Journal of Neutron Research 22 (2020) 257–264
DOI 10.3233/JNR-190140
IOS Press

^{64}Cu production by 14 MeV neutron beam

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Abstract. ^{64}Cu is an emerging radionuclide of great interest in personalized nuclear medicine. It is produced by a cyclotron via the reaction ^{64}Ni(p,n)^{64}Cu. This production method increased during the last decades, because small biomedical cyclotrons can be easily installed close to the nuclear medicine department of a hospital. As a matter of fact, ^{64}Ni is a very expensive target material. For this reason, an alternative ^{64}Cu production method was investigated at ENEA by using the quasi-monochromatic 14 MeV fusion neutron beam made available at the Frascati Neutron Generator (FNG) located at the ENEA – Frascati Research Center. In particular, two nuclear reactions were studied: ^{65}Cu(n,2n)^{64}Cu and ^{64}Zn(n,p)^{64}Cu. The radiochemical analysis of the activated samples was performed at the ENEA-NMLNWM laboratory located in ENEA-Casaccia Research Center. The activity measurements were carried out at the ENEA-INMRI, located in the ENEA-Casaccia Research Center, with high metrological level conditions and by assuring their traceability to the ^{64}Cu primary activity standard here developed and maintained. A prediction of the ^{64}Cu production by means of the high-brilliance 14 MeV neutron source named Sorgentina is also discussed.

Keywords: ^{64}Cu, fast neutrons, radiopharmaceuticals, direct activity measurements

1. Introduction

Copper-64 (^{64}Cu) features a half-life $T_{1/2}$ = 12.7004(20) hours and a maximum energy of the β^+ emission $E_{\beta+}$ = 653.1(2) keV, with emission probability $I_{\beta+}$ = 17.52(15)%. The maximum energy of the β^- emission is $E_{\beta-}$ = 579.4(7) keV, with $I_{\beta-}$ = 38.48(26)% [1]. ^{64}Cu is an emerging radionuclide in nuclear medicine. In fact, for its physical and chemical characteristics,^{64}Cu is suitable for both imaging and treatment, i.e. it is a so-called theranostic radionuclide [9]. Because of the increasing interest in Nuclear Medicine for this radionuclide, many researches around the world have been carried out by using different production methods, such as the ones performed by neutron accelerators [19,20], by biomedical cyclotrons [23], and fission nuclear reactors [28]. The production method based on biomedical cyclotrons increased in the last decades, because such small facilities are now available in many Nuclear Medicine Departments where the radionuclide is produced. The method used

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to produce Copper-64 is based on the ^{64}Ni(p,n)^{64}Cu reaction. As a matter of fact, ^{64}Cu production by ^{64}Ni is very expensive, because of the high cost of the enriched nickel target material (^{64}Ni abundance in the natural Ni is 0.92%). In Italy ^{64}Cu is presently produced by the A.C.O.M. Srl Italian Company [4] with which ENEA started in the 2018 a research activity devoted to the investigation of alternative routes for ^{64}Cu production, namely by means of 14 MeV fusion neutrons. In particular, the Frascati Neutron Generator (FNG) facility of 14 MeV neutrons is an accelerator-driven 14 MeV neutron source operating at the ENEA – Frascati Research Center. Two reactions were investigated: ^{65}Cu(n,2n)^{64}Cu and ^{64}Zn(n,p)^{64}Cu [5]. The ^{64}Cu produced by the two reactions was then chemically characterized at the Laboratory for Waste Management of Nuclear Material (NMLNWM) located in the ENEA-Casaccia Research Center and the activity measurements of the sample prepared were carried out, with high-metrological conditions, at the Laboratory of Radioactivity of the Italian National Institute of Ionizing Radiation Metrology (INMRI), located at the ENEA-Casaccia Research Center.

2. The 14 MeV neutron fusion reactions for ^{64}Cu production

The reactions involving 14 MeV fusion neutrons are mostly inelastic endoergic processes characterized by thresholds, typically lying in the MeV region. The cross sections for inelastic reaction ^{65}Cu(n,2n)^{64}Cu and ^{64}Zn(n,p)^{64}Cu are reported in Fig. 1.

