

PROTON INDUCED REACTIONS ON ^{159}Tb AND ^{139}La FOR PRODUCING ^{159}Dy AND ^{139}Ce

H.E. Hassan, F.S. Al-Saleh*, K.F. Hassan, A. Sayed and Z.A. Saleh

Cyclotron Facility, Nuclear Research Center, Atomic Energy Authority, Cairo 13759, Egypt.
*Physics Department, Girls College of Education in Riyadh, P.O.Box 27329, Riyadh 11417,
Saudi Arabia.

Abstract

The radionuclides ^{159}Dy ($T_{1/2}=144$ d) and ^{139}Ce ($T_{1/2}=137.64$ d) are potentially important for medical purposes. The first radioisotope is important for transmission imaging and bone mineral analysis. The other one is convenient as a standard source. In the present work the possibility of their production at MGC-20 cyclotron was investigated through the (p,n) reactions on $^{\text{nat}}\text{Tb}$ and $^{\text{nat}}\text{La}$. Excitation functions were measured from the respective threshold of $^{159}\text{Tb}(p,n)^{159}\text{Dy}$ and $^{139}\text{La}(p,n)^{139}\text{Ce}$ reactions up to 18 MeV and 14.7 MeV, respectively, using the stacked-foil technique. Radioactivity of the produced radionuclides was measured using high resolution gamma ray spectrometer. Natural copper foils were used as monitors. Nuclear model calculations were performed using ALICE-91, ALICE-IPPE and EMPIRE-2.19 codes. The experimental results were compared with the theoretical calculations as well as with the previously reported data. Integral yields for the produced ^{159}Dy and ^{139}Ce were also calculated.

Key Words: *Cross-Section/ Excitation Function/ Nuclear Model Calculation/ Radionuclides / Yield / ^{159}Dy / ^{139}Ce*

INTRODUCTION

Nuclear data play a very important role in the choice of a radioisotope for a medical application. Nuclear structure and the decay data determine the suitability of a radioisotope for diagnostic application while the nuclear reaction data study the possibility of its production in a pure form. The production of non-conventional radionuclides, however, demands detailed nuclear data work covering both experimental investigations and nuclear model calculations. Radioisotopes produced by charged-particle nuclear reactions find important applications in medicine [1-3]. The radionuclide ^{159}Dy ($T_{1/2}=144$ d, EC =100%) is a pure Auger electron and X-ray emitter and has been gained interest in transmission imaging and bone mineral analysis. On the other hand the radionuclide ^{139}Ce ($T_{1/2}=137.6$ d, EC=100%)

is convenient as a standard source for calibration of γ -ray. Recently the ^{139}Ce line source was used to determine attenuation maps for single photon emission computer tomography (SPECT) [4]. In the present work the cross sections of the $^{159}\text{Tb}(p,n)^{159}\text{Dy}$ and $^{139}\text{La}(p,n)^{139}\text{Ce}$ reactions were measured using samples of natural terbium and lanthanum (isotopic abundance:100% and 99.91% respectively). The possibility of producing ^{159}Dy and ^{139}Ce radioisotopes using the MGC-20 cyclotron is studied.

EXPERIMENTAL

Sample Preparation

The convention stacked-foil technique was applied for cross-section measurements. Thin layers of terbium oxide, Tb_4O_7 (purity 99.9%; supplied by Russland Co.), and lanthanum oxide, La_2O_3 (purity 99.9999%; supplied by Hicol b.v.-chemicals), on a high purity aluminum backing of thickness 250 μm were prepared by sedimentation technique [5,6]. After drying the samples, accurate thicknesses of the sedimented layer were determined by weighing each aluminum foil and subtract its weight before sedimentation. Each sample was covered with 10 μm Al foil to protect the formed layer in the stack during irradiation and measurements.

