International Atomic Energy Agency



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# INTERNATIONAL NUCLEAR DATA COMMITTEE

# Summary Report of the Third Research Co-ordination Meeting on DEVELOPMENT OF REFERENCE CHARGED-PARTICLE CROSS SECTION DATABASE FOR MEDICAL RADIOISOTOPE PRODUCTION

Vrije Universiteit Brussel, Brussels, Belgium 28 September - 2 October 1998

Prepared by

Pavel OBLOŽINSKÝ IAEA Nuclear Data Section Vienna, Austria

November 1998

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## Abstract

The report summarizes results of the final coordination meeting of the IAEA research project on "Development of Reference Charged-Particle Cross Section Database for Medical Radioisotope Production". Details are given on the status of the Database and preparation of the TECDOC, two major results of the project. Actions and deadlines are specified towards finalizing these results in the near future.

November 1998

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# **1** Summary of the Meeting

### **Objectives and Participation**

The third Research Co-ordination Meeting (RCM) on "Development of Reference Charged-Particle Cross Section Database for Medical Radioisotope Production" was held at the Vrije Universiteit Brussel, Brussels, Belgium, from 28 September - 2 October 1998. The local host of the RCM was Prof. A. Hermanne.

The purpose of the meeting was to review evaluations to be included in the reference charged-particle database for medical radioisotope production and beam monitoring, to review overall results achieved by the CRP, and to discuss the draft of the related IAEA Technical Document (TECDOC). Prof. S.M. Qaim of the Institut für Nuklearchemie, Forschungszentrum Jülich, Jülich, Germany, served as chairman of the Meeting. The detailed Agenda is attached (Appendix 1).

The meeting was attended by the chief scientific investigators of all seven laboratories The participating laboratories were participating in the project, and by one consultant. represented by A. Hermanne (Vrije Universiteit Brussel, Brussels, Belgium), Zhuang (China Institute of Atomic Energy, Beijing, China). Youxiang S.M. Oaim (Forschungszentrum Jülich GmbH, Jülich, Germany), M.G. Mustafa (Lawrence Livermore National Laboratory, Livermore, California, USA), T.F. Tárkányi (Hungarian Academy of Sciences, Debrecen, Hungary), Y. Shubin (Institute of Physics and Power Engineering, Obninsk, Russia), and F.M. Nortier (National Accelerator Centre, Faure, South Africa). Furthermore, K. Gul (Pakistan Institute of Nuclear Science and Technology, Islamabad, Pakistan) attended as a consultant. For details see Appendix 2.

### **Main Conclusions**

It was concluded that good progress has been made since the last CRP meeting, and the goals initially defined for the project will be met. The CRP is producing two major results:

- Reference Charged-Particle Cross Section Database for Medical Radioisotope Production, to be completed in May 1999, and
- TECDOC "Charged-Particle Cross Section Database for Medical Radioisotope Production", second draft to be ready also in May 1999.

## **2 Progress Reports and Presentations**

All seven participating laboratories presented their progress reports at the meeting. These reports reflect a substantial amount of work performed by the participating laboratories since the last meeting in Cape Town. April 1997. See Appendix 3 for more details.

The work done by K. Gul (Islamabad, Pakistan) in support of the present CRP was presented. It was felt that this work represents a very useful contribution to the project. For the full report see Appendix 4.

# **3** Recommendations for the Database

All reactions to be included into the Reference Charged-Particle Cross Section Database for Medical Radioisotope Production were discussed. The final list includes 21 monitor reactions (8 for protons, 4 for deuterons, 3 for <sup>3</sup>He and 6 for  $\alpha$  particles), 11 reactions for gamma emitters (as well as 5 other associated reactions), and 10 reactions for positron emitters. The following recommendations were made:

# 3.1 Beam Monitor Reactions

## Protons

## a. ${}^{27}Al(p,x){}^{22}Na$

Spline fit seems good, do new Pade fit (action Shubin), do final selection (action Tárkányi).

Energy range: 30-100 MeV.

## b. <sup>27</sup>Al(p,x)<sup>24</sup>Na

Add to the list, compile and select (action Tárkányi), do Pade fit (action Shubin) and spline fit (action Zhuang), do model calculations (actions Mustafa, Shubin, Zhuang), do final selection (Tárkányi).

Energy range: 40-100 MeV.

## c. $^{nat}Ti(p,x)^{48}V$

Revise selection of experimental data including new measurements from Debrecen, Brussels and Jülich) (action Tárkányi), do new spline fit (action Zhuang) and new Pade fit (action Shubin), do final selection (action Tárkányi). Energy range: threshold - 50 MeV.

## d. $^{nat}Ni(p,x)^{57}Ni$

Add new experimental data from Jülich up to 20 MeV (action Tárkányi), do new Pade fit (action Shubin) and new spline fit (action Zhuang), do final selection (action Tárkányi). Energy range: threshold - 50 MeV.

## e. $^{nat}Cu(p,x)^{56}Co$

Pade fit seems good, do new spline fit (action Zhuang), do final selection (action Tárkányi). Include statement for users that it is a cumulative cross section. Energy range: 50-100 MeV.

## f. $^{nat}Cu(p,x)^{62}Zn$

Using Scholten selection, do new Pade fit (action Shubin) and new spline fit (action Zhuang), do final selection (action Tárkányi). Energy range: 15-50 MeV.

## g. $^{nat}Cu(p,x)^{63}Zn$

Using Scholten selection, do new Pade fit (action Shubin) and new spline fit (action Zhuang), do final selection (action Tárkányi). Energy range: 5-50 MeV.

### h. $^{nat}Cu(p,x)^{65}Zn$

Using Scholten selection, do new Pade fit (action Shubin) and accept it as recommended data.

Energy range: 3-100 MeV.

### Deuterons

## a. <sup>27</sup>Al(d,x)<sup>22</sup>Na

Accept Pade fit as recommended data. Energy range: 30 - 80 MeV.

# b. <sup>27</sup>Al(d,x)<sup>24</sup>Na

Do collection and selection of experimental data (action Tárkányi), do Pade fit (action Shubin) and spline fit (action Zhuang), do final selection (action Tárkányi). Energy range:15-80 MeV.

## c. $^{nat}Ti(d,x)^{48}V$

Remove from the list due to substantial discrepancies in selected experimental data.

## d. <sup>nat</sup>Fe(d,x)<sup>56</sup>Co

Accept Pade fit as recommended data. Energy range: 8-40 MeV.

### e. $^{nat}Ni(d,x)^{61}Cu$

Accept Pade fit as recommended data. Energy range: 3-20 MeV.

## <sup>3</sup>He particles

## a. <sup>27</sup>Al(<sup>3</sup>He,x)<sup>22</sup>Na

Conditionally accept the spline fit, check unpublished data from Brookhaven (action Tárkányi), if necessary do new spline fit (action Tárkányi, Zhuang). Energy range: 10-100 MeV.

## b. <sup>27</sup>Al(<sup>3</sup>He,x)<sup>24</sup>Na

Add into the list (action Tárkányi, Shubin and Zhuang) Energy range: to be defined (action Tárkányi).

## c. $^{nat}Ti(^{3}He,x)^{48}V$

Include all selected data in the energy range close to the threshold, increase uncertainties (action Tárkányi), do new Pade fit (action Shubin) and spline fit (action Zhuang), do final selection (action Tárkányi).

Energy range: 10-100 MeV.

## d. <sup>nat</sup>Cu(<sup>3</sup>He,x)<sup>66,67</sup>Ga,<sup>65</sup>Zn

Remove from the list due to variety of problems (conflicting data for <sup>65</sup>Zn, very small cross sections for <sup>67</sup>Ga).

## Alpha particles

a.  ${}^{27}Al(\alpha, x){}^{22}Na$ 

Accept spline fit as recommended data. Energy range: 40-100 MeV.

b.  ${}^{27}Al(\alpha, x){}^{24}Na$ 

Add into the list, do selection of experimental data (action Tárkányi), do Pade fit (action Shubin) and spline fit (action Zhuang), do final selection (action Tárkányi). Energy range: 50-100 MeV.

c. <sup>nat</sup>Ti( $\alpha$ ,x)<sup>51</sup>Cr

Accept Pade fit as recommended data. Energy range: 6-40 MeV.

d.  $^{nat}Cu(\alpha, \mathbf{x})^{66}Ga$ 

Accept Pade fit as recommended data. Energy range:10-30 MeV.

e.  $^{nat}Cu(\alpha, \mathbf{x})^{67}Ga$ 

Accept Pade fit as recommended data. Energy range: 18-50 MeV.

f.  $^{nar}Cu(\alpha, x)^{65}Zn$ 

Accept Pade fit as recommended data. Energy range: 18-45 MeV.

# 3.2 Gamma Emitters

a. <sup>67</sup>Zn(p,n)<sup>67</sup>Ga

Obninsk Alice and Gul (Islamabad) calculations were found to be very near to the selected data. It is, however, suggested that the fit be accepted as recommended data (action Hermanne).

Energy range: threshold - 25 MeV.

## b. <sup>68</sup>Zn(p,2n)<sup>67</sup>Ga

Remove Kopecky data, add new Debrecen and Brussels data (action Tárkányi), then fit with Pade and spline (action Shubin and Zhuang), do final selection (action Hermanne). Energy range: threshold - 30 MeV.

# c. ${}^{82}$ Kr(p,2n) ${}^{81}$ Rb

Only one data set of Kovacs *et al* is available, but renormalisation of <sup>nat</sup>Kr data will be done up to 20 MeV (action Nortier), do new fits (action Shubin, Zhuang), do final selection (action Hermanne).

Energy range: threshold - 30 MeV.

