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REVIEW OF NUCLEAR DATA RELATED TO THORIUM FUEL CYCLE

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ABSTRACT

A knowledge of accurate and reliable nuclear data for the materials in any fuel cycle is essential in assessing its technical feasibility and economic potential. In this review, the nuclear parameters, in which a reactor physicist is usually interested, are discussed for the heavy elements associated with the thorium cycle, such as Th^{232}, U^{233}, Pa^{233} and U^{234} and the fission products. The existing data for these materials and the gaps that have to filled up are described. Temperature dependent effective group capture cross-sections for thorium, which were evaluated with the available resonance parameters, are also tabulated.

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INTRODUCTION

The ultimate feasibility and economic potential of any thorium based fuel cycle depends among other things upon the availability of accurate and reliable nuclear parameters of the associated materials in the thorium fuel cycle. The fact that the value of eta[*] for U^{233} remains greater than 2 over the entire energy range, shows that breeding can be achieved in the U^{233} - Th cycle in thermal as well as fast reactors. The knowledge of nuclear parameters is therefore essential over all the energy regions encountered in the different types of reactors. The accuracy required of cross sections depends upon the influence these have on reactor safety and economics. The specific influence varies with each reactor and therefore a large variety of conditions have to be evaluated in order to specify the cross section needs in detail. Attempts are being made in various laboratories to improve the experimental techniques and obtain the data to a satisfactory accuracy. Integral measurements such as infinite dilution and effective resonance integrals for the fertile and fissile isotopes are also being carried out. These are essential to provide data which cannot be readily obtained in a differential form as a function of energy. It is also necessary to perform the reactor lattice experiments to determine the physics of various U^{233}-Th systems. In this review, the nuclear parameters of the heavy elements associated with the thorium cycle, such as Th^{232}, U^{233}, Pa^{233} and U^{234} and the fission products are discussed. The nuclear parameters in which a reactor physicist is usually interested in his design calculations are first discussed.

Nuclear Data of Interest to the Reactor Physicist:

The total cross section for neutrons at a given energy is defined as the sum of partial cross sections viz.

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[*] Number of neutrons emitted per neutron absorbed

$$\sigma \text{ total} = \sigma_f + \sigma_{n\gamma} + \sigma_{in} + \sigma_{el} + \sigma_{n,2n} + \sigma_{n,\alpha} + \sigma_{n,p}$$

where $\sigma_f, \sigma_{n\gamma}, \sigma_{in}, \sigma_{el}, \sigma_{n,2n}, \sigma_{n,\alpha}$ and σ_{np} refer to the fission, capture, inelastic scattering, elastic scattering, (n,2n) reaction, (n,γ) reaction and (n,p) reaction cross sections respectively. These processes will be discussed in detail below.

Fission:

The data required from this process are the fission cross sections at different energies for the fertile and fissile isotopes; the average number of neutrons emitted per fission, $\overline{\nu}$ and its energy dependence $\frac{d\overline{\nu}}{dE}$; the fission spectrum $\chi(E)$ i.e. the fraction of the total number of neutrons produced in fission in a particular energy interval dE at E.

The fission spectrum is usually obtained from an expression of the type:

$$\chi(E) = ae^{-bE} \sinh(cE)^{1/2}$$

where the constants a, b and c are fitted from experimental results for different nuclei.

Capture:

The radiative capture cross sections as a function of energy are required for the different materials under consideration. Many of these elements exhibit resonance behaviour in their reaction cross-sections. For such of them, data is also needed regarding the resonance parameters for the resonances which occur at different energies. The presence of strong resonances causes fluctuations in the neutron spectrum of a reactor and it is necessary to specify an effective cross section in such resonance regions. This is done by evaluating the so called "resonance integral" which is defined as the lethargy integrated cross section required to produce the true reaction rate when multiplied by the flux existing in the absence of the resonance. Resonance absorption plays a vital role in the prediction of the behaviour of a reactor and a knowledge of the related resonance parameters for different nuclei is essential.