Irradiation

Three stacks of terbium and one of lanthanum were irradiated with 18 MeV and 14.7 MeV, respectively, on the beam line of the MGC-20 cyclotron, Atomic Energy Authority, Cairo, Egypt. Irradiation time was 3 hours for each stack and the incident beam current was 300-500 nA. Several beam current monitors of 50 μm Cu foils were placed in the stack to determine the actual beam intensity on the samples. The cross section data of the monitor reactions $^{\text{nat}}\text{Cu}(p, nx)^{62,63}\text{Zn}$ were taken from the literature [7]. Knowing the density and isotopic composition of the samples, energy degradation within the stack was calculated using the computer program (STACK) which is based on tabulated data for stopping power of charged particles [8,9]. The decay characteristics obtained from table of isotopes [10] for ^{159}Dy and ^{139}Ce are given in table (1).

Table (1) Decay characteristic of the investigated reactions product

Produced nuclide	$T_{1/2}$	E_γ (keV)	Mode of decay	I_γ (%)
^{159}Dy	144.4 d	58.00	EC (100%)	2.22
^{139}Ce	137.64 d	165.86	EC (100%)	80.0

Measurement of Radioactivity

The activity of the irradiated samples was measured non destructively without chemical separation. The gamma ray spectrometer consists of HPGGe detector model CANBERRA-Gr7023 of relative efficiency 50% connected to multichannel analyzer module (TRUMB-

8192) installed in PC. The resulting spectra were analyzed using the software Gamma Vision, version 5.1. The counting time was adjusted to get a good counting statistics. The measured energy resolution was 2.0 keV for ^{60}Co gamma ray line of energy 1332.5 keV. To reduce the background, the detector was shielded with a lead cylinder of thickness 10 cm having an additional low activity internal copper shield inside. Absolute efficiency curves for the detector were deduced using standard gamma ray sources at the same experimental conditions of the measured spectra.

Calculation of the Nuclear Reactions Cross Section

The absolute activity values of ^{159}Dy and ^{139}Ce at EOB were determined using their characteristic γ - ray spectrum. Since both Tb and La are mono-isotopic elements, any correction for isotopic abundance of the target was not necessary. The reaction cross-section was calculated using the well-known activation formula depending on the decay rates [11]. The sources of uncertainty in the measured cross sections were estimated and found to be: beam current 10%, detector efficiency 3-5%, statistical counting 1-2 % and decay data 1%. The overall uncertainty in the measured cross-section values amounts to be 15-18%.

Nuclear Model Calculation

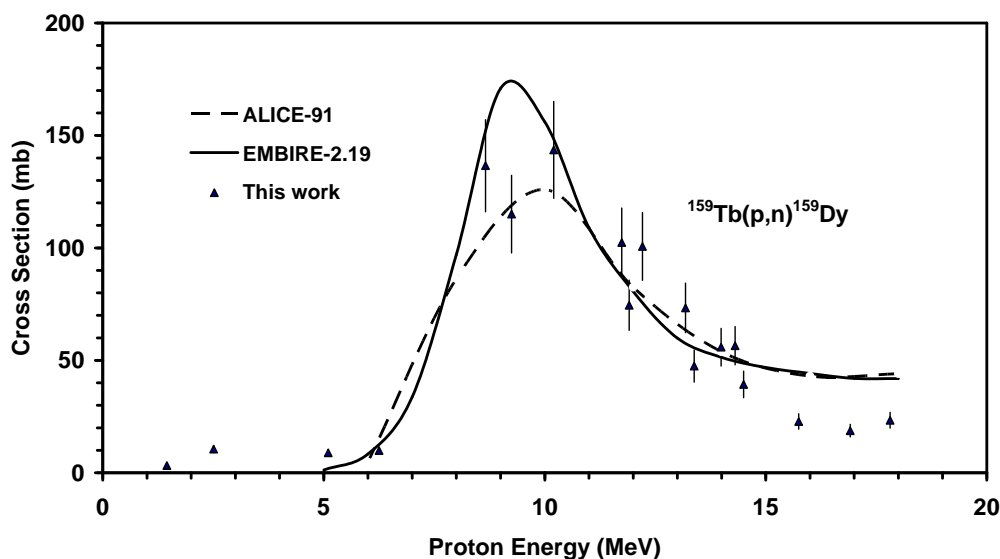
Nuclear model calculations are of considerable interest in studying the reactions producing radioisotopes, in particular in prediction of unknown reaction cross section data. In this work two computer codes have been used for calculating the reaction cross-sections theoretically, namely ALICE and IMPIRE. We used here ALICE-91 and its modified versions ALICE-IPPE. This code was originally developed by Blann in 1975 [12] and modified to ALICE-91 which is based on the hybrid, the geometry-dependent hybrid (GDH) or the hybrid Mont Carlo simulation (HMS) pre-equilibrium models and the Weisskopf-Ewing evaporation formalism [13]. The ALICE-IPPE code is a version of the ALICE code modified by the Obninsk group [14] to include the generalized super fluid level density model and pre-equilibrium cluster emission.