### d. <sup>nat</sup>Kr(p,x)<sup>81</sup>Rb

Deselect <sup>nat</sup>Kr data of Kovacs *et al*, renormalise Kovacs data on enriched <sup>82</sup>Kr to <sup>nat</sup>Kr up to 20 MeV, add Acerbi data to both sets of Steyn data (action Nortier), do fits (action Shubin, Zhuang), do final selection (action Hermanne). Energy range: threshold - 30 MeV.

### e. ${}^{111}Cd(p,n){}^{111}In$

It was suggested during the last meeting that Obninsk should do new Alice calculations with smaller energy steps around the peak. This was done and the agreement is good. Accept Alice as recommended data.

Energy range: threshold -30 MeV.

### f. <sup>112</sup>Cd(p,2n)<sup>111</sup>In

There was a 10% inconsistency between the experimental data and the Obninsk Alice calculations around the peak. During the last meeting it was decided to resolve this discrepancy by performing new cross section measurements. No new measurement was done. Rescale Alice by about 10% (exact value action Hermanne, rescaling action Hermanne), do new spline fit (action Zhuang), do final selection (action Hermanne). Energy range: threshold - 35 MeV.

## g. $^{nat}Cd(p,x)^{111}In$

This reaction was dropped from the final list.

# h. <sup>123</sup>Te(p,n)<sup>123</sup>I

The Obninsk Alice results were rescaled to reproduce the experimental peak value based on data of Scholten *et al* and Mahunka *et al*. The calculations were then accepted as the recommended values.

Energy range: threshold - 20 MeV.

### i. <sup>124</sup>Te(p,2n)<sup>123</sup>I and <sup>124</sup>Te(p,n)<sup>124</sup>I

The theoretical results from Obninsk for the (p,2n) reaction are accepted after rescaling the curve so that the experimental peak value of Scholten et al was reproduced, no action needed.

Energy range: threshold - 30 MeV.

The (p,n) reaction is important in connection with impurity consideration. The rescaled theoretical results from Obninsk are accepted as recommended values. Energy range: threshold - 30 MeV.

## j. ${}^{127}I(p,5n){}^{123}Xe \text{ and } {}^{127}I(p,3n){}^{125}Xe$

The Obninsk Alice results for the (p,5n) reaction were rescaled so that the experimental peak value of selected data was in agreement. It is suggested that Pade smoothing is done on the renormalised Alice results (action Shubin), final selection (action Hermanne). Energy range: threshold - 100 MeV.

In order to be able to estimate the  $^{125}I$  impurity in  $^{123}I$ , it is recommended that the (p,3n) reaction be treated the same way as the (p,5n) reaction (actions Nortier, Shubin), final selection (action Hermanne).

Energy range: threshold - 100 MeV if data available.

## k. <sup>124</sup>Xe(p,2n)<sup>123</sup>Cs and <sup>124</sup>Xe(p,pn)<sup>123</sup>Xe

In view of the great importance of these reactions for production of <sup>123</sup>I, they should be included. Data collection to be done by Jülich (action Qaim) and sent to Tárkányi for dissemination, do model calculation (actions Shubin, Mustafa and Gul), do fitting (action Shubin, Zhuang), do final selection (action Hermanne). Energy range: threshold - 40 MeV.

## l. <sup>203</sup>Tl(p,3n)<sup>201</sup>Pb, <sup>203</sup>Tl(p,2n)<sup>202</sup>Pb and <sup>203</sup>Tl(p,4n)<sup>200</sup>Pb

The data for the (p,3n) reaction were considered up to 35 MeV and the Obninsk Alice results were found to support the higher values of Hermanne et al. However, considering all the selected results, it was decided that rescaling of Alice is necessary (action Hermanne), new spline fit will be done (action Zhuang), final selection (action Hermanne). Energy range: 20-35 MeV.

Furthermore, due consideration should be given to the  ${}^{203}Tl(p,2n){}^{202}Pb$  and  ${}^{203}Tl(p,4n){}^{200}Pb$  reactions which lead to the impurities  ${}^{202}Tl$  and  ${}^{200}Tl$ , respectively. Do also here data compilation, data selection, model calculation, spline fit and Pade fit (actions Hermanne Mustafa, Shubin, Gul, Zhuang).

Energy range: 20-35 MeV.

# 3.3 Positron Emitters

### a. ${}^{14}N(p,\alpha){}^{11}C$

Large number of available data showing large scatter. Do thorough screening, select consistent data, consider nuclear structures of compound system and product nucleus, deduce energy positions of expected resonances (action Qaim). Do Pade and spline fittings and send results in fine energy steps (actions Shubin, Zhuang), do final selection (action Qaim).

Energy range: threshold - 25 MeV.

## b. ${}^{16}O(p,\alpha){}^{13}N$

Select data, do energy adjustment, suggest resonances (action Qaim). Do Pade and spline fittings (actions Shubin, Zhuang) in fine energy steps, do final selection (action Qaim). Energy range: threshold - 20 MeV.

## c. <sup>14</sup>N(d,n)<sup>15</sup>O

Selected data fitted well both with Pade and spline fittings. Accept Pade as recommended data, submit results in fine energy steps (action Shubin). Energy range: threshold - 15 MeV.

## d. <sup>15</sup>N(p,n)<sup>15</sup>O

Compile and select data (action Qaim), do Pade and spline fits in fine energy steps (actions Shubin, Zhuang), do final selection (action Qaim). Energy range: threshold - 15 MeV.

## e. <sup>18</sup>O(p,n)<sup>18</sup>F

Only Ruth data are reliable. Suggest resonances (action Qaim). Do Pade and spline fittings (actions Shubin, Zhuang) in fine energy steps, do final selection (action Qaim). Energy range: threshold - 15 MeV.

## f. $^{nat}Ne(d,\alpha)^{18}F$

Three sets of consistent data available (Nozaki, Backhausen, Fenyvesi), Pade and spline fittings are good. Submit fits in fine energy steps (actions Shubin, Zhuang). Energy range: threshold - 20 MeV.

## g. <sup>69</sup>Ga(p,2n)<sup>68</sup>Ge

Reselect data (action Hermanne), do new Pade and spline fittings with <u>all</u> selected data (actions Shubin, Zhuang), do final selection (action Qaim). Energy range: threshold - 40 MeV.

## h. <sup>nat</sup>Ga(p,x)<sup>68</sup>Ge

Results for the  ${}^{69}\text{Ga}(p,2n){}^{68}\text{Ge}$  reaction were taken from spline fitting and those for the  ${}^{71}\text{Ga}(p,4n){}^{68}\text{Ge}$  reaction from SPEC. Again spline fitting was done. Result accepted but not as recommended data. Produce data sheet (action Tárkányi). Energy range: threshold - 100 MeV.

## i. <sup>85</sup>Rb(p,4n)<sup>82</sup>Sr

Show selected experimental data together with SPEC and Alice calculations. Accept but not as recommended data.

Energy range: threshold - 70 MeV.

## j. <sup>nat</sup>Rb(p,x)<sup>82</sup>Sr

Show selected experimental data together with SPEC and Alice calculations. Do not show fitting. Accept but not as recommended data. Energy range: threshold - 100 MeV.

# 3.4 Actions and deadlines

## 1. Actions by all evaluators

• See the above text.

## 2. Actions on all contributors to experimental evaluation

- Provide description of selection of experimental data (actions Tárkányi, Hermanne, Nortier, Qaim, Scholten, 1 December 1998.)
- Provide errors to selected data points, either stated uncertainties or 15 (minimum) to 20 (maximum) % (actions Tárkányi, Hermanne, Nortier, Qaim, Scholten, 1 December 1998).

## 3. Other actions

- Supply the fitted data in steps of 0.1 MeV, except for positron emitters where very fine steps may be needed (actions Shubin, Zhuang, 15 February 1999). Select the data for the recommended table (action Tárkányi, Hermanne and Qaim as required).
- Send copies of all file transfers to Tárkányi (action as needed).

### 4. Deadlines for completion of the Database

- Selection of experimental data will be done by 1 December 1998 (actions Tárkányi, Nortier, Hermanne, Scholten).
- Results will be sent to Shubin and Zhuang by 15 December 1998 (action Tárkányi).
- Fits will be sent to Tárkányi, Hermanne and Qaim as required, by 15 February1999 (actions Shubin, Zhuang).
- Database will be completed by 31 May 1999 (action Tárkányi).

# **4** Recommendations for the TECDOC

The draft n° 1 was discussed and the recommendations were made concerning the outline and contents, style and format of the document. The title of the TECDOC will be "Charged-Particle Cross Section Database for Medical Radioisotope Production", subtitle "Diagnostic Radioisotopes and Monitor Reactions". The total length will be about 300 pages.

