

Modified method of α determination in the $1/E^{1+\alpha}$ epithermal neutron spectrum of reactor

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Abstract A modified method has been developed for the α determination in the $1/E^{1+\alpha}$ epithermal neutron spectrum of reactor. It is based on the Cd-covered and without Cd-covered irradiations of two monitors. This method was applied to determine the α value in the channels of Dalat reactor and the results were compared with those obtained by the other methods. It appeared that the results of the modified method were in quite good agreement with those of other methods. It also showed that the modified method was simple in practical uses and a good application in the experiment of α determination in the reactor irradiation channels.

Keywords α Value · Dalat reactor · Modified method

Introduction

As reported by Schumann and Albert [1] and by Ryves [2], the epithermal neutron flux in reactor irradiation channels is not proportional to $1/E$, but rather $1/E^{1+\alpha}$, where α is a small positive or negative constant and a measure of the epithermal neutron flux deviation from the ideal distribution $1/E$, and E is the neutron energy. The α values are smaller than unity in absolute value, and vary between the irradiation channels in the same reactor.

In the ideal case, the resonance integral for a $1/E$ epithermal neutron spectrum is written as:

$$I_0 = \int_{E_{Cd}}^{\infty} \frac{\sigma(E)}{E} dE \quad (1)$$

with: E_{Cd} is the effective Cd cut-off energy (=0.55 eV).

The resonance integrals, defined according to Eq. 1 and tabled in literatures, are not valid in a non-ideal case. In the non-ideal case, the resonance integral for a $1/E^{1+\alpha}$ epithermal neutron spectrum is defined as:

$$I_0(\alpha) = \int_{E_{Cd}}^{\infty} \frac{\sigma(E) \cdot 1 eV^\alpha}{E^{1+\alpha}} dE. \quad (2)$$

It indicates that the resonance integrals for practical uses are a function of α and thus of the irradiation position. Thus, in the (n, γ) -activation analysis with reactor neutrons (NAA) using comparator method, α should be known to preserve the accuracy of the analysis results.

The α value should be determined either by experiment or by calculation. In experiment, several techniques have been developed by De Corte et al. [3–5], namely the “Cd-covered multi-monitor” method, the “Cd-ratio for multi-monitor” method, the “bare multi-monitor” method. However, in these methods, the α values are found from implicit functions by the iterative method on a computer. Consequently, they are merely approximate methods.

In this work, a modified method for the determination of α parameters in reactor irradiation channels is to be presented. In that, the α parameter is written as an explicit formula. The results of the α determination in irradiation channels of Dalat reactor using the modified method are also being reported.

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Base of modified method

From Eq. 2, the resonance integral of isotope *i* in the non-ideal case should be written as:

$$I_{0i}(\alpha) = \left(\frac{I_{0i} - 0.426\sigma_{0i}}{(\bar{E}_{ri})^\alpha} + \frac{0.426\sigma_{0i}}{(2\alpha + 1)(E_{Cd})^\alpha} \right) 1 \text{ eV}^\alpha \quad (3)$$

where σ_{0i} —2,200 m s⁻¹ cross-section of nuclide *i*, \bar{E}_{ri} —effective resonance energy (eV) of nuclide *i*.

Note that Eq. 3 is only valid when $E_{Cd} = 0.55 \text{ eV}$, since $0.426 = 2(E_0/E_{Cd})^{1/2}$ with $E_0 = 0.025 \text{ eV}$ and $E_{Cd} = 0.55 \text{ eV}$.

Accordingly, $Q_{0i}(\alpha) = I_{0i}(\alpha)/\sigma_{0i}$ can be written (in eV unit) as below:

$$Q_{0i}(\alpha) = \left(\frac{Q_{0i} - 0.426}{(\bar{E}_{ri})^\alpha} + \frac{0.426}{(2\alpha + 1)(E_{Cd})^\alpha} \right). \quad (4)$$

