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REACTIONS IN PLUTONIUM
COMPOUNDS CALCULATED FROM
HAUSER-FESBACH REACTION THEORY

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NEUTRON SPECTRA FROM (α ,n) REACTIONS
IN PLUTONIUM COMPOUNDS CALCULATED
FROM
HAUSER-FESHBACH REACTION THEORY

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ABSTRACT

A sequence of computer programs was developed to calculate the yield and energy spectrum of neutrons from (α, n) reactions in materials having an alpha emitter in intimate contact with an (α, n) reacting nuclide. Alpha particle fluxes were calculated from emitter concentrations, alpha decay lifetimes, and charged particle stopping power of the material medium. Experimental (α, n) total cross sections were used where available, with branching ratios to final excitation states and angular distributions calculated from Hauser-Feshbach theory and optical model penetrabilities. The neutron spectrum in PuO_2 and PuC with natural ^{17}O , ^{18}O , and ^{13}C isotopic composition was calculated for plutonium isotopes 238, 239, and 240.

INTRODUCTION

The energy of typical alpha particles emitted by high Z isotopes is 4 to 5.5 MeV. Their range is on the order of 4 cm in air and about 10^{-3} to 10^{-4} times that distance in solids. Hence, most alpha particles emitted in solids by alpha decay come to rest in the same solid unless its dimensions are on the order of a millimeter or less. The alpha particle loses energy primarily to electrons by Coulomb collisions, with large numbers of electrons along the track receiving an energy on the order of a few electron volts each. Hence, the slowing down of the alpha particle is essentially continuous with energy lost per unit of path length a function of the instantaneous energy. The great majority of the alpha particles slow to rest in the matter, picking up electrons to become ordinary helium atoms.

In materials with certain light nuclides in intimate contact with the alpha emitter, however, the alpha particles can undergo a nuclear reaction instead of merely slowing to rest. Typically, on the order of one alpha in 10^5 will experience such a reaction in compounds involving an element some of whose isotopes are favorable for (α, n) reactions. These reactions are more probable from (1) nuclides with low charge ($z \lesssim 20$) so that alpha particles can penetrate, and (2) nuclides having a sufficiently high reaction energy ($Q_0 \gtrsim 3$ MeV) that the reaction can go at available alpha energies. Some nuclides meeting these requirements and their reactions and maximum reaction energies Q_0 are:

The oxides of plutonium and plutonium nitrates in water solution will produce (α ,n) reaction neutrons by virtue of the traces of ${}^{17}\text{O}$ and ${}^{18}\text{O}$ present (.037% and .204%, respectively) in the oxygen, while plutonium carbides will produce (α ,n) reaction neutrons from the ${}^{13}\text{C}$ present as 1.1% of the carbon.

The object of the present study was to calculate the spectra and yields of (α ,n) neutrons to confirm and supplement direct measurements. We have attempted to (1) compute a realistic flux spectrum of alpha particles based on experimental stopping power, (2) use available (α ,n) experimental cross sections, and (3) calculate branching ratios to final excitation states and center of mass angular distributions of emitted neutrons directly from theory. The calculated neutron spectra and yields are intended to confirm experimental spectra and yields observed with plutonium compounds to be used in fast reactor fuels.

Reaction Kinematics and Counting

Given a flux of alpha particles $\Phi(E_\alpha) \Delta E_\alpha$ in a narrow energy band E_α to $E_\alpha + \Delta E_\alpha$ in a material having number density n_T of target atoms whose cross section is $\sigma_T(E_\alpha)$ for (α ,n) reaction with alphas of energy E_α , the number of reactions per unit volume per second will be

$$\Delta \dot{N} = \phi(E_\alpha) \Delta E_\alpha n_T \sigma_T(E_\alpha). \quad (1)$$

The energy of the emitted neutron, however, is a function of the angle at which it is emitted in the center of mass frame and of the reaction energy Q for the particular reaction and residual nucleus excitation state. The neutron center of mass (CM) energy E_n^O is unique for a given alpha laboratory energy E_α , given target species, and given residual nucleus excitation state:

$$E_n^O = \frac{m_F}{m_F + m_n} \left(\frac{m_T}{m_T + m_\alpha} E_\alpha + Q \right). \quad (2)$$

Here m_n , m_α , m_T , and m_F are neutron, alpha, target nucleus, and residual nucleus masses, respectively. The corresponding neutron laboratory energy E_n is a function of CM angle θ :

$$E_n = E_n^O + \frac{m_n m_\alpha}{(m_\alpha + m_T)} E_\alpha + 2 \frac{(m_n m_\alpha)^{1/2}}{m_\alpha + m_T} (E_n^O E_\alpha)^{1/2} \cos \theta. \quad (3)$$

Here CM angle θ is measured from the direction of the incident alpha in the laboratory frame. From relation (3) we can see that the neutrons from the (α, n) reaction that are produced with laboratory energies E_n between E_n^1 and E_n^2 for a given target and reaction energy Q are those emitted with CM angle between θ_1 and θ_2 , where $\cos \theta_1$ is the value of $\cos \theta$ satisfying relation (3) when E_n has the value E_n^1 . $\cos \theta_2$ and E_n^2 are similarly related.