## 4.1 Revised outline

- Foreword (Obložinský, 1 page)
- 1. Introduction (Qaim, 6 pages)
  - 1.1. Data needs
  - 1.2. Reactions studied
  - 1.3. Overview of the present TECDOC

## 2. Experimental Evaluation (20 pages)

- 2.1. Compilation of experimental data (Tárkányi)
- 2.2. Analysis of experimental data (Tárkányi)
- 2.3. Methods of fitting (Obložinský)
  - 2.3.1. Pade fit (Shubin)
  - 2.3.2. Spline fit (Zhuang)

References

### 3. Theoretical Evaluation (Mustafa, 25 pages)

- 3.1. Nuclear reaction models
- 3.2. Codes and calculations
- 3.3. Discussion
- References

### 4. Beam Monitor Reactions (Tárkányi, 100 pages)

4.1. Protons

<sup>27</sup>Al(p,x)<sup>22</sup>Na <sup>27</sup>Al(p,x)<sup>24</sup>Na <sup>nat</sup>Ti(p,x)<sup>48</sup>V <sup>nat</sup>Ni(p,x)<sup>57</sup>Ni <sup>nat</sup>Cu(p,x)<sup>56</sup>Co <sup>nat</sup>Cu(p,x)<sup>62</sup>Zn <sup>nat</sup>Cu(p,x)<sup>63</sup>Zn <sup>nat</sup>Cu(p,x)<sup>65</sup>Zn

- 4.2. Deuterons  ${}^{27}Al(d,x){}^{22}Na$   ${}^{27}Al(d,x){}^{24}Na$   ${}^{nat}Fe(d,x){}^{56}Co$  ${}^{nat}Ni(d,x){}^{61}Cu$
- 4.3. <sup>3</sup>He particles <sup>27</sup>Al(<sup>3</sup>He,x)<sup>22</sup>Na <sup>27</sup>Al(<sup>3</sup>He,x)<sup>24</sup>Na <sup>nat</sup>Ti(<sup>3</sup>He,x)<sup>48</sup>V
- 4.4. Alpha particles  ${}^{27}Al(\alpha,x){}^{22}Na$   ${}^{27}Al(\alpha,x){}^{24}Na$   ${}^{nat}Ti(\alpha,x){}^{51}Cr$   ${}^{nat}Cu(\alpha,x){}^{66}Ga$   ${}^{nat}Cu(\alpha,x){}^{67}Ga$  ${}^{nat}Cu(\alpha,x){}^{65}Zn$

### 5. Production Reactions for Diagnostic Radioisotopes (Tárkányi, 150 pages)

- 5.1. Gamma emitters  ${}^{67}Zn(p,n){}^{67}Ga$   ${}^{68}Zn(p,2n){}^{67}Ga$   ${}^{82}Kr(p,2n){}^{81}Rb$   ${}^{nat}Kr(p,x){}^{81}Rb$   ${}^{111}Cd(p,n){}^{111}In$   ${}^{122}Cd(p,2n){}^{111}In$   ${}^{122}Cd(p,2n){}^{123}I$   ${}^{124}Te(p,2n){}^{123}I$  and  ${}^{124}Te(p,n){}^{124}I$   ${}^{127}I(p,5n){}^{123}Xe$  and  ${}^{127}I(p,3n){}^{125}Xe$   ${}^{124}Xe(p,2n){}^{123}Cs$  and  ${}^{124}Xe(p,pn){}^{123}Xe$  ${}^{203}Tl(p,3n){}^{201}Pb, {}^{203}Tl(p,2n){}^{202}Pb$  and  ${}^{203}Tl(p,4n){}^{200}Pb$
- 5.2. Positron emitters  ${}^{14}N(p,\alpha){}^{11}C$   ${}^{16}O(p,\alpha){}^{13}N$   ${}^{14}N(d,n){}^{15}O$   ${}^{15}N(p,n){}^{15}O$   ${}^{18}O(p,n){}^{18}F$   ${}^{20}Ne(d,\alpha){}^{18}F$   ${}^{69}Ga(p,2n){}^{68}Ge$   ${}^{nat}Ga(p,x){}^{68}Ge$   ${}^{85}Rb(p,4n){}^{82}Sr$  ${}^{nat}Rb(p,x){}^{82}Sr$

Appendix: Guidance to Users of Data (Qaim, 10 pages maximum)

Contributors to Drafting and Review (2 pages)

# 4.2 Comments on individual chapters

## 1. Introduction

- Justify beam monitor reactions, particularly for <sup>3</sup>He and alpha particles.
- Explain why CRP focused on diagnostic radioisotopes.

## 2. Experimental Evaluation

- Describe methodology, provide illustrative figures.
- Explain description of the fitting procedures, provide more general introduction on Pade fit, provide illustrative figures.

## 3. Theoretical Evaluation

- Expand to about 25 pages, provide illustrative figures.
- Mention appropriate theory for light nuclei.
- Explain the role of the theory in the present CRP (guidance rather than full evaluation).

## 4. Beam Monitor Reactions

Each reaction will be described in 4-5 pages, including:

- Bibliography for experimental data.
- Description of selection (1/2 page maximum).
- Fig. 1: All experimental data.
- Fig. 2: Selected experimental data compared with theory.
- Fig. 3: Selected experimental data with fitted curves.
- Justification for recommended data (1 paragraph).
- Table with recommended data (generally in 0.5 MeV steps, to be adjusted as needed).

# 5. Production Reactions for Diagnostic Radioisotopes

The same conclusions apply as for the chapter 4. In addition, there will be:

- Fig. 4: Yields.
- Table with yields in 3 units in 0.5 MeV steps.

# 4.3 Deadlines

The following deadlines were agreed upon towards the completion of the TECDOC:

- Chapters 1, 2, and 3 will be completed by 1 December 1998 (Qaim, Tárkányi, Mustafa).
- Text for Chapters 4, 5 and if possible also for Appendix will be prepared by 1 May 1999 (Tárkányi, Qaim).
- All figures for Chapters 4 and 5 will be prepared by 15 May 1999 (Tárkányi).
- Using the above input, the draft n° 2 will be completed by the end of May 1999 (Obložinský).

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## Final Research Co-ordination Meeting on Development of Reference Charged-Particle Cross Section Database for Medical Radioisotope Production

Brussels, Belgium 28 September - 2 October 1998

# AGENDA

### Monday, 28 September

#### 09:00-09:20 Opening Session

- Opening address
   (M. Despotin, Vice-Rector, University Brussels; P. Obložinský, IAEA Vienna)
- Adoption of Agenda
- Local announcements

### 09:20-12:30 Presentations

- Progress Reports by chief scientific investigators
  - A. Hermanne: Progress Report for Brussels
  - M. Mustafa: Progress Report for Livermore
  - M. Nortier: Progress Report for Cape Town
  - S.M. Qaim: Progress Report for Jülich
  - Y. Shubin: Progress Report for Obninsk
  - F. Tárkányi: Progress Report for Debrecen
  - Y. Zhuang: Progress Report for Beijing
- Other reports by participants
  - K. Gul: Calculations of production cross sections of Zn and Ga radioisotopes through charged particle bombardment of Cu and Zn isotopes in 3-26 MeV energy range
     A. Hermanne: Activation of accelerator shielding

### 12:30-14:00 Lunch Break

### 14:00-18:00 Review of Database: Old List of 14 Reactions from Cape Town

- Monitor reactions (Obložinský/Hermanne)
- Gamma emitters (Obložinský/Hermanne)

### Tuesday, 29 September

### 08:30-12:30 Review of Database: New List of 26 Reactions from Cape Town

- Monitor reactions (discussion leader: Tárkányi)

### 12:30-14:00 Lunch Break

### 14:00-18:00 Review of Database: New List of 26 Reactions from Cape Town

- Gamma emitters (discussion leader: Hermanne)

## Wednesday, 30 September

### 08:30-12:30 Review of Database: New List of 26 Reactions from Cape Town

- Positron emitters (discussion leader: Qaim)

## 12:30-14:00 Lunch Break

## 14:00-17:30 Review of Database: New List of 26 Reactions from Cape Town

- Continuation

## 17:30-21:00 Evening Programme

- Visit to the cyclotron company IBA
- Meeting Dinner (wives invited)

## Thursday, 1 October

## 08:30-12:30 Review of TECDOC

- Structure, content and layout
- Editing of chapters
  - 1. Introduction (Qaim)
  - 2. Experimental evaluation (Tárkányi)
  - 3. Theoretical evaluation (Mustafa)
  - 4. Reference cross section database (Hermanne)
  - 5. Validation of data (Nortier)
  - 6. Yields of production reactions (Tárkányi)
  - Attachment (Qaim)

## 12:30-14:00 Lunch Break

## 14:00-16:00 Review of TECDOC

- Editing of chapters

## 16:00-18:00 Drafting of the meeting report

## Friday, 2 October

### 08:30-12:30 Concluding Session

- Adoption of the meeting report
- Summary of CRP results

### 12:30-14:00 Lunch Break

### 14:00-17:00 Concluding Session

- Final discussion
- Adjournment

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### Final Research Co-ordination Meeting on Development of Reference Charged-Particle Cross Section Database for Medical Radioisotope Production

Brussels, Belgium 28 September - 2 October 1998

# LIST OF PARTICIPANTS

#### **BELGIUM**

#### **Alex Hermanne**

Cyclotron Department Vrijie Universiteit Brussel Laarbeklaan, 103 B-1090 Brussels Tel.: +32-2-477-48-60 Fax: +32-2-477-48-55 E-mail: aherman@vucy.vub.ac.be

#### <u>CHINA</u>

Youxiang Zhuang China Nuclear Data Centre China Institute of Atomic Energy P.O. Box 275 (41) 102413 Beijing Tel.: +86-10-935-7830 Fax: +86-10-935-7008 E-mail: yxzhuang@mipsa.ciae.ac.cn

#### **GERMANY**

Syed M. Qaim Institut für Nuklearchemie Forschungszentrum Jülich GmbH Postfach 1913 D-52425 Jülich Tel.: +49-2461-61-3282 Fax: +49-2461-61-2535 E-mail: s.m.qaim@fz-juelich.de

#### **HUNGARY**

Ferenc Tárkányi Institute of Nuclear Research Hungarian Academy of Sciences Bem ter 18/c P.O. Box 51 H-4001 Debrecen Tel.: +36-52-417-266 Fax: +36-52-416-181 E-mail: tarkanyi@atomki.hu

### <u>RUSSIA</u>

Yuri Shubin Theoretical Division Department of Nuclear Physics Institute of Physics and Power Engineering 249020 Obninsk, Kaluga Region Tel.: +7-084-399-8611 Fax: +7-095-883-3112 or +7-095-230-2326 E-mail: shubin@ippe.rssi.ru

### SOUTH AFRICA

Meiring Nortier Radioisotope Production Group National Accelerator Centre P.O. Box 72 713I Faure Tel.: +27-21-843-3820 Fax: +27-21-843-3525 E-mail: meiring@nac.ac.za

