AUSTRALIAN ATOMIC ENERGY COMMISSION

RESEARCH ESTABLISHMENT

LUCAS HEIGHTS

DETERMINATION OF THE THERMAL NEUTRON

CAPTURE CROSS SECTION OF SAMARIUM-148

by

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ABSTRACT

The thermal neutron capture cross section of ^{148}Sm has been measured by mass spectrometer abundance measurements before and after irradiation of an isotopically enriched sample of ^{148}Sm.

A value of 4.73 ± 0.11 barns was found.

CONTENTS

1. INTRODUCTION

Harvey (1960) listed ^{148}Sm as a nuclide for which no measurements of neutron capture cross section had been made. Cook and Wall (1967) theoretically evaluated the thermal cross section and their result of 4.9 barns was estimated as correct to within one order of magnitude.

The cross section of this nuclide can be important in considerations of neutron economy at high burnup because it provides a link between ^{147}Sm, which terminates the 147 mass chain, and ^{149}Sm, which has an extremely high capture cross section. ^{148}Sm is also formed by the reaction

$$^{147}Pm \ (n,\gamma) \ ^{148}Pm \xrightarrow{\beta^-} \ ^{148}Sm$$
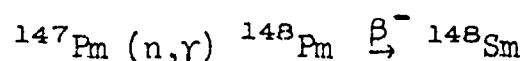
Since both ^{148}Sm and its (n,γ) daughter ^{149}Sm are 'stable' isotopes, mass spectrometric measurements of isotopic abundances both before and after irradiation of a ^{148}Sm sample offer a method of determining the capture cross section. The method is complicated by the extremely high capture cross section of ^{149}Sm since reaction rates of even low initial abundances of ^{149}Sm are appreciable. Two methods explained below can be used to allow for the ^{149}Sm (n,γ) ^{150}Sm reaction, but it should be noted that unless the target material has a very low ^{149}Sm content, error propagation can be serious.

2. THEORY

2.1 Method of Measurement of ^{149}Sm/^{148}Sm Changes

The system can be represented by the reaction chain:

$$A \ (n,\gamma) \ B \ (n,\gamma) \ C$$
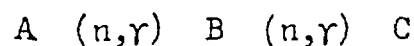
The reaction rates are

$$\frac{dA}{dt} = -\sigma_A \ \emptyset \ A \qquad \qquad .....(1)$$

and

$$\frac{dB}{dt} = \sigma_A \ \emptyset \ A - \sigma_B \ \emptyset \ B \quad , \qquad ......(2)$$

where σ_A is the cross section for the reaction A (n,γ) B,

σ_B is the cross section for the reaction B (n,γ) C,

A, B, C are the concentrations of each nuclide,

$\emptyset$ is the neutron flux,   and

t is time .

Solution of these equations and substitution lead to

$$\frac{\sigma_A}{\sigma_B - \sigma_A} = \left[B/A - (B_o/A_o) \, f \right] / (1-f) \quad , \qquad \ldots (3)$$

where B_o/A_o is the pre-irradiation isotopic ratio, and

$$f = \exp\left[-(\sigma_B - \sigma_A) \, \emptyset \, t \right] \quad , \qquad \ldots (4)$$

$$\simeq \exp\left[-\sigma_B \, \emptyset \, t \right] \text{ since } \sigma_B >> \sigma_A \quad ,$$

$$\simeq \text{ fraction of original } {}^{149}Sm \text{ remaining.}$$

The fraction f can be easily determined by simultaneously irradiating a sample where B_o/A_o is close to unity, so that the growth of ^{149}Sm is negligible compared to its burnout.

In this control sample then

$$f = (B/A)/(B_o/A_o) \qquad \ldots (5)$$

The fraction f can also be measured from the growth of ^{150}Sm in the control sample using the relation

$$1 - f = \left[(C/A) - (C_o/A_o) \right] / (B_o/A_o) \qquad \ldots (6)$$

2.2 Method of Measurement of $(^{149}Sm + {}^{150}Sm)/^{148}Sm$ Changes

Since the cross section of ^{150}Sm is much less than 1 per cent of that of ^{149}Sm, and their concentrations are of the same order, one can neglect the reaction rate of ^{150}Sm. Under these circumstances, the growth of $(^{149}Sm + {}^{150}Sm)$ will represent the reaction rate of ^{148}Sm.

Using the terminology of Section 2.1 this can be written

$$(B/A + C/A) - (B_o/A_o + C_o/A_o) = 1 - \exp(-\sigma_A \, \emptyset \, t), \qquad \ldots (7)$$

from which,

$$\sigma_A \, \emptyset \, t = - \ell n \left[1 - (B/A + C/A) + (B_o/A_o + C_o/A_o) \right] \quad .$$

Since

$$\ell n \, (1 + x) = x - \tfrac{1}{2} x^2 + \tfrac{1}{3} x^3 - \tfrac{1}{4} x^4 + \ldots \ldots \ldots$$

and in this case x is of the order of 10^{-4}, we can neglect all but the first term in the series.