The ^{65}Cu cross section features a threshold at about 10 MeV, then reaching an almost constant value of about 850 mb at 14 MeV. The ^{64}Zn cross section features a threshold at about 2 MeV and a maximum at about 11 MeV of 250 mb, decreasing to about 150 mb at 14 MeV.

2.1. The ENEA Frascati Neutron Generator (FNG)

The Frascati Neutron Generator (FNG) [2] is schematically represented in Fig. 2.

FNG is a compact accelerator-driven neutron source, relying on both Deuteron–Deuteron (D-D) and Deuteron–Tritium (D-T) fusion reactions, in turn producing almost monochromatic 2.5 MeV and 14 MeV neutrons with a maximum neutron emission rate of 10^9 s^{-1} and 10^{11} s^{-1}, respectively.

Deuterons are accelerated by means of an electrostatic accelerator up to 300 kV and 1 mA current is delivered onto a Titanium target, loaded with Deuterium or Tritium, where D-D or D-T fusion reactions (namely D + T → n + ^{4}He + 17.6 MeV and D + D → n + ^{3}He + 3.27 MeV) take place.

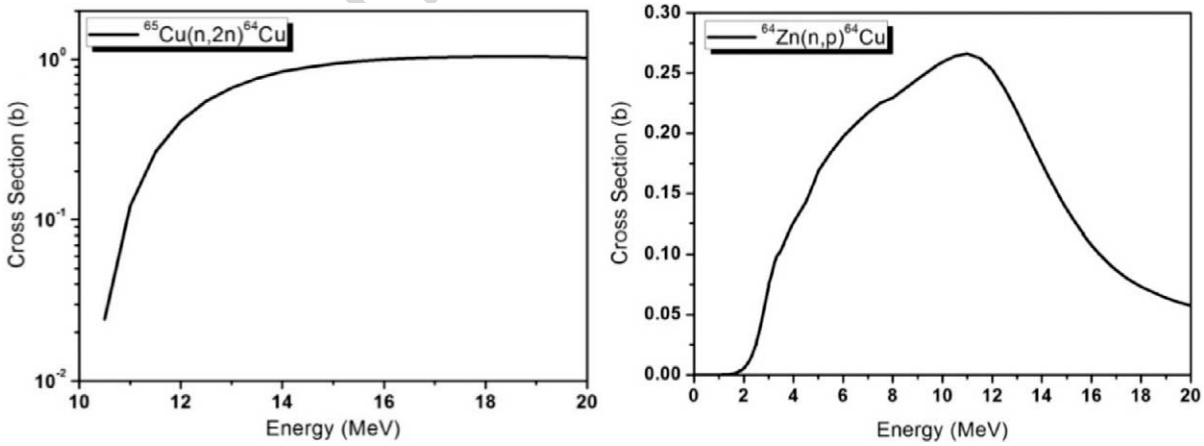

Fig. 1. (left) Inelastic (n,2n) cross section of ^{65}Cu in the region above the threshold reaction energy; (right) inelastic (n,p) cross section of ^{64}Zn in the region above the threshold reaction energy (data from Ref. [5]).

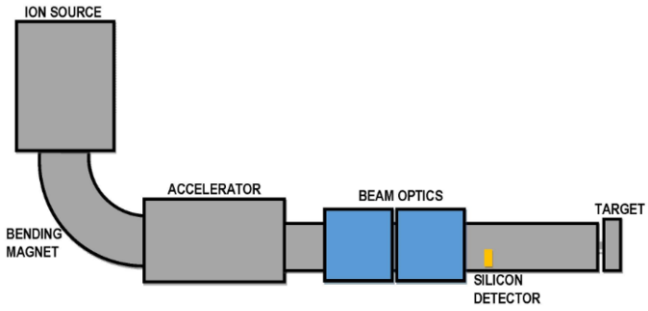

Fig. 2. Schematic layout of the Frascati Neutron Generator (FNG).

