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COMPARISON OF NUCLEAR DATA OF ^{64}Cu PRODUCTION USING AN ACCELERATOR BY TALYS 1.0 CODE

by

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The extensive use of ^{64}Cu ($T_{1/2}$ = 12.7 h) as a positron and electron emitter radioisotope in recent years has ensured its potential to serve a dual role in the development of molecular agents in PET and radioimmunotheraphy drugs in oncology. The TALYS 1.0 code was used to calculate excitation functions for induced proton, deuteron and alfa-particles on ^{64}Zn, ^{66}Zn, ^{67}Zn, ^{68}Zn, ^{70}Zn, ^{62}Ni, and ^{64}Ni up to 50 MeV. According to the data acquired by the TALYS 1.0 code, thick-target integral yield of the induced charged particles on the enriched targets was achieved.

Key words: ^{64}Cu, TALYS-1.0, excitation function, integral yield

INTRODUCTION

Due to extensive applications of the ^{64}Cu radioisotope in radioimmunotheraphy (RIT) and as a positron emission tomography agent in PET imaging, various methods have been used to produce it. An additional reason for giving the production of trouble-free radiolabeling of ^{64}Cu extra consideration lies in insoluble complexes. ^{64}Cu is widely labeled as a DTPA-peptide, DOTA-peptide, or monoclonal antibody in colon cancer studies. Moreover, ^{64}Cu antibodies have shown promising results as RIT agents [1]. Research has also shown that ^{64}Cu-PTSM is a potential agent for tumor blood-flow quantification and hypoxia [2, 3].

The production of ^{64}Cu (of which physical-decay parameters are to be sought in $T_{1/2}$ = 12.7 h, E_β = 0.58 MeV, E_β = 0.66 MeV, I_β = 38.4%, I_β = 17.8 %, IEC = = 43.8% [4]) is mostly founded on proton and deuteron irradiation of enriched Zn and Ni targets by a cyclotron. Abbas *et al.* [5] used an enriched ^{64}Zn target to produce ^{64}Cu via deuteron induced on the target. Zweit *et al.* [6] produced no carrier added ^{64}Cu using an enriched ^{64}Ni target irradiated by deuteron particles. Szelesenyi *et al.* [7, 8] used the same target bombarded by proton particles. Cyclotron production of ^{64}Cu was also accomplished using $^{66/68}$Zn targets. The production of ^{64}Cu is possible by a reactor using fast and thermal neutron irradiations [5-13].

In this work, theoretical excitation functions of the ^{64}Cu production were calculated using probable methods, such as those within the 50 MeV particle energy range, using the TALYS 1.0 code as a systematic study of particle-induced activations of metal targets. Theoretical calculations of the production yield and calculations of the required thickness of the targets proved to suggest possible optimum reactions in the production of ^{64}Cu.

METHODS

Excitation functions of ^{64}Cu production via $^{66/67/68/70}$Zn + p, ^{64}Ni + p, $^{64/66/67/68}$Zn + d, ^{64}Ni + d, ^{62}Ni + α, and ^{64}Zn + n reactions were calculated by the TALYS 1.0 code of up to 50 MeV. An optimum energy range to avoid the formation of radionuclide impurities, decrease the excitation functions of inactive impurity production or, even, to avoid their presence altogether while taking full benefit of the excitation functions was determined. Therefore, the feasibility of the production of ^{64}Cu via various nuclear reactions was investigated. To determine the economical routine and theoretical calculations of nuclear reactions for predicting excitation functions, the TALYS 1.0 code developed by Koning *et al.* [14] for low/medium energy accelerators was used. TALYS 1.0 is a computer

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code system for the analysis and prediction of nuclear reactions. The basic objective behind its construction is the simulation of nuclear reactions that involve neutrons, photons, protons, deuterons, tritons, 3He-particle, and alpha-particles in the 1 keV-200 MeV energy range for target nuclides of mass 12 and heavier. Several optical potential models were used for protons. Cross-section calculations were performed using the framework of the Exciton model for equilibrium and pre-equilibrium emission of protons. For deuterons, tritons, ^{3}He, and alpha particles, a simplification of the folding approach of Watanabe was used [15, 16].

