East African Journal of Biophysical and Computational Sciences

Journal homepage : https://journals.hu.edu.et/hu-journals/index.php/eajbcs

Theoretical Model Predictions for Production of Medically Used Radionuclides on Alpha Induced with Cobalt-59 At Energies of 25 - 172 MeV

Y. Tiguaded, F. K. Amanuel

Department of Applied Physics, Hawassa University, P.O.Box 05, Hawassa, Ethiopia

KEYWORDS:

COMPLET code; Excitation function; Radionuclide production; Reaction cross section; TALYS-1.95 code

ABSTRACT

This study used the theoretical nuclear model codes COMPLET and TALYS-1.95 to make theoretical predictions of the medically important production cross-sections for Chromium-51, Manganese-54, Iron-59, Cobalt-59 and Cobalt-60 radionuclides produced in the interaction of alpha- projectile with Cobalt-59 target ≈ 25 – 172 MeV alpha-energies. The results were compared with the measured values in the EXFOR data library. Pearson's correlation coefficient indicates a strong and positive correlation between the predicted and the previously measured medically important production cross-sections for Chromium-51, Manganese-54, Iron-59, Cobalt-59, and Cobalt-60 radionuclides. Further, the results showed that except for Chromium-51, the COMPLET code predicts more successful outcomes than the TALYS-1.95.

INTRODUCTION

Nuclear reaction cross-section data are very important to the field of medical radiobiology in both diagnostic imaging and targeted therapy (Kebede, 2021), which is crucial for the optimized production of radionuclides. In nuclear medicine, radionuclides are used for various useful applications, such as diagnosis, therapy, prevention of many serious ailments, and research to evaluate metabolic, physiologic, and pathologic conditions of the human body (Aydin *et al*., 2007). The successful production and usage of these radionuclides extends to oncology, cardiology, and even psychiatry through imaging procedures where information about the function of every major organ and tissue of the human body can be generated. Many radionuclides used in nuclear medicine are produced in cyclotrons, accelerators, or nuclear reactors, and production is an important and constantly evolving issue. In addition to this, different radionuclides play significant roles in technological applications of importance to our daily lives and scientific research (Aydin *et al*., 2007; Kilinç *et al*.,2016).

**Corresponding author:*
Email: a_fessahatsion@yahoo.com +251911567435

https://dx.doi.org/10.4314/eajbcs.v5i1.1S

The Positron Emission Tomography (PET) imaging technique is widely used for planning, early diagnosis of cancer, and evaluation of the treatment response in patients with cancer. This imaging technique is also used to study diseases of the heart, brain, thyroid, etc. (Noori *et al*., 2017) for example, Cobalt-57($T_{1/2}$ =272 d) is used as a marker to estimate organ size and for in vitro diagnostic kits. Similarly, Chromium-51 radionuclide ($T_{1/2}$=28 d) is used to label red blood cells and quantify gastro-intestinal protein loss (Aydin *et al*., 2007; Kilinç *et al*., 2016).

Production cross sections for charged particles, especially nuclear reactions on metals that are induced by alpha, are important in medical radioisotope production (Mohamed, 2006; Demir *et al*., 2017). Accordingly, reasonable comparative theoretical reaction model studies with an experiment using light-charged projectiles (proton, deuteron, and alpha) are beneficial (Qaim *et al*., 2016; Tárkányi *et al*., 2019; Amanuel, 2023) because of the non-availability of experimental cross-section data for the production of medical radionuclides, particularly in alpha-induced reactions, which are limited and still need further investigations.

To optimize the production routes, charged particle-induced cross-sections are desired. To optimize the radioisotope produced, a full knowledge of the excitation function is necessary, which helps maximize the yield of the desired product and minimize the radioactive impurities (Qaim *et al*., 2002).

In radionuclide production, accurate reaction cross-section data are required for well-controlled and maximized production routes (Mohamed, 2006; Qaim, 2010). Nuclear reaction model-based computer codes can be essential in predicting production cross-sections, particularly for radionuclides whose experimental data are either unavailable or have significant discrepancies. In addition, theoretical model predictions have played a crucial role in creating optimized reference cross-section data, particularly in producing medically useful radionuclides (Koning *et al*., 2013) that were calculated using the Monte Carlo nuclear reaction simulation codes TALYS 1.95 and COMPLETE.

Despite all efforts, one of the crucial aspects of the reaction mechanisms study is finding optimized production routes for radionuclides, particularly for medically used radionuclides. Moreover, it is evident that the non-availability of experimental cross-section data for producing medically useful radionuclides, particularly in alpha-induced reaction, are very limited and need further investigation. Therefore, the present work focuses on finding optimized production routes for medically useful radionuclides produced in the reaction of α-projectile with ^{59}Co-target, more specifically, to evaluate the nuclear data for the production of Chromium-51, Manganese-54, Iron-59, Cobalt-59, and Cobalt-60 on alpha-induced Cobalt-59 at alpha energy of positron-emitting radionuclides.

