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Positron emission intensities in the decay of ^{64}Cu, ^{76}Br and ^{124}I

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Summary. The relatively long-lived positron emitters ^{64}Cu ($t_{1/2} = 12.7$ h), ^{76}Br ($t_{1/2} = 16.2$ h) and ^{124}I ($t_{1/2} = 4.18$ d) are finding increasing applications in positron emission tomography (PET). For precise determination of their positron emission intensities, each radionuclide was prepared *via* a charged particle induced reaction in a "no-carrier-added" form and with high radionuclidic purity. It was then subjected to γ-ray and X-ray spectroscopy as well as to anticoincidence beta and $\gamma\gamma$-coincidence counting. The positron emission intensities measured were: ^{64}Cu (17.8 ± 0.4)%, ^{76}Br (58.2 ± 1.9)% and ^{124}I (22.0 ± 0.5)%. The intensity of the weak 1346 keV γ-ray emitted in the decay of ^{64}Cu was determined as (0.54 ± 0.03)%. Some implications of the precisely determined nuclear data are discussed.

1. Introduction

In Positron Emission Tomography (PET) the so called "organic" positron emitters ^{11}C ($t_{1/2} = 20.3$ min), ^{13}N ($t_{1/2} = 10.0$ min), ^{15}O ($t_{1/2} = 2.0$ min) and ^{18}F ($t_{1/2} = 110$ min) are commonly used. They are all relatively short-lived and are almost pure positron emitters. On the other hand, for studying slow biochemical processes, interest in positron emitters of somewhat longer half-lives is growing. Some of the important radionuclides in the latter category are ^{64}Cu ($t_{1/2} = 12.7$ h), ^{73}Se ($t_{1/2} = 7.1$ h), ^{76}Br ($t_{1/2} = 16.2$ h), ^{86}Y ($t_{1/2} = 14.7$ h), ^{124}I ($t_{1/2} = 4.18$ d) *etc.* (for more discussion cf. [1]).

A drawback of many of the non-conventional relatively long-lived positron emitters is that the positron emission intensity is often rather low and not exactly known. Furthermore, the decay of those radionuclides is generally associated with γ-rays. Both those properties lead to some uncertainty in the quantitation of tomographic scans. For the more promising radionuclides it is therefore imperative to determine some of the relevant decay data with higher accuracy.

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The decay data of most of the non-conventional positron emitters were determined in the context of nuclear structure studies; the recent related work deals more with evaluations of older data (cf. [2] and references cited therein to original works) rather than with new measurements. Since many of the radioactive samples used in older measurements were prepared without radiochemical separations, they were radionuclidically not pure. It appeared therefore important to prepare samples of very high purity and perform some new measurements on a few of the important radionuclides. Three radionuclides, namely ^{64}Cu, ^{76}Br and ^{124}I, were investigated in this study. They have been attracting considerable attention in recent years for use in PET studies.

The radionuclide ^{64}Cu decays by three modes, *viz.* β^- emission, electron capture (EC) and β^+ emission. According to the latest evaluation [2] the positron emission intensity corresponds to 17.4%. The experimentally reported I_{β^+} values, however, range between 17.4 and 19.0%. With the EC a very weak γ-transition of energy 1346 keV has also been reported. Its intensity could range between 0.47 and 0.60%. The present work is devoted to an experimental determination of the contributions of the three decay processes mentioned above as well as to a precise measurement of the intensity of the weak γ-ray at 1346 keV.

The radionuclides ^{76}Br and ^{124}I have been fairly well investigated [2]. The evaluated β^+ emission intensity in the decay of ^{76}Br is given as 56%. However, the radioactive sample used contained a mixture of ^{76}Br and ^{77}Br, demanding a significant correction while analysing the X-ray decay curves. Since in the present work ^{76}Br of high radionuclidic purity could be produced, it was thought worthwhile to determine the β^+ emission intensity again. In the case of ^{124}I the reported I_{β^+} values range between 22.0 and 26.0% and the evaluated value is 22.8%. In a recent work [3], based on γ-ray spectroscopic studies we showed that the β^+ emission intensity in the case of ^{124}I is 22.0%. The intensity was now remeasured using $\gamma\gamma$-coincidence and beta counting.

2. Experimental

The decay parameters were determined by using samples of very high radionuclidic purity. The counting methods included γ-ray and X-ray spectroscopy, $\gamma\gamma$-coincidence counting and anticoincidence β proportional counting. The salient features of the techniques used are described.