According to the SRIM (The Stopping and Range of Ions in Matter) code, the required thickness of the target was calculated for any reaction [17]. The physical thickness of the target layer is chosen in such a way that for a given beam/target angle geometry (90°), the incident beam of the target layer be excited with the predicted energy. To minimize the thickness of the target layer, 6° geometry is preferred; so required layer thickness will be less with the coefficient of 0.1.

The thick target integral yields were deduced using measured cross-sections by the Simpson numerical integral method from eq. 1. As the yield equation states, the enhancement of incident energy, beam current and use of an enriched target increase the yield as follows

$$\text{Yield} \quad H(1 \quad e^{\lambda t}) I/M \; [\sigma(E)/S_P(E)]\mathrm{d}E \quad [\text{mCi} \quad \text{A}^{-1}\text{h}^{-1}]^{*}$$

where H is the isotope abundance [%], M – the mass number [g], (E) – the cross-section [mb]**, S_p – the stopping power [MeVcm^{2}mg^{-1}], I – the beam current [A], and t – the bombardment time [h].

RESULTS AND DISCUSSION

^{64}Cu production practicability via proton particles

^{66}Zn(p, x)^{64}Cu. A beneficial range of proton energy for producing ^{64}Cu from a ^{66}Zn target is 33 to 43 MeV of a maximum cross-section of 80.238E-03 b in 41 MeV. The obtained production yield of ^{64}Cu in the chosen energy range is 6.43 mCi/μAh; nevertheless, ^{65}Zn (244.26 d) and ^{64}Zn (stable) radiochemical impurities of the cross-sections are considerably higher than (^{64}Cu) generates in the said energy range. The recommended thickness of the target is 140.0 μm for geometry 6° of the beam toward target, in agreement with the optimum energy range of 33-43 MeV, using the SRIM code.

^{67}Zn(p, α)^{64}Cu. The excitation function data acquired from the code predicted that target irradiation lead to the creation of radioactive and inactive impurities of equal or higher cross-sections compared to the entire utilizable energy range of ^{64}Cu, which could make it an unsuitable method for producing ^{64}Cu. The maximum cross-section obtained, that of 39.74E-03 b in 14 MeV energy and theoretical thick-target yields, is 1.10 mCi/μAh. The recommended thickness of the target is 44.38 m.

^{68}Zn(p, x)^{64}Cu. The excitation functions of ^{64}Cu production via the ^{68}Zn (*p, x*)^{64}Cu reaction in the energy range of 20 to 30 MeV, exploitable to produce ^{64}Cu, showed that this reaction appears not to be suitable for impurities produced through very high cross-sections. The maximum cross-section of the copper-64 radioisotope is 58.63E-03 b in the 25 MeV energy range and the theoretical thick-target yield is 3.46 mCi/μAh. The recommended thickness of the target is 102.20 m.

^{70}Zn(p, x)^{64}Cu. Proton irradiation of the ^{70}Zn target produces isotopic and non-isotopic impurities of equal or higher cross-sections compared to the entire utilizable range of ^{64}Cu. The maximum cross-section gain of 39.87E-03 b in the 46 MeV energy range and theoretical thick-target yield is 3.50 mCi/μAh. Within this energy range in its entirety, radionuclidic impurities by this method consist of ^{66}Zn (stable), ^{67}Zn (stable), ^{68}Zn (stable), ^{69}Zn (56.4 m), ^{70}Zn (5 10^{14} y), ^{67}Ga (3.26 d), ^{68}Ga (67.629 m), and ^{69}Ga (stable). The recommended thickness of the target is 159.0 m.

