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DETERMINATION OF NEUTRON-INDUCED ACTIVATION CROSS SECTIONS USING NIRR-1

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ABSTRACT
Thermal Activation cross-sections for the (n, γ) reaction were experimentally measured using NIRR-1 facilities. The irradiated target isotopes were ^{71}Ga, ^{109}Ag, ^{55}Mn ^{94}Zr; ^{96}Zr; ^{238}U, ^{74}Se, ^{75}As and ^{48}Ca. In order to obtain reliable activation cross sections, careful attention was paid to neutron irradiation and to the measurement of induced activity. The values obtained for the ten (n, γ) reactions were compared against the comprehensive evaluated values listed in Mughabghab, (2003) and other evaluated data libraries. The values were found to compare well with values evaluated by Mughabghab and those from other libraries.

Keywords: RNAA, Thermal capture cross section, Resonance integral, k_0-factors.

INTRODUCTION

A database of activation cross sections for neutron energies (of up to 20MeV) is required for the design of a D-T fusion reactor, neutron dosimetry and neutron shielding in a reactor facility, and for confirming predictions based on reaction calculations. The (n, γ) cross section σ_0 is a basic constant and the reliability of the literature data should be a matter of concern; as the accuracy and consistency of σ_0's from different libraries are often troublesome (De Corte et al, 1987) thus making a selection of reliable data difficult. A number of the cross section data compiled, in several evaluated data libraries, are evaluated mainly on the basis of theoretical calculations. Experimental data, if available, are used to normalize the calculated values and to improve their accuracy and reliability. In cases where no measurements have been made, the systematics for the majority of the reactions are used to predict the cross sections.

Recently, a number of investigations were conducted to measure activation cross sections σ_0 and isomeric cross section ratios I_0 of some nuclides whose σ_0 and I_0 are either doubtful or have never been measured (Andrej, 2002). More recently, the IAEA set up the Working Party on Evaluation Co-operation (WCEC) Subgroup-23 (SG-23) to assemble, verify and validate an evaluated data library for the minor fission products by reviewing and making use of existing evaluations, running nuclear model calculations and performing partial re-evaluation work (Andrej, 2002). The purpose of the exercise was to compare the thermal capture cross sections and resonance integrals from the SG-23 library to the equivalent values in the k_0 database. It was discovered that 12 out of 30 nuclides studied showed differences from 5% to greater than 20%.

The Nigeria Research Reactor-1, NIRR1, is designed for use in NAA thus, its neutron flux remains stable and constant during both irradiations of the 'bare' and the 'Cd-covered' foils.. The k_0-standardized method is widely accepted for multi-element NAA with respect to variations in neutron spectrum parameters and gamma-ray counting geometry (De Corte, 1987). The k_0-standardized method, developed in the 1970s by Simonits et al., (1975) involves the simultaneous irradiation of a sample and a neutron flux monitor, such as gold, and the use of a composite nuclear constant called k_0-factor. The k_0-factor is independent of irradiation and measuring conditions thus, eliminating the problems of non-availability of standards for certain elements and the inaccuracy caused by poorly prepared in-house standards (Jonah et al, 2005). Our main objective is to determine activation cross section data in the framework of the IAEA Coordinated Research Project No.13728 entitled "Reference Database for NAA" using the NIRR-1 Facilities.

MATERIALS AND METHODS

In the k_0 standardization of NAA, the concentration of the analyte 'a' is obtained from its measured isotope/gamma ray as

$$\rho_a = \frac{\left(N_P / Wt_m SDC\right)_a}{\left(N_P / wt_m SDC\right)_{Au}} x \frac{1}{k_{0,Au}(a)} x \frac{f + Q_{0,Au}(\alpha)}{f + Q_{0,a}(\alpha)} x \frac{\varepsilon_{P,Au}}{\varepsilon_{P,a}} \qquad (1)$$

where 'Au' refers to the co-irradiated gold monitor [^{197}Au (n,γ) ^{198}Au, E_γ=411.8 keV] and N_p is the number of counts in the full energy peak, W the weight of the gold monitor, W the weight of the sample, S the saturation factor, D the decay factor, C the counting factor, t_d the decay time, t_{ur} the irradiation time, t_m the measuring time, f the thermal to epithermal neutron flux ratio, α the epithermal neutron flux distribution, approximated by a $1/ E^{1+\alpha}$ dependence,