We know that, α value is much smaller than unity in absolute value. In practice, in reactor irradiation channels, the absolute α value is less than 0.2 (in most cases, $|\alpha| < 0.1$ and this condition is satisfactory in reactor core). We suggest a substitution $Q_{0i}(\alpha)$ from Eq. 4 by the following approximate formula:

$$Q_{0i}(\alpha) = Q_{0i} \exp(-a_i(\ln \bar{E}_{ri})^\alpha) \quad (5)$$

where a_i is constant for each nuclide and determined by fitting the values of $Q_{0i}(\alpha)$, which are calculated from Eq. 4 in the range of $|\alpha| \leq 0.2$, according to the fitting function (5). Note that, a_i for each nuclide is dependent on the sign of α . In this work, a_i values are determined by fitting $Q_{0i}(\alpha)$ using Kaleigraph program. The values of a_i , correlation and coefficient r of fitting function, and the relevant nuclear data for nuclides chosen as α -monitors are given in Table 1.

As the comparison of $Q_{0i}(\alpha)$ between formula (4) and (5), the results of calculating $Q_{0i}(\alpha)$ for $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$, $^{64}\text{Zn}(n, \gamma)^{65}\text{Zn}$ and $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$ for negative and positive α are respectively shown in Tables 2 and 3. It indicates that, when α values are negative and less than 0.2 in absolute value, the differences of $Q_{0i}(\alpha)$ calculated from these formula are less than 0.08% for $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$, 0.8% for $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$, and about 2% for $^{64}\text{Zn}(n, \gamma)^{65}\text{Zn}$.

In the case of positive α values, $Q_{0i}(\alpha)$ values for $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ match each other very well with the differences less than 0.04%, while $Q_{0i}(\alpha)$ values for $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$ and $^{64}\text{Zn}(n, \gamma)^{65}\text{Zn}$ are in good agreement with $\alpha < 0.16$. When α values are more than 0.16, the differences of $Q_{0i}(\alpha)$ values calculated from these formula can be more than 2% for $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$ and $^{64}\text{Zn}(n, \gamma)^{65}\text{Zn}$. However, in most cases, $|\alpha| < 0.1$ and this condition is satisfactory in reactor

Table 1 Nuclear data and a_i factor for the nuclides chosen as α -monitors, correlation coefficient r of fitting function

Nuclide	\bar{E}_{ri} (eV)	Q_{0i}	a_i		r
			$\alpha < 0$	$\alpha > 0$	
$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$	5.65	15.70	1.0013 ± 0.0006	0.9903 ± 0.0003	0.9999
$^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$	6260	5.306	0.9603 ± 0.0001	0.8770 ± 0.0009	0.9999
$^{64}\text{Zn}(n, \gamma)^{65}\text{Zn}$	2560	1.908	0.8797 ± 0.0002	0.7165 ± 0.0011	0.9997

Data for Au and Zn were taken from De Corte [6] and Zr data from Simonits [7]

Table 2 The values $Q_{0i}(\alpha)$ for monitors calculated from formula (4) and (5) with α in interval [0,−0.2]

α	$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$		$^{64}\text{Zn}(n, \gamma)^{65}\text{Zn}$		$^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$	
	$Q_{0i}(\alpha)$ from (4)	$Q_{0i}(\alpha)$ from (5)	$Q_{0i}(\alpha)$ from (4)	$Q_{0i}(\alpha)$ from (5)	$Q_{0i}(\alpha)$ from (4)	$Q_{0i}(\alpha)$ from (5)
−0.00	15.700	15.700	1.908	1.908	5.306	5.306
−0.02	16.251	16.254	2.172	2.190	6.251	6.275
−0.04	16.822	16.827	2.481	2.514	7.374	7.423
−0.06	17.413	17.421	2.840	2.887	8.712	8.780
−0.08	18.027	18.036	3.260	3.314	10.304	10.380
−0.10	18.663	18.672	3.750	3.805	12.199	12.276
−0.12	19.323	19.331	4.322	4.368	14.453	14.530
−0.14	20.008	20.013	4.990	5.015	17.137	17.186
−0.16	20.719	20.719	5.771	5.758	20.333	20.328
−0.18	21.458	21.451	6.683	6.610	24.137	24.144
−0.20	22.225	22.207	7.750	7.590	28.667	28.541