^{64}Ni(p, n)^{64}Cu. Induced protons on enriched ^{64}Ni resulted in isotopic and radionuclide impurities in the energy range of up to 50 MeV. Within the boundary of 3-11 MeV incident particle, ^{64}Ni (stable), ^{61}Co (1.65 h), and ^{64}Cu (12.7 h) are produced; the maximum probability of the reaction for producing ^{64}Cu (maximum cross-section of 767.25E-03 b) occurred in 10 MeV. The theoretical thick-target yield and optional thickness are 11.62 mCi/mA h and 23.89 μm, respectively (fig. 1).

^{64}Cu production practicability via deuteron irradiation

^{64}Zn(d, x)^{64}Cu. In the reaction, 15-30 MeV projectile energies are purposed to decrease the produc-

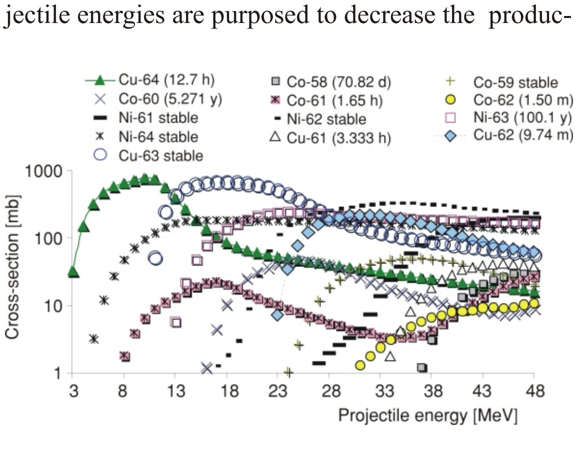

Figure 1. Excitation function of ^{64}Ni(p, n)^{64}Cu reaction using TALYS-1.0 code

* 1 Ci = 3.7 10^{10} Bq
**1 b = 10^{-28} m^2

tion of impurities. The maximum cross-section of 72.61E-03 b occurs in the 22 MeV energy range, with a theoretical thick-target yield of 3.20 mCi/mAh. This reaction is highly undesirable for generating ^{64}Zn (stable) with a maximum cross-section of 1080E-03 b in 18 MeV. Instead of the desired result, ^{65}Zn (244.26 d), ^{65}Ga (15.2 min), ^{64}Ga (2.63 min), ^{63}Zn (38.47 min) and ^{63}Cu (stable) with non-connivent excitation functions are produced. The suggested thickness of the target is 84.82 m.

^{66}Zn(d, x)^{64}Cu. In order to produce ^{64}Cu during the deuteron irradiation of the target, the maximum cross-section of 25.88E-03 b is created in the 9 MeV energy range. The calculated yield is 0.16 mCi/ Ah. This reaction is unattractive for its high impurities, produced after bombardment. In the chosen energy range, ^{67}Zn (114.34E-03 b, stable), ^{66}Zn (673.62E-03 b, Stable), ^{67}Ga (239.62 E-03 b, 3.26 d), and ^{66}Ga (69.55 E-03 b, 9.49 h) will be produced.

^{67}Zn(d, x)^{64}Cu. The best range of deuterons' offered energy for the production of ^{64}Cu via this reaction was found to be in the 15-25 MeV range. The maximum cross-section obtained in the 18 MeW energy range was 45.137E-03 b, with a theoretical thick-target yield of 1.10 mCi/ Ah. The production of contaminants during this unwelcome reaction overshadows the desired result by far.

^{68}Zn(d, x)^{64}Cu. ^{64}Cu production in the 30-50 MeV energy range is limited by its poor cross-sections; in 50 MeV, its cross-section is only 21.68E-03 b. This reaction, as said before, is not desirable at all.

^{64}Ni(d, 2n)^{64}Cu. When using the ^{64}Ni(d, 2n)^{64}Cu reaction to produce ^{64}Cu, the best range of incident energy was assumed to be that of 7-23 MeV. Maximum cross-section of ^{64}Cu was obtained 738.025E-03 b in 14 MeV. To take full benefit of the excitation function and to avoid the formation of the radionuclide and isotope impurities, energy range to produce no-carried added of ^{64}Cu should be 5-15 MeV. The physical thickness of the target and theoretical production yield in view of the chosen range are 38.73 μm and 11.59 mCi/mA·h, respectively (fig. 2).

