



## Standardization, decay data measurements and evaluation of $^{64}\text{Cu}$

M.-M. Bé<sup>a,\*</sup>, P. Cassette<sup>a</sup>, M.C. Lépy<sup>a</sup>, M.-N. Amiot<sup>a</sup>, K. Kossert<sup>b</sup>, O.J. Nähle<sup>b</sup>, O. Ott<sup>b</sup>, C. Wanke<sup>b</sup>, P. Dryak<sup>c</sup>, G. Ratel<sup>d</sup>, M. Sahagia<sup>e</sup>, A. Luca<sup>e</sup>, A. Antohe<sup>e</sup>, L. Johansson<sup>f</sup>, J. Keightley<sup>f</sup>, A. Pearce<sup>f</sup>

<sup>a</sup> CEA-LIST/LNE-LNHB, CE Saclay, 91191 Gif-sur-Yvette, France

<sup>b</sup> PTB, Bundesallee 100, 38116 Braunschweig, Germany

<sup>c</sup> CMI, Radiova 1, CZ-102 00, Prague 10, Czech Republic

<sup>d</sup> BIPM, Pavillon de Breteuil, 12 bis Grande Rue, 92312 Sèvres Cedex, France

<sup>e</sup> IFIN-HH, Magurele, Ilfov county, RO-077125, Bucharest, Romania

<sup>f</sup> NPL, Hampton Road, Teddington, TW11 0LW, UK

### ARTICLE INFO

Available online 28 February 2012

#### Keywords:

$^{64}\text{Cu}$  standardization

$^{64}\text{Cu}$  decay scheme

Half-life

$\gamma$ -Ray intensities

x-Ray intensities

### ABSTRACT

The purposes of this study were to create national activity standards of  $^{64}\text{Cu}$ , to make possible the definition of an international key comparison reference value and to determine the decay data in order to improve the decay scheme. Four laboratories measured the activity of a  $^{64}\text{Cu}$  solution; these results were compared through the International Reference System. Moreover, the laboratories carried out new measurements of the photon emission intensities and of the half-life. A new decay scheme was derived from these new values and the previously published ones.

© 2012 Elsevier Ltd. All rights reserved.

### 1. Introduction

$^{64}\text{Cu}$  ( $T_{1/2}=12.7$  h) is a radionuclide decaying by  $\beta^+$  emission or electron-capture transition to  $^{64}\text{Ni}$ , and by  $\beta^-$  emission to  $^{64}\text{Zn}$ . This nuclide is commonly used as dosimeter to determine the neutron flux in a nuclear reactor, moreover it is to be used as a radiopharmaceutical for PET imaging.

However, before this study, there was no international traceability established for this radionuclide through key comparisons. Thus, EURAMET project 1085 was proposed with the following aims:

- to create national activity standards of  $^{64}\text{Cu}$  which can be disseminated to practitioners in the medical field through secondary standards;
- to establish an international metrological infrastructure bringing significant input to the key comparison data base (KCDB) at the *Bureau International des Poids et Mesures* (BIPM);
- to determine nuclear decay data such as the  $\beta^+/\beta^-$  branching ratio, absolute emission intensities of x-rays, annihilation and gamma photons, and the half-life with high accuracy;
- to evaluate and publish an updated decay scheme, based on the results coming from this project and the previously published results as well.

Participants in this EURAMET project were as follows: LNE-LNHB (coordinator, France), PTB (Germany), CMI (Czech Republic), NPL (United Kingdom) and IFIN-HH (Romania). The first part of the exercise was devoted to activity measurements and the first four participants sent their results to the BIPM for participation in the International Reference System (SIR) (Ratel, 2007). In parallel, the participants measured the half-life and the photon emission intensities; these results and those previously published were exploited to establish a new decay scheme. The new data set was then used by two participants to analyze their activity measurement results. The influence of the new data is not negligible for most of the methods utilized for activity determination. The updated decay scheme parameters are described in the second part of this study.

### 2. Decay scheme

$^{64}\text{Cu}$  decays by  $\beta^-$  emission to the ground state of  $^{64}\text{Zn}$  and by  $\beta^+$  emission in competition with electron capture (EC) to  $^{64}\text{Ni}$  (Fig. 1). Several determinations of the  $\beta^-$  transition probability were reported ranging from 38.06(3)% (Wermann et al. 2002) to 39.04(33)% (Christmas et al. 1983), this 1% difference in absolute value being carried forward to the ( $\beta^+$ +EC) branch.

