Model Calculations for Proton Induced Nuclear Reaction on Zinc at Low Energy

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ABSTRACT

Excitation functions of the reactions \( \text{natZn}(p,xn)\text{66,67,68Ga} \) were measured from their respective thresholds up to 18 MeV, using the conventional stacked-foil technique. The radioactivity was determined via high resolution \( \gamma \)-ray spectrometry. Nuclear model calculations were performed using the codes, ALICE-IPPE, EMPIRE and TALYS. In some cases, good agreement was found between the experimental and theoretical data, while in others considerable deviations were observed. From the experimental data the expected integral yields of the three investigated radionuclides were calculated.

Key Words: Zn/Stacked-Foil/Proton/Excitation Function/Model Calculations.

INTRODUCTION

The three radioisotopes \( \text{66Ga} \), \( \text{67Ga} \) and \( \text{68Ga} \) are well known and widely used in the field of nuclear medicine. \( \text{67Ga} \) has become one of the most frequently employed cyclotron produced radioisotope over the last two decades and is a widely used single photon marker for detecting the presence of malignancy and for diagnosis of inflammatory diseases. The positron emitter \( \text{68Ga} \), usually obtained through a \( \text{68Ge (t}(1/2) = 288 \text{ d)} \text{68Ga} \) generator, is employed at PET centers for blood–brain barrier investigations, in diagnosis of some tumor diseases of liver and other organs and for transmission measurements for encoding calibration and normalization of detector efficiencies of PET scanners. Recently, \( \text{66Ga} \) was proposed for studying some slow dynamic processes by PET. It is noticed that the use of natural zinc for practical purposes is very limited because of the lower yield and/or high contaminations \(^1\).

Recently, the status of nuclear data for medical radioisotopes produced by accelerators has been reviewed. This summarizes the available data on experimental measurements of cross-sections and thick target yields for medical radioisotopes of current interest besides the presentation of exhaustive reference material. The statuses also shows that although many reactions were frequently studied in the past, especially in the above-mentioned energy range, results of new precise cross-sections certainly could be useful for some data bases even for the most commonly used radioisotopes including \( \text{67Ga}, \text{111In}, \text{123I} \) and \( \text{201Tl} \)\(^1\).

This article reports on the cross-sections and production yields of \( \text{66Ga}, \text{67Ga} \) and \( \text{68Ga} \), have been measured for the compilation of the existing data base. The cross-sections and the production yields are plotted in the energy range from 4 to 17.6 MeV to give the excitation functions and thick target yield functions for \( \text{66Ga}, \text{67Ga} \) and \( \text{68Ga} \) in natural zinc. The results and the relevant errors are compared with the data published in the literature.
EXPERIMENTAL

The experimental technique used and the method of the data evaluation are similar to those described in our earlier investigations (2,3,4). The activation method and stacked foil irradiation technique were used to produce and to detect the radioactive products.

High-purity Zn foils (51.83 μm thick) and high-purity Ti (13.1 μm thick) monitor foils (99.99%, Good-fellow foils) were irradiated with the external proton beam of Debrecen MGC-20 cyclotron facility. The Proton beam energies were defined by an analyzing magnet. The foils diameters were 10 mm and their individual thicknesses were determined by weighting.

Two irradiation experiments have been done. The first stack contains 20 foils, 10 Ti and 10 Zn. The primary proton incident energy was 18 MeV, the energy interval in the stack varies from 17.6 up to 8.7 MeV, due to the energy loss in the foil the energy degradation was about 0.6 MeV and 1.2 MeV in the first and the last foil respectively. In the second experiment the primary proton incident energy was 14.5 MeV, the stack contained 18 foils 9 Zn and 9 Ti, the energy intervals was from 14.4 to 3 MeV. From the two experiments an energy overlap region were covered (14.5-8.5 MeV) to insure the reproducibility of the results. The targets were irradiated for 30 minutes with 100 nA of proton beams. The intensity of the beam current was kept constant during the irradiation.

Ti foils measured over the whole energy range served to monitor the exact beam intensity along the target stack. High-resolution gamma spectrometer was used to detect and to identify the radioactive reaction products. The activity of the irradiated samples was measured non-destructively, without any chemical separation of the produced activities. The activity measurements started one hour after End Of Bombardment EOB. The well-known activation formula with the standard corrections for decay during irradiation, cooling and measurement were used for cross-section determination. The energy degradation of the bombarding particles in the foils of the irradiated targets was determined from the polynomial approximation method of Andersen and Ziegler (5).