MATERIALS AND METHODS

Several theoretical nuclear reaction model-based computer codes have been used to predict radionuclide production cross-sections (Koning *et al*., 2013; Amanuel, 2023). In this work, predictions of production cross sections for ^{51}Cr, ^{54}Mn, ^{59}Fe, $^{\text{and}}$ ^{57,60}Co radionuclides produced in the interaction of alpha-projectile with Cobalt-59 target via (a, x) channel were carried out using the computer codes TALYS-

1.95 and COMPLETE. The results were compared with the experimental data (Michel *et al*., 1980).

These codes were selected because they have been successful and widely used for predicting production cross-sections and evaluating reaction data (Amanuel, 2023). The present work also used the default values of the level density nuclear model TALYS-1.95 computer codes.

TALYS-1.95 Code

TALYS-1.95 code is an advanced version of the TALYS code family with additional features. TALYS was initially developed in 1998 when it was decided to implement the combined knowledge of nuclear reactions into one single software package which integrates the pre-equilibrium, direct, optical model, statistical, and fission nuclear reaction models and for all the open reaction channels it gives prediction in one calculation scheme (Qaim *et al*., 2016). A Monte Carlo reaction code that simulates all types of nuclear reactions; it runs on a Linux operation system and is written in the FORTRAN programming language. One of the possible outcomes of using a Monte Carlo method for nuclear data evaluation is that a series of correlations can be extracted from the previous results. Therefore, the objective and vision of its construction were to provide a complete and accurate simulation of nuclear reactions that involve neutrons, photons, protons, deuterons, tritons, ^{3}He, and alpha particles in the 1 keV-200 MeV energy range, with some exceptions. The code's data was based on the reference input parameter library through an optimal combination of reliable nuclear models, resilience, and ease of use (Koning *et al*., 2013).

The theory of excitation functions for the production of medical radioisotopes are obtained with alpha-induced reactions (a,x) for some radioisotopes used in medicine that are important for the development of improved nuclear reaction theory and for many medical applications were calculated by using TALYS 1.95 code. In this code, the reaction cross section for entrance channel α and exit channel β can be expressed, in general, using Hauser-Feshback (Hauser-Feshback, 1952) formalism as follows:

$$\sigma_{\alpha\beta} = \frac{\pi}{k^2} \sum_J \frac{(2J+1)}{(2i_\alpha+1)(2I_\alpha+1)} \frac{\sum_{s,\ell} T_\ell(\alpha) \sum_{s',\ell'} T_{\ell'}(\beta)}{\sum_\alpha \sum_{s,\ell} T_\ell(\alpha)} \quad (1)$$

Where s is the channel spin, T_ℓ represents the transmission coefficients, and l is the orbital angular momentum. The Hauser-Feshback formula is simplest for the energy-average angle integrated cross-section of statistical reactions (reaction cross-section leading to a single final state).

COMPLETE code

Computer code COMPLETE is a modified and advanced version of the Alice code family with additional physics, corrections, and capabilities and has been used to predict production cross-sections (Asres *et al*., 2018). This code has successfully predicted numerous nuclear data sets, particularly for the production of medical radionuclides (Yiğit and Tel, 2013; Asres *et al*., 2019). This code employs the Weisskopf-Ewing (Weisskopf and Ewing, 1940) formulation for compound nucleus (CN) emission and the hybrid, as well as geometry-dependent hybrid, model of Blann for PE emission of particles

(Blann and Vonach, 1983). In the complete code, level densities of residual nuclei play an important role in deciding the shapes and absolute values of excitation functions (Akkoyun *et al*., 2015). This code uses the Weisskopf-Ewing formulation to predict reaction cross-sections as follows:

$$\sigma_\alpha \, d\varepsilon_\beta = \sigma_C(\alpha) \frac{(2i_\beta+1)u_\beta\varepsilon_\beta\sigma_C(\beta)\omega(u_\beta)d_\beta}{\sum_\alpha \int_0^{E_\alpha^m}(2i_\alpha+1)u_\alpha\varepsilon_\alpha\sigma_C(\alpha)\omega(u_\alpha)d_\alpha} \quad (2)$$

Where u_α is the reduced mass of the ejectile α, and $\sigma_C(\alpha)$ is the cross-section for the formation of the CN.

Comparison between experimental and Theoretical results

The theoretical and experimental reaction cross-sections are plotted against the projectile energies and shown in Figs. 1–5. Pearson's correlation coefficient quantifies the level of mutual statistical dependence between two variables (Baak *et al*., 2020). Typically, their values range from -1 to +1 or 0 to +1, where 0 means no statistical association, +1 means the strongest possible association, and −1 means the strongest negative relation.

