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Modeling of (α, n) Reactions on Light Nuclei

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α interactions on light nuclei in the resonance range are an important source of neutrons via (α, n) processes in present (oxide) and future (carbide, nitride) nuclear fuels. The purpose of this work is the modeling of such (α, n) interactions on oxygen isotopes to get a set of nuclear parameters which can subsequently be used to calculate the intensity and spectrum of the emitted neutrons.

***KEYWORDS:* modeling (α, n) reactions, oxygen isotopes, Reich-Moore approximation, validation, neutron energy spectrum, (α, n) cross section**

I. Introduction

Evaluated nuclear data are essential for a large range of applications, in particular in the electro-nuclear field. Such data are derived from nuclear reaction models adjusted on experimental measurements.

Among these data, those concerning the α particle interactions on light nuclei are essential for calculating neutron emission via (α, n) processes in present (oxide) and future (carbide, nitride) nuclear fuels. Examples of application are reprocessing, packaging, transportation and storage of radioactive waste, neutron source term evaluation in critical reactors and sub-critical reactors (Accelerator Driven Systems).

The goal of this work is the modeling of (α, n) reactions on oxygen isotopes to extract the resonance parameters. An application of this work will be to use these parameters to obtain the number and energy spectrum of neutrons produced by (α, n) reactions in oxide nuclear fuels. This is motivated by the fact that, in past studies of $PuO_2 - UO_2$ fuel arrays, unexplained discrepancies of 40% have been observed between calculations and experiments.

II. Determination of Oxygen Isotope Resonance Parameters

1. Fitting Procedure

The first step of the present work is the modeling of the (α, n) reactions cross section of the oxygen isotopes. We used the SAMMY code[1)] which relies on the Reich and Moore approximation of the R-matrix theory. From the Bayessian fitting procedure between the calculated and the experimental data, a set of nuclear resonance parameters was determined. In its most recent version, the SAMMY code allows the study of the in- and out-going charged particle channels. An important validation work of this module has been made at CEA Cadarache in collaboration with the author of the code (N. Larson[2)]).

An immediate difficulty is the retrieval of the experimental data. The (α, n) data on oxygen isotopes are not currently studied experimentally. The ideal configuration would be the simultaneous analysis of all the channels leading to the same compound nucleus ($(n, n), (n, \alpha), (\alpha, \alpha)$ and finally (α, n)) by least squares fitting. Unfortunately, the only accurate experimental data available are the $^{17}O(\alpha, n)^{20}Ne$ reaction measured by J. K. Bair and F. X. Haas[3)] in 1973 and the $^{18}O(\alpha, n)^{21}Ne$ reaction measured by J. K. Bair and H. B. Willard[4)] in 1962.

In 1978, J. K. Bair and J. Gomez del Campos[5)] obtained a new value for the absolute cross section of the $^{NAT}O(\alpha, n)$ reaction. This measurement being more accurate, they compared it to the values of the $^{17,18}O(\alpha, n)$ reactions previously measured. Following the conclusion of their comparison, we had to re-normalize the two early experimental data sets by a factor of 1.35.

2. The $^{17}O(\alpha, n)^{20}Ne$ and $^{18}O(\alpha, n)^{21}Ne$ Reactions

The experimental data used to fit the $^{17}O(\alpha, n)^{20}Ne$ reaction (**Figure 1**) and the $^{18}O(\alpha, n)^{21}Ne$ reaction (**Figure 2**), are the re-normalized cross section data of J. K. Bair and F. X. Haas[3)] and J. K. Bair and H. B. Willard.[4)] For the first reaction, the cross section has been measured for α-particles having an energy between 2.0 MeV and 5.3 MeV.

