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Activity measurement of a ^{64}Cu sample activated by a 14 MeV neutron beam

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Summary. — A new activity measurement system based on two NaI(Tl) $5'' \times 5''$ cylindrical crystals, placed at 180°, was built at the ENEA Italian National Institute of Ionizing Radiation Metrology (INMRI) and characterized by Monte Carlo simulations in FLUKA code. It was used to determine the activity of a ^{64}Cu source, produced at ENEA's Frascati Neutron Generator (FNG) by irradiating an enriched ^{64}Zn sample with a 14 MeV neutron beam. Radiochemical treatments of the activated sample were performed at the ENEA Nuclear Material characterization Laboratory and Nuclear Waste Management (NMLNWM) to separate ^{64}Cu from the zinc target and other impurities. The radioactive solution resulting from the process was measured using the Triple-to-Double Coincidence Ratio (TDCR) technique. The comparison of ^{64}Cu activity in the solid sample and in the radioactive solution allowed to assess the separation efficiency. This study opened interesting perspectives in the production of short-lived radionuclides for nuclear medicine applications by means of 14 MeV neutron beams. ^{64}Cu is, in fact, an emerging theranostic radionuclide currently produced with cyclotrons, irradiating very expensive ^{64}Ni targets. From the achieved results, a prediction on the production of ^{64}Cu with a high-brilliance 14 MeV neutron source, under construction at ENEA, was made.

1. – Introduction

The development of personalised nuclear medicine relies on the availability of new radionuclides. In particular, radiometals are receiving increasing attention since they can be used to label relevant bio-molecules (such as peptides), finding applications in both therapeutic and diagnostic techniques. This feature opens the door to the theranostic approach, based on the use of a pair of radionuclides with very similar or identical

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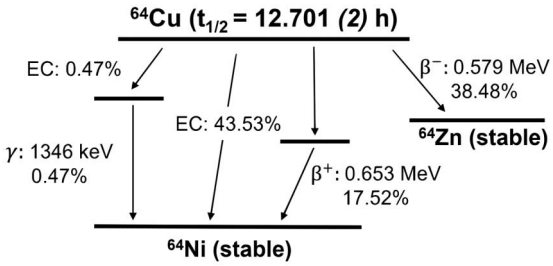

Fig. 1. – Decay scheme of ^{64}Cu.

chemical properties to label the same bio-molecule. One isotope (β^+- or γ-emitter) is used to visualize the distribution of the labelled compound and the acquired data allow to tune the dose deposition delivered by the second isotope (β^--, Auger- or α-emitter). Among radiometals, ^{64}Cu ($t_{1/2} = 12.701$ h) is of particular interest because of its unique decay scheme featuring three different decay routes, namely electron capture (EC), β^+ decay and β^- decay (see fig. 1). The low-energy positron emission makes ^{64}Cu suitable for Positron Emission Tomography (PET) technique, providing high-resolution images. The absence of an abundant γ-ray emission does not compromise the imaging process and reduces dosimetric or radiobiological concerns. On the other hand, the Auger electrons emitted as a result of EC decay and the β^- particles can be used for targeted radionuclide therapy [1]. These characteristics earned ^{64}Cu the name of theranostic agent [2]. Moreover, focusing on the β^+ route, ^{64}Cu can be used as a diagnostic partner for the therapeutic ^{67}Cu ($t_{1/2} = 61.83$ h), providing a very promising theranostic pair.

Due to the increasing interest for this radionuclide in nuclear medicine, several ^{64}Cu production methods have been proposed, such as the use of neutron generators [3, 4], fission nuclear reactors [5] and biomedical cyclotrons by irradiating both solid [6-8] and liquid [9] targets. The most widely used production method is based on the reaction ^{64}Ni(p,n)^{64}Cu which allows to obtain high production yields with low energy protons, easily reachable with small medical cyclotrons. Although these accelerators are available in many nuclear medicine departments, the availability of ^{64}Cu is still limited. The main reason is the low natural abundance of ^{64}Ni, which makes it extremely expensive.