The number of neutrons produced per unit volume per unit time in a neutron energy range from E_n^1 to E_n^2 is calculated by program NSPEC as

$$\int_{E_n^1}^{E_n^2} N(E_n) dE_n = 2\pi \sum_{\{T, Q\}} n_T \sum_i \Delta E_\alpha^i \int_{\cos \theta_1}^{\cos \theta_2} \phi(E_\alpha^i) \frac{d\sigma_T^Q(E_\alpha^i, \theta)}{d\Omega} d(\cos \theta). \quad (4)$$

Here $N(E_n)$ is the number of neutrons produced per unit volume per unit range of neutron energy E_n , and Q is the reaction energy for transition to a particular residual nucleus excitation state. $\Phi(E_\alpha^i)$ is the alpha flux per unit energy at alpha laboratory energy E_α^i , while ΔE_α^i is the energy width of the i th flux interval. The number of (α, n) reacting nuclides per unit volume of target species T is n_T , while $d\sigma_T^Q(E_\alpha^i, \theta)/d\Omega$ is the differential cross section for emission of the neutron into CM solid angle $d\Omega$ at CM angle θ from initial α direction from an (α, n) reaction on target species T with residual nucleus being left in a state corresponding to reaction energy Q . The integration limits $\cos \theta_1$ and $\cos \theta_2$ are obtained by substituting the lower and upper neutron energy limits E_n^1 and E_n^2 , respectively, into relation (3) with E_α set to E_α^i in relation (3) and relation (2) for the i th alpha energy interval. In our calculations we have typically used neutron energy intervals $E_n^2 - E_n^1 = .05$ MeV. The alpha energy interval size has been primarily determined by the requirement that we reasonably represent the alpha energy integral which we are approximating by a sum. The alpha particle flux is a smooth function of energy from 2 MeV up to the ~ 5 MeV alpha birth energies; by contrast, the (α, n) cross sections have intricate structure with $\sim 10^{-2}$ MeV width.

NSPEC requires differential angular cross sections for each residual nucleus excitation state. NSPEC is currently accepting this information as a set of Legendre coefficients W_{TL}^Q , where

$$\frac{d\sigma_T^Q}{d\Omega} = \sum_L W_{TL}^Q P_L(\cos \theta) \quad (5)$$

For target nucleus and alpha energy such that only the ground state of the residual nucleus in (α, n) reactions is energetically accessible, the Legendre coefficients for the neutron angular distribution frequently will have been determined experimentally. This is the case for $^{13}\text{C}(\alpha, n)^{16}\text{O}$ for α energies below 5 MeV.

When multiple excitation states of the residual nucleus are energetically accessible, the branching and angular distribution information is generally not available. For these cases we used Hauser-Feshbach theory and the optical model to calculate the coefficient array W_{TL}^Q in program HAU. Some manipulation of optical model parameters was performed to improve agreement of calculated (α, n) total cross sections with experimental totals. The SCALE code was then used to scale the calculated Legendre coefficients (and hence partial cross sections) to give agreement with experimental totals. The averaging in energy of experimental cross sections is reasonable because: (1) the optical model is expected to produce energy-averaged cross sections without resonances, (2) the neutron spectrum is reasonably determined by the averaged cross sections and is somewhat insensitive to intricate resonance structure, and (3) smoothing saves computer time by allowing use of fewer energy points.

NSPEC Operations

The function of the NSPEC code is to read parameters describing the material medium and the alpha particle production rates, to calculate the alpha flux in the material, to read the array W_{TL}^Q of Legendre coefficients describing the angular differential partial cross sections to final states by (α, n) processes, and then to compute neutron production per unit volume in an array of discrete neutron energy intervals according to relation (4).

NSPEC reads the set EBIRTH(I) of alpha particle birth energies and the corresponding rates NDOT(I) of alpha emission in number per unit volume per second. The user supplies this information according to the alpha emitters present. NSPEC reads a set Z(I) of atomic numbers of the elements present in the material medium and the corresponding mass fractions FMASS(I). The flux of alphas per unit energy range at energy E is given by

$$\phi(E) = \frac{\sum_I \dot{n}_{\alpha I}}{\left(\frac{dE}{ds}\right)_E}, \quad (6)$$

where $\sum_I \dot{n}_{\alpha I}$ is the sum of the rates of alpha emission per unit volume at alpha energies higher than E. The energy loss per unit path length dE/ds is calculated as

