Study of induced activity of $^{167}$Ho from different neutron capture paths

T S L Morais$^1$ and M S Dias$^2$

Nuclear and Energy Research Institute (IPEN-CNEN/SP)
Nuclear Metrology Laboratory (LMN) – Research Reactor Center (CRPq)
Av. Professor Lineu Prestes 2242, Cidade Universitária, 05508-000
São Paulo, SP, Brazil

$^1$thales.morais@usp.br, $^2$msdias@ipen.br

Abstract. The main purpose of this study is to predict the induced activity of $^{167}$Ho produced by $^{165}$Ho(n,γ)$^{166m}$Ho(n,γ)$^{167}$Ho, $^{165}$Ho(n,γ)$^{166}$Ho(n,γ)$^{167}$Ho and $^{166m}$Ho(n,γ)$^{167}$Ho reactions to choose the best path to measure the cross section with lowest uncertainty. The activation and decay scheme was established starting from the $^{165}$Ho target and considering single, double and triple neutron capture reactions. The activity results were deduced from differential activation equations and decay rates for all reaction products. The calculations were performed considering samples which were taken from a stock solution supplied by the Electrotechnical Laboratory (Japan) for purposes of an international comparison.

1. Introduction
To predict the induced activity from neutron capture reactions is an important step in planning a neutron cross section measurement. The Nuclear Metrology Laboratory (LMN) at the IPEN, in São Paulo, has been involved in improving the accuracy of neutron cross sections by irradiations at the IEA-R1 research reactor. For the measurement of the thermal neutron cross section and the resonance integral of the $^{166m}$Ho(n,γ)$^{167}$Ho reaction it is necessary to know all the different paths that $^{167}$Ho can be produced by neutron capture reactions. As a result, the best method of irradiation can be chosen in order to reduce the uncertainties in the measurements. The difficulty of conducting experiments with radioactive targets is due to the complex decay and activation schemes. This is one of the reasons why the thermal neutron cross section and the resonance integral data are scarce in the literature for the $^{166m}$Ho(n,γ)$^{167}$Ho reaction, when compared with reactions with stable isotopes.

2. Methodology
The activation and decay scheme was established starting from $^{165}$Ho considering single, double and triple neutron capture reactions (see Figure 1) and the calculations were performed considering samples with 20 MBq/g of Ho and 0.31 g/ml of HoCl$_3$ in 1N HCl according to stock solutions supplied by the Electrotechnical Laboratory (Japan) for purposes of an international comparison$^3$.

The variation in the number of atoms of the target nucleus $^{165}$Ho ($N_s$) is given by:

$$\frac{dN_s}{dt} = -N_s\sigma_{5,6}\phi - N_s\sigma_{5,8}\phi$$  \hspace{1cm} (1)
Where:

- $\sigma_{5,6m}$ = cross section for $^{165}$Ho(n,γ)$^{166m}$Ho reaction;
- $\sigma_{5,6}$ = cross section for $^{165}$Ho(n,γ)$^{166}$Ho reaction;
- $\phi$ = neutron flux.

**Figure 1. Activation and decay scheme starting from $^{165}$Ho** [1][2]

The first term refers to the production rate of $^{166m}$Ho and the second refers to the production rate of $^{166}$Ho. Solving the differential equation (1) by the method of separation of variables, the result is:

$$N_5(t) = e^{-\sigma_{5,6m}^\phi + \sigma_{5,6}^\phi} \cdot e^{K_1}$$  \hspace{1cm} (2)

Where $K_1$ is a constant and $t$ is the irradiation time. When $t = 0$, $N_5 = N_5^0$. Where $N_5^0$ is the number of atoms of $^{165}$Ho in the sample at the beginning of the irradiation. Replacing this information in the equation (2) one obtains $e^{K_1} = N_5^0$, therefore:

$$N_5(t) = N_5^0 \cdot e^{-\sigma_{5,6m}^\phi + \sigma_{5,6}^\phi}$$  \hspace{1cm} (3)

The variation of $^{166m}$Ho atoms in the sample, must consider the formation of $^{166m}$Ho atoms through the activation of $^{165}$Ho, the decay of the $^{166m}$Ho, and its activation forming $^{167}$Ho, as follows:

$$\frac{dN_{6m}}{dt} = N_5^0 \sigma_{5,6m}^\phi - \lambda_{6m} N_{6m} - N_{6m} \sigma_{6m}^\phi$$  \hspace{1cm} (4)

Where:

- $\lambda_{6m}$ = decay constant of $^{166m}$Ho;
- $\sigma_{6m}$ = cross section for $^{166m}$Ho(n,γ)$^{167}$Ho reaction.