### 3. Activity and decay scheme data measurements

In this exercise, each participating laboratory obtained its own  $^{64}\text{Cu}$  radioactive solution and sent its result of activity measurement, as well as an ampoule of the same solution, to the BIPM for

\* Corresponding author.

E-mail address: [mmbe@cea.fr](mailto:mmbe@cea.fr) (M.-M. Bé).

participation in the SIR. Then, the comparison of the results has been realized through the “equivalent activity” given by the SIR (Ratel, 2007). All results obtained in this exercise and reported below, are given with combined standard uncertainties.

### 3.1. PTB experimental results

The PTB carried out activity measurements by means of a  $4\pi\beta\text{-}\gamma$  coincidence counting equipment with a proportional counter in the  $\beta$  channel and two NaI(Tl) detectors in the  $\gamma$  channel. In addition, PTB made use of a Tri-Carb 2800 TR and a Wallac 1414 liquid scintillation (LS) counters; the efficiencies were computed according to the CIEMAT/NIST (C/N) method. This part was already described in Wanke et al. (2010), hence only the results are listed here. The activity values of the solution were 2699(14) and 2721(22) kBq/g for LS counting and  $4\pi\beta\text{-}\gamma$  coincidence counting respectively (Fig. 2); with a final result sent to the SIR of 2705(12) kBq/g (Michotte et al., 2009).

A determination of the half-life (12.704(5) h), of the 511 keV photon emission intensity (35.12(22)%) and, of the 1346 keV  $\gamma$ -ray intensity (0.474(5)%) were also carried out at PTB (Wanke et al., 2010).

### 3.2. NPL experimental results

The NPL employed two activity standardization techniques, namely  $4\pi(\text{LS})\text{-}\gamma$  Digital Coincidence Counting (Keightley and Watt, 2002) utilizing an NPL built LS counter (with two photomultiplier tubes operating in coincidence for background reduction purposes) along with a NaI(Tl) detector, as well as the C/N

efficiency tracing technique (using both Tri-Carb 2700 and Beckman 6500 LS counters).

For the  $4\pi(\text{LS})\text{-}\gamma$  Digital Coincidence Counting (DCC), several difficulties arose when the 511 keV annihilation photon peak was employed as a gamma gate, due to positron annihilation-in-flight effects. The efficiency extrapolation was thus developed from the use of the 1345 keV gamma ray (with an associated increase on the data collection intervals due to the low photon emission intensity for the associated gamma transition).

Three different LS cocktails were employed for the C/N measurements (UltimaGold AB, ReadySafe and EcoScint A) over five standardization runs, with the preferred results (finally submitted to the SIR) based upon the use of 10 ml of ECOSCINT A, with 0.1 ml of carrier/active solution and 0.2 ml of tritiated/distilled water.

Finally, two results of the activity were obtained and sent to the SIR: 71800(510) and 72150(460) kBq/g for the LS(C/N) counting and  $4\pi(\text{LS})\text{-}\gamma$  coincidence counting respectively.

A series of accurately weighed aliquots of the standardized solution were dispensed to six 10 R Schott vials and measured in the master NPL Secondary Standard Ionization Chamber. This yielded a calibration factor of 1.955(18) pA/MBq, which is directly transferable to users of the commercially available systems ISOCAL IV and FIDELIS.

Furthermore, the decay of  $^{64}\text{Cu}$  was tracked over a period of 110 h in a system comprising two identical Centronic IG-25 ionization chambers (with the second chamber operated at reverse voltage bias for background reduction purposes). The rate of voltage-change across a standard feedback capacitor (300.63 pF) employed in a current integrating electrometer was monitored for over 2200 cycles of up to 180 s. The half-life was estimated as 12.702(8) h at  $k=1$ .

### 3.3. CMI experimental results

The CMI made use of the  $4\pi\beta\text{-}\gamma$  coincidence technique with a proportional counter in the  $\beta$  channel, two NaI(Tl) detectors in the  $\gamma$  channel and applied the efficiency extrapolation method. Several  $\gamma$ -ray energy window settings were tested. The optimal energy window and the linear extrapolation range were derived using software procedures (the detailed description of the method will be published elsewhere).

