



**Measurement of Cross Sections of Proton Induced Nuclear
Reactions on Ti, Ni, Zn, Cd, and Au up to 30 MeV and
Their Application in Radioisotope Production**

PhD. thesis

by

Ferenc Szelecsényi dr. univ.

**Lajos Kossuth University
Debrecen, Hungary
1997**

Ezen értekezés a KLTE "**Fizika**" doktori program "**Magfizika**" (II) alprogramja keretében készítettem **1995-1997** között és ezúton benyújtom a KLTE doktori PhD fokozatának elnyerése céljából.

Debrecen, 1997 augusztus 22.

a jelölt aláírása

Tanúsítom, hogy **Dr. Szelecsényi Ferenc** doktorjelölt **1995-1997** között a fent megnevezett doktori alprogram keretében irányításommal végezte munkáját. Az értekezésben foglaltak a jelölt önálló munkáján alapulnak, az eredményekhez önálló alkotó tevékenységével meghatározóan hozzájárult. Az értekezés elfogadását javaslom.

Debrecen, 1997 augusztus 22

a témavezető aláírása

Contents

1. Introduction	5
1.1. Importance of cross section data for practical applications.....	5
1.2. Production of radioisotopes with cyclotron.....	6
2. Background of the work	7
2.1. Background of the work, motivations.....	7
2.2. The aim of this study.....	8
2.3. List of the original publications.....	10
3. Basic equations, model codes	15
3.1. Mechanism of the nuclear reaction.....	15
3.2. Cross sections of a nuclear reaction.....	15
3.3. Target yields.....	17
3.4. Model codes for calculation of excitation functions.....	17
4. Experimental techniques	19
4.1. Stacked-foil technique	19
4.1.1. Principle of the method.....	19
4.1.2. Application of monitor reactions in 'stacked-foils' type cross section measurements.....	19
4.2. Targets.....	20
4.2.1. Targets for cross section measurements.....	20
4.2.2. Targets for routine radioisotope production.....	21
4.3. Accelerators.....	22
4.4. Irradiations circumstances and measurements of activity.....	22
4.5. Calculation of the cross sections, energy degradation and their errors.....	25
4.6. Yield measurements and routine productions.....	26
5. Results and conclusions	27
5.1. Ti+p processes.....	27
5.1.1. Study of the Ti+p reactions.....	27
5.1.2. Application of the results.....	28
5.2. Ni+p processes.....	28
5.2.1. Study of the Ni+p reactions.....	28
5.2.2. Application of the results.....	30
5.3. Zn+p processes.....	31

5.3.1. <i>Study of the Zn+p reactions</i>	31
5.3.2. <i>Application of the results</i>	33
5.4. <i>Cd+p processes</i>	34
5.4.1. <i>Study of the Cd+p reactions</i>	35
5.4.2. <i>Application of the results</i>	36
5.5. <i>Au+p processes</i>	36
5.5.1. <i>Study of the Au+p reactions</i>	36
5.5.2. <i>Application of the results</i>	37
6. Summary	38
7. Összefoglalás	41
8. Acknowledgements	46
9. References	47
10. Appendix: photocopies of the publications related to the thesis	53

1. Introduction

1.1 Importance of cross section data for practical applications

Cross section data for charged particle induced nuclear reactions, especially proton induced ones, are required in a number of practical applications (medical radioisotope production, nuclear wear measurement, radiobiology etc.). The excitation functions which represent variations of cross sections for particular reactions with incident energy, can be used in the field of radioisotope production to

- a) determine the particle energies and optimal energy ranges required for a particular reaction type.
- b) calculate the radioisotope production yield which can be expected for a particular reaction and a given target matrix.
- c) calculate, especially for medical radioisotope production, the production yields for radionuclidic impurities, which help to determine the need for isotopically enriched target materials.

In spite of the increasing need for well measured cross sections and the sometimes rich experimental information the status of the cross section data was not satisfactory at the end of the last decade. The reported cross sections from different groups often showed unacceptable deviations both in the values of the cross sections and in their energy scale. Unfortunately, these problems existed even in the case of those reactions which were used for production of the most frequently employed medical radioisotopes. Additionally, for some reactions important for other than radioisotope production (i.e. nuclear wear measurement, monitor reactions etc.) the cross section data were measured with relative high errors or they were missing in some energy regions of interest.

From practical point of view the 'accurate' knowledge of the amount of the product nuclei in a given target material is 'vital' for the majority of practical applications. However, for any (medical) radioisotope the optimum conditions for production will depend on many factors. A large quantity of data would be required to determine production yields and contaminants of all competing reactions. Only if these data are available can a 'proper' assessment be made of the advantages and limitations of the various production methods. The situation is even more complex if indirect production regimes are assumed to take advantage of half-life discrimination of contaminants in the decay of the activation products.

Quite often only production yield data are available in the literature. While such data are important for production purposes, a better understanding of the relationship between the various factors contributing to product and contaminant yield(s) is obtained from excitation function data. In particular, it is easier to assess the consequences of a variation in irradiation conditions, which for various reasons might be difficult to be made identical at different accelerators.

Indeed, excitation functions are just the beginning. There remain many more factors that are important in production of the required radionuclide. These include suitable chosen 'targetry' (i.e. target composition and material, target construction and transfer of dissipated heat) and radiochemical procedures (i.e. remote target handling, chemical processing, radiochemical and radiopharmaceutical quality assurance etc.)

There is an additional but not really practical application of well measured cross section values. A number of nuclear model codes have been developed first to gain theoretical understanding of nuclear reaction processes involved. Later these codes have also been employed by several authors to calculate excitation functions for charged particle induced reactions of medical interest. However, the accuracy with which excitation functions can be

reproduced using these codes depends on several factors. The measure for the accuracy of calculated excitation function is the experimental data. Since recently several evaluated experimental cross section data bases are available for different charged particle induced reactions, there is a possibility to improve the predicting capabilities of the different codes.

It is not suggested, however, that one should rely on calculated excitation functions, but in the planning of new production or analytical methods, the calculated activation cross section data could be ‘helpful’ if the experimental data are missing or not satisfactory (Nowotny and Uhl, 1987).

Due to the increased need recently arisen for well measured charged particle cross section data several nuclear data centers all around the world have started the collection and evaluation of these data. Additionally, several laboratories are also involved in measurements (and sometimes in re-measurement) of charged particle reactions relevant for practical purposes. The Nuclear Data Section of the International Atomic Energy Agency, Vienna, Austria has been coordinating these efforts since many years and makes available the collected data through the EXFOR data base of the Agency (*cf* Schwerer and Lemmel, 1996). Our laboratory in Debrecen, Hungary is also participating both at the experimental and the evaluation works. We perform cross section (re)measurements for: **a**) reactions used for monitoring p, d, ³He and alpha particle beams, **b**) reactions used for medical radioisotope production (*cf* Oblozinský, 1997).

1.2 Production of radioisotopes with cyclotron

In those cases when radioisotopes can be produced for practical purposes only by charged particle induced nuclear reactions, the cyclotrons are the most commonly used accelerators. (Especially for routine or commercial production and for some type of nuclear wear measurement.) It is also important to note that due to their available energy ranges and the easy access to cyclotron beams (i.e. several nuclear research centers have cyclotron(s)), the majority of the cross section measurements were (and are) also done with the help of these accelerators.

Cyclotrons are technically less complicated than other accelerators and the particle beam currents obtainable are high. On the other hand, they are also cost efficient. The majority of the cyclotrons involved in radionuclide business and industrial applications are ‘proton only’ accelerators. Cyclotrons used for radionuclide production are usually classified in for levels, I-IV, (using the classification of Wolf and Jones (1983)) depending on their maximum proton beam energy. (They are also called low, medium or high energy machines.) The big level III ($E_{p,max}=45$ MeV) and IV ($E_{p,max}=200$ MeV) cyclotrons can be found in larger research centers or industrial laboratories. However, for the production of the most frequently used PET isotopes only, level I ($E_{p,max}=10$ MeV) and II ($E_{p,max}=20$ MeV) cyclotrons are suitable. Level I cyclotrons, having a rather low beam energy often require the use of isotopically enriched target materials in order to produce sufficient amounts of the desired radionuclides. When using level II cyclotrons enriched target materials are often used as well, mostly in order to avoid unwanted side reactions, resulting in radionuclidic contamination of the product.

2. Background of the work

2.1 Motivations, background of the work

The multiparticle MGC-20E cyclotron of the Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), Debrecen, Hungary was put into operation in 1985. Since it is the only cyclotron in Hungary, one of its duties was and still is to provide radioisotopes for Hungarian medical centers having nuclear medicine department. According to the needs of the nuclear medicine community we had to elaborate the routine production of some frequently used SPECT (Single Photon Emission Computer Tomography) radioisotopes (^{67}Ga , ^{111}In and ^{123}I) at the beginning. Some years later when a PET center (Positron Emission Tomography) was established on the premises of our Institute, the production of some PET radioisotopes and chemical compounds labeled with these radionuclides become more important (^{11}C , ^{13}N , ^{15}O , ^{18}F , ^{61}Cu , ^{64}Cu etc.).

One of my duties at the Cyclotron Department of ATOMKI during the last decade was to investigate the production possibility of $^{61,64}\text{Cu}$, $^{66,67,68}\text{Ga}$, and ^{111}In radioisotopes. This work included not only the design and development of beam-lines used for routine radionuclide production, but the elaboration of production routes of the different radionuclides as well. I also took part in the routine production work of the above isotopes. Some results of these works were already published in my thesis for the university doctor's degree (Szelecsényi 1988).

When we started our work in the field of routine radioisotope production we decided to analyze the literature results concerning radionuclide production to gain knowledge for production optimization. Taking into account the available maximum charged particle energies of the MGC-20E cyclotron (this is a level II accelerator; *cf* Mahunka *et al.* 1988) we could select only (p,n) and (p,2n) reactions on highly enriched (>90%) target materials for practical production purposes.

Surveying the information on excitation functions and yields for the possible reactions, we found several measurements. However, we could conclude that in some cases:

- a) the published cross sections were associated with relative high errors and/or the excitation functions were measured in less detailed for production purposes.
- b) the reported cross sections from different groups often showed unacceptable deviations both in the values of the cross sections and in their energy scales.
- c) cross section data for the reactions resulting contaminant radionuclides not only had the above problems but they were also missing in some energy regions.
- d) the reported calculated and/or measured integral thick target yields showed significant differences.

Due to the above problems we could not employ the published results on cross sections and yields for our purposes. In order to have reliable nuclear data for our radioisotope production program we decided to evaluate the available data bases and to conduct some new detailed measurements if it was necessary. We also noticed during our search for cross section data that there were also problems with these cross sections and/or thick target yield values at higher energies. Since the available maximum proton energy of the majority of the cyclotrons used for routine or commercial radioisotope production are usually below 30 MeV, we also planned to extend our investigations up this energy. We thought that this way the results of our investigations will be useful not only for our radioisotope program, but other laboratories also could use these evaluations and the new data for improving their efforts in the field of routine production.

It is well known that the material of the target backing is very important in radioisotope production from the point of view of the chemistry used. Usually, electroplating methods have been developed for the routine targets. Due to the chemistry used in our laboratory the enriched targets (both for production and cross section measurements) were electroplated onto nickel and gold. (In the case of cross section measurements the backings were thin Au and Ni metal foils.) According to our experiences proton bombardment of Au and Ni produces several well measurable nuclear processes. Since monitor reactions were planned to use in all of our cross section measurements we also decided to study the possibility of using of some of the reactions induced in these target backings for monitor purposes. (The use of a monitor reaction makes possible to measure the bombarding beam flux in those cases when the use a Faraday-cup is difficult or even not possible.) We also wanted to investigate these possibilities because the number of the well measured proton monitor reactions below 30 MeV were also limited at the beginning of our investigations (*cf* Schwerer and Okamoto, 1989). Additionally, to increase the number of the available monitor reactions we decided to measure the $^{nat}\text{Ti}+p$ reactions, too.

2.2 The aim of the study

In the light of the above mentioned problems and duties **the aim of this study** could be summarized as follows:

- A) Cross section measurements on highly enriched Zn, Ni and Cd for investigating the proton production routes of the medically important ^{61}Cu , ^{64}Cu , ^{66}Ga , ^{67}Ga , ^{68}Ga , and, ^{111}In radionuclides up to 30 MeV.
- B) Cross section measurements on enriched Zn, Ni and Cd for investigating the proton production routes for reactions resulting contaminant radionuclides.
- C) Investigation and optimization the routine production way of the $^{66,67,68}\text{Ga}$ and ^{111}In with small and medium sized (up to 30 MeV) cyclotrons.
- D) Investigation of $^{nat}\text{Ti}+p$, $^{nat}\text{Ni}+p$, and $^{nat}\text{Au}+p$ processes from the point of view of their potential use for monitoring proton beam performance.
- E) Compilation and evaluation of the available cross sections of the above reactions to resolve the discrepancies found among the literature values. Creating ‘recommended’ cross sections and/or thick target yield data bases of Zn+p, Cd+p and Au+p processes for practical applications up to 30 MeV. Perform model calculations in the case of some Ni+p and Zn+p reactions to check the prediction capability of some computer codes.

Below 18 MeV the irradiations were planned to do in Debrecen, Hungary but for the measurements with higher energies (for the (p,2n), (p,pn) and (p,3n) processes) we had to find medium sized cyclotrons of other laboratories. The list of the collaborating partners and the types of their accelerators can be seen in Table 4.3.1. The reactions which were selected on the basis of the above list are collected in Table 2.1 together with the investigated energy regions and the potential use of the reactions or the reaction product(s). These reactions are ordered by increasing atomic number of the product nuclei.

Table 2.1. List of the investigated nuclear reactions

Target	Reaction	Energy (MeV)	Product nuclei	Importance of the investigated reaction or the product nuclei
<i>Ti+p reactions</i>				
^{nat}Ti	$^{nat}\text{Ti}(p,x)$	$E_p \leq 30$	^{43}Sc	side reaction
^{nat}Ti	$^{nat}\text{Ti}(p,x)$	$E_p \leq 18$	^{44}Sc	side reaction

^{nat}Ti	$^{nat}\text{Ti}(p,x)$	$E_p \leq 18$	^{47}Sc	side reaction
^{nat}Ti	$^{nat}\text{Ti}(p,x)$	$E_p \leq 18$	^{48}V	possible monitor reaction
Ni+p reactions				
^{nat}Ni	$^{nat}\text{Ni}(p,x)$	$E_p \leq 30$	^{55}Co	PET radioisotope used mainly in labeling bleomycin and cerebral flow studies (<i>cf</i> Reiner <i>et al.</i> , 1997), possible monitor reaction
^{nat}Ni	$^{nat}\text{Ni}(p,x)$	$E_p \leq 30$	^{56}Co	possible monitor reaction
^{nat}Ni	$^{nat}\text{Ni}(p,x)$	$E_p \leq 30$	^{57}Co	possible monitor reaction
$^{61}\text{Ni}(89\%)$	$^{61}\text{Ni}(p,\alpha)$	$E_p \leq 18.7$	^{58}Co	side reaction
$^{64}\text{Ni}(95\%)$	$^{64}\text{Ni}(p,\alpha)$	$E_p \leq 18.7$	^{61}Co	side reaction
^{nat}Ni	$^{nat}\text{Ni}(p,x)$	$E_p \leq 44$	^{57}Ni	possible monitor reaction
$^{61}\text{Ni}(89\%)$	$^{61}\text{Ni}(p,n)$	$E_p \leq 18.7$	^{61}Cu	PET radioisotope, especially in chelate form it is used for blood flow studies in heart and brain (<i>cf</i> Green <i>et al.</i> , 1990)
$^{64}\text{Ni}(95\%)$	$^{64}\text{Ni}(p,n)$	$E_p \leq 18.7$	^{64}Cu	PET radioisotope, (see its application at ^{61}Cu)
Zn+p reactions				
$^{66}\text{Zn}(99\%)$	$^{66}\text{Zn}(p,2n)^{65}\text{G}$ a+ $^{66}\text{Zn}(p,pn)^{65}\text{Z}$ n	$E_p \leq 26.5$	^{65}Zn	side reaction, also a candidate for use in nuclear wear measurements
$^{66}\text{Zn}(99\%)$ $^{67}\text{Zn}(92\%)$ $^{68}\text{Zn}(99\%)$	$^{66}\text{Zn}(p,n)$ $^{67}\text{Zn}(p,2n)$ $^{68}\text{Zn}(p,3n)$	$E_p \leq 26.5$ $E_p \leq 26.5$ $E_p \leq 26.5$	^{66}Ga	PET radioisotope used for studying some slow dynamic processes such as lymphatic transport (<i>cf</i> Goethals <i>et al.</i> , 1988); also a candidate for use in nuclear wear measurements
$^{67}\text{Zn}(92\%)$ $^{68}\text{Zn}(99\%)$	$^{67}\text{Zn}(p,n)$ $^{68}\text{Zn}(p,2n)$	$E_p \leq 26.5$ $E_p \leq 26.5$	^{67}Ga	SPECT radioisotope, used for detecting the presence of malignancy and for diagnosis of inflammatory diseases (<i>cf</i> Noujaim, 1981); also a candidate for use in nuclear wear measurements
$^{67}\text{Zn}(98\%)$	$^{68}\text{Zn}(p,3n)$	$E_p \leq 26.5$	^{68}Ga	PET radioisotope, used for blood-brain barrier investigation (<i>cf</i> Qaim, 1987)
Cd+p reactions				
$^{111}\text{Cd}(96\%)$	$^{111}\text{Cd}(p,3n)$	$E_p \leq 30$	^{109}In	side reaction
$^{111}\text{Cd}(96\%)$ $^{112}\text{Cd}(98\%)$	$^{111}\text{Cd}(p,2n)$ $^{112}\text{Cd}(p,3n)$	$E_p \leq 30$ $E_p \leq 30$	^{110}In	side reaction
$^{111}\text{Cd}(96\%)$ $^{112}\text{Cd}(98\%)$	$^{111}\text{Cd}(p,n)$ $^{112}\text{Cd}(p,2n)$	$E_p \leq 30$ $E_p \leq 30$	^{111}In	SPECT radioisotope, used mainly for labeling of cellular blood components and monoclonal antibodies (<i>cf</i> Kirchner <i>et al.</i> , 1983)
Au+p reactions				

^{197}Au	$^{197}\text{Au}(p,pn)$	$E_p \leq 18$	^{196}Au	possible monitor reaction
^{197}Au	$^{197}\text{Au}(p,n)$	$E_p \leq 18$	^{197}Hg	possible monitor reaction

It took us more than eight years till we could perform the experimental works (in collaboration with several research groups from different countries), the data evaluations and the publication of the results for the reactions listed in Table 2.1. The necessity of studying some of the above reactions was already proposed in my thesis for the university doctor's degree (Szelecsényi 1988). From this point of view this study is a continuation of that work.

2.3 List of the original publications

This work was based on the following papers, which are referred to in the text of this thesis by their bold Roman numerals. The photocopies of these publications can be found at the end of the thesis in the Appendix.