Elastic Scattering

Because of the anisotropies involved in the elastic scattering of neutrons, the angular distribution of the scattered neutrons in addition to the elastic scattering cross section σ_{el}, are required in the reactor

calculations. The elastic scattering cross section is obtained as

$$\sigma_{el} = \sigma_{total} - \sigma_{ne}$$

where σ total is obtained from transmission measurements and the non-elastic cross section σ_{ne} obtained from 'sphere techniques'.

The experimental measurements of angular distributions in certain energy regions are supplemented by calculations using nuclear models like Optical model etc. Another quantity, $\bar{\mu}$, which is the average value of the cosine of the angle of scattering, has been widely used in making what is known as the transport approximation in which a correction for the forward bias of elastic scattering is made by substituting elastic transport (σ_{eltr}) and total transport (σ_{tr}) cross sections, for σ_{el} and σ_{tot}, viz

$$\sigma_{eltr} = \sigma_{el}(1 - \bar{\mu})$$
$$\sigma_{tr} = \sigma_{tot} - \bar{\mu}\sigma_{el}$$

Curves of $\bar{\mu}$ versus energy have been extensively plotted for different elements.

Inelastic Scattering

This is the main mechanism of neutron energy loss in fast reactors. The quantities required in this process are the cross section σ_{in} for this process, the energy distribution of inelastically scattered neutrons and their angular distribution.

σ_{in} is usually measured by threshold detectors, time of flight and gamma ray measurements.

Deviations from isotropy are comparatively small in this process and hence isotropic scattering is assumed.

When the energy of the incident neutron is such that only the first few levels of the target are excited, energy distribution cross sections are calculated from the excitation curves for individual levels. For energies above 1-1.5 MeV in heavy nuclei and 3-3.5 MeV in light nuclei, energy distribution of neutrons is calculated from the

statistical model or the evaporation model using a formula of the type

$$N(E) = E e^{-E/\theta}$$

where θ is the so called nuclear temperature given by Weisskopf as

$$\theta = \left(\frac{E}{\nu A}\right)^{1/2}$$

where E is the incident energy, A is the target mass and ν is a constant determined from experimental results. Recently, a higher value of ν was reported by Dr S. Yiftah [11] at the Third Geneva Conference based on the latest experimental $\theta(E,A)$ values. This higher value of ν has the effect of shifting the "evaporated" neutrons towards lower energies or in other words softening the predicted neutron spectra in fast reactors. Revised inelastic scattering cross sections resulting from these new parameters have been shown by Dr Yiftah to give better agreement between calculated and experimental critical masses. We have to take note of such developments as and when they take place.

The reactions (n,α), (n,p) and $(n,2n)$ require very high threshold energies and hence are not usually considered for reactor calculations.

Thorium - 232.

The cross sections of several materials have been measured with sufficient accuracy in the thermal energy. The emphasis on measurements hitherto has been in the thermal or fast energy regions and little data exist in the intermediate energy ranges. Current experiments are directed towards this gap and results are becoming available.

The important data required for fertile materials like thorium is on the resonance parameters which are necessary to evaluate the effective resonance integrals and the effective multi-group cross sections. Until recently the resonances in thorium had been resolved only upto 1 KeV. Two sets of data have been available from Harwell [1] and Columbia University [2]. There are some uncertainities with regard to the radiation width Γ for thorium. Columbia gives an average value of 40 mv. (millivolt) for Γ_γ whereas Harwell value varies from 18 mv. to 30 mv. These uncertainties in the resonance data are reflected in the comparison between the calculated and measured infinite dilution resonance absorption integral
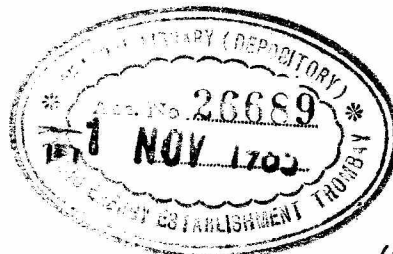
for Th^{232}, which has not been found satisfactory at all. Nordheim[3]
has attempted to bridge the gap by choosing a value of $\Gamma\gamma = 34$ mv along
with the Harwell parameters and has shown a very good agreement between
the calculated and experimental results of not only the infinite dilution
resonance integral, but also for finite rods of Th metal and ThO2. The
fitting he obtains with the measurements of Weitman[4] is extremely good.
Data on the resonance parameters for Th^{232} obtained at Columbia
University[5] were presented at the U.S.A.E.C. Cross Section Advisory
Group meeting in August 1963. Resolved resonance parameters were measured
up to 3 KeV in this data. These parameters were employed to perform the
resonance integral calculations using Nordheim's method[6]. Temperature
dependent effective group cross sections were evaluated for the lower
eleven groups of a 22-group cross section set[7], covering both the
unresolved and resolved resonance regions with the potential scattering
cross section per thorium atom at 40 barns.

