AN ALTERNATIVE APPROACH TO THE DETERMINATION OF
EPITHERMAL FLUX-SHAPING FACTOR (α) FOR k_0-NAA

Y.A. Ahmed¹
Centre for Energy Research and Training, Ahmadu Bello University, Zaria, Nigeria
and
The Abdus Salam International Centre for Theoretical Physics, Trieste, Italy,

I.M. Umar, I.O.B. Ewa, A.S. Ajuji
Centre for Energy Research and Training, Ahmadu Bello University, Zaria, Nigeria,

B.J.B. Nyarko and E.H.K. Akaho²
National Nuclear Research Institute, Ghana Atomic Energy Commission, Accra, Ghana.

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¹ Junior Associate of ICTP. Corresponding author: yaahmed1@gmail.com
Abstract

The deviation of the epithermal flux spectra at reactor irradiation channel from the ‘ideal’ 1/E-shape to the ‘non-ideal’ $1/E^{1+\alpha}$ has to be corrected when applying $k_0$ standardization of the neutron activation analysis. Cadmium ratio method is one of the popular methods used in the determination of this deviation (flux-shaping) factor but the equations involved demand iteration or graphical solution. Interactive computer programs (in BASIC and C++) were developed in this study to ease the otherwise cumbersome and challenging method of solving the cadmium ratio equations. The programs developed were used in the determination of the epithermal flux-shaping factor ($\alpha$) for inner irradiation channel 5 and outer irradiation channel 7 of the Ghana Research Reactor-1 by irradiating Gold and Zirconium foils with and without Cadmium cover. The programs were also used to reproduce results reported earlier for inner channel 1 and outer channel 7 of the same reactor as a test of performance. The results obtained are – 0.107 for inner channel 5, -0.1246 for inner channel 1, -0.0241 for outer channel 6 and – 0.173 for outer channel 7. This result suggests that the interactive and less-cumbersome iteration programs in this study can be adopted for the determination of epithermal flux-shaping factor ($\alpha$) since the values obtained follow similar trends reported for low power research reactors and the programs were able to reproduce results reported for the earlier characterized channels of the same reactor.
Introduction
From the various experimental methods available for the determination of the epithermal flux-shaping factor ($\alpha$), the Cd ratio method is known to yield accurate results\(^1\). However, the equations involved demand iteration methods, which can manually be cumbersome and results take time to be achieved.

The knowledge of $\alpha$-values in irradiation channel is necessary\(^2\) for the conversion of resonance integral ($I_o$) and other parameters like the ratio of resonance integral to thermal cross-section ($Q_0$), valid only in ideal situation, to $I_o$ ($\alpha$) and $Q_0$ ($\alpha$) respectively for application in real reactor neutron activation analysis situations. The epithermal flux-shaping factor is applied mainly in the calibration of an irradiation channel for nuclear data measurements\(^3\) and in performing reactor-neutron activation without standards\(^4\) (e.g., using the $k_o$-standardization method). The latter requires the most accurate $\alpha$-determinations possible like the “Cd-Covered” method or the “Cd-ratio” method\(^5\). For most analytical cases, the Cd ratio method using the $^{197}$Au-$^{96}$Zr-$^{94}$Zr sets provide acceptable precision and accuracy as tested and verified extensively (Table 1) in the Ghent Thesis reactor and the Budapest WWR-M reactor\(^2\) -\(^5\) and recently used for the channels of the GHARR-1\(^6\) -\(^9\), the Korean HANARO Research Reactor\(^10\) and the Nigerian Research Reactor-1\(^11\).

Theory
In terms of the HOGDAHL convention\(^12\) the concentration of an element by instrumental NAA ($k_0$-standardization method) for ideal spectra (Gold-comparator data with an asterisk) is given by:

$$\text{Conc(ppm)} = \frac{I_{sp} \cdot \varepsilon_{\gamma} \cdot \{f + Q_0^\ast\}}{I_{sp} \cdot k_o \cdot \varepsilon_{\gamma} \cdot \{f + Q_o\}}$$

$$I_{sp} = \frac{N_p}{t_m \cdot S \cdot D \cdot C \cdot m}$$

where $N_p$ = net peak counts, $t_m$ = measurement time, $m$ = sample weight,

$k_o$ = k-zero of gold, $\varepsilon_{\gamma}$ = detector efficiency

$S$ = saturation factor = $1 - e^{-\lambda t_r}$

$D$ = decay factor = $e^{-\lambda t_d}$

$C$ = counting factor = $\left(1 - e^{-\lambda t_c}\right)$

$\lambda = \frac{Ln2}{T_{1/2}}$

$\lambda = \frac{Ln2}{T_{1/2}}$

$t_r$ = irradiation time, $t_d$ = decay time, $t_c$ = counting time.
Table 1: Experimentally determined values of $\alpha$ for different research reactors