- I** Tárkányi F., Szelecsényi F., Kovács Z., Sudár S.: Excitation Functions of Proton Induced Nuclear Reactions on Enriched ^{66}Zn , ^{67}Zn , ^{68}Zn . Production of ^{67}Ga and ^{66}Ga . *Radiochimica Acta* **50** (1990) 19-26

Cited by:

1. Qaim S.M. in Proceeding of the IAEA Advisory Group Meeting on Intermediate Energy Nuclear Data for Applications. 9-12 October, 1990, Vienna, Austria, p.45
2. Hermanne A. *et al.* in Abstracts of International Conference on Nuclear Data for Science and Technology, 13-17 May, 1991. Jülich, Germany (1991) p.259
3. Ramavataram S. in Nuclear Data Sheets **62** (1991) 529
4. Rösch F. *et al.* in ZfK Preprint, Dresden, Germany, ZfK-745 (1991) 1
5. Mirzadeh S. *et al.* in Proceedings of International Conference on Nuclear Data for Science and Technology. 13-17 May, 1991, Jülich, Germany. Ed.: S.M.Qaim. Springer Verlag. (1992) p.619
7. Iljinov A.S. *et al.* in Production of Radionuclides at Intermediate Energies. Landolt-Börnstein, New Series, Vol.13A, Group I.: Nuclear and Particle Physics. Berlin, Springer Verlag (1991) 1
6. Hermanne A. *et al.* in Proceedings of International Conference on Nuclear Data for Science and Technology. 13-17 May, 1991, Jülich, Germany. Ed.: S.M.Qaim. Springer Verlag. (1992) p.616
8. Weinreich R. in Proceedings of the IVth International Workshop on Targetry and Target Chemistry. 9-12 September, 1991 Villigen, Switzerland, PSI-Proceedings 92-01, 1992, p.50
9. Tóth Gy. Egyetemi doktori értekezés. Debrecen, MTA Atommagkutató Intézet. (1992) p.1
10. Jao Y. *et al.* in Journal of Radioanalytical and Nuclear Chemistry, Articles **195** (1995) 279
11. Iljinov A.S. *et al.* in Production of Radionuclides at Intermediate Energies. Landolt-Börnstein, New Series, Vol.13D, Group I.: Nuclear and Particle Physics. Berlin, Springer Verlag (1994)
12. Dmitriev S. N. *et al.* in Fizika Elementarnykh Chastits i Atomnogo Yadra **27** (1996) 977

- II** Tárkányi F., Szelecsényi F., Kopecký P.: Excitation Functions of Proton Induced Nuclear Reactions on Natural Nickel for Monitoring Beam Energy and intensity. *International Journal of Radiation Applications and Instrumentation Part "A": Applied Radiation and Isotopes* **42** (1991) 513-517

Cited by:

1. Piel H. in Bestimmung der Anregungsfunktionen von (p,xn)-Reaktionen an Ni-Isotopen. Diplomarbeit, Universität zu Köln, Germany (1991)
2. Weinreich R. in Proceedings of the IVth International Workshop on Targetry and Target Chemistry. 9-12 September, 1991, Villigen, Switzerland, PSI-Proceedings 92-01 (1992) p.50
3. Sonck M. in Diplomwork in Engineering, Vrije Universitet, Fakulteit Toegepaste Wetenschappen Brussel, Belgium, 1993 p.130
4. Sudár S. *et al.* in Physical Review **C50** (1994) 2408
5. Sonck M. in Diplomwork for Certification in Biomedical and Clinical Engineering. Vrije Universitet, Fakulteit Toegepaste Wetenschappen, Brussel, Belgium, 1994 p.40

6. Ijtinov A.S. in Production of Radionuclides at Intermediate Energies. Landolt-Börnstein, Numerical Data and Functional Relationships in Science and Technology. (Ed. Schopper H.) New Series, Vol.13D, Group I., Springer Verlag, Berlin, Germany, (1994)
7. Sonck M. *et al.* in Applied Radiation and Isotopes **47** (1996) 445
8. Schiekel Th. *et al.* in Nuclear Instruments and Methods in Physics Research **B114** (1996) 91
9. Mahunka I. *et al.* in Journal of radioanalytical and Nuclear Chemistry, Letters **213** (1996) 135
10. Reimer P. *et al.* in Abstracts of the XIIth International Symposium on Radiopharmaceutical Chemistry, 15-19 June, 1997, Uppsala, Sweden (1997) p.299
- III** Kopecký P., Szelecsényi F., Molnár T., Mikecz P., Tárkányi F.: Excitation Functions of (p,xn) Reactions on ^{nat}Ti: Monitoring of Bombarding Proton Beams.
Applied Radiation and Isotopes **44** (1993) 687-692
Cited by:
1. Ditrói F. in The Thin Layer Activation Method and its Applications in Industry. IAEA-TECHDOC-924, IAEA, Vienna, Austria, 1997, p.29
- IV** Boothe T.E., McLeod T.F., Plitnikas M., Kinney D., Tavano E., Feijoo Y., Smith P. Szelecsényi F.: Commercial and PET Radioisotope Manufacturing with a Medical Cyclotron.
Nuclear Instruments and Methods in Physics Research **B79** (1993) 926-928
- V** Szelecsényi F., Blessing G., Qaim, S.M.: Excitation Functions of Proton Induced Nuclear Reactions on Enriched ⁶¹Ni and ⁶⁴Ni: Possibility of Production of No-Carrier-Added ⁶¹Cu and ⁶⁴Cu at a Small Cyclotron.
Applied Radiation and Isotopes **44** (1993) 575-580
Cited by:
1. McCarthy *et al.* in Journal of Labelled Compounds and Radiopharmaceuticals **37** (1995) 830
2. Philpott G. W. *et al.* in Journal of Nuclear Medicine **36** (1995) 1818
3. McCarthy *et al.* in Proceedings of Sixth Workshop on Targetry and Target Chemistry. Vancouver, B.C., Canada, 17-19 August 1995, (Eds. Link J. M. and Ruth T.J.) Triumf Publishing Office, 1996 p.92
4. Blower P.J. *et al.* in Nuclear Medicine and Biology **23** (1996) 957
5. Smith T. *et al.* in Radiochimica Acta **75** (1996) 65
6. Takacs S. *et al.* in Radiochimica Acta **76** (1997) 15
- VI** Sudár S., Szelecsényi F., Qaim, S.M.: Excitation Function and Isomeric Cross-Section Ratio for the ⁶¹Ni(p,α)⁵⁸Co^{m.g} Process.
Physical Review **C48** (1993) 3115-3118
- VII** Tárkányi F., Szelecsényi F., Kopecký P., Molnár T., Andó L., Mikecz P., Tóth Gy., Rydl A.: Cross Section of Proton Induced Nuclear Reactions on Enriched ¹¹¹Cd and ¹¹²Cd for the Production of ¹¹¹In for Use in Nuclear Medicine.
Applied Radiation and Isotopes **45** (1994) 239-249
Cited by:
1. Nortier *et al.* in Applied Radiation and Isotopes **45** (1994) 239
2. Stöcklin G. in Radiochimica Acta **70** (1995) 249
- VIII** Szelecsényi F., Boothe T. E., Tavano E., Plitnikas M., Feijoo Y., Takács S., Molnár T., Tárkányi F.: Compilation and Evaluation of Cross Sections Thick Target Yields for ¹¹¹In Production.
Proceedings of the International Conference on Nuclear Data for Science and Technology. (Ed. Dickens J.K.), 9-13 May, 1994, Gatlinburg, TN, USA. American Nuclear Society Inc. La Grange Park, 1994 pp.1042-1044

- IX** Szelecsényi F., Boothe T.E., Tavano E., Plitnikas M., Tárkányi F.: Compilation of Cross Sections/Thick Targets Yields for ^{66}Ga , ^{67}Ga and ^{68}Ga Production Using Zn Target up to 30 MeV Proton Energy.
Applied Radiation and Isotopes **45** (1994) 473-500
Cited by:
1. Stöcklin G. in *Radiochimica Acta* **70** (1995) 249
2. Schwarzbach R. *et al.* in *Applied Radiation and Isotopes* **46** (1995) 329
3. Dmitriev S. N. *et al.* in *Fizika Elementarnykh Chastits i Atomnogo Yadra* **27** (1996) 977
4. Ditrói in IAEA-TECHDOC-924, Vienna, Austria, 1977 p.29
- X** Qaim S. M., Uhl M., Rösch F., Szelecsényi F.: Excitation Functions of (p, α) Reactions on ^{64}Ni , ^{78}Kr , and ^{86}Sr .
Physical Review **C52** (1995) 733-739
- XI** Sonck M., Hermanne A. Szelecsényi F., Takács S. and Tárkányi F.: Study of the $^{\text{nat}}\text{Ni}(p,x)^{57}\text{Ni}$ Process up to 44 MeV for Monitor Purposes.
Applied Radiation and Isotopes **48** (1997) accepted for publication as a technical note.
- XII** Szelecsényi F., Boothe T.E., Takács S., Tavano E., Tárkányi F.: Evaluated Cross Section and Thick Target Yield Data Bases of Zn+p Processes for Practical Applications.
Applied Radiation and Isotopes **48** (1997) accepted for publication.
- XIII** Szelecsényi F., Takács S., Fenyvesi A., Szücs Z., Tárkányi F., Heselius, S.-J., Bergman, J., Boothe T.E.: Study of the $^{197}\text{Au}(p,pn)^{196\text{m1,m2,g}}\text{Au}$ and $^{197}\text{Au}(p,n)^{197\text{m}}\text{Hg}$ Reactions and Their Application for Proton Beam Monitoring in Radioisotope Production.
Accepted for publication in *Proceedings of International Conference on Nuclear Data for Science and Technology*, 19-23 May 1997, Triest, Italy,

Part of the results published in the following articles were also used in this thesis, but the copies of these articles are not enclosed.

- XIV** Szelecsényi F., A ^{67}Ga radioizotóp előállítás MGC ciklotronnal. (Production of ^{67}Ga with MGC-20 Cyclotron)
Thesis for university doctor's degree (supervisors: Tárkányi F. and Mahunka I.) MTA Atomki, Debrecen, Hungary (1988) pp. 1-104
- XV** Szelecsényi F., Tárkányi F., Kovács Z., Bergman, J., Heselius, S.-J., Solin, O.: Production of ^{66}Ga and ^{67}Ga .
Acta Radiologica Supplementum **376** (1991) 62-63
- XVI** Tárkányi F., Szelecsényi F., Takács S.: Determination of Effective Bombarding Energies and Fluxes Using Improved Stacked Foil Technique
Acta Radiologica. Supplementum **376** (1991) 72-73
Cited by:
1. Sonck M. *et al.* in *Applied Radiation and Isotopes* **47** (1996) 445

XVII Szelecsényi F., Tárkányi F., Andó L., Mikecz P., Tóth Gy., Kopecký P., Rydl A.: Excitation Functions of Proton Induced Nuclear Reactions on ^{111}Cd and ^{112}Cd . Production of ^{111}In .

Proceedings of International Conference on Nuclear Data for Science and Technology. (Ed.: S.M.Qaim) 13-17 May, 1991, Jülich, Germany. Springer-Verlag, Berlin, Germany (1992) pp. 603-605

Cited by:

1. Sonck M. in Diplomwork in Engineering, Vrije Universitet, Fakuliteit Toegepaste Wetenschappen Brussel, Belgium, 1993 p.130
2. Sonck M. in Diplomwork for Certification in Biomedical and Clinical Engineering. Vrije Universitet, Fakuliteit Toegepaste Wetenschappen, Brussel, Belgium, 1994 p.40
3. Iljinov A.S. in Production of Radionuclides at Intermediate Energies. Landolt-Börnstein, Numerical Data and Functional Relationships in Science and Technology. (Ed. Schopper H.) New Series, Vol.13D, Group I., Springer Verlag, Berlin, Germany, (1994) p.1

XVIII Tárkányi F., Szelecsényi F. and Kopecký, P.: Cross Section Data for Proton, ^3He and α -particle Induced Reactions on $^{\text{nat}}\text{Ni}$, $^{\text{nat}}\text{Cu}$ and $^{\text{nat}}\text{Ti}$ for Monitoring Beam Performance.

Proceedings of International Conference on Nuclear Data for Science and Technology. (Ed.: S.M.Qaim), 13-17 May, 1991, Jülich, Germany. Springer-Verlag, Berlin, Germany (1992) pp. 529-532

Cited by:

1. Rösch F. *et al.* in Radiochimica Acta **61** (1993) 1
2. Sonck M. *et al.* in Abstracts for the 13th International Conference on the Application of Accelerators in Research and Industry. (Ed. Duggan J. L.) 1994, p.116
3. Hermanne A. *et al.* in Proceedings of the International Conference on Nuclear Data for Science and Technology. (Ed. Dickens J.K.) 9-13 May, 1994, Gatlinburg, TN, USA, American Nuclear Society Inc., La Grange Park, 1994 p.1039
4. Sonck M. in Diplomwork in Engineering, Vrije Universitet, Fakuliteit Toegepaste Wetenschappen Brussel, Belgium, 1993 p.130
5. Sonck M. in Diplomwork for Certification in Biomedical and Clinical Engineering. Vrije Universitet, Fakuliteit Toegepaste Wetenschappen, Brussel, Belgium, 1994 p.40
6. Csák Cs. in Diplomamunka, MTA Atomki, Debrecen 1994
7. Merchel S. *et al.* in Diplomarbeit. Universität zu Köln, Germany (1995)
8. Denzler F.O *et al.* in Radiochimica Acta **69** (1995) 209
9. Sonck M. *et al.* in Applied Radiation and Isotopes **47** (1996) 445
10. Ditrói in IAEA-TECHDOC-924, Vienna, Austria, 1977 p.29

XIX Szelecsényi F., Boothe, T.E., Takács S., Tárkányi F., Tavano, E., Plitnikas, M.: Nuclear Data Relevant to the Production of ^{67}Ga : A Critical Comparison of Excitation Functions / Thick Target Yield Data for the $^{67}\text{Zn}(p,n)$ and $^{68}\text{Zn}(p,2n)$ Nuclear Reactions.

Proceedings of the Fifth International Workshop on Targetry and Target Chemistry. (Eds. Dahl R. *et al.*), 19-23 September, 1993, Upton, NY. USA. published by Brookhaven National Laboratory (1994) BNL-61149, pp.234-242

Cited by

1. Ditrói in IAEA-TECHDOC-924, Vienna, Austria, 1977 p.29

XX Szelecsényi F., Boothe, T.E., Tavano, E., Plitnikas, M., Feijoo, Y., Takács S., Tárkányi F., Szücs Z. New Cross Section Data for $^{66,67,68}\text{Zn}+p$ Reactions up to 26 MeV.

Proceedings of the International Conference on Nuclear Data for Science and Technology. (Ed. Dickens J.K.), 9-13 May, 1994, Gatlinburg, TN, USA American Nuclear Society Inc. La Grange Park, (1994) pp.393-396

Cited by

1. Ditrói in IAEA-TECHDOC-924, Vienna, Austria, 1977 p.29

3. Basic equations, model codes

3.1 Mechanism of the nuclear reaction

In the case of the majority of irradiations for routine radioisotope production, the bombarding particle beam energies are usually less than 50 MeV. Up to this energy there are three important models which can be used to describe those interactions of energetic particles with the target nucleus where a new nucleus is formed.

a) *Compound nucleus reaction*: This reaction can be treated as a two-step process. In the first step the incident particle which stays in the nucleus for a relative long time (10^{-15} s) delivers its energy to many nucleons in the target nucleus, and this energy is rapidly distributed throughout the nucleus. The incident particle itself becomes indistinguishable from other nucleons in the compound nucleus, and we can say that the compound nucleus ‘forgets’ the way in which it was formed. Due to this forgetfulness of the compound nucleus, the second step is independent and unrelated to the first step. The excitation energy of the compound nucleus is equal to the kinetic energy introduced by the incident particle plus its binding energy. This energy is statistically distributed among the nucleons, and each nucleon is rapidly colliding with the others and changing its energy. It may happen that by chance enough energy is concentrated on one nucleon, that they can leave the nucleus, and a particle(s) emission takes place.

b) *Precompound reaction*: With increasing projectile energy (above 10 MeV) sometimes a particle emission can occur even before the whole energy could distribute evenly among the nucleons of the compound nucleus.

c) *Direct reaction*: This process becomes probable when the projectile spends less time in the vicinity of the target nucleus. (10^{-22} s) The interaction between the incident particle and the target nucleus takes place usually close to the surface of the nucleus. Only a few nucleons of the target nucleus take part in this process.

3.2 Cross sections of a nuclear reaction

The probability of a nuclear reaction at a given projectile energy is known as the cross section of the nuclear reaction. From the point of view of radioisotope production we only interested in the total activity of the product nuclei that can be produce during a given irradiation. If the beam current constant during the irradiation, the number of the projectile is the same when they enter and leave the target and the target consist of only one isotope, this activity (**A**) can be expressed as the function of the cross sections ($\sigma = \sigma(E)$) as:

$$A = (\Phi N_A) (1 - e^{-\lambda t}) \int_{E_{out}}^{E_{in}} [dE/dx]^{-1} \sigma(E) dE \quad (3.2.1)$$

where: Φ beam flux

N_A number of target atoms per surface unit

λ decay constant

t irradiation time
 dE/dx stopping power
 E_{in} energy of the bombarding beam on the surface of the target
 E_{out} outgoing beam energy

On the other hand the measured activity can be used to calculate this (**integral**) cross section. If the target is 'very thin', the above integral can be substituted for:

$$\int_{E_{out}}^{E_{in}} [dE/dx]^{-1} \sigma(E) dE \approx [\Delta E/\Delta x]^{-1} \sigma^* \Delta E \quad \text{if } E_{in} \approx E_{out} \quad (3.2.2)$$

where: $\Delta E = E_{in} - E_{out}$
 Δx thickness of the target
 σ^* average cross section in the target

For the majority of the nuclear reactions it is true that

$$\sigma^* < |\sigma(E_{in}) - \sigma(E_{out})| \quad (3.2.3)$$

and

$$\sigma^* \rightarrow \sigma(E) \quad \text{if } \Delta x \rightarrow 0 \quad (3.2.4)$$

If the activation technique is used for cross section measurement, we can evaluate only this average integral cross section. In practice, the following equation is used to calculate the average cross section in mbarn (1 barn = 10^{-27} m²):

$$\sigma^* = \alpha C \lambda M / I_g \epsilon \Phi R e^{-\lambda t^*} (1 - e^{-\lambda t^{**}}) (1 - e^{-\lambda t^{***}}) \quad (3.2.5)$$

where: α constant (0.461268186)
 C measured photopeak area
 λ decay constant (1/h)
 M molar weight of the target
 I_g intensity of the measured gamma photon
 ϵ absolute efficiency of the detector
 Φ beam flux (1/s)
 R target thickness (g/cm²)
 t^* cooling time (time between EOB and the start of the measurement [h])
 t^{**} measuring time [h]
 t^{***} irradiation time [h]

In the field of radioisotope production sometimes more than one reaction can contribute at the same time to the formation of the final product. By measuring only the activity of the final product the cross sections of the participating reactions can not be separated. For practical reasons, however, it is usually calculated the so called **elemental** cross sections (summed cross sections weighted by isotopic abundances) on targets which are not monoisotopic. (It is also used in the case monitor reactions.). In the case of cumulative production routes the entire cross sections (production after the full decay of the parent nuclei) are also deduced and called **cumulative cross sections**.

3.3 Target yields

The ‘yield of a nuclear reaction’ (Y) as a ratio of the number of the nucleus formed in a nuclear reaction to the number of the bombarding particles hitting the target can be defined in different ways. The most frequently employed definition (so called physical yield) calculates the yield at the end of a very short irradiation time and has the analytical meaning of the slope of the function of the produced radionuclides versus irradiation time. Several contributions are available in the literature which discuss the different yield definitions employed for practical purposes (*cf* Bonardi, 1988). However, in the field of radioisotope production the above mentioned physical yield and two other ‘target yields’ are mainly used:

1) ‘The ‘1h/1 μ A yield’ is a yield after $t=1$ h irradiation time that is produced by a beam current of 1 μ A can be calculated as

$$Y_{1h/1\mu A} = (\Phi N_L / A) (1 - e^{-\ln 2 / T_{1/2}}) \int_{E_{out}}^{E_{in}} [dE/dx]^{-1} \sigma(E) dE \quad (3.3.1)$$

where: N_L Avogadro constant

A atomic weight (amu)

$T_{1/2}$ half-life of the product nucleus

2) *Saturation yield* (Y_{EOSB} : end of saturation bombardment yield) is the activity that is produced by a beam current of 1 μ A if the irradiation time $t \gg T_{1/2}$. This saturation yield is given by:

$$Y_{EOSB} = (\Phi N_L / A) \int_{E_{out}}^{E_{in}} [dE/dx]^{-1} \sigma(E) dE \quad (3.3.2)$$

3.4 Model codes for calculation of excitation functions

For reactions where no experimental data are available the application of nuclear reaction theory seems to be a feasible method to get more information on the data required. A number of nuclear model codes were developed first to gain theoretical understanding of the nuclear reaction processes involved. Later they have been also used to calculate excitation functions for charged particle induced reactions of medical interest and compared their results with experimental ones. (*cf* Pavlik, 1988)

Most of the computer codes in use to calculate these cross sections employ a combination of the compound nucleus evaporation model and a model for preequilibrium emission (*cf* Nowotny and Uhl, 1987) The compound nucleus evaporation model is based on Bohr’s concept of the compound nucleus which decay is assumed to be independent from its formation. The theoretical treatment of the compound nucleus reactions on the other hand, is based essentially on statistical assumptions. For the description of the preequilibrium emission the exciton model (Griffin, 1966) and the hybrid model (and its extension the geometry dependent hybrid model) (Blann, 1971a, 1971b and 1972) are mainly used.