The p-wave contribution to the capture cross sections in the
unresolved resonance region (groups 12 to 15) was also evaluated by
resonance integral calculations. The s-wave and p-wave parameters used
in the calculations and the capture cross sections at four temperatures
in the unresolved resonance region are tabulated with s-wave and p-wave
contributions separately in Table 1. The radiative capture width $\Gamma\gamma$
was assumed to have a value of 40 mv. for s-waves and 30 mv for p-waves
in the unresolved resonance region. For the resolved resonances, most
of the levels have a $\Gamma\gamma = 35$ mv . The temperature dependent capture
cross sections for groups 12 to 22 are given in Table 2.

Very recently, more data on the resonance parameters of Th^{232} and
U^{238} have been published[8] from Columbia University in which resonances
have been resolved upto 4000 eV. The average of the first 10 values of
$\Gamma\gamma$ for Th^{232} is found to be 19 mv and for the subsequent levels it is
given as $\sim$35 mv. These parameters can be used to calculate the infinite
dilution resonance integral and the resonance integral for the thorium
metal and oxide rods to see how well they agree with the measurements.
There are still some uncertainties regarding the s-wave and p-wave strength
functions for calculations in the unresolved resonance regions and

suggestions for using different Γ_γ values for s-waves and p-waves may also have to be considered. It appears now that these may be cleared up very soon and reliable effective multi-group cross sections for thorium as a function of temperature and potential scattering cross section per thorium atom can be obtained in the very near future.

Another area of importance for Th^{232} is in the fast inelastic and elastic scattering cross sections which will have to be measured accurately. Statistical model calculations are being currently used to get the inelastic scattering matrix elements.

Uranium-233

The total cross section in the thermal energy range has been measured fairly accurately ($\pm 1\%$). The fission cross section is probably accurate to the same degree at this energy, but the scattering cross section is still uncertain. Attempts to fit the resonance cross sections at low energies (1 to 5 eV) by multi level analysis have not been very successful[9]. The cross sections at energies greater than 10 eV are known only to $\pm 10\%$ accuracy. No direct measurements of radiative capture exist for U^{233}. Measurements of α, the ratio of capture to fission cross section, have been made from 30 KeV to 1 MeV. The capture cross sections are obtained from σ_f and α values.

Some of the cross sections are taken to be the same as for U^{235} in the regions of existing gaps. At present, it is believed that the contribution to the Doppler effect from U^{233} is probably negligible in large fast reactors. One of the main reasons for the lack of sufficient data for U^{233} has been the non-availability of this material in adequate quantities to carry out experiments extensively. It is hoped that in due course as more quantity of this material becomes available for all types of integral experiments, the situation of the nuclear parameters for this element will improve fast.

Protactinium-233

Since Pa^{233} is an intermediate product (with a relatively long 27 day half-life) in the formation of U^{233} from Th^{232}, its cross

section can appreciably affect both the reactivity and the U^{233} yield.

Thermal capture cross section and the gross resonance integral of this element are quoted [12] as 48 $\pm$ 5 barns and 920$\pm$90 barns respectively. But the current results on the resonance parameters account for only about half of the measured resonance integral.

All the cross sections in the fast regions are either obtained by interpolation from data on neighbouring nuclei or computing them theoretically. σ_f is believed to closely resemble that of U^{238}.