Table 3 The values $Q_{0i}(\alpha)$ for monitors calculated from formula (4) and (5) with α in interval [0,0.2]

α	$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$		$^{64}\text{Zn}(n, \gamma)^{65}\text{Zn}$		$^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$	
	$Q_{0i}(\alpha)$ from (4)	$Q_{0i}(\alpha)$ from (5)	$Q_{0i}(\alpha)$ from (4)	$Q_{0i}(\alpha)$ from (5)	$Q_{0i}(\alpha)$ from (4)	$Q_{0i}(\alpha)$ from (5)
0.00	15.700	15.700	1.908	1.908	5.306	5.306
0.02	15.169	17.171	1.681	1.701	4.511	4.541
0.04	14.656	14.659	1.487	1.517	3.844	3.886
0.06	14.161	14.165	1.320	1.353	3.282	3.326
0.08	13.683	13.687	1.176	1.207	2.810	2.847
0.10	13.222	13.226	1.053	1.076	2.413	2.437
0.12	12.777	12.779	0.947	0.960	2.078	2.085
0.14	12.348	12.349	0.856	0.857	1.797	1.785
0.16	11.933	11.933	0.777	0.764	1.560	1.528
0.18	11.532	11.530	0.709	0.681	1.360	1.308
0.20	11.146	11.142	0.651	0.608	1.192	1.119

core; as for channel R4V4 of the DR-3 reactor (Ri ϕ , Denmark), $\alpha = 0.158 \pm 0.011$ [8], Eq. 5 can be a good use to replace Eq. 4 in α -practice. It can be seen later in the discussion about the error estimation of the method.

Thus, in the two-detector method of Ryves using Cd-ratio modified by De Corte et al. [4], α can be found as the root of the equation:

$$\frac{(R_{Cd2} - 1)}{(R_{Cd1} - 1)} = \frac{\left(\frac{I_{01}}{\sigma_{01}} - 0.426\right) (\overline{E_{r1}})^{-\alpha} + \frac{0.426}{(2\alpha+1)E_{Cd}^{\alpha}}}{\left(\frac{I_{02}}{\sigma_{02}} - 0.426\right) (\overline{E_{r2}})^{-\alpha} + \frac{0.426}{(2\alpha+1)E_{Cd}^{\alpha}}} \tag{6}$$

Note that:

$$Q_{0i}(\alpha) = \frac{Q_{0i} - 0.426}{E_{ri}^{\alpha}} + \frac{0.426}{(2\alpha + 1).0.55^{\alpha}}$$

Equation 6 can be written as:

$$\frac{(R_{Cd2} - 1)}{(R_{Cd1} - 1)} = \frac{Q_{01}(\alpha)}{Q_{02}(\alpha)} \tag{7}$$

Where i is denotes the i th monitor and R_{Cdi} is Cd ratio of i th monitor.

Substituting Eq. 5 into Eq. 7, it can be written:

$$\frac{(R_{Cd2} - 1)}{(R_{Cd1} - 1)} = \frac{Q_{01}(\alpha)}{Q_{02}(\alpha)} = \frac{Q_{01} \exp(-a_1(\ln \overline{E_{r1}})\alpha)}{Q_{02} \exp(-a_2(\ln \overline{E_{r2}})\alpha)} \tag{8}$$

and thus, α parameter can be written:

$$\alpha = \frac{1}{(a_2 \ln \overline{E_{r2}} - a_1 \ln \overline{E_{r1}})} \ln \left(\frac{(R_{Cd2} - 1)Q_{02}}{(R_{Cd1} - 1)Q_{01}} \right) \tag{9}$$

In the case of Cd-covered co-irradiation of two-detector, from base activation equation, α is easily written:

$$\alpha = \frac{1}{(a_2 \ln \overline{E_{r2}} - a_1 \ln \overline{E_{r1}})} \ln \left(\frac{A_{sp1}k_{0Au(1)}\varepsilon_2Q_{02}}{A_{sp2}k_{0Au(2)}\varepsilon_2Q_{01}} \right) \tag{10}$$

$$A_{spi} = \frac{A_{pi}}{wSDC}$$

where A_{pi} —measured average activity of the full-energy peak, $A_{pi} = N_{pi}/t_m$ with N_{pi} —net number of counts under the full-energy peak collected during measuring time t_m ; w —weight of the irradiated element; $S = 1 - e^{-\lambda t_{irr}}$; λ = decay constant, t_{irr} = irradiation time; $D = e^{-\lambda t_d}$; t_d = decay time; $C = \frac{1-e^{-\lambda t_m}}{\lambda t_m} k_{0Au(i)}$ is k_0 -factor of i th isotope to gold (see Ref. [3]) and ε_i is full-energy peak detection efficiency of energy E_i .