Measurements of photon emission intensities were also carried out, using a calibrated HPGe (p-type) detector with a crystal diameter of 63 mm, a crystal thickness of 59 mm and, an Al

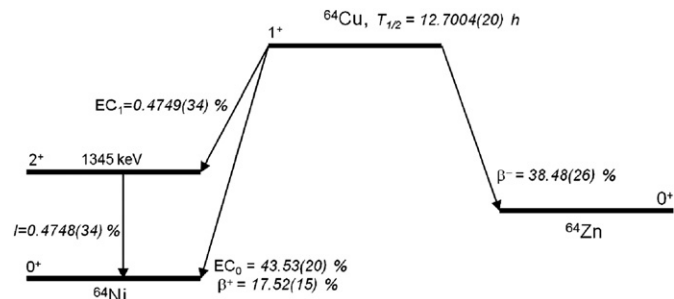


Fig. 1.  $^{64}\text{Cu}$  decay scheme. The values in italic were determined in this exercise.

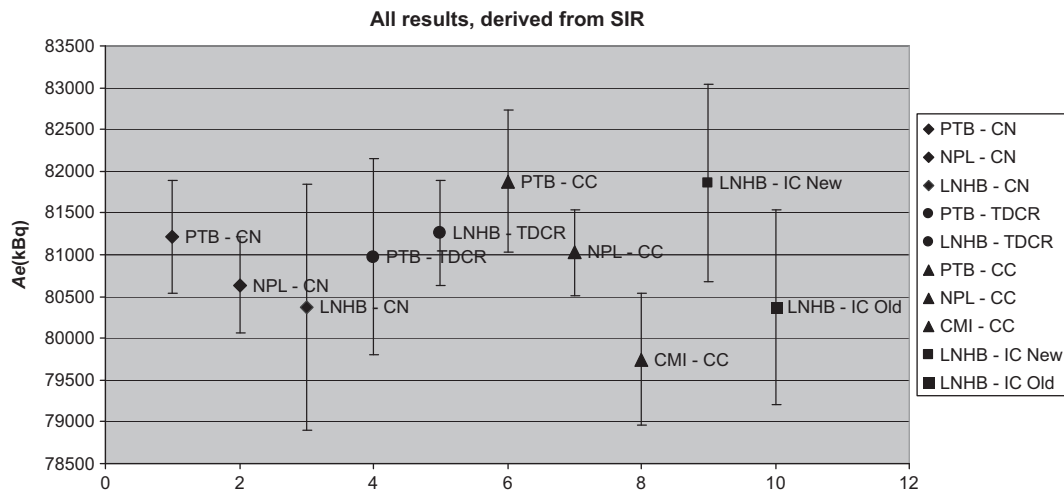


Fig. 2. Results of activity obtained by various methods.

window of 1.5 mm. The measurements were performed with a source–detector distance of 25 cm. The detector efficiency curve was established using a simulation code (MCNP) validated by experimental measurements of standard sources. Coincidence summing effects were calculated and found negligible. Weighed drops of a  $\text{CuCl}_2$  solution in HCl were deposited onto thin polyester foil.

For the determination of the 511 keV photon intensity, the sources were covered by different types of absorbers: plexiglas, aluminum, copper and tin disks, in order to assess the correction due to in-flight annihilation. The estimated values for this effect, which have been used, are 1.01%, 1.05%, 1.10%, and 1.14% for each of the absorbers respectively. Consequently such corrections were applied to the four measurement results and the mean of the four results adopted as a final value. The 511 keV peak areas were determined from the calculated continuous spectra as 4% of the total spectrum areas.

Finally, the 511 keV photon intensity was found to be 34.99(38)% and the 1346 keV  $\gamma$ -ray intensity being 0.476(6)%, where the uncertainties are given as combined standard uncertainties including the components on the activity result and all correction factors.

### 3.4. LNHB experimental results

The LNHB made use of the LS counting technique with a locally developed variant of the CIEMAT/NIST method and the Triple to Double Coincidence Ratio (TDCR) method to measure the activity of a  $\text{CuCl}_2$  solution in HCl as a solvent. The TDCR method was applied by considering the weighted sum of calculated triple- and double- coincidence detection efficiencies for each branch:  $\beta^+$ ,  $\beta^-$  and electron capture. As the relationship between the TDCR and the detection efficiency is not a bijective function (one TDCR value can correspond to three values of detection efficiency), the identification of the relevant location on the “efficiency vs. TDCR” curve was obtained making use of quenched sources and assuming that the effect of quenching is a decrease of the detection efficiency. Detection efficiency was then calculated taking into account only the results of unquenched sources. However, the TDCR method is very sensitive to the ratio of the electron capture probability over the sum of the beta plus and minus probabilities:  $EC/(\beta^- + \beta^+)$  and it appeared that the determination of the detection efficiency was possible only with the ratio value derived from the new decay scheme as described in Section 5. The results of activity measurements were as follows: 808(12) for the variant of C/N method and 817(6) kBq/g for TDCR method using the new decay scheme data. This latter value was adopted for participation in the SIR.