#### **UNITED STATES OF AMERICA**

Mohammed G. Mustafa Lawrence Livermore National Laboratory P.O. Box 808, L-22 Livermore, CA 94550 Tel.: +1-925-422-5523 Fax: +1-925-423-0925 E-mail: mustafa1@llnl.gov

#### **CONSULTANT**

# PAKISTAN

Khunab Gul Scientific Information Division Pakistan Institute of Nuclear Science and Technology P.O. Nilore Islamabad Tel.: +92-847-60109 Fax: +92-51-9290275 E-mail: hakhan@isb.comsats.net.pk *LAEA* Pavel Obložinský Nuclear Data Section Wagramer Strasse 5 A-1400 Vienna Tel.: +43 1 2600 21712 Fax: +43 1 20607 E-mail: oblozinsky@iaeand.iaea.or.at

# **Extended Abstracts of Presented Papers**

1.	A. Hermanne (Brussels, Belgium) Progress Report of the VUB Cyclotron Group Brussels
2.	M.G. Mustafa (Livermore, California, U.S.A.) Progress Report of the Lawrence Livermore National Laboratory
3.	<ul><li>F.M. Nortier (Faure, South Africa)</li><li>Progress Report of IAEA Research Agreement on "Determination of Excitation</li><li>Functions for Medical Radioisotopes"</li></ul>
4.	S.M. Qaim and B. Scholten (Jülich, Germany) Contributions to the Standardization of Nuclear Data for Medical Radioisotope Production
5.	Yu. N. Shubin (Obninsk, Russia) Progress Report on IAEA Research Contract on "Development of Charged Particle Cross Section Calculation Methods for Medical Radioisotope Production"
6.	<ul> <li>F. Tárkányi, S. Takács, F. Szelecsényi (Debrecen, Hungary)</li> <li>Development of Reference Charged Particle Cross Section Data Base for</li> <li>Medical Radioisotope Production</li></ul>
7.	Youxiang Zhuang (Beijing, China) Recommendation of Charged Particle Cross Section for Medical Radioisotope Production

### Progress report of the VUB Cyclotron group Brussels, Prof A. Hermanne

The results obtained in the period April 1997 - September 1998 in the frame of the research on charged particle reactions of nuclear reactions used for beam monitoring and production of medically relevant radionuclides are summarized below.

1. Experimental tasks given at the Cape Town Meeting to the Brussels group.

- The <sup>nat</sup>Ti(p,x)<sup>48</sup>V reaction was reassessed (in collaboration with the Debrecen group) in an irradiation of 25 foils of 25  $\mu$  thick pure Ti in a 15 MeV proton beam. The measured cross sections confirm the earlier values of the maximal cross section value of this reaction.

The results are ready for a common Brussels- Debrecen publication and for insertion in the experimental database.

- The reactions induced on 7.6m thick <sup>nat</sup>Cu by a particles were reassessed in a 41 and 34MeV α-beam (in collaboration with the Debrecen group). The values for the cross sections of the reactions yielding <sup>65</sup>Zn, <sup>67</sup>Ga and <sup>66</sup>Ga were found to be in good accordance with our previous results and will be included in the experimental database. The results are ready for a common Brussels- Debrecen publication.
- The 3 other tasks where less successful.

\* We did not succeed in finding adequate material to produce thin  $^{112}Cd$  targets, hence the  $^{112}Cd(p,2n)^{111}In$  reaction was not remeasured.

\* After reevaluation of the available data of our early 1997 measurements on enriched <sup>68</sup>Zn and <sup>nat</sup>Zn of the <sup>68</sup>Zn(p,2n)<sup>67</sup>Ga reaction and as we had some problems in making enriched <sup>68</sup>Zn targets we decided no to make a new irradiation in 1998. The new data are included in the data set but do not resolve the problems of value and energy of the maximum of the cross section.

\* the intercomparison of monitor reactions (mixed Ni, Cu, Ti stacks) in beams of the different light particles was postponed although reassessment of cross sections at some energies were performed in different experiments.

2. Data compilation and analysis.

- Excitation function for γ-emitters : <sup>209</sup>Bi(α,2n)<sup>211</sup>At.
   Only a limited number of useful data were found in the EXFOR or in the (older) literature. The data of Stickler and Hofstetter (1974) are completely discrepant.
   The data of Lambrecht et al. (1985), Ramler et al. (59) and Kelly and Segré (1949) are consistent and were selected and sent for evaluation.
- Excitation function for positron emitters: <sup>69</sup>Ga(p,2n)<sup>68</sup>Ga.
   Only 3 references containing cross section values were found and are available in the EXFOR database. The publication by Cohen and Newman (1955) contains only 1 data point. The data of Porile (1963) and Levkovskii (1991) are generally in good agreement and together they form the selected data base sent for evaluation.

- 3. Preparation of TECDOC.
- The texts on methods of fitting to experimental data and on systematics were collected (Shubin and Zhouang) and slightly edited (Sections 2.4, and 2.6.) The values for the fits to experimental data for the following reactions were collected from Shubin (PADE) and Zhouang (SPLINE) :  $nat Ti(p,x)^{48}V$ ;  $^{nat}Cu(p,x)^{56}Co; {^{nat}Al(p,x)^{22}Na; {^{nat}Cu(p,x)^{65}Zn; {^{111}Cd(p,n)^{111}In.}}$ Tables and graphs with all available fit data were prepared. (Section 2.5.)
- The data for the recommended values for 5 monitor reactions were collected from Shubin (ALICE). For some of these reactions a better selection of the experimental data has still to be performed.

The data for the reactions  $^{nat}Cu(p,x)^{56}Co$  and  $^{nat}Al(p,x)^{22}Na$  are still missing.

- The data for the recommended values for 8 reactions for production of  $\gamma$ -emitters were collected from Shubin (ALICE) and 1 from Zhouang (Systematics). Tables of values and graphs were prepared for these reactions. (Chapter 4.)
- The data for the reactions to be evaluated as defined in Cape Town and calculated by Shubin (PADE and ALICE) and by Zhuoang (SPLINE and Recommended) are available in Brussels but not yet edited (format problem for Zhouang data).

An experiment that can give some useful additional information for the CRP is now being analyzed : 5 stacks containing foils of <sup>nat</sup>Mo, <sup>100</sup>Mo, Ni, Ti, Cu, Fe, Al, Zr, W were irradiated in 30MeV and 50MeV beams of deuterons at LLN. Results for Al. Ti and Fe are available and confirm the excitation cures already evaluated in this CRP.

Other irradiations performed and analysis ongoing :

- \* 10 $\mu$  Ni foils with 42MeV  $\alpha$  (yielding <sup>62</sup>Zn, <sup>57</sup>Ni, <sup>61</sup>Cu, <sup>58</sup>Co, <sup>56</sup>Co, <sup>61</sup>Co)
- \* 35 $\mu$  Fe with 42 and 31 MeV  $\alpha$
- \* 8 $\mu$  Nb with 41 and 28MeV  $\alpha$  and with 20MeV deuterons
- \* W foils with deuterons and protons (partly published)

# Progress Report Third CRP Meeting, Brussels, Belgium September 28-October 2, 1998

M. G. Mustafa Lawrence Livermore National Laboratory Livermore, California, USA

The Livermore group has made cross section calculations for the following reactions since the last CRP meeting in South Africa. Two codes were used in the calculations: Alice-91 and HMS-Alice.

#### **Monitor reactions**

$^{27}Al(d,x)^{22}Na$	<sup>27</sup> Al( <sup>3</sup> He,x) <sup>22</sup> Na	$^{27}$ Al(a,x) $^{22}$ Na
$^{nat}Ti(d,x)^{48}V$	<sup>nat</sup> Ti( <sup>3</sup> He,x) <sup>48</sup> V	<sup>nat</sup> Ti(a,x) <sup>51</sup> Cr
<sup>1</sup> Fe(d,x) <sup>56,57</sup> Co	<sup>114</sup> Cu( <sup>3</sup> He,x) <sup>66,67</sup> Ga, <sup>65</sup> Zn	<sup>nat</sup> Cu(a,x) <sup>66,67</sup> Ga, <sup>65</sup> Zn
<sup>nat</sup> Ni(d,x) <sup>61</sup> Cu		
Gamma emitter r	reactions	

 $^{67}$ Zn(p,n) $^{67}$ Ga  $^{68}$ Zn(p,2n) $^{67}$ Ga

The Alice-91 code is the latest released version of the standard Alice code at Livermore, which uses hybrid or geometry dependent hybrid precompound models and Weisskopf-Ewing evaporation for the equilibrium part of the reaction process. The HMS-Alice code is based on a new precompound Monte Carlo simulation model. The model permits unlimited multiple precompound emission for each interaction and may be used to calculate exclusive particle spectra and yields. The evaporation part of the calculation is done with the usual Weisskopf-Ewing formalism, as in the Alice–91 code. The Monte Carlo precompound formulation is available at present for neutron and proton induced reactions, but not for deuteron, <sup>3</sup>He or alpha particle induced reactions. The Livermore group therefore used HMS-Alice for proton induced reactions in this CRP, but used the Alice-91 code for other reactions.