$$\left(\frac{dE}{ds}\right)_E = \sum_i \rho_i \frac{dE}{dM_i}, \quad (7)$$

where dE/dM_i is the energy loss per unit (mass/area) of the i th element of the material medium traversed by the alphas. We approximate that the energy lost per unit (mass/area) traversed of each material is independent of the density and of the presence of other elements. SUBROUTINE FLUX calculates alpha flux ϕ at a discrete set of energy points, obtaining the dE/dM_i information from a table lookup. A table of stopping powers¹ dE/dM_i for alphas was built into the subroutine for 38 energies from .05 to 48 MeV and for 14 elements from beryllium ($Z = 4$) to uranium ($Z = 92$). Stopping powers for an arbitrary element and energy are obtained by first quadratically interpolating on energy for three adjacent Z values, then quadratically interpolating on atomic number.

With alpha particle flux calculated as a function of energy, NSPEC reads in a target number density n_T followed by a reaction energy Q for one final state, a set of alpha particle energies, and a set of Legendre coefficients $W_{TL}^Q(E_\alpha)$ describing the angular differential partial cross section at each alpha energy. The energy intervals in the cross section information read in determines the size of the interval ΔE_α^i for which $\phi(E_\alpha^i) \Delta E_\alpha^i$ is numerically computed. The alpha flux is numerically summed in an energy interval halfway to the next lower and next higher cross section energy, except at the lowest and highest energy cross section read in for a given

T and Q. The integration of the Legendre polynomial between the two values of $\cos \theta$ corresponding to lower and upper edges of the neutron energy bucket is performed by evaluating the analytical indefinite integral of the Legendre polynomial at the upper and lower limits.

Program HAU Operations

The computation of the array W_{TL}^Q of Legendre coefficients describing the partial cross sections is the function of Program HAU. Program HAU uses the Hauser-Feshbach theory of those nuclear reactions which proceed through compound nucleus states, with transmission coefficients calculated on the basis of the nuclear optical model.

The Hauser-Feshbach model² is a statistical model which assumes that, at the excitation energy of the compound nucleus, there are many levels of all types. That is, in an energy interval of interest to the experiment, many compound nucleus levels can be excited. The interferences between the values of orbital angular momentum ℓ which contribute to compound nucleus formation and those values ℓ' which contribute to the decay of a compound nucleus angular momentum J and parity π are neglected. In this approximation, the only information retained from the initial state by the compound nucleus is total angular momentum J (which will be compound nuclear angular momentum), parity π , total energy, and of course the rigorously conserved quantum numbers like charge and baryon number.

The cross section for formation of a compound nucleus by an incident particle of relative orbital angular momentum ℓ is given in the optical model as

$$\sigma_{\ell}^R = (2\ell + 1) \pi k^{-2} T_{\ell}(E) \quad (8)$$

where k is the relative wave number (equal to $p/\hbar$, where p is relative momentum and $\hbar$ is Planck's constant divided by 2π) and E is center of mass energy of the initial two body (target nucleus and incident particle) system. The transmission coefficient $T_\ell(E)$ is the ratio of the probability of absorption of an ℓ wave particle to the maximum quantum mechanically allowed probability and has in it the information about the interaction of the initial nucleus and incident particle. Parenthetically, one black nucleus model has maximum quantum mechanically possible absorption of incident particles in orbital angular momentum states up to some cutoff value ℓ_c , that is $T_\ell(E) = 1$ for $\ell \leq \ell_c$, $T_\ell = 0$ for $\ell > \ell_c$.

The cross section for formation of a compound nucleus of total angular momentum J by particles with relative orbital angular momentum ℓ is taken as σ_ℓ^R multiplied by the probability that ℓ and the channel spin j combine vectorially to give J , where the probability is taken purely from angular momentum basis algebra:

$$\sigma_\ell^R (c_{\ell j J}^{o m m})^2 = (2\ell + 1) \pi k^{-2} T_\ell(E) (c_{\ell j J}^{o m m})^2. \quad (9)$$

The channel spin j is the vector sum of initial nuclear spin i and incident particle spin I , and m is the projection of j on the initial relative momentum. The quantum numbers j , ℓ , and J correspond to the angular momenta $\underline{j}$, $\underline{\ell}$, and $\underline{J}$. The indexed c 's are Clebsch-Gordan coefficients.