In D-T mode, the source neutron emission rate is determined by means of an absolute measurement, the so-called associated alpha particle technique. The D-T reaction produces in the final state a 14.1 MeV neutron and a 3.6 MeV alpha particle, respectively. A calibrated silicon detector, placed inside the drift vacuum tube at 2 m from the target and subtending a small and well-defined solid angle to the interaction point, provides the absolute number of the 14.1 MeV neutrons produced at the target by counting the alpha particles from the reaction. Simulations, benchmarked by means of experimental measurements, provide a neutron yield and flux determination within an uncertainty of 3%. In D-D mode, the 2.5 MeV neutron emission rate is determined using foil activation technique, where a ^{115}In solid target is irradiated and the gamma activation, following the ^{115}In(n,n')^{115m}In reaction channel, is measured by means of an absolutely energy calibrated High-Purity Germanium detector.

The D-D neutron emission is measured at ±7% uncertainty. The neutron source is simulated using the MCNP code [27], while the D-T reactions are simulated using a custom subroutine which reproduces the neutron emissivity of FNG source considering the angular and energy distribution. The subroutine has been tested in many benchmark experiments, reproducing very accurately experimental results [10].

2.2. *Cu production at ENEA-FNG*

^{64}Cu was produced at ENEA-FNG facility via the two reactions mentioned above, i.e. ^{65}Cu(n,2n)^{64}Cu and ^{64}Zn(n,p)^{64}Cu, using both natural and enriched target of copper and zinc, respectively. Before irradiating the metallic targets, simulations of the two reaction channels were performed by using MCNP Monte Carlo [27], with the JEFF3.1.1 libraries, and the FISPACT-II [6] code, with TENDL2015 nuclear data libraries, considering both the characteristics of the FNG neutron beam and the targets. These simulations allowed to forecast, for a defined neutron yield, target-beam geometry and irradiation time, the ^{64}Cu activity that could be produced at ENEA-FNG. At the same time, they allowed to understand better the two reaction mechanisms and the radionuclides different from ^{64}Cu, i.e. impurities, which are activated by the 14 MeV neutron beams in the two reactions considered. In particular, for the first reaction, ^{64}Cu was produced both using natural and enriched copper target. In the first case, a ^{64}Cu activity amount was obtained by irradiating a target of natural copper (69.15% ^{63}Cu, 30.85% ^{65}Cu), of mass $m = 1.8940 \pm 0.0001$ g, with a neutron yield of 3.8 10^{10} s^{-1}, for an irradiation time of 16 min and positioning the target at a distance of 2.8 cm from the neutron source. In the second case, a ^{64}Cu activity amount was produced by irradiating a target of enriched copper (99.7% ^{65}Cu, 0.3% ^{63}Cu), of mass $m = 0.09994 \pm 0.00001$ g, with a neutron yield of 4.3 10^{10} s^{-1}, for an irradiation time of 101 minutes and positioning the target at a distance of 1.5 cm from the neutron source. The second reaction was, differently, carried out by using both natural (49.17% ^{64}Zn) and enriched (99.40% ^{64}Zn) zinc targets, with mass of $m = 1.7661 \pm 0.0001$ g and $m = 0.918089 \pm 0.000001$ g, respectively. The two zinc targets were irradiated: the first with a neutron yield of 2.5 10^{11} s^{-1}, at a distance of 3 cm from the neutron source and for an irradiation time of 60 minutes; the second one with a neutron yield of 4.0 10^{11} s^{-1}, at a distance of 1 cm from the neutron source and for an irradiation time of 90 minutes.