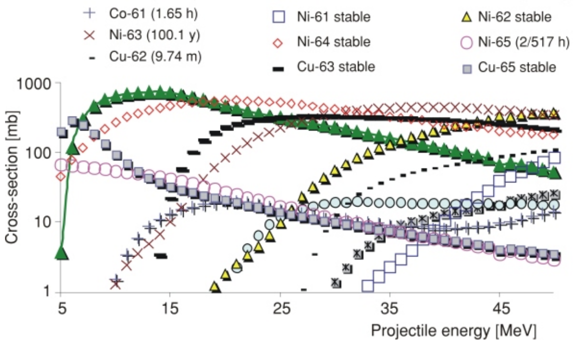

Figure 2. Excitation function of ^{64}Ni(d, 2n)^{64}Cu reaction using TALYS-1.0 code

^{64}Cu production practicability via alpha irradiation

^{62}Ni (, x)^{64}Cu. An induced alpha on the ^{62}Ni target makes a maximum cross-section of 383.159 mb in the 31 MeV range, along with the escorted radioisotope and isotope impurities. To decrease the production of such impurities, an energy range of 25-35 MeV can be chosen. Thus, the theoretical yield and target thickness in accordance with this energy range would be 2.05 mCi/μAh and 7.93 m, in that order (fig. 3).

^{64}Cu production practicability via fast neutron irradiation of ^{64}Zn

^{64}Zn (n, p)^{64}Cu. The feasibility of ^{64}Cu production using fast neutrons has shown that it can be produced with a maximum excitation function of 227.682E-03 b in the 11 MeV energy range. By means of this method, it is possible to produce a no-carrier-added radioisotope of ^{64}Cu. However, ^{64}Zn of cross-sections much higher than ^{64}Cu will be in all desirable regions for producing ^{64}Cu (fig. 4).

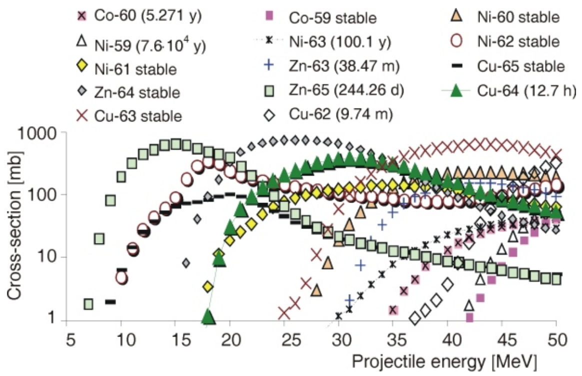

Figure 3. Excitation function of ^{62}Ni(, 2n)^{64}Cu reaction using TALYS-1.0 code

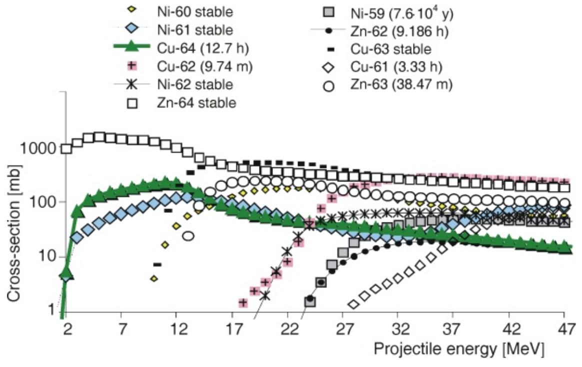

Figure 4. Excitation function of ^{64}Zn(n, p)^{64}Cu reaction using TALYS-1.0 code

TALYS 1.0 CODE CERTIFICATION

In order to define the precision of TALYS 1.0 code calculations, experimental excitation functions in several decay channels were compared with the TALYS 1.0 code data. The average error between the ^{64}Cu experimental excitation function and data acquired from the code through the induced proton on the ^{nat}Zn target was about 20% (fig. 5) [12,18-20].