The EMPIRE-2.19 code [15] is based on double differential hybrid Monte Carlo simulation (HMS) routine which gives the levels population cross-sections of residual nuclei. The code enables flexible input and modulation of parameters including level density, optical model parameters (OPM) and level scheme. The cross section for the investigated nuclear reactions was calculated and compared with the experimental results. More details will be discussed below.

RESULTS AND DISCUSSION

$^{159}\text{Tb}(p,n)^{159}\text{Dy}$ reaction was measured in the frame of this work in the energy range from threshold energy up to 17.8 MeV. The experimentally determined cross section values are given in table (2) while fig.1 shows the representation of the excitation function for the investigated nuclear reaction. The measured excitation function shows a distinct maximum cross-section amounting to about 143.5 mb at 10.2 MeV proton energy. No earlier

experimental data were found in the literature. The theoretically calculated cross-section values are also presented in fig (1) for comparison. A satisfactory agreement is achieved between our results and those calculated using the two computer codes.



Fig(1) Excitation function for $^{159}\text{Tb}(p,n)^{159}\text{Dy}$ reaction

Table(2) Measured cross-sections for $^{159}\text{Tb}(p,n)^{159}\text{Dy}$ reaction.

Energy (MeV)	Cross Section (mb)
17.8 ± 1.2	23.4 ± 3.5
16.9 ± 1.3	18.7 ± 2.8
15.7 ± 1.1	22.8 ± 3.4
14.5 ± 0.9	39.3 ± 5.9
14.3 ± 1.1	56.5 ± 8.5
13.9 ± 0.8	55.5 ± 8.4
13.3 ± 0.9	47.5 ± 7.1
13.1 ± 1.2	73.4 ± 11.0
12.2 ± 1.1	100.7 ± 15.1
11.9 ± 1.4	74.5 ± 11.2
11.7 ± 1.2	102.4 ± 15.4
10.2 ± 0.7	143.5 ± 21.4
9.2 ± 0.9	115.0 ± 17.3
8.6 ± 1.3	136.5 ± 20.5
6.2 ± 1.5	9.9 ± 1.5
5.1 ± 1.4	8.9 ± 1.3
2.5 ± 0.9	10.5 ± 1.3
1.4 ± 1.0	3.2 ± 0.5