3. Cu master solution preparation at ENEA-NMLNWM

^{64}Cu produced via 14 MeV neutron reaction on zinc target was also separated, after irradiation, at the ENEA-NMLNWM Laboratory. The procedure, hereafter described, allowed to produce a purified liquid solution of ^{64}Cu useful for direct activity measurements described in the next section. Radiochemical methods for ^{64}Cu depend on the target material and its purity. Amongst the ones that have been successfully applied, the Ion Exchange Chromatography method [21] is the most effective for ^{64}Cu separation, especially from zinc target material and other impurities, as well as in copper (enriched or natural) targets. In this last case the separation on activated ^{64}Cu is possible only from impurities but not from non-activated copper. In this work a strong anion exchanger absorbent resin was used (Dowex®-1 × 8 or equivalent) using gradient elution with 8M, 4M, 2M HCl and distilled water Cu separation from Zn and other impurities.

The target materials consist of Zn or Cu, both natural or enriched. For enriched targets, ^{64}Zn pellets of 99.40% enrichment and a purity of 99.95% and ^{65}Cu of 99.7% enrichment and a purity of 99.99% (Trace Sciences Internal Corporation [7], Ontario-Canada). The natural target materials are ^{nat}Zn 99.95% and ^{nat}Cu 99.99% in foils (Goodfellow [8], UK).

The pieces, after irradiation, underwent a complete dissolution treatment based on digestion with *Aqua Regia* (HNO_3/HCl 1:3 v/v) using 5 ml of acidic solution for each gram of target material. After complete digestion, the solution was heated until it reduces to dryness and then the residue dissolved in few ml of concentrated HCl (2–3 ml). Less concentrated acid can be also used (8M HCl), however the target weight has to be considered to avoid undesired volume increase and pH shift. The solution was loaded on a ∅0.7 × 10 cm Luer-Lock® ended disposable columns packed with 5 g of dry Dowex®-1 × 8 preconditioned with 8M HCl (approx. 20 ml). Using gradient elution with 8M, 4M, 2M of HCl and distilled water the ^{64}Cu was collected in the fraction at 2M of HCl. The zinc if recycling and reuse were needed, was collected in the last fraction with distilled water.

4. Cu activity measurements at ENEA-INMRI

The activity measurements of the irradiated material (when possible also in liquid solution obtained by proper radiochemical treatments as described in the previous section) were carried out at the Radioactivity Laboratory of the ENEA-INMRI. The measurements were performed in high-metrological conditions by assuring their traceability to the ^{64}Cu primary standard developed [16] and maintained [22] at ENEA-INMRI and transferred, by calibration procedures, to the end-users [15]. In particular, three absolute metrological techniques were applied to carry out these measurements. All the three techniques are based on the high-efficiency 4π counting methods: (1) the $4\pi\gamma$-integral counting technique, based on the well-type NaI(Tl) 5″ × 5″ detector [13,26]; (2) the γ-γ coincidence technique based on two NaI(Tl) 5″ × 5″ detectors [25] and (3) the Triple-to-Double-Coincidence-Ratio (TDCR) method [14] based on $4\pi\beta$-integral counting made by liquid scintillation detectors [11].

The first one was applied to the activity measurements of the ^{64}Cu produced via the (n,2n) reaction, because of the impossibility to radio-chemically separate the ^{64}Cu from of ^{65}Cu of the irradiated target. The second one was applied to the ^{64}Cu produced via the (n,p) reaction before the dissolution, taking into account the β^+ emission of ^{64}Cu. The last one was applied to the ^{64}Cu produced the (n,p) reaction after the dissolution, because of the possibility to separate by radiochemical techniques ^{64}Cu from^{64}Zn. Both reactions were studied using natural and enriched targets of copper and zinc provided by A.C.O.M. S.r.l. Company and bought from the GoodFellow. The impurities activated in the natural target materials were identified and quantified by the high-energy resolution ENEA-INMRI HPGe spectrometer, a robustly calibrated and routinely strictly quality-controlled detector.

In particular, the preliminary ^{64}Cu activity concentration (A_C) results and the corresponding uncertainties $u(A_C)$, obtained for the (n,2n) reaction in the conditions specified in Sect. 2.2, are reported, at the end-of-irradiation (EOI) time, for the two kind of targets (natural- and enriched-copper), in Table 1.