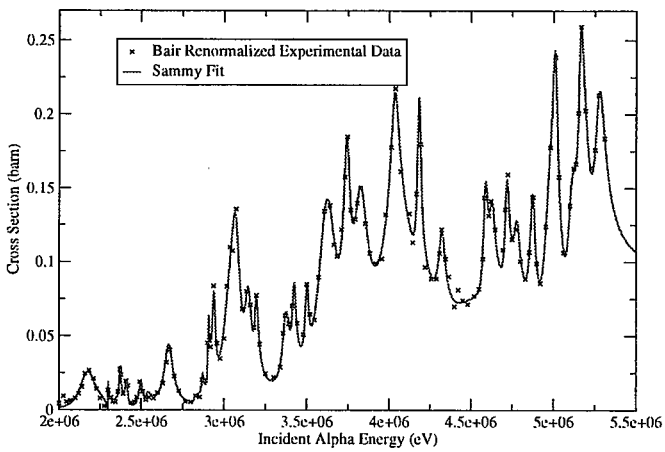

Fig. 1 $^{17}O(\alpha, n)^{20}Ne$ reaction fit realized with SAMMY from Bair re-normalized experimental data.

For the second reaction, the cross section has been measured for α-particles having an energy between 2.4 MeV and 5.2 MeV.

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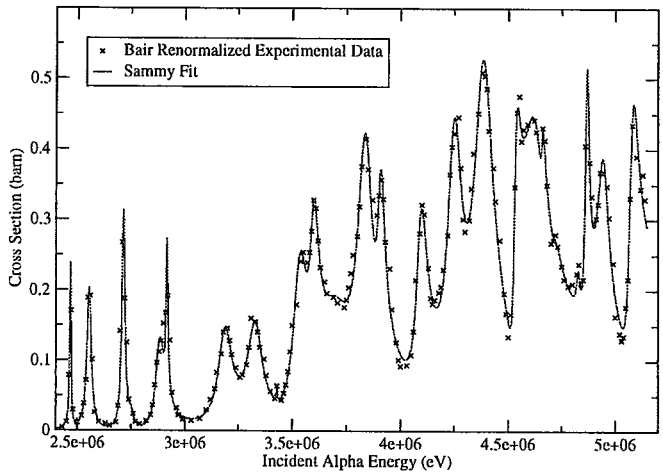

Fig. 2 $^{18}O(\alpha,n)^{21}Ne$ reaction fit realized with SAMMY from Bair re-normalized experimental data.

3. Validation of the SAMMY Module for Charged Particles

Contrary to other codes, SAMMY allows one to calculate the cross sections for charged particle channels. In order to validate SAMMY in this configuration, we have used the reciprocity principle. This principle shows that it is possible to create a compound nucleus either from the entrance channel or from the exit channel. It requires a mathematical transformation to switch from a reaction to the reverse reaction.

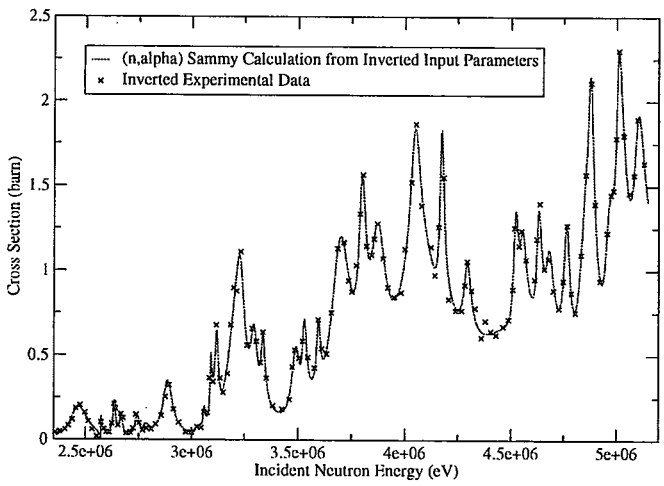

Fig. 3 $^{20}Ne(n,\alpha)^{17}O$ SAMMY calculation vs inverted experimental data.