Those reaction model codes which used the compound nucleus model and a model for preequilibrium emission have been successfully applied to calculate excitation functions of

nuclear reactions with neutrons, protons and alpha particles both in entrance and exit channel. The applicability of these codes, however, limited in the case of light nuclei. For nuclei with mass numbers less than about 20 reliable predictions of excitation functions cannot be expected, due to the individual character of most light elements

We have to point out, however, that the theoretical calculations cannot replace experiments at all as the model parameters like optical model parameters, level density parameters and preequilibrium model parameters need adjustment to experimental data to supply results with sufficient accuracy. To obtain accurate results each calculations of unknown cross sections should be based on parameters which simultaneously reproduce available experimental data in the mass region of interest. The degree of the agreement may then give an estimate for the accuracy of the unknown cross sections.

4. Experimental techniques

For the cross sections measurements we planned to use the most frequently employed way:

- A) Activation method using stacked-foil technique.
- B) Target stacks activation by beams of cyclotrons.
- C) Beam intensity monitoring with Faraday cup and monitor reactions.
- D) Measurement of the activity of the irradiated foils with standard gamma ray spectrometry without chemical separation.

4.1 The stacked- foil technique

4.1.1 Principle of the method

One of the most frequently used methods for the measurement of the cross sections of a nuclear reaction is the so called ‘stacked-foil technique’. In this method several targets (usually thin foils) are placed behind each other and they are irradiated in one step. The simultaneous irradiation of the samples (the stack) is not only economical but also could assure a good relative accuracy. The energy degradation of the entering particle beam along the stack is usually determined by calculations using the stopping power formulae and tables of different authors. (*cf* Williamson *et al.*,1966; Andersen and Ziegler J.,1977). The energy value related to the cross section is the mean of the energy values entering and leaving the given foil (taking into account the change of the excitation function). However, this technique has some disadvantages, especially in those cases when numerous targets are activated and when the experiment was made without the necessary precautions. The main problems arising from the above mentioned cases are as follows:

- a) The beam intensity may changes along the stack due to ‘outscattering’ and/or ‘outfocusing’ of the bombarding particles.
- b) The beam energy can not be estimated properly in the stack due to the errors of the (1) thickness of the samples, (2) stopping power calculation, (3) incident particle energy [which is an accelerator related problem].
- c) Broadening of the beam energy.
- d) In some cases efficiency of the target cooling.

The problems of a) and e) can be solved with proper experimental set-up and low beam current used during the activation, however, experiences showed that after 8-10 MeV energy absorption in the stack, the ‘calculated’ energy may shift 1-2 MeV in comparison to the ‘measured’ value. (It is very common especially in those cases when targets are prepared by electrodeposition methods.) To decrease the influence of the errors mentioned in b) and c) to the estimation of the ‘actual’ beam energy, one has to decrease the number of the foils in the stack and/or has to use so called monitor reaction(s). (See later in this chapter).

4.1.2 Application of monitor reactions in ‘stacked-foils’ type cross section measurements

Though there are many factors which cause errors in obtaining experimentally the cross sections, one of the most important systematic error comes from the estimation of the beam flux. The beam flux is usually obtained by measuring the electric current induced by the

incident beam in a Faraday-cup. However, if precaution is not made for preventing the escape of secondary electrons or ions which may be caused by bombarding particles in the gas (which remained in the system due to insufficient vacuum), the measurement of beam intensity may cause an unexpected error of the cross sections.

Instead to measure the cross sections each time absolutely, it is more easier to measure them relative to those of a selected reaction (called later as monitor reaction) whose cross sections is already very well known in the interested energy region. The use of a monitor reaction has also an advantage in those cases when the use a Faraday-cup is difficult or even not possible. A monitor reaction should satisfy at least the majority of the following conditions (Hashizume, 1988).

- 1) The target element(s) should be isotopically pure or disturbances of quantitative determination of reaction product caused by other isotope products should be small.
- 2) The absolute cross sections should be known precisely in wide range of energies of the incident particle.
- 3) The excitation function should change smoothly as the incident particle energy increases. Reactions which have a sharp resonance(s) should be avoided.
- 4) The effects of secondary particles induced by primary reactions should be small.
- 5) The half-life of the reaction product(s) should not to be too short or very long as compared it to the irradiation time.
- 6) The target foil should be obtained without difficulty.
- 7) The target should be stable during the activation. Materials with low melting point should be avoided.
- 8) The reaction product(s) should be remain in the target material during and after the activation.
- 9) The activity of the reaction product(s) should be determined accurately and easily. The nuclear data of the product(s) should be accurately known.

Prior to our measurements only some reactions were suggested to use for proton beam intensity monitoring up to 30 MeV. For example: $^{12}\text{C}(p,pn)^{11}\text{C}$, $^{59}\text{Co}(p,pn)^{58}\text{Co}$, $^{63}\text{Cu}(p,n)^{63}\text{Zn}$, $^{65}\text{Cu}(p,2n)^{62}\text{Zn}$, $^{\text{nat}}\text{Cu}(p,x)^{61}\text{Cu}$ (*cf* Schwerer and Okamoto, 1989).

In the practice, several monitor foils (sometimes different materials) are mounted among the target foils to increase the reliability of this method. If we can assure that different monitor reactions (with different threshold energies) are induced in these foils then simultaneous beam intensity and energy evaluation can be done as it was demonstrated by us in **XVI**.

4.2 Targets

4.2.1 Targets for cross section measurements

Isotopically enriched materials were used to prepare targets in those cases when the investigated reactions were important for routine radioisotope production. The targets used for monitor cross section measurements were commercially available thin metal foils with natural isotopic composition usually obtained from the Goodfellow Metals, Cambridge, England.

The enriched Ni materials were purchased from Isotec Inc., Miamisburg, OH, U.S.A. while the Zn and Cd materials were supplied by Technabexport, Moscow, Russia/USSR. The isotopic composition of the above materials are collected in the appropriate tables of **V**, **I** and **VII** for Ni, Zn and Cd, respectively. In the case of enriched materials we have prepared the samples by electrodeposition on commercially available high purity thin metal foils (thickness ranged from 5-20 μm ; purchased also from Goodfellow). The description of the electrodeposition methods developed by us are described in more detailed in **I** and **VII** for Zn

and Cd, respectively. In the case of the Cd targets the chemical procedure was developed by our former colleague Dr. P. Mikecz. The Ni material was electrodeposited onto thin Au foils by using the method developed by Piel *et al.* (1992).

4.2.2 Targets for routine radioisotope production

Targets for routine radioisotope production ('production targets') were prepared using almost the same chemical methods which were used in the case of the enriched thin targets. In the case of production targets (both external or internal irradiations) the body of the target backing is usually made from copper. This material is relatively cheap, easy to fabricate and its mechanical and heat transfer properties are relative good. The surface of the copper is usually covered (electroplated) with a thin layer of other element to prevent the dissolving of the copper during the chemical separation. Table 4.2.1 shows the typical parameters of the targets which were used for cross section measurements and routine radioisotope production.

Table 4.2.1. Material, weight and size of the targets used for cross section measurements and routine production.

Target material	Backing (and covering)	Thickness (mg/cm ²) [weight]	Diameter (mm)	Reference
^{nat} Ti	-	10, 35	14	III
^{nat} Ni	-	8.9, 17.7, 26.6	20	II
⁶¹ Ni	Au	0.4 to 1.2	12	V
	Cu (Au)	[~140 mg for production]	12	V
⁶⁴ Ni	Au	0.4 to 1.2	12	V
	Cu (Au)	[~140 mg for production]	12	V
⁶⁶ Zn	Ni	15 (average)	10	I
	Cu (Ni)	[~250 mg for production]	12	I
⁶⁷ Zn	Ni	15 (average)	10	I
	Cu (Ni)	[~250 mg for production]	12	I
⁶⁸ Zn	Ni	15 (average)	10	I
	Cu (Ni)	[~250, mg for production] [4000-5000 mg for production]	12 ~18 cm ²	I IV
¹¹¹ Cd	Ni	15 (average)	10	VII
	Cu (Ni)	[~500 mg for production]	12	VII
¹¹² Cd	Ni	15 (average)	10	VII
	Cu (Ni)	[~400 mg for production]	12	VII

The estimation of the effective thickness of thin samples, especially the electrodeposited ones, is often difficult. The targets are usually individually weighed before activation (in case of

enriched materials both the backing and the sample+backing) for determination of target thicknesses. However, this method cannot show the possible surface inhomogeneities. Inhomogeneous targets can cause serious problems during the evaluation of the effective beam energy in the targets. To decrease the error caused by this problem, we have checked the uniformity of the targets by measuring the thickness along different axes of the enriched targets. Our results for example in the case of enriched Zn targets can be seen in Fig.2. of I. Using this method we could select out not only the ‘unproper’ targets, but it was also possible to locate the ‘useful surface’ (i.e. where the inhomogeneity was less than 3-5 %) of each target. Knowing this ‘useful surface’ of the sample one can select the right size of the beam collimator to be used during the irradiation(s). According to our measurements, the inhomogeneity of the prefabricated thin metal foils was less than 2%)

4.3 Accelerators

All irradiations for cross section measurements took place at external beams of different cyclotrons. The list of the accelerators used for irradiation of the targets and the list of the investigated reactions/routinely produced isotopes are collected in Table 4.3.1. The incident proton energies were derived from the frequency and actual extraction radius of the cyclotrons. These energies, however, were checked using analyzing magnets for the Rez and Debrecen experiments, and by an iteration method based on measured activities of different monitor foils at Brussels, Jülich, Miami Beach and Turku (see more detailed in XVI). This way these incident beam energies were determined with an accuracy of ± 0.2 - 0.3 MeV.

Table 4.3.1. List of the cyclotrons used during our experiments.

Type of Cyclotron	Institute	Proton energy	Investigated reactions, [routinely produced radioisotope]
MGC-20E	Institute of Nuclear Research (ATOMKI) Debrecen, Hungary	$E_p \leq 18$ MeV	$^{nat}\text{Ti}+p$, $^{nat}\text{Ni}+p$, $^{66,67,68}\text{Zn}+p$, $^{111,112}\text{Cd}+p$ [$^{66,67}\text{Ga}$, ^{111}In]
MGC-20E	Abo Akademi Accelerator Laboratory, Turku, Finland	$E_p \leq 18$ MeV	$^{66,67,68}\text{Zn}+p$, $\text{Au}+p$
CV-28	KFA Forschungsanlage Jülich, Germany	$E_p \leq 18.7$ MeV	$^{nat}\text{Ni}+p$, $^{66,67,68}\text{Zn}+p$, $\text{Au}+p$, $^{61,64}\text{Ni}+p$
U-120M	Rez, Czech Republic	$E_p \leq 35$ MeV	$^{nat}\text{Ti}+p$, $^{nat}\text{Ni}+p$, $^{111,112}\text{Cd}$
CS-30	Cyclotron Department of Mount Sinai Medical Center, Miami Beach, Florida, U.S.A	$E_p \leq 26.5$ MeV	$^{66,67,68}\text{Zn}+p$, [^{67}Ga , ^{111}In]
CGR-560	Vrije Universiteit Brussel (VUB) Brussels, Belgium	$E_p \leq 44$ MeV	$^{nat}\text{Ni}+p$

4.4 Irradiation circumstances and measurement of activity

Targets used for cross section measurements were placed in target holders, which usually served as Faraday-cup for beam current measurements. The target holders were placed in the vacuum chamber except in the case of the Turku experiments where they were separated from the vacuum system of the cyclotron with two thin foils ('exit foils'; stainless steel foil of 12.5 μm thickness). The surface of these foils were cooled by He gas of 1.2 bar. Table 4.4.1. gives the summary of the experimental circumstances of our experiments.

Table 4.4.1. Summary of the pertinent experimental circumstances

Reaction	Incident proton energy (MeV)	Number of targets	Irradiation time (T_{irr}) and Beam current (I)	References and Remarks
$^{\text{nat}}\text{Ti}+\text{p}$	$E_p=30.2$ $E_p=18.0$ $E_p=14.5$	48 14 7	$T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=120$ min, $I=200$ nA	III
$^{\text{nat}}\text{Ni}+\text{p}$	$E_p=43.7$ $E_p=40.4$ $E_p=40.4$ $E_p=37.5$ $E_p=36.5$ $E_p=34.5$ $E_p=32.6$ $E_p=32.25$ $E_p=31.25$ $E_p=30.0$ $E_p=28.75$ $E_p=26.7$ $E_p=22.0$ $E_p=22.0$ $E_p=21.75$ $E_p=18.0$	5 7 5 7 5 5 5 7 5 30 7 8 22 17 9 10	$T_{\text{irr}}=53$ min, $I=2.8$ μA $T_{\text{irr}}=5$ min, $I=5$ μA $T_{\text{irr}}=20$ min, $I=8$ μA $T_{\text{irr}}=5$ min, $I=5$ μA $T_{\text{irr}}=20$ min, $I=8$ μA $T_{\text{irr}}=20$ min, $I=8$ μA $T_{\text{irr}}=20$ min, $I=8$ μA $T_{\text{irr}}=5$ min, $I=5$ μA $T_{\text{irr}}=20$ min, $I=8$ μA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=10$ min, $I=5$ μA $T_{\text{irr}}=5$ min, $I=5$ μA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=60$ min, $I=100$ nA $T_{\text{irr}}=10$ min, $I=5$ μA $T_{\text{irr}}=60$ min, $I=150$ nA	II (for $E_p=30, 22, 22$ and 18 MeV) XI (for $E_p=43.7, 40.4, 40.4, 37.5, 36.5, 34.5, 32.6, 32.25, 31.25, 28.75, 26.7$ and 21.75 MeV)
$^{61}\text{Ni}+\text{p}$	$E_p=18.7$ $E_p=18.7$ $E_p=18.7$ $E_p=18.7$ $E_p=18.7$ $E_p=18.7$ $E_p=18.7$ $E_p=18.7$	7 6 5 5 5 5 5 13	$T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=60$ min, $I=150$ nA $T_{\text{irr}}=300$ min, $I=200$ nA	V Al foils of different thicknesses were used for energy degradation in front of targets
$^{64}\text{Ni}+\text{p}$	$E_p=14.9$ $E_p=18.7$ $E_p=18.7$ $E_p=18.7$ $E_p=18.7$ $E_p=18.7$	2 5 5 5 5 4	$T_{\text{irr}}=30$ min, $I=150$ nA $T_{\text{irr}}=30$ min, $I=150$ nA $T_{\text{irr}}=30$ min, $I=150$ nA $T_{\text{irr}}=30$ min, $I=150$ nA $T_{\text{irr}}=30$ min, $I=150$ nA $T_{\text{irr}}=30$ min, $I=150$ nA	V Al foils of different thicknesses were used for energy degradation in front of targets
$^{66}\text{Zn}+\text{p}$	$E_p=26.0$ $E_p=26.0$ $E_p=26.0$	8 5 5	$T_{\text{irr}}=30$ min, $I=600$ nA $T_{\text{irr}}=60$ min, $I=200$ nA $T_{\text{irr}}=60$ min, $I=200$ nA	I (for $E_p=22, 18$ and 14 MeV), XII (for $E_p=26.0$ MeV)

	$E_p=26.0$	5	$T_{irr}=70\text{ min, }I=200\text{ nA}$	Al foils of different thicknesses were used for energy degradation in front of targets in the case of $E_p=26.0\text{ MeV}$ irradiations
	$E_p=22.0$	6	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
	$E_p=18.0$	6	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
	$E_p=18.0$	6	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
	$E_p=14.0$	3	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
$^{67}\text{Zn+p}$	$E_p=26.0$	4	$T_{irr}=60\text{ min, }I=600\text{ nA}$	I (for $E_p=22, 18$ and 14 MeV), XII (for $E_p=26.0\text{ MeV}$) Al foils of different thicknesses were used for energy degradation in front of targets in the case of $E_p=26.0\text{ MeV}$ irradiations
	$E_p=26.0$	4	$T_{irr}=60\text{ min, }I=200\text{ nA}$	
	$E_p=26.0$	5	$T_{irr}=30\text{ min, }I=200\text{ nA}$	
	$E_p=26.0$	5	$T_{irr}=30\text{ min, }I=200\text{ nA}$	
	$E_p=22.0$	6	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
	$E_p=18.0$	8	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
	$E_p=18.0$	8	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
$^{68}\text{Zn+p}$	$E_p=26.0$	1	$T_{irr}=60\text{ min, }I=600\text{ nA}$	I (for $E_p=22, 18$ and 14 MeV), XII (for $E_p=26.0\text{ MeV}$) Al foils of different thicknesses were used for energy degradation in front of targets in the case of $E_p=26.0\text{ MeV}$ irradiations
	$E_p=26.0$	5	$T_{irr}=60\text{ min, }I=200\text{ nA}$	
	$E_p=26.0$	4	$T_{irr}=30\text{ min, }I=200\text{ nA}$	
	$E_p=26.0$	4	$T_{irr}=30\text{ min, }I=200\text{ nA}$	
	$E_p=26.0$	3	$T_{irr}=60\text{ min, }I=200\text{ nA}$	
	$E_p=22.0$	8	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
	$E_p=18.0$	7	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
	$E_p=18.0$	6	$T_{irr}=30\text{ min, }I=120\text{ nA}$	
$^{111}\text{Cd+p}$	$E_p=30.2$	7	$T_{irr}=30\text{ min, }I=200\text{ nA}$	VII
	$E_p=22.0$	10	$T_{irr}=30\text{ min, }I=200\text{ nA}$	
	$E_p=18.0$	14	$T_{irr}=60\text{ min, }I=200\text{ nA}$	
$^{112}\text{Cd+p}$	$E_p=30.2$	12	$T_{irr}=30\text{ min, }I=200\text{ nA}$	VII
	$E_p=22.0$	8	$T_{irr}=30\text{ min, }I=200\text{ nA}$	
	$E_p=18.0$	5	$T_{irr}=60\text{ min, }I=200\text{ nA}$	
$^{197}\text{Au+p}$	$E_p=18.7$	4	$T_{irr}=12\text{ min, }I=150\text{ nA}$	XIII
	$E_p=18.7$	4	$T_{irr}=12\text{ min, }I=150\text{ nA}$	
	$E_p=18.7$	4	$T_{irr}=12\text{ min, }I=150\text{ nA}$	
	$E_p=18.7$	4	$T_{irr}=12\text{ min, }I=150\text{ nA}$	
	$E_p=18.7$	4	$T_{irr}=12\text{ min, }I=150\text{ nA}$	
	$E_p=18.7$	4	$T_{irr}=12\text{ min, }I=150\text{ nA}$	
	$E_p=18.7$	4	$T_{irr}=12\text{ min, }I=150\text{ nA}$	
	$E_p=18.7$	4	$T_{irr}=12\text{ min, }I=150\text{ nA}$	
	$E_p=17.0$	20	$T_{irr}=90\text{ min, }I=200\text{ nA}$	

The activity of the irradiated samples was measured in all cases by standard gamma-ray spectrometry without chemical separation. (Except in the case of $^{64}\text{Ni}(p,\alpha)^{61}\text{Co}$ reaction [see in **X**] where four targets were dissolved before the activity measurement.) The detectors were Ge(Li) or HpGe detectors which were coupled to PC-based analyzers (Aptec, Nucleus, Camberra etc.). Special attention was paid to the correct determination of the efficiency of the detectors and to the correction of pile-up and cascade losses. Due to the large source to detector distance (usually higher than 25 cm) the corrections for the finite source diameter was also negligible. The energy and efficiency calibration of the systems were done using standard calibration sources. Detailed list of these sources can be found in the appropriate sections of our articles (**I, II, V, VI, VII, X, XI and XIII**). The recoil effect of residual nuclei

for electrodeposited targets was neglected because of the backing materials. The activity loss caused by recoil in the case of the monitor foils was always checked experimentally by activity measurement and was found to be negligible.

4.5 Calculation of the cross sections, energy degradation and their errors

The average cross sections (σ^*) [in mbarn (1 barn = 10^{-27} m²)] were calculated from the measured activities using the standard activation equation (see Eq. 3.2.5).