For this purpose, ENEA has started in 2018 a research program focused on the production of ^{64}Cu by means of 14 MeV neutrons from nuclear fusion produced by the Frascati Neutron Generator (FNG) at the ENEA Frascati Research Center [10, 11].

The aim of the study presented in this paper is to precisely measure the activity of ^{64}Cu, produced through the ^{64}Zn(n,p)^{64}Cu reaction by irradiating a 99.40% enriched ^{64}Zn target. The activated ^{64}Cu was chemically treated at the Nuclear Material Waste Management Laboratory (NMLNWM) located at the ENEA-Casaccia Research Centre to separate copper from zinc and other impurities in the irradiated target, in turn preparing a liquid solution. The ^{64}Cu activity measurements were carried out under high-metrological conditions at the Laboratory of Radioactivity of the Italian National Institute of Ionizing Radiation Metrology (INMRI), situated in the ENEA-Casaccia Research Center, using different detectors. A new system, named 4π γ-γ detector and based on two large volume NaI(Tl) cylindrical crystals, was built in the framework of this study and characterized by means of Monte Carlo simulations using the FLUKA code [12]. This system was used to assess the activity of the irradiated sample a few

hours after the irradiation as well as of the liquid solution, obtained by means of the separation process. The latter was also measured with a HIDEX 300SL "Metro" version detector, based on the Triple-to-Double Coincidence Ratio (TDCR) technique. The comparison between the two ^{64}Cu activity measurements allowed to estimate the efficiency of the separation process. From the achieved results, a prediction on the production of ^{64}Cu with a high-brilliance 14 MeV neutron source, under construction at the ENEA Brasimone Research Center, was made.

2. – Experimental methodologies

2˙1. *^{64}Cu activity measurements at ENEA-INMRI.* – The activity measurements of ^{64}Cu were performed under high-metrological conditions at the ENEA-INMRI Laboratory, using two different detectors based on independent physical principles.

The first detector, built and characterized in the framework of this study, makes use of the 4π γ-γ coincidence technique. It is composed of two high-efficiency NaI(Tl) $5'' \times 5''$ cylindrical crystals, placed at 180° along the z-axis (fig. 2(a)). The upper detector is movable and allows to manually introduce the source; the lower one is fixed and is surrounded by a 5-mm-thick lead jacket for shielding purposes. Each crystal is coupled to a photomultiplier (BURLE $5''$ S83006E), connected to a high-voltage generator (SILENA HV7712) with a maximum voltage of 2 kV, and a preamplifier (ORTEC 113). The electronic signal outputs are recorded by a CAEN DT5724 digitizer. The energy calibration of the detector was performed using a multi-gamma source, while Monte Carlo simulations by means of the FLUKA code (fig. 2(b)) were developed to determine the counting efficiency in any experimental geometry. The calibration was tested with a ^{137}Cs point-like source certified by INMRI, whose activity was measured within a deviation of 0.06%. The FLUKA simulations allow to calculate the counting efficiency of the two crystals within an uncertainty of 0.2%.

The second detector, used to assess the ^{64}Cu activity after the separation process, was a HIDEX 300SL "Metro" version. It is based on the TDCR technique, a fundamental

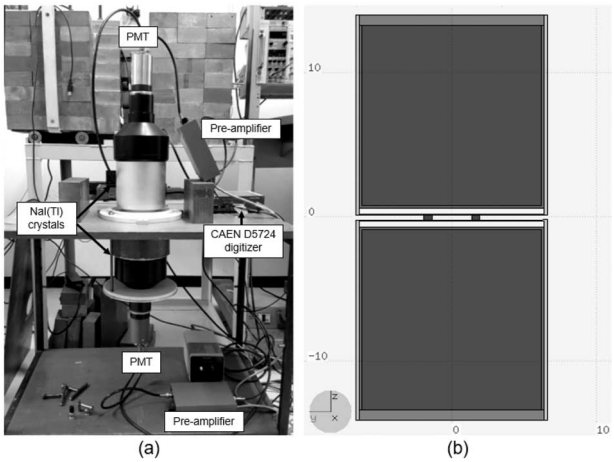

Fig. 2. – (a) Picture of the ENEA-INMRI 4π γ-γ detector. (b) Sketch of the geometric model implemented in the FLUKA simulations.