$Q_{o,}$= I_o/σ_o is the ratio of resonance integral to thermal neutron capture cross section at neutron velocity of 2200 ms^{-1} and ε_P the full energy peak detection efficiency.
Furthermore, in Eq. (1), the $k_{0,Aa}$ (a) factor is defined as

$$k_{0,Au}(a) = \frac{M_{Au}\ \theta_a\ \sigma_{0,a}\ \gamma_a}{M_a\ \theta_{Au}\ \sigma_{0,Au}\ \gamma_{Au}} \qquad (2)$$

Where M is the atomic weight, θ is the isotopic abundance, σ_0 is the 2200 m/s (n,γ) cross section, and γ is the absolute gamma ray intensity (emission probability).

Eq. (1) is written in the Hogdahl formalism (modified for a 1/ $E^{1+\alpha}$ epithermal flux shape for which it is required that the cross section in the thermal neutron region varies as $1/v$. This requirement is fulfilled to a good approximation by most analytically interesting (n, γ) reactions.
The conversions from the Q_0 values to Q_0 (α) and from s_0 values to s_0 (α), is given by

$$Q_0(\alpha) = \left[\frac{Q_0 - 0.429}{(\overline{E}_r)^\alpha} + \frac{0.429}{(2\alpha+1)(E_{Cd})^\alpha}\right](1\,eV)^\alpha \qquad (3)$$

where $\overline{E}_r$ is the effective resonance energy. The (1 eV)$^\alpha$ term (numerically unity) originates from the definition of the epithermal neutron flux in a $1/E^{1+\alpha}$ distribution. The above treatise holds for the case where the analytical isotope is directly formed by (n, γ) reaction.

As to the measurement of Q_0 values, the well-known cadmium ratio method (in the Hogdahl formalism) was used, and

$$Q_0(\alpha) = \frac{f}{F_{Cd}R_{Cd} - 1} \qquad (4)$$

followed by conversion of Q_0 (α) to Q_0 according to Eq. (3)

Experimental

The neutron-induced nuclear reactions, whose activation cross sections were measured include:- ^{71}Ga (n, γ)^{72}Ga; ^{109}Ag (n, γ)^{110m}Ag; ^{55}Mn (n, γ)^{56}Mn; ^{94}Zr (n, γ)^{95}Zr; ^{96}Zr (n, γ) ^{97}Zr; ^{238}U (n, γ)^{239}U; ^{74}Se (n, γ)^{75}Se; ^{75}As (n, γ)^{76}As; ^{48}Ca (n, γ)^{49}Ca. The thermal cross sections σ_0; resonance integrals, I_0, or the ratio of resonance integrals to thermal cross sections Q_0, from references, (Andrej, 2002) show large differences of between 5-20%.
The description of the monitors and the nuclear data of the reactions measured or reevaluated are as given in Tables 1 and 2.

Table 1: Description of monitor foils and oxides used.

Target	Material description	Diameter, cm	Range of mass, mg
Au	Al-0.1% Au; 0.1mm thick, IRMM-530	0.8	12-14
Mn	Mn-1% Al foil, 0.1mm thick, SP89070 IRMM.	0.6	3-5
Zr	99.8% Zr foil; 1mm thick, Goodfellow	0.6	42-45
U	U-0.2% Al foil, 0.1mm thick, NS2017 IRMM.	0.6	43-53
Se	SeO_2 powder, Merck KGaA	-	51-63
Ag	Ag_2O powder, Merck KGaA	-	50-51
Ga	Ga_2O_3 powder, Riedel-de-Haën.	-	52-62
Ca	CaO powder, Merck KGaA	-	54-57
As	As_2O_3 powder, Riedel-de-Haën.	-	52-54

Table 2: uclear data of monitor foils and oxides used.