2.1 Preparation of radioactive sources

2.1.1 Preparation of ^{64}Cu samples

The production of this radionuclide was done *via* two nuclear processes, namely ^{66}Zn$(d, \alpha)^{64}$Cu and ^{nat}Zn$(d, x)^{64}$Cu. In the first case highly enriched ^{66}Zn (99.0%) and in the second ^{nat}Zn was electroplated on a Ni-backing. Irradiations were done at the compact cyclotron CV 28 of the Forschungszentrum Jülich with 14 MeV deuterons for 2 h at a beam current of about 2 μA.

The separation of radiocopper from the irradiated target was carried out using liquid-liquid extraction [4]. For this purpose the irradiated zinc was removed from the Ni-backing with a few drops of conc. HCl which was then evaporated to dryness. The residue was taken up in 1 mL of 1 M HCl and again evaporated to dryness. It was then dissolved in 10 mL of 1 M HCl. The radiocopper (partly contaminated with radiogallium) was extracted by shaking the mixture three times with 10 mL of a 0.01% solution of dithizone in CCl_4. The combined organic fractions were washed with 10 mL of 0.1 M HCl to remove traces of zinc. Radiocopper was back-extracted into the aqueous phase by treating the organic phase twice with 10 mL of 7.2 M HCl to which a few drops of 30% H_2O_2 were added. The organic phase was discarded. The aqueous phase was treated with 5 mL of CCl_4 and three times with 5 mL of di-isopropyl ether (saturated with 7.2 M HCl) to remove the traces of radiogallium. The remaining aqueous solution was reduced to about 1 mL by evaporation and finally adsorbed on a filter paper which then served as the radiocopper sample.

The radiochemical separation procedure developed was very effective. The separation yield was only 50–60% but for the present measurements the yield was not critical; important was the radionuclidic purity. While using the ^{66}Zn$(d, \alpha)^{64}$Cu reaction the separated product was pure ^{64}Cu, with absolutely no radionuclidic impurity. In spectroscopic studies only samples prepared *via* this method were used. The nuclear process ^{nat}Zn$(d, x)^{64}$Cu led to ^{64}Cu with some contamination from ^{61}Cu ($t_{1/2} = 3.4$ h, $E_\gamma = 283$ keV; $I_\gamma = 12.5\%$), formed *via* the ^{64}Zn$(d, \alpha n)^{61}$Cu reaction. The mixture of ^{61}Cu and ^{64}Cu was, however, deliberately produced, and used to determine the efficiency of the X-ray detector (see below).

2.1.2 Preparation of ^{76}Br samples

The production of this radionuclide was done *via* the ^{76}Se$(p, n)^{76}$Br reaction. About 3 mg of enriched ^{76}Se (99.8%) was placed between two aluminium foils and irradiated with low-energy protons at the compact cyclotron CV 28 for 45 min at a beam current of 100 nA. Since the radiobromine was not chemically separated, the projectile energy incident on the ^{76}Se target had to be adjusted to 7.3 MeV using some Al or Cu absorbers. At this energy the ^{76}Se$(p, pn)^{75}$Se reaction was not possible [5]. On the other hand, some small contribution from ^{77}Br ($t_{1/2} = 57.04$ h), possibly formed *via* the ^{76}Se$(p, \gamma)^{77}$Br reaction on the ^{76}Se target as well as *via* the ^{77}Se$(p, n)^{77}$Br reaction on the small amount of ^{77}Se (0.05%) present in the enriched ^{76}Se sample, could not be avoided. However, since that radionuclide is not a β^+ emitter, it did not interfere in the positron related studies on ^{76}Br. Only in X-ray spectroscopy a very small correction ($< 0.2\%$, see Sect. 3.3) had to be considered. After the irradiation, ^{76}Se was dissolved gently in a few drops of conc. HNO_3 and the solution was diluted. About 300 μL of that solution was then spread on an Al foil and evaporated. The sample had a thickness of < 1 mg ^{76}Se/cm^2. For X-ray spectroscopy this sample was used as such; for other measurements it was covered with a thin Al foil.