Calculations predicted that one of the greatest yields will be achieved from deuteron bombardment of an enriched nickel-64 target (11.59 mCi/mAh), although ^{63}Cu (stable), ^{65}Cu (stable) and $^{65/64}$Ni (2.517 h, stable) are produced in the entire energy range; so, it is not possible to produce ^{64}Cu as a no-carrier-added product. Its no-carrier-added production is possible via ^{67}Zn(p, α)^{64}Cu nuclear reaction associated with $^{66/67}$Zn (stable) and ^{67}Ga (3.261 d) impurities; proton bombardment of ^{67}Zn (41% isotope abundance) has as a consequence a yield of 41.0 MBq/ A h at EOB; regardless of the brilliant characteristic of the reaction, a much higher cross-section of ^{67}Ga impurity than with ^{64}Cu makes this reaction undesirable. Production yield of ^{64}Cu via the ^{64}Ni(p, n)^{64}Cu reaction at 10 MeV proton energy is 11.62 mCi/mAh. There are ^{61}Co (1.65 h) ^{61}Ni (stable), and ^{64}Ni (stable) impurities with 6.61 mb and 96.71 mb cross-sections in this energy range, respectively, that are inconsiderable to compare ^{64}Cu production (767.254 mb). The production of copper-64 can be attained using a reactor. According to TALYS 1.0 computation data, it can be produced as a no-carrier-added using a fast neutron flux of up to 10 MeV. Only ^{64}Zn (stable) and ^{61}Ni (stable) impurities are present. The maximum excitation function of ^{64}Cu occurs by neutron flux of 11 MeV (227.682 mb); in the said energy, ^{63}Cu (stable) has an excitation function of about 69 mb. The choice of flux of 10 MeV to produce a no-carrier-added decreases the ^{64}Cu excitation function for about 5%. Many researchers have determined experimental excitation functions of ^{64}Cu production by the fast neutron flux. Their data is in good agreement with TALYS 1.0 code outcomes. For instance, Mannhart *et al.* [21] has reported a maximum excitation function of ^{64} Cu (228.5 mb) occurring in 12.69 MeV neutron energy, Xiaolong *et al.* [22] has reported an excitation function of 243 mb in 11.4 MeV and Qaim *et al.* [23] has reported it as 132 mb in 14 MeV.

^{66}Zn(p, x)^{64}Cu could not lead to the production of a no-carrier-added of ^{64}Cu while accompanied by ^{64}Zn (stable) and ^{65}Zn (244.26 d) impurities that have not relinquished their excitation functions. Despite the desirable yield of ^{64}Cu at EOB (237.9 MBq/ Ah), the needed high thickness (140 mm) of the target is another disadvantages of this method.

^{68}Zn(p, x)^{64}Cu, ^{70}Zn(p, x)^{64}Cu, ^{67}Zn(d, x)^{64}Cu, and ^{68}Zn(d, x)^{64}Cu reactions are much more undesirable than the prior ones not only because of the high thickness of the targets needed, but also because of the many impurities that go along with the product.

^{64}Cu production via induced alpha by ^{62}Ni(, x)^{64}Cu reaction can not result in no-carrier added ^{64}Cu production; there will be $^{59/61/62}$Ni (7.6 10^4 y, stable), $^{65/63}$Zn (244.26 d, 38.47 m), and $^{63/65}$Cu (stable) impurities. Finally, their poor yield makes these reactions unwanted for our purposes.

Hermanne *et al.* [10], showed that thick target yields for 20 MeV incident deuterons on highly enriched ^{64}Ni targets could reach 800 MBq/mAh which represents a 15% difference compared to our calculations (678.97 MBq/ Ah) using the TALYS 1.0 code (tab. 1). Daraban *et al.* [11] reported the acquired yield of 844 MBq/ Ah in the 9-20.5 MeV range via the same production method.