In parallel, four  $^{64}\text{Cu}$  BIPM glass ampoules were measured using a Vinten type 671 ionization chamber (IC), filled with nitrogen at a pressure of about  $10^6$  Pa and with an effective volume of 10.5 L. The activity was determined using the PENELOPE simulation code (Salvat et al., 2008) using the method described in de Vismes et al. (2003) and then, without requiring a standardization value. However, the IC calibration is quite dependent on the decay scheme data, and therefore, the results were analyzed using both decay schemes. An activity value of 808(8) kBq/g was derived from the previous decay scheme and 823(8) kBq/g from the new one. Both results differ by about 1.8%, however they are in agreement to each other and with the other results.

The measurements of photon emission intensities were performed using two n-type high-purity germanium (HPGe) detectors, equipped with conventional electronics, based on  $100\text{ cm}^3$  germanium crystal with  $500\text{ }\mu\text{m}$  Be window. These spectrometers were calibrated using standard point sources, the source-to-detector distance being 10 cm.

The 1346 keV  $\gamma$ -ray intensity was found to be 0.472(12)% and, the 511 keV photon emission 35.1(3)%. For this latter event, the measurement was performed using an aluminum container to force the annihilation of the positrons close to the source deposit, and a corrective factor for in-flight annihilation was applied (Lépy et al., 2010).

LNHB measured also the two major x-ray emission intensities at 7.47 keV ( $K\alpha$ ) and 8.30 keV ( $K\beta$ ). The spectra recorded were obtained in the calibration geometrical conditions, thus the photons intensities were derived using the efficiency curve, without geometrical and coincidence corrective factors. The peak areas were determined using the COLEGRAM software (Ruellan et al., 1996), taking into account the natural Lorentzian width of x-rays. The results are 14.41(15)% and 2.01(3)% for  $K\alpha$  and  $K\beta$  respectively.

All these findings will be described in a forthcoming publication.

### 3.5. IFIN-HH experimental results

In this context, the IFIN-HH carried out an activity measurement but could not send an ampoule for participation in the SIR. The measurements were performed, using the  $4\pi\beta$ - $\gamma$  coincidence technique with a proportional counter in the  $\beta$  channel, a NaI(Tl) detector in the  $\gamma$  channel, in two conditions. First, in the  $\beta$  channel, the radiations due to  $\beta^{+/-}$  decay and electron capture were measured and, the 511 keV and 1346 keV rays in the gamma channel. Then, only the radiations due to  $\beta^{+/-}$  decay in the  $\beta$  channel and the 511 keV photons in the  $\gamma$  channel were measured. The mean of the two values was adopted as the final result. Details are presented in Sahagia et al. (2012).

In addition, IFIN-HH followed the  $^{64}\text{Cu}$  decay with an ionization chamber and determined a half-life value of 12.696(12) h with the original vial, prior to its opening, to obtain significant ionization current values.

The photon emission intensity measurements were performed with a calibrated gamma-ray spectrometer consisting of an HPGe (p-type) detector (29% relative efficiency), associated electronics and software. Several corrections were applied, including the annihilation-in-flight process (Lépy et al., 2010). The photon emission intensities for the 1346 keV and 511 keV  $\gamma$ -rays were found to be 0.481(17)% and 35.3(12)% respectively (Luca et al., 2012).

## 4. Comparison of activity measurement results

The participating laboratories carried out their measurements and, at the same time, some of them sent an ampoule filled with an aliquot of the same radioactive solution to the BIPM, where they were compared with matched standard sources of  $^{226}\text{Ra}$  using a pressurized ionization chamber filled with 2 MPa nitrogen (Ratel, 2007). An equivalent activity,  $A_e$ , was determined for each submitted sample, making possible the comparison of radioactive solutions with different levels of activity and at any moment.

The results of activity measurements were kindly provided by the BIPM for this EURAMET 1085 exercise (Table 1).