In general, the data of this study have been analyzed after the theoretical data have been generated using the computer codes TALYS-1.95 and COMPLETE. The theoretical and experimental total cross-section results are compared using Pearson's correlation coefficient. Correlation is a measure of association between two variables. The mathematical description is given by Tárkányi *et al*. (2019).

$$R=\frac{\sum_{i=1}^{N}(X_{T_i}-\langle X_T\rangle)(X_{E_i}-\langle X_E\rangle)}{(N-1)(S_{XT})(S_{XE})} \quad (3)$$

Where;

$$\langle X_T\rangle=\frac{1}{N}\sum_i^N(X_{T_i}) \quad (4)$$

$$S_X=\sqrt{\frac{1}{N-1}\sum_{i-1}^{N}(X_{T_i}-\langle X_T\rangle)} \quad (5)$$

$$\langle X_E\rangle=\frac{1}{N}\sum_{i-1}^N(X_{E_i}) \quad (6)$$

$$S_X=\sqrt{\frac{1}{N-1}\sum_{i-1}^{N}(X_{E_i}-\langle X_E\rangle)} \quad (7)$$

Where R is the correlation coefficient and unit less, $\langle X_T\rangle$ and $\langle X_E\rangle$ are the mean theoretical and experimental reaction cross-sections, respectively, X_{T_i} and X_{E_i} are the theoretical and experimental total cross-sections of the i^{th} value, respectively, N is the number of the theoretical and experimental data, S_{XT} and S_{XE} are the standard deviations of the theoretical and experimental total cross-sections respectively. If $0 \leq R \leq 0.3$, the correlation is weak and positive, $0.3 \leq R \leq 0.7$ describes a moderate correlation, and $0.7 \leq R \leq 1$, the correlation is strong (Baak *et al*., 2020).

RESULTS AND DISCUSSION

The present work investigated the excitation functions of medically important Chromium-51, Mangenes-54, Iron-59, Cobalt-59, and Cobalt-60 radionuclides produced in the interaction of alpha-projectile with Cobalt-59 target at 25–172 MeV alpha-energies. In addition, the experimentally measured excitation functions available in the literature (Michel *et al*., 1980) were compared using the nuclear reaction-model codes TALYS-1.95 and COMPLET.

In COMPLETE code, the level density parameter a, which predominantly affects the equilibrium state components of a cross-section, is calculated from the expression:

$$a = \frac{A_C}{K} \quad (8)$$

Where *A* is the nucleon number of *a* compound system, and *K* is an adjustable constant, which may be varied to match the experimental data. For the present system, a representative ^{59}Co (α, x) ^{60}Co reaction, the value of K was varied (K = values of 8, 10, and 12 were used) to match the experimental data. A value of K = 10 in general reproduced satisfactorily experimentally measured EFs for Cobalt-60 residue. This value is consistently used for other residues populated in the interaction of the alpha-projectile with the target Cobalt-59. For the same representative Cobelt-60 residue, the initial exciton numbers n_o = 4 (2,2,0), 5 (2,2,1) were varied, and it was found that a value of n_o = 4 (2,2,0) better reproduced the measured excitation function (Michel and Brinkmann, 1980). For COMPLETE code prediction, K = 10 and n_o = 4 are consistently used for all residues populated in the interaction of the alpha-projectile with the target Cobalt-59.

Production of Chromium-51 radionuclide

The theoretically predicted and experimentally measured excitation functions for Crominum-51 radionuclide produced via the (α, x) channel in the interaction of alpha-projectile with the Cobalt-59 target are shown in Figure 1. By using the TALYS-1.95 code, predicted cross-section values, except at alpha energies of 90–120 MeV, are in very good agreement with the measurements of Michel and Brinkmann (1980). It may further be seen that COMPLETE code predicted cross-section values that were generally in satisfactory agreement with the measured values of Michel and Brinkmann (1980).

It may be observed from Table 1 that Pearson's correlation coefficient values for TALYS-1.95 and COMPLETE code predicted cross-section values confirm strong positive correlations between the theoretically predicted and experimentally measured production cross-sections (Michel and Brinkmann, 1980).