Natural Ti was used as monitor foils for energy and beam intensity measurements. Therefore the natTi(p,x)48V reaction was investigated. As 48Sc(43.67 h) formed from the reaction natTi(p,x)48Sc decays with the two gamma lines (983,1312 KeV) which are the same for 48V, the monitor foils were measured 20 days later after end of bombardment to insure the complete decay of 48Sc isotope.

The reference cross-section data for the reaction natTi(p,x)48V were taken from (6) and the recommended database of the IAEA (7). The experimental data in comparison with IAEA data are given in (Fig.1).

\[ \text{Cross section (mb)} \]

\[ \text{Proton energy (MeV)} \]

Fig. (1): Experimental excitation functions for the natTi(p,x)48V monitor reaction.
Activity measurements were performed at long detector/sample distance to avoid dead time corrections and to minimize geometrical effects (calibration performed with point sources). Monitor foils and Zn foils were measured at the same source to detector distance for eliminating possible relative inaccuracies in efficiency calibration. The decay data of the investigated nuclei were taken from the Nudat 2.5\(^8\) and are represented in (Table 1). The Q-values of the contributing processes were taken from the T2 database of Los Alamos\(^9\). The resulting uncertainties of the linearly contributing processes were calculated according to the well-accepted summation rules.

Table (1): The decay data of the investigated nuclei

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Decay mode(%)</th>
<th>E(_γ) (keV)</th>
<th>I(_γ) (%)</th>
<th>Contributing reactions</th>
<th>Q-value (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{66})Ga</td>
<td>9.49 h</td>
<td>EC(7) (\beta^+(93))</td>
<td>833.5 1039.3 1333 1918.6</td>
<td>5.89 37 1.2 2.1</td>
<td>(^{66})Zn(p,n)(^{66})Ga (^{67})Zn(p,2n)(^{66})Ga (^{68})Zn(p,3n)(^{66})Ga</td>
<td>-5.957 -13.2 3.2</td>
</tr>
<tr>
<td>(^{67})Ga</td>
<td>3.261 d</td>
<td>EC (100)</td>
<td>93.31 184.5 300.2 393.5</td>
<td>39.2 21.2 16.8 4.68</td>
<td>(^{67})Zn(p,n)(^{67})Ga (^{68})Zn(p,2n)(^{67})Ga</td>
<td>-1.782 -11.98</td>
</tr>
<tr>
<td>(^{68})Ga</td>
<td>67.6 min</td>
<td>EC(38) (\beta^+(62))</td>
<td>1077</td>
<td>3</td>
<td>(^{68})Zn(p,n)(^{68})Ga (^{70})Zn(p,3n)(^{68})Ga</td>
<td>-3.7 -19.4</td>
</tr>
</tbody>
</table>

**NUCLEAR REACTIONS CROSS SECTION CALCULATIONS**

The absolute activity values of the interested radionuclides were determined using their characteristic \(\gamma\)-ray lines. The reaction cross-section was calculated using the well-known activation formula depending on the decay rates\(^{10}\). The sources of uncertainty in the measured cross sections were estimated and found to be: beam current 9-12\%, detector efficiency 3-4\%, statistical counting 4-6\% and decay data 0-1\%. The overall uncertainty in the measured cross-section values amounts were found 10-14\%.

**NUCLEAR MODEL CALCULATIONS**

With a view to validate the data and testing the predictive power of the nuclear theory, reaction cross sections were calculated theoretically using the nuclear model codes ALICE-IPPE, TALYS-1.2 and EMPIRE-03. A brief description is given below.

**A. ALICE-IPPE Calculations**

The code ALICE-IPPE is a modified version of the exciton model code ALICE, originally developed by Blann\(^{11}\). The modifications introduced by the Obninsk group\(^{12}\) include:

(a) treatment of the level density in the frame of the generalized super fluid model,

(b) consideration of the pre-equilibrium cluster emission (d, t, \(^3\)He, \(^4\)He), and

(c) estimation of direct interactions in cluster emission.