2.1.3 Preparation of ^{124}I samples

For production of this radionuclide the ^{124}Te$(p, n)^{124}$I process was used. A 99.8% enriched $^{124}TeO_2$ target on a Pt-backing was irradiated at the compact cyclotron CV 28 with 14 MeV protons for 3 h at a beam current of about 10 μA. The irradiated material was allowed to "cool" for 60 h (to let ^{123}I decay out), and then the radioiodine was separated *via* the standard dry distillation process [3]. A part of the radioactivity collected in 0.02 M NaOH solution was adsorbed on a filter paper which then served as the ^{124}I source. This was a very pure sample; the only radionuclidic impurity found was ^{125}I at a level of $< 0.1\%$.

2.2 Measurement of radioactivity

Four methods were utilized to determine the radioactivity of each radioactive sample. The details are given below.

2.2.1 Beta counting

For this purpose a very stable gas flow proportional counter (Berthold, Multi-Logger LB 5310) in low-level anticoincidence mode was used; the background was about 2 counts/min. Each sample was counted in a fixed 2π geometry for a long enough time to accumulate at least 20 000 counts (statistical uncertainty $< 1\%$). Measurements were repeated over several half-lives and then a decay curve analysis was performed. As the samples were thin and the β^+ (or β^-) end-point energies were not very low, the self-absorption correction was negligible ($< 1\%$). The counting efficiency of the detector was determined using a set of beta standards, like ^{14}C, ^{147}Pm, ^{99}Tc, ^{90}Sr, ^{36}Cl, ^{210}Bi, *etc.* (supplied by NEN, USA). For beta end-point energies > 0.8 MeV, a constant absolute counting efficiency of $(40 \pm 2)\%$ was achieved.

2.2.2 $\gamma\gamma$-coincidence counting

For an accurate determination of the radioactivity of a positron emitter a $\gamma\gamma$-coincidence counting system is very suitable. We made use of such a system available in the Abteilung Nuklearchemie of the University of Cologne. It consists of two 7.5 cm × 12.5 cm NaI(Tl) detectors, each shielded by lead, and placed in the basement of the building to decrease the cosmic background. Each detector is equipped with its own power supply and the necessary electronics. The energy region of interest can be selected *via* a single channel discriminator. The count rate could be measured singly in each detector or in the coincidence mode using a coincidence circuit with a time resolution of 1 μs. The efficiency of coincidence counting was determined for

various distances between the detectors using a ^{22}Na standard, equidistant from the two detectors. For a distance of 20 cm, the counting efficiency was about 7%. An important requirement in these measurements is the complete annihilation of the positron prior to detection of the annihilation quanta. Each sample was therefore placed in a 4 mm thick Cu-capsule and the ^{22}Na standard was also placed in the same geometry. Each sample was counted repeatedly over several half-lives and a decay curve analysis was performed to check the radionuclidic purity of the sample.

2.2.3 Gamma ray spectroscopy

For γ-ray spectroscopy several HPGe detectors (EG & G ORTEC) of various volumes were used. Each detector was coupled to a 4 k or 16 k multichannel analyser and a PC. The distance between the sample and the detector was at least 10 cm and the dead time was kept below 5%. This reduced the real coincidence and pile-up losses to $< 1\%$. The absolute efficiency of each detector was determined using γ-ray standards supplied by the PTB Braunschweig or Amersham International.

In the present work, special attention was directed to the determination of radioactivity *via* the annihilation peak. In this regard, two points were important:

(a) use of a thick absorber between the sample and the detector for complete annihilation of the positron. This was particularly important in the case of ^{76}Br which has a high positron end-point energy of about 4.0 MeV. Similar to $\gamma\gamma$-coincidence measurements, each sample was placed in a 4 mm thick Cu-capsule, and the standards were also placed in the same geometry.
(b) careful analysis of the area under the 511 keV annihilation peak which is known to be much broader than a normal photopeak. The region of interest (ROI) in the spectrum was properly chosen and the peak area integration performed accordingly.

The length of each measurement was chosen such that good counting statistics were obtained for each photopeak of interest. As in beta and $\gamma\gamma$-coincidence counting, the γ-ray spectroscopic measurements were repeated several times to check whether the photopeak areas decreased according to the half-life of the corresponding radionuclide. The analysis of the γ-ray peaks in the spectrum (other than the annihilation peak) was done using the software Gamma Vision (EG & G ORTEC).