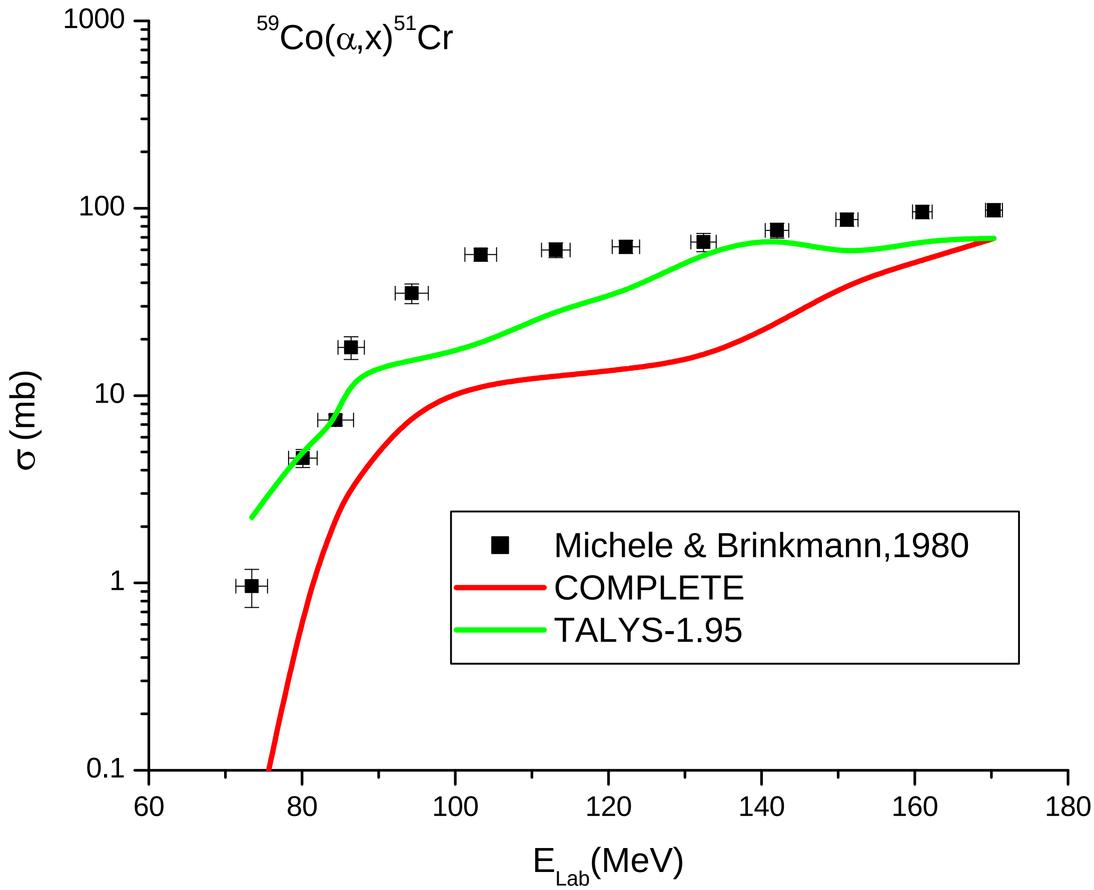

Figure 1. The experimentally measured and theoretically predicted excitation functions for medically used Crominum-51.

Production of Manganese-54 Radionuclide

The experimentally measured production cross-sections for Manganese-54 radionuclide from the literature are compared with the theoretical predictions obtained using the COMPLET and TALYS-1.95 codes. Fig. 2 displays the measured excitation functions along with theoretical predictions for Manganese-54radionuclide produced via the (α, x) channel in the interaction of alpha-projectile with a Cobalt-59 target at 25 MeV–172 MeV. Using COMPLETE code, predicted cross-section values except for 60 MeV–90 MeV are in very good agreement with the cross-section values measured by Michel and Brinkmann (1980). Figure 2 shows that the predicted cross-section values using the TALYS-1.95 code generally underestimate the measured values of Michel and Brinkmann (1980). The COMPLETE code predicted production cross sections for Manigenes-54 radionuclide to have a peak value of 111 mb at ≈170 MeV.

In addition, it may be observed from Table 1 that, for Manganese-54 radionuclide, Pearson's correlation coefficient values between theoretically predicted on COMPLETE and experimentally measured production cross sections confirmed moderately positive correlations. Pearson's correlation coefficient values between theoretically predicted using TALYS-1.95 and experimentally measured production cross-sections confirmed strong positive correlations.