The peak area analysis was done using the built-in subroutine of the MCA's. In those cases where multiple peak separation was necessary, we have used a flexible gamma-ray analysis program FGM (Székely, 1985). The cross section data published in our articles are 'elemental' cross sections, i. e. they describe the formation cross sections of activation products assuming the target to be monoisotopic and if targets with natural isotopic composition were activated. They include the contribution of all nuclear reactions leading directly, or through possible decay processes of some intermediate nuclei, to the formation of the products under investigation. (In those cases where cumulative production routes are also exist, the entire cross section (production after the full decay of the parent nuclei) was deduced for practical purposes.) Since our enriched materials contained less than 100% of the nuclei to be investigated, especially in the case of ⁶¹Ni (88.84%) or ⁶⁷Zn (91.5%), special corrections were applied in calculating the (absolute) cross section values. In the first step cross sections were computed on the assumption of 100% enriched targets. Since samples having different isotopic compositions were irradiated, usually not at the same energies, the calculated cross sections were fitted with a cubic-spline for making the corrections for the real isotopic composition. Using these fitted curves, correction functions were then calculated as a function of energy. During the last step, the measured individual points were corrected in accordance with these correction functions. Using this method we could give absolute cross section values for the following reactions: ⁶¹Ni(p,n)⁶¹Cu, ⁶¹Ni(p,2n)⁶⁰Cu, ⁶⁶Zn(p,n)⁶⁶Ga, ⁶⁷Zn(p,n)⁶⁷Ga, ⁶⁷Zn(p,2n)⁶⁶Ga, ⁶⁸Zn(p,2n)⁶⁷Ga, ⁶⁸Zn(p,3n)⁶⁶Ga, ¹¹¹Cd(p,2n)¹¹⁰In, ¹¹¹Cd(p,n)¹¹¹In, ¹¹²Cd(p,3n)¹¹⁰In and ¹¹²Cd(p,2n)¹¹¹In. In the case of our 'recommended cross sections/integral thick target yield data bases', the acceptable literature results published through 1996 were evaluated (together with our values) via curve fitting to supply values for practical applications such as routine radioisotope production, nuclear wear measurements etc. The fitting procedure of the acceptable data sets was as follows: First, the individual data sets were fitted separately and then values were calculated for the same energy points from the fitted data set in 0.5 MeV increments. The error of these fitted values in a given energy point was estimated by summing up the error of the nearest experimental value and the deviation of the fitted value in the energy point in question from the interpolated value calculated from the two nearest experimental data points. In the second step, weighted average values and their corresponding uncertainties were computed from the fitted ones at each grid point. Finally, to eliminate the significant discontinuities arising from the fact that different energy ranges were investigated by different groups, the weighted averages were fitted once again and the errors were calculated (Takács *et al.*, 1994). These final evaluated cross sections and their associated uncertainties are tabulated in 0.5 MeV increments in appropriate Tables of articles **I-XIII**. The yields based on integration of the "fitted" excitation curves using the stopping power of Andersen and Ziegler (1977) are also listed in above Tables of **I-XIII**.

The total errors associated with our cross sections were calculated by summing up quadratically the possible individual relative errors: counting statistics, 1-5%; detector efficiency and geometry, 3-5%; decay data, 1-2%; number of target nuclei, 5-7% and incident bombarding particle intensity, 3-5%. The total estimated error of the absolute cross sections - where the error

propagation through correction for real isotopic composition was also taken into account - varied from 9 to 15%

The energies in the middle of the targets and the monitor foils were initially derived from the energy degradation calculation using the formulae and data of Williamson *et al.*, (1966) and Andersen and Ziegler (1977). The estimated errors in the energy scales were varied from ± 0.2 to ± 0.8 MeV along the irradiated stacks, taking into account the energy error of the incident beams, the beam broadening, and the error propagation.

4.6 Yield measurements and routine productions

Targets prepared for routine production were also used for yield measurements with low and high beam currents. Increasing the beam current step by step it was possible to check the behavior of the targets during production circumstances and we could also find the maximum beam currents which could be used without damaging the target. For these studies external and internal target stations of different cyclotrons were used (Rez, Czech Republic, Miami Beach, Florida, U.S.A., Debrecen, Hungary). Results on yield measurements can be found in **I**, **IV**, **VII** and **XIV**. In the above laboratories beam currents up to 50-150 μA were used for routine production. A vertical beam-line system used also for routine radioisotope production was designed by us and was put into operation in Debrecen, Hungary. Solid, molten (liquid) and gas target materials can be handled and irradiated (up to 50 μA) safely with this system. (See more detailed in **XIV**.) Commercial production of ^{67}Ga and ^{111}In is also going on in these centers for many years.

5. Results and conclusions

5.1 $Ti+p$ processes

The activation of ^{nat}Ti targets with protons results in the formation of several scandium and vanadium radioisotopes up to 30 MeV. In this study we have investigated only those reactions which produced ^{43}Sc , ^{44}Sc , ^{47}Sc , and ^{48}V . The radiation characteristics of these nuclides and the Q-values of the contributing reactions are summarized in Table 1 of **III**. Our measured production cross sections are tabulated in Table 2 (for ^{48}V up to 30 MeV) and in Table 3 (for ^{43}Sc , ^{44}Sc and ^{47}Sc up to 18 MeV) of **III**. These cross sections are also presented in the appropriate Figures of **III** in comparison with the previously published data.

5.1.1 Study of $Ti+p$ reactions

$^{46}Ti(p,\alpha)^{43}Sc$ reaction: Our measurement resulted in 27 cross section values from 9 to 18 MeV. The excitation function curve of this reaction seems to reach its maximum at about 17 MeV (about 3.8 mb). The presented results show good agreement with the values of Michel *et al.*(1978a) in the comparable energy region (from 13 to 18 MeV). Our work has also extended their results to 9 MeV with 17 new cross section values that were published by us for the first time.

$^{47}Ti(p,\alpha)^{44}Sc$ reaction: The bombardment of titanium with protons populates two isomeric states of ^{44}Sc . Our systematic investigation resulted in 21 and 20 cross section values for the meta and the ground states, respectively, from 9 to 18 MeV. For the cross section ratios 20 values was also calculated by us. The excitation function curve of the meta state increases over the whole investigated energy region while the curve of the ground state seems to show a maximum at about 17 MeV (about 4.2 mb). Since in the studies of Michel *et al.*(1978a, 1980) cross sections both for the meta and the ground states were published only above 13 MeV, our values (11, 10 and 10 for the meta, ground and meta/ground ratios, respectively) are completely new below 13 MeV. The presented values both for meta and ground states show fairly good agreement with the results of Michel *et al.*(1978a, 1980). However, in the case of the cross section ratios, an energy shift of about 0.5-1 MeV can be seen in the energy scale.

$^{48}Ti(p,2p)^{47}Sc$ reaction: We have measured 23 cross section values for this reaction up to 18 MeV. The excitation function curve of this reaction increases over the whole investigated energy region. Surveying the information on the excitation function of this (p,2p) reaction, we found 3 earlier works. They reported values only above 10 MeV, therefore our 3 values below this energy are completely new. Our excitation function curve shows an energy shift of about 1 MeV in comparison with the results of Michel *et al.*(1978a, 1980). The only data point of Brodzinski *et al.*(1971) at 15 MeV is much lower than the value of our work and those of Michel *et al.*(1978a, 1980).

$^{nat}Ti(p,x)^{48}V$ process: The ^{48}V radioisotope could be produced through four independent reaction channels up to 30 MeV. Although several authors have reported cross section values for the $^{nat}Ti(p,x)^{48}V$ process prior to our measurement, we performed the first systematic study from the threshold of this process up to 30 MeV (69 cross section values). The excitation function curve shows a maximum of about 380 mb at about 12.6 MeV. The results of the present investigation in general show good agreement with the values of the previous works (Brodzinski *et al.*,(1971); Michel *et al.*(1978a, 1980); Tanaka *et al.* (1959).

5.1.2 Application of the results

A) Monitoring of proton beam: From the point of view of their potential use as monitor reactions only the $^{nat}\text{Ti}(p,x)^{48}\text{V}$ process has practical significance in the investigated energy region. (The other reactions have very low cross section values up to 30 MeV.) Our detailed critical review of the results of the previous works showed that this process seems to be well measured and its cross section data base (including our recent values) can be used for proton beam monitoring in different applications (see more detailed in **III**.) An evaluation work to create ‘recommended’ cross section values for this process are in progress by us.

B) Other applications: The cross section results on $^{46}\text{Ti}(p,\alpha)^{43}\text{Sc}$, $^{47}\text{Ti}(p,\alpha)^{44}\text{Sc}$ and $^{48}\text{Ti}(p,2p)^{47}\text{Sc}$ reactions could serve as reliable experimental data bases for different model calculations. These values also can be used for example to estimate cosmogenic radionuclide production.

5.2 Ni+p processes

The activation of natural nickel with protons results in the formation of several cobalt, nickel and copper radioisotopes up to 44 MeV. We have investigated in this work only those nuclear processes that produced the following radionuclides: ^{55}Co , ^{56}Co , ^{57}Co , ^{58}Co , ^{61}Co , ^{57}Ni , ^{60}Cu , ^{61}Cu and ^{64}Cu . The decay characteristics of ^{55}Co , ^{56}Co and ^{57}Co and the Q-values of the contributing reactions are summarized in Table 1 of **II** while these data for ^{58}Co and ^{61}Co can be found in **VI** and **X**, respectively. In the case of ^{57}Ni , ^{60}Cu , ^{61}Cu and ^{64}Cu the above data are listed in Table 1 of **II** (for nickel) and Table 2 of **V** (for coppers). Our measured cross sections of nuclear processes leading to the formation of the above radioisotopes are tabulated in Table 2 of **II** (for ^{55}Co , ^{56}Co and ^{57}Co up to 30 MeV), Table 1 of **VI** (for ^{58}Co up to 19 MeV) Table 1 of **X** (for ^{61}Co up to 19 MeV), Table 2 of **II** (for ^{57}Ni up to 30 MeV), Table 1 of **XI** (for ^{57}Ni up to 44 MeV), Table 3 of **V** (for ^{60}Cu and ^{61}Cu up to 19 MeV) and Table 4 of **V** (for ^{64}Cu up to 19 MeV). The experimental cross sections are also presented in the appropriate Figures of **II** (for ^{55}Co , ^{56}Co , ^{57}Co and ^{57}Ni), **V** (for ^{60}Cu , ^{61}Cu and ^{64}Cu), **VI** (for ^{58}Co), **X** (for ^{61}Co) and **XI** (for ^{57}Ni) in comparison with the data reported earlier.

5.2.1 Study of Ni+p reactions

$^{nat}\text{Ni}(p,x)^{55}\text{Co}$ process: ^{55}Co is formed through four independent reaction channels on natural Ni below 30 MeV. We have performed a detailed study of these processes up to 30 MeV (54 cross section values). The excitation function curve of this process reaches its maximum of about 23 mb at about 17 MeV. The results of the present investigation are in good agreement with the values of the three previous works (Kaufman 1960; Michel *et al.*,1978b and Tanaka *et al.*,1972), however, the excitation function curve of Tanaka *et al.*(1972) near the threshold shows a small energy shift (about 0.5 MeV) in comparison with our results.

$^{nat}\text{Ni}(p,x)^{56}\text{Co}$ process: ^{56}Co can be produced via two direct reactions and one cumulative process on natural Ni up to 30 MeV. We have measured 26 cross section values in this energy region. The excitation function curve of this process does not reach its maximum up to 30 MeV. In the case of this process three authors have reported cross sections prior to our study. The results of the present investigation show good agreement with the values of the previous works (Aleksandrov *et al.*,1987; Michel *et al.*,1978b and Tanaka *et al.*,1972), except for the results of Aleksandrov *et al.*(1987) who obtained significantly lower value at 30 MeV.

$^{nat}\text{Ni}(p,x)^{57}\text{Co}$ process: Five direct reactions and a cumulative one contribute to the formation of ^{57}Co on natural Ni over the energy region studied. The result of our detailed study was 40 new cross section values up to 30 MeV. The excitation function curve shows a peak at about 24 MeV ($\sigma_{\text{max}}=450$ mb). Three measurements have been reported for this process in the literature through 1996. The results of the present investigation show close agreement with the values of these works (Kaufman 1960; Michel *et al.*, 1978b and Tanaka *et al.*, 1972).

$^{61}\text{Ni}(p,\alpha)^{58}\text{Co}$ reaction: The bombardment of enriched ^{61}Ni with protons populates two isomeric states of ^{58}Co . Our systematic investigation on highly enriched nickel resulted in 14 and 13 cross section values for the meta and the meta+ground states, respectively, from 5 to 19 MeV. For the m/m+g ratios 13 values was also calculated by us. The excitation function curves of both the meta and meta+ground states reach their maximum at about 15 MeV ($\sigma_{\text{max}}=35$ and 58 mb, respectively). As far as we know, our measurement describes the first systematic study of this (p, α) process. Up till now only one work (Tanaka *et al.*, 1972) has published values for the total cross sections in the investigated energy region, but they used natural target. Although those data are somewhat higher than our values, taking into account the error limits the agreement is good. Cross section data for $^{61}\text{Ni}(p,\alpha)^{58\text{m}}\text{Co}$ reaction were reported for the first time by us.

$^{64}\text{Ni}(p,\alpha)^{61}\text{Co}$ reaction: 24 cross section values was measured for this reaction from 5 to 19 MeV. The excitation function curve of the reaction shows a peak at about 16.5 MeV (about 18.7 mb). As far as we know, our measurement not only described the first systematic study of this process on enriched ^{64}Ni target, but also reported cross section values for the first time in the literature.

$^{nat}\text{Ni}(p,x)^{57}\text{Ni}$ process: Six reactions contribute to the formation of ^{57}Ni in the case of proton bombardment of natural nickel below 44 MeV: $^{58}\text{Ni}(p,pn)^{57}\text{Ni}$, $^{58}\text{Ni}(p,2n)^{57}\text{Cu}\rightarrow^{57}\text{Ni}$, $^{58}\text{Ni}(n,2n)$, $^{60}\text{Ni}(p,p3n)$, $^{60}\text{Ni}(p,4n)\rightarrow^{57}\text{Ni}$ and $^{60}\text{Ni}(n,4n)$. The neutrons responsible for the $^{58}\text{Ni}(n,2n)$ and $^{60}\text{Ni}(n,4n)$ reactions are coming from different (p,xn) processes induced in the Ni itself as well as from the target holder unit, other materials in the target stack and from the aperture in front of the target stack. Our systematic study resulted in 142 cross section values for this process. Taking into account the isotopic composition of the natural nickel and the type of the contributing reactions, the excitation function curve of this process shows only one peak at about 26 MeV. The maximum cross section value is about 175 mb. A survey of the information on cross sections of the $^{nat}\text{Ni}(p,x)^{57}\text{Ni}$ process yielded 18 additional investigations prior to our study. (Aleksandrov *et al.*, 1987; Barrandon *et al.*, 1975; Bodemann *et al.*, 1995; Brinkman *et al.*, 1977; Brinkman *et al.*, 1979; Cohen *et al.*, 1955; Ewart and Blann 1964; Furukawa *et al.*, 1990; Haasbroek *et al.* 1976; Kaufman 1960; Levkovski 1991; Michel *et al.*, 1978b; Michel *et al.*, 1980; Michel *et al.*, 1996; Piel *et al.* 1992; Stück 1983; Tanaka *et al.* 1972; and Zhuravlev *et al.* 1984). All the published excitation functions were measured on ^{nat}Ni except in the measurements of Levkovski (1991) and Tanaka *et al.* (1972). Although the irradiation energies varied between 25 and 100 MeV, all the excitation functions below 45 MeV show very good agreement with each other, except for the measurements of Ewart and Blann (1964), Haasbroek *et al.* (1976) and for the single value of Zhuravlev *et al.* (1984) at 22 MeV. To check the contribution of the neutron induced reactions to the cross section values of this (p,xn) process in a given target arrangement, it is necessary to put some Ni foils in a place (into the stack) where the energy of the beam entering those foils is lower than the threshold energy of the (p,xn) process. In some separate experiments (see more detailed in **XI**) an ‘effective’ production cross section (related to the incident total proton flux) was calculated by us to allow a comparison with the values found in charged particle activation. Our results were lower than 0.02 mb for the 26.7 MeV proton beam while at 42.9 MeV we found 0.0002 mb. Since in a stacked foil experiment this neutron contribution will be nearly uniform for all the foils in a single experiment, a worst case estimation will be obtained by comparing the above

values with the lowest real charged particle cross section. According to our investigations we can state that the interference of the neutron induced processes is generally <0.1%, for all possible experimental set ups. (See also in Hermanne *et al.*,1997.)

$^{61}\text{Ni}(p,n)^{60}\text{Cu}$ reaction: The excitation function of this reaction was measured by us for the first time below 19 MeV (9 cross section values). Due to the relatively large amount of ^{60}Ni in our target material the contribution of the $^{60}\text{Ni}(p,n)^{60}\text{Cu}$ reaction was not negligible near the threshold. For subtracting the contribution of this process we used the data of Piel *et al.*(1992). The excitation function curve of this reaction increases over the whole investigated energy region.

$^{61}\text{Ni}(p,n)^{61}\text{Cu}$ reaction: Our measurements resulted in 38 cross section values up to 19 MeV. Surveying the available information on the excitation function of the $^{61}\text{Ni}(p,n)^{61}\text{Cu}$ reaction, we found 8 earlier works. It is worth pointing out that prior to our investigation only in one work enriched target material was used (Jonson *et al.*, 1960) for measurements below 5.5 MeV. Our excitation function curve shows a maximum at about 12 MeV ($\sigma_{\text{max}} = 430$ mb). Till the onset of the $^{62}\text{Ni}(p,2n)^{61}\text{Cu}$ reaction our values show good agreement with the data of Piel *et al.*(1992), Michel *et al.*(1978b) and Blosser and Handley (1955). The results of Tanaka and Furukawa (1959) and Barrandon *et al.*(1975) are significantly higher than our values. Furthermore, the energy scale of those investigations seems to be shifted to lower energy region by about 1.5 and 1.8 MeV, respectively, in comparison to our measurements. Two other reports (Blaser *et al.*, 1951 and Johnson *et al.*, 1960) dealing with measurements in the low energy region up to 6.7 MeV also show energy shifts of about 0.8 MeV.

$^{64}\text{Ni}(p,n)^{64}\text{Cu}$ reaction: We have performed a detailed study of this reaction up to 19 MeV on highly enriched target (23 cross sections). Our excitation function curve shows a peak at about 10.5 MeV (about 720 mb). For the $^{64}\text{Ni}(p,n)^{64}\text{Cu}$ reaction four measurements have been reported earlier. All of them used natural Ni targets. The values of Blaser *et al.*(1951), Tanaka and Furukawa (1959) and Tanaka *et al.* (1972) are comparable to our values, however, with a little higher maximum in the case of the last two authors. The only data point of Blosser and Handley (1955) at 12 MeV is very high. Our measurements thus described the first accurate study.

5.2.2 Application of the results

A) *Radioisotope production:* Among the radioisotopes produced by proton bombardment of nickel, copper radionuclides have already found application in PET centers. In our study (V) we investigated the practical production possibility of ^{61}Cu and ^{64}Cu using highly enriched target materials and a low energy accelerator. On the bases of the above cross section measurements we have calculated the integral thick target yields of the $^{61}\text{Ni}(p,n)$, $^{61}\text{Ni}(p,2n)$ and $^{64}\text{Ni}(p,n)$ reactions. In this region the optimum energy range for the production of both ^{61}Cu and ^{64}Cu was found to be 12→9 MeV. The calculated thick target yield of ^{61}Cu amounted to 17.5 mCi/μAh. For production of ^{64}Cu the yield was 6.71 mCi/μAh. We had to calculate the yield of the $^{61}\text{Ni}(p,2n)^{60}\text{Cu}$ reaction because this nuclide is the major contamination during the production of ^{61}Cu . Recently, the ^{60}Cu was also suggested to use as a PET isotope (Bass *et al.*,1997) but according to our present study this isotope can be produced only at higher energy cyclotron (over 20 MeV) using this reaction.

We have developed a simple anion-exchange chromatographic method for the separation of radiocopper from the activated Ni target. We have also elaborated a technique for recovery of the very expensive enriched target materials. These methods are described in V.

B) Monitoring of proton beam: On the bases of our detailed critical review of the numerous literature results (see more detailed in **II** and **XI**) and our new cross section measurements, the following processes seems to be well measured and can be used for proton beam monitoring purpose: $^{nat}\text{Ni}(p,x)^{55}\text{Co}$ (up to 30 MeV), $^{nat}\text{Ni}(p,x)^{56}\text{Co}$ (up to 30 MeV), $^{nat}\text{Ni}(p,x)^{57}\text{Co}$ (up to 30 MeV) and $^{nat}\text{Ni}(p,x)^{57}\text{Ni}$ (up to 45 MeV). It is worth to mention that the $^{nat}\text{Ni}(p,x)^{57}\text{Ni}$ process was recently added to the list of the suggested proton monitors up to 100 MeV by the Nuclear Data Section of IAEA, Vienna, Austria. Further detailed evaluation as well as experimental works are in progress for this reaction.