2.2.4 X-ray spectroscopy

In order to determine the intensity of electron capture (EC) decay, the radionuclidically pure samples were subjected to X-ray spectroscopy. For this purpose a Si(Li) detector, especially suitable for counting X-rays of energies between 3 and 30 keV was used. It was coupled to the usual electronics, a 4 k multichannel analyser and a PC. The detector resolution (FWHM) at 14.4 keV γ-ray of ^{57}Co was 45 eV. In general k_α and k_β lines of each radionuclide were investigated. The spectrum analysis was done again using the software Gamma Vision (EG & G ORTEC) and the decay of both X-ray peaks was followed.

The decay of ^{64}Cu is accompanied by the emission of the K_α and K_β lines of the product nickel having energies of 7.47 and 8.26 keV, respectively. Due to the relatively low energy of the radiation involved, the self-absorption of the radiation in the sample and the efficiency of the detector needed to be carefully considered. The calibration of the detector was done using a mixed ^{61,64}Cu source produced *via* the $^{\text{nat}}$Zn$(d, x)^{61,64}$Cu processes (see above). At first the absolute activity of ^{61}Cu was determined *via* γ-ray spectrometry using the characteristic γ-ray of energy 283 keV ($I_\gamma = 12.5\%$). Thereafter, the same sample was counted on the X-ray detector at periodic intervals for about 30 h and, from the decay curve analysis of the K_α and K_β lines of the daughter element nickel, the count rates of both ^{61}Cu and ^{64}Cu were obtained. A comparison of the count rates of ^{61}Cu obtained using the two X-rays with the absolute activity of ^{61}Cu, determined *via* the γ-ray spectrometry, gave the efficiency of the detector (including self-absorption) for the 7.47 and 8.26 keV X-rays, respectively. Those efficiency values were then used for determining the radioactivity of ^{64}Cu *via* X-ray spectrometry.

The overall efficiency calibration of the detector was done using standard X-ray sources ^{48}V ($K_\alpha = 4.5$ keV), ^{55}Fe ($K_\alpha = 5.9$ keV), ^{57}Co ($E_\gamma = 14.4$ keV) and ^{125}I ($K_\alpha = 27.5$ keV, $E_\gamma = 35.5$ keV). While measuring the ^{76}Br and ^{124}I samples, besides corrections for the detector efficiency, self-absorption corrections also needed to be taken into account. In the case of ^{124}I, due to the use of a weighless source on filter paper, the self-absorption correction was neglected. For ^{76}Br the source thickness was about 1 mg/cm^2. The self-absorption of the 12 keV X-rays in selenium was estimated by interpolation of the absorption values for Cu and Sn given in [6]. It was found to be $< 3\%$.

2.3 Normalised count rates and estimation of uncertainties

The count rate of each radionuclide in each measurement was first extrapolated to end of bombardment (EOB). Thereafter, it was corrected for self-absorption (only in case of X-ray counting related to ^{76}Br). Finally it was corrected for the counting efficiency of the detector for the geometry used. The normalised count rates thus obtained corresponded practically to absolute disintegration rates except for correction for the intensity of the counted radiation. In cases where absolute disintegration rates were needed, the latter correction was also applied (see Sect. 3.3).

The uncertainties involved in the determination of normalised count rates were similar to those encountered in nuclear reaction cross section measurements, except for those in particle flux measurement. As in almost all cases the same sample was counted at the four different types of detectors mentioned above, the uncertainties in particle flux and chemical yield determination were of no significance. The major random uncertainties involved in the present work were associated with the determination of peak areas (2%), and corrections for self-absorption, pile-up and coincidence losses (2%). Due to the use of thin sources, the uncertainty in self-absorption correction in case of X-ray counting was also within this value. The major systematic uncertainty was related to the efficiency of each detector (3–6) %. The total

uncertainty in each normalised count rate was obtained by combining all the individual uncertainties in quadrature; it amounted to between 4 and 7%. It should be mentioned that in those cases where the absolute activity was determined, the total uncertainty was still within the range of 4–7%, since the uncertainty in the intensity of the used γ-ray was relatively small. In measurement of ratios of activities, on the other hand, the total uncertainty was smaller.

3. Results and discussion

3.1 Test on quantitative measurement of annihilation radiation

The annihilation of a positron mostly leads to the formation of two γ-quanta, each of energy 511 keV, which are correlated by 180°. The best way of measuring the annihilation radiation is therefore *via* a $\gamma\gamma$-coincidence system where the two detectors are correlated by 180°. In recent years, with the common availability of high-resolution HPGe detectors, the γ-ray spectroscopy has become the method of choice for the measurement of radioactivity. It is, however, observed that the automated peak analysis programmes do not give satisfactory results in the case of the 511 keV annihilation radiation since that peak is much broader than the normal photopeak.