The reduced  $\chi^2/(n-1)$  of this set of five data is equal to 1; then it is accepted as consistent. The arithmetic mean is 80815 kBq with a standard deviation of 296 and the weighted mean is 81018 kBq with an internal uncertainty of 238 kBq and an external uncertainty of 242 kBq. As the internal and the external uncertainty show quite similar values the uncertainty evaluation made by the laboratories can be considered as realistic; consequently, the weighted mean value of the comparison is an excellent estimator of the equivalent activity value.

In addition to the values submitted to the SIR, PTB and LNHB employed other methods to determine the activity; all results

**Table 1**  
Results of SIR activity measurements.

Laboratory-method	Reference date	Equivalent activity $A_e/(kBq)$	Combined standard uncertainty
PTB-4 $\pi$ (LS) $\beta$ C/N+	13/02/2009	81 396	385
4 $\pi\beta$ -NaI(Tl) $\gamma$ CC			
NPL-4 $\pi$ (LS) $\beta$ - $\gamma$ (DCC)	17/11/2009	81 029	514
NPL-4 $\pi$ (LS) $\beta$ C/N		80 635	573
CMI-4 $\pi$ ( $\beta, e_A$ )-NaI(Tl) $\gamma$ CC	16/09/2009	79 748	788
LNHB-4 $\pi$ (LS) $\beta$ TDCR	18/03/2010	81 265	630

(derived from the SIR equivalent activity) are graphically shown in Fig. 2.

In 2011, PTB re-analyzed its results and derived an activity value by means of the TDCR method. This method failed when using evaluated data as published in 2004 (Bé et al., 2004). For some LS samples the experimental TDCR value was lower than computed ratios of the triple and double efficiencies. The situation changed when using the decay data as presented in this paper. When applying the TDCR method with updated data, PTB obtained an activity of  $A=2691(35)$  kBq/g, which is consistent with the other PTB results, as well as with the overall data set. This PTB study then confirmed the observation previously pointed out by LNHB.

To conclude, the activity measurements carried out by various methods led to results in good agreement, in spite of the fact that some techniques seemed, such as the TDCR or the coincidence counting methods, were difficult to use.

The LS techniques (C/N and TDCR) depend on the branching ratios, in particular on the ratio  $EC/(\beta^- + \beta^+)$ , any change of its value will affect similarly as the activity value.

When determining the activity value, the LNHB made use of the LS counting with the TDCR method. The old decay data did not allow calculating ratios of the triple and double counting efficiencies compatible with the experimental TDCR value for an unquenched source. Only when using the new decay data as presented in this work this was possible. Later, these findings were confirmed at PTB.

This point underlines the importance of having accurate nuclear decay data, as even the so-called primary measurement methods can be very dependent on the radionuclide decay scheme.

## 5. $^{64}\text{Cu}$ decay scheme data

Several procedures can be followed to determine the decay scheme of  $^{64}\text{Cu}$ .

Combining the new decay data obtained within the scope of this exercise with those previously published in literature, a new decay scheme was derived, for details see Bé et al. (2011).

In this evaluation, we tried to introduce results coming from methods other than ionizing radiation measurements, in order to minimize the inherent correlation in the data.

### 5.1. $\beta^-$ and $\beta^+$ transitions

The probabilities of the  $\beta^-$ ,  $\beta^+$ , and EC branches were determined by a series of separate, but partially correlated, measurements by Christmas et al. (1983), Kawada (1986), and Qaim et al. (2007). Thus, Christmas et al. (1983) included, in their analysis, a least-squares fit to the various measured quantities and ratios of quantities, in order to account for covariances.

Another kind of investigation made by mass spectrometry measurements of the number of atoms of  $^{64}\text{Ni}$  and  $^{64}\text{Zn}$  produced in the decay of a  $^{64}\text{Cu}$  sample (Wermann et al., 2002) led to the determination of the  $P_{\beta^-}$  branching ratio.

#### • $\beta^-$ transition

The published measured probabilities of the  $\beta^-$  transition are listed in Table 2. The weighted mean of the set of four values has been adopted in this evaluation, i.e.  $P_{\beta^-} = 38.48(26)\%$ ; from which a matching ( $\beta^+ + \text{EC}$ ) branch of 61.52(26)% was derived.

#### • $\beta^+$ transition

Two methods are possible to derive the  $P_{\beta^+}$  probability value:

- From published measured probabilities of the  $\beta^+$  transition (Table 3).
- From theoretical calculations.