C) Model calculations: In the case of proton induced reactions on ^{61}Ni and ^{64}Ni we have also performed statistical model calculations taking into account preequilibrium effects to describe the strong (p,n) [or (p,2n)] and the weaker (p, α) reaction channels. These calculations were performed with codes STAPRE (Uhl and Strohmaier, 1976) for $^{61}\text{Ni}+p$ reactions and with MAURINA (Marinov *et al.*, 1984) and DWUCK (Uhl, 1995) for $^{64}\text{Ni}+p$ reactions. The experimental excitation functions were described well by the calculations over the whole investigated energy range for the $^{61}\text{Ni}(p,n)^{61}\text{Cu}$, $^{61}\text{Ni}(p,2n)^{60}\text{Cu}$, $^{61}\text{Ni}(p,\alpha)^{58m}\text{Co}$ and $^{61}\text{Ni}(p,\alpha)^{58m+g}\text{Co}$ reactions. The isomeric cross section ratio for the isomeric pair $^{58}\text{Co}^{m,g}$ formed in the (p, α) process was also reproduced well by this model. These results can be seen in Figures 1-5 of **VI**. In the case of the $^{64}\text{Ni}(p,n)^{64}\text{Cu}$ and $^{64}\text{Ni}(p,\alpha)^{61}\text{Co}$ reactions the strong channel was described well with the model, but an acceptable agreement between the experimental and theoretical cross section data for the $^{64}\text{Ni}(p,\alpha)^{61}\text{Co}$ reaction was available only if the influence of a direct three-nucleon pickup process was also taken into account. Our results can be found in **X**.

5.3 Zn+p processes

Proton activation of enriched zinc targets (^{66}Zn , ^{67}Zn and ^{68}Zn) produces four radioactive gallium (^{65}Ga , ^{66}Ga , ^{67}Ga and ^{68}Ga) and one zinc (^{65}Zn) radioisotopes up to 30 MeV. The ^{65}Ga decays completely to ^{65}Zn . The radiation characteristics of these nuclides are summarized in Table 2 of **I** (and Table 1 of **IX**). Our measured cross section values are tabulated in Tables 3-5 of **I** (for ^{65}Zn , ^{66}Ga , ^{67}Ga and ^{68}Ga , up to 22 MeV) and in Table 1 (for ^{65}Zn , ^{66}Ga and ^{67}Ga up to 26 MeV) of **XII**. (See also Tables 1-3 of **XX**.) The cross sections are also presented in the appropriate Figures of **I** and **XII** (and **XX**) in comparison with the available data reported earlier.

5.3.1 Study of Zn+p reactions

$^{nat}\text{Zn}(p,x)^{65}\text{Zn}$ process: There are two proton induced nuclear reactions on ^{66}Zn that result in ^{65}Zn , namely the $^{66}\text{Zn}(p,pn)^{65}\text{Zn}$ and the $^{66}\text{Zn}(p,2n)^{65}\text{Ga}\rightarrow^{65}\text{Zn}$ reactions. In our study we have measured only the cumulative cross sections (23 values) up to 26 MeV. The excitation function curve of this process reaches its maximum over the investigated energy region at about 24 MeV ($\sigma_{\text{max}}=880$ mb). A recent survey of the information on cross sections of the $^{66}\text{Zn}(p,x)^{65}\text{Zn}$ process found three investigations prior to our measurements. (Bonardi and Birattari 1983; Hermanne 1994 and Levkovski 1991). Our cross section data show good agreement with the values of the previous works. It is worth pointing out that prior to our investigation only in one work enriched target material was used (Levkovski, 1991).

$^{66}\text{Zn}(p,n)^{66}\text{Ga}$ reaction: Our systematic investigation on highly enriched zinc resulted in 44 cross section values up to 26 MeV. The curve of the excitation function of this reaction shows a peak of about 640 mb at about 12.5 MeV. Since in the previous studies (11) only natural targets were used, (except Levkovski 1991) their values can be used for comparison only up to the threshold energy of the $^{67}\text{Zn}(p,2n)$ reaction (up to about 13 MeV). Cross sections were reported by Blaser *et al.*, (1951); Barrandon *et al.* (1975); Blosser and Handley (1955); Bonardi and Birattari (1983); Hermanne (1994); Hille *et al.* (1972); Howe (1958); Kopecky (1990); Levkovski (1991); Little and Lagunas-Solar (1983) and Nortier *et al.* (1991) through 1996. The published cross section measurements are in acceptable agreement with each other, especially in the case of the position of the cross section maximum (except the result of Little and Lagunas-Solar 1983). This maximum, however, varies in magnitude from 450 to 700 mb. It is also worth pointing out that prior to our investigation only one work reported reliable cross section values from 13 to 26 MeV (Levkovski 1991).

$^{67}\text{Zn}(p,2n)^{66}\text{Ga}$ reaction We have measured 32 cross section values for this reaction up to 26 MeV. The excitation function curve of this process seems to reach its maximum at about 21 MeV ($\sigma_{\text{max}} = 470$ mb). Although three authors have published cross section values for this reaction in the past, only the values of Levkovski (1991) can be used for comparison with our data. The published values of Hermanne (1994) and Little and Lagunas-Solar (1983) were not 'real' measured ones, because they used natural targets in their experiments and the contribution of the $^{66}\text{Zn}(p,n)$ reaction was separated only on the bases of systematics of excitation functions of the neighboring nuclei. Our data support the values of Levkovski (1991) over the whole comparable energy region.

$^{68}\text{Zn}(p,3n)^{66}\text{Ga}$ reaction: Due to the relative high threshold energy of this reaction we could measure only 6 cross section values. The excitation function curve of this (p,3n) reaction increases up to 26 MeV. In analyzing the published literature results, only two works were found that used highly enriched ^{68}Zn target (Levkovski 1991, and McGee *et al.* 1970). The problems of the results of Hermanne (1994) and Little Lagunas-Solar (1983) was already discussed above. Our cross sections show relatively good agreement with the results of Levkovski (1991). The only data point of McGee *et al.* (1970) at 30 MeV, however, seems to be very low in comparison with the data of Levkovski (1991).

$^{67}\text{Zn}(p,n)^{67}\text{Ga}$ reaction: Our systematic investigation resulted in 43 cross section values between the threshold energy of this (p,n) reaction and 26 MeV. The excitation function curve of this reaction shows a peak of about 670 mb at about 10 MeV. For the $^{67}\text{Zn}(p,n)$ reaction seven authors have presented cross section values up to 1997 (Blaser *et al.*, 1951; Barrandon *et al.* 1975; Bonardi and Birattari 1983; Hermanne 1994; Johnson *et al.* 1960; Kopecky 1990; Levkovski 1991; Little and Lagunas-Solar 1983 and Nortier *et al.* 1991). Unfortunately, the majority of the studies used natural zinc targets (except Johnson *et al.* 1960 and Levkovski 1991), therefore their values can be used for comparison only up to the threshold energy of the $^{68}\text{Zn}(p,2n)$ reaction (up to about 12 MeV). In analyzing the presented values it can be concluded that the majority of the results show good agreement with each other and with our values except the data of Barrandon *et al.* (1975), Little and Lagunas Solar (1983) and Levkovski (1991). According to the review of these data (see in IX) the results of our present investigation are the only reliable data that could be used for this (p,n) reaction above 12 MeV.

$^{68}\text{Zn}(p,2n)^{67}\text{Ga}$ reaction: We have performed a systematic study for this reaction up to 26 MeV. In the investigated energy region we have measured 39 cross section values. Our curve for this reaction shows a maximum of 660 mb at about 20 MeV. Although this reaction is the most frequently used for commercial ^{67}Ga production, only two works (Levkovski, 1991 and McGee *et al.*, 1970) were found in the literature prior to our present work that used highly enriched ^{68}Zn . There are, however, some 'production cross section' measurements for the $^{\text{nat}}\text{Zn}(p,xn)^{67}\text{Ga}$ process which results can also be used for comparison between 17 and 26 MeV (simply

multiplying the values with 100/18.8). In this energy range the contribution of the $^{67}\text{Zn}(p,n)^{67}\text{Ga}$ reaction can be neglected (<7% in this energy range) due to the low isotopic abundance of ^{67}Zn in natural zinc matrix and the magnitude of the cross section values of the $^{67}\text{Zn}(p,n)^{67}\text{Ga}$ reaction over this energy region (see the section of cross sections of the $^{67}\text{Zn}(p,n)^{67}\text{Ga}$ reaction). The influence of the $^{70}\text{Zn}(p,4n)^{67}\text{Ga}$ process to the production cross sections is also negligible because of the very low isotopic abundance of ^{70}Zn in natural zinc (0.62%). This way we could use the results of the following authors: Bonardi and Birattari (1983), Hermanne (1994), Kopecký (1990) and Nortier *et al.*(1991). On the bases of the usable results (including ours) it can be seen that the majority of the cross section measurements show acceptable agreement with each other for the (p,2n) reaction except the results of the Levkovski (1991), Little and Lagunas-Solar (1983) and McGee *et al.*(1970). Similar to the case of the above reaction our present investigation is the only one that has reliable cross section values close to the onset of the (p,2n) reaction (up to 17 MeV).

$^{68}\text{Zn}(p,n)^{68}\text{Ga}$: Our measurement resulted in 10 cross section values from 11 to 22 MeV. The excitation function of this reaction reaches its maximum of about 900 mb at about 12 MeV. A search for cross section values yielded ten works in the literature. (Barrandon *et al.*1975; Blaser *et al.*1951; Esat *et al.*1981; Hermanne 1994, Hille *et al.*1972; Howe 1958; Johnson *et al.*1960; Levkovski 1991; McGee *et al.*1970 and Nortier *et al.*1990). Our values show acceptable agreement with the majority of the published values (except Hermanne 1994; Levkovski 1991 and McGee *et al.*1970).

5.3.2 Application of the results

A) *Evaluated cross sections and thick target yield data bases:* ^{67}Ga is one of the most frequently employed SPECT radioisotope. Recently, ^{66}Ga and ^{68}Ga have also found application in PET investigations (see more detailed in **IX**). The evaluation as well as the ‘standardization’ of the cross sections and yields of their most frequently employed production reactions were already suggested by the Nuclear Data Section of the IAEA (Lambrecht, 1988) We have decided therefore to create recommended data bases for the $^{\text{nat}}\text{Zn}(p,xn)^{65}\text{Zn}$, $^{66}\text{Zn}(p,n)^{66}\text{Ga}$, $^{67}\text{Zn}(p,n)^{67}\text{Ga}$, $^{67}\text{Zn}(p,2n)^{66}\text{Ga}$, $^{68}\text{Zn}(p,n)^{68}\text{Ga}$, $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$ and $^{68}\text{Zn}(p,3n)^{66}\text{Ga}$ reactions up to 30 MeV. In the first step of this study we have compiled and partly evaluated those cross section measurement and thick target yield calculations (and measurements) which were published in the literature up to 1994. The results of this work are collected of **IX**. After a critical review of the available (till 1996) experimental results, the acceptable data sets were evaluated using the earlier mentioned cubic spine method. (See more detailed in 4.5) The point of views of the selections of the data as well as the recommended cross sections and the calculated integral thick target yield values are collected (in 1 MeV increments) in Table 3 of **XII** (and partly in **XIX**). In the work of **XII** we have also compared our yield calculations with the measured yields (measured both on natural or enriched targets) The majority of the presented values verified the reliability of our calculations.

B) *Routine radioisotope production:* On the basis of our cross section measurements and the recommended cross section and integral thick target yield data bases, we have evaluated the optimal production conditions for ^{66}Ga , ^{67}Ga and ^{68}Ga . Optimum production energy ranges were calculated for the following reactions: $^{66}\text{Zn}(p,n)^{66}\text{Ga}$ [(energy range: 18→6 MeV, yield: 25.6 mCi/μAh)], $^{67}\text{Zn}(p,2n)^{66}\text{Ga}$ [energy range: 30→14 MeV, yield: 38 mCi/μAh], $^{68}\text{Zn}(p,3n)^{66}\text{Ga}$ [energy range: 30→25 MeV, yield: 1.7 mCi/μAh], $^{67}\text{Zn}(p,n)^{67}\text{Ga}$ [energy range: 18→5 MeV, yield: 2.8 mCi/μAh], $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$ [energy range: 30→14 MeV, yield:

7.2 mCi/ μ Ah], $^{68}\text{Zn}(p,n)^{68}\text{Ga}$ [energy range: 18→4 MeV, yield: 284 mCi/ μ Ah]. At the MGC-20E cyclotron Debrecen, Hungary, the $^{67}\text{Zn}(p,n)$ reaction was employed for production of ^{67}Ga ($E_p=14$ MeV for routine production) for many years (XIV), while in Miami Beach, Florida, the $^{68}\text{Zn}(p,2n)$ reaction was used for high scale production at the CS-30 accelerator ($E_p=26$ MeV for routine production) (IV). For the separation of radiogallium from the zinc matrix (^{67}Zn), an ionexchange separation method was also developed and used in Debrecen, Hungary. Thanks to the efforts made in the field of improving efficiency of the targetry and the chemistry used, it was possible to reach almost 90% of the yields expected from our data bases. Details of these works can be found in I, IV and XIV.

C) *Model calculations:* The excitation functions for the $^{66}\text{Zn}(p,n)^{66}\text{Ga}$, $^{67}\text{Zn}(p,2n)^{66}\text{Ga}$, $^{66}\text{Zn}(p,n)^{67}\text{Ga}$, $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$ and $^{68}\text{Zn}(p,n)^{68}\text{Ga}$ reactions were calculated by us using the model code STAPRE (Uhl and Strohmaier, 1976) which is a statistical model that takes into account preequilibrium effects to describe the strong (p,n) and (p,2n) reaction channels. In general the experimental excitation functions were described well by these calculations over the whole investigated energy range (up to 25 MeV). Although the shape of the excitation curves were reproduced well, some deviations in absolute cross sections were always found (10-30%).

D) *Other applications:* For studying wear and/or corrosion processes, especially in industry, the thin layer activation technique (TLA) is an effective method. By measuring the decrease of radioactivity during the removal of the surface layer the material loss can be estimated very precisely. These applications, however, require adequate knowledge of the relative distribution of the radioactivity as a function of depth (calibration curve). This calibration curve could be determined by grinding away the activated layer step by step and measuring the remaining activity or by calculation using nuclear data. It is obvious that well measured cross sections play a key role in this calculation. Since zinc alloys and compounds are widely used in many fields of industry (cf. Wagner *et al.*, 1981), it could be important to investigate the feasibility of this technique for those materials which contain zinc. The ^{65}Zn and $^{66,67,68}\text{Ga}$ can be produced via Zn+p reactions with high activation cross sections. These excitation functions are now - thanks to our efforts - well measured and evaluated, therefore they seem to be useful for the mentioned industrial purposes. On the other hand the gallium radioisotopes, especially the ^{68}Ga , have relatively short half-lives which limit their applications to relatively quick removal processes. For calculating the activated depth for TLA analysis and for radiation dose estimations we have computed the total activity versus penetration depth and the distribution of activity as a function of the penetration depth for ^{65}Zn , ^{66}Ga and ^{67}Ga which are produced by different incident proton energies from 5 to 20 MeV (in 1 MeV increments) in a material which contains homogeneously distributed natural zinc. These calculated curves are shown in the appropriate Figures of XII. For these calculations our 'recommended' excitation functions were employed. Unfortunately, until now we have found only one experimental calibration curve in the literature (Herkert, 1975) which to compare our calculations. The comparison of the two calibration curves (at $E_p=25.196$ MeV) measured by Herkert (1975) and calculated by us show excellent agreement with each other. This 'integral test' verifies the reliability of evaluated cross section data for this reaction. We believe that, based on this result, the other calculations (i.e. for $^{66,67}\text{Ga}$) would also be useful for practical purposes.

5.4 Cd+p processes

The activation of highly enriched ^{111}Cd and ^{112}Cd with protons up to 30 MeV resulted in the formation of indium radioisotopes of ^{109}In , ^{110}In , ^{111}In and ^{110}In , ^{111}In , respectively. The decay characteristics of these nuclides and the Q-values of the contributing reactions are summarized in Table 2 of **VII**. Our measured cross sections of those reactions which produced the above isotopes are tabulated in Table 1 and 2 of **VII**. These cross sections are also presented in the appropriate Figures of **VII** (and **XVII**) in comparison with earlier reported data.

5.4.1 Study of Cd+p cross sections

$^{111}\text{Cd}(p,3n)^{109}\text{In}$ reaction: Proton bombardment of ^{111}Cd with protons populates three isomeric levels of ^{109}In . The short-lived meta states decays completely to the ground state. The total production cross sections were measured in this study after these isomers had decayed to the ground state. We have evaluated 7 cross section values from 20 to 30 MeV. The excitation function curve of this reaction increases over the whole investigated energy region. A comparison with the data of Marten *et al.*(1985) showed good agreement between the two data sets, but our values are systematically lower than those obtained by them. Our three cross section values below 20 MeV were reported for the first time.

$^{111}\text{Cd}(p,2n)^{110}\text{In}$ reaction: Two longer lived isomeric states of ^{110}In are formed via this reaction. Our systematic investigation on highly enriched cadmium resulted in 17 cross section values both for the meta and the ground states from 13 to 30 MeV. For the cross section ratios 17 values was also calculated by us. The excitation function curves of both the meta and ground states reach their maximum in the investigated energy region. For the meta state the maximum can be found at about 23 MeV ($\sigma_{\text{max}}=300$) while these data for the ground state are $E_{\text{peak}}=20$ MeV and $\sigma_{\text{max}}=300$ mb. Our data show good agreement with the values of Marten *et al.*(1985) and Otozai *et al.*(1966) for both the investigated states. As far as we know, our two data point below 13.5 MeV were measured for the first time for the ground state.

$^{111}\text{Cd}(p,n)^{111}\text{In}$ reaction: The bombardment of ^{111}Cd with protons populates two isomeric levels of ^{111}In . The short-lived meta states ($T_{1/2}=7.6$ min) decays completely to the ground state. Our systematic study has yielded 31 cross section values for the meta+ground states from 3.8 to 30 MeV. The excitation function curve of the total cross section shows a peak ($\sigma_{\text{max}}=810$ mb) at about 11.8 MeV. In this case seven authors have reported cross sections through 1996. (Blaser *et al.*,1951; Marten *et al.*,1985; Nortier *et al.*,1990; Otozai *et al.*,1966; Skakun *et al.*,1975; Skakun *et al.*, 1979 and Wing *et al.*,1962). We got very good agreement with all the earlier measured data both on energy scale and cross section values.

$^{112}\text{Cd}(p,3n)^{110}\text{In}$ reaction: Similar to the (p,n) reaction on ^{111}Cd , the products of the $^{112}\text{Cd}(p,3n)$ reaction are the same isomeric states of this indium radioisotope: $^{110\text{m}}\text{In}$ and $^{110\text{g}}\text{In}$. Their excitation curves, however, are increasing from the threshold up to 30 MeV. Since only one author (Otozai *et al.*,1966) presented cross sections for these reactions below 30 (only two data points for each process), our study was the first systematic investigation in this field which resulted in 8 cross section values for both processes (and for the cross section ratios) from 23 to 30 MeV. Although it was difficult to compare these results with the values of Otozai *et al.*(1966), the values seem to support each other.

$^{112}\text{Cd}(p,2n)^{111}\text{In}$ reaction: Both the meta and the ground states of ^{111}In are formed during proton bombardment of ^{112}Cd . We have measured 25 cross sections for the total ^{111}In production. Our excitation function curve shows a maximum of about 1000 mb at about 19 MeV. The values of the two previous investigations published by Otozai *et al.*(1966) and

Skakun *et al.*(1975) agree well with our values, however, our data close to the peak are a little scattered.

5.4.2 Application of the results

A) *Evaluated cross section/thick target yield data bases:* Since ^{111}In is also a very frequently used radioisotope of nuclear medicine, the evaluation as well as the ‘standardization’ of the cross sections and yields of its most frequently employed production reactions were suggested by the Nuclear Data Section of the IAEA, Vienna, Austria (Lambrech, 1988). We have performed such a work for $^{111}\text{Cd}(p,n)$ and $^{112}\text{Cd}(p,2n)$ reactions for the first time. After a critical review of the available experimental results the acceptable data sets were evaluated using the earlier mentioned cubic spine method. (See more detailed in 4.5) These recommended cross sections and integral thick target yield values are (in 1 MeV increments) collected in Table 1 and 2 of **VIII**.

