Some Studies in Activation Foils for Measuring Low-Level Induced Activity for the Neutron Radiation Protection

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Abstract. For the measurement of induced activity in the activation foils, various methods and instruments are used. When the activity is measured by using a beta counter it is very important to know the beta attenuation in the activation foils. The beta attenuation in the foil depends on the beta spectrum of the activation product, the material and the thickness of the material. The beta attenuation has been evaluated theoretically as well as experimentally. The detailed investigations of beta attenuation in the foils are presented in this paper. The experimental study was conducted using an absolute system, the $4\pi\beta$-$\gamma$ coincidence system. For the various foils like Au, Mn, Co, Al and Mg used in this study, it is found that both theoretical and experimental values for the beta attenuation are matching. The attenuation of betas in the foils varies with beta energy spectrum and the materials and it varies from 7% to 92%. The foil activation method is very accurate for the neutron fluence rate measurement. In foil activation method the induced activity is correlated to the neutron fluence rate. This activity is measured using different counters. $4\pi\beta$-$\gamma$ coincidence system is the primary standard for the activity measurement. If the activity is very low this system cannot be used for the measurement. $4\pi\beta$ gas flow type proportional counters are used for the measurement of activity. For this the efficiency of the foil should be known accurately. Evaluation of the beta efficiency of different activation foils is discussed in this paper. Gold, Manganese and Cobalt foils are used for thermal neutrons and Magnesium and Aluminium foils are used for fast neutron measurements.

KEYWORDS: Beta attenuation, induced activity, activation foils, $4\pi\beta$ proportional counter, $4\pi\beta$-$\gamma$ coincidence system, neutron fluence.

1. Introduction

Materials, which are activated by exposure to neutrons, can serve in principle as neutron detector\textsuperscript{[1]}. The activity of the product radioisotope can often be used for the measurement of the amount and energy distribution of neutron fluence rate which induced the radioactivity. In neutron measurements by induced activity, the stable material is exposed to the neutron fluence rate for a measured period of time. Following the irradiation, the material is removed from the neutron field, and a determination is made of the activity induced in it. Through the choice of materials with different cross-sections, it is possible to make measurements down to low fluxes and up to the largest values obtainable with a controlled reactor. The detector may be thin foils or small pellets and consequently it is possible to introduce them in the medium in which the measurements are to be made without the introduction of voids. Further, the effect of neutron absorption can be made negligible either by the selection of the foil or by application of a small correction. Since the irradiations are made first and the activity measurements follow at a later time, it is sometimes possible to obtain measurements at locations where it would be otherwise very difficult or impossible to instrument them. The cross-section for the production of radioactivity varies with the neutron energy; different foils have different response curves\textsuperscript{[2]}. Therefore it is possible to select, within limits, materials appropriate for various neutron energy ranges. The induced activities in the foils can be measured using a $4\pi\beta$-$\gamma$ coincidence system, the primary standard for the activity measurement. But when the activity is low, it is difficult to standardize the activity using this system, $4\pi\beta$ gas flow system is used as it has good efficiency. The beta rays undergo significant self-absorption for the usual thicknesses. The theoretical calculation of the beta attenuation of gold, manganese, cobalt, magnesium and aluminium foils and its practical verification using $4\pi\beta$ flow type proportional counter is discussed in this paper.
2. Materials and Methods

2.1 Gold ($^{197}$Au$^{197}$) \[ \text{Reaction: }^{197}$Au (n, $\gamma$) $^{198}$Au \]

Gold is the most widely used detector for low-energy neutron fluence measurements and is often chosen as a standard.\[^3\] Gold can be obtained with very high purity and the natural abundance for the $^{197}$Au isotopes is 100%. The cross-section is 98.8 barns for neutrons of 0.025eV and can reach 3x10^4 barns for 4.9eV, which is the resonance energy. The resonance integral is 1560 barns with a 44-barn component for the ‘1/v’ part of the cross-section curve. Measurements with thick gold foils are not valid unless special methods are used. The specific gravity of gold is large and the 0.96 MeV beta rays undergo significant self absorption for the usual thickness. Beta ray counting therefore gives rise to some problems when activity is not uniformly distributed in the foil, as is the case for epithermal neutron measurements. Gamma-ray counting is therefore preferable. The radio-isotope produced, $^{198}$Au has a 2.7 days half-life and is a beta-gamma emitter. The maximum beta ray energy is 0.96 MeV and the gamma ray energy 0.412 MeV. Since the beta and gamma rays are emitted in coincidence, an absolute measurement of the $^{198}$Au is possible by the 4$\pi$ beta-gamma coincidence method. Gold is used bare and under cadmium shield to obtain the thermal and epithermal neutron fluences.