Uranium-234

The build-up of this isotope in a fast reactor affects the reactivity coefficients and makes them less negative. The measured cross sections for this isotope are very meagre and the present data comprise of values obtained from comparison with the isotope like Pu^{240}, but U^{234} has a lower threshold for fission than Pu^{240}.

Fission Products

Knowledge of the gross fission-product cross sections is also necessary to predict the performance of a reactor. The estimates of gross cross sections are made from the fission product yields and cross section data for the individual fission fragments, together with theoretical calculations. The probable error associated with these calculated thermal cross sections appears to be $\pm$ 10%. The error may be higher in the case of high energy cross sections. A set of group capture cross sections for the fission products resulting from the fission of U^{233}, U^{235} and Pu^{239} have been published by Garrisan and Roos[10], which are obtained from experimental measurements and statistical estimates.

To sum up, it can be said that there are still many gaps in the basic physics data to be filled, related to thorium utilization. Efforts to obtain the basic nuclear data, integral data and reactor lattice data are all essential to the development of thorium fuel cycle.

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Table 1

s and p-Wave Contributions to the Capture Cross Sections
of Thorium (Barns).

s-waves: $\Gamma_n^o = 1.27 \times 10^{-3}$ ev; $D = 20.1$ ev; $\Gamma_\gamma = 0.04$ ev

p-waves $\dfrac{\langle \Gamma_n^o \rangle}{D} = 1 \times 10^{-4}$; $D = 20.1$ for $J = \frac{1}{2}$; $\Gamma_\gamma = 0.03$ ev
$= 10.05$ for $J = \frac{3}{2}$

$\sigma_p = 40$ barns.

E_L (KeV)	(ℓ, J) = 0, $\frac{1}{2}$	1, $\frac{1}{2}$	1, $\frac{3}{2}$	Total $\sigma_{n\gamma}$	0, $\frac{1}{2}$	1, $\frac{1}{2}$	1, $\frac{3}{2}$	Total $\sigma_{n\gamma}$
	T= 300°K				T = 750° K			
15.0	0.2075	0.0835	0.2237	0.5147	0.2157	0.0843	0.2259	0.5259
9.1	0.3006	0.0905	0.2227	0.6138	0.3181	0.0915	0.2252	0.6348
4.0	0.4459	0.0876	0.1970	0.7305	0.4851	0.0886	0.1994	0.7731
2.8	0.6224	0.0751	0.1588	0.8563	0.6992	0.0759	0.1608	0.9359

E_L (KeV)	(ℓ, J) = 0, $\frac{1}{2}$	1, $\frac{1}{2}$	1, $\frac{3}{2}$	Total $\sigma_{n\gamma}$	0, $\frac{1}{2}$	1, $\frac{1}{2}$	1, $\frac{3}{2}$	Total $\sigma_{n\gamma}$
	T = 1500° K				T = 2500° K			
15.0	0.2206	0.0847	0.2271	0.5324	0.2236	0.0848	0.2274	0.5358
9.1	0.3292	0.0919	0.2264	0.6475	0.3362	0.0922	0.2271	0.6555
4.0	0.5117	0.0891	0.2006	0.8014	0.5292	0.0894	0.2013	0.8199
2.8	0.7555	0.0764	0.1618	0.9919	0.7945	0.0767	0.1624	1.0336

Table 2

Temperature Dependent Capture Cross Sections of Thorium (Barns)

$$\sigma_p = 40 \text{ barns}$$

j	E_L (KeV)	Temperature			
		300° K	750° K	1500° K	2500° K
12	15.0	0.515	0.526	0.532	0.536
13	9.1	0.614	0.635	0.648	0.656
14	4.0	0.731	0.773	0.801	0.819
15	2.8	0.856	0.936	0.992	1.0336
16	2.0	0.995	1.144	1.253	1.328
17	1.4	1.348	1.577	1.759	1.894
18	1.0	1.07	1.254	1.395	1.498
19	0.55	1.088	1.323	1.528	1.692
20	0.30	1.03	1.288	1.54	1.758
21	0.10	0.895	1.119	1.359	1.576
22	0.03	0.723	0.829	0.958	1.087