B) *Radioisotope production:* We have also investigated the practical production possibility of ^{111}In using highly enriched target materials and low and middle energy cyclotrons (**VII**). On the bases of the above mentioned evaluated thick target yield data base (**VIII**) and the calculations presented in **VII**, the optimum production energy range for the $^{111}\text{Cd}(p,n)$ reaction was found to be 18→7 MeV, while this data for the $^{112}\text{Cd}(p,n)$ reaction was 30→15 MeV. The calculated thick target yields amounted to 2.0 and 7.6 mCi/μAh, respectively. We have also evaluated the yields of the $^{111}\text{Cd}(p,2n)^{110}\text{In}$, $^{112}\text{Cd}(p,3n)^{110}\text{In}$ and $^{111}\text{Cd}(p,n)^{109}\text{In}$ reactions because these nuclides are the major contaminations during the production of ^{111}In . Using these yield values one can calculate the contamination level in the case of those targets whose enrichment level is less than 100%. The measured thick target yields verified the reliability of the yields which were suggested by us as recommended ones in **VIII**. Details on the routine production can also be found in **VII**.

C) *Other application:* The measured cross sections (and recommended values) as well as the cross section ratios can be used for testing the predicting capabilities of the different model codes.

5.5 Au+p processes

Only a few gold and mercury radioisotopes are formed during proton bombardment of $^{\text{nat}}\text{Au}$ targets with protons up to 30 MeV. We have investigated in detail in **XIII** only those reactions which resulted in the production of ^{196}Au and the ^{197}Hg . The decay mode of these radioisotopes are discussed in detail in **XIII**. Our measured cross sections of the $\text{Au}(p,pn)$ and $\text{Au}(p,n)$ nuclear reactions up to 18 MeV are tabulated in Table 1 (for ^{196}Au) and Table 2 (for $^{197\text{m}}\text{Hg}$), respectively of **XIII**. These cross sections are also presented in the appropriate Figures of **XIII** in comparison with earlier reported data.

5.5.1 Study of Au+p cross sections

$^{197}\text{Au}(p,pn)^{196}\text{Au}$ reaction: Proton bombardment of gold populates three metastable states of ^{196}Au . The two metastable states decays to the ground state. We have measured the total cross sections (14 cross section values) for this reaction below 18 MeV. The excitation function

curve of the (p,pn) reaction starts above 12 MeV and increases over the whole investigated energy region. In the case of the above reaction there is, however, an other contributing process namely the $^{197}\text{Au}(n,2n)$ which results the same residual. The neutrons responsible for this reaction are coming from the different (p,xn) processes induced in the Au itself as well as from the targets, target holder unit and from the aperture in front of the target stack. To check the contribution of this process to the cross section values of the (p,pn) reaction in a given target arrangement, it is necessary to put some gold foils in a place (into the stack) where the energy of the beam entering those foils is lower than the threshold energy of the (p,pn) reaction. In our case the contribution of these processes resulted an average 0.045 mb which was subtracted from the final results. In accordance with our estimation and some special measurements, the contribution of this neutron induced process, even in the worst case, is small and negligible. Surveying the available information on the excitation function of the (p,pn) reaction up to 30 MeV, we found 5 earlier works. The results of the present investigation show very good agreement with the values of the previous works (Birattari and Bonardi 1980; Nagame *et al.*, 1990; Sosniak 1958) in the comparable energy regions.

$^{197}\text{Au}(p,n)^{197}\text{Hg}$ reaction: The proton activation of gold target produce to isomeric states of ^{197}Hg . The meta state decays almost completely to the ground state. Our measurements for the meta state resulted in 70 new cross section values from 5 to 18 MeV. The excitation function curve of this reaction seems to reach its maximum at about 12 MeV (about 39 mb). Prior to our measurement six authors have presented cross sections for the $^{197}\text{Au}(p,n)^{197\text{m}}\text{Hg}$ nuclear reaction through 1997 (Alderliesten *et al.* 1975; Gritsina *et al.* 1963; Hansen *et al.* 1962; Tilbury and Yaffe 1963; and Vandenbosch and Huizenga 1960). Below 6 MeV, however, only we have reported cross section values (3 values) As it can be concluded the reported cross sections show deviations both in the values of the cross sections and their energy scales. No other measurements was found that supported totally the results of our detailed study.

5.5.2 Application of the results

A) *Evaluated cross section data base.* On the bases of the available experimental works only the $^{197}\text{Au}(p,n)^{196}\text{Au}$ nuclear reaction seems to be well measured up to 30 MeV. Using our results and the cross section values of the above authors (except the results of Tilbury and Yaffe (1963) it was possible and worth-while to fit the cross section data sets of this (p,pn) reaction up to 30 MeV. The fitting procedure was the same as in the case of the Zn+p, and Cd+p processes. The results are collected in 1 MeV increments in Table 1 (Fig.1) of **XIII**.

B) *Proton beam monitoring:* As it was already mentioned above the $^{197}\text{Au}(p,n)^{196}\text{Au}$ nuclear reaction can be used up to 30 MeV for monitoring the proton beam performance. Till some new precise independent measurement below and above 18 MeV for the $^{197}\text{Au}(p,n)^{197\text{m}}\text{Hg}$, we suggest to use for monitor purposes the results of the present work since the same technique was employed to measure both the above reactions and the cross section values for the (p,pn) reaction show very good agreement with results of other investigators.

6. Summary

The aim of this study was to investigate the production possibility of some medically important radioisotopes of copper ($^{61,64}\text{Cu}$), gallium ($^{66,67,68}\text{Ga}$) and indium (^{111}In) at low and/or medium energy ($E_p \leq 30$ MeV) cyclotrons. Although several factors contribute to the optimization of the production of medical radioisotopes, one of the most important factors in this process is the knowledge of the excitation function of the reaction selected for production purpose. On the base of well measured cross sections the expectable maximum yield for the required radioisotope and the contaminating ones, the energy regions used for production etc. can be easily calculated. Our preliminary reviews of the nuclear data relevant to production of the above radioisotopes showed that the status of the cross sections and yield data of the selected reactions was not satisfactory. (In some cases these values were contradicting or they were missing in some energy regions).

In this work we have focused our efforts mainly on the above problems of these data. To have reliable nuclear data relevant to production of the required radioisotopes we have measured new cross section values as well as we have critically compared and evaluated of the available experimental results.

In the frame of this thesis we have measured the excitation function of those (p,n) and (p,2n) nuclear reactions which resulted in the production of the above mentioned radioisotopes on enriched nickel, zinc and cadmium targets. Besides these production reactions we have also investigated those processes that resulted in the most important contaminating radionuclides. In our experiments highly enriched materials electroplated onto thin metal backing foils (Ni, Au) were used. Some of the nuclear reactions induced in these backings (and in Ti foils) were also measured to investigate their usefulness for proton beam monitoring up to 30 MeV. Additionally, we have also investigated the possibilities of employing the improved data bases in fields other than routine radioisotope production. In the case of some Zn+p and Ni+p reactions model calculations were also performed and the calculated excitation functions were compared with the experimentally obtained ones to check the prediction capabilities of some model codes.

The results of this study can be summarized as follows:

1) We have measured in detail the excitation functions of eight reactions which resulted in the formation of $^{61,64}\text{Cu}$, $^{66,67,68}\text{Ga}$ and ^{111}In at low and/or medium energy accelerators ($^{61}\text{Ni}(p,n)^{61}\text{Cu}$ (up to 19 MeV), $^{64}\text{Ni}(p,n)^{64}\text{Cu}$ (up to 19 MeV), $^{66}\text{Zn}(p,n)^{66}\text{Ga}$ (up to 26 MeV), $^{67}\text{Zn}(p,n)^{67}\text{Ga}$ (up to 26 MeV), $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$ (up to 26 MeV), $^{68}\text{Zn}(p,n)^{68}\text{Ga}$ (up to 26 MeV), $^{111}\text{Cd}(p,n)^{111}\text{In}$ (up to 30 MeV) and $^{112}\text{Cd}(p,2n)^{111}\text{In}$ (up to 30 MeV). The results of this study increased the number of the reliable data which are needed for yield calculations. On the other hand, the critical review of the previous results showed that our values in some energy regions are the only available or reliable cross sections. (For example in the cases of $^{61}\text{Ni}(p,n)^{61}\text{Cu}$, $^{66}\text{Zn}(p,n)^{66}\text{Ga}$, $^{67}\text{Zn}(p,n)^{67}\text{Ga}$ and $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$ reactions.) Our values together with the reliable earlier published cross sections can be used to calculate the expected yields with increased accuracy.

2) We have also measured the cross sections of reactions resulting the most important contaminating radioisotopes ($^{61}\text{Ni}(p,2n)^{60}\text{Cu}$, $^{67}\text{Zn}(p,2n)^{66}\text{Ga}$, $^{68}\text{Zn}(p,3n)^{66}\text{Ga}$, $^{111}\text{Cd}(p,3n)^{109}\text{In}$, $^{111}\text{Cd}(p,2n)^{110}\text{In}$ and $^{112}\text{Cd}(p,3n)^{110}\text{In}$). Our results were published in some cases for the first time in the whole (or in the part of the) investigated energy regions ($^{61}\text{Ni}(p,2n)^{60}\text{Cu}$, $^{111}\text{Cd}(p,3n)^{109}\text{In}$, $^{111}\text{Cd}(p,2n)^{110}\text{In}$). (It is worth pointing out that in the case of

the production of a radioisotope, only those longer-lived radioisotopes are considered contaminating ones which are chemically not separable from the required product(s).) Using our present data and the acceptable previous results, the selection of the optimum energy ranges for production purposes become more reliable.

3) Precise measurements of the excitation function was done in the case of the Ti+p, Ni+p and Au+p processes to check their usefulness for proton beam monitoring. On the basis of our detailed investigations 7 processes were selected and suggested for this purpose: $^{nat}\text{Ti}(p,x)^{48}\text{V}$ (up to 30 MeV), $^{nat}\text{Ni}(p,x)^{55}\text{Co}$ (up to 30 MeV), $^{nat}\text{Ni}(p,x)^{56}\text{Co}$ (up to 30 MeV), $^{nat}\text{Ni}(p,x)^{57}\text{Co}$ (up to 30 MeV), $^{nat}\text{Ni}(p,x)^{57}\text{Ni}$ (up to 44 MeV), $^{nat}\text{Au}(p,pn)^{196}\text{Au}$ (up to 30 MeV) and $^{nat}\text{Au}(p,n)^{197m}\text{Hg}$ (up to 18 MeV). We have suggested them for proton beam monitoring for the first time. These processes are already accepted by other research groups and they are frequently used in their studies.

4) During this works we could also measure some reactions on our targets which have less importance from the point of use as monitor or routine radioisotope production reactions. However, these values could increase the data base of experimentally measured reactions. In some cases these reactions were measured by us for the first time ($^{61}\text{Ni}(p,\alpha)^{58m}\text{Co}$, $^{64}\text{Ni}(p,\alpha)^{61}\text{Co}$) and/or we could extend their investigated energy regions ($^{46}\text{Ti}(p,\alpha)^{44}\text{Sc}$, $^{47}\text{Ti}(p,\alpha)^{44m,g}\text{Sc}$, $^{48}\text{Ti}(p,2p)^{47}\text{Sc}$, $^{61}\text{Ni}(p,\alpha)^{58}\text{Co}$, $^{111}\text{Cd}(p,2n)^{110g}\text{In}$, and $^{197}\text{Au}(p,n)^{197m}\text{Hg}$). The cross section values of the above reactions could be important to fields other than radioisotope production (i.e. radiation safety, accelerator technology, astrophysical calculations etc.)

5) In the case of all measured processes, compilation and critical review of the available experimental data was always performed and published together with the new results. After eliminating the systematic inconsistencies found in some contributions it was possible to select those measurements which can be used for further evaluation and for nuclear data 'standardization' works. Part of this investigation, especially for Zn+p reactions were published separately.

6) On the basis of our measurements and the reliable experimental works, we have created 'recommended' cross section data bases for $^{nat}\text{Zn}(p,x)^{65}\text{Zn}$, $^{66}\text{Zn}(p,n)^{66}\text{Ga}$, $^{67}\text{Zn}(p,2n)^{66}\text{Ga}$, $^{68}\text{Zn}(p,3n)^{66}\text{Ga}$, $^{67}\text{Zn}(p,n)^{67}\text{Ga}$, $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$, $^{68}\text{Zn}(p,n)^{68}\text{Ga}$, $^{111}\text{Cd}(p,n)^{111}\text{In}$ and $^{112}\text{Cd}(p,2n)^{111}\text{In}$ and $^{197}\text{Au}(p,pn)^{196}\text{Au}$ reactions. We have also calculated recommended yield values for those reactions which are used for routine production. Comparing our yield predictions with measured thick target yields, we could verify the reliability of our values. From practical point of view these 'standardized' data also could serve to (i) establish and to maintain international uniformity; (ii) improve the accuracy, where this would become necessary; and (iii) help other laboratories during the improvement of their radioisotope production program. These recommended data bases are already accepted by several laboratories involved in radionuclide production.

7) Optimum energy ranges for the production of $^{61,64}\text{Cu}$, $^{66,67,68}\text{Ga}$ and ^{111}In have been calculated by us using our cross section measurements and/or the recommended yield data. Production routes of ^{67}Ga (via $^{67}\text{Zn}(p,n)$ and $^{68}\text{Zn}(p,2n)$ reactions) and ^{111}In (via $^{111}\text{Cd}(p,n)$ and $^{112}\text{Cd}(p,2n)$ processes) was also elaborated using small and medium energy cyclotrons. Using our data routine and commercial production of ^{67}Ga and ^{111}In was put into practice in Debrecen, Hungary, Miami Beach, Florida, U.S.A. and Rez, Czech Republic). A vertical beam-line system used for routine radioisotope production was also designed and put into

operation in Debrecen, Hungary. Solid, molten (liquid) and gas target materials can be handled and irradiated (up to 50 μA) safely with this system.

8) Statistical model calculations taking into account precompound effects were performed by us to describe the excitation functions of the following reactions: $^{61}\text{Ni}(p,n)$, $^{61}\text{Ni}(p,2n)$, $^{61}\text{Ni}(p,\alpha)$, $^{64}\text{Ni}(p,n)$, $^{64}\text{Ni}(p,\alpha)$, $^{66}\text{Zn}(p,n)$, $^{67}\text{Zn}(p,2n)$, $^{67}\text{Zn}(p,n)$, $^{68}\text{Zn}(p,2n)$ and $^{68}\text{Zn}(p,n)$. It has been concluded that the model codes used could describe well the investigated reaction channels, however, some deviations in absolute cross sections were always found especially in the case of Zn+p reactions.

9) For the first time we have investigated the usefulness of cross section data in Thin Layer Activation technique (TLA) used for nuclear wear measurements of those materials which contain zinc. On the basis of our recommended Zn+p data bases we have suggested to use the $^{\text{nat}}\text{Zn}(p,x)^{65}\text{Zn}$ and $^{\text{nat}}\text{Zn}(p,x)^{66,67,68}\text{Zn}$ processes for the mentioned industrial purpose. We have published curves for total activity versus penetration depth and for distribution of activity as a function of penetration depth for ^{65}Zn , ^{66}Ga and ^{67}Ga (which are produced by different incident proton energies). Our calculations were compared and verified by experimentally measured calibration curve.

7. Összefoglaló

Protonok által kiváltott magreakciók hatáskeresztmetszetének mérése Ti, Ni, Zn, Cd és Au céltárgyakon a 30 MeV-ig terjedő energiatartományban, valamint ezek alkalmazása radioizotópok előállítására

Előzmények

A Magyar Tudományos Akadémia debreceni Atommagkutató Intézetében 1985-ben helyezték üzembe Magyarország első ciklotronját. Az MGC-20E típusú izokron gyorsító üzemidejének jelentős részében orvosi célú radioizotópokat állított (és azóta is állít) elő. Kezdetben a hagyományos izotópdiagnosztikai vizsgálatokhoz használható SPECT (Single Photon Emission Tomography) radioizotópok, valamint az ezekkel jelzett vegyületek (radiofarmakonok) termelésével foglalkoztak (pl. ^{67}Ga , ^{111}In , ^{123}I). A jelzett vegyületek az ország számos kórházában, klinikáján kerültek felhasználásra. Az 1990-es évek elején létrejött PET Centrum (Positron Emission Tomography) igényei alapján, mára már az ún. PET radiofarmakonok képviselik a termelt izotópok döntő részét (^{11}C , ^{13}N , ^{15}O , ^{18}F , stb.).

Amikor 1982-ben csatlakozhattam az intézet ciklotron alkalmazásokkal foglalkozó csoportjához, egyik feladatomban kaptam a rutin izotóptermeléshez szükséges technikai rendszerek tervezését, üzembeállítását, valamint - figyelembe véve a ciklotron lehetőségeit - a termelendő radioizotópok előállításának, termelési körülményeinek optimalizálását. Ezen feladatok megoldásának tudományos megalapozása érdekében jelentős alkalmazott kutatási feladatokat (gerjesztési függvények mérése, hozamok meghatározása stb.) kellett elvégezni. A jelen dolgozatban az ezekkel a munkákkal kapcsolatos kutatásaimat, vizsgálataimat és a kapott eredményeket foglaltam össze.

A megoldandó feladatok

Ismeretes, hogy számos tényezőt kell figyelembe venni ahhoz, hogy a kérdéses radioizotópot optimálisan lehessen termelni egy adott gyorsítóval (pl. a gyorsítható részecskefajta száma és ezek maximális energiái, a nyalábintenzitások, a termeléshez választott magreakciók hozamai, a szennyező izotópok fajtái és hozamai, a besugárzási technikák (targetry), az alkalmazható kémiai elválasztási módszerek stb.).

A nukleáris adatok pontos ismerete alapvető fontosságú mind a tervezésnél, mind a termelések során, mivel ezek ismeretében:

a) Az optimális magreakció könnyen kiválasztható, a termeléshez szükséges energiatartomány (céltárgyvastaság, izotópösszetétel) meghatározható.

b) Az elérhető hozamok pontosan kiszámíthatóak.

c) A hozamok ismeretében pedig a termelés más tényezőinek hatásfoka is pontosan meghatározhatóvá válik. (Pl. a nagy intenzitású besugárzások relatív hozamcsökkenése, a céltárgyfeldolgozás és a radioizotóp kinyerése, valamint a kémiai eljárások miatt bekövetkező esetleges aktivitásveszteségek, stb.)

Már a kezdetekben nyilvánvaló volt, hogy izotóptermelési célokra elsősorban protonok által kiváltott magreakciók jöhetnek szóba, és figyelembe véve a debreceni ciklotron lehetőségeit, azok is csak dúsított izotópösszetételű céltárgyanyagok alkalmazása esetén.

Mivel munkánk kezdetén nem rendelkezünk elegendő tapasztalattal az izotóptermelési feladatok megoldásához, első lépésként összegyűjtöttük és elemeztük az ilyen jellegű irodalmakat. Tapasztalataink szerint a fellelhető források által mért hatáskeresztmetszetek és

hozamادات sok esetben: **(a)** jelentősen eltértek egymástól, **(b)** hiányosak voltak (bizonyos energiatartományokban vagy reakciókra hiányoztak az adatok), **(c)** mérési pontatlanságuk miatt nem voltak felhasználhatók céljainkra. Ezen problémák mindegyike jelentkezett azon izotópokkal kapcsolatban is, melyek termését munkám során feladatul kaptam, nevezetesen a $^{61,64}\text{Cu}$, $^{66,67,68}\text{Ga}$ és a ^{111}In .

A vizsgálataink fő célja és motivációja tehát az volt, hogy a termeléshez szükséges nukleáris adatok részletes és kellő pontosságú mérésével tudományosan megalapozzuk a fenti radioizotópok optimális termelését a debreceni ciklotron körülményei között.

Az adatok elemzéséből az is kiderült azonban, hogy nem csak az általunk elérhető 18 MeV-es protonenergiáig vannak problémák a közölt adatokkal. Ezért vizsgálatainkat kiterjesztettük 30 MeV-ig, mely energia jelenleg a legoptimálisabbnak tekinthető izotóptermelésre (figyelembe véve az üzemeltetési költségeket és az elérhető hozamokat). Reményeink szerint, így az adatainkat a legtöbb izotóptermelő centrum munkájában is fel tudják majd használni.

A hatáskeresztmetszet mérésekhez használandó céltárgyainkat olyan fém hátlapokra készítettük, melyekben természetesen szintén számos jól mérhető (p,n) és (p,2n) reakció megy végbe. Mivel a mérések során egyébként is használunk külön un. monitor fóliákban lejátszódó reakciókat a nyalábáramok monitorizálására, felvetődött a gondolat, hogy a hátlapokban végbemenő reakciók is betölthetnék a monitor reakciók szerepét. Ilyen módon nem szükséges további más fóliákat alkalmazni monitorizálási célokra. Meg kell azt is jegyezni, hogy munkánk kezdetén - az elmúlt évtized végén - az általunk vizsgálandó energiatartományban az adatok elégtelensége miatt, csupán néhány reakciót lehetett monitorizálásra felhasználni.