The calculations for this CRP are based on global optical potentials included in the code, and two level density options: Fermi gas level density or shell dependent level densities given by the model of Kataria and Ramamurthy. For deuteron induced reactions Livermore studied the effects of the deuteron breakup in the entrance channel using a microscopic theory developed by Tamura and Udagawa (TU) coupled with a modified Alice-91 code. In this approach the TU model was used to deduce the spectra of neutrons and protons transferred into the target nucleus (stripping to bound states and breakupcapture in the continuum); these were then assumed to initiate three quasiparticle cascades. The deuteron cross section that does not undergo stripping and breakup-capture was assumed to initiate a separate preequilibrium cascade. Detailed calculations were done for deuteron induced reactions on <sup>48</sup>Ti. The results were used to choose the precompound initial exciton number parameters for the remaining deuteron induced reactions for this CRP. The total number of initial excitons thus chosen in the calculation is two, i. e., one proton and one neutron above the Fermi surface. The <sup>3</sup>He induced reactions are also known to have a sizeable breakup cross section. This problem has not been addressed here. Therefore, the choice of the initial exciton number for <sup>3</sup>He induced reactions and also for alpha induced reactions is arbitrary. Four excitons, two protons and 1 neutron (and one hole) are used for <sup>3</sup>He induced reactions, and four excitons, two protons and two neutrons, for alpha induced reactions. Because of the global nature of these parameters, the calculated cross sections for this CRP should be used as a guide rather than a fit to the experimental data. Using a single global parameter choice allows an estimation of the predictive powers of the models; varying parameters to fit each data set precludes this.

The calculated cross sections have been compared with selected experimental data. The results are shown in seventeen separate figures. Exceptionally poor fit to the data are found for all the reactions on <sup>27</sup>Al target, and <sup>nat</sup>Cu(<sup>3</sup>He,x) <sup>65</sup>Zn and <sup>nat</sup>Cu(a,x) <sup>65</sup>Zn reactions. The fit to the data for the rest of the reactions was good.

# **Progress Report from NAC, Faure**

## IAEA Research agreement No. 8601/CF on "Determination of Excitation Functions for Medical Radioisotopes"

F.M. Nortier, National Accelerator Centre, P.O. Box 72, Faure 7131, South Africa

This report summarises the progress in the work done at the NAC since the second Research Coordination Meeting on "Development of Reference Charged-Particle Cross Section Database for Medical Radioisotope Production" in Cape Town, April 1997.

### 1. <sup>27</sup>Al(p,X)<sup>22</sup>Na and <sup>nat</sup>Cu(p,X)<sup>56</sup>Co monitor reactions

According to the Work Plan agreed upon at the previous meeting in Cape Town a critical analysis of the published cross section data for the  ${}^{27}Al(p,X){}^{22}Na$  and  ${}^{nat}Cu(p,X){}^{56}Co$  monitor reactions was performed. In the case of  ${}^{27}Al(p,X){}^{22}Na$  a total of 19 data sets were available. In view of the abundance of published data only 15 sets for which the data were available in numerical form (either from the publication or in EXFOR) were considered. Of these, 10 data sets were selected for evaluation. In the case of  ${}^{nat}Cu(p,X){}^{56}Co$  a total of 6 data sets were critically analysed and 3 sets were selected for evaluation

### 2. $^{127}$ I(p,5n) $^{123}$ Xe

A critical analysis was also performed of the published cross section data for the  $^{127}I(p,5n)^{123}Xe$  reaction employed in the production at intermediate energies of the  $\gamma$ -emitter,  $^{123}I$ . A total of 9 papers with experimental data were found of which 3 present numerical values and values for 3 more were available in EXFOR. Figures from the remaining 3 were digitised. Experimental values from 7 papers were used to determine an "experimental peak value" which was subsequently supplied to the theoretical group at Obninsk for re-scaling of the theoretical curve.

#### 3. Additional nuclear reactions

The NAC group was also responsible for the compilation and critical analysis of published cross section data for the following additional nuclear reactions:

γ-emitters:	$^{nat}$ Kr(p,xn) $^{81}$ Rb,	${}^{82}$ Kr(p,2n) ${}^{81}$ Rb
$\beta$ -emitters:	$^{nat}Rb(p,xn)^{82}Sr,$	$^{85}$ Rb(p,4n) $^{82}$ Sr

A total of 9 papers, presenting 10 data sets (6 for  ${}^{nat}Kr(p,xn)^{81}Rb$ ; 1 for  ${}^{82}Kr(p,2n)^{81}Rb$ ; 2 for  ${}^{nat}Rb(p,xn)^{82}Sr$ ; 1 for  ${}^{85}Rb(p,4n)^{82}Sr$ ) were found in the literature search. Four data sets (given in figure form only) were digitised and all the data were critically analysed. Apart from in the case of the  ${}^{nat}Kr(p,xn)^{81}Rb$  reaction where 3 data sets were not selected, all other data were selected for evaluation of the relevant excitation functions.

### 4. Inter-comparison of monitor reactions

A foil stack consisting of Al and Cu foils and mounted in a Faraday cup was irradiated at NAC with a 66 MeV proton beam for an inter-comparison of experimental cross section data for the  ${}^{27}\text{Al}(p,X){}^{22}\text{Na}$  and  ${}^{nat}\text{Cu}(p,X){}^{62}\text{Zn}$  monitor reactions. However, the results are not yet available since the data analysis still needs to be completed.

### 5. Integral measurements

Thick-target irradiations in a Faraday cup for integral measurements for the  ${}^{127}I(p,5n){}^{123}Xe \rightarrow {}^{123}I$  and  ${}^{nat}Rb(p,xn){}^{82}Sr$  production routes have also not been done yet.

# Contributions to the Standardization of Nuclear Data for Medical Radioisotope Production

by S.M. Qaim and B. Scholten, Institut für Nuklearchemie, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

Progress Report (Research Agreement No. 8598/CF)

The progress achieved in the work since the second CRP meeting in April 1997 is summarized below:

### a) Information collection and dissemination

During the last meeting, we had accepted the responsibility to collect all available information on the following nuclear reactions commonly used in the production of important  $\beta^-$  emitters:

A thorough search of the literature data was done, and results obtained both via activation and neutron spectrum measurement techniques were taken into account. Due to strong structure effects in the light mass region considered here, the excitation functions show large fluctuations: often it is not possible to distinguish whether the fluctuation in the data is genuine or whether the values have large errors. Through a careful screening, several erroneous results were deleted, and the whole information was sent to Dr. F. Tárkányi at Debrecen for onward transmission to the evaluators. The effort involved was much more than anticipated. The material reached the evaluators with a delay of about two months.

### b) Experimental measurements

- During the production of <sup>15</sup>O via the <sup>14</sup>N(d,n)-process small amounts of <sup>13</sup>N and <sup>11</sup>C are also formed. The cross sections of the contributing processes were not known with high accuracy. We measured (in cooperation with ATOMKI, Debrecen) the excitation functions of the <sup>14</sup>N(d,t)<sup>13</sup>N and <sup>14</sup>N(d,αn)<sup>11</sup>C reactions from their respective thresholds up to 12.3 MeV. Furthermore, some data on the important production reaction <sup>14</sup>N(d,n)<sup>15</sup>O were also obtained which have been included in the on-going evaluation of this process.
- Measurements on the excitation function of the <sup>18</sup>O(p,n)<sup>18</sup>F reaction initiated about a year ago were continued using 95% enriched <sup>18</sup>O<sub>2</sub>. Several irradiations were done in the energy range of 10 to 20 MeV, both at Jülich and Debrecen, and data analysis is in progress.
- A new  $\beta^-$  emitter  ${}^{120g}I$  (T<sub>1/2</sub> = 1.35 h) was suggested for PET studies. Its production methods, viz.  ${}^{122}Te(p,3n)$  and  ${}^{120}Te(p,n)$ -reactions, were investigated in detail using highly enriched isotopes as target materials.

## c) Future plans

- Completion of measurements on the <sup>18</sup>O(p,n)<sup>18</sup>F process
- Formulation of guidelines (together with Dr. F. Tárkányi) on the measurement and use of evaluated nuclear data
- Identification of discrepancies in the evaluated data
- Contributions to the TEC DOC on the CRP

# PROGRESS REPORT

# on Agency Research Contract No. 8599/R1/R2

# DEVELOPMENT OF CHARGED PARTICLE CROSS SECTION CALCULATION METHODS FOR MEDICAL RADIOISOTOPE PRODUCTION

Yu. N. Shubin

Institute of Physics and Power Engineering 249020 Obninsk, Russia

Time period covered: 1997/04/15 to 1998/09/22

September 1998, Obninsk

### **SUMMARY**

The activity on the development of charged particle cross section calculation methods for medical radioisotope production was carried out according to second year working plan outlined in Summary Report of 2nd CRP Meeting, Faure, South Africa, 7-10 April 1997 (IAEA, INDC(NDS-371) in several directons:

I. The evaluation of first 14 reactions have been completed.

a) Evaluation of proton induced reactions for 7 beam monitor reaction excitation functions on Ti, Ni, Cu and Al.

According to the recommendation of the 2nd CRP Meeting and working plan:

We recalculated the  $^{nat}Ni(p,x)^{57}Ni$  reaction cross section in the energy range up to 50 MeV and table of numerical data were prepared.

The  $^{nat}Ti(p,x)^{48}V$  reaction excitation function was recalculated with small energy steps and numerical tables were prepared. Experimental data were fitted also using Pade approximation method developed in Obninsk, IPPE. Numerical tables of the approximant were prepared also.

For the  $^{nat}Cu(p,x)^{56}Co$  reaction after a critical analysis of experimental data (performed in Faure) the good description of the experimental data was achieved using Pade approximation method and selected experimental data.

The same procedure was applied to the  ${}^{27}Al(p,x){}^{22}Na$  excitation function using selected experimental data. A rather good desciption of experimental data was achieved with Pade approximation method and the numerical data tables were prepared for the approximant.

Excitation functions for proton induced reactions on Cu were recalculated with a smaller energy steps. Numerical data tables for the  $^{nat}Cu(p,x)^{62}Zn$ ,  $^{nat}Cu(p,x)^{63}Zn$  and  $^{nat}Cu(p,x)^{65}Zn$  reactions were prepared. For the  $^{nat}Cu(p,x)^{65}Zn$  reaction the fit to the selected experimental data was done and tables of numerical data were prepared.

b) Theoretical evaluation of proton induced reactions for production of gamma-emitter In-111, I-123 and Tl-201 were completed.