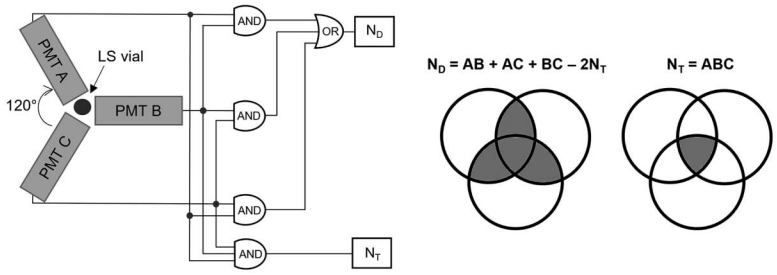

Fig. 3. – Schematic representation of the TDCR technique.

method in liquid scintillation counting developed to determine the activity of pure β- and pure EC-emitters in solution [13].

In liquid scintillation counting, the radioactive solution is mixed with a liquid scintillation cocktail, consisting mainly of scintillator molecules dissolved in organic solvent. When ionising radiation is emitted into the vial, a fraction of its energy is transferred to the fluorescent molecules, which emit light photons. If the efficiency of the system is known, the radiation activity can be determined by measuring the count rate [14].

The TDCR method requires the use of a three-photomultiplier (PMT) counter and a coincidence logic electronics which has five outputs, *i.e.*, three double coincidences, a triple coincidence and the logic sum of the double coincidences (fig. 3) [14].

The method is based on a physical and statistical model of the distribution of scintillation photons and their detection probability in the three-PMTs counter. Measuring the count rate in double (N_D) and triple (N_T) coincidence, the activity of the radioactive solution is given by

$$A = \frac{N_D}{\phi_D} = \frac{N_T}{\phi_T}, \tag{1}$$

where ϕ_D and ϕ_T are the counting efficiency in double and triple coincidence, respectively, which have to be assessed. For this purpose, it is possible to define the TDCR parameter as the ratio between the triple and double coincidence counting efficiency. The TDCR parameter is derived from counting statics laws and it is a non-linear equation. However, for a large number of disintegrations it tends to the ratio between the triple and double coincidence counting rate, which is experimentally known. Numerical calculation methods allow to derive the counting efficiency, which has one solution in the case of pure β-emitters and up to three solutions in the case of EC-emitters [14].

2˙2. *The Frascati Neutron Generator and enriched ^{64}Zn target irradiation.* – ^{64}Cu was produced via the ^{64}Zn(n,p)^{64}Cu reaction by irradiating a 99.40% enriched ^{64}Zn target with a 14 MeV neutron beam at the Frascati Neutron Generator (FNG) facility, in operation at the ENEA Frascati Research Centre. FNG, represented in fig. 4, is a compact accelerator-driven neutron source producing almost monochromatic 14 MeV neutrons with a nominal neutron emission rate of $10^{11}\,\mathrm{s}^{-1}$ [15]. A mixed beam of atomic and molecular deuterium ions is produced by the ion source and then analyzed by a 90° bending magnet. Only mono-atomic deuterium ions (D^+) are selected and accelerated by means of an electrostatic accelerating tube up to 300 kV. A D^+-current of 1 mA is