The branching to final states is then calculated assuming time reversal invariance on a similar model. The probability of a given final partition of particles, energy state E' , channel spin j' , and orbital angular momentum ℓ' from a compound nucleus of given J and parity π is taken as proportional to the probability of formation of the compound nucleus with given J and π from the time reversed final state. We used the differential angular cross section

form given by McCarthy³ for transition to a two-body final state with quantum numbers α' with center of mass angle θ from an initial state with quantum numbers α :

$$\begin{aligned} \sigma(\alpha|\alpha';\theta) = & \sum_L (4k^2)^{-1} \sum_{j\pi} (2I+1)^{-1} (2i+1)^{-1} \sum_{\ell j} T_\ell(\alpha) \\ & \times \sum_{\ell' j'} \frac{T_{\ell'}(\alpha')}{\sum_{\lambda} T_{\lambda}(\alpha')} Z(\ell j \ell J, j L) Z(\ell' j' \ell' J, j' L) \\ & \times (-1)^{j-j'} P_L(\cos \theta) \end{aligned} \quad (10)$$

Here i and I are initial nucleus and incident particle intrinsic spins, respectively. The denominator summation $\sum_{\lambda} T_{\lambda}(\alpha')$ runs over all two body final states consistent with a given J , π , and incidence CM energy. The quantum numbers lumped together under the sets α' are the internal quantum numbers of the two final bodies and their CM kinetic energy. The Z coefficients are related to the Racah coefficients by

$$Z(\ell_1 \ell_2 J_2, sL) = \{(2\ell_1+1)(2\ell_2+1)(2J_1+1)(2J_2+1)\}^{1/2} c_{\ell_1 \ell_2 L}^{\circ \circ \circ} W(\ell_1 J_1 \ell_2 J_2, sL). \quad (11)$$

The Racah coefficients can be generated from one of a number of standard expressions. The Racah coefficient expression used on our calculations is that given by Rose⁴, Equation (6.7), p. 110.

The form above given by McCarthy for the differential angular compound nucleus cross sections was selected as most convenient for our purposes, in that it gives directly the Legendre coefficients previously called W_{TL}^0 for the two body states of interest. They are the coefficients of $P_L(\cos \theta)$ for the final states α' which are states of neutron plus residual nucleus in energetically

accessible excitation state. In the (α, n) reaction calculations, one must supply the information about all energetically allowed states accessible from probable compound nucleus states, including alpha particle plus nucleus states.

A limitation of the Hauser-Feshbach reaction theory stems from the assumption of no interference between incoming and outgoing orbital angular momentum states. Only moduli squared of spherical harmonics will appear in the angular distribution (no cross terms), which implies that only even order Legendre polynomials appear in the angular distribution. Hence the calculated angular distribution in the center of mass system will be symmetric about 90° . Deviation from Hauser-Feshbach predictions frequently appears as a forward peaking of the cross section.

The transmission coefficients $T_\ell(E)$ for insertion into the Hauser-Feshbach expression were calculated by numerically solving by fourth order Runge-Kutta techniques the differential equation for the radial wave function. The potential was taken as spin dependent and of the form

$$\begin{aligned}
 V(r) = & V_0 \frac{1}{1 + e^{(r-r_a)/a}} + V_{so} \frac{s \cdot \ell}{\hbar} \left(\frac{\hbar}{m_\pi c} \right)^2 \frac{1}{r} \frac{d}{dr} \frac{1}{1 + e^{(r-r_a)/a}} \\
 & + V_c(r) \\
 & + i \left(W_0 (x * \frac{1}{1 + e^{(r-r_a)/a}} + (1-x) e^{-\frac{(r-r_b)^2}{b^2}} \right) \\
 & + W_{so} \frac{s \cdot \ell}{\hbar} \left(\frac{\hbar}{m_\pi c} \right)^2 \frac{1}{r} \frac{d}{dr} \frac{1}{1 + e^{(r-r_a)/a}} \quad (12)
 \end{aligned}$$

The Coulomb potential $V_c(r)$ is taken as

$$V_c(r) = \begin{cases} z_1 z_2 e^2 / r & r > r_a \\ z_1 z_2 e^2 (3 - (r/r_a)^2) / 2r & r \leq r_a \end{cases} \quad (13)$$

The parameters describing the interaction are nuclear potential ranges r_a and r_b , nuclear surface thickness parameters a and b , well depths V_a , W_a , V_{so} , and W_{so} , and division factor x giving relative weights of volume and surface contributions to the imaginary part of the potential. The "centrifugal barrier" term provides an additional effective potential term in the radial wave function for a given ℓ . The step size in the radial wave function integration was incremented through five values in integrating from the low r asymptotic form of the regular wave function to an r value effectively beyond the nuclear potential range in about 200 steps. The transmission coefficient T_ℓ is determined from the coefficient c_ℓ , which is a ratio of combinations of regular and irregular Coulomb wave functions in the large r region. Continuity of logarithmic derivative is imposed:

$$\frac{U_\ell^+}{U_\ell^-} = \frac{F_\ell^- + c_\ell^+ (G_\ell^- + iF_\ell^-)}{F_\ell^+ + c_\ell^- (G_\ell^+ + iF_\ell^+)} \quad (14)$$