For example, we can switch from the $^{17}O(\alpha,n)^{20}Ne$ reaction to the $^{20}Ne(n,\alpha)^{17}O$ reaction with the energy and cross section transformations hereafter:

$$E_n = [(B_\alpha - B_n) + \frac{m_{o17}}{m_{o17}+m_\alpha}E_\alpha]\frac{m_{Ne20}+m_n}{m_{Ne20}} \quad (1)$$

where E_n is the neutron incident energy, E_α is the α energy, B_α is the α binding energy in the ^{21}Ne and B_n is the neutron binding energy in the ^{21}Ne.

$$\sigma_{n\alpha} = \frac{k_\alpha^2}{k_n^2}\frac{g_n}{g_\alpha}\sigma_{\alpha n} \quad (2)$$

where $\sigma_{\alpha n}$ is the (α,n) reaction cross section, $\sigma_{n\alpha}$ is the (n,α) reaction cross section, k_α is the in-coming wave number, k_n is the out-going wave number, g_n is the neutron channel statistical spin factor and g_α is the α channel statistical spin factor.

Figure 3 shows that the $^{20}Ne(n,\alpha)^{17}O$ reaction cross section calculation is consistent with the $^{17}O(\alpha,n)^{20}Ne$ reaction cross section fit.

4. Charged Particles Reactions Specificities

In studying reactions induced by charged particles, we notice that for a given J^π (corresponding to the spin and the parity of the compound nucleus state) the number of entrance or exit channels leading to the same compound nucleus state is often greater than one. Consequently, several neutron and alpha channel spins are involved.

For instance, the spin and parity of the various particles in the $^{18}O(\alpha,n)^{21}Ne$ reaction are:

$$\begin{cases} s^\pi_{o^{18}} & = 0^+ \\ s^\pi_\alpha & = 0^+ \\ s^\pi_{Ne^{20}} & = 3/2^+ \\ s^\pi_n & = 1/2^+ \end{cases}$$

Therefore, the value of the entrance channel spin (S_{in}) determined by the following inequality

$$|s_{o^{18}} - s_\alpha| \leq S_{in} \leq s_{o^{18}} + s_\alpha$$

is

$$S_{in} = 0$$

The reciprocal inequality $|s_{Ne} - s_n| \leq S_{out} \leq s_{Ne} + s_n$, determines the possible values of the exit channel spin

$$S_{out} = 1 \; or \; 2$$

Now, the spin (J) of the compound nucleus is finally $|S-l| \leq J \leq (S+l)$ where S is the entrance or the exit channel spin and l the orbital momentum of the in- or outgoing particule.

Table 1 gives the J^π values calculated from a channel spin S^π and an orbital momentum l taking into account parity conservation. For $J^\pi = 2^+$, we can see that we have only one entrance channel but four exit channels (one from the channel spin $S^\pi = 1^+$ and three from the channel spin $S^\pi = 2^+$). In the R-matrix theory, a partial width is associated with each channel (defined by J, π, l, S quantum numbers) leading to the same compound nucleus state. Consequently in the previous example, one width is associated with the α channel and four widths are associated with the neutron channel.

Table 1 J^π spin parity values of the compound nucleus calculated from a channel spin S^π and an orbital momentum l for the $^{18}O(\alpha,n)^{21}Ne$ reaction.

CN nucleus spin J^π	α channel spin $S^\pi = 0^+$	neutron channel spin $S^\pi = 1^+$	neutron channel spin $S^\pi = 2^+$
$l=0$ $\pi=+$	0^+	1^+	2^+
$l=1$ $\pi=-$	1^-	$0^-,1^-,2^-$	$1^-,2^-,3^-$
$l=2$ $\pi=+$	2^+	$1^+,2^+,3^+$	$0^+,1^+$ $2^+,3^+,4^+$
$l=3$ $\pi=-$	3^-	$2^-,3^-,4^-$	$1^-,2^-$ $3^-,4^-,5^-$
$l=4$ $\pi=+$	4^+	$3^+,4^+,5^+$	$2^+,3^+$ $4^+,5^+,6^+$

III. Preliminary Results

We present in this section the preliminary results obtained with the SAMMY code. **Table 2** gives the resonance parameters of the $^{17}O(\alpha,n)^{20}Ne$ reaction. This reaction is characterized by only one neutron partial width and three α partial widths.