Excitation functions for the  ${}^{111}Cd(p,n) {}^{111}In$  and  ${}^{112}Cd(p,2n) {}^{111}In$  reaction were recalculated with smaller energy steps and tables of numerical data were prepared.

Excitation function for the  ${}^{123}$ Te(p,n) ${}^{123}$ I reaction was recalculated with smaller energy steps and normalized to experimental peak value of 665 mb. Tables of numerical data were prepared.

For the  ${}^{124}$ Te(p,2n) ${}^{123}$ I reaction the excitation function was recalculated with smaller energy steps an normalised to the experimental peak value of 995 mb.

Excitation function for the  ${}^{127}I(p,5n){}^{123}Xe$  reaction was recalculated with smaller energy steps and normalized to peak value 350 mb at 55 MeV proton energy. Tables of numerical data were prepared also.

For the  ${}^{203}$ Tl(p,3n) ${}^{201}$  reaction the excitation function was recalculated with smaller energy steps and detailed tables of numerical data were prepared.

c) Theoretical calculations of deuteron and alpha induced reactions for selected beam monitor reactions:  $Al(d,x)^{22}Na$ ,  $^{nat}Ti(d,x)^{48}V$ ,  $^{nat}Fe(d,x)^{56}Co$ ,  $^{nat}Ni(d,x)^{61}Cu$ ,  $Al(\alpha,x)^{22}Na$ ,  $^{nat}Ti(\alpha,x)^{51}Cr$ ,  $^{nat}Cu(\alpha,x)^{66,67}Ga$ ,  $^{65}Zn$  were performed.

d) Excitation functions for selected positron emitters were calculated.

II. The calculations for new 27 reactions listed in the Summary Report of the Second CRP Meeting in Cape Town using the ALICE-IPPE code were performed and Pade approximation method was used for the description of the experimental data and creation of Pade approximants for all these reactions. The corresponding Figures and Tables of numerical data have been prepared.

The following reactions were analysed:

Monitor reactions

Monitor reactions for deuterons

 $AI(d,x)^{22}Na, {}^{nat}Ti(d,x)^{48}V, {}^{nat}Fe(d,x)^{56}Co, {}^{nat}Ni(d,x)^{61}Cu,$ 

Monitor reactions for 3He particles

 $Al({}^{3}He, x), {}^{nat}Ti({}^{3}He, x){}^{61}Cu, {}^{nat}Cu({}^{3}He, x){}^{66}Ga, {}^{nat}Cu({}^{3}He, x){}^{67}Ga, {}^{nat}Cu({}^{3}He, x){}^{65}Zn,$ 

Monitor reactions for  $\alpha$  particles

Al
$$(\alpha, x)^{22}$$
Na, <sup>nat</sup> Ti $(\alpha, x)^{51}$ Cr, <sup>nat</sup>Cu $(\alpha, x)^{66}$ Ga, <sup>nat</sup>Cu $(\alpha, x)^{67}$ Ga, <sup>nat</sup>Cu $(\alpha, x)^{65}$ Zn

Excitation functions for y-emitters

 ${}^{67}Zn(p,n){}^{67}Ga, {}^{68}Zn(p,2n){}^{67}Ga, {}^{209}Bi(p,2n){}^{211}At, {}^{nat}Kr(p,x){}^{81}Rb, {}^{82}Kr(p,x){}^{81}Rb,$ 

Excitation functions for positron emitters

<sup>14</sup>N(p, $\alpha$ )<sup>11</sup>C, <sup>16</sup>O(p,  $\alpha$ )<sup>13</sup>N, <sup>14</sup>N(d,x)<sup>15</sup>O, <sup>18</sup>O(p,n)<sup>18</sup>F, <sup>20</sup>Ne(d,  $\alpha$ )<sup>18</sup>F, <sup>69</sup>Ga(p,2n)<sup>68</sup>Ge, <sup>85</sup>Rb(p,4n)<sup>82</sup>Sr, <sup>nat</sup>Rb(p,x)<sup>82</sup>Sr.

III. Contribution to the CRP final report describing the models and code used was prepared and sent to IAEA Nuclear Data Section (see INDC(NDS)-410, 1998).

Two papers " Evaluation of calculation methods for excitation functions for production of radioisotopes iodine, thallium and other elements" and "Possibility of thallium-199 production by proton bombardment of mercury-200" were prepared, sent to editor and accepted for the publication in *Applied Radiation and Isotopes*. Numerical data were sent to the IAEA Nuclear Data Section.

### Development of Reference Charged Particle Cross Section Data Base for Medical Radioisotope Production

#### **Progress Report**

Submitted to Third Research Coordination Meeting Brussel, 28 September-02 October, 1998

### F. Tárkányi, S. Takács, F. Szelecsényi Cyclotron Department, Institute of Nuclear Research of the Hungarian Academy of Sciences H-4001 Debrecen, Pf. 51, Hungary

After the Second Research Co-ordination Meeting in Cape Town, South Africa, 7-10 April 1997 the activity of the Debrecen (ATOMKI) charged particle group in the CRP tasks can be summarized as follows:

#### **Compilation and critical selection:**

Excitation functions of monitor reactions:

According to the adopted work plan of the evaluation procedure the ATOMKI group has compiled all the proposed monitor reactions for deuterons, <sup>3</sup>He- and alpha particles:

Reaction	Number of compiled work	Number of selected work
$^{nat}Al(d,x)^{22}Na$	4	2
$^{nat}Ti(d,x)^{48}V$	4	4
$^{nat}Fe(d,x)^{56}Co$	8	6
$^{nat}Ni(d,x)^{61}Cu$	5	3
$^{nat}Al(^{3}He,x)^{22}Na$	7	6
$^{nat}Ti(^{3}He,x)^{48}V$	3	3
<sup>nat</sup> Cu( <sup>3</sup> He,x) <sup>66</sup> Ga	6	5
<sup>nat</sup> Cu( <sup>3</sup> He,x) <sup>67</sup> Ga	3	3
$^{nat}Cu(^{3}He,x)^{65}Zn$	5	5
$^{nat}Al(\alpha,x)^{22}Na$	13	10
$^{nat}Ti(\alpha,x)^{51}Cr$	9	6
$^{nat}Cu(\alpha,x)^{66}Ga$	17	10
$^{nat}Cu(\alpha,x)^{67}Ga$	14	8
$^{nat}Cu(\alpha,x)^{65}Zn$	15	9

### Excitation functions of gamma-emitters

The experimental data of the following reactions were investigated:

Reaction	Number of compiled work	Number of selected work
	-	
<sup>67</sup> Zn(p,n) <sup>67</sup> Ga	9	6
$^{68}$ Zn(p,2n) $^{67}$ Ga	11	8

#### Excitation functions of positron emitters

In collaboration with the INC, Forschungszentrum Jülich, Jülich, Germany the following reactions were also compiled.:

Reaction	Number of compiled work	Number of selected work
$^{14}N(p,\alpha)^{11}C$	12	9
$^{16}O(p,\alpha)^{13}N$	11	10
$^{14}N(d,n)^{15}O$	4	4
$^{18}O(p,n)^{18}F$	5	5
$^{20}$ Ne(d, $\alpha$ ) <sup>18</sup> F	6	6

#### **Experimental measurements**

To clear the discrepancies and to complete the existing experimental data base new measurements were also performed. Excitation functions of **16 reactions** were remeasured or measured for the first time, among them:

- Additional measurements were done on <sup>68</sup>Zn(p,2n)<sup>67</sup>Ga nuclear reaction in collaboration with VUB Brussel. The obtained new result were included in the compiled data base. The new results were sent for publication to the Journal of Radioanalytical Chemistry. The new results were also included into the database of present CRP compilation.
- New measurements were done in Jülich-Debrecen collaboration on the <sup>18</sup>O(p,n)<sup>18</sup>F reaction, but the experiments at low energies are still not completed.
- New measurements were done in Brussel-Debrecen collaboration on  $^{nat}Ti(p,x)^{48}V$  monitor reaction. The results of the new measurements are prepared for publication.
- New series of measurements were performed on  $^{nat}Cu(\alpha,x)$  reactions up to 40 MeV in Brussel-Debrecen collaboration. The new results were included into the data base of
present CRP compilation. The publication of the new results in the Nuclear Instruments and Methods is in progress.

- To complete the not satisfactory experimental data base of deuteron induced reactions new series of measurements were performed on iron, copper, aluminum, titan and nickel up to 50 MeV deuterons in Brussel-Debrecen collaboration. The evaluation of the experimental data is in progress.
- In spite of the very intensive experimental work, there was no time to perform important experiments for validation of the recommended data for intercomparison of the monitor reactions.

### **Compilation in EXFOR format**

The list of all nuclear reactions related to CRP and still not compiled into accepted international data base EXFOR format were prepared. About **150 work** used in the CRP was collected for further compilation in EXFOR. The list were transferred to Data Centers for additional checking. Part of the references were already compiled by the Debrecen Charged Particle Data Group. The remaining publications were redistributed between CP Data Centers according to Data Center agreement.

## CRP TECDOC

The preparation of the related chapters of the TECDOC is in progress. During the preparation it was concluded that to avoid duplications and to make more practical the use of the TECDOC for every day users, small modification of the outline of the TECDOC seems to be necessary. According to the evaluation process and the methods have to be discussed in the first part only briefly and the experimental data, the results of the theoretical calculations, recommended cross section and yield data have to be presented for each investigated reaction together. Taking into account the minimal number of required figures and tables and the number of the investigated reactions, the TECDOC can reach 500 pages.