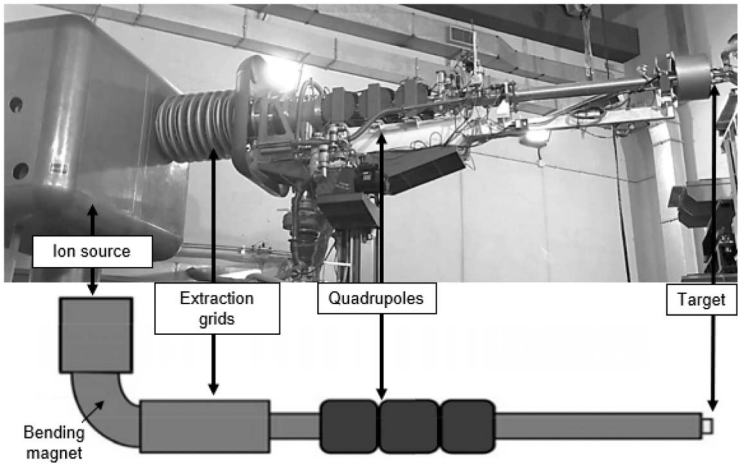

Fig. 4. – The Frascati Neutron Generator (FNG).

then delivered onto a titanium target, loaded with tritium, where the fusion reaction $D + T \rightarrow n + {}^4He + 17.6\,MeV$ takes place [16].

The neutron emission rate is determined by means of an absolute measurement, counting the produced α-particles with a small silicon surface barrier detector incorporated in the beam line. This technique allows to assess the neutron yield and flux with an uncertainty of 4% [17]. A Monte Carlo simulation in MCNP code [18] and a customised sub-routine were implemented to reproduce the neutron emission of FNG considering the angular and energy distribution. The subroutine has been tested in many benchmark experiments, reproducing experimental results very accurately [19].

The irradiated 99.40% enriched ^{64}Zn target was supplied by Trace Science International Corporation and its isotopic composition is reported in table I. The sample had an irregular shape (fig. 5(a)) and mass $m = (0.918089 \pm 0.000001)\,g$. It was placed at a distance from the FNG's neutron emitting source of about 1 cm (fig. 5(b)) and irradiated over 75 min with a neutron emission rate of $4 \times 10^{10}\,s^{-1}$. Monte Carlo simulations were performed using the MCNP and FISPACT-II [20] codes in order to calculate the neutron flux at the target position and to make quantitative predictions of the activity of ^{64}Cu and other impurities produced, for a defined target-beam geometry and irradiation conditions. The target was modelled as a parallelepiped with dimensions $10\,mm \times 3\,mm \times 3\,mm$ (fig. 5(c)). The calculations provided a ^{64}Cu activity concentration at the end of irradiation (EoI) of 118 kBq/g, with an uncertainty of about 8%. Besides ^{64}Cu, the only relevant radionuclides activated in the sample were ^{63}Zn and ^{60}Co, produced by the secondary reactions ^{64}Zn(n, 2n)^{63}Zn and ^{64}Zn(n, pα)^{60}Co, respectively. At EoI, a P-type HPGe detector in operation at the ENEA-FNG laboratory was used to verify the activation of the sample. The radionuclides of interest, together with their decay characteristics and the γ-lines used to identify them are reported in table II.

2˙3. *Radiochemical separation process at ENEA-NMLNWM.* – After the irradiation, the activated enriched ^{64}Zn sample underwent a complete dissolution treatment by means of aqua regia (HNO_3/HCl 1:3 v/v) using 5 mL of acid solution for each gram of target material. The solution was heated to dryness and then the metal residue was dissolved

TABLE I. – *Isotopic composition of the enriched ^{64}Zn target supplied by Trace Science International Corporation.*

Isotope	^{64}Zn	^{66}Zn	^{67}Zn	^{68}Zn	^{70}Zn
[%]	99.40	0.39	0.04	0.17	<0.01

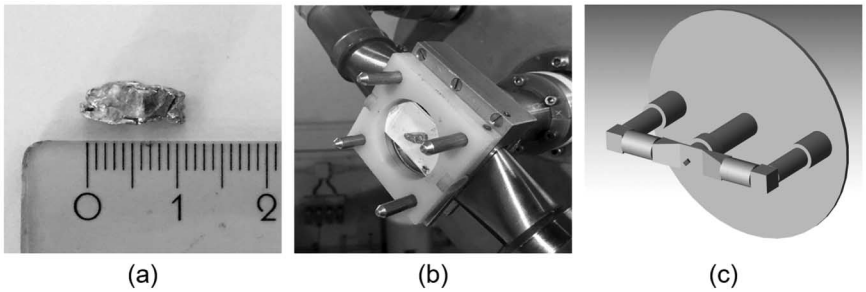

Fig. 5. – (a) Enriched ^{64}Zn sample; experimental (b) and simulated (c) setup of the enriched ^{64}Zn sample irradiation at the FNG facility.