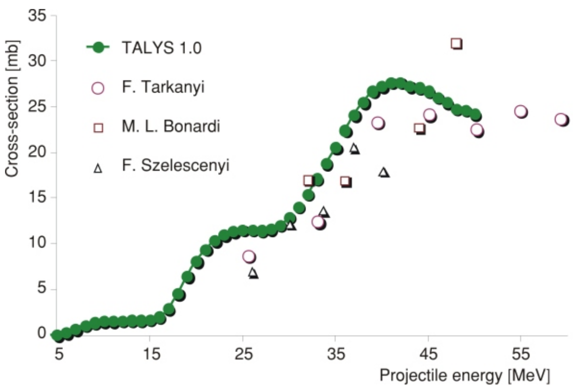

Figure 5. Excitation function of ^{nat}Zn(p, n)^{64}Cu reaction using TALYS-1.0 code

Table 1. Nuclear data of ^{64}Cu production via various nuclear reactions using TALYS 1.0 code

Reaction	Beneficial energy range [MeV]	Maximum cross-section [mb]	Theoretical yield [MBq/ A·h]	Target thickness [m]	Isotopic abundance [%]
^{66}Zn(p, x)^{64}Cu	33 43	80.23	6.43	140.00	27.90
^{67}Zn(p,)^{64}Cu	5 14	39.74	1.10	44.38	4.10
^{68}Zn(p, x)^{64}Cu	20 30	58.63	3.46	102.20	18.80
^{70}Zn(p, x)^{64}Cu	40 50	39.87	3.50	159.00	0.60
^{64}Ni(p, n)^{64}Cu	3 11	767.25	11.62	23.89	0.91
^{64}Zn(d, x)^{64}Cu	15 30	72.61	3.20	84.82	48.60
^{66}Zn(d, x)^{64}Cu	5 10	25.88	0.16	13.00	27.90
^{67}Zn(d, x)^{64}Cu	15 25	45.14	1.10	51.76	4.10
^{64}Ni(d,2n)^{64}Cu	5 15	738.02	11.59	23.81	0.91
^{62}Ni(, x)^{64}Cu	25 35	383.159	2.05	7.93	14.20
^{64}Zn(n, p)^{64}Cu	10-11	227.68	–	–	0.78

CONCLUSIONS

^{64}Zn(n, p)^{64}Cu using fast neutrons is one of the best methods for an attainable high yield and ^{64}Cu production with minimal impurities. ^{67}Zn(p,)^{64}Cu can be used as a routine method, but it needs a separation process on $^{67/66}$Zn and ^{67}Ga from ^{64}Cu; also, in the chosen energy range, there will be a little ^{63}Cu impurity of 18.17 mb maximum cross-section.

^{64}Ni(d, 2n)^{64}Cu result in a high yield, while Cu stable isotopic impurities will be produced along with ^{64}Cu radioisotope. Also, the ^{64}Ni(p, n)^{64}Cu process can be an efficient and economic route for the production of millicuries of the radioisotope.

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Махди САДЕГИ, Солалех СЕЈЕДИ, Зохрех ГОЛАМЗАДЕХ

ПОРЕЂЕЊЕ НУКЛЕАРНИХ ПОДАТАКА ПОМОЋУ TALYS 1.0 КОДА У АКЦЕЛЕРАТОРСКОЈ ПРОИЗВОДЊИ ^{64}Cu

Новије екстензивно коришћење ^{64}Cu ($T_{1/2}$ = 12.7 h) као позитронског и електронског емитера радиоизотопа, потврдило је његову двојну улогу у развоју молекуларних средстава за ПЕТ и лекова радиоимуне терапије у онкологији. TALYS 1.0 код употребљен је да се израчунавају ексцитационе функције индукованих протона, деутерона и алфа-честица на ^{64}Zn, ^{66}Zn, ^{67}Zn, ^{68}Zn, ^{70}Zn, ^{62}Ni и ^{64}Ni, до енергије од 50 MeV. На основу података прикупљених TALYS 1.0 кодом, постигнут је интегрални принос наелектрисаних честица индукованих на обогаћеним метама у виду дебелог штита.

Кључне речи: ^{64}Cu, TALYS 1.0, ексцитациона функција, интегрални принос