and the lithium spectrum; 507 mRem/hr using the "West method" data and the boron spectrum and finally; 456 mRem/hr using the "West method" data and the lithium data. As stated previously, it is believed that the data used with the SOURCES code overpredicted the sodium (α,n) neutron yield, and therefore, the "best estimate" of the neutron radiation level associated with an unshielded DHLW canister is approximately 500 mRem/hr. The same four cases were repeated for the cask geometry. The resulting neutron dose rates on the surface of the cask ranged from 125 mRem/hr to 74 mRem/hr, with a "best estimate" value of approximately 85 mRem/hr. No method uncertainty or design assurance factors have been included in any of these values. The design radiation levels for the cask were a total surface dose rate of 50 mRem/hr and a neutron dose rate of 10 mRem/hr. These calculations obviously indicated the need for the addition of neutron shielding material to the cask design.

The results of the two methods of neutron source calculations are judged to show very good agreement with the major uncertainties being the sodium target cross sections and yields, and the sodium (α,n) neutron energy spectrum. It is also noted again that the assumption of the homogeneity of the waste/glass mixture is critical but thought to be realistic. Both of the methods used in these calculations are significantly more detailed and believed to provide more realistic values than those used in Ref. 1 to estimate the CHLW neutron levels. These calculations definitely confirm the significance of the (α,n) contribution to the neutron source levels in borosilicate glass waste forms. It is clear that these sources must be accounted for in the design of radiation shielding for the handling and transportation of this waste. An experimental program is in progress at the Savannah River Plant to confirm the magnitude of the neutron source levels in DHLW. Additional development of cross section libraries and neutron energy spectra is still needed, and additional experimental confirmation of the data and calculational methods is certainly desired.

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