TABLE II. – *Activated radionuclides and their decay properties used to identify them* [21].

Radionuclide	Half-life	Decay mode: [%]	E_γ [keV]	BR [%]
^{64}Cu	12.701 (2) h	EC, β^+: 61.5	1345.77 (6)	0.475 (11)
		β^-: 38.5	–	–
^{63}Zn	38.47 (5) min	EC, β^+: 100	669.62 (5)	8.2 (3)
^{60}Co	1925.28 (14) d	β^-: 100	1332.492 (4)	99.9826 (6)

in a few mL of 8 M HCl. The ^{64}Cu separation from the zinc target material and impurities was performed using a strong anion exchanger absorbent resin (Dowex®-1 × 8), preconditioned with 8 M HCl. Using gradient elution with 8 M, 4 M, 2 M and distilled water, the ^{64}Cu was collected in the 2 M fraction of HCl. Zinc was collected in the last fraction with distilled water and can be recovered to be reused. The solution containing ^{64}Cu was then reduced in volume and diluted with distilled water to lower its acidity level.

3. – Analysis and results

3˙1. *Pre-separation ^{64}Cu activity measurement*. – The preliminary measurement with the HPGe detector at the ENEA-FNG facility confirmed the presence of ^{64}Cu, ^{63}Zn and ^{60}Co in the sample (fig. 6). The first ^{64}Cu activity measurement was carried out at ENEA-INMRI about two hours after the EoI with the 4π γ-γ detector. The sample was placed between the two NaI(Tl) crystals and measured with both of them. The counting efficiency was assessed by means of the FLUKA simulation, considering an

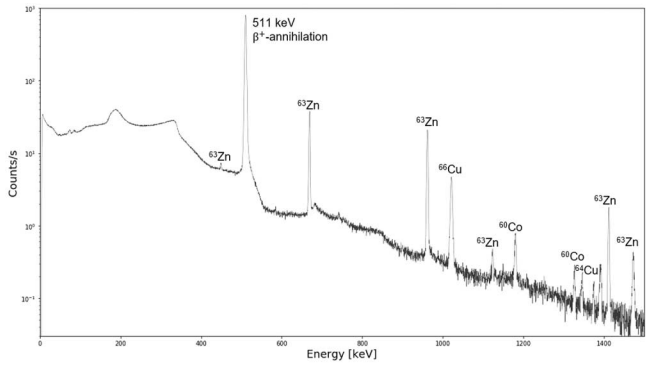

Fig. 6. – The activated enriched ^{64}Zn sample spectrum, recorded by the ENEA-FNG HPGe detector.

extended source of ^{64}Cu. Given the presence of the β^+-emitter ^{63}Zn, it was necessary to separate the two contributions to the 511 keV annihilation peak, by taking into account readings at different times exploiting the different half-life of the two radionuclides [22].

For a sample containing n radionuclides, the background corrected count rate, ρ, is a function of the time and is defined as

$$\rho(t) = \sum_{k=0}^{n-1} A_k \epsilon_k f_k e^{-\lambda_k t}, \tag{2}$$

where k is the index on the radionuclide, A_k is its EoI activity to be determined, λ_k is the decay constant, $f_k = \frac{1-e^{-\lambda_k \cdot RT}}{\lambda_k \cdot RT}$ is the correction factor for measurement duration and ϵ_k is the counting efficiency of the detector.