\[\text{Beta Efficiency of Various Thickness Gold Foils}\]

![Beta Efficiency of Various Thickness Gold Foils](image)

Fig.1

2.2 Manganese ($^{55}$Mn$^{55}$) \[ \text{Reaction: }^{55}$Mn(n, $\gamma$) $^{56}$Mn \]

Manganese is a good thermal neutron detector with a strictly 1/v cross-section in the thermal region. It is normally used in the form of pure metal or as Mn-Ni alloy. Its density is 7.3 gm/cm$^3$. The cross-section is 13.3 barns for neutrons of 0.025eV and can reach 8 barns at 337 eV, which is the resonance energy. The resonance integral is 14 barns with a 6 barn component for the ‘1/v’ part of the cross-section curve.\[^3\] The specific gravity of manganese is low compared with gold and the 2.85 MeV beta rays undergo less self absorption in thin foils or the usual thickness. Both gamma and beta-ray counting is preferable in manganese foils. The radio-isotope produced, $^{56}$Mn has a 2.58 hrs. half-life and is a beta-gamma emitter. The maximum beta ray energy is 2.85 MeV and the gamma ray energy 0.847 MeV. Since the beta and gamma rays are emitted in coincidence, an absolute measurement of the $^{56}$Mn is possible by the 4$\pi$ beta-gamma coincidence method. Manganese is also used bare and under cadmium shield to obtain the thermal and epithermal neutron fluences.
2.3 Cobalt ($\gamma$Co$^{59}$) 

Cobalt is also a thermal neutron detector with a strictly $1/v$ cross-section in the thermal region. It is used in the form of pure metal. Its density is $8.85\text{g/cm}^3$. The cross-section is $20.2\text{ barns}$ for neutrons of $0.025\text{eV}$\cite{2}. The specific gravity of cobalt is high; the $0.312\text{ MeV}$ low-energy beta rays undergo very high self absorption in the usual thickness. So gamma ray counting is preferable in cobalt foils. The radio-isotope produced, $^{60}\text{Co}$ has a 5.27 yrs. half-life and is a beta-gamma emitter. The single beta ray energy is $0.312\text{ MeV}$ and the gamma ray energies are $1.173\text{ MeV}$ and $1.332\text{ MeV}$. Since the beta and gamma rays are emitted in coincidence, an absolute measurement of the $^{60}\text{Co}$ is possible by the $4\pi$ beta-gamma coincidence method. Cobalt can also used bare and under cadmium shield to obtain the thermal and epithermal neutron fluences.
2.4 Magnesium ($^{24}$Mg)  

Reaction: $^{24}$Mg(n, p)$^{24}$Na

This element is sometimes chosen for giving information on the high energy part of the spectrum because it has a relatively high effective threshold (7MeV) and can be obtained easily with a suitable purity. The natural abundance for $^{24}$Mg is 79%. Its density is 1.738 g/cm$^3$. The average cross-section integrated over the fission spectrum is 1.46mb$^3$. The usual shape, like that of most of the fast neutron detectors is a pellet with 1 to 3 mm thickness and various diameters. Such discs are obtained by stamping from sheet. The irradiation produces radioisotope $^{24}$Na which has a 15 hour half-life and emits a 1.39 MeV beta ray and 1.37 and 2.75 MeV gamma rays. Either beta or gamma radiations can be counted. The absolute activity can be detected using $4\pi$$\beta$$\gamma$ coincidence system.

![Beta Efficiency of Various Thickness Magnesium Foils](image1.png)

Fig.4

2.5 Aluminium ($^{27}$Al)  

Reaction: $^{27}$Al(n, $\alpha$)$^{24}$Na

Aluminium can be obtained with a high purity. The natural abundance for the $^{27}$Al isotope is 100%. Its density is 2.699 g/cm$^3$. The usual detector shape is a metallic disc with various diameters and thickness. This reaction is used for high–energy neutrons. The effective threshold is about 8 MeV and the activity produced ($t_{1/2} = 15$ hrs.) is measured by beta (1.39 MeV) or gamma (1.37 MeV, 2.75 MeV) counting $^{24}$Mg(n, p)$^{24}$Na. The average cross-section for a fission spectrum is 0.73 mb$^3$.