In the present work, the excitation functions of the deuteron induced reactions on \(^{nat}\)Zr investigated experimentally were also calculated from their respective thresholds up to 10 MeV. In all the calculations, standard recommended input data were used and no additional fitting parameter was done.
B. EMPIRE-03 Calculations

The code EMPIRE-03\(^{(13)}\) makes use of the Hauser-Feshbach and the exciton model formalisms. Furthermore, it combines several other modern features described below. In these calculations the standard library of input parameters was used which includes the nuclear masses, optical model parameters, ground state deformations, discrete levels and decay schemes, level densities, moments of inertia (MOMFIT), and \(\gamma\)-ray strength functions. The direct contribution was determined via the Coupled Channel calculation using the built in ECIS03 code. The particle transmission coefficients were generated via the spherical optical model using the computer code (ECIS03) and the default set of global parameters: for neutrons and protons from Koning, Delaroche \(^{(14)}\) and for alpha particles from McFadden and Satchler\(^{(15)}\). In the calculation the Multi Step Direct, Multi Step Compound, Hauser-Feshbach model with width fluctuation correction (HRTW), the DEGAS and PCROSS codes were used. These codes conserve the particle flux by dividing the absorption cross section of the optical model between the different types of reaction mechanisms. For the level densities, the HF-BCS microscopic level densities were used.

C. TALYS Calculations

In TALYS code the pre-equilibrium particle emission is described using the two-component exciton model\(^{(16,17)}\). The mode implements new expressions for internal transition rates and new parameterization of the average squared matrix element for the residual interaction obtained using the optical model potential. The phenomenological model\(^{(18)}\) is used for the description of the pre-equilibrium complex particle emission (deuteron, triton and a-particle). The contribution of direct processes in inelastic scattering is calculated using the ECIS-97 code incorporated in TALYS. The equilibrium particle emission is described using the Hauser–Feshbach model. The nuclear level density for equilibrium states is calculated using different nuclear models\(^{(19)}\). The cross sections for total neutron non-elastic interactions with nuclei have been calculated using the optical potential.

RESULTS AND DISCUSSION

Cross Section Data

Three proton induced reactions on zinc, namely \(^{nat}\text{Zn}(p,xn)^{66}\text{Ga}, \(^{nat}\text{Zn}(p,xn)^{67}\text{Ga}\) and \(^{nat}\text{Zn}(p,xn)^{68}\text{Ga}\) have been investigated from threshold energy up to 18 MeV.

Experimental cross section data for proton induced reactions on \(^{nat}\text{Zn}\) are given in (Table 2). The estimated uncertainties of energy and cross section values are also presented.

Excitation Functions

The data were plotted as a function of proton energy and the excitation curves obtained were compared with the results of nuclear model calculations as well as with the literature experimental data (cf. EXFOR\(^{(20)}\), 2010, and other relevant citations given below at appropriate places). The results are discussed below.
Table(2): Measured cross-sections of the investigated reactions of deuterons with *nat*Zn

<table>
<thead>
<tr>
<th>Proton energy(MeV)</th>
<th>Cross section (mb)</th>
<th>66Ga</th>
<th>67Ga</th>
<th>68Ga</th>
</tr>
</thead>
<tbody>
<tr>
<td>17.6±0.66</td>
<td>75.8±8.6</td>
<td></td>
<td></td>
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<tr>
<td>16.7±0.70</td>
<td>94.6±10.7</td>
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<tr>
<td>15.9±0.71</td>
<td>114.0±12.8</td>
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<tr>
<td>15.0±0.74</td>
<td>137.8±15.5</td>
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<tr>
<td>14.2±0.78</td>
<td>163.2±18.4</td>
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<td></td>
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<tr>
<td>14.0±0.80</td>
<td>154.2±18.8</td>
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<tr>
<td>13.2±0.82</td>
<td>172.8±19.5</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>13.0±0.83</td>
<td>170.5±19.2</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>12.2±0.87</td>
<td>177.0±20.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12.0±0.88</td>
<td>172.3±19.4</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>11.1±0.93</td>
<td>170.0±19.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11.0±0.94</td>
<td>163.8±18.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10.0±1.00</td>
<td>154.5±17.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9.7±1.00</td>
<td>152.0±17.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8.7±1.10</td>
<td>117.6±15.9</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>8.4±1.10</td>
<td>128.1±14.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7.0±1.30</td>
<td>93.7±10.6</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5.3±1.50</td>
<td>5.8±0.7</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>3.5±2.10</td>
<td>5.8±0.7</td>
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</tbody>
</table>