#### **Recommendations**

The compilation and evaluation process formulate some modification on the list of the reactions. The status of several monitor reactions is not satisfactory for monitoring. From other side several monitor reactions earlier not considered because of the sensitivity to the neutron background have to be included.

• It is necessary to extend the list of the investigated reactions:

Important monitor reactions:  ${}^{nat}Al(p,x)^{24}Na$  ${}^{nat}Al(d,x)^{24}Na$   $^{nat}Al(^{3}He,x)^{24}Na$  $^{nat}Al(a,x)^{24}Na$  $^{nat}Fe(p,x)^{56}Co$ 

Gamma-emitters:  $^{124}$ Xe(p,x) $^{123}$ I

Positron -emitters: <sup>11</sup>B(p,n)<sup>11</sup>C

• the data are important according to survey of isotope production cyclotrons, therefore it is recommended to publish the results and the guidance to users of the data not only in form of TECDOC but also in a referred Journal or a book. It requires detailed and thorough work. Most of the paper work could be done only after clearing disagreements, accepting final data.

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Journal of Radioanalytical and Nuclear Chemistry(1998)(in Print)

#### RECOMMENDATION OF CHARGED PARTICLE CROSS SECTION FOR MEDICAL RADIOISOTOPE PRODUCTION

Youxiang Zhuang (China Institute of Atomic Energy, Beijing 102413)

The combined method of experimental evaluation with theoretical calculation and systematics research is the best one to get the recommended values.

The recommended values in my evaluation are taken from

1. Fitting values of evaluated experimental data, if there are enough and accurate experimental data; and it is not necessary to do careful theoretical calculation.



 Combined values of fitting values of experimental data with supplementary theoretical values, if evaluated experimental data are deficient.











## Calculation of the production cross sections of Zn and Ga radioisotopes through charged particle bombardment of Cu and Zn isotopes in 3-26 MeV energy range

## K. Gul

## Pakistan Institute of Nuclear Science and Technology P.O. Nilore, Islamabad, Pakistan

Calculations for the excitation functions of reactions used for the production of  ${}^{62}$ Zn,  ${}^{63}$ Zn,  ${}^{65}$ Zn,  ${}^{65}$ Ga,  ${}^{66}$ Ga,  ${}^{67}$ Ga and  ${}^{68}$ Ga radioisotopes through the bombardment of  ${}^{63}$ Cu,  ${}^{65}$ Cu with protons, helions and  $\alpha$ -particles and of  ${}^{66}$ Zn,  ${}^{67}$ Zn and  ${}^{68}$ Zn with protons in 3-26 MeV energy range have been carried out using statistical and pre-equilibrium nuclear reaction models. The calculations have been compared with reported measurements and discussed.

#### I. Introduction

Radioisotopes produced through charged particle beams find wide applications in the field of medicine [1-4]. Therefore, the knowledge of precise cross sections of various reactions used for energy and flux determination of charged particle beams and production of radioisotopes of high purity is desirable [5]. Some of the radioisotopes of interest include <sup>62</sup>Zn, <sup>63</sup>Zn, <sup>65</sup>Zn, <sup>65</sup>Ga, <sup>66</sup>Ga, <sup>67</sup>Ga and <sup>68</sup>Ga. Reactions induced in Cu by

protons and  $\alpha$ -particles are utilized for energy and flux monitoring of these particles used for production of different types of medical radioisotopes. The reactions of protons with the various isotopes of Zn are used for the production of Ga radioisotopes for medical applications. Several measurements of the excitation functions of these reactions have been reported but the data are highly discrepant. The evaluation based on the discrepant data will contain large uncertainties. Therefore an evaluation based on the standard nuclear reaction models using consistent sets of parameters is highly desirable for identification of discrepant data. The present paper gives a description of the calculated excitation functions of ten reactions induced in the two isotopes of Cu by protons, helions and  $\alpha$ -particles and seven reactions induced by protons in three isotopes of Zn. The details of these calculations and numerical data are given in Refs. [6-12]. The calculations are compared with reported measurements and discussed.

## **II.** Model Calculations

Calculations for the excitation functions were carried out using HFMOD code [13] for the statistical model and PREMOD code [14] for the pre-equilibrium model calculations using the concept of sequential emission. In general Perey potentials [15] were used for protons and Wilmore-Hodgson potentials [16] were used for neutrons. The potentials of  $\alpha$ -particles were taken from Ref. [17]. Deuteron potentials were taken from Ref. [18]. The potentials of helions were taken from Ref. [19]. Weisskopf single-particle model was used for the calculation of the emission probabilities of photons. Energy level densities were calculated on the basis of the formalism of Dilg *et al.* [20] assuming a rigid body moment of inertia for nuclei. Same values of energy level density parameters were used for the common intermediate and residual nuclei for the sake of consistency of the calculations.

## III. Results and discussion

## A. ${}^{63}Cu(p,n){}^{63}Zn, {}^{63}Cu(p,2n){}^{62}Zn \text{ and } {}^{65}Cu(p,n){}^{65}Zn \text{ reactions}$

The calculation for the excitation function of  ${}^{63}Cu(p,n){}^{63}Zn$  reaction is compared with reported measurements [21-33] and the evaluation of Vukolov and Chukreev [34] in Fig.1. There is a very good agreement between the calculation and measurements up to 10 MeV except for the cross sections at 6.3 MeV reported by Blaser et al. [26], at 9.3 MeV reported by Chackett et al. [31] and at 9.85 MeV reported by Meyer and Hintz [33] which are relatively high. Beyond 10 MeV the data are highly discrepant. Between 14 and 24 MeV the calculation is in good agreement with the measurements of Colle et al. [22] and Grutter [24]. In this energy region the data of Ghoshal [27] and Meadows [28] and Aleksandrov et al. [32] are high. In 10-14 MeV energy region the calculation shows slightly higher trend as compared with the data of Colle et al. [22]. The calculation for the excitation function of <sup>63</sup>Cu(p,2n)<sup>62</sup>Zn reaction is shown and compared with reported measurements [24, 27-28, 32, 35-38] in Fig.2. The data of Ghoshal [27] rises rapidly beyond 18 MeV. The calculation is in good agreement with the data of Kopecky [35] and Grutter [24] in 19-24 MeV region. The calculation gives slightly higher cross section values as compared with measurements in 15-19 MeV range. The calculation for <sup>65</sup>Cu(p,n)<sup>65</sup>Zn reaction is shown and compared with reported measurements [22-26, 29, 31, 35, 36, 39-43] in Fig.3. All the data seem in reasonably good agreement with the calculation up to 11 MeV except the data of Howe [29] which show a higher trend. The data of Kopecky [35], Colle et al. [22] and Grutter [24] are in good agreement with the calculation. The data of Kormali et al. [42] show slightly higher trend implying a shift of energy by about 1 MeV towards the higher energy side. The data of Williams and Fulmer [40] and Gadioli et al. [41] have higher cross section values probably due to a shift of the data towards higher energy side.

## B. ${}^{65}Cu({}^{3}He,n){}^{67}Ga and {}^{65}Cu({}^{3}He,2n){}^{66}Ga reactions$

The calculation for the excitation function of  ${}^{65}Cu({}^{3}He,n){}^{67}Ga$  reaction is shown in Fig.4 and compared with one measurement [44] due to the availability of the numerical data only in this case. As can be seen the calculation is high by about a factor of two compared with the data of Bryant *et al.* [44]. The calculation for the excitation function of  ${}^{65}Cu({}^{3}He,2n){}^{66}Ga$  reaction is shown and compared with reported measurements [44-47] in Fig.5. The calculation is high by about a factor of two compared with the data of Bryant *et al.* [44]. The data of Gulchert *et al.* [46] and Lebowitz *et al.* [47] are very low. The data of Bissem *et al.* [45] have the indication that cross section of the reaction could brobaly be higher than previously reported values. A new measurement for the excitation function of  ${}^{65}Cu({}^{3}He,2n){}^{66}Ga$  reaction seems desirable.

# C. ${}^{63}Cu(\alpha,n){}^{66}Ga, {}^{63}Cu(\alpha,2n){}^{65}Ga, {}^{63}Cu(\alpha,np){}^{65}Zn, {}^{65}Cu(\alpha,n){}^{68}Ga and {}^{65}Cu(\alpha,2n){}^{67}Ga reactions$

The calculation for the excitation function of  ${}^{63}Cu(\alpha,n){}^{66}Ga$  reaction is compared with reported measurements [21, 44, 48-51] in Fig.6. The calculation agrees very well with the data of Stelson and McGowan [51] which have precise energy values of incident  $\alpha$ -particles. The agreement of the calculation with the data of Bryant *et al.* [44] and Zweit *et al.* [48] is better at higher energies than at lower energies which may be due to a large energy uncertainty inherent in the stacked-foil technique for the lower energy data. The data of Porile and Morrison [50] and Rizvi *et al.* [49] show a poor agreement with the calculation both being very low in magnitude. The data of Porile and Morrison [50] are not only very low but the position of their maximum cross section is also shifted towards the higher energy side by about 2 MeV. The calculations for the excitation functions of  ${}^{63}Cu(\alpha,2n){}^{65}Ga$  and  ${}^{63}Cu(\alpha,np){}^{65}Zn$  reactions and the accumulated yield of  ${}^{65}Zn$  [48-50, 52-54] in Fig.7. The experimental data are highly discrepant. The calculation agrees partly with the data of Rizvi *et al.* [49], Zweit *et al.* [48] Porile and Morrison [50] and Porges [52]. The data of Lin and Alexander [54] are systematically higher compared with the calculation.