Several samples of each of the three radionuclides under consideration (^{64}Cu, ^{76}Br and ^{124}I) were measured *via* the two methods, *viz.* $\gamma\gamma$-coincidence counting and γ-ray spectroscopy using different counting geometries and a thick enough absorber to ensure complete annihilation of positrons. The peak area analysis of the annihilation radiation in the case of γ-ray spectroscopy was done manually. The count rates obtained *via* the two methods were extrapolated to EOB and corrected for the efficiency of the respective detector. A typical result for each radionuclide is given in Table 1. The results agree within about 2%. Considering that the coincidence method entails an uncertainty of about 3% and the γ-ray spectroscopic method an uncertainty of about 5%, the results reported here may be regarded as in excellent agreement. Thus a careful γ-ray spectroscopic analysis of the annihilation radiation leads to results comparable to those obtained *via* $\gamma\gamma$-coincidence counting.

3.2 Decay data of ^{64}Cu

Each ^{64}Cu sample of very high radionuclidic purity was counted using different detectors. Of special interest was the X-ray spectrum since the energies involved are low. A typical spectrum is reproduced in Fig. 1. Evidently, only the K_α and K_β lines of the decay product Ni at 7.47 and 8.26 keV, respectively, are present. The absence of a γ-ray at 67 keV confirmed that no ^{61}Cu impurity was present. Another interesting aspect was the investigation of the high-energy part of the γ-ray spectrum; it is depicted in Fig. 2. Besides the strong positron annihilation peak at 511 keV, a weak 1346 keV γ-ray is clearly visible. In addition, the 1461 keV γ-ray of the ^{40}K natural background is also distinguishable. This clean spectrum allowed an accurate determination of the intensity of the 1346 keV γ-ray.

For each sample, the count rate from each detector was extrapolated to EOB and corrected for the efficiency of the detector. In the case of X-rays, correction was also applied

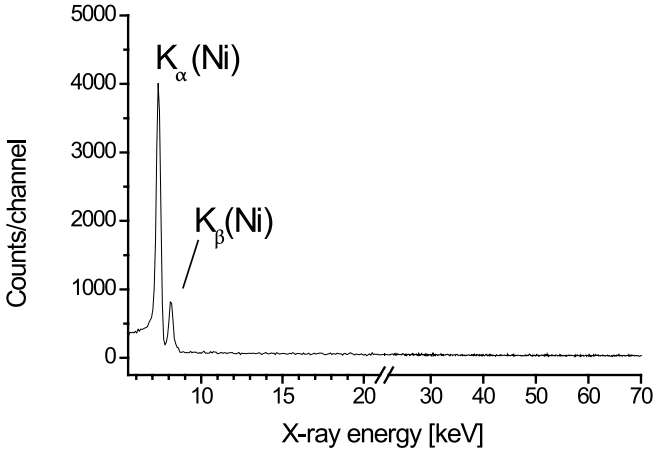

Fig. 1. X-ray spectrum of a radionuclidically pure ^{64}Cu sample taken with a Si(Li) detector.

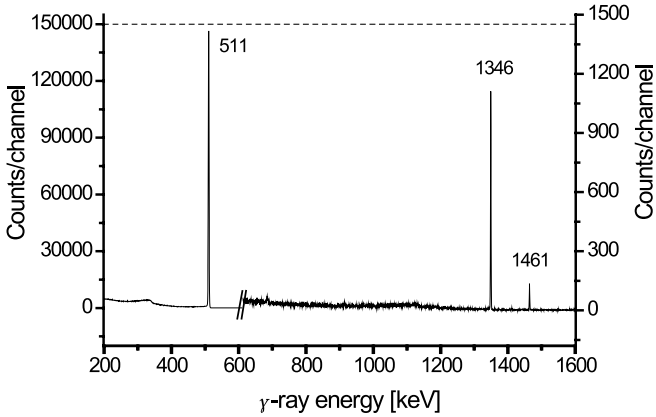

Fig. 2. γ-ray spectrum of a radionuclidically pure ^{64}Cu sample taken with a HPGe detector.