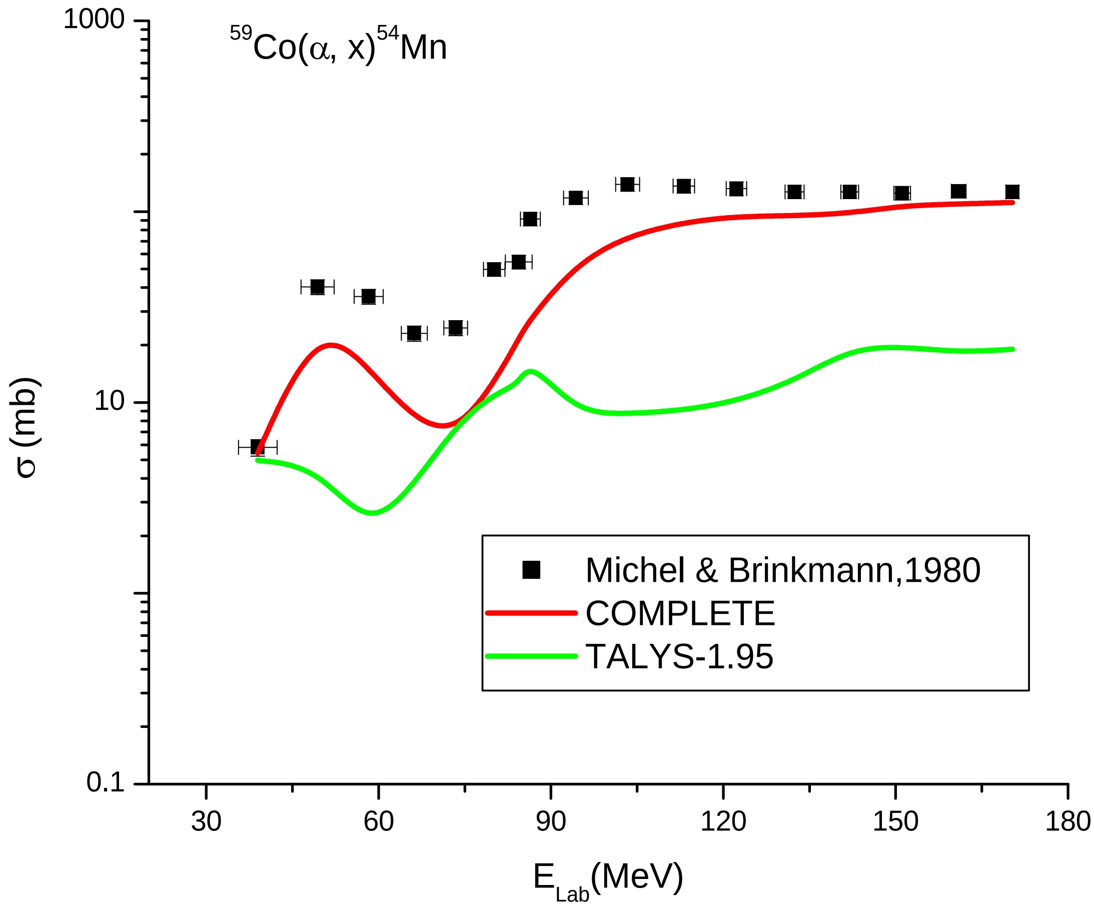

Figure 2. The experimentally measured and theoretically predicted excitation functions for medically used Manganese-54.

Production of Iron-59 radionuclide

Figure 3 displays the experimentally measured excitation functions along with theoretical predictions obtained using TALYS-1.95 and COMPLETE codes for Iron-59 radionuclide produced via (α, x) channel in the interaction of alpha-projectile with the Cobalt-59 target. Using COMPLETE code, the predicted cross-section values in the energy range 55–172 MeV generally agree with the experimental measurements of Michel and Brinkmann (1980). However, the prediction of COMPLETE code below 55 MeV underestimates the cross-section values measured by Michel and Brinkmann (1980). It may further be observed from Figure 3 that the predicted cross-section values at low energy using the TALYS-1.95 code are in satisfactory agreement with the measured cross-section values of Michel and Brinkmann (1980). On the contrary, the predicted cross-section values at a high energy range using the TALYS-1.95 code underestimate the measured cross-section values of Michel and Brinkmann (1980).

In addition, it may further be observed from Table 1 that Pearson's correlation coefficient values for Iron-59 radionuclide confirmed strong and positive correlations between the cross-section values predicted using the COMPLETE code and the values measured by Michel and Brinkmann (1980).