Table 2 Resonance parameters for the $^{17}O(\alpha,n)^{20}Ne$ reaction

E_α (keV)	J^π	$\Gamma_{\alpha1}$ (meV)	$\Gamma_{\alpha2}$ (meV)	$\Gamma_{\alpha3}$ (meV)	Γ_n (meV)
2170	1/2+	2.4E+7			9.2E+7
2300	1/2+	5.3E+5			4.5E+6
2374	3/2+	9.2E+6	4.8E+6		1.5E+6
2413	3/2-	1.0E+6	2.1E+5		1.9E+7
2500	1/2-	5.2E+6			2.7E+7
2540	1/2+	3.9E+7			2.6E+6
2668	3/2-	3.4E+7	1.6E+7		1.3E+7
2825	1/2+	1.2E+6			3.6E+7
2871	1/2-	1.7E+6			1.2E+7
2908	3/2-	7.8E+6	2.6E+6		3.1E+6
2939	3/2+	1.9E+6	1.0E+7		1.1E+7
3040	5/2+	5.5E+7	9.0E+6	1.2E+7	2.6E+7
3069	7/2-	1.9E+6	5.6E+6	8.2E+6	4.6E+7
3148	3/2+	4.2E+6	2.0E+7		2.4E+7
3197	3/2-	3.8E+6	5.6E+6		1.8E+7
3378	5/2-	2.9E+7	7.5E+6	7.8E+6	1.3E+7
3425	5/2+	8.0E+6	6.2E+6	8.7E+6	7.5E+6
3502	7/2+	1.5E+7	3.8E+6	3.8E+6	3.8E+6
3625	11/2-	6.8E+7	2.5E+6	1.5E+7	4.5E+7
3741	13/2-	5.5E+6	3.5E+6	2.0E+6	3.6E+7
3828	11/2+	5.3E+7	1.7E+7	7.2E+6	2.5E+7
4035	5/2-	2.9E+7	7.5E+6	7.8E+6	1.3E+7
4068	5/2+	8.0E+6	6.2E+6	8.7E+6	7.5E+6
4182	7/2+	1.5E+7	3.8E+6	3.8E+6	3.8E+6
4324	11/2-	6.8E+7	2.5E+6	1.5E+7	4.5E+7
4591	7/2-	4.0E+6	4.6E+6	8.1E+6	1.7E+7
4608	3/2+	1.9E+7	2.4E+8		2.8E+8
4635	5/2+	1.3E+7	9.6E+6	9.8E+6	1.9E+7
4720	9/2+	9.3E+6	8.5E+6	4.0E+6	1.4E+7
4778	1/2+	3.0E+8			1.3E+8

Table 2 (continued)

E_α (keV)	J^π	$\Gamma_{\alpha1}$ (meV)	$\Gamma_{\alpha2}$ (meV)	$\Gamma_{\alpha3}$ (meV)	Γ_n (meV)
4782	5/2-	1.3E+7	1.0E+7	1.0E+7	1.3E+7
4875	9/2-	8.1E+6	8.2E+6	8.4E+6	1.4E+7
4987	11/2+	1.4E+7	1.4E+7	2.2E+7	4.2E+7
5010	13/2-	5.7E+6	8.3E+6	7.5E+6	1.8E+7
5117	9/2+	1.5E+7	1.5E+7	1.5E+7	3.8E+7
5168	11/2+	9.8E+6	1.1E+7	1.3E+7	1.1E+7
5172	13/2+	1.7E+7	1.9E+7	2.4E+7	3.5E+7
5281	13/2-	3.5E+7	7.2E+6	6.9E+6	2.9E+7