<table>
<thead>
<tr>
<th>Research Reactor</th>
<th>Monitors</th>
<th>Chann</th>
<th>$\alpha$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>TRIGA MARK II Ljubljana, Yugoslavia.</td>
<td>$^{197}$Au-$^{94}$Zr</td>
<td>CC</td>
<td>-0.057±0.010</td>
<td>[2]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>PT</td>
<td>-0.046±0.011</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>CR25</td>
<td>-0.014±0.006</td>
<td></td>
</tr>
<tr>
<td>WWR-M, Budapest, Hungary</td>
<td>$^{197}$Au-$^{238}$U-$^{98}$Mo-$^{100}$Mo-$^{64}$Zn</td>
<td>MILA</td>
<td>-0.014±0.006</td>
<td>[3]</td>
</tr>
<tr>
<td>DR-3, RisØ, Denmark</td>
<td>$^{197}$Au-$^{238}$U-$^{98}$Mo-$^{100}$Mo-$^{64}$Zn</td>
<td>R4V4</td>
<td>-0.158±0.11</td>
<td>[3]</td>
</tr>
<tr>
<td>THETIS, Gent, Belgium</td>
<td>$^{197}$Au-$^{238}$U-$^{98}$Mo-$^{100}$Mo-$^{64}$Zn</td>
<td>7</td>
<td>-</td>
<td>[3]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>14</td>
<td>0.0713±0.0088</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>16</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.0298±0.0089</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.0792±0.0089</td>
<td></td>
</tr>
<tr>
<td>SLOWPOKE-2 Halifax, Canada</td>
<td>$^{197}$Au-$^{94}$Zr-$^{96}$Zr</td>
<td>5</td>
<td>-0.0427</td>
<td>[6]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1</td>
<td>-0.0098</td>
<td></td>
</tr>
<tr>
<td>GHARR-1, Accra, Ghana</td>
<td>$^{197}$Au-$^{94}$Zr-$^{96}$Zr</td>
<td>1</td>
<td>-0.104</td>
<td>[6]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6</td>
<td>-0.026</td>
<td></td>
</tr>
<tr>
<td>CIRUS, Bhabha Atomic Centre, India</td>
<td>$^{197}$Au-$^{94}$Zr-$^{96}$Zr</td>
<td>PT</td>
<td>-0.011±0.004</td>
<td>[4]</td>
</tr>
</tbody>
</table>

However, owing to non-ideality of neutron spectra, the expression yields:

\[
\text{Conc}(ppm) = \frac{I_{sp} e^*_r \left\{ f + Q_o^*(\alpha) \right\}}{I_{sp} k_o e^*_r \left\{ f + Q_o(\alpha) \right\}}
\]

as $Q_o$ transforms to an alpha-dependent term using the following identities:

\[
E_r \to \overline{E}_r^\alpha, \quad I_o \to I_o(\alpha), \quad Q_o \to Q_o(\alpha)
\]

for which we obtain the following expressions:

\[
I_o(\alpha) = \left\{ \frac{I_o - 0.429\sigma_o}{\overline{E}_r^\alpha} + \frac{0.429\sigma_o}{(2\alpha + 1)\overline{E}_{cd}^\alpha} \right\} \overline{E}_{cd}^\alpha
\]

and

\[
Q_o(\alpha) = \frac{Q_o - 0.429}{\overline{E}_r^\alpha} + \frac{0.429}{(2\alpha + 1)\overline{E}_{cd}^\alpha}
\]

where $E_r$ = effective resonance energy \( \overline{E}_{cd} = \text{Cd cut-off energy} \approx 0.55\text{eV} \)

From the above expressions \( \Rightarrow \) alpha for the reactor channels is required.
The Triple monitor set of $^{197}\text{Au}-^{94}\text{Zr}-^{96}\text{Zr}$ allows for quick determination of alpha and measurement of $R_{Cd}$ of Au and Zr via their $I_{sp}$ values. When two or more monitors are irradiated under uniform flux (i.e., $f_1 \approx f_2$) with and without Cd cover alternately, their Cd ratio equation is of the form:

$$f = Q_{0,i} (\alpha) F_{c,i} (R_{Cd,i} - 1) \frac{G_{e,i}}{G_{th,i}}$$

Under uniform flux, $f_1 \approx f_2$ and for $G_{e,i} \approx G_{th,i} \approx 1$

$$\frac{f_1}{f_2} \approx 1 \approx \frac{(R_{Cd} - 1)_1}{(R_{Cd} - 1)_2} = \frac{Q_{0,2} (\alpha)}{Q_{0,1} (\alpha)} = f(\alpha) \quad (3A)$$