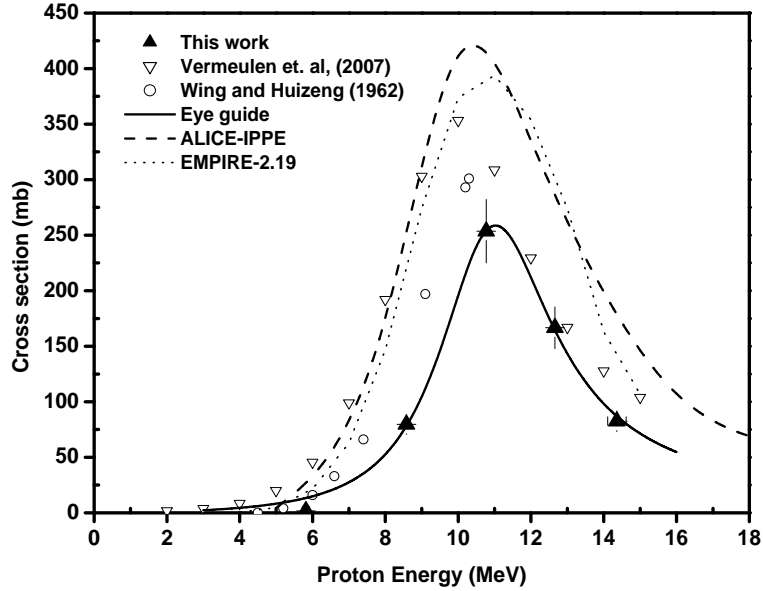


Fig.(2) Excitation function for $^{139}\text{La}(p,n)^{139}\text{Ce}$ reaction

The reaction cross-section and excitation function for $^{139}\text{La}(p,n)^{139}\text{Ce}$ were measured in the proton energy range 1.0 MeV up to 14.36 MeV. Table (3) gives the experimentally measured cross-sections and their uncertainty values. Fig (3) shows representation of the excitation function as compared with the previously available measured data. We have to mention that reaction of protons with ^{nat}La populates two states in ^{139}Ce , the ground state which decays by 99.99% EC ($T_{1/2}=137.64\text{d}$) to ^{139}La and the isomeric state with ($T_{1/2}=54.8\text{ sec}$) which decays internally by M4 γ -ray transition to the ^{139}Ce ground state. Hence the cross-section presented in fig (3) is the cumulative cross-section $\bar{\sigma}_{m+g}$. It is clear that there is an acceptable agreement between our results and those obtained by Vermeulen et al., 2007 [16], in the high energy range of the curve while their values are higher in the low energy part of the curve. We have to mention also that the distribution in the measured excitation function is narrower than that reported by them. This difference may be explained due to the technique they used for obtaining the excitation function, where thick-target yields were measured in a different energy windows in order to establish a thick-target production rate curve. The corresponding excitation function was then extracted by means of differential procedure. Theoretical calculation using ALICE-IPPE code was also carried out and presented in the figure for comparison. The shape of the excitation function curve and the position of the maximum cross-section value measured experimentally agree with the theoretical calculation but with a lower absolute value. The results obtained previously by Wing et al., 1962 [17], lie between our values and those obtained by Vermeulen et al., 2006.

Table(3) Cross section values for the reaction $^{139}\text{La}(p,n)^{139}\text{Ce}$

Energy (MeV)	Cross Section (mb)
14.4 ± 0.3	82.5 ± 9.3
12.7 ± 0.1	166.7 ± 18.8
10.8 ± 0.1	253.6 ± 28.6
8.6 ± 0.1	79.6 ± 8.0
5.8 ± 0.1	1.7 ± 0.2

Calculation of Integral Yields

From the measured excitation function of the nuclear reactions $^{159}\text{Tb}(p,n)^{159}\text{Dy}$ and $^{139}\text{La}(p,n)^{139}\text{Ce}$, the yield of ^{159}Dy and ^{139}Ce radionuclides were calculated. The results are presented in fig. (4) and (5). The optimization of a nuclear process for the production of a radioisotope at a cyclotron involves a selection of the projectile energy range that will maximize the yield of the product and minimize that of radioactive impurities if present. From the excitation function curve of $^{159}\text{Tb}(p,n)^{159}\text{Dy}$, it is clear that the optimum energy range for the production of ^{159}Dy is from $E_p = 14 \rightarrow 6$ MeV. This gives integral yield of $0.9 \text{ MBq}/\mu\text{Ah}$. On the other hand the optimum energy range for the production of ^{139}Ce is $E_p = 14.3 \rightarrow 8$ MeV with a value of $8.57 \text{ MBq}/\mu\text{Ah}$ integral yield.