![Beta Efficiency of Various Thickness Aluminium Foils](image2.png)

Fig.5
3. Calculation of the Beta Efficiency of the Foils

For accurate measurement of the dose, the induced activity should be measured precisely. For this activity measurement we can use a $4\pi \beta$ detector. When we use a $4\pi \beta$ detector, the $\beta$ attenuation in the foils should be known. This can be calculated mathematically \cite{7,8,9,10,11,12}. The present work consists of the theoretical calculation of the $\beta$ attenuation of different foils, and its experimental verification.

3.1 Theoretical Calculation

\[
\mu_{Al} = \frac{17}{(E_{\text{max}})^{1.43}} \text{ cm}^2/\text{gm}
\]

\[
\mu_z \propto (Z/A)_T \cdot e^{1/3} \cdot Z_{T}^{1/3}
\]

\[
\mu_{Al} \propto (Z/A)_{Al} \cdot e^{1/3} \cdot Z_{Al}^{1/3}
\]

\[
\mu_z = \frac{\mu_{Al} \cdot [(Z/A)_T \cdot e^{1/3} \cdot Z_{T}^{1/3}]}{[(Z/A)_{Al} \cdot e^{1/3} \cdot Z_{Al}^{1/3}]}
\]

Where, ‘$T$’ is the target nuclide, ‘$Al$’ aluminium, ‘$E_{\text{max}}.$’ is the maximum beta energy emitted by the product radioisotope, ‘$\mu_z$’ attenuation of the betas in the foil, ‘$\mu_{Al}$’ attenuation of the betas in aluminium.

Beta Efficiency, \( \varepsilon_{\beta} = \frac{1-e^{-\mu t}}{\mu t} \)

Where ‘$\mu$’ is the beta attenuation in the foil and ‘$t$’ is the thickness of the foil in gm/cm$^2$. \cite{2}

3.2 Experimental Verification

Five foils of gold (100%) of diameter 0.8cm, manganese-nickel alloy (11% nickel) of diameter 1.25cm, cobalt (100%) of diameter 1.0cm each were irradiated in the thermal column of Apsara reactor at 200KW power for 1 hour and five foils each of magnesium (78.99%) of diameter 1.0cm, aluminium (100%) of diameter 1.0cm were irradiated in the F-7 location of Apsara reactor at 100KW power for 30 minutes \cite{5,6}. Each foil was counted on at least two different times in a $4\pi \beta \gamma$ coincidence counter. All foils were supported in the counter on VYNS film 10$\mu$g/cm$^2$ in thickness.

$4\pi \beta \gamma$ coincidence system is the primary standard for the activity measurement. If there is an ideal source emitting a beta particle and a prompt converted gamma ray and this source is counted by two detectors one responding only to beta ray and other only to gamma-ray and if the disintegration rate of the source is $N_0$ then the counting rate,

\[
N_{\beta} = N_0 \varepsilon_{\beta}
\]

\[
N_{\gamma} = N_0 \varepsilon_{\gamma}
\]

\[
N_C = N_0 \varepsilon_{\beta} \varepsilon_{\gamma}
\]

\[
N_0 = \frac{N_{\beta} N_{\gamma}}{N_C}
\]
Where \( N_{\beta}, N_{\gamma}, N_C \) are count rates in the beta channel, gamma channel, and coincidence channel respectively, and \( \epsilon_{\beta}, \epsilon_{\gamma} \) are the efficiency of beta and gamma channel respectively. But the above equation shows that the disintegration rate is independent of the efficiency of either detector and hence independent of self absorption. The above equation is true for an ideal source assumed above, but in the real case there are a number of correction factors. But these correction factors turned out to be small if efficiency of either of the detector is 100%. In this case the efficiency of the beta detector is nearly 100%. Therefore in \( 4\pi\beta-\gamma \) coincidence counting system all the correction factors are very small and the source activity can be determined with very high accuracy.