Az elvégzendő vizsgálatainkat a fentiek alapján az alábbiakban tudtuk összefoglalni:

A) Hatáskeresztmetszet mérések végzése dúsított Zn, Ni és Cd céltárgyakon 30 MeV-ig azon magreakciókra melyekkel az alábbi orvosi célra használandó radioizotópok termelhetők : ^{61}Cu , ^{64}Cu , ^{66}Ga , ^{67}Ga ^{68}Ga és ^{111}In .

B) Hatáskeresztmetszet mérések végzése dúsított Zn, Ni és Cd céltárgyakon 30 MeV-ig azon magreakciókra melyekkel a fenti radioizotópok szennyezői párhuzamosan előállnak.

C) A $^{66,67,68}\text{Ga}$ és a ^{111}In rutin termelési körülményeinek kidolgozása kis- és közép energiás ciklotronokra ($E_p \leq 30$ MeV).

D) $^{nat}\text{Ti}+p$, $^{nat}\text{Ni}+p$ és $^{nat}\text{Au}+p$ folyamatok gerjesztési függvényeinek meghatározása 30 -ig, és annak vizsgálata, hogy közülük mely reakciók alkalmasak proton nyalábok intenzitásának monitorizálására.

E) Az irodalomban a fenti reakciókra közölt hatáskeresztmetszet és hozamادات összegyűjtése, és kritikai elemzése. Ajánlott hatáskeresztmetszet és hozam adat adatbázisok létrehozása a fontosabb termelésre használandó reakciók esetében. Modell számolások végzése, hogy vizsgáljuk a különböző computer programok felhasználhatóságát az izotóptermelésben használandó nukleáris adatok becslésére, és a kísérleti adatok ellentmondásainak tisztázására.

Kísérleti eszközök és alkalmazott módszerek

A termelési reakciók méréséhez használt céltárgyakat dúsított izotópösszetételű anyagokból galvanizálási eljárással készítettük. A monitor reakciókhoz és a dúsított céltárgyak hátlapjaiként vékony gyári fém fóliák (nikkel és arany) kerültek felhasználásra. A hozammérésekhez és a termelésekhez használt targetek hátlapjaiként rezet használtunk, melyet előzőleg nikkellel vagy arannyal vontunk be. Ezek a targetek is ugyanolyan galvanizációs eljárással készültek, mint a hatáskeresztmetszet mérésekhez használtak.

A vizsgált magreakciók hatáskeresztmetszet adatainak meghatározását az un. aktivációs módszerrel végeztük. Egyszerre több céltárgyat sugároztunk be (szedvics technika), így

viszonylag gyorsan (kis relatív hibával) lehetett a vizsgált reakció gerjesztési függvényét, vagy annak egy szakaszát meghatározni. A céltárgytartó egységeink úgy lettek kialakítva (Faraday-kamra), hogy közvetlenül abszolút áramot mérhessünk rajtuk. A céltárgyak közé monitor fóliákat is helyeztünk, melyekkel szintén monitoroztuk a nyalábok energiáját és intenzitását.

A besugárzások 18 MeV alatti részét a debreceni gyorsítónál végeztük, míg az ennél nagyobb energiákon történő mérésekre nemzetközi együttműködések keretében került sor (Brüsszel (Belgium), Jülich (Németország) Miami Beach (Florida, Egyesült Államok), Řež (Cseh köztársaság) valamint Turku (Finnország)). A besugárzások során általában kis nyaláb-áramokat (100-200 nA) és a vizsgálandó izotóp felezési idejének megfelelően választott besugárzási időket (10 perc-3 óra) használtunk.

A fóliák aktivitását az esetek döntő többségében kémiai szeparáció nélkül mértük meg. Spektrumméréseinknél különböző típusú detektor-jelfeldolgozó rendszer összeállításokat használtunk. (HpGe, Ge(Li) detektorok, Aptec, Ortec stb. sokcsatornás analizátorok (MCA)). Valamennyi mérésünknel az aktivált minták úgy voltak elhelyezve, hogy a detektorok szempontjából pontszerű forrásnak tekinthessük őket, illetve hogy a detektorokat érő fotonok ne terheljék túl a mérőrendszereket (a holtidő kisebb mint 5 %). A mérőrendszerek hatásfok hitelesítését standard (3-5% abszolút pontosság) hitelesítő források segítségével végeztük.

A hatáskeresztmetszet adatokat a mért csúcsterületek felhasználásával és az un. aktivációs egyenlet segítségével számoltuk ki. A nyalábok céltárgyban történő energiacsökkenését irodalmi táblázatokkal és standard számítógépes programokkal határoztuk meg, illetve monitor reakciókat használtunk azok ellenőrzésére. (A fóliásor egy adott tagjához a fóliába be- valamint kilépő energiaértékeknek, a gerjesztési függvény változásának figyelembe vételével számolt átlagát rendeltük.) Ezen energia adat hibáját a ciklotronból kilépő nyaláb kezdeti bizonytalanságának és a megelőző céltárgyak ismeretében számoltuk ki. A hatáskeresztmetszet adatokhoz hibaként a legfontosabb bizonytalansági tényezők (a céltárgy-vastagság, a detektor hatásfok, a fluxusmérés, a csúcsterület, a felhasznált nukleáris adatok, a mérési geometria meghatározásának hibái) négyzetösszegének négyzetgyökét rendeltük.

A rutin izotóptermelési feladatok ellátására függőleges elrendezésű nyalábvéget és speciális besugárzó rendszert építettünk fel a debreceni ciklotronnál, mely biztosítja a gyors és kontrollálható nyalábvezetést, a céltárgyak megbízható kezelését, valamint a céltárgyakban keletkező hő elvezetését. Ezen rendszer kiválóan alkalmas szilárd, cseppfolyós valamint gáz halmazállapotú céltárgyanyagok besugárzására is. Hozamméréseink nagy részét is ezzel a berendezéssel végeztük. A (p,2n) reakcióval történő indium és gallium termelések hozamméréseire illetve a rutin termelésekre a Řež-i (Cseh köztársaság) valamint a Miami Beach-i (Florida, U.S.A.) gyorsítók besugárzási rendszereit használtuk. Termelések esetén ezekre az izotópokra az igényelt aktivitástól mennyiségétől függően 10-36 órás besugárzásokat végeztünk 50-150 μ A nyalábtintenzitások mellett.

Eredményeink

A vizsgálataink során szerzett tapasztalatok és új tudományos eredmények (melyek elérésében minden esetben meghatározó szerepem volt) az alábbiakban foglalhatók össze:

1) Új, az eddigieknél részletesebb méréseket végeztünk a széleskörben használatos $^{66,67,68}\text{Ga}$, ^{111}In és $^{61,64}\text{Cu}$, radioizotópok termeléséhez szükséges magreakciók gerjesztési függvényeinek meghatározására. A vizsgált reakciók és energiatartományok a következők voltak $^{61}\text{Ni}(p,n)^{61}\text{Cu}$ (19 MeV-ig), $^{64}\text{Ni}(p,n)^{64}\text{Cu}$ (19 MeV-ig), $^{66}\text{Zn}(p,n)^{66}\text{Ga}$ (26 MeV-ig), $^{67}\text{Zn}(p,n)^{67}\text{Ga}$ (26 MeV-ig), $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$ (26 MeV-ig), $^{68}\text{Zn}(p,n)^{68}\text{Ga}$ (26 MeV-ig), $^{111}\text{Cd}(p,n)^{111}\text{In}$ (30 MeV-ig) és $^{112}\text{Cd}(p,2n)^{111}\text{In}$ (30 MeV-ig). Méréseink segítségével sikerült

teljessé tennünk a fenti reakciók gerjesztési függvényeit a termelésekhez szükséges energiatartományokban, mely adatok bizonyos energiatartományokban eddig vagy nem álltak rendelkezésre, vagy ezekkel kapcsolatban csak egyetlen, mások által meg nem erősített mérést közöltek az irodalomban. (Ilyen reakciók pl. a $^{61}\text{Ni}(p,n)^{61}\text{Cu}$, $^{66}\text{Zn}(p,n)^{66}\text{Ga}$, $^{67}\text{Zn}(p,n)^{67}\text{Ga}$ és a $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$.) A megbízhatónak tűnő irodalmi adatokkal kiegészítve méréseinket lehetőség nyílt arra, hogy a várható hozamadatokat az eddigieknél nagyobb pontossággal számolhassuk ki.

2) A fenti vizsgálatokhoz kapcsolódva mértük az un. mellék reakciók hatáskeresztmetszeteit is, melyek ugyanazokon (vagy a targetben jelenlevő egyéb) magokon mennek végbe és a legfontosabb un. szennyező radioizotópokat eredményezik. Az ilyen reakciókra vonatkozó mérések tapasztalataink szerint még hiányosabbak, mint a termelési reakciókkal kapcsolatos adatok. Az általunk vizsgált 6 reakció alapján jelentősen kiegészítettük és pontosítottuk a korábbi ismereteket a ^{60}Cu , ^{66}Ga , ^{109}In és ^{110}In előállításával kapcsolatban ($^{61}\text{Ni}(p,2n)^{60}\text{Cu}$, $^{67}\text{Zn}(p,2n)^{66}\text{Ga}$, $^{68}\text{Zn}(p,3n)^{66}\text{Ga}$, $^{111}\text{Cd}(p,3n)^{109}\text{In}$, $^{111}\text{Cd}(p,2n)^{110}\text{In}$ és $^{112}\text{Cd}(p,3n)^{110}\text{In}$). A fentiek közül a $^{61}\text{Ni}(p,2n)^{60}\text{Cu}$ reakciót elsőként mértük a reakció küszöbenergiájától 19 MeV-ig, míg a kadmiumon végbemenő reakciók esetében ugyancsak mi közöltünk először adatokat bizonyos energiatartományokra vonatkozólag. (Meg kell jegyeznünk a szennyezőkkel kapcsolatban azt, hogy valamely radioizotóp termelése esetén azokat a hosszú felezési idejű izotópokat kell szennyezőknek tekinteni, melyek kémiaiilag nem elkülöníthetőek.) Méréseink alapján, az optimális termelési energiaintervallumok (ahol a szennyezők mennyisége a felhasználók által megkívánt szint alatt marad, vagy egyáltalán nem termelődnek) pontosabb meghatározása vált lehetővé.

3) A korábbiaknál részletesebb hatáskeresztmetszet méréseket végeztünk néhány Ti+p, Ni+p és Au+p folyamattal kapcsolatban, olyan új reakciókat keresve, melyeket proton nyalábok intenzitásának monitorizálására is jól fel lehet használni. Eredményeink alapján hét reakció, illetve folyamat került kiválasztásra, melyeket elsőként mi javasoltunk a fenti célra ($^{nat}\text{Ti}(p,x)^{48}\text{V}$ (30 MeV-ig), $^{nat}\text{Ni}(p,x)^{55}\text{Co}$ (30 MeV-ig), $^{nat}\text{Ni}(p,x)^{56}\text{Co}$ (30 MeV-ig), $^{nat}\text{Ni}(p,x)^{57}\text{Co}$ (30 MeV-ig), $^{nat}\text{Ni}(p,x)^{57}\text{Ni}$ (44 MeV-ig), $^{nat}\text{Au}(p,pn)^{196}\text{Au}$ (30 MeV-ig) és $^{nat}\text{Au}(p,n)^{197m}\text{Hg}$ (18 MeV-ig). A kapott visszajelzések alapján eredményeinket azóta már számos laboratórium izotóptermelési programjában használják.

4) Hatáskeresztmetszet méréseink során számos olyan reakciót is mértünk, melyek jelenleg nem játszanak számottevő szerepet sem az izotóptermelésben sem a nyalábok monitorizálásában. Ezen eredmények azonban kiegészítik a töltött részecskék által kiváltott magreakciókra vonatkozó rendelkezésre álló adatokat. Ezen folyamatokat vagy elsőnek mértük ($^{61}\text{Ni}(p,\alpha)^{58m}\text{Co}$ és $^{64}\text{Ni}(p,\alpha)^{61}\text{Co}$), vagy bővítettük a vizsgált energiatartományokat ($^{46}\text{Ti}(p,\alpha)^{43}\text{Sc}$, $^{47}\text{Ti}(p,\alpha)^{44m,g}\text{Sc}$, $^{48}\text{Ti}(p,2p)^{47}\text{Sc}$, $^{61}\text{Ni}(p,\alpha)^{58}\text{Co}$, $^{111}\text{Cd}(p,2n)^{110g}\text{In}$ és $^{197}\text{Au}(p,n)^{197m}\text{Hg}$). A kapott adatok számos, az izotóptermelésen kívüli terület számára is hasznosak lehetnek (pl. sugárvédelem, gyorsító technológia, asztrofizikai kutatások stb.).

5) A mérésekhez kapcsolódva minden esetben elvégeztük az addig megjelent irodalmi adatoknak a korábbiaknál sokkal részletesebb kompilálását és kritikai elemzését. Kiszűrtük azon problémákat melyek a kísérleti körülmények nem megfelelő megválasztásából, illetve a hibás számolásokból következtek be. Az így összeállított hatáskeresztmetszet és hozamadat adatbázisokat a későbbiekben felhasználtuk un. "ajánlott" adatokat tartalmazó adatbázisok létrehozására. A kompilálási eredményeket melyek a Zn+p reakciókra vonatkoztak, külön publikációban is hozzáférhetővé tettük.

6) Saját mérési eredményeinkre és a megbízható irodalmi adatokra támaszkodva elsőként hoztunk létre ajánlott (recommended) hatáskeresztmetszet és hozamadat adatbázisokat a 30 MeV-ig terjedő energiatartományban az alábbi reakciókra $^{nat}\text{Zn}(p,x)^{65}\text{Zn}$, $^{66}\text{Zn}(p,n)^{66}\text{Ga}$, $^{67}\text{Zn}(p,2n)^{66}\text{Ga}$, $^{68}\text{Zn}(p,3n)^{66}\text{Ga}$, $^{67}\text{Zn}(p,n)^{67}\text{Ga}$, $^{68}\text{Zn}(p,2n)^{67}\text{Ga}$, $^{68}\text{Zn}(p,n)^{68}\text{Ga}$, $^{111}\text{Cd}(p,n)^{111}\text{In}$, $^{112}\text{Cd}(p,2n)^{111}\text{In}$ és $^{197}\text{Au}(p,pn)^{196}\text{Au}$. Számolt hozamaink összehasonlítása irodalmi mérésekkel megerősítette adataink használhatóságát az izotóptermelés területén.

7) A mért gerjesztési függvények alapján meghatároztuk a $^{61,64}\text{Cu}$, $^{66,67,68}\text{Ga}$ és ^{111}In radioizotópok termeléséhez szükséges optimális energiatartományokat, figyelembe véve a rendelkezésre álló céltárgyanyagok dúsítási fokát és az elérhető nyalábenergiákat. Ezen adatok alapján folynak a rutin besugárzások Debrecenben, Řež-ben (Cseh Köztársaság) és Miami Beach-ben (Florida, Egyesült Államok) a ^{67}Ga ($^{67}\text{Zn}(p,n)$ és $^{68}\text{Zn}(p,2n)$) és a ^{111}In ($^{111}\text{Cd}(p,n)$ és $^{112}\text{Cd}(p,2n)$) előállítására.

A debreceni ciklotronhoz kifejlesztettünk egy olyan függőleges elrendezésű izotóptermelő rendszert is, mely egyaránt alkalmas szilárd, cseppfolyós és gáz halmazállapotú céltárgyanyagok nagyáramokkal történő besugárzására.

8) Számolásokat végeztünk a $^{61}\text{Ni}(p,n)$, $^{61}\text{Ni}(p,2n)$, $^{61}\text{Ni}(p,\alpha)$, $^{64}\text{Ni}(p,n)$, $^{64}\text{Ni}(p,\alpha)$, $^{66}\text{Zn}(p,n)$, $^{67}\text{Zn}(p,2n)$, $^{67}\text{Zn}(p,n)$, $^{68}\text{Zn}(p,2n)$ és $^{68}\text{Zn}(p,n)$ reakciók gerjesztési függvényeinek leírására különböző magreakció elméleten alapuló modell programok segítségével. (A számolások során a felhasznált programok figyelembe veszik a közbenső magok képződésével járó folyamatokat és az un. preequilibrium kibocsátásból származó járulékokat is.). Megállapítható volt, hogy a modell számítások jól reprodukálják a gerjesztési függvényeket, habár a hatáskeresztmetszet adatok becslése rendre eltért a mért adatoktól. (pl a $\text{Zn}+p$ reakciók esetén). A számítások felhívták a figyelmet a pontos kísérleti adatok szükségességére.

9) Részletesen vizsgáltuk az általunk mért hatáskeresztmetszet adatok felhasználhatóságát a kopásvizsgálatokra használt vékonyréteg aktivációs technikában (TLA). A $\text{Zn}+p$ reakciók ajánlott adatait figyelembe véve, javasoltuk a $^{nat}\text{Zn}(p,x)^{65}\text{Zn}$ és a $^{nat}\text{Zn}(p,x)^{66,67,68}\text{Zn}$ folyamatok használatát cink tartalmú anyagok un. hitelesítő görbéinek számolásokkal történő meghatározására. Számolásainkat a rendelkezésre álló néhány kísérleti adattal is összehasonlítottuk, melyek igazolták adataink ilyen célú használhatóságát is.

Az értekezés eredményeiről 13 közleményben (megjelent illetve elfogadott tudományos cikk) számoltunk be. (A dolgozat témájához még további 7 cikk kapcsolódik.) A fenti publikációkra 1997 májusáig 49 hivatkozás történt. A cikkek (13) fénymásolatai a jelen dolgozat függelékében megtalálhatóak.

8. Acknowledgements

I am greatly indebted to my supervisor Dr Ferenc Tárkányi PhD, head of the Cyclotron Department of ATOMKI, Debrecen, Hungary for his active support during the many years of work and for the constructive criticism and many fruitful discussions during the preparation of this manuscript.

I am also grateful to all present and former colleagues and the directorate of ATOMKI, Debrecen, Hungary who in one way or another have given their support during the performance of this work.

I direct my sincerest thanks to my co-authors Dr. Jörgen Bergman, PhD, Mr. Gerrit. Blessing, Dr. Thomas E. Boothe, PhD, Mr. Yolvi Feijoo, Prof. Dr. Alex Hermanne, PhD, Dr. Sven-Johan Heselius, PhD, Mr. Don Kinney, Dr. Peter Kopecký, PhD, Dr. Zoltán Kovács, PhD, Dr. Imre Mahunka, PhD, Dr. Thomas F. McLeod, PhD, Dr. Pál Mikecz, MSc, Dr. Tamás Molnár, MSc, Mr. Michael Plitnikas, Prof. Dr. Sayed M. Qaim, PhD, Prof. Dr. Frank Rösch, PhD, Mrs. Patty Smith, Dr. Olof Solin, PhD, Dr. Michel Sonck, PhD, Dr. Sándor Sudár, PhD, Dr. Sándor Takács, MSc, Mr. Emanuel Tavano, Dr. Ferenc Tárkányi, PhD, and Prof. Dr. M. Uhl, PhD (deceased). Their collaboration in various parts of the study has been necessary for the progress of this work.

I am grateful to Prof. Dr. Sayed M. Qaim of Institute für Nuklearchemie, Forschungsanlage Jülich GmbH, Jülich, Germany, and Dr Thomas E. Boothe of Cyclotron Facility of Mount Sinai Medical Center of Greater Miami, Miami Beach, Florida, U.S.A. for making it possible for me to stay at their departments and conduct cross section measurements related to this work as an IAEA fellow (six months) and as a full time staff member (two years), respectively,

I am much indebted to the technical staffs of the cyclotrons in Abo, Finland; Brussels, Belgium; Debrecen, Hungary; Jülich, Germany; Miami Beach, Florida, U.S.A and Rez, Czech Republik.

Finally I owe my warmest thanks to my wife Lilian for her help and patience through the years of this work. I also thank my daughter Eva-Ibolya for her understanding attitude during my absences.

9. References

Alderliesten C., Bousshid O., Jahn P. and Maier-Börcke C. (1975) Measurement and hybrid model analysis of (particle,xnyp) excitation functions. *Ann. Report Jülich, Germany*.p 38.

Andersen H.H. and Ziegler J.F. (1977) Hydrogen stopping powers and ranges in all elements. In *The Stopping and Ranges of Ions in Matter*. Vol.3 (Ed. Ziegler J.F.), Pergamon Press, New York, U.S.A.

Aleksandrov V. N., Semyonova M. P. and Semyonov V. G. (1987) Excitation functions for radionuclides produced by (p,x) reactions of copper and nickel. *At. Energ.* **62**, 411.