The epithermal deviation factor could be obtained from equation (3A) or by the slope of the straight line when plotting graph of:

$$\log\left(\frac{E_{r,i}^{\alpha}}{k_{0,Au}(i)E_{p,j}F_{c,j}Q_{0,j}(\alpha)G_{e,j}}\right) \quad \text{Versus} \quad \log(E_{r,i}) \quad (3B)$$

where $i = \text{isotope} 1, 2, 3, \ldots N \quad G_{e,i} = \text{epithermal neutron self-shielding factor for the ith monitor, } G_{th,i} = \text{thermal neutron self-shielding factor for the ith monitor. Other terms are as defined above.}$

Recent authors in this field preferred the slope method (Equation 3B) due to the fact that the manual iteration of Equation 3A could be cumbersome because of large possibility of trial-values for several step lengths. This work simplifies this iteration by developing simple computer programs.

**The Q-Basic program**

It was developed based on the linear method. To iterate the function $f(\alpha)$ (Equation 3A) within a specified step-length (Equation 4)

$$\alpha_{n+1} = f(\alpha) \quad (4)$$

Choice of trial $\alpha$-values for every step increment depends on the iteration result obtained at each step and finite point of convergence. It was noticed that a trial value of -0.5 makes the function undefined (non-convergence). To avoid this, the program is designed to skip oscillatory points (Fig. 1), assign a unity solution to it, and send a warning signal to the user. The result of this program shows that, it is slow in converging and so a faster method had to be developed in C++ language.
The C++ Program

The program estimates the number of iterations necessary to achieve convergence for a given function by relying on the derivative of the function (normally yields a linear first order convergence). If \( f'(\alpha) \) is not equal to zero, it indicates that it does not vanish at the solution [a divergence situation (Equation 7)]. In situations like this, we require that the function \( f(\alpha) \) be differentiable, its graph should have a definite slope at each point and with a unique tangent line (Fig. 2). By extending the tangent line to the curve of \( f(\alpha) \) at an initial alpha value say \( \alpha_0 \) to intersect the \( \alpha \)-axis at \( \alpha_1 \) and similarly \( \alpha_n \) to \( \alpha_{n+1} \), we obtain an acceptable value of alpha that satisfies \( f(\alpha) \). Analytically it means:

\[
\alpha_1 = \alpha_0 - \delta \alpha
\]

\ldots

\[
\alpha_n = \alpha_{n-1} - \delta \alpha \quad \ldots \quad \alpha_{n+1} = \alpha_n - \delta \alpha
\]  

(5)

The tangent to the curve at point \( \{\alpha_0 f(\alpha_0)\} \) on the graph (Fig. 2) is thus:

\[
f'(\alpha_0) = \frac{f(\alpha_0)}{\delta \alpha} \Rightarrow \delta \alpha = \frac{f(\alpha_0)}{f'(\alpha_0)}
\]  

(6)

So that substituting Equation 6 into 5 we have:

\[
\alpha_1 = \alpha_0 - \frac{f(\alpha_0)}{f'(\alpha_0)}
\]  

(7)

In general Equation 7 could be written as:

\[
\alpha_{n+1} = \alpha_n - \frac{f(\alpha_n)}{f'(\alpha_n)}
\]  

(8)

The Newton-Raphson method\(^\text{14}\) was found to converge faster than the linear method of the Q-Basic program. However, it fails to converge if \( f'(\alpha_n) \) becomes very small or vanishes and/or second term in the RHS of Equation 8 becomes large (causing the iteration to diverge and derivative of the function in Equation 8 vanishes). This is another difficulty that encourages the use the divided difference approximation method usually known\(^\text{14}\) as Secant method.

The secant method iterates between any two selected points (Equation 10), say \( \alpha_{n-1} \) and \( \alpha_n \), and a Secant line that intersect \( \alpha \)-axis at a point say \( \alpha_{n+1} \) is drawn. The \( f'(\alpha_n) \) in Equation 8 is replaced by the definition of \( f'(\alpha) \).