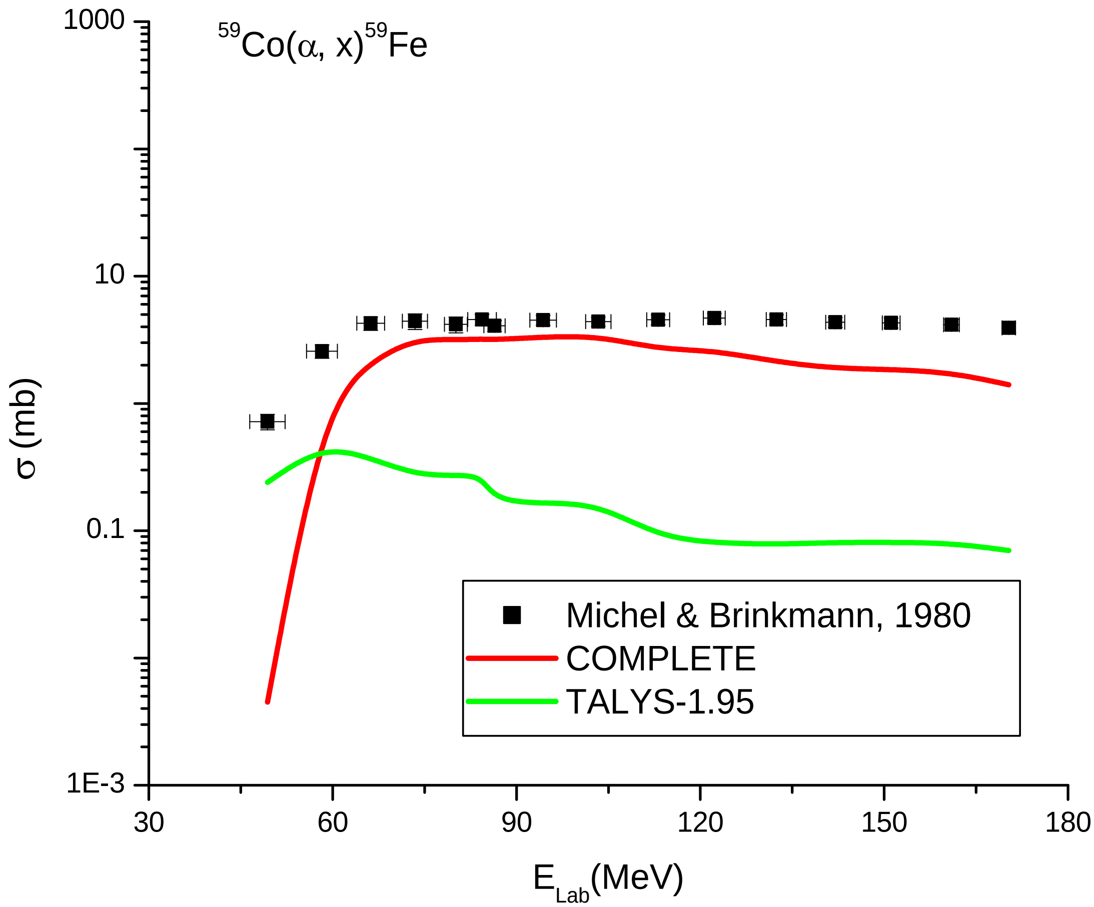

Figure 3. The experimentally measured and theoretically predicted excitation functions for medically used Iron-59.

Production of Cobalt-57 radionuclide

In Figure 4, the measured excitation functions for the medically used Cobalt-57 radionuclide produced via the complex (α, x) channel in the interaction of alpha-projectile with Cobalt-59 target are displayed together with the theoretically predicted excitation functions by using the TALYS-1.95 and COMPLETE codes. Using COMPLETE code, predicted production cross-sections are in very good agreement with the measured values of Michel and Brinkmann (1980). Using the TALYS-1.95 code, predicted production cross sections in the energy range 30 MeV–90 MeV are in very good agreement with the measured values (Michel and Brinkmann, 1980). However, predicted production cross sections using the TALYS-1.95 code at energies above 90 MeV are in satisfactory agreement with the measured values of Michel and Brinkmann (1980). The COMPLETE predicted production cross sections for Cobalt-57 radionuclide have a maximum value of ≈207 mb at ≈39 MeV. In addition, it may further be observed from Table 1 that Pearson's correlation coefficient values for Cobalt-57 radionuclide confirmed strong and positive correlations between the cross-section values predicted and the values measured by Michel and Brinkmann (1980).

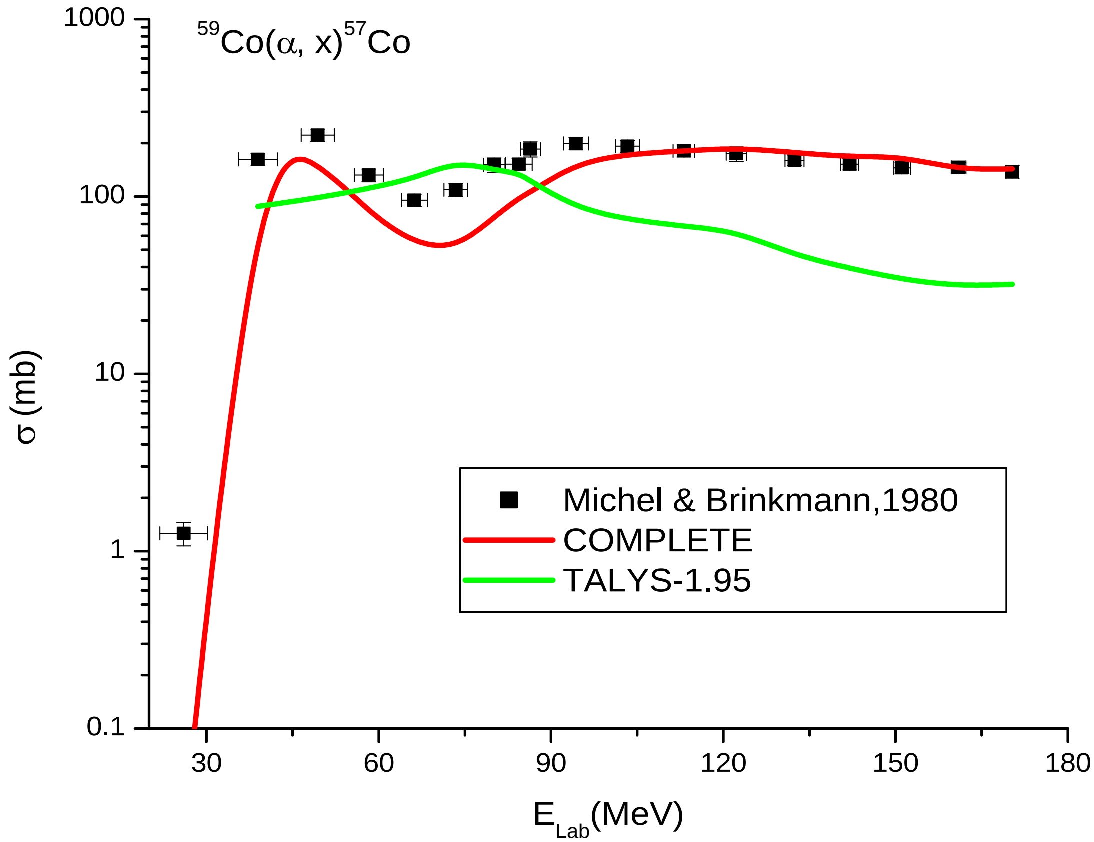

Figure 4. The experimentally measured and theoretically predicted excitation functions for medically used Cobalt-57