Using the LOGFT program, the ratio  $P_{\text{ECO}}/P_{\beta^+}$  is 2.485(25), from the  $P_{\beta^+} + P_{\text{ECO}} = 61.05(26)\%$  above, the  $P_{\beta^+}$  value is derived being 17.52(15)%.

This latter value has been obtained by an independent method and it is less correlated than the results of direct measurements. Moreover, it can be noted that the weighted mean of the last four values, listed in the Table 3, 17.59(9) is very close to 17.52(15). Thus, the value  $P_{\beta^+} = 17.52(15)\%$ , computed as previously indicated, has been adopted in this evaluation.

**Table 2**  
Measured probabilities of the  $\beta^-$  transition.

References	$P_{\beta^-}$ (%)	Comments
Christmas et al. (1983)	39.04(33)	4 $\pi\beta$ (LS)- $\gamma$ coincidence counting
Kawada (1986)	38.34(56)	4 $\pi\beta$ (PC)- $\gamma$ coincidence counting
Wermann et al. (2002)	38.06(30)	Mass spectrometry
Qaim et al. (2007)	38.4(12)	2 $\pi\beta$ (PC)- $\gamma$ anticoincidence counting
<b>Weighted mean</b>	<b>38.48(26)</b>	<b>Adopted</b>

**Table 3**  
Measured probabilities of the  $\beta^+$  transition.

References	$P_{\beta^+}$ (%)	Comments
Christmas et al. (1983)	17.86(14)	Ge(Li) spectrometry
Kawada (1986)	17.93(20)	HPGe $\gamma$ spectrometry
Qaim et al. (2007)	17.8(4)	$\gamma$ - $\gamma$ coincidence counting
Wanke et al. (2010)	17.56(11)	HPGe $\gamma$ spectrometry
Euromet 1085—CMI	17.69(19)	HPGe $\gamma$ spectrometry
Euromet 1085—LNHB	17.55(15)	HPGe $\gamma$ spectrometry
Euromet 1085—IFIN-HH	17.65(60)	HPGe $\gamma$ spectrometry
Weighted mean	17.68(6)	WM of the seven values
Weighted mean	17.58(8)	WM of the last four values

**Table 4**  
Measured values of the 1345 keV  $\gamma$ -ray intensity.

Reference	$I_{\gamma 1345}$ (%)	Comments
Christmas et al. (1983)	0.471(11)	
Kawada (1986)	0.487(20)	
Qaim et al. (2007)	0.54(3)	outlier
Wanke et al. (2010)	0.474(5)	
Euromet 1085—CMI	0.476(6)	Section 3.3
Euromet 1085—LNHB	0.472(12)	Section 3.4
Euromet 1085—IFIN-HH	0.481(17)	Section 3.5
<b>Weighted mean</b>	<b>0.4748(34)</b>	<b>Adopted</b>

**Table 5**

Experimental results of the total electron capture transition probability.

References	Total $P_{EC}$ (%)	Comments
Christmas et al. (1983)	43.10(46)	$4\pi\beta(\text{LS})-\gamma$ coincidence counting
Kawada (1986)	43.73(52)	$4\pi\beta(\text{PC})-\gamma$ coincidence counting+HPGe $\gamma$ spectrometry+ $4\pi\beta-\gamma$ coincidence counting
Qaim et al. (2007)	43.8(14)	Si(Li) x-ray spectrometry

**Table 6**

Half-life measured values.

Reference	$T_{1/2}$ (h)	Comments
Heinrich and Philippin (1968)	12.701(11)	
Merritt and Taylor (1972)	12.701(7)	
Dema and Harbottle (1973)	12.699(2)	Mean of 22 values
Ryves and Zieba (1974)	12.704(6)	
Rutledge et al. (1980, 1982)	12.701(3)	
Abzouzi et al. (1989)	12.700(3)	Original uncertainty $\times 3$
Wanke et al. (2010)	12.704(5)	
Euromet 1085—IFIN-HH	12.696(12)	Section 3.5 and (Luca et al., 2012)
Euromet 1085—NPL	12.702(8)	Section 3.2
<b>Adopted</b>	<b>12.7004(20)</b>	

**Table 7**

Decay scheme data comparison.