Here U_ℓ^+ and U_ℓ^- are the radial wave function and its derivative at the connection point for $j = \ell + \frac{1}{2}$ (+) and $j = \ell - \frac{1}{2}$ (-) for neutrons. For alpha particle states, the incident or outgoing particle spin is zero, channel spin j is equal to orbital angular momentum ℓ , and the $\pm$ labels are not relevant. The functions F_ℓ and G_ℓ are the regular and irregular Coulomb wave functions, which reduce to the Riccatti-Bessel functions for neutron states. The transmission coefficient T_ℓ is then found from^[5]

$$T_\ell = \frac{4\ell}{2\ell + 1} [\text{Im } c_\ell^- - |c_\ell^-|^2] + \frac{4(\ell + 1)}{2\ell + 1} [\text{Im } c_\ell^+ - |c_\ell^+|^2] \quad (15)$$

for neutron states, and

$$T_\ell = 4(\text{Im } c_\ell - |c_\ell|^2) \quad (16)$$

for alpha particle states.

SCALE Operations

The calculated array of Legendre coefficients for neutron final states from program HAU should hopefully produce a cross section bearing some relation to the actual (α, n) cross sections obtainable from alpha beams on thin targets. In practice an automated search for optical model parameters to produce even the total (α, n) cross section would require large amounts of computer time. Hence the procedure used was to find plausible optical model parameters which produced rough qualitative features of the total (α, n) cross section, then to scale all the Legendre coefficients according to a factor which gives the smoothed experimental total:

$$W_{TL}^Q(E_\alpha)' = \frac{\bar{\sigma}_T(E_\alpha)}{4\pi \sum_Q W_{TO}^Q(E_\alpha)} W_{TL}^Q(E_\alpha). \quad (17)$$

Here $W_{TL}^Q(E_\alpha)'$ is the scaled Legendre coefficient while $W_{TL}^Q(E_\alpha)$ is the Legendre coefficient calculated by HAU. The averaged cross section $\bar{\sigma}_T(E_\alpha)$ is obtained by integrating the experimental thin target (α, n) cross sections over an interval and replacing the resonance resolved cross sections by a polygraph having the same area under the curve. The denominator expression

$$4\pi \sum_Q W_{TO}^Q(E_\alpha) \quad (18)$$

gives the total (α, n) cross section at energy E_α as calculated by HAU. The summation runs over those final states having a neutron which are accessible from target T by (α, n) reaction with an alpha of energy E_α .

The smoothing of the experimental cross sections before scaling the HAU output Legendre coefficients was done to allow use of a courser alpha energy mesh from program HAU. Program HAU Legendre coefficients were generated at typically about 40 energy values in the 1-6 MeV range. Experimental cross sections were reduced to a polygraph with each rectangle centered at an alpha energy point at which HAU had generated its partial cross section Legendre coefficients.

PuO₂ Calculations

The contributing nuclides to (α, n) reactions in PuO₂ are ¹⁷O and ¹⁸O, which are .037% and .204%, respectively, of natural oxygen. The cross section information from HAU was generated using the spin, parity, and energy level assignments diagrammed in Figure 1 for ¹⁷O, ¹⁸O, ²⁰Ne, and ²¹Ne.

For ¹⁷O(α, n)²⁰Ne the initial state has

$$^{17}\text{O} \text{ ground state spin } i = 5/2$$

$$^{17}\text{O} \text{ ground state parity} = +$$

$$^4\text{He} \text{ spin } I = 0$$

$$^4\text{He} \text{ parity} = +$$

While energy levels, spins, and parities of other ¹⁷O states accessible with 5.5 MeV alphas by (α, α') interaction are

$$.871 \text{ MeV} \quad \frac{1}{2} \quad +$$

$$3.06 \text{ MeV} \quad \frac{1}{2} \quad -$$

$$3.85 \text{ MeV} \quad \frac{5}{2} \quad -$$

To determine what excitation states of ²⁰Ne are energetically accessible from ¹⁷O(α, n)²⁰Ne, we first compute the reaction energy Q_0 for the ground state from the mass excesses given in Table of Isotopes⁶:

$$\begin{aligned} Q_0 &= \Delta_{\text{He}} + \Delta_{^{17}\text{O}} - \Delta_n - \Delta_{^{20}\text{Ne}} \\ &= 2.4248 + (-.808) - 8.074 - (-7.042) \\ &= .5848 \text{ MeV} \end{aligned}$$

The (neutron plus ^{20}Ne) states energetically accessible from $^{17}\text{O}(\alpha, n)^{20}\text{Ne}$ with alphas of laboratory energy E_α are those for which

$$\frac{m_{^{17}\text{O}}}{m_{\text{He}} + m_{^{17}\text{O}}} E_\alpha + Q_0 - E_x > 0 .$$

Here E_x represents ^{20}Ne excitation energy measured from ground state. With $E_\alpha = 5.5$ MeV, the ^{20}Ne states (energy, spin, and parity) accessible are

0 MeV	0+
1.63 MeV	2+
4.25 MeV	4+
4.97 MeV	2-

These states with their spins, parities, and Q values were supplied to HAU as competing final states.