Thus:

\[
f'(\alpha) = \lim_{y \to 0} \frac{f(\alpha + y) - f(\alpha)}{y} \approx \frac{f(\alpha + y) - f(\alpha)}{y}
\]  

(9)

If \( \alpha = \alpha_n, y = \alpha_{n-1} - \alpha_n \) then Equation 8 transforms to:

\[
f'(\alpha_n) \approx \frac{f(\alpha_{n-1}) - f(\alpha_n)}{\alpha_{n-1} - \alpha_n}
\]  

(10)
So that on substituting Equation 10 in Equation 8 we have:

\[ \alpha_{n+1} = \alpha_n - \frac{f(\alpha_n)(\alpha_n - \alpha_{n-1})}{f(\alpha_n) - f(\alpha_{n-1})} = \frac{\alpha_{n-1}f(\alpha_n) - \alpha_n f(\alpha_{n-1})}{f(\alpha_n) - f(\alpha_{n-1})} \]  

(11)

This was the method adopted for the development of the C++ Computer program used for the determination of the epithermal flux-shaping factor (\( \alpha \)) in this work.

Fig. 1 Oscillatory behaviour of \( f(\alpha) \) for a linear iteration

Fig. 2 \( f(\alpha) \) for Newton-Raphson and Secant methods
Table 2: Nuclear parameters of suitable monitors

<table>
<thead>
<tr>
<th>Channel</th>
<th>Monitor</th>
<th>$Q_o$</th>
<th>$E_r$</th>
<th>$E_{cd}$</th>
<th>$R_{cd}$ (Expt.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5 (Inner)</td>
<td>Au-197</td>
<td>15.71</td>
<td>5.65</td>
<td>0.55</td>
<td>2.10</td>
</tr>
<tr>
<td></td>
<td>Zr-96</td>
<td>251.6</td>
<td>338</td>
<td>0.55</td>
<td>2.68</td>
</tr>
<tr>
<td></td>
<td>Zr-94</td>
<td>5.06</td>
<td>6260</td>
<td>0.55</td>
<td>2.68</td>
</tr>
<tr>
<td>7 (Outer)</td>
<td>AU-197</td>
<td>15.71</td>
<td>5.65</td>
<td>0.55</td>
<td>3.57</td>
</tr>
<tr>
<td></td>
<td>Zr-96</td>
<td>251.6</td>
<td>338</td>
<td>0.55</td>
<td>3.57</td>
</tr>
<tr>
<td></td>
<td>Zr-94</td>
<td>5.06</td>
<td>6260</td>
<td>0.55</td>
<td>3.52</td>
</tr>
</tbody>
</table>

Experimental

Four samples of standard Gold solution and four of Zirconium foils with masses between 4.7 and 5.0 mg were prepared and packed into plastic polyethylene vial for irradiation. Two of the prepared samples each of Au (one Cd covered and one bare) and Zr were sent to the outer irradiation channel (No.7) and another two sets to the inner irradiation channel (No.5) of GHARR-1 research reactor for one-hour irradiation. The samples were irradiated with the reactor operating at half thermal power of 15 kW with neutron flux of $5 \times 10^{11}$ n cm$^{-2}$ s$^{-1}$ in one of the inner irradiation channels. The nuclear parameters for the monitors are shown in Table 2. Counting of the samples was carried out at a geometry of 7.2 cm from the top of the detector using a gamma-ray spectroscopy system consisting of an N-Type High Purity Germanium (HPGe) detector model GR2518 with relative efficiency of 25% and an energy resolution of 1.8 keV (FWHM) at 1332.5 keV gamma ray of $^{60}$Co. The efficiency parameters of the detector were obtained by fitting the efficiency curve with polynomial function using parameters determined earlier$^{15}$.

Results and Discussion

For completeness and comparison of the merits of each computer program described above, both Q-Basic and C++ programs developed were used in this work to determine the alpha of the inner channel (5) and outer channel (7) of the Ghana Research Reactor-1 (GHARR-1) with Au and Zr serving as our monitor pairs. The epithermal flux-shaping factor ($\alpha$) obtained in this study is $-0.107$ for the inner channel No. 5 and $-0.173$ for the outer channel No. 7 of the Ghana Research Reactor-1 (Table 4).
Table 3: Iteration results obtained for four channels of GHARR-1