*nat*Zn(p,xn)66Ga reaction

The main contributing processes for the formation of 66Ga radionuclide however are the 66Zn(p,n), 67Zn(p,2n) and 68Zn(p,3n) reactions (see Table 1). The excitation function for the formation of 66Ga is represented in (Fig. 2) along with the available literature data up to 18 MeV. The excitation function shows a maximum cross-section of 177 mb ± 20 mb at about 12MeV proton energy.

![Fig.(2): Experimental excitation function for the *nat*Zn(p,xn)66Ga reaction.](image)

The present work agrees with Al-Saleh et al. (1), uddin et al. (21) Szecencenyi et al. (22) Norrier et al. (23) and Little et al. (24) in the energy region higher than 14 MeV but lower than all of them in the energy region lower than 14 MeV. Barrandon et al. (25) results are much lower than the experimental results in the whole energy range.
The results of the theoretical calculations using the ALICE-IPPE, TALYS-1.2 and EMPIRE-03 codes are in acceptable agreement with the present experimental data and the literature as well.

**nat**$^\text{Zn}(p,xn)^{67}\text{Ga}$ reaction

The main contributing processes for the formation of $^{67}\text{Ga}$ radionuclide however are the $^{67}\text{Zn}(p,n)$ and $^{68}\text{Zn}(p,2n)$ reactions (see Table 1). Fig. 3 represents the excitation function for the formation of $^{67}\text{Ga}$ along with the available literature data up to 18 MeV. The excitation function shows a maximum cross-section of 127 mb±15mb at about 18 MeV.

![Excitation Function Graph](image)

**Fig.(3): Experimental excitation function for the nat**$^\text{Zn}(p,xn)^{67}\text{Ga}$ reaction.

The present work agrees with Al-Saleh et al.\(^{(1)}\), uddin et al.\(^{(2)}\), Szelecenyi et al.\(^{(21)}\), and Nortier et al.\(^{(22)}\) in the whole energy range. Little et al.\(^{(24)}\) and Barrandon et al.\(^{(25)}\) results are much lower than the experimental results.

The results of the theoretical calculations using the ALICE-IPPE, TALYS-1.2 and EMPIRE-03 codes are in acceptable agreement with the present experimental data and the literature as well.

**nat**$^\text{Zn}(p,n)^{68}\text{Ga}$ reaction

The main contributing process for the formation of $^{68}\text{Ga}$ radionuclide however is the $^{68}\text{Zn}(p,n)$ (see Table 1). Fig. 4 represents the excitation function for the formation of $^{68}\text{Ga}$ along with the available literature data up to 18 MeV. The excitation function shows a maximum cross-section of 179mb±18mb at about 12.2 MeV.
Fig. (4): Experimental excitation function for the $^{nat}Zn(p,n)^{68}Ga$ reaction.

The present work agrees with Al-Saleh et al. (1) Szelecenyi et al. (22) Nortier et al. (23) and Tárányi et al. (26) in the whole energy range.

The results of the theoretical calculations using the ALICE-IPPE, TALYS-1.2 and EMPIRE-03 codes are in acceptable agreement with the present experimental data and the literature as well.

CALCULATED YIELDS

From the eye guides of the excitation functions for the formation of $^{66,67,68}Ga$ radioisotopes given in Figs. 2-4, differential and integral yields were calculated. The integral yields are shown in Fig.5 as a function of proton energy.

The yields of $^{66}Ga$, $^{67}Ga$ and $^{68}Ga$ at $17\rightarrow 5$ MeV amounted to 305, 14.5 and 2362 MBq/μA·h respectively. Thus sufficient quantities of $^{66}Ga$, $^{67}Ga$ and $^{68}Ga$ can be produced at a small-sized cyclotron.
CONCLUSION

Proton-induced activation cross-sections were measured for the formation of the $^{66,67,68}$Ga on a natural zinc targets up to 18 MeV. The present measurement over this energy range confirms the earlier published data. The used theoretical nuclear model codes can describe well the investigated reaction in the whole interested proton energy range. The quality and success of each code was also represented.

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