The potential parameters for the interaction of the alpha particles with the ^{17}O compound nucleus were based on those found by Satchler⁷ for interaction of alphas with a variety of nuclei at 28 MeV. The imaginary part of the potential had to be drastically reduced to produce reasonable cross sections for the (α, n) processes. The parameter values arrived at in the potential expressions (12) and (13) for the interaction of alphas with ^{17}O were

$$V_0 = 50. \text{ MeV}$$

$$W_0 = 4. \text{ MeV}$$

$$x = 0.$$

$$r_a = 1.2 A^{1/3} + 1.3 \text{ Fermis} \quad A = 17$$

$$r_b = r_a$$

$$a = .55 \text{ Fermis}$$

$$b = .48 \text{ Fermis}$$

The parameters for the interaction of the neutrons with the residual ^{20}Ne were based on a variety of sources, including fits by Holmquist and Wiedling⁸ to neutron elastic scattering on Al and S at 4 MeV. Parameters used were

$$V_o = 47.9 \text{ MeV}$$

$$W_o = 7.35 \text{ MeV}$$

$$V_{so} = 8.00 \text{ MeV}$$

$$W_{so} = 0.$$

$$a = .72 \text{ fermis}$$

$$b = .48 \text{ fermis}$$

$$r_a = r_b = 1.13 A^{1/3} \text{ fermis} \quad A = 20$$

$$x = 0.$$

The resulting (α, n) cross sections show rather poor agreement with experiment. Because the contribution of ^{17}O to the neutron yields is much less than that of ^{18}O , this was not considered serious. Scaling (α, n) partial cross sections to agree with the experimental total (α, n) cross sections should, of course, correct the contribution to the yield. The calculated $^{17}\text{O}(\alpha, n)^{20}\text{Ne}$ cross sections are shown in Figure 2.

Somewhat better agreement of the calculated (α, n) cross sections with experimental values was obtained for $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$. The ground state of ^{18}O is a $0+$ state. States of ^{18}O accessible by (α, α') reaction with 5.5 MeV alphas have energies, spins, and parities given by

1.98 MeV 2+

3.55 MeV 4+

3.63 MeV 0+

3.92 MeV 2+

4.45 MeV 1-

States of ^{21}Ne accessible by $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ with 5.5 MeV alphas have energies, spins, and parities

0 MeV	$\frac{3}{2} +$
.351 MeV	$\frac{5}{2} +$
1.75 MeV	$\frac{7}{2} +$
2.80 MeV	Not reported in <u>Table of Isotopes</u> . $\frac{1}{2} -$, $\frac{3}{2} -$ suggested.
2.87 MeV	$\frac{1}{2} +$

In practice, only the three lowest states were read in as possible final states. This conservative choice bypassed the question of spin of the 2.80 MeV state. The calculated cross sections are shown in Figure 3. The cross sections obtained by averaging the experimental thin target $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ cross sections of Bair and Willard⁹ are shown for comparison. The averaged experimental cross sections shown were used in scaling the calculated cross section Legendre coefficients by the prescription (17).

The thin target cross sections were available only up to 5.145 MeV for ^{18}O and 5.31 MeV for ^{17}O . Outside of this range the theoretical cross sections from HAU were scaled according to the ratio of experimental to theoretical cross sections at the end points of the experimental data. There is, of course, some error introduced by this procedure, particularly for $^{238}\text{PuO}_2$, in which 72% of the alphas are emitted at 5.50 MeV.

The (α, n) neutron source spectra calculated by this procedure for PuO_2 with natural oxygen concentrations are shown in Figures 4, 5, and 6 for plutonium 238, 239, and 240, respectively. The alpha flux in PuO_2 for $^{238}\text{PuO}_2$ is shown in Figure 7. Calculated yields and neutron spectrum parameters appear in Table 1.

Table 1. Calculated PuO_2 (α, n) Neutron Spectrum Characteristics

<u>Plutonium Isotope</u>	<u>(α, n) neutron yields (neutrons/gm Pu/sec)</u>	<u>Mean Energy (MeV)</u>	<u>Most Probable Energy (MeV)</u>
238	1.06×10^4	2.14	2.32
239	31.1	2.02	2.37
240	116.	2.03	2.37

PuC Calculations

The $^{13}\text{C}(\alpha, n)^{16}\text{O}$ reaction in PuC is distinctly different in one feature from corresponding (α, n) reactions in plutonium oxides. The closed shell structure of ^{16}O results in a high excitation energy for the first excited state. Hence for alphas below about 5.02 MeV, only the ground state is energetically accessible. Hence, the Legendre coefficients for the angular distribution of neutrons are those for transitions to the ground state. No branching calculations are needed below 5.02 MeV, and the angular distribution information is available from experiment. The Legendre coefficient ratios W_L/W_0 , $1 \leq L \leq 6$, from the work of Barnes, Belote, and Risser¹⁰ were used in the 2.00 to 3.45 MeV alpha energy range in conjunction with the (α, n) total cross sections ($= 4\pi W_0$) of Bair and Haas.¹¹ For alpha energies from 3.64 to 4.98 MeV, Legendre coefficients from the work of Kerr, Morris, and Risser¹² were used.