Thus

$$\sigma_A \, \emptyset \, t = (B/A + C/A) - (B_o/A_o + C_o/A_o) \qquad \ldots (8)$$

Substituting for $\emptyset$ t from Equation 4 leads to

$$\frac{\sigma_A}{\sigma_B - \sigma_A} = \frac{-\left[(B/A + C/A) - (B_o/A_o + C_o/A_o) \right]}{\ell n \, f} \qquad \ldots (9)$$

2.3 Measurement of Effective Reactor Cross Section $\hat{\sigma}$ for ^{149}Sm

The methods described in Paragraphs 2.1 and 2.2 lead to ratios of effective cross sections for ^{148}Sm and ^{149}Sm. Since ^{149}Sm has a large resonance near the thermal region (in Westcott (1960) notation g > 1), its effective cross section will be strongly dependent on the thermal distribution of the irradiating neutrons, and this will be reflected in the derived cross section for ^{148}Sm. Thus it is necessary to determine $\hat{\sigma}$ for ^{149}Sm under the irradiation conditions used for the main experiment. This can be done by simultaneous irradiation of the $^{148}Sm - {}^{149}Sm$ control sample and an appropriate thermal flux monitor such as Al-0.5% Co wire.

3. EXPERIMENTAL

3.1 Materials

Separated ^{148}Sm and ^{149}Sm were obtained from Oak Ridge National Laboratory, U.S.A. Two grades of ^{148}Sm were used. The former (Sample 1309(a) of Series KV) contained only 0.032 per cent ^{149}Sm and was used as the irradiation target. The other, less isotopically pure, was used for mixing with the ^{149}Sm as the irradiation control sample. The materials were in the form of Sm_2O_3.

3.2 Preparation of Solutions

The samples were weighed, dissolved in minimum amounts of HNO_3 and made up to volume to give concentrations of about 300 µg/ml. Two solutions were prepared, one of the 'pure' ^{148}Sm and the other of approximately equal amounts of ^{148}Sm and ^{149}Sm.

3.3 Preparation of Targets

Samples of variable amounts in the range of 1 to 10 µg were prepared by dispensing the solutions into silica ampoules each about $\tfrac{3}{4}$ in. long by $\tfrac{1}{4}$ in. o.d. The solutions were evaporated overnight at 80°C before sealing off. Six ampoules of each solution were prepared and the 12 ampoules were sealed into an aluminium irradiation can.

3.4 Irradiation Details

The can was irradiated for one reactor period (566 hours) in hole C1 of Rig X6 in HIFAR. At the end of this period, three standard and three sample ampoules were removed, and the remainder irradiated for a further reactor period of 478 hours (the second period in hole C3 was several days less than usual as a result of mechanical faults in the irradiation rig).

3.5 Specimen Mounting for Mass Spectrometry

Unirradiated samples were mounted by direct pipetting onto a heated tungsten centre filament of an MS5 bead. The irradiated samples were treated as follows: after the ampoule had been opened about 5 μl of 0.5 M HNO_3 was added, the ampoule warmed to assist dissolution, and the solution then mounted as above. Amounts of the order of 1 μg of Sm were used.

3.6 Mass Spectrometry

Samples were run in A.E.I. Ltd. Mass Spectrometer Type MS5 at a filament current of about 3.6A, where large Sm^+ beams ($\sim 10^{-12}$A) were obtained. Care was taken to run all the ^{148}Sm specimens before those containing appreciable ^{149}Sm so that 'memory' effects would not be troublesome. Cyclic recording of the mass range 147 to 150 was employed.

3.7 Flux Measurements

Two flux monitors of Al-0.5% Co wire (Sigmund Cohn Corp., Mount Vernon, N.Y.) were irradiated together with two samples of the ^{148}Sm - ^{149}Sm control. After 24 days' irradiation the monitors and one control sample were removed, fresh monitors were added and the can was re-irradiated for a further 24 days.

The monitors were weighed and their activities measured in a calibrated ion chamber.

4. RESULTS

4.1 Mass Spectrometry Results

^{148}Sm SAMPLES

Irradiation Time (hours)	149/148		150/148	
	Mean	Std. Error	Mean	Std. Error
0	3.402×10^{-4}	0.02×10^{-4}	0.300×10^{-4}	0.01×10^{-4}
556	1.677×10^{-4}	-	2.53×10^{-4}	-
1044	1.094×10^{-4}	0.006×10^{-4}	3.518×10^{-4}	0.004×10^{-4}

^{148}Sm - ^{149}Sm CONTROL SAMPLES

Irradiation Time (hours)	149/148		150/148	
	Mean	Std. Error	Mean	Std. Error
0	0.5803	0.002	0.869×10^{-2}	0.005×10^{-2}
556	0.2402	-	0.3402	-
1044	0.1058	-	0.4828	-
1044	0.1083	-	0.4894	-

4.2 Effective Cross Section for ^{149}Sm

The separate irradiation carried out to determine the effective cross section of ^{148}Sm gave the following results:

Irradiation Time (hours)	149/148		150/148		Wt (mg) Al-Co alloy (0.5% Co)		Activity (μCi) at mid point of irrad.	
	Mean	Std. Error	Mean	Std. Error				
0	0.5803	0.0053	0.869×10^{-2}	0.012×10^{-2}	-		-	
515	0.2467	0.0012	0.3408	0.0012	20.1	19.9	44.7	44.3
1066	0.1036	0.0005	0.4862	0.001	20.1*	19.9*	46.3*	48.0*

* additional to 515 hours' irradiation value

Integrated flux was calculated using nuclear data for cobalt recommended by Wade and Bailey (1967), and epithermal index (Westcott r) value measured in a geometrically similar irradiation facility by Boldeman, Lang and Nicholson (1962).