The resonance parameters of the $^{18}O(\alpha,n)^{21}Ne$ reaction are given in **Table 3**. The reaction is described by only one α partial width and four neutron partial widths. For the gamma partial width, we have taken the constant values from the reference[10] (Γ_γ= 2.0 eV for the $^{17}O(\alpha,n)^{20}Ne$ and Γ_γ= 4.0 eV for the $^{18}O(\alpha,n)^{21}Ne$ reaction).

Table 3 Resonance parameters for the $^{18}O(\alpha,n)^{21}Ne$ reaction

E_α (keV)	J^π	Γ_α (meV)	Γ_{n1} (meV)	Γ_{n2} (meV)	Γ_{n3} (meV)	Γ_{n4} (meV)
2468	2+	1.1E+6	1.7E+6	1.4E+6	2.2E+6	1.4E+6
2555	1-	5.5E+6	-7.5E+5	1.8E+7		
2714	2+	3.8E+6	3.9E+6	3.4E+6	2.8E+6	3.1E+6
2750	0+	1.3E+6	2.2E+7			
2884	1-	7.4E+6	1.4E+5	4.2E+7		
2916	2+	1.6E+6	2.2E+6	2.5E+6	2.3E+6	2.3E+6
3189	3-	5.3E+6	1.5E+7	9.3E+6	1.5E+7	3.5E+7
3328	4+	4.5E+6	1.9E+7	2.5E+7	2.5E+7	1.0E+7
3432	1-	1.3E+5	2.8E+6	2.8E+6		
3537	2+	1.6E+7	6.6E+6	3.8E+6	2.1E+7	3.3E+7
3603	4+	4.5E+6	2.1E+6	2.2E+6	1.8E+7	2.4E+7
3704	1-	1.5E+8	2.3E+7	9.1E+6		
3836	5-	1.3E+7	2.3E+6	6.7E+7	1.9E+6	8.9E+5
3910	4+	5.2E+6	2.1E+7	1.3E+6	8.2E+6	9.1E+6
4098	2+	1.8E+7	1.1E+7	9.1E+6	9.1E+6	1.6E+7
4251	3-	2.3E+7	2.2E+7	2.3E+7	8.3E+6	1.4E+6
4383	5-	2.1E+7	1.9E+7	5.0E+6	5.0E+7	5.4E+6
4537	5-	5.1E+6	8.5E+6	8.9E+6	8.7E+6	6.9E+6
4625	6+	1.7E+7	2.4E+7	3.0E+6	3.9E+7	6.1E+7
4660	4+	8.0E+5	5.2E+6	5.2E+6	4.6E+6	3.9E+6
4718	2+	8.2E+5	1.0E+7	9.4E+6	1.0E+7	1.1E+7
4827	0+	8.6E+6	1.0E+7			
4868	3-	9.0E+6	3.1E+6	5.1E+6	4.0E+6	1.8E+6
4940	6+	5.8E+6	1.8E+6	5.4E+7	9.4E+5	5.7E+5
5087	6+	8.8E+6	1.1E+7	1.5E+7	1.1E+7	1.3E+7

IV. Evaluation of the Neutron Source (α,n) Reactions in Oxide Fuels

1. Aim of This Section

In order to test the new SAMMY evaluation, we decided to compute the neutron source (in level and energy spectrum) from the nuclear parameters deduced with the SAMMY fit and to compare the results with integral measurements performed

on an experimental arrangement made of $PuO_2 - UO_2$ fuel subassemblies. The source calculation is to be done with another program (ALPHAEN) which has been written by V. Benzi.[6] For a given energy E_α of the in-coming α particle, ALPHAEN calculates the neutron yield $Y(E_\alpha)$, the neutron source $S_{\alpha,n}$ and the neutron energy spectrum in a homogeneous mixture containing natural Oxygen and various kinds of α-emitters. These neutrons are emitted in oxide mixture or compounds, like UO_2 or PuO_2 by (α, n) reactions or spontaneous fission.

