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

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Abstract. The main purpose of this study is to predict the induced activity of ^{167}Ho produced by $^{165}\text{Ho}(n,\gamma)^{166\text{m}}\text{Ho}(n,\gamma)^{167}\text{Ho}$, $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}(n,\gamma)^{167}\text{Ho}$ and $^{166\text{m}}\text{Ho}(n,\gamma)^{167}\text{Ho}$ 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 $^{166\text{m}}\text{Ho}(n,\gamma)^{167}\text{Ho}$ 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 $^{166\text{m}}\text{Ho}(n,\gamma)^{167}\text{Ho}$ 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_5) is given by:

$$\frac{dN_5}{dt} = -N_5\sigma_{5,6\text{m}}\phi - N_5\sigma_{5,6}\phi \quad (1)$$



Where:

$\sigma_{5,6m}$ = cross section for $^{165}\text{Ho}(n,\gamma)^{166m}\text{Ho}$ reaction;

$\sigma_{5,6}$ = cross section for $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ reaction;

ϕ = neutron flux.

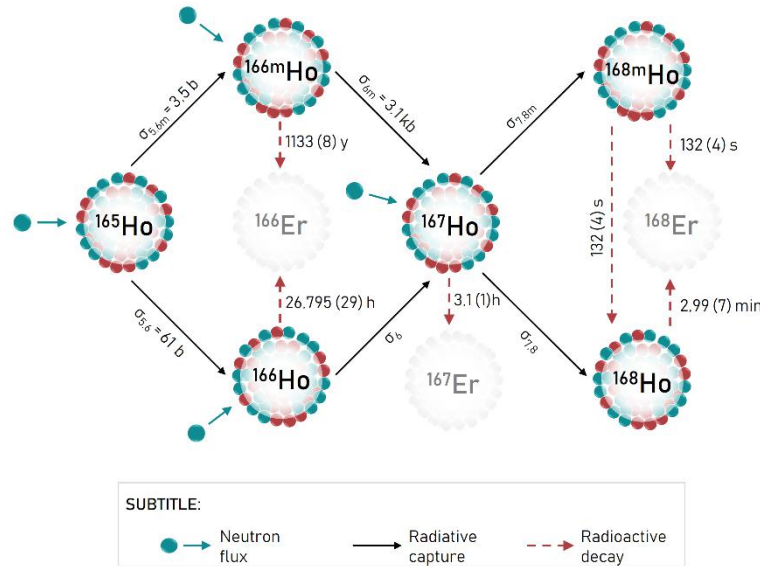


Figure 1. Activation and decay scheme starting from ^{165}Ho [1][2]

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:

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

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:

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

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:

$$\frac{dN_{6m}}{dt} = N_5 \sigma_{5,6m} \phi - \lambda_{6m} N_{6m} - N_{6m} \sigma_{6m} \phi \tag{4}$$

Where:

λ_{6m} = decay constant of ^{166m}Ho ;

σ_{6m} = cross section for $^{166m}\text{Ho}(n,\gamma)^{167}\text{Ho}$ reaction.

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

$$N_{6m}(t) = \frac{N_5^0 \sigma_{5,6m} \phi e^{-(\sigma_{5,6m} + \sigma_{5,6})\phi t}}{\lambda_{6m} - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_{6m})\phi} + K_2 e^{-(\lambda_{6m} + \sigma_{6m}\phi)t} \tag{5}$$

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:

$$N_{6m}(t) = \frac{N_5^0 \sigma_{5,6m} \phi}{\lambda_{6m} - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_{6m}) \phi} \left[e^{-(\sigma_{5,6m} + \sigma_{5,6}) \phi t} - e^{-(\lambda_{6m} + \sigma_{6m} \phi) t} \right] + N_{6m}^0 e^{-(\lambda_{6m} + \sigma_{6m} \phi) t} \quad (6)$$

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

$$N_6(t) = \frac{N_5^0 \sigma_{5,6m} \phi}{\lambda_6 - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_6) \phi} \left[e^{-(\sigma_{5,6m} + \sigma_{5,6}) \phi t} - e^{-(\lambda_6 + \sigma_6 \phi) t} \right] \quad (7)$$

Where:

λ_6 = decay constant of ^{166}Ho ;

σ_6 = cross section for $^{166}\text{Ho}(n,\gamma)^{167}\text{Ho}$ reaction.