Table 1. Typical results on measurement of annihilation radiation.

Nuclide	Normalised count rate		Deviation [%]
	$\gamma\gamma$-coincidence	γ-ray spectroscopy	
^{64}Cu	477 753	486 730	1.8
^{76}Br	29 585	30 200	2.1
^{124}I	1790	1806	0.9

Table 2. Measured ratios of various types of radiation emitted by ^{64}Cu.

Decay modes	Radiation considered	Ratio
$\frac{\beta^+}{\beta^- + \beta^+}$	$\frac{(A_{511\ \mathrm{keV}}) \times 0.5}{\text{beta activity}}$	0.317 ± 0.008
$\frac{\beta^+}{\mathrm{EC}}$	$\frac{(A_{511\ \mathrm{keV}}) \times 0.5}{A_{K_\alpha\ \text{X-ray}}}$	0.407 ± 0.012
$\frac{\gamma\text{-ray}}{\beta^+}$	$\frac{(A_{1346\ \mathrm{keV}})}{(A_{511\ \mathrm{keV}}) \times 0.5}$	0.0304 ± 0.0015

Table 3. Intensities of decay modes of ^{64}Cu.

Decay mode	Intensity [%]			
		Literature		
	This work	Christmas *et al.* [7]	Kawada [8]	Wermann *et al.* [9]
β^+ emission	17.8 ± 0.4	17.86 ± 0.14	17.93 ± 0.20	
β^- emission	38.4 ± 1.2	39.04 ± 0.33	38.34 ± 0.56	38.06 ± 0.30
EC	43.8 ± 1.4	43.10 ± 0.46	43.73 ± 0.52	
1346 keV γ-ray (following EC)	0.54 ± 0.03	0.471 ± 0.01	0.487 ± 0.02	

for the fluorescence yield. The normalised count rates thus obtained were then used to build ratios of the contributions of various types of radiation emitted by ^{64}Cu. The results are given in Table 2. Each ratio is based on 3 to 5 independent measurements and each deviation is the square root of a quadratic sum of statistical and systematic uncertainties; in case of ratio measurements the latter were rather small.

The measured ratios were iteratively treated such that the relative percentages of the three modes of decay (β^-, β^+ and EC) added up to 100%. From the positron emission intensity thus determined and the measured ratio of the 1346 keV γ-ray to the annihilation radiation, the absolute intensity of the weak 1346 keV γ-ray was also obtained. The results are summarized in Table 3. The estimated uncertainty in each decay mode is 2–3% and in the 1346 keV γ-ray about 5%. For comparison, the data reported in three most recent publications [7–9] are also shown. The results for the β^-, β^+ and EC branchings agree with recent accurate measurements [7–9]. Based on known knowledge on the decay of ^{64}Cu [2, 7–9] and the data measured in this work, the decay scheme with the updated intensities of the various transitions is shown in Fig. 3. It should be mentioned that the β^- branching is very well established, also through a mass-spectrometric analysis of the daughter ^{64}Zn [9]. The β^+ branching and the intensity of EC decay are also now fairly well established. The intensity of the 1346 keV γ-ray was, however, not determined so precisely in those measurements [7, 8]. In both cases ^{64}Cu was produced *via* the ^{63}Cu$(n, \gamma)^{64}$Cu reaction so that the source was neither very pure nor very thin. In the 4π $\beta\gamma$-coincidence counting used by Kawada [8], for example, the background rate around 1340 keV gamma region was 20%. Christmas *et al.* [7], on the other hand, compared the peak area of the 1346 keV γ-ray, determined using a Ge detector, with the absolute activity of ^{64}Cu measured *via* liquid scintillation counting. Since in the present work a highly pure weighless source was used and the ratio of the 1346 keV γ-ray to the annihilation radiation was determined using the same detector, we believe our result to be more accurate.

There was some uncertainty about the EC transition to the excited 2^+ state of ^{64}Ni. The literature values vary between 0.46 and 0.60% [8]. Since the internal conversion coefficient of the 1346 keV transition to the ground state of ^{64}Ni is expected to be very small (10^{-4}), it is concluded that the intensity of the EC transition to the 2^+ state of ^{64}Ni is (0.54 ± 0.03)%, *i.e.* the same as that of the subsequent γ-transition.