Barrandon J. N., Debrun J. L., Kohn A. and Spear R. H. (1975) Étude du dosage de Ti, V, Cr, Fe, Ni, Cu et Zn par activation avec des protons d'énergie limitée a 20 MeV. *Nucl. Instrum. Methods* **127**, 269.

Bass L. A., McCarthy D. W., Jones L. A., Cutler P. D., Shefer R. E., Klinkowstein R. E., Schwarz S. W., Cutler C. S., Lewis J. S., Anderson C. J. and Welch M. J. (1997) High purity production and potential applications of copper-60 and copper-61. *Abstract of the XIIIth International Symposium on Radiopharmaceutical Chemistry*, 15-19 June 1997, Uppsala, Sweden, p. 325.

Birattari C. and Bonardi M. (1980) Funzioni di eccitazione per le reazioni nucleari (p,xn) e (p,pxn) su targhette d'oro e studio del generatore del radioisotopo a vita ultrabreve ^{195m}Hg , *Report INFN/TC- 80/17, Frascati, Italy*, p. 1.

Blann M. (1971a) Hybrid model for preequilibrium decay in nuclear reactions. *Phys. Rev. Lett.* **27**, 337.

Blann M. (1971b) Hybrid model for preequilibrium decay in nuclear reactions. erratum *Phys. Rev. Lett.* **27**, 700.

Blann M. (1972) Importance of the nuclear density distribution on pre-equilibrium decay. *Phys. Rev. Lett.* **28**, 757.

Blaser J.-P., Boehm F., Marmier P. and Peaslee D. C. (1951) Fonctions d'excitation de la réaction (p,n).I. *Helv. Phys. Acta* **24**, 3.

Blosser H. G. and Handley T. H. (1955) Survey of (p,n) reactions at 12 MeV. *Phys. Rev.* **100**, 1340.

Bodemann R., Michel R., Rosel R., Herpers U., Holmqvist B., Conde H. and Malmberg P. (1995) Production of radionuclides from target elements ($22 \leq Z \leq 29$) by proton-induced reactions up to 100 MeV. *Progress Report on Nuclear Data Research in the Federal Republic of Germany for the Period April 1, 1994 to March 31, 1995* (ed. Qaim S. M.), *INDC(Ger)-040, Jül-3086*, p. 27.

Bonardi M. and Birattari C.(1983) Optimization of irradiation parameters for Ga-67 production from Zn(p,xn) nuclear reactions. *J.Radioanal.Chem.* **76**, 311.

Bonardi M. (1988) The contribution to nuclear data for biomedical radioisotope production from the Milan cyclotron laboratory. *Proceedings of the IAEA Consultants' Meeting on Data Requirements for Medical Radioisotope Production*. 20-24 April 1987, Tokyo, Japan (Ed. Okamoto .K), INDC(NDS)-195/GZ, IAEA, Vienna, Austria, p.98.

Brinkman G. A., Helmer J. and Lindner L. (1977) Nickel and copper foils as monitors for cyclotron beam intensities. *Radiochim. Radioanal. Letters* **28**, 9.

Brinkmann G. A (1979) Integrale Anregungsfunktionen für Protonen and Alpha-induzierte Reactionen an Targetelementen $22 \leq Z \leq 28$. Dissertation, Universität zu Köln, Cologne, Germany.

Brodzinski R. L., Rancitelli L. A., Cooper J. A. and Wogman N. A. (1971) High energy proton spallation of titanium. *Phys. Rev.* **C4**, 1250.

Browne E. and Firestone R.B. (1986) *Table of Radioactive Isotopes*, Wiley and Sons, London, UK.

Cohen B. L., Newman E. and Handley T. H. (1955) (p,pn)+(p,2n) and (p,2p) cross sections in medium weights elements. *Phys. Rev.* **99**, 723.

Esat M. T., Spear R. H., Zyskind J. L., Shapiro M. H., Fowler W. A. and Davidson J. M. (1981) Test of global Hauser-Fresbach calculations for proton-induced reactions on ^{68}Zn . *Phys. Rev.* **C23**, 1822.

Ewart H. A. and Blann M. (1964) Nuclear cross section for charged particle induced reactions. *Preprints ORNL CPX-1, CPX-2*, Oak Ridge, U.S.A.

Furukawa M., Shinohara A., Narita M. and Kojima S. (1990) Production of $^{59,63}\text{Ni}$ from natural Ni irradiated with protons up to $E_p = 40$ MeV and decay of ^{56}Ni . *Ann. Report Institute for Nuclear Study, University of Tokyo, Japan*, p. 35.

Goethals P., Coene M., Slegers G., Agon P., Deman J. and Schelstraete K. (1988) Cyclotron production of carrier-free ^{66}Ga as a positron emitting label of albumin colloids for clinical use. *Eur. J. Nucl. Med.* **14**, 152.

Green M. A., Mathias C. J., Welch M. J., McGuire A. H., Perry D., Fernandez-Rubio F., Perlmutter J. S., Raichle M. E. and Bergmann S. R. (1990) Copper-62-labelled pyruvaldehyde bis(N4-methylthiosemicarbazone)copper(II): synthesis and evaluation as a positron emission tomography tracer for cerebral and myocardial perfusion. *J. Nucl. Med.* **31**, 1989.

Griffin J. J. (1966) Statistical model of intermediate structure. *Phys. Rev. Lett.* **17**, 478.

Gritsina V.T., Klucharev A. P., Remayev V. V. and Reshetova L. N. (1963) Ratio of the cross sections for formation of the isomer and ground states of nuclei in the (p,n)-reaction at energies from the threshold to 20 MeV. *J. Exp. Theor. Phys.* **44**, 1770.

Haasbroek F. J., Steyn J., Neirinckx R. D., Burdzik G. F., Cogneau M. and Wanet P. (1976) Excitation functions and thick target yields for radioisotopes induced in natural Mg, Co, Ni and Ta by medium energy protons. *Appl. Radiat. Isot.* **28**, 533.

Hansen L. F., Jopson R. C., Mark H. and Swift C. D. (1962) Ta¹⁸¹(p,n)W¹⁸¹ and Au¹⁹⁷(p,n)Hg¹⁹⁷ excitation functions between 4 and 13 MeV. *Nucl. Phys.* **30**, 389.

Hashizume A. (1988) Monitor reactions for the production of radioisotopes for medical use. in *Proceedings of the IAEA Consultants' Meeting on Data Requirements for Medical Radioisotope Production*. 20-24 April 1987, Tokyo, Japan (Ed. Okamoto .K), INDC(NDS)-195/GZ, IAEA, Vienna, Austria, p. 44.

Herkert A. (1975) Die Aktivierung von metallischen Maschinenteilen mit geladenen Teilchen zur Durchführung von Verschleissmessungen. KFK 2096, Karlsruhe, Germany.

Hermanne A. (1994) Private communication.

Hermanne A., Sonck M., Takács S., Szelecsényi F. and Tárkányi F. (1997) Influence of secondary neutrons on cross section determination of proton and deuteron induced reactions on ^{nat}Ni targets. To be published in *Proceedings of the International Conference on Nuclear Data for Science and Technology*, 19-24 May 1997, Triest, Italy.

Hille M., Hille P., Uhl M. and Weisz W. (1972) Excitation functions of (p,n) and (α,n) reactions on Ni, Cu and Zn. *Nucl. Phys.* **A198**, 625.

Howe H. A. (1958) (p,n) cross sections of copper and zinc. *Phys. Rev.* **109**, 2083.

Johnson C. H., Galansky A. and Inskeep C. N. (1960) Cross sections for (p,n) reactions in intermediate weight nuclei. *Report ORNL-2910*, Oak Ridge, U.S.A.

Kaufman S. (1960) Reactions of protons with ⁵⁸Ni and ⁶⁰Ni. *Phys. Rev.* **117**, 1532.

Kavanagh T.M. and Bell R. E. (1961) Cross sections of (p,pxn) reactions in Au¹⁹⁷. *Can. J. Phys.* **39**, 1172.

Kirchner P. T. (Senior editor) (1983) *Nuclear Medicine Review Syllabus*. Society of Nuclear Medicine, New York, NY, U.S.A.

Kopecký P. (1990) Cross sections and production yields of ⁶⁶Ga and ⁶⁷Ga for proton reactions in natural zinc. *Appl. Radiat. Isot.* **41**, 606.

Lambrecht R. M., Waters S. L., Hanlin L., Qaim S. M., Umewaza H., Beyer G. J., Heinzl H., Bonardi M., Nozaki T., Hashizume A., Lagunas-Solar M. C., Kitao K. and Berényi D. (1988) Summary of conclusion and recommendations. *Proceeding of the IAEA Consultant' Meeting on Data Requirements for Medical Radioisotope Production*, 20-24 April 1987, Tokyo, Japan, INDC(NDS)-195/GZ, IAEA Vienna, Austria, p.13.

Levkovskii V. N. (1991) *Cross-section of Medium Mass Nuclide Activation (A=40-100) by Medium Energy Protons and Alpha Particles (E=10-50 MeV)*. Inter-Vesi, Moscow USSR.

Little F. E. and Lagunas-Solar M. C. (1983) Cyclotron Production of ^{67}Ga . Cross sections and thick-target yields for the $^{67}\text{Zn}(p,n)$ and $^{68}\text{Zn}(p,2n)$ reactions. *Int. J. Appl. Radiat. Isot.* **34**, 631.

Mahunka I., Tárkányi F., Andó L., Szelecsényi F., Fenyvesi A., Ditrói F. and Takács S. (1988) Medical project at the MGC cyclotron in Debrecen. Proceedings of the Fourth Symposium on the Medical Application of Cyclotrons 28-31 May, 1986, Turku, Finland (Eds. Nántö V. and Suolinna E-M) *Ann. Univ. Turkuensis* **D27**, 138

Marinov A., Oelert W., Gopal S., Berg G. P. A., Bojowald J., Hürlimann W., Katayama I., Martin S. A., Mayer-Böricke C., Meissburger J., Römer J. G. M., Rogge M., Tain J. L., Turek P., Zemlo L., Mooy R. B. M., Glaudemans P. W. M., Brant S., Paar V., Vouk M. and Lopac V. (1984) Study of proton-hole states in ^{61}Co using the $^{62}\text{Ni}(d,^3\text{He})^{61}\text{Co}$ reaction at 78 MeV. *Nucl. Phys.* **A431**, 317.

Marten M., Schüring A., Scobel W. and Probst H. J. (1985) Preequilibrium neutron emission in $^{109}\text{Ag}(^3\text{He},xn)$ and $^{111}\text{Cd}(p,xn)$ reactions. *Z. Phys.* **A322**, 93.

McGee T., Rao C. L., Saha G. B. and Yaffe L. (1970) Nuclear interactions of ^{45}Sc and ^{68}Zn with protons of medium energy. *Nucl. Phys.* **A150**, 11.

Michel R., Brinkmann G., Weigel H. and Herr W. (1978a) Proton-induced reactions on titanium with energies between 13 and 45 MeV. *J. inorg. Nucl. Chem.* **40**, 1845.

Michel R., Weigel H. and Herr W. (1978b) Proton-induced reactions on nickel with energies between 12 and 45 MeV. *Z. Phys.* **A286**, 393.

Michel R. and Brinkmann G. (1980) On the depth-dependent production of radionuclides ($44 < A < 59$) by solar protons in extraterrestrial matter. *J. Radionucl. Chem.* **59**, 467.

Michel R., Bodemann R., Busemann H., Daunke R., Gloris M., Lange H.-J., Klug B., Krins A., Leya I., Lüpke M., Neumann S., Reinhardt H., Schnatz-Böttgen M., Herpers U., Schieken Th., Sudbrock F., Holmqvist B., Conde H., Malmberg P., Suter M., Dittrich-Hannen B., Kubik P.-W., Synal A. (1997) Cross sections for the production of residual nuclides by low- and medium-energy protons from the target elements C, N, O, Mg, Al, Si, Ca, Ti, Mn, Fe, Co, Ni, Cu, Sr, Y, Zr, Nb, Ba and Au. to be published in *Nucl. Instr. Meth. Phys. Res.* **B129**, 153.

Nagame Y., Sueki K., Baba S., and Nakahara H. (1990) Isomeric yield ratios in proton-, ^3He and α -particle-induced reactions on ^{197}Au . *Phys. Rev.* **C41**, 889.

Nortier F. M., Mills S. J. and Steyn G. F. (1990) Excitation functions and production rates of relevance to the production of ^{111}In by proton bombardment of $^{\text{nat}}\text{Cd}$ and $^{\text{nat}}\text{In}$ up to 100 MeV. *Appl. Radiat. Isot.* **41**, 1201.

Nortier F. M., Mills S. J. and Steyn G. F. (1991) Excitation functions and yields of relevance to the production of ^{67}Ga by proton bombardment of $^{\text{nat}}\text{Zn}$ and $^{\text{nat}}\text{Ge}$ up to 100 MeV. *Appl. Radiat. Isot.* **42**, 353.

Noujaim A. A. (editor) (1981) Gallium-67 and related elements. *Int. J. Nucl. Med. Biol.* **8**, 1.

Nowotny R., Uhl M. (1987) Calculation of excitation functions for charged particle induced reactions. in *Handbook on Nuclear Activation Data, IAEA Technical Report Series No.273, IAEA, Vienna, Austria, ISBN 92-0-135087-2 p.441.*

Oblozinský P.(editor) (1997) Summary Report of the Second Research Co-ordination Meeting on Development of Reference Charged-Particle Cross Section Database for Medical Radioisotope Production. 7-10 April 1997, Cape Town, South Africa, *IAEA Report INDC(NDS)-371, Distrib.: G+MR, IAEA, Vienna, Austria.*

Otozai K., Kume S., Mito A., Okamura H., Tsujoni R., Kanchiku Y., Katoh T. and Gotoh H. (1966) Excitation functions for the reactions induced by protons on Cd up to 37 MeV. *Nucl. Phys.* **80**, 335.

Pavlik A. (1988) Calculation of excitation functions for radioisotope production. *Proceeding of the IAEA Consultant' Meeting on Data Requirements for Medical Radioisotope Production*, 20-24 April 1987, Tokyo, Japan, INDC(NDS)-195/GZ, IAEA Vienna, Austria, p.124.

Piel H., Qaim S. M. and Stöcklin G. (1992) Excitation functions of (p,xn)-reactions on ^{nat}Ni and highly enriched ⁶²Ni: possibility of production of medically important radioisotope ⁶²Cu at a small cyclotron. *Radiochimica Acta* **57**, 1.

Qaim S. M. (1987) Cyclotron production of generator radionuclides. *Radiochim. Acta* **41**, 111.

Reimer P., Qaim S. M., Blessing G., Spellerberg S., Coenen H. H., (1997) Production of ⁵⁵Co and ⁵⁷Co via (p,x) reaction on ⁵⁸Ni. *Abstracts of the XIIth International Symposium on Radiopharmaceutical Chemistry*, 15-19 June 1997, Uppsala, Sweden, p. 299.

Schwerer O. and Lemmel H. D. (1996) Co-ordination of the nuclear reaction data centers. INDC(NDS)-360 Distr. G,NC., IAEA, Vienna, Austria,

Schwerer O. and Okamoto K. (1989) Status report on cross-sections of monitor reactions for radioisotope production. INDC(NDS)-218/GZ+, IAEA Vienna, Austria.

Skakun E. A., Kljucharev A. P., Rakivnenko Yu. and Romanij I. A. (1975) Excitation functions of (p,n)- and (p,2n)-reactions on cadmium isotopes. *Izv. AN. SSSR, ser fiz.* **39**, 24.

Skakun E. A., Iordakesku A., Lutsik V. A., Rakivnenko Yu. and Romanij I. A (1979) Excitation functions and isomeric ratios for ¹¹¹Cd(p,n)^{111m,g}In and ¹¹³Cd(p,n)^{113m}In. *Abstracts of XXIXth Symp. on Nuclear Spectroscopy and Nuclear Structure*, Riga, 1979, Nauka, Leningrad, p.20.

Sosniak J. (1958) PhD Thesis, McGill University, Montreal Canada.

Stück R. (1983) Protonen-induzierte Reaktionen an Ti, V, Mn, Fe, Co,und Ni. Messung und Hybrid Modell Analyse. Integraler Anregungsfunktionen und ihre Anwendung in Modellrechnungen zur Production kosmogener Nuklide. Dissertation, Universität zu Köln, Cologne, Germany.

Székely G. (1985) FGM- a flexible gamma-spektrum analysis program for a small computer. *Comput. Phys. Commun.* **34**, 313.

Szelecsényi F. (1988) A ^{67}Ga radioizotóp előállítás MGC ciklotronnal (Production of ^{67}Ga with MGC-20 Cyclotron). Thesis for university doctor's degree (supervisors: Tárkányi F. and Mahunka I.), ATOMKI, Debrecen, Hungary, p.1.

Takács S., Vasváry L. and Tárkányi F. (1994) Remeasurement and compilation of excitation function of proton induced reactions on iron for activation techniques. *Nucl. Instrum. and Meth. Phys. Res.* **B89**, 88.

Tanaka S. and Furukawa H. (1959) Excitation function for (p,n) reactions with titanium, vanadium, chromium, iron and nickel up to $E_p=14$ MeV. *J. Phys. Soc. Japan* **14**, 1269.

Tanaka S., Furukawa M. and Chiba M. (1972) Nuclear reaction of nickel with protons up to 56 MeV. *J. Inorg. Nucl. Chem.* **34**, 2419.

Tilbury R.S. and Yaffe L. (1963) Nuclear isomers $\text{Hg}^{197m,g}$, $\text{Hg}^{195m,g}$ and $\text{Au}^{196m,g}$ formed by proton bombardment of gold with protons of energies from 8 to 60 MeV. *Can. J. Chem.* **41**, 2634.

Uhl M. and Strohmaier B. (1976) Computer code for particle induced activation cross section and related quantities. *IRK Report No76/01*, Vienna, Austria.

Uhl.M. (1995) unpublished results.

Vandenbosch R. and Huizenga J.R. (1960) Isomeric cross-section ratios for reactions producing the isomeric pair $^{197,197m}\text{Hg}$. *Phys. Rev.* **120**, 1313.

Wagner K., Langenstrassen R., Ludwig S. and Menzel M. (1981) Zum Einsatz der radioaktiven Markierung und der Neutronenaktivierungsanalyse für die Untersuchung von Schichten auf Reibkörpern und von Verschießvorgängen. *Schmierungstechnik* (ISSN 0036-6226) **12**, 74.

Williamson C. F., Boujot J. P. and Picard J. (1966) Tables of range and stopping power of chemical elements for charged particles of energy 0.05 to 500 MeV. *Rapport CEA-R-3042*, Saclay, France.

Wing J. and Huizenga J. R. (1962) (p,n) cross sections of V^{51} , Cr^{52} , Cu^{63} , Cu^{65} , Ag^{107} , Ag^{109} , Cd^{111} , Cd^{114} and La^{139} from 5 to 10.5 MeV. *Phys Rev.* **128**, 280.

Wolf A.P. and Jones W.B. (1983) Cyclotrons for biomedical radioisotope production. *Radiochimica Acta* **34**, 1.

Zhuravlev B. V., Grusha O. V. Ivanova S. P., Trykova V.I. and Shubin Yu. N. (1984) Analysis of neutron spectra in 22 MeV proton interactions with nuclei. *Yadarnaya. Fizika* **39**, 264.

10. Appendix

Photocopies of the publications related to the thesis.

Measurement of Cross Sections of Proton Induced Nuclear Reactions on Ti, Ni, Zn, Cd and Au up to 30 MeV and Their Application in Radioisotope Production

Értekezés a doktori (PhD) fokozat megszerzése érdekében a **fizika** tudományában.

Írta: **Dr. Szelecsényi Ferenc** okleveles **fizikus**

Készült a Kossuth Lajos Tudományegyetem **Fizika** doktori programja (**Magfizikai** (II) alprogramja) keretében.

Témavezető: **Dr. Tárkányi Ferenc** a **fiz. tud. kandidátusa**

Elfogadásra javaslom: 1997 augusztus 22.

.....

Jelölt a doktori szigorlatot 1997..november 12 ...-n eredményesen letette:

a bizottság elnöke: Dr. ..Berényi Dénes..

.....

Az értekezést bírálóként elfogadásra javaslom:

Dr. Kiss Ádám..... ..

Dr. Kiss Árpád Zoltán..... ..

Jelölt az értekezést 1997 november 28.....-n sikeresen megvédte:

A védési bizottság elnöke: Dr. ..Csikai Gyula..... ..

Debrecen, 1997.