Considering n measurements starting at $t_0, t_1, \ldots, t_{n-1}$, it is possible to set up the following linear system:

$$\begin{pmatrix} \epsilon_0 f_0 e^{-\lambda_0 t_0} & \epsilon_1 f_1 e^{-\lambda_1 t_0} & .. & \epsilon_{n-1} f_{n-1} e^{-\lambda_{n-1} t_0} \\ \epsilon_0 f_0 e^{-\lambda_0 t_1} & \epsilon_1 f_1 e^{-\lambda_1 t_1} & .. & \epsilon_{n-1} f_{n-1} e^{-\lambda_{n-1} t_1} \\ .. & .. & .. & .. \\ \epsilon_0 f_0 e^{-\lambda_0 t_{n-1}} & \epsilon_1 f_1 e^{-\lambda_1 t_{n-1}} & .. & \epsilon_{n-1} f_{n-1} e^{-\lambda_{n-1} t_{n-1}} \end{pmatrix} \begin{pmatrix} A_0 \\ A_1 \\ .. \\ A_{n-1} \end{pmatrix} = \begin{pmatrix} \rho(t_0) \\ \rho(t_1) \\ .. \\ \rho(t_{n-1}) \end{pmatrix}, \tag{3}$$

whose solution gives the $\{A_k\}$ vector of radionuclide activity values.

Such solutions are only acceptable if the equations (eq. (2)) forming the system are linearly independent, *i.e.*, if the minimum time interval between the different measurements

$$\Delta\tau = \min\{|t_0 - t_1|, |t_0 - t_2|, \ldots, |t_0 - t_{n-1}|, \ldots, |t_1 - t_2|, \ldots, |t_1 - t_{n-1}|, \ldots\} \tag{4}$$

is sufficiently large.

If the source containing n radionuclides is measured l times, it is possible to set up $N = \frac{l!}{(l-n)!n!}$ linear systems.

The obtained N values of A_k, correlated with the different parameters $\Delta\tau$, are distributed as a Gaussian function (fig. 7). The activity of the k-th radionuclide is then

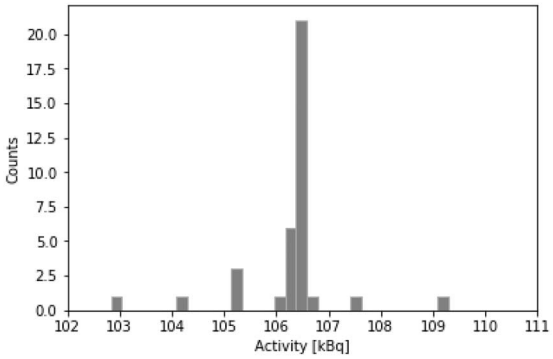

Fig. 7. – Distribution of the ^{64}Cu activity values.

given by the mean value of the distribution and the uncertainty is the standard deviation of the mean value. Considering the sample mass of (0.918089 ± 0.000001) g, the activity concentration of ^{64}Cu was found to be (116.00 ± 0.04) kBq/g, in agreement with the simulated one within a deviation lower than 2%.

3˙2. *Post-separation ^{64}Cu activity measurement.* – The procedure used for dissolving the zinc sample and separating ^{64}Cu from the target material and other impurities is described in sect. **2˙3**. It provided a ^{64}Cu radioactive solution with mass $m = (7.733028 \pm 0.000001)$ g, from which a point-like source to be measured with the 4π γ-γ detector was prepared depositing a few drops on a plastic disc (fig. 8(a)) and letting them dry. In the same way, a liquid source was prepared for the measurement with the HIDEX detector by depositing a few drops of solution into a 20 ml glass vial containing the Ultima Gold$^{\text{TM}}$ scintillation cocktail (fig. 8(b)).

The deposited masses were assessed with an analytical balance (METTLER TOLEDO XP56) and found to be $m = (0.148928 \pm 0.000001)$ g and $m = (0.493835 \pm 0.000001)$ g for the point-like and the liquid source, respectively.

The point-like source was placed between the two NaI(Tl) detectors and repeatedly measured with both of them. The counting efficiency was assessed with the FLUKA simulation considering a point-like source of ^{64}Cu, since in this case it was the only radionuclide present in the sample. Its activity concentration was then derived from the annihilation peak and measured to be (13.3 ± 0.2) kBq/g.