![Block Diagram of the 4\( \pi \)\( \beta-\gamma \) Coincidence System](image)

**Fig. 6** Block Diagram of the 4\( \pi \)\( \beta-\gamma \) Coincidence System

To find the activity of a radionuclide the system is switched on half an hour before the experiment. The gas flow is on for half an hour to flush out all air present in the counter. By putting a source the EHT plateau and the resolution plateau are taken. The curves are given below for the system used. The by keeping the source inside the counter photo peak of the NaI (TI) scintillator is determined and the window of the gamma channel is adjusted in such a way that the main photo peak counts are collected. The count rate in the beta channel, gamma channel and the coincidence channels were noted and the activity is calculated using the above formula.

![Operating plateau of coincidence unit](image)

**Fig. 7** Operating Plateau of 4\( \pi \beta-\gamma \) Coincidence System
The absolute disintegration rate of the foil was measured by the $4\pi\beta$-$\gamma$ coincidence technique. For $\beta$ detection a pill-box type $4\pi\beta$ counter made from aluminium was used with burshane as the counting gas. Two 1 mil thick tungsten wires served as anodes. The EHT is in the range from 2000V to 3000V for $\beta$-rays. The plateau slope is about 0.1% per 100V. Operating point is selected at the centre of the plateau. The gamma detector is a counting assembly consisting of a 1.75”X 2” NaI (Tl) crystal coupled to a suitable photo multiplier tube. Care was taken to see that there was sufficient absorber between NaI crystal and the foil so that the highest energy beta ray was completely absorbed before reaching the crystal. The coincidence circuit has a resolving time of about 2-4 $\mu$sec. To compensate for any relative instrumental delay of the pulses, a delay unit is incorporated in one of the channels. The overall efficiency of the beta counter was then calculated by using the expression:

$$\varepsilon_\beta = \frac{N_\beta}{N_0} = \frac{(N_c - 2\tau_R N_\beta' N_\gamma')(1 - N_c\tau_D)(1+K)}{N_\gamma[1 - \tau_R (N_\beta' + N_\gamma')](1 - N_\beta\tau_D)}$$

Where, $N_0$ is the absolute disintegration rate, $N_\beta'$, $N_\gamma'$, $N_\varepsilon'$ are the count rates corrected by background. $\tau_R$ is the resolving time of the coincidence circuit and $\tau_D$ is the dead time. $K$ is the correction term for decay scheme, internal conversion effect and $\gamma$-efficiency of the beta counter.

Table-1 Theoretical and Experimental Beta Attenuation Factor of Various Neutron Activation Foils

<table>
<thead>
<tr>
<th>S.No.</th>
<th>Activation Foils Used</th>
<th>Beta Energy (MeV)</th>
<th>Thickness (gm/cm²)</th>
<th>Theoretical Beta Attenuation Factor</th>
<th>Experimental Beta Attenuation Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Gold</td>
<td>0.952</td>
<td>0.0398</td>
<td>39.02</td>
<td>39.85</td>
</tr>
<tr>
<td>2.</td>
<td>Manganese</td>
<td>2.006</td>
<td>0.0170</td>
<td>6.08</td>
<td>6.87</td>
</tr>
<tr>
<td>3.</td>
<td>Cobalt</td>
<td>0.314</td>
<td>0.1237</td>
<td>92.52</td>
<td>92.58</td>
</tr>
<tr>
<td>4.</td>
<td>Magnesium</td>
<td>1.39</td>
<td>0.0242</td>
<td>11.94</td>
<td>11.96</td>
</tr>
<tr>
<td>5.</td>
<td>Aluminium</td>
<td>1.39</td>
<td>0.0387</td>
<td>17.99</td>
<td>18</td>
</tr>
</tbody>
</table>
4. Results and Conclusion

The theoretically calculated and practically verified beta efficiencies of gold, manganese, cobalt, magnesium and aluminium are plotted and given as Fig.1 to Fig.5. It is found that the beta efficiencies decrease with increase of thickness of the foils. For the various foils like Au, Mn, Co, Al and Mg used in this study, it is found that both theoretical and experimental values for the beta attenuation follows the same pattern and are almost matching. The experimental beta efficiency is found to be always less than the theoretical because of the overall error involved in the measurements. The attenuation of betas in the foils varies with beta energy spectrum and the materials and it varies from 7% to 93% for the foils of usual thickness. The results are given in Table-1. For the neutron measurements in the low flux region using these activation foils, the beta attenuation factor of the respective foil thickness should be applied for getting the correct absolute activity and hence the accurate neutron dose.

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