Thus, in experiment using pairs of ^{197}Au – ^{94}Zr and ^{197}Au – ^{64}Zn , we only need the determination of R_{Cdi} ratios (Cd-ratio method) or A_{spi} (Cd-covered irradiation only) of monitors. Then α will be calculated using Eqs. 9 or 10, respectively.

Error estimation of the method

Errors of the method should be estimated in two kinds: systematic and statistical errors. In this report, the errors due to approximation of Eq. 5 are considered as systematic errors, while the errors of the variables in Eqs. 9 and 10 using calculation of α are statistical errors. The ^{197}Au – ^{94}Zr and ^{197}Au – ^{64}Zn pairs were applied. The choice of ^{197}Au – ^{94}Zr and ^{197}Au – ^{64}Zn monitor pairs is very suitable for the experiment of the α -determination. The reason is the $\overline{E_{ri}}$ effective resonant energies of these isotopes are in wide region ($\overline{E_r} (^{197}\text{Au}) = 5.65 \text{ eV}$, $\overline{E_r} (^{64}\text{Zn}) = 2,560 \text{ eV}$, $\overline{E_r} (^{94}\text{Zr}) = 6,260 \text{ eV}$) and nuclear parameters are suitable for reactor irradiation. Moreover, the sample preparation is particularly easy, the product nucleus have a simple decay scheme and their cross-section have been determined in detail and with high accuracy.

Error estimation due to approximation of Eq. 5

Reviewing Tables 2 and 3, we see that the $Q_{0i}(\alpha)$ -values of $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ calculated from Eqs. 4 and 5 are in very

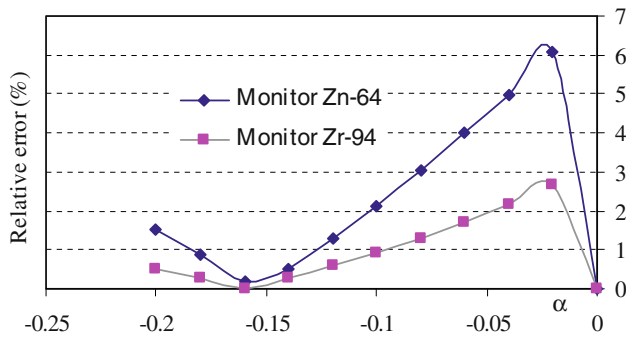


Fig. 1 Survey of ζ_α -values on α when $\alpha < 0$

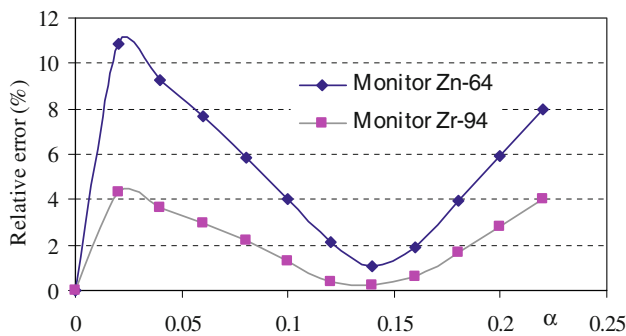


Fig. 2 Survey of ζ_α -values on α when $\alpha > 0$

good agreement with the difference of less than 0.08% in the range of $|\alpha| \leq 0.2$. We can think that they are quite coincident. The uncertainty of the α -values depends only on the differences of the $Q_{0i}(\alpha)$ -values of $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$ and $^{64}\text{Zn}(n, \gamma)^{65}\text{Zn}$ calculated from Eqs. 4 and 5, respectively. From Eq. 5, it can be written as:

$$\alpha = (\ln Q_0(\alpha) - \ln Q_0) / a \ln \bar{E}_r. \quad (11)$$

From the error propagation equation, the percentile error or uncertainty due to approximation of Eq. 5 can be written as:

$$\zeta_\alpha = \frac{\sigma_\alpha}{\alpha} = \frac{1}{\alpha} \frac{1}{a \ln \bar{E}_r} \frac{\Delta Q_0(\alpha)}{Q_0(\alpha)} \quad (12)$$

where σ_α is absolute uncertainty of α ; $\Delta Q_0(\alpha)$ is the difference of the $Q_{0i}(\alpha)$ -values calculated from Eqs. 4 and 5. From Eq. 5, ζ_α is dependent upon each isotope chosen as monitor and inverse proportion to α -value. The survey of ζ_α -values on α , in which $|\alpha| < 0.2$, for ^{94}Zr and ^{65}Zn were carried out in Figs. 1 and 2.

From Figs. 1 and 2, the systematic uncertainty (ζ_α) of the ^{197}Au - ^{94}Zr pair is lower than that of ^{197}Au - ^{64}Zn one. It is obvious that Q_0 of ^{94}Zr ($Q_0 = 5.306$) is bigger than one of ^{64}Zn ($Q_0 = 1.908$). Furthermore, $\Delta Q_0(\alpha)$ of ^{64}Zn calculated from Eqs. 4 and 5 is bigger than one of ^{94}Zr and the effective resonance energy of ^{94}Zr ($\bar{E}_{ri} = 6,260$ eV) is also bigger than the one of ^{64}Zn ($\bar{E}_{ri} = 2,560$ eV).

Statistical errors

This error can be estimated from the errors of the variables in Eqs. 9 or 10. The absolute uncertainty in α can be calculated from the uncertainties of the variables (denoted x_j) which determine α in Eqs. 9 or 10:

$$\sigma_\alpha = \sqrt{\sum_j \sigma_{x_j}^2 \left(\frac{\partial \alpha}{\partial x_j} \right)^2} \quad (13)$$

where $\partial \alpha / \partial x_j$ and σ_{x_j} are the corresponding partial derivatives and the uncertainties of x_j -variables, respectively.

According to the customary error propagation theory, the error propagation functions can be written as:

$$Z_\alpha(x_j) = \left| \left(\frac{\partial \alpha}{\alpha} \right) / \left(\frac{\partial x_j}{x_j} \right) \right| = \left| \frac{\partial \alpha}{\partial x_j} \frac{x_j}{\alpha} \right|. \quad (14)$$

And relative error:

$$s_\alpha(x_j) = Z_\alpha(x_j) \frac{\Delta x_j}{x_j}. \quad (15)$$

Applying the above formula Eqs. 14–9, we get:

$$Z_\alpha(\bar{E}_{r1}) = \left| \frac{a_1}{a_2 \ln \bar{E}_{r2} - a_1 \ln \bar{E}_{r1}} \right| \quad (16)$$

$$Z_\alpha(\bar{E}_{r2}) = \left| \frac{a_2}{a_2 \ln \bar{E}_{r2} - a_1 \ln \bar{E}_{r1}} \right| \quad (17)$$

$$Z_\alpha(a_1) = \left| \frac{a_1 \ln \bar{E}_{r1}}{a_2 \ln \bar{E}_{r2} - a_1 \ln \bar{E}_{r1}} \right| \quad (18)$$

$$Z_\alpha(a_2) = \left| \frac{a_2}{a_2 \ln \bar{E}_{r2} - a_1 \ln \bar{E}_{r1}} \right| \quad (19)$$

$$Z_\alpha(Q_{01}) = Z_\alpha(Q_{02}) = \left| \frac{1}{\alpha a_2 \ln \bar{E}_{r2} - a_1 \ln \bar{E}_{r1}} \right| \quad (20)$$