The measured activity concentration values for the reaction (n,2n), reported in Table 1, are in good agreement with the simulated ones. In fact, by generating 10^7 events, the value of 12.71 10^3 Bq g^{-1} for natural copper target

Table 1
Activity concentration of ^{64}Cu produced via (n,2n) reaction

Target	A_c (Bq g^{-1})	$u(A_c)$ (Bq g^{-1})
Natural copper	13.0910^3	0.2010^3
Enriched copper	1067.5310^3	40.3510^3

Table 2
Activity concentration of ^{64}Cu produced via (n,p) reaction

Target	A_c (Bq g^{-1})	$u(A_c)$ (Bq g^{-1})
Natural zinc	7.87 10^3	0.12 10^3
Enriched zinc	115.83 10^3	4.20 10^3

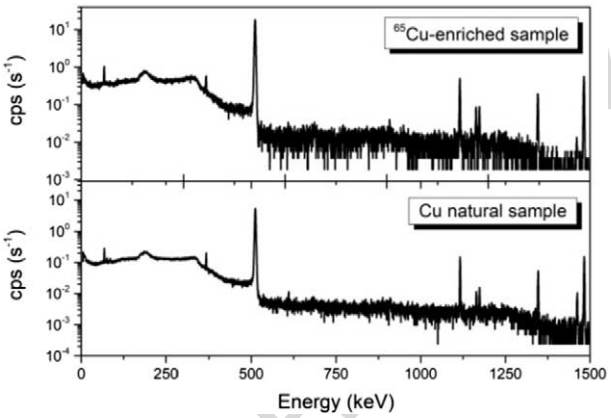

Fig. 3. (top panel) Spectrum of the enriched copper target after irradiation at FNG; (bottom panel) spectrum of the natural copper target after irradiation at FNG.

and the value of 1060.76 10^3 Bq g^{-1} for enriched copper target were obtained by the Monte Carlo codes used. The uncertainty on this data are mainly due to the statistical components.

Differently, the preliminary ^{64}Cu activity concentration results, obtained for the reaction (n,p) in the conditions specified in the Sect. 2.2, are reported, at the EOI, for the two kind of targets (natural- and enriched-zinc), in Table 2, together with the respective uncertainties $u(A_c)$.

Due to the very irregular shape of the zinc target used, the simulation for (n,p) reaction is still under study. A very preliminary result for the natural zinc target shows a deviation of $\Delta(\%) = (1 - A_{C\_sim}/A_{C\_exp}) = +7.62\%$ between the simulated activity concentration (A_{C_sim}) and the experimental one (A_{C_exp}) reported in Table 2. No simulated data are still available for enriched zinc target, but due to very good agreement between simulated and experimental results obtained in the previous (n,2n) reaction for enriched target, and taking into account the deviation $\Delta(\%)$ as above, one can infer that also for enriched target the deviation between simulated and experimental results will be no greater than +8%. In Figs 3 and 4 the typical spectra recorded by means of a calibrated HPGe spectrometer after irradiation of the samples are shown. A definitive and extensive definition of the results is already ongoing.

Thanks to the ENEA-INMRI high-energy resolution HPGe spectrometer, well calibrated by the high-level metrological standard available in the Radioactivity Laboratory of the Institute, a first experimental estimation of the impurities activated in the target was also performed. In particular, a preliminary impurities analysis was carried out on the (n,2n) reaction highlighting a presence, in the irradiated sample, of the radionuclide ^{65}Ni ($T_{1/2}$ = 2.5172(3) h [3]) with an activity concentration of about 1.3 10^{-2} times lower than the ^{64}Cu activity concentration reported in Table 1. For zinc targets the impurity data analysis is still in progress. All the data of activity presented in this paper are given in unit Bq.g^{-1} because this is, in fact, the unit used in the International System (SI) of Units,