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

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

Where:

λ_7 = decay constant of ^{167}Ho ;

σ_7 = sum of cross sections for $^{167}\text{Ho}(n,\gamma)^{168}\text{Ho}$ and $^{167}\text{Ho}(n,\gamma)^{168m}\text{Ho}$ 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:

$$\begin{aligned} N_7(t) = & \frac{N_5^0 \sigma_{5,6} \sigma_6 \phi^2}{\lambda_6 - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_6) \phi} \left[\frac{e^{-(\sigma_{5,6m} \phi + \sigma_{5,6} \phi) t} - e^{-(\lambda_7 + \sigma_7 \phi) t}}{\lambda_7 - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_7) \phi} + \frac{e^{-(\lambda_6 + \sigma_6 \phi) t} - e^{-(\lambda_7 + \sigma_7 \phi) t}}{\lambda_6 - \lambda_7 + (\sigma_6 - \sigma_7) \phi} \right] + \\ & + \frac{N_5^0 \sigma_{5,6m} \sigma_6 \phi^2}{\lambda_{6m} - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_{6m}) \phi} \left[\frac{e^{-(\sigma_{5,6m} \phi + \sigma_{5,6} \phi) t} - e^{-(\lambda_7 + \sigma_7 \phi) t}}{\lambda_7 - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_7) \phi} + \frac{e^{-(\lambda_{6m} + \sigma_{6m} \phi) t} - e^{-(\lambda_7 + \sigma_7 \phi) t}}{\lambda_{6m} - \lambda_7 + (\sigma_{6m} - \sigma_7) \phi} \right] + \\ & + N_{6m}^0 \sigma_{6m} \phi \left[- \frac{e^{-(\lambda_{6m} + \sigma_{6m} \phi) t} - e^{-(\lambda_7 + \sigma_7 \phi) t}}{\lambda_{6m} + \sigma_{6m} \phi - \lambda_7 - \sigma_7 \phi} \right] \end{aligned} \quad (9)$$

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

$$A_7(t) = A_7^{5,6}(t) + A_7^{5,6m}(t) + A_7^{6m}(t) \quad (10)$$

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

Multiplying the equation (9) by λ_7 we get $A_7(t)$, so the following equations were determined:

$$A_7^{5,6}(t) = \frac{\lambda_7 N_5^0 \sigma_{5,6} \sigma_6 \phi^2}{\lambda_6 - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_6) \phi} \left[\frac{e^{-(\sigma_{5,6m} \phi + \sigma_{5,6} \phi)t} - e^{-(\lambda_7 + \sigma_7 \phi)t}}{\lambda_7 - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_7) \phi} + \frac{e^{-(\lambda_6 + \sigma_6 \phi)t} - e^{-(\lambda_7 + \sigma_7 \phi)t}}{\lambda_6 - \lambda_7 + (\sigma_6 - \sigma_7) \phi} \right] \quad (11)$$

$$A_7^{5,6m}(t) = \frac{\lambda_7 N_5^0 \sigma_{5,6m} \sigma_{6m} \phi^2}{\lambda_{6m} - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_{6m}) \phi} \left[\frac{e^{-(\sigma_{5,6m} \phi + \sigma_{5,6} \phi)t} - e^{-(\lambda_7 + \sigma_7 \phi)t}}{\lambda_7 - (\sigma_{5,6m} + \sigma_{5,6} - \sigma_7) \phi} + \frac{e^{-(\lambda_{6m} + \sigma_{6m} \phi)t} - e^{-(\lambda_7 + \sigma_7 \phi)t}}{\lambda_{6m} - \lambda_7 + (\sigma_{6m} - \sigma_7) \phi} \right] \quad (12)$$

$$A_7^{6m}(t) = \lambda_7 N_{6m}^0 \sigma_{6m} \phi \left[\frac{e^{-(\lambda_{6m} + \sigma_{6m} \phi)t} - e^{-(\lambda_7 + \sigma_7 \phi)t}}{\lambda_{6m} + \sigma_{6m} \phi - \lambda_7 - \sigma_7 \phi} \right] \quad (13)$$

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 [3]. As can be seen, the predominant contribution comes from the $^{166m}\text{Ho}(n,\gamma)^{167}\text{Ho}$ reaction.

Induced Activity of ^{167}Ho

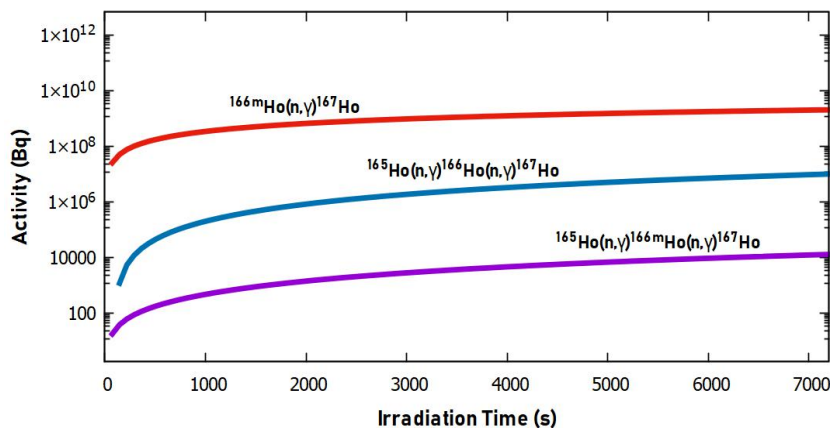


Figure 2. Induced Activity of ^{167}Ho as a function of irradiation time.

Acknowledgments

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