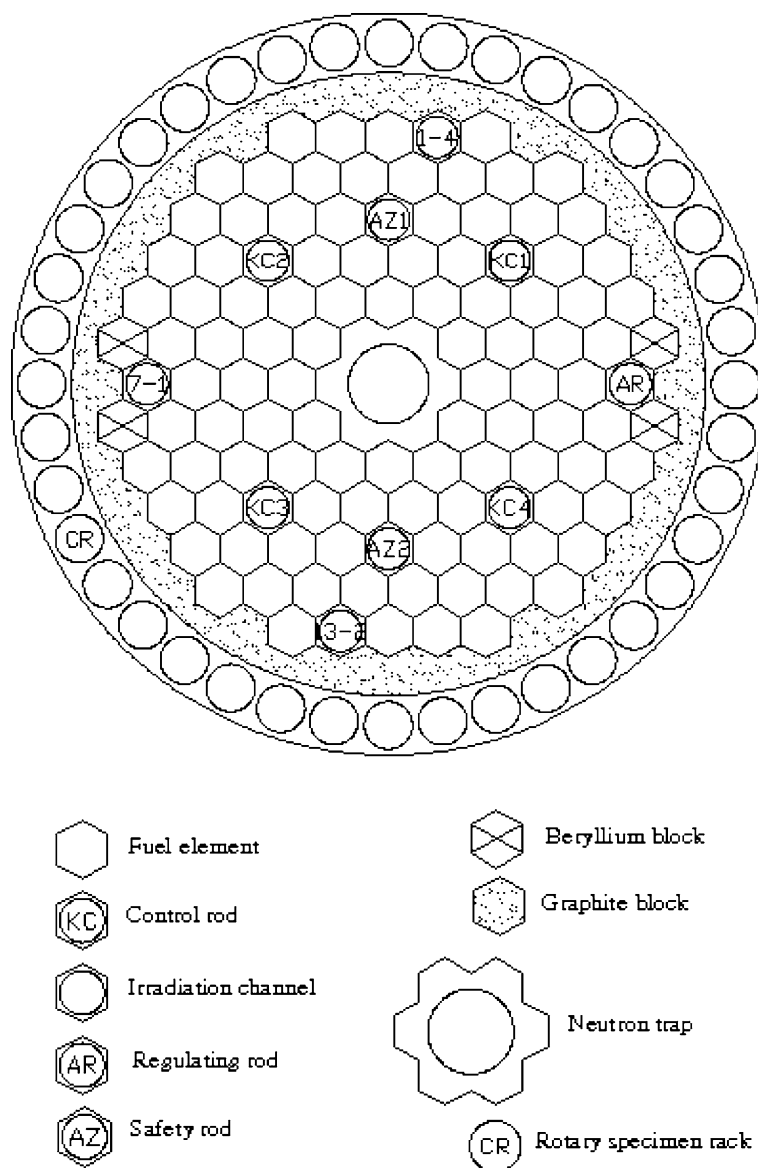
$$Z_\alpha(R_{Cd1}) = \left| \frac{1}{\alpha (R_{Cd1} - 1) (a_2 \ln \bar{E}_{r2} - a_1 \ln \bar{E}_{r1})} \right| \quad (21)$$

$$Z_\alpha(R_{Cd2}) = \left| \frac{1}{\alpha (R_{Cd2} - 1) (a_2 \ln \bar{E}_{r2} - a_1 \ln \bar{E}_{r1})} \right|. \quad (22)$$

In the case of Cd-covered co-irradiation of two-detector, the α value is calculated using Eq. 10, and we obtain Eqs. 16–19 for the error propagation functions of \bar{E}_{ri} and a_i . The other variables can be calculated as following:

$$\begin{aligned} Z_\alpha(A_{sp1}) &= Z_\alpha(A_{sp2}) = Z_\alpha(k_{0Au(1)}) = Z_\alpha(k_{0Au(2)}) \\ &= Z_\alpha(\varepsilon_1) = Z_\alpha(\varepsilon_2) = \left| \frac{1}{\alpha (a_2 \ln \bar{E}_{r2} - a_1 \ln \bar{E}_{r1})} \right|. \end{aligned} \quad (23)$$