Production of Cobalt-60 Radionuclide

Figure 5 displays the experimentally measured and theoretically predicted excitation functions for the medically used Cobalt-60 radionuclide produced in alpha-projectile interaction with the Cobalt-59 target via complex (α, x) channel. The cross-section values predicted using COMPLETE codes overestimate the cross-section values measured by Michel and

Brinkmann (1980). However, predicted production cross sections in the energy range 30 MeV – 45 MeV are in very good agreement with the measured values of Michel and Brinkmann (1980). The prediction of the TALYS-1.95 code underestimates the cross-section values of the measurement of Michel and Brinkmann (1980). In addition, the maximum value of the production cross section for Cobalt-60 radionuclide obtained using the COMPLETE code is about ≈343 mb at ≈39 MeV. It may further be observed from Table 1 that Pearson's correlation coefficient values between theoretically predicted, and experimentally measured production cross-sections confirmed strong and positive associations.

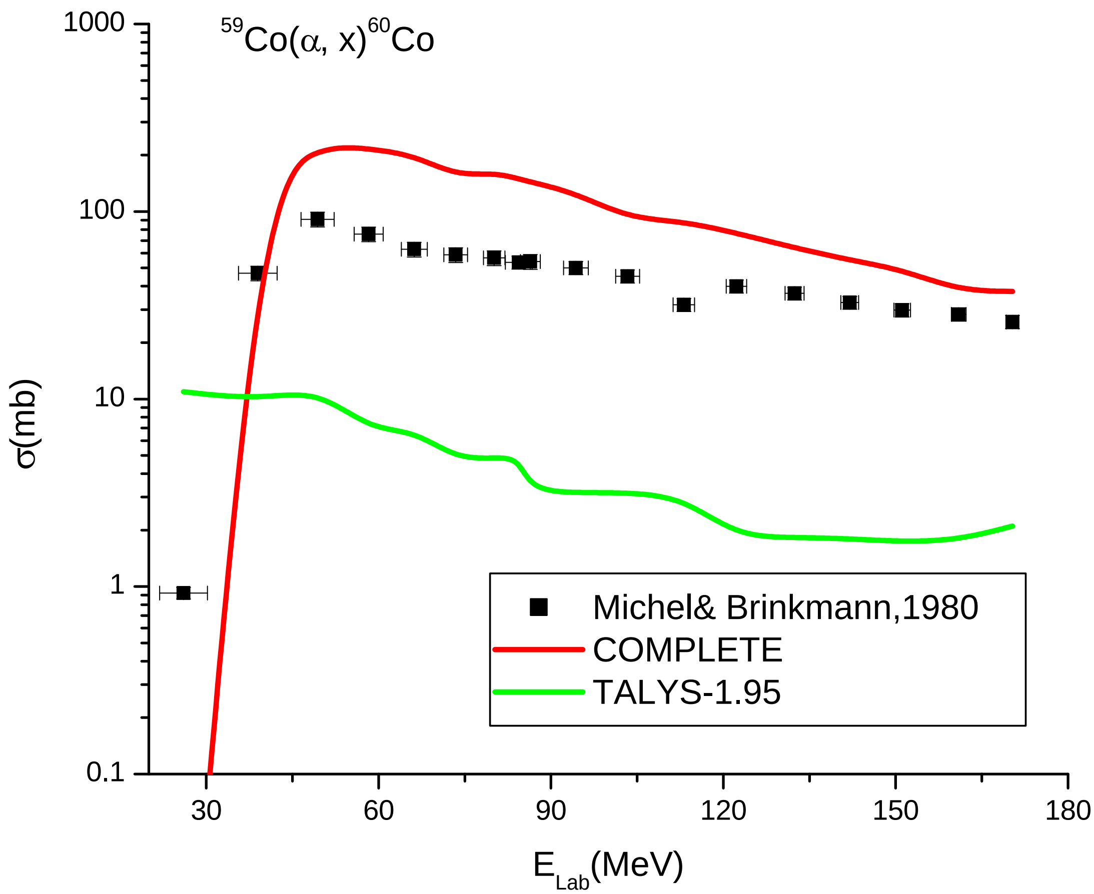

Figure 5. The experimentally measured and theoretically predicted excitation functions for medically used Cobalt-60.

Table 1. Pearson's correlation coefficient, R, between experimental measurements by Michel and Brinkmann (1980) and theoretical predictions

Radionuclide	TALYS-1.95 [COMPLETE]
^{51}Cr	0.921 [0.86]
^{54}Mn	0.65 [0.92]
^{59}Fe	- [0.84]
^{57}Co	0.5 [0.76]
^{60}Co	0.4 [0.96]

CONCLUSION

Excitation functions for the production of radionuclides from the alpha-bombardment of Cobalt-59 were studied for alpha-energies from 25 to 172.5 MeV. The theoretical nuclear reaction model codes COMPLET and TALYS-1.95 were used to make predictions of the medically important production cross-sections for Chromium-51, Mangenes-54, Iron-59, Cobalt-59, and Cobalt-60 radionuclides produced in the interaction of alpha-projectile with Cobalt-59 target ≈ 25 - 172 MeV alpha-energies. The results were compared with the measured values in the EXFOR data library. Pearson's correlation coefficient indicates a strong and positive correlation between the predicted and previously measured production cross-sections for medically important Chromium-51, Mangenes-54, Iron-59, Cobalt-59, and Cobalt-60 radionuclides. Further, the results show that except for Chromium-51, the COMPLETE code predicts more successful outcomes than the TALYS-1.95.