Reference	$P_{\beta^+}$ (%)	$P_{EC0}$ (%)	$P_{\beta^-}$ (%)	$EC/(\beta^- + \beta^+)$
1-Helmer (Bé et al., 2004)	17.86(14)	42.6(5)	39.0(3)	0.758(10)
2-This work	17.52(15)	43.53(20)	38.48(28)	0.786(10)
$\Delta_{\text{Rel}} [(1-2)/1] \times 100$	-1.9	+2.1	+1.3	+3.6

### 5.2. 1345 keV $\gamma$ -ray emission intensity and transition probability

A 1345 keV  $\gamma$ -ray intensity ( $I_{\gamma 1345}$ ) of 0.4748(34)% was deduced from the measured values as given in Table 4.

For this transition of E2 multipolarity, the internal-conversion coefficients ( $\alpha$ ) were interpolated from the tables of Band and Trzhaskovskaya (2002) using the computer code BrLcc (Kibédi et al., 2008) with the so called “frozen orbital” approximation, so as:  $\alpha_{\text{TOTAL}} = 0.394(6) 10^{-4}$ .

From  $\alpha_{\text{TOTAL}}$  and  $I_{\gamma 1345} = 0.4748(34)\%$ , the 1345 keV gamma transition probability is  $P_{\gamma 1345} = 0.4749(34)\%$ .

### 5.3. Electron capture transitions

The electron capture probability  $P_{EC1}$  to the first excited level in  $^{64}\text{Ni}$  was deduced being:  $P_{EC1} = P_{\gamma 1345} = 0.4749(34)\%$ . From:  $P_{\beta^+} + P_{EC0} = 61.05(26)\%$  and  $P_{\beta^+} = 17.52(15)\%$ , then  $P_{EC0} = 43.53(20)\%$ .

The sum  $P_{EC} = P_{EC0} + P_{EC1} = 44.00(20)\%$  can be compared with the three experimental results obtained for the total electron capture probability  $P_{EC}$  in Table 5.

### 5.4. Half-life

Since the 1930s, the half-life of  $^{64}\text{Cu}$  has been extensively studied and about 23 values have been published. In this study, only the most accurate ones, with uncertainties less than 0.012 h, have been accepted for statistical processing. In addition, three laboratories determined the half-life during the present exercise. The values used in this evaluation are listed in Table 6. The adopted half-life is 12.7004(20) h. More detailed comments are

placed on the Decay Data Evaluation Project website (<http://www.nucleide.org/NucData.htm>).

## 6. $^{64}\text{Cu}$ adopted decay scheme

The final decay scheme as established following this EURAMET 1085 exercise, is shown in Fig. 1. It differs noticeably from the previous evaluation of Helmer (Bé et al., 2004). The main characteristics are compared in Table 7. The electron capture probability to the ground state differs by 1%, in absolute value and the ratio  $EC/(\beta^- + \beta^+)$  by about 3%.

## 7. Conclusions

In the activity measurement part, four laboratories submitted results to the SIR for entries into the Key Comparison Database, from which a Key Comparison Reference Value will be established, allowing the international traceability of future  $^{64}\text{Cu}$  activity measurements and the creation of national secondary standards.

Moreover, the participating laboratories seized the opportunity to measure some of the decay data. These latter values and other published results were used to establish a new decay scheme. From the activity measurements it was noteworthy that the TDCR method can be applied only with the new decay scheme data. The results obtained with this technique are in good agreement with those obtained with the coincidence counting technique, this point supporting the new decay scheme data.