Above 5.02 MeV, the first two excited states of ^{16}O become accessible. In this range, it was again necessary to obtain Legendre coefficients for the partial cross sections from Hauser-Feshbach theory. By using the potential parameters listed below, reasonable agreement of calculated and experimental cross sections in the 5.00 to 5.15 MeV alpha energy range was obtained.

$^{13}\text{C} + \alpha$ states

$$r_a = 1.2 A^{1/3} + 1.3 \text{ Fermis} \quad A = 13$$

$$r_b = 1.2 A^{1/3}$$

$$a = .55 \text{ Fermis}$$

$$b = .74 \text{ Fermis}$$

$$x = 0.$$

$$V_o = 50. \text{ MeV}$$

$$W_o = 4. \text{ MeV}$$

$^{16}_0 + n$ states

$$r_a = 1.13 A^{1/3} \text{ Fermis}$$

$$r_b = 1.13 A^{1/3} + 1.3 \text{ Fermis}$$

$$a = .72 \text{ Fermis}$$

$$b = .98 \text{ Fermis}$$

$$V_o = 47.9 \text{ MeV}$$

$$W_o = 7.35 \text{ MeV}$$

$$V_{so} = 8.00 \text{ MeV}$$

$$W_{so} = 0. \text{ MeV}$$

$$x = 0.$$

The same potential parameters were used to generate the cross section information on up to 5.60 MeV, and these cross sections were used in the NSPEC calculations. No experimental $^{13}\text{C}(\alpha, n)^{16}\text{O}$ cross sections were available above 5.15 MeV. The calculated partial cross sections in the interval from 5. to 5.6 MeV are shown in Figure 8.

It is to be noted that the experimental information includes both odd and even Legendre coefficients, while Hauser-Feshbach theory gives nonzero Legendre coefficients only for even order. The accuracy of Hauser-Feshbach theory declines as the number of accessible states becomes very small. Hence, the presence of nonzero odd Legendre coefficients in the energy range where only one residual nucleus state is accessible by (α, n) reaction (although several states are accessible by (α, α')) is not surprising. The odd Legendre coefficients do appear to oscillate about the value zero as a function of alpha energy over an approximate two MeV interval, but the value of W_L/W_o for L odd at certain energies can be of order unity.

The $^{13}\text{C}(\alpha, n)^{16}\text{O}$ neutron spectrum in PuC calculated with this input is shown in Figure 9, 10, and 11. Calculated yields and neutron spectrum parameters appear in Table 2.

Limitations and Sources of Error

The spectra and yields calculated by our procedure have three primary sources of error: (1) Hauser-Feshbach theory may not be adequate to describe the reactions, (2) the optical model parameters may be less than adequately optimized, (3) the cross section information has a fairly high uncertainty ($\pm 25\%$) and is missing in certain key regions.

Hauser-Feshbach theory predicts symmetry about 90° in the center of mass frame. In practice some peaking in the forward (i.e., incident alpha particle) direction may be expected to occur. The statistical assumptions in the theory we have used assume that the compound nucleus loses all memory of the momentum of the incident particle except in translation velocity of the body as a whole. The direct knocking out of a neutron from the target nucleus or stripping of two protons and a neutron from an incident alpha should result in a neutron distribution more highly peaked in the forward direction and hence, more energetic. Because of differences in energy resolution, cross section normalization and target thickness in experiments observing forward and laterally emitted neutrons and those detecting neutrons emitted at all angles, it is difficult to assess the amount of forward peaking that is occurring. The work of Bonner, Kraus, Marion, and Schiffer¹³ shows that the neutron emission from (α, n) reactions on ^{10}B and ^{11}B is isotropic for alpha energies below about 3.75 MeV. The $^{13}\text{C}(\alpha, n)^{16}\text{O}$ neutron emission from their work is non-isotropic, as is also our calculated emission. One can not readily estimate the anisotropy of $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ neutrons from their work. A more important test of Hauser-Feshbach, however, is the presence of

Table 2. Calculated PuC (α ,n) Neutron Spectrum Characteristics

<u>Plutonium Isotope</u>	<u>(α,n) Neutron Yield (Neutrons/gm Pu/sec)</u>	<u>Mean Energy (MeV)</u>	<u>Most Probable Energy (MeV)</u>
238	1.49×10^4	4.87	4.37
239	37.3	4.68	4.37
240	137.8	4.68	4.37

forward-backward symmetry in the center of mass frame. From Relation (2) and Relation (3) we see that the difference of laboratory energies of a neutron emitted forward and one emitted backward by $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ reaction from a 5 MeV alpha with ^{21}Ne going to ground state is

$$\Delta E = 1.46 \text{ MeV.}$$

Hence, a marked peaking of the neutron distribution in the forward direction could result in a significant shifting of the neutron spectrum to higher energies.