Replacing equation (3) into equation (4), multiplying both sides by $e^{(\lambda_{6m} + \sigma_{6m})t}$ and solving the differential equation, the result is:

$$N_{6m}(t) = \frac{N^0_5 \sigma_{5,6m}^\phi e^{-(\sigma_{5,6m}^\phi + \sigma_{5,6}^\phi)t}}{\lambda_{6m} - (\sigma_{5,6m}^\phi + \sigma_{5,6} - \sigma_{6m}^\phi)} + K_2 e^{(\lambda_{6m} + \sigma_{6m})t}$$  \hspace{1cm} (5)
Where $K_2$ is a constant.
Considering that initially ($t = 0$) there was a certain amount of $^{166m}$Ho atoms ($N_{6m}^0$), one obtains:

$$N_{6m}(t) = \frac{N_{6m}^0 \sigma_{5,6m} \phi}{\lambda_{6m} - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_{6m}) \phi} \left[ e^{-\left(\sigma_{5,6m} + \sigma_{5,6} - \sigma_{6m}\right)t} - e^{-\left(\lambda_{6m} + \sigma_{6m}\right)t} \right] + N_{6m}^0 e^{-\left(\lambda_{6m} + \sigma_{6m}\right)t} \tag{6}$$

The number of $^{166}$Ho atoms in the sample can be calculated in a similar way, as follows:

$$N_6(t) = \frac{N_6^0 \sigma_{5,6m} \phi}{\lambda_6 - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_6) \phi} \left[ e^{-\left(\sigma_{5,6m} + \sigma_{5,6} - \sigma_6\right)t} - e^{-\left(\lambda_6 + \sigma_6\right)t} \right] \tag{7}$$

Where:
- $\lambda_6$ = decay constant of $^{166}$Ho;
- $\sigma_6$ = cross section for $^{166}$Ho(n,γ)$^{167}$Ho reaction.

Analyzing the production of $^{167}$Ho from $^{166}$Ho and $^{166m}$Ho.

$$\frac{dN_7}{dt} = N_{6m} \sigma_{6m} \phi + N_6 \sigma_6 \phi - \lambda_7 N_7 - N_7 \sigma_7 \phi \tag{8}$$

Where:
- $\lambda_7$ = decay constant of $^{167}$Ho;
- $\sigma_7$ = sum of cross sections for $^{167}$Ho(n,γ)$^{168}$Ho and $^{167}$Ho(n,γ)$^{168m}$Ho reactions.

The first term refers to the activation of $^{166m}$Ho, the second refers to the activation of $^{166}$Ho, the third refers to the radioactive decay of $^{167}$Ho and the fourth refers to the activation of $^{167}$Ho.

Analogously to what has already been presented and replacing the values of $N_{6m}$ and $N_6$ according to equations (6) and (7), we have:

$$N_7(t) = \frac{N_7^0 \sigma_{5,6} \phi^2}{\lambda_6 - (\sigma_{5,6} + \sigma_{5,6} - \sigma_6) \phi} \left[ e^{-\left(\sigma_{5,6} + \sigma_{5,6} - \sigma_6\right)t} - e^{-\left(\lambda_6 + \sigma_6\right)t} \right] +$$

$$+ \frac{N_{6m}^0 \sigma_{5,6m} \sigma_{6m} \phi^2}{\lambda_{6m} - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_{6m}) \phi} \left[ e^{-\left(\sigma_{5,6m} + \sigma_{5,6} - \sigma_{6m}\right)t} - e^{-\left(\lambda_{6m} + \sigma_{6m}\right)t} \right] +$$

$$+ N_{6m}^0 \sigma_{6m} \phi \left[ e^{-\left(\lambda_{6m} + \sigma_{6m}\right)t} - e^{-\left(\lambda_6 + \sigma_6\right)t} \right] \tag{9}$$

Since the induced activity of $^{167}$Ho is given by,

$$A_7(t) = A_7^{167}(t) + A_7^{166m}(t) + A_7^{166}(t) \tag{10}$$

The terms in equation (10) are the activities of $^{167}$Ho from $^{165}$Ho(n,γ)$^{166}$Ho(n,γ)$^{167}$Ho, $^{165}$Ho(n,γ)$^{166m}$Ho(n,γ)$^{167}$Ho and $^{166m}$Ho(n,γ)$^{167}$Ho reactions, respectively.

Multiplying the equation (9) by $\lambda_7$ we get $A_7(t)$, so the following equations were determined:
3. Results

Figure 2 shows the activity (in Bq) of $^{167}$Ho as a function of irradiation time, considering samples with 10 mg from a solution with 20 MBq/g of $^{166m}$Ho and 0.31 g/ml of HoCl$_3$ in 1N HCl\textsuperscript{[3]}. As can be seen, the predominant contribution comes from the $^{166m}$Ho(n,$\gamma$)$^{167}$Ho reaction.

![Induced Activity of $^{167}$Ho](image)

Figure 2. Induced Activity of $^{167}$Ho as a function of irradiation time.

Acknowledgments

The authors are indebted to the National Counsel of Technological and Scientific Development (CNPq), from Brazil, for partial support of the present research work.

References

[1] International Atomic Energy Agency IAEA (2018) The Live Chart of Nuclides. https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html

[2] Harada H, Wada H, Nakamura S, Furutaka K and Katoh T (2000) “Measurement of Effective Neutron Capture Cross Section of $^{166m}$Ho using Two Step Irradiation Technique”, Journal of Nuclear Science and Technology, 37:9, 821-823.

[3] Hino, Matui, Yamada, Takeuchi S, Onoma, Iwamoto and Kogure. “Absolute measurement of $^{166m}$Ho radioactivity and development of sealed sources for standardization of gamma-ray emitting nuclides”, Applied radiation and isotopes, 52:3 (2000) 545-549.