## References

- Abzouzi, A., Antony, M.S., Ndocko Ndongue, V.B., 1989. J. Radioanal. Nucl. Chem. 135, 455.
- Band, I.M., Trzhaskovskaya, M.B., 2002. Dirac-Fock internal conversion coefficients. At. Data Nucl. Data Tables 88, 1.
- Bé, M.-M., Chisté, V., Dulieu, C., Mougeot, X., C., Browne, E., Chechev, V., Kuzmenko, N., Kondev, F., Nichols, A., Luca, A., Galan, M., Arinc, A., Huang, X., 2011.  $^{64}\text{Cu}$  in table of radionuclides, monographie BIPM-5, ISBN 13 978 92 822 2234 8 (set) and ISBN 13 978 92 822 2235 5 (CD). CEA/LNE-LNHB, 91191 Gif-sur-Yvette, France and BIPM, Pavillon de Breteuil, 92312 Sèvres, France.
- Bé, M.-M., Chisté, V., Dulieu, C., Browne, E., Chechev, V., Kuzmenko, N., Helmer, R., Nichols, A., Schönfeld, E., Dersch, R., 2004.  $^{64}\text{Cu}$  in table of radionuclides, monographie BIPM-5, ISBN 92-822-2207-7 (set) and ISBN 92-822-2205-5 (CD). CEA/BNM-LNHB, 91191 Gif-sur-Yvette, France and BIPM, Pavillon de Breteuil, 92310 Sèvres, France.
- Christmas, P., Judge, S.M., Ryves, T.B., Smith, D., Winkler, G., 1983. Nucl. Instrum. Methods 215, 397.
- Dema, I., Harbottle, G., 1973. A 'best value' for the half-life of  $^{64}\text{Cu}$ . Radiochem. Radioanal. Lett. 15, 261.
- de Vismes, A., Amiot, M.-N., 2003. Towards absolute activity measurements by ionisation chambers using the PENELOPE Monte-Carlo code. Appl. Radiat. Isot. 59 (4), 267–272.
- Heinrich, F., Philippin, G., 1968. Helv. Phys. Acta 41, 431.
- Kawada, Y., 1986. Int. J. Appl. Radiat. Isot. 37, 7.
- Keightley, J.D., Watt, G.C., 2002. Digital Coincidence Counting (DCC) and its use in the corrections for out-of-channel gamma events in  $4\pi\beta-\gamma$  coincidence counting. Appl. Radiat. Isot. 56, 205–210.
- Kibédi, T., Burrows, T.W., Trzhaskovskaya, M.B., Davidson, P.M., Nestor Jr., C.W., 2008. Evaluation of theoretical conversion coefficients using BrLcc. Nucl. Instrum. Methods Phys. Res. A589, 202.
- Lépy, M.-C., Cassette, P., Ferreux, L., 2010. Measurement of beta-plus emitters by gamma-ray spectrometry. Appl. Radiat. Isot. 68, 1423–1427.
- Luca, A., Sahagia, M., Antohe, A., . Measurements of  $^{64}\text{Cu}$  and  $^{68}\text{Ga}$  half-lives and gamma-ray emission intensities. Appl. Radiat. Isot., in this issue.

- Merritt, J.S., Taylor, J.G.V., 1972. Report AECL-4257, 25.
- Michotte, C., Courte, S., Ratel, G., Kossert, K., Nähle, O., 2009. BIPM comparison BIPM.RI(II)-K1.Cu-<sup>64</sup> of the activity measurements of the radionuclide <sup>64</sup>Cu. Metrologia 46 (Technical Supplement), S06010.
- Qaim, S.M., Bisinger, T., Hilgers, K., Nayak, D., Coenen, H.H., 2007. Positron emission intensities in the decay of <sup>64</sup>Cu, <sup>76</sup>Br and <sup>124</sup>I. Radiochem. Radioanal. Lett. 95, 67.
- Ratel, G., 2007. The Système International de Référence and its application in key comparisons. Metrologia 44 (4), S7–S16.
- Ruellan, H., Lépy, M.-C., Etchevery, M., Plagnard, J., Morel, J., 1996. A new spectra processing code applied to the analysis of <sup>235</sup>U and <sup>238</sup>U in the 60–200 keV energy range. Nucl. Instrum. Methods Phys. Res. A 369, 651–656.
- Ryves, T.B., Zieba, K.J., 1974. J. Phys. (London) A7, 2318.
- Rutledge, A.R., Smith, L.V., Merritt, J.S., 1980. Report AECL-6692.
- Rutledge, A.R., Smith, L.V., Merritt, J.S., 1982. Report NBS-SP-626, p. 5.
- Sahagia, M., Luca, A., Antohe, A., Ivan, C., . Standardization of <sup>64</sup>Cu and <sup>68</sup>Ga by the 4πPC-γ coincidence method and calibration of the ionization chamber. Appl. Radiat. Isot. in this issue.
- Salvat, F., Fernandez-Varea, J.M., Sempau, J., 2008. PENELOPE-2008. A code system for Monte Carlo simulation of electron and photon transport. In: Workshop Proceedings, Barcelona, Spain.
- Wanke, C., Kossert, K., Nähle, O.J., Ott, O., 2010. Activity standardisation and decay data of <sup>64</sup>Cu. Appl. Radiat. Isot. 68 (7–8), 1297–1302.
- Wermann, G., Alber, D., Pritzkow, W., Riebe, G., Vogl, J., Görner, W., 2002. Appl. Radiat. Isot. 56 (1–2), 145.