For each irradiation period, $\hat{\sigma}$ was calculated from both depletion of ^{148}Sm and growth of ^{150}Sm. Results are:

Irradiation Time (hours)	$\hat{\sigma}$ from ^{149}Sm depl. (barns)	$\hat{\sigma}$ from ^{150}Sm growth (barns)
515	82,890	82,920
1,066	81,360	81,730
Mean	82,230 ± 400 (1 S.E.)	

4.3 ^{148}Sm Cross Section Results

Results of the 556-hour irradiation gave values for ^{148}Sm $\hat{\sigma}$ of 3.46 barns by Method 2.1, and 4.80 barns by Method 2.2. The disagreement arises from large error magnifications incurred in the subtraction in Equations 3 and 9. Since the samples irradiated for 1,044 hours had significantly lower error magnifications, subsequent calculations were confined to these, and are detailed below.

Values for 'f' (Equation 4) were calculated from both depletion of ^{149}Sm (Equation 5) and growth of ^{150}Sm (Equation 6) in the control samples. Four values were thus obtained and a mean and standard error calculated, after appropriate weighting to allow for error propagation in the result derived. (At this level of burnout, the depletion measurements are 4.3 times more precise than the growth measurements). A value for f = 0.183 ± 0.002 was obtained. Results were then calculated for ^{148}Sm $\hat{\sigma}$ using Equations 3 and 9. Standard errors were calculated for each, using conventional error propagation methods and assuming independence of variances.

Results are:

Method 2.1 (Equation 3) $\hat{\sigma}$ = 4.72 ± 0.08 barns

Method 2.2 (Equation 9) $\hat{\sigma}$ = 4.78 ± 0.17 barns

These provide a grand mean of 4.73 barns with a standard error of 0.08.

5. DISCUSSION

The measured cross section can be related to the 2,200 m/s cross section σ_0 by the Westcott (1960) relation

$$\hat{\sigma} = \sigma_0 \, (g + r \, s)$$

Cook (1967 - private communication) believes that ^{148}Sm has no large resonances and that they are well spaced from the thermal region (i.e. s ≯ 20 and g = 1). Since Boldeman, Lang and Nicholson (1962) found a value for r = 0.0004 in an irradiation facility similar to that used for the experiment, it can be concluded that

$$\hat{\sigma} - \sigma_0 < 0.04 \ .$$

In other words, the value of σ_0 is equal to $\hat{\sigma}$ within less than one standard error of the above measurement.

Systematic errors could arise from several sources. One is the ^{59}Co cross section, on which all these cross section data depend. The value used was 37.4 barns. Another possible source of error is self-shielding during irradiation of

the control samples, that is, those containing a high abundance of ^{149}Sm. However, a uniform film that exhibits 1 per cent attenuation has a thickness of about 50 µg/cm^2 of ^{149}Sm. It is expected that these samples were much thinner than this, and further, since it is unlikely that the specimens were spread to the same thickness any appreciable self-shielding effects would be reflected in inter-specimen discrepancies. That no such discrepancies were observed is further evidence that self-shielding effects were negligible.

An independent check on these effects is affored by the ^{149}Sm $\hat{\sigma}$ measurement above. If this is combined with σ_0 of 41,500 (G.E.C. Chart of the Nuclides, 9th ed., 1966) one derives a g value of 1.981, corresponding to a Maxwellian temperature of 138°C (Westcott 1960). The measured temperature of the graphite near the irradiation position was 130°C.

A possible source of mass spectrometry error is the familiar occurrence of background peaks such as hydrocarbons over a wide mass range and alkali aggregates such as ^{23}Na$_3$, ^{39}K, ^{41}K$^+$ having m/e = 149. In general, these unwanted peaks are easily resolved by reducing the collector slit width to about one third its normal setting of 0.020 in. When source conditions are such that the Sm peaks are 'clean', the slit is returned to normal and recording is commenced. A further check is to observe the constancy of the ^{147}Sm/^{148}Sm samples. This is particularly sensitive to the alkali aggregates since ^{23}Na$_3$; ^{39}K$_2$$^+$ appears at m/e = 147, and is about thirteen times as abundant as the 149 interference.

If the likely error in the ^{59}Co cross section (0.6 barn) is compounded with the standard error of the experiment a final result for σ_0 is 4.73 ± 0.11 barns.

6. ACKNOWLEDGEMENTS

The author is indebted to Mr. J. L. Cook and Dr. J. L. Symonds for informative discussions, and to Dr. G. C. Lowenthal for ion chamber measurements.

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