The calculation for  ${}^{65}$ Cu( $\alpha$ ,n) ${}^{68}$ Ga reaction is shown and compared with reported measurements [21, 44, 49-52] in Fig.8. The calculation agrees very well with the data of Bryant *et al.* [44] and Hille *et al.* [21] upto 22 MeV and show slightly higher trend beyond 22 MeV. The peak of the data of Porile and Morrison [50] seems to be shifted by about 3 MeV towards the high energy direction and shifting their data downward by 3 MeV improves the agreement of their data with the calculation. The data of Porges [52] and Rizvi *et al.* [49] are very low. The calculation for the excitation function of  ${}^{65}$ Cu( $\alpha$ ,2n) ${}^{67}$ Ga reaction is compared with the reported measurements [44, 49, 50, 52, 55, 56] in Fig.9. The data of Watson *et al.* [56] at 26.3 MeV which agree within 9% with the calculation have reported standard deviation less than ±3% and should therefore provide a standard for assessing the discrepancy of the data in this region. As in the case of  ${}^{65}$ Cu( $\alpha$ ,n) ${}^{68}$ Ga reaction, the data of Porile and Morrison [50] agree better if they are shifted downward in energy by 3 MeV. The data of Porges [52] are on the lower side while the data of Bryant *et al.* [44] are on the higher side.

# D. ${}^{66}Zn(p,n){}^{66}Ga, {}^{66}Zn(p,2n){}^{65}Ga, {}^{66}Zn(p,np){}^{65}Zn, {}^{67}Zn(p,n){}^{67}Ga, {}^{67}Zn(p,2n){}^{66}Ga, {}^{68}Zn(p,n){}^{68}Ga and {}^{68}Zn(p,2n){}^{67}Ga reactions$

A compilation of the measurements done before 1993 has been reported by Szelecsenyi *et al.* [57]. Experimental data for comparison with calculations have been taken from this compilation where these data were reported in graphical form only. The calculation for the excitation function of  ${}^{66}Zn(p,n){}^{66}Ga$  reaction has been compared with reported measurements [21, 29, 58-64] in Fig.10. Recent data of Hermanne taken from Ref. [64] have also been included for comparison. The measurements of Howe [29], Hille *et al.* [21], Tarkanyi *et al.* [58] and Szelecsenyi *et al.* [59] agree very well within the

errors with the calculation keeping in view the large energy uncertainty inherent in the stacked-foil technique for the lower energy protons. The data of Kopecky [60] and Barrandon *et al.* [61] have low magnitude of cross sections. The data of Hermanne *et al.* [62] and the data of Hermanne taken from Ref. [64] have lower cross section values below 15 MeV. The peak of the excitation function of the reaction reported by Little and Lagunas-Solar [63] appears to be shifted by 1-2 MeV towards the higher energy side and has a low cross section value. The calculation beyond 19 MeV overestimates the measurements which are in better agreement with each other. The calculations for the excitation functions of <sup>66</sup>Zn(p,np)<sup>65</sup>Zn as well as <sup>66</sup>Zn(p,2n)<sup>65</sup>Ga reactions and their sum have been shown in Fig.11. There are no separate measurements for (p,np) and (p,2n) reactions. However the measurements of the accumulated yields of these reactions i.e. <sup>66</sup>Zn(p,x)<sup>65</sup>Zn reaction have been reported by Tarkanyi *et al.* [58] and Szelecsenyi *et al.* [59] which are in good agreement with the calculation.

The calculation for the excitation function of  ${}^{67}Zn(p,n){}^{67}Ga$  reaction is compared with the reported measurements [26, 39, 58-61, 63-64] in Fig.12. The calculation has a good agreement within the errors with the data of Tarkanyi *et al.* [58], Szelecsenyi *et al.* [59], Kopecky [60], the data of Hermanne taken from Ref. [64] and the data of Blaser *et al.* [26]. The peak of the data of Little and Lagunas-Solar [63] shows energy shift towards the higher energy side also for this reaction. The data of Barrandon *et al.* [61] agree with the calculation upto the peak value but drop down very quickly beyond the peak of the excitation function. The calculation for the excitation function of  ${}^{67}Zn(p,2n)^{66}$ Ga reaction is compared with reported measurements [58, 59, 63, 64] in Fig.13. The data of Tarkanyi *et al.* [58] are in good agreement with the calculation. However, the data of Szelecsenyi *et al.* [59] agree with the calculation upto 18 MeV beyond which measurements are low in magnitude. The data of Little and Lagunas-Solar [63] is very low and as for their other data there is also a shift of energy towards the higher energy side. The data of Hermanne taken from Ref. [64] have lower magnitude.

The calculation for  ${}^{68}Zn(p,n){}^{68}Ga$  reaction is compared with reported measurements [21, 26, 29, 39, 58, 61, 62, 64-67] in Fig.14. Only a limited number of data of Johnson et al. [39] and Esat et al. [65] have been included in the figure for the sake of clarily. All data are in good agreement within the errors with the calculation except the data of Hermanne et al. [62] and the data of Hermanne taken from Ref. [64] which seem shifted by 2 MeV to the lower energy side and the 10 MeV cross section of McGee et al. [67] which is very low. The shifting of the data of Hermanne et al. [62] by 2 MeV in the increasing direction brings their data in good agreement with the calculation. The calculation for  ${}^{68}Zn(p,2n){}^{67}Ga$  reaction is compared with reported measurements [58, 59, 62-64, 67, 68] in Fig.15. The calculation is in general in very good agreement with the data of Tarkanyi et al. [58] and Hermanne et al. [68]. The data of Szelesenyi et al. [59] have low cross section values. The data of Little and Lagunas-Solar [63] are low in magnitude and have energy calibration problem. The data of McGee et al. [67] are very low. The earlier data of Hermanne et al. [62] appear shifted by 2 MeV to the lower side and shifting their data by 2 MeV in the higher energy direction brings the data in good agreement with the calculation.

Calculation for the excitation function of  $^{nat}Zn(p,xn)^{66}Ga$  reaction is compared with reported measurements [21, 29, 58, 64, 66] in Fig.16. Above 14 MeV it is almost in good agreement with the data of Szelecsenyi *et al.* [64] and Nortier *et al.* [66]. Below 14 MeV however the data of Szelecsenyi *et al.* [64] and Nortier *et al.* [66] are low. The data of Howe [29] Hille *et al.* [21] and Tarkanyi *et al.* [58] are in good agreement with the calculation. The calculation for the excitation function of  $^{nat}Zn(p,xn)^{67}Ga$  reaction is compared with reported measurements [64, 66] in Fig.17. The calculation is in good agreement with reported measurement of Hermanne taken from Ref. [64] and data of Nortier *et al.* [66].

## Summary

The nuclear reaction model calculations resulted in validation of some of the experimental data relevant to medical radioisotopes production. Some of the measurements were found shifted in energy and could be brought in better agreement with the validated data if shifted appropriately in energy. Some highly discordant data were also identified.

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## **Figure Captions**

- Fig.1 Comparison of the calculation with measurements of the excitation function of <sup>63</sup>Cu(p,n)<sup>63</sup>Zn reaction.
- Fig.2 Comparison of the calculation with measurements of the excitation function of  ${}^{63}Cu(p,2n){}^{62}Zn$  reaction.
- Fig.3 Comparison of the calculation with measurements of the excitation function of <sup>65</sup>Cu(p,n)<sup>65</sup>Zn reaction.
- Fig.4 Comparison of the calculation with measurements of the excitation function of <sup>65</sup>Cu(<sup>3</sup>He,n)<sup>67</sup>Ga reaction.
- Fig.5 Comparison of the calculation with measurements of the excitation function of <sup>65</sup>Cu(<sup>3</sup>He,2n)<sup>66</sup>Ga reaction.
- Fig.6 Comparison of the calculation with measurements of the excitation function of <sup>63</sup>Cu(<sup>4</sup>He,n)<sup>66</sup>Ga reaction.
- Fig.7 Comparison of the calculation with measurements of the excitation function of  ${}^{63}Cu({}^{4}He,x){}^{65}Zn$  reaction.
- Fig.8 Comparison of the calculation with measurements of the excitation function of <sup>65</sup>Cu(<sup>4</sup>He,n)<sup>68</sup>Ga reaction.
- Fig.9 Comparison of the calculation with measurements of the excitation function of <sup>65</sup>Cu(<sup>4</sup>He,2n)<sup>67</sup>Ga reaction.

- Fig.10 Comparison of the calculation with measurements of the excitation function of <sup>66</sup>Zn(p,n)<sup>66</sup>Ga reaction.
- Fig.11 Comparison of the calculation with measurements of the excitation function of  ${}^{66}Zn(p,x){}^{65}Zn$  reaction.
- Fig.12 Comparison of the calculation with measurements of the excitation function of <sup>67</sup>Zn(p,n)<sup>67</sup>Ga reaction.
- Fig.13 Comparison of the calculation with measurements of the excitation function of <sup>67</sup>Zn(p,2n)<sup>66</sup>Ga reaction.
- Fig.14 Comparison of the calculation with measurements of the excitation function of <sup>68</sup>Zn(p,n)<sup>68</sup>Ga reaction.
- Fig.15 Comparison of the calculation with measurements of the excitation function of  ${}^{68}$ Zn(p,2n) ${}^{67}$ Ga reaction.
- Fig.16 Comparison of the calculation with measurements of the excitation function of  $^{nat}Zn(p,xn)^{66}Ga$  reaction.
- Fig.17 Comparison of the calculation with measurements of the excitation function of <sup>nat</sup>Zn(p,xn)<sup>67</sup>Ga reaction.



Fig.1















Fig.8





Fig.10









Proton Energy(MeV)




Fig.16



Nuclear Data	a Section		e-mail: services@iaeand.iaea.or.at
International Atomic Energy Agency P.O. Box 100 A-1400 Vienna		Energy Agency	fax: (43-1)26007 cable: INATOM VIENNA telex: 1-12645 atom a
Austria			telephone: (43-1)2600-21710
or	nline:	TELNET or FTP: iaeand.iaea.or.at	
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