Atomic Weight	Target	product	$T_{½}$	E_r	θ,%	E_γ, keV	γ, %
74.92	^{75}As	^{76}As	26.32h	106	100	559.1	44.6
						559.2	46.2
						563.2	1.6
						657.1	6.4
						1212.9	1.8
						1212.9 + 1216.1	5.5
						1216.1	3.7
196.97	^{197}Au	^{198}Au	2.695d	5.65	100	411.8	95.53
54.94	^{55}Mn	^{56}Mn	2.579h	468	100	846.8	99
						1810.7	27.2
						2113.1	14.3
91.22	^{95}Zr	^{96}Zr	34.97d	6260	17.28	765.8	99

Table 2 Continue

Atomic Weight	Target	product	T½	Er	θ,%	Eγ, keV	γ, %
238.03	^{238}U	^{239}U	23.45m	16.9	99.2746	74.7	50
78.96	^{74}Se	^{75}Se	119.8d	29.4	0.9	121.1	16.41
						136	56.02
						264.7	58.6
						279.5	24.73
						400.7	11.13
107.87	^{109}Ag	^{110m}Ag	249.76d	6.08	48.17	446.8	3.657
						620.4	2.776
						657.8	94.74
						677.6	10.8621
						687	6.49
						706.7	17.0242
						744.3	4.661
						763.9	22.36
						818	7.323
						884.7	72.86
						937.5	34.31
						1384.3	24.35
						1475.8	3.989
						1505	13.11
						1562.3	1.184
69.72	^{71}Ga	^{72}Ga	14.1h	154	39.9	630	24.37
						834	99.55
						894.2	9.842
						1050.7	6.921
						2201.7	26.06
						2491	7.472
						2501.8	20.52
						2507.9	13.05
40.08	^{48}Ca	^{49}Ca	8.718m	1330000	0.187	3084.4	91.7

Source: DeCorte *et al.* (986)

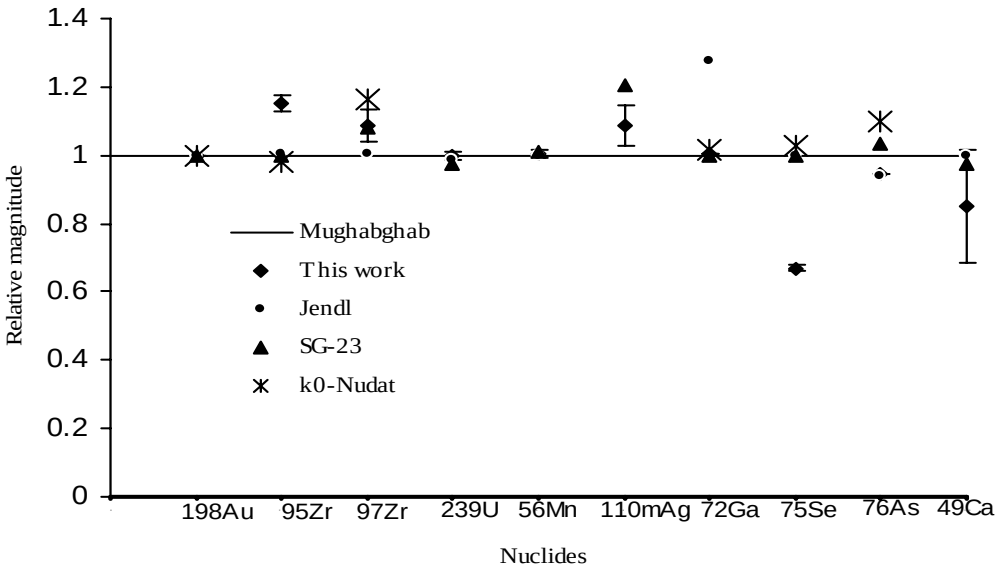

Fig.1. Graphical comparison of Mughabghab with our result and other evaluated data

Table 3: Comparison of Calculated Activation cross section σ_0 and libraries.

Target	Product	Mughabghab	SG-23	k_0/Nudat	Jendl	This work
^{197}Au	^{198}Au	98.65 ± 0.09	-	-	-	98.74 ± 0.10
^{94}Zr	^{95}Zr	0.0499 ± 0.0024	0.04992	0.051	0.04981	0.0433 ± 0.01
^{96}Zr	^{97}Zr	0.0229 ± 0.001	0.02118	0.0197	0.0228	0.0213 ± 0.01
^{238}U	^{239}U	2.68 ± 0.019	2.75*	-	2.717	2.7252 ± 0.03
^{55}Mn	^{56}Mn	13.36 ± 0.05	13.2*	-	13.41	13.3055 ± 0.14
^{109}Ag	^{110m} Ag	4.7 ± 0.20	3.90*	-	90.53	4.2403 ± 0.26
^{48}Ca	^{49}Ca	1.09 ± 0.07	1.12*	-	1.093	0.8502 ± 0.24
^{71}Ga	^{72}Ga	4.73 ± 0.15	4.735(1.8)	4.650(0.8)	3.709	4.7857 ± 0.05
^{74}Se	^{75}Se	51.8 ± 1.2	51.86(2.7)	50.50(4.8)	51.8	77.418 ± 9.14
^{75}As	^{76}As	4.23 ± 0.08	4.092(6.3)	3.85(4.8)	4.5	4.3776 ± 0.03