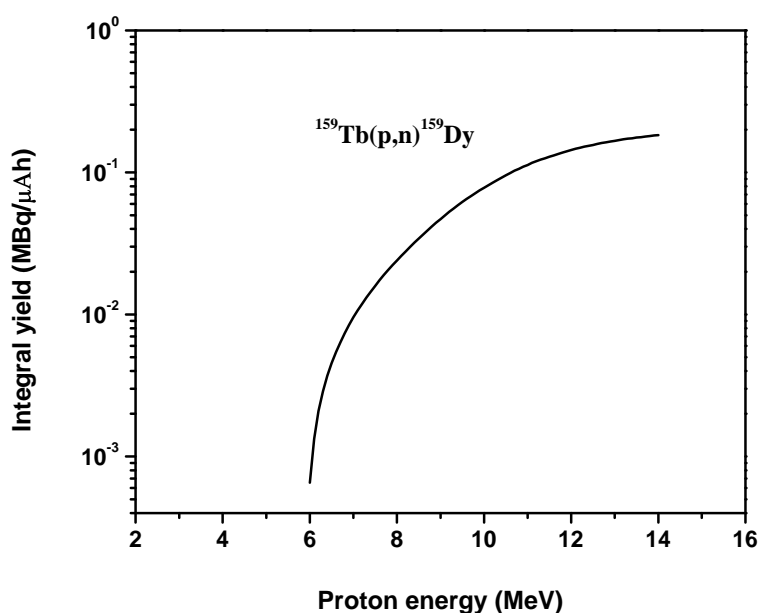
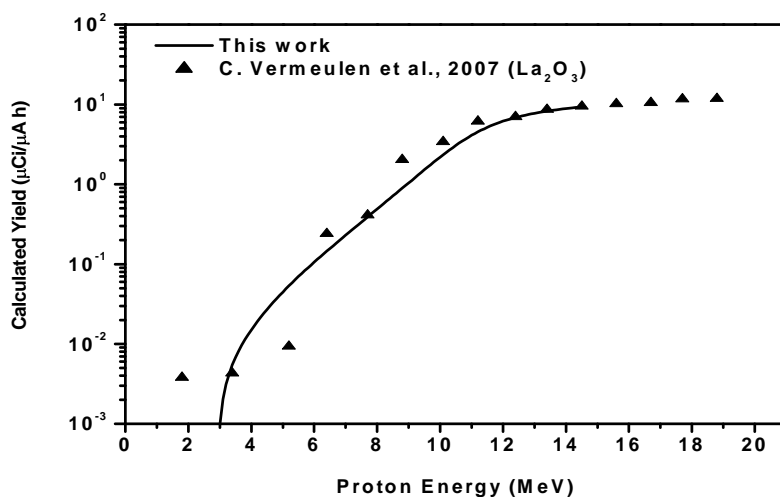


Fig (3) Integral yield for the production of ^{159}Dy



Fig(4) Integral yield for the production of ^{139}Ce

CONCLUSION

From a nuclear data point of view, both reactions $^{159}\text{Tb}(p,n)^{159}\text{Dy}$ and $^{139}\text{La}(p,n)^{139}\text{Ce}$ are suitable for producing ^{159}Dy and ^{139}Ce radionuclides using low energy cyclotrons. A satisfactory agreement was achieved between the present results and the literature data for the production of ^{139}Ce especially in the high energy range of the excitation function curve. Comparison with the theoretical calculations using ALICE-IPPE code shows that the theoretical calculation over predict the measured data. In case of $^{159}\text{Tb}(p,n)^{159}\text{Dy}$ reaction there are no previously reported data to compare with. Only the experimental results were compared with the theoretical calculations using two codes. The excitation function curve produced using ALICE-91 code is in good agreement with the experimental results up to proton energy 15 MeV. The EMPIRE-2.19 code over estimate the present data in the energy range $E_p=7\text{--}11\text{MeV}$.