3.3 Positron emission intensities in the decay of ^{76}Br and ^{124}I

Like ^{64}Cu, each ^{76}Br sample was counted using different detectors. The X-ray spectrum is shown in Fig. 4. The occurrence of X-rays of only the daughter Se shows that no non-isotopic radionuclidic impurity (*e.g.* ^{75}Se) was present. The possible contribution of the ^{77}Br impurity, which gives the same X-rays as ^{76}Br, was estimated from the decay curve analysis of the X-rays as well as through a calculation of the formation probability of ^{77}Br *via* the ^{76}Se$(p, \gamma)^{77}$Br reaction on highly enriched ^{76}Se and the ^{77}Se$(p, n)^{77}$Br reaction on

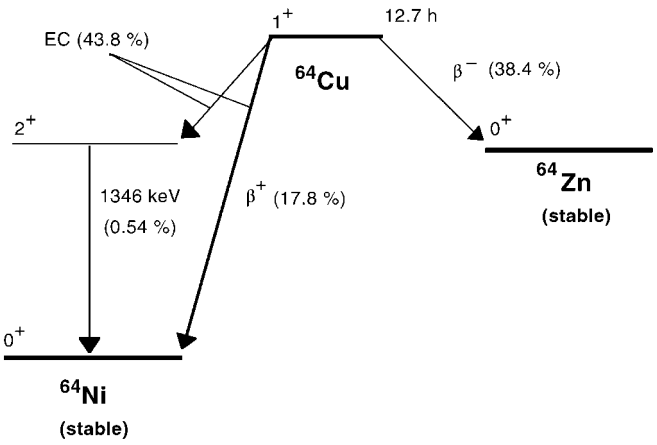

Fig. 3. Decay scheme of ^{64}Cu with intensities of emitted radiations as determined in this work.

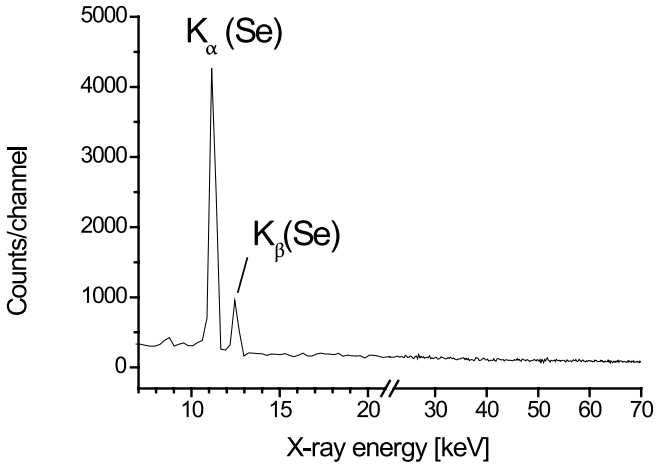

Fig. 4. X-ray spectrum of a radionuclidically pure ^{76}Br sample taken with a Si(Li) detector.

Table 4. Methods used for measurement of the positron emission intensity of ^{76}Br.

Counting methods used	Calculational formula for determining I_{β^+} [%]
Beta counting and γ-ray spectroscopy	$\frac{A_{\beta^+}}{(\text{absolute activity } [^{76}\text{Br}])^a} \times 100$
γ-ray spectroscopy	$\frac{(A_{511\text{ keV}}) \times 0.5}{(\text{absolute activity } [^{76}\text{Br}])^a} \times 100$
γ-ray and X-ray spectroscopy	$\frac{(A_{511\text{ keV}}) \times 0.5}{(A_{511\text{ keV}}) \times 0.5 + (A_{K_\alpha\text{X-ray}})} \times 100$

a: Absolute activity of ^{76}Br (at EOB) was determined using the 559 keV γ-ray ($I_\gamma = 0.74$).

Table 5. Positron branching in the decay of ^{76}Br.

Method of measurement	Intensity [%]
Beta counting	59.3 ± 2.4
γ-ray spectroscopy	59.2 ± 1.8
X-ray spectroscopy	56.6 ± 1.7
Weighted average	$\mathbf{58.2 \pm 1.9}$

the 0.05% ^{77}Se impurity present in the irradiated enriched ^{76}Se target. The cross section data for the two reactions were deduced from the literature [5, 10]. The contribution of ^{77}Br activity relative to the ^{76}Br activity was estimated to be $< 0.2\%$ and was therefore neglected.

The positron emission intensity of ^{76}Br was determined *via* three