Acknowledgment

The authors thank Dr Zelalem A. for his helpful scientific discussions on the present work. However, all opinions and any errors are the author's responsibility alone.

References

Akkoyun S., Bayram T. and Kara S. O. 2015. Photonuclear Reaction Cross Sections for Gallium Isotopes. In Journal of Physics: Conference Series Vol. 590, No. 1, IOP Publishing.

Amanuel K. F. 2023. Production of 68Ge, 68Ga, 67Ga, 65Zn, and 64Cu important radionuclides for medical applications: theoretical model predictions for α-particles with 66Zn at≈ 10–40 MeV. *Radiochim. Acta*. **111**(3): 159-167.

Asres Y. H., Mathuthu M. and Birhane M. D. 2018. Analysis of reaction cross-section production in neutron induced fission reactions on uranium isotope using computer code COMPLET. *Appl. Radiat. Isot*. **139**: 81-85.

Asres Y. H., Mathuthu M. and Ferede Y. A. 2019. Investigation of nuclear reaction mechanisms of Nickel isotopes at various energies induced by alpha particles. *J. Phys. Commun*. **3**(11): 115018.

Aydin A., Şarer B. and Tel E. 2007. New calculation of excitation functions of proton-induced reactions in some medical isotopes of Cu, Zn and Ga. *Appl. Radiat. Isot*. **65**(3): 365-370.

Baak M., Koopman R., Snoek H. and Klous S. 2020. A new correlation coefficient between categorical, ordinal and interval variables with Pearson characteristics. *Comput. Stat. Data Anal*. **152**: 107043.

Blann M. and Vonach H. K. 1983. Global test of modified pre-compound decay models. *Phys. Rev. C*. **28**: 1475–1492.

Demir N. K., Çetin B., Akkurt İ. and Noori S. S. 2017. Calculations of Double Differential Cross Sections on 56Fe, 63Cu and 90Zr Neutron Emission in Proton Induced Reactions. *Acta Physica. Polonica. A.* 132:1181-1185.

Girma A. and Amanuel F. K. G. 2019. Investigating entrance channel influences in the fusion of some heavy–ion systems. *J. Appl. BiotechnolBioeng*. **6**(5): 247-252.

Hauser W., Feshbach H. 1952. The inelastic scattering of neutrons. Phys. Rev. C87, 366–373.

Kebede B. Z. 2021. Investigation of α-induced Reaction on Copper Isotopes for Energy Range of 15-50 MeV. *Nucl. Sci.* **6**(2): 12-17.

Kilinç F., Karpuz N. and Çetın B. 2016. Calculation of the (p, n) reaction cross section of radionuclides used for PET applications. *Acta. Physica. Polonica A.* **130**(1): 318-319.

Koning A., Hilaire S. and Goriely S. 2013. Talys-1.6 a nuclear reaction program. User Manual, NRG, The Netherlands.

Michel R. and Brinkmann G. 1980. Alpha-induced reactions on cobalt. *Nucl. Phys. A.* **338**(1): 167-189.

Mohamed M. B. 2006. Study of the excitation functions for some cyclotron produced Radionuclides. *Open Chem.* **16**(1): 810-816.

Noori S., Akkurt İ. and Demir N. 2017. Comparison of Excitation Functions of Longer and Shorter Lived Radionuclides. *Acta. Physica. Polonica. A*. **132**(3): 1186-1188.

Qaim S. 2010. Radiochemical determination of nuclear data for theory and applications. *J. Radioanal. Nucl. Chem.* **284**(3): 489-505.

Qaim S. M., Spahn I., Scholten B. and Neumaier B. 2016. Uses of alpha particles, especially in nuclear reaction studies and medical radionuclide production. *Radiochim. Acta*. **104**(9): 601-624.

Qaim S.M., Tárkányi F.T., Obložinský P., Gul K., et al. 2002. Charged-Particle cross section database for medical radioisotope production. *J. Nucl. Sci. Technol*. **39**: 1282–1285.

Tárkányi, F.T., Ignatyuk, A.V., Hermanne, A. *et al.* 2019. Recommended nuclear data for medical radioisotope production: diagnostic positron emitters. *J. Radioanal. Nucl. Chem.* **319:** 533–666.

Weisskopf V. F. and Ewing D. H. 1940. On the yield of nuclear reactions with heavy elements. *Phys. Rev*. **57**: 472–485.

Yiğit M. and Tel E. 2013. Alpha production cross sections for some target fusion structural materials up to 35 MeV. *J. Fusion Energy*. 32, 442-450.

.