<table>
<thead>
<tr>
<th>Program</th>
<th>Channel No</th>
<th>$R_{cd}$ Ratio</th>
<th>$Q_0(\alpha)$Ratio</th>
<th>Deviation</th>
<th>$\alpha$-Value</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>C++</td>
<td>5 (inner)</td>
<td>0.6547619</td>
<td>0.6547619</td>
<td>Nil</td>
<td>-0.1077</td>
<td>This work</td>
</tr>
<tr>
<td>C++</td>
<td>7 (outer)</td>
<td>1.019841</td>
<td>1.019841</td>
<td>Nil</td>
<td>-0.17308</td>
<td>“</td>
</tr>
<tr>
<td>Q-Basic</td>
<td>5 (inner)</td>
<td>0.6547619</td>
<td>0.6547619</td>
<td>Nil</td>
<td>-0.1076</td>
<td>“</td>
</tr>
<tr>
<td>C++</td>
<td>1 (inner)</td>
<td>0.7351351</td>
<td>0.7351351</td>
<td>Nil</td>
<td>-0.1246</td>
<td>This work</td>
</tr>
<tr>
<td>Q-Basic</td>
<td>1 (inner)</td>
<td>0.7351351</td>
<td>0.7351351</td>
<td>Nil</td>
<td>-0.1249</td>
<td>“</td>
</tr>
<tr>
<td>Unreported</td>
<td>6 (outer)</td>
<td>0.3813946</td>
<td>0.3762887</td>
<td>0.0051</td>
<td>-0.0261</td>
<td>[6]</td>
</tr>
<tr>
<td>C++</td>
<td>6 (outer)</td>
<td>0.3813946</td>
<td>0.3813946</td>
<td>Nil</td>
<td>-0.0241</td>
<td>This work</td>
</tr>
<tr>
<td>Q-Basic</td>
<td>6 (outer)</td>
<td>0.3813946</td>
<td>0.3813946</td>
<td>Nil</td>
<td>-0.0240</td>
<td>“</td>
</tr>
</tbody>
</table>

The results reported earlier\textsuperscript{6} for other channels of the same reactor were also recalculated to validate the reliability of the new programs in this study. Although no information as to the type of method adopted by Akaho and Nyarko 2002 for iteration, yet, when the reported input nuclear data were computed with the C++ and Q-Basic programs of this study, the Akaho and Nyarko 2002 results were reproduced except for the inner channel 1(Table 3) which has a deviation of about 9.6%. The deviation could be attributed to the $Q_0(\alpha)$ ratio (0.6387489), which was found to have a difference of 0.0963862 from the $R_{cd}$ ratios (0.7351351) of the two monitors used. For an iteration program, which normally allows a convergence at a null difference between the $Q_0(\alpha)$ and cadmium ratios, a difference of that order is quite significant. However, the results can be said to have similar trend as obtained for low power research reactors (Table 1).
Table 4: Epithermal flux-shaping factor determined for GHARR-1 research reactor.

<table>
<thead>
<tr>
<th>Channel No.</th>
<th>Monitor</th>
<th>$Q_\gamma(\alpha)$</th>
<th>$E_r$</th>
<th>$\alpha$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>5(inner)</td>
<td>$^{197}$Au</td>
<td>18.91788</td>
<td>5.65</td>
<td>- 0.107</td>
<td>This Work</td>
</tr>
<tr>
<td></td>
<td>$^{96}$Zr</td>
<td>4711.0421</td>
<td>338</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$^{94}$Zr</td>
<td>12.39631</td>
<td>6260</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1(inner)</td>
<td>$^{197}$Au</td>
<td>18.793</td>
<td>5.65</td>
<td>- 0.104</td>
<td>[6]</td>
</tr>
<tr>
<td></td>
<td>$^{96}$Zr</td>
<td>446.826</td>
<td>338</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$^{94}$Zr</td>
<td>12.615</td>
<td>6260</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7(outer)</td>
<td>$^{197}$Au</td>
<td>21.2039</td>
<td>5.65</td>
<td>- 0.173</td>
<td>This work</td>
</tr>
<tr>
<td></td>
<td>$^{96}$Zr</td>
<td>689.209</td>
<td>338</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$^{94}$Zr</td>
<td>21.64129</td>
<td>6260</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6(outer)</td>
<td>$^{197}$Au</td>
<td>16.423</td>
<td>5.65</td>
<td>- 0.0261</td>
<td>[6]</td>
</tr>
<tr>
<td></td>
<td>$^{96}$Zr</td>
<td>290.599</td>
<td>338</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$^{94}$Zr</td>
<td>6.336</td>
<td>6260</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Conclusion**

The two programs developed in this study were used to determine the epithermal flux-shaping-factor ($\alpha$) for the inner channel 5 and the outer channel 7 of the Ghana Research Reactor-1. The results obtained for these channels were – 0.107 for channel 5 and – 0.173 for channel 7. This result suggests that the interactive and less-cumbersome iteration programs in this study can be adopted for the determination of epithermal flux-shaping factor ($\alpha$) since the values obtained follow similar trends reported for low power research reactors and the programs were able to reproduce results reported for the earlier characterized channels of the same reactor.

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References