Fig. 3 Ground plan of Dalat reactor



Experimental

As the first check, in this work, the ^{197}Au – ^{94}Zr pair was applied. The \overline{E}_{ri} effective resonant energies of these isotopes are in wide region ($\overline{E}_{r}(^{197}\text{Au}) = 5.65 \text{ eV}$, $\overline{E}_{r}(^{96}\text{Zr}) = 248 \text{ eV}$, $\overline{E}_{r}(^{94}\text{Zr}) = 6,260 \text{ eV}$) and nuclear parameters are suitable for reactor irradiation. Moreover, the irradiation of these monitor foils with bare and Cd-cover, we can simultaneously determine the α value by “bare multi-monitor” method as presented in [3], as a comparison. The experiment of the α -determination was carried out in the channels 7-1, 1-4 and neutron trap of Dalat reactor. These irradiation positions are situated in reactor core (Fig. 3).

As standard materials, 0.0115% Au–Al wire (diam. 1 mm) and high-purity Zr foils of 0.127 mm thickness

were used. In this case, the monitor foils of Au and Zr with bare and Cd-covered were simultaneously irradiated in the mentioned reactor channels. The irradiation durations were 20 min and 1 h, respectively. In both cases, decaying and measuring time were 20–24 h and 30 min. The counting was performed with a 70 cm³ coaxial GeHP detector paired to a 4096 channels analyzer. The results which were summarized in Table 4, were compared with those obtained by the three-detector method without Cd [3] and by a α -determination method using neutron spectrum calculated by MCNP code [9].

As an example for the error estimation of α , we carried out the results of the error estimation of α in 7-1 channel of Dalat reactor using R_{Cd} method (Eq. 9). Indeed, with $\alpha = 0.044$, the uncertainty of R_{Cd} in the experiment about 1%, the uncertainties Q_{oi} and \overline{E}_{ri} from report [6, 7], we

Table 4 α -Values in irradiation channels of Dalat reactor

Channels	$^{197}\text{Au}-^{94}\text{Zr}$ (Eq. 9), α (10^{-2})	$^{197}\text{Au}-^{94}\text{Zr}$ (Eq. 10), α (10^{-2})	Three-detector method with Cd, α (10^{-2})	Calculated α (10^{-2}) [9]
Neutron trap	-3.1 ± 0.5	-3.3 ± 0.5	-3.5 ± 1.0	-3.3 ± 0.5
1-4 Channel	-3.6 ± 0.5	-3.4 ± 0.5	-3.8 ± 1.1	-3.4 ± 0.5
7-1 Channel	-4.4 ± 0.5	-4.5 ± 0.5	-4.8 ± 1.0	-4.8 ± 0.4

used Eqs. 16–22 and obtained: $s_x(\overline{E_{rZr}}) \approx 1.4\%$, $s_x(\overline{E_{rAu}}) \approx 1.4\%$, $s_x(a_{Au}) \approx 0.03\%$, $s_x(a_{Zr}) \approx 0.13\%$, $s_x(Q_{0Au}) \approx 6.5\%$, $s_x(Q_{0Zr}) \approx 7\%$, $s_x(R_{CdAu}) \approx 5.2\%$, $s_x(R_{CdZr}) \approx 7.6\%$. The uncertainty of α in total was about 13% (with $\zeta_x \approx 2\%$). Similarly, in the case using the method of Cd-covered co-irradiation of two-detectors ($\alpha = 0.045$), the uncertainty of α estimated from Eqs. 16–20 and Eq. 23 was about 15%, whereas using of the three-detector method without Cd, the uncertainty of α was about 30%.

The comparison of the α values in Table 4 shows that the values calculated from Eqs. 9 or 10 are in good agreement with each other. Moreover, they agree well with α -determination method using neutron spectrum calculated by MCNP code [9] and the three-detector method without Cd.

Conclusion

From Table 4, it is obvious that the modified method presented in this report is suitable for rapid α -determination in experiment. This is in complete agreement with the results of the other methods. Moreover, the results from studying on the error due to approximation of Eq. 5 suggest

that in case absolute α value is less than 0.25 (this condition is satisfactory in irradiation channels located in the reactor core), the using of this method is quite possible with reasonable errors.

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