RESULTS AND DISCUSSIONS

For these determinations, the cadmium ratio was used. The nuclear data for the nuclides used and the cross section for gold, which is the universal comparator, were taken from DeCorte et al., (2003). The resulting activation cross sections of the 10 (n, γ) reactions obtained were compared to the evaluated values by Mughabghab, 2003, the SG-23 library, 2002, k_0/Nudat as well as other nuclear data libraries and is as presented in Table 3 and discussed in 1 to 10 below.

1. ^{197}Au (n, γ) ^{198}Au reaction.
   Though this is not among the reactions whose activation cross section is in doubt, but our result is in excellent agreement with the evaluated data from the nuclear data libraries, SG-23 and Mughabghab within a range of 0.1%.
2. ^{94}Zr (n, γ) ^{95}Zr reaction.
   Although the evaluated data from the libraries and Mughabghab agree with our results to within 13%, the values from the different libraries agree with each other to within almost the same margin.
3. ^{96}Zr (n, γ) ^{97}Zr reaction.
   Our result completely agrees with the results of the SG-23 but differ slightly with results from JENDL-3.3 and Mughabghab to within 7%. The k_0-Nudat value differs from JENDL-3.3 and Mughabghab by about 14%.
4. ^{238}U (n, γ) ^{239}U reaction.
   Our result completely agrees with the evaluated data reported in the libraries and are also in excellent agreement with Mughabghab within a range of 2%
5. ^{55}Mn (n, γ) ^{56}Mn reaction.
   The evaluated data reported by Mughabghab are in agreement with our results to within 0.4%. The results from JENDL-3.3 also differ from Mughabghab by about the same margin.
6. ^{109}Ag (n, γ) ^{110m}Ag reaction.
   The evaluated data reported by Mughabghab is in agreement with our result to within 10%., but differs significantly from results listed by JENDL-3.3.
7. ^{48}Ca (n, γ) ^{49}Ca reaction.
   The evaluated data reported by Mughabghab and JENDL-3.3 vary significantly with our results to within 22%. The results from IAEA-TECDOC, 1990 also differ from our results by about 26%.
8. ^{71}Ga (n, γ) ^{72}Ga reaction.
   The evaluated data reported by Mughabghab and the other nuclear data libraries are in excellent agreement with our results to within 1.2%. Only the results from JENDL-3.3 differ from our result by about 22%.
9. ^{74}Se (n, γ) ^{75}Se reaction.
   The most significant difference from evaluated data reported by Mughabghab and the other nuclear data libraries is in this reaction, where the agreement with our results is to within 49%.
10. ^{75}As (n, γ) ^{76}As reaction.
    The evaluated data reported by Mughabghab and JENDL-3.3 are in excellent agreement with our results to within 3.5%. The results from SG-23 and k_0-Nudat differ from our result by about 7% and 14% respectively.

A graphical comparison of our results with reported values from evaluated data libraries is also presented in Figure 1. The k_0-values for the various gamma energies of these 10 isotopes were also calculated and presented along with the other calculated parameters as shown in Table 3. The results also compare favorably with the evaluated values by Mughabghab and other compilations by De Corte et al, 2003. A graphical comparison of our values with Mughabghab is also given in Figure 1.

CONCLUSION

The cadmium ratio was used to determine the thermal cross sections of ten (n, γ) reactions. Using the reaction ^{197}Au (n, γ)^{198}Au as the standard, the results from seven of the nine reactions were found to be in excellent agreement to within 7% of those reported from evaluated data libraries. The results from the reactions ^{48}Ca (n, γ) ^{49}Ca and ^{74}Se (n, γ) ^{75}Se, were found to differ significantly (22% and 49%) from the evaluated data libraries.

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