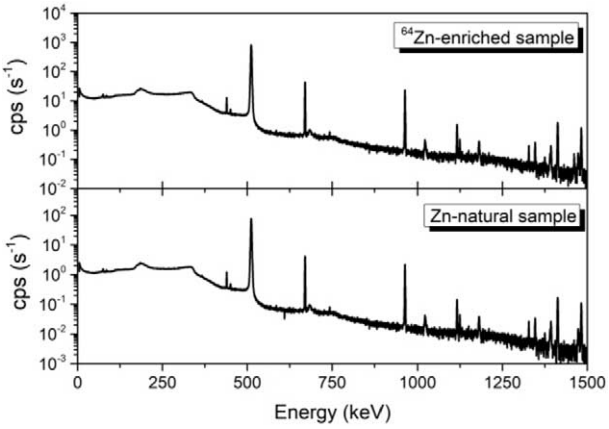

Fig. 4. (top panel) Spectrum of the enriched zinc target after irradiation at FNG; (bottom panel) spectrum of the natural zinc target after irradiation at FNG.

maintained at the Bureau International des Poids et Mesures (BIPM, Sèvres – France) for the activity as physical quantity. The activity values reported in the previous tables must be compared with the neutron yield used for irradiating the target during the experiments performed; it is not convenient for this kind of neutron facility, like FNG, to use other parameters (such as the deuteron current per hour) to give the results of activity because all the other parameters depend from different working conditions of the facility, such as its power, the amount of the ^{3}H loaded in the target, etc. and the only parameter which is significant in this case is the neutron yield actually used that must be measured with high precision as in these experiments.

5. Perspectives for higher brilliance 14 MeV neutron sources

While ENEA-FNG in its present configuration is well suited for testing physical processes related to radioisotopes production by means of inelastic neutron reactions involving 14 MeV fusion, a 14 MeV neutron source featuring a higher neutron emission rate (above 10^{14} s^{-1}) might be well suited for extensive production of these radioisotopes. Following the assessment discussed in [12,17] for ^{99}Mo/^{99m}Tc, it is possible to make a first rough estimate of the specific activity for ^{64}Cu that can be produced at a 10^{15} (taken as reference) 14 MeV neutron source, such as the New Sorgentina Fusion Source (NSFS), well described in [18,24]. This estimation can be made thanks to the results of the ^{64}Cu achieved in these experiments performed using the ENEA-FNG neutron beams and taking into account the simulation of the two reactions channels above benchmarked by experimental data measured and analysed at ENEA-INMRI. In fact, the activities reported in Tables 1 and 2 should be in the case of NSFS facility scaled for the ratio between the neutron yield of NSFS and the neutron yield of FNG. In particular, taking into account that the maximum ^{64}Cu quantity tolerable by the human body is 10 mg, it was possible to estimate, from the activity data measured by the reaction ^{65}Cu(n,2n)^{64}Cu reported in Table 1, the maximum activity of ^{64}Cu achievable with the NSFS Facility for this kind of reaction. In fact, for an irradiation time of 3 times the ^{64}Cu half-life the estimated ^{64}Cu activity was 1.02 10^{10}Bq roughly. Differently, for the other reaction channel using Zinc enriched target, the achievable ^{64}Cu activity was estimated to be 3.5 10^{10}Bq roughly for the same above irradiation time.

6. Conclusions

Thanks to the expertise available at ENEA in the FSN department, concerning both monochromatic FNG neutron source and radioactivity measurements facility at INMRI, the ^{64}Cu production via fast and quasi-monochromatic

neutrons, of 14.1 MeV energy about, produced at the ENEA-FNG facility was investigated for the first time at ENEA, exploiting two different reaction channels, namely ^{65}Cu(n,2n)^{64}Cu and ^{64}Zn(n,p)^{64}Cu by measuring, at ENEA-INMRI in high metrological level conditions, the activity of the ^{64}Cu produced including a first estimation of the impurities activated both in natural and enriched samples. So, this work may open new interesting perspectives for an alternative production route for this radionuclide, which is very interesting for application in nuclear medicine. It is clear that all the processes considered in this work are still under investigation and the situation will be clearer when the analysis data will be completed and new experiments will be performed.

Acknowledgements

The authors acknowledge Dr P. De Felice (from ENEA-INMRI) and Dr. M. Pillon (from ENEA-FNG) for useful discussions.

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