We are currently focussing our efforts on replacing the ALPHAEN (α, n) average cross sections (based on the statistical and optical models) by the pointwise SAMMY cross sections calculated with the Reich and Moore approximation of the R-matrix. Thus we expect a finer description of the neutron energetic spectrum within the fuel and so a more accurate calculation of the integral experiment.

2. The ALPHAEN Structure

The ALPHAEN code[6] calculates the energy spectrum of the neutrons emitted by (α, n) reaction in oxide nuclear fuels. The energy distribution of the emitted neutron is given by

$$F(E_n) = 2\pi N_0 \sum_i \sum_k \rho_i \int_{E_{\alpha min}}^{E_{\alpha max}} \frac{\sigma_{i,k}(E_\alpha, \theta)}{b_{i,k}(E_\alpha)} \Phi(E_\alpha) dE_\alpha \tag{3}$$

$b_{i,k}$ is a simple function of the α-particle energy E_α. The differential cross section $\sigma_{i,k}(E_\alpha, \theta)$ is expressed in terms of Legendre polynomial coefficients $C_{i,k,L}(E_\alpha)$ defined for an isotope i, a Legendre degree L and an excited state k of the residual nucleus :

$$\sigma_{i,k}(E_\alpha, \theta) = \sum_L C_{i,k,L}(E_\alpha) P_L(\cos\theta) \tag{4}$$

The Legendre coefficients are included in the block data of the code. They have been previously calculated using the optical model and the Hauser-Feshbach statistical model. It must be stressed that the cross section is a function of the residual nucleus excitation energy, on which the emitted neutron energy also depends.

The expression of the α-particule flux is

$$\phi(E_\alpha) = \sum_k \sum_r N_k \frac{\lambda_{k,r} \delta_{k,r}(E_\alpha)}{-\frac{dE}{dx}} \tag{5}$$

$$with\ \delta_{k,r} = \begin{cases} 1 & \text{if } E_\alpha \leq E_{k,r} \\ 0 & \text{if } E_\alpha > E_{k,r} \end{cases}$$

N_k is the density of the k-th isotope emitting α-particles with energie $E_{k,r}$, r represents the number of raies per emitter, $\lambda_{k,r}$ is the decay constant of the r-th ray of the k-th emitter. The ratio $(-dE/dx)$ represents the stopping power of the α-particle in the mixture. It is assumed that the stopping power in the mixture is given by the so-called "additivity rule" of Bragg and Kleeman.[7] For a plutonium fuel, the adopted stopping power is given by the empirical formula of Perry and Wilson.[8] For all the other elements, ALPHAEN adopts the stopping powers evaluated by Ziegler.[9]

V. Conclusion and Outlook

In this paper, the resonance parameters resulting from a new SAMMY evaluation of $^{17,18}O(\alpha,n)^{20,21}Ne$ reactions have been presented. Our search for experimental data for the analysis of the $^{17}O(\alpha,n)^{20}Ne$ and $^{18}O(\alpha,n)^{21}Ne$ reactions has pointed out the crucial lack of microscopic data on light isotopes, especially for in-coming charged particles. In some cases, the lack of some experimental data may be overcome by the use of the reciprocity principle.

Our contribution to the validation of the SAMMY charged particle module has highlighted the existence of numerous entrance and exit channels leading to the same compound state and the problem of the indetermination of the associated partial widths.

Concerning the neutron source calculation in oxide fuels, we still have to calculate the $C_{i,k,L}$ Legendre polynomial coefficients from the microscopic cross sections description. Thus, we hope to obtain a finer description of the neutron energetic spectrum of the so-called intrinsic source inside the fuels and a better match with some integral data.

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