ACKNOWLEDGMENT

The Authors are greatly indebted to Dr. E.K. Elmaghraby, experimental nuclear physics department, nuclear research center, AEA, Cairo, Egypt, for his help in carrying out the theoretical calculations using EMBIRE code.

REFERENCES

- [1] G. Stöcklin, S.M.Qaim, F. Röscher; *Radiochimica Acta*, 70/71, 249 (1995).
- [2] S.M. Qaim; *Radiochimica Acta*, **89**, 189 (2001).
- [3] S.M. Qaim; "Cyclotron production of medical radionuclides", *Handbook of Nuclear Chemistry*, Edited by A.Vértes, S.Nagy and Z. Klencsár, Kluwer Academic Publishers, The Netherlands, page 47 (2003).

- [4] H. du Raan, P.D. du Toit, A. van Aswegen, M.G. Lotter, C.P. Herbst, T.N. van der Walt, A.C. Otto; *Med. Phys.*, 27, 1523(2000).
- [5] H.E. Hassan, S.M. Qaim, Yu. Shubin, A. Azzam, M. Morsy and H.H. Coenen; *Appl. Radiat. and Isot.*; 60, 899 (2004).
- [6] F. R?sch, S.M. Qaim, G. St?cklin, *Radiochimica Acta*; **61**, 1(1993).
- [7] F. F. T?rk?nyi, S. Tak?cs, K. Gul, A. Hermanne, M.G. Mustafa, M. Nortier, P. Oblo?insk?y, S.M. Qaim, B. Scholten, Yu. N. Shubin, Z. Youxiang, *Beam Monitor Reactions in Charged Particle Cross-Section Database for Medical Radioisotope Production*. IAEA TECDOC-1211, IAEA, Vienna, 2001, p. 49. Available from: <<http://www-nds.iaea.org/medical>>.
- [8] C.F. Williamson, J.P. Boujot, J. Picard., "Tables of range and stopping power of chemical elements for charged particle of energy range 0.05 to 500 MeV", Report CEA-R-3042, Saclay, France, (1966).
- [9] H.H. Andersen, J.F. Ziegler, "Handbook of Stopping Cross-Sections of Energetic Ions and Elements", Vol. 3, Pergamon, New York, (1997).
- [10] R.B. Firestone, L.P. Eckstr?m, *WWWTable of Radioactive Isotopes*, Version 2.1, January 2004. URL: <<http://ie.lbl.gov/toi>>.
- [11] Sc.D. Frank Helus, G. Lelio, Sc.D. Colombetti; *Radionuclides production*, CRC press Inc., Boca Raton, Florida, (2000).
- [12] M. Blann; *Ann. Rev. Nucl. Sci.*; 25, 123(1975).
- [13] M. Blann; Lawrence Livermore National Laboratory Report; UCRL-JC-109052 (1991).
- [14] A.I. Dityuk, A.Yu. Konobeyev, V.P. Lunev and Yu.N. Shubin, "New Version of the Advanced Computer Code ALICE-IPPE", IAEA, Vienna, Report INDC(CCP)-410 (1998).
- [15] M. Herman , R. Capote, B. Carlson, P. Oblo?insk?y, M.Sin, A. Trakov, V. Zerkin, "EMPIRE-II, Nuclear reaction model code version 2.19", IAEA, Vienna, Austria, (2005).
- [16] C. Vermeulen, G.F. Steyn, F.M. Nortier, F. Szelecs?nyi, Z. Kov?cs, S.M. Qaim, Production of ^{139}Ce by proton-induced reactions on ^{141}Pr and $^{\text{nat}}\text{La}$; *Nucl. Instr. and Meth. B*, 255, 331(2007)
- [17] J. Wing, J.R. Huizenga, *Phys. Rev.* 128, 280(1962)

This document was created with Win2PDF available at <http://www.win2pdf.com>.
The unregistered version of Win2PDF is for evaluation or non-commercial use only.