Fig. 8. – Preparation of the point-like (a) and liquid (b) source.

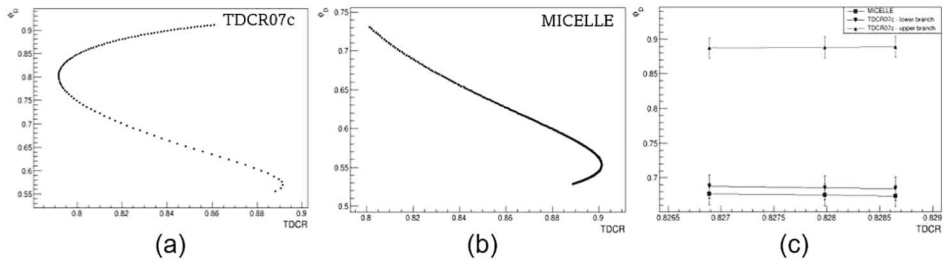

Fig. 9. – The counting efficiency in double coincidence (ϕ_D) as a function of the TDCR parameter, calculated with the TDCR07c (a) and the MICELLE (b) code; (c) comparison of the obtained counting efficiencies.

The liquid source was repeatedly measured with the HIDEX detector. Two codes were used to calculate the counting efficiency, namely an INMRI implementation of the TDCR07c code [23] and the MICELLE code [24]. The former uses experimental data, characteristic of the detector used, to solve the TDCR equation numerically; the latter implements a Monte Carlo simulation to calculate the efficiency as a function of the TDCR parameter. The counting efficiencies in double coincidence (ϕ_D) obtained with the two codes are shown in fig. 9(a) and in fig. 9(b). Since ^{64}Cu is an EC-emitter, the efficiency as a function of the TDCR parameter is characterised by two clearly separated branches. Comparing the efficiency values obtained with the two codes, it was observed that the MICELLE code provides results compatible within the uncertainty with those obtained from the lower branch of the TDCR07c implementation (fig. 9(c)). The latter were used for the calculation of the ^{64}Cu activity concentration, which resulted in (13.3 ± 0.6) kBq/g. The ^{64}Cu activities measured with the two detectors are in agreement within a deviation lower than 0.05%. The comparison between the measured activities of the solid sample and of the radioactive solution made it possible to estimate the efficiency of the separation process, which was found to be $\epsilon = (96 \pm 2)\%$.

4. – Conclusion and outlooks

The experiment described in this paper allowed to assess the ^{64}Cu activity produced by irradiating a solid 99.40% enriched ^{64}Zn sample with a 14 MeV neutron beam at the ENEA-FNG facility. Activity measurements of ^{64}Cu before and after radiochemical separation were carried out under high-metrological conditions at the ENEA-INMRI laboratory, using detectors based on different physical principles. The measurement of the metallic sample provided a ^{64}Cu EoI activity of (116.00±0.04) kBq/g, which is in good agreement with the value expected from the simulations in FISPACT-II (118 kBq/g). The radioactive solution provided by the radiochemical treatments was measured with both the TDCR and the 4π γ-γ method and was found to be (13.3 ± 0.3) kBq/g. The comparison of these two values allowed to estimate the efficiency of the separation process, resulting in (96 ± 2)%.

The precise results achieved in the activity measurements allowed to benchmark the Monte Carlo simulations implemented for the experiment and open interesting perspectives in the production of radionuclides for medical applications, such as ^{64}Cu, by means of a high-brillance 14 MeV neutron source. In fact, knowing that in nuclear medicine activities in the order of magnitude of GBqs are required, FNG turned out not to be

suitable for routine production. For this reason, a new facility characterized by a neutron emission rate above $10^{14}\,\mathrm{s}^{-1}$ is under design at ENEA's Brasimone Research Center. The new 14 MeV neutron generator should allow to achieve ^{64}Cu activity concentration 10^3–10^4 times higher than that produced in the present experiment.

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