We have no information readily available to check the branching to residual nucleus excitation states predicted by Hauser-Feshbach. An experimental check would involve counting decay gammas from the excited states of the residual nucleus.

Assuming adequacy of Hauser-Feshbach theory, there remains the possibility of improving the potential parameters describing the interaction. The agreement of the calculated and measured $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ cross-sections with our choice of parameters was only qualitative. While scaling for agreement with experimental totals corrects the predicted yield, it does nothing to correct errors in branching to residual nucleus excitation states resulting from poorly chosen parameters. Further optimization might be helpful.

The cross sections for $^{17}\text{O}(\alpha, n)^{20}\text{Ne}$ used had a stated uncertainty of $\pm 25\%$. Presumably, the other cross sections used have a similar uncertainty. A systematic understating of cross sections may account for calculated neutron yields being persistently lower than those reported for bulk materials.

An additional source of error in our calculations is the alpha flux computation. If the stopping power obtained from tables or the quadratic interpolation on atomic charge were systematically in error for a material present, the calculated alpha flux would be in error. Since the stopping

power of plutonium is extrapolated rather than interpolated (highest Z element in the table used is uranium); the stopping power attributed to it may be too high, the calculated alpha flux and calculated (α, n) neutron yields then being too low. The error in yield from this source is probably only a few percent, less than the cross section uncertainty.

Discussion of Results

Examined grossly, the neutron spectra which we have calculated appear plausible. The mean and most probable energies for our calculated $\text{PuO}_2(\alpha, n)$ spectra, however, are somewhat lower than those previously measured or calculated.^{14,15,16} If this proves to be an erroneous prediction, the cause is probably one of those discussed in the preceding section. For conservative shielding calculations, it may be wise to also utilize a conservative "hard" spectrum in which all residual nuclei are in the ground state. One needs a corresponding "hard" assumption for the angular distribution. The ground states of ^{18}O and ^{21}Ne have spins of 0 and 3/2, respectively. Hence a change of at least one unit orbital angular momentum must be accomplished by the alpha absorption and neutron emission. One might be tempted for this reason to take a $|Y_{10}|^2$ angular distribution as the conservative case. We believe, however, that the alpha particle is more likely to carry the orbital angular momentum change than is the neutron. We believe, therefore, that isotropic neutron emission plus all transitions to ground state provides a sufficiently conservative "hard" spectrum for shielding calculations. Such a spectrum is shown in Figure 12.

Owing to the fairly large uncertainty in the experimental thin target cross sections, one should be cautioned against assigning a greater significance to the calculated yields than is justified.

The plutonium carbide (α, n) neutron spectrum calculations are much less ambiguous than those for plutonium oxide. Because of the lack of

ambiguity as to final nucleus excitation state in $^{13}\text{C}(\alpha, n)^{16}\text{O}$ below 5.02 MeV and the availability of experimentally supported angular distribution information, the calculated neutron production for ^{239}PuC and ^{240}PuC should be quite reliable. The ^{238}PuC neutron spectrum and yield will be less reliable because of the greater alpha energy range through which one must rely on calculated cross sections and branching. The leading source of error in the PuC calculations appears to be in cross section uncertainties and inadequate energy range of cross sections.

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Figure 1. Energy levels, spins, and parities of (α, n) reacting nuclides and residual nuclei.

Figure 2. Calculated and group averaged experimental $^{17}\text{O}(\alpha, n)^{20}\text{Ne}$ cross sections.

Figure 3. Calculated and group averaged experimental $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ cross sections.

NEUTRON SPECTRUM FROM (ALPHA,n) REACTION IN $^{238}\text{PuO}_2$

NEUTRONS/CC/SEC/.05 MEV ENERGY INTERVAL $\times 10^4$

Figure 4.

Neutron spectrum in $^{238}\text{PuO}_2$
from (α,n) reactions assuming
Hauser-Feshbach branching and angular
distribution. Vertical axis assumes
single crystal density 11.46 gm/cm^3

E_{LAB}

MEV

Figure 7. Alpha flux in $^{238}\text{PuO}_2$.

Figure 8. Calculated $^{13}\text{C}(\alpha, n)^{16}\text{O}$ cross sections above ^{16}O first excited state threshold.

NEUTRON SPECTRUM FROM (ALPHA, N) REACTION IN ^{238}PuC

NEUTRON SPECTRUM FROM (ALPHA, N) REACTION IN ^{240}Pu C

Figure 11.

Calculated neutron spectrum
from (α,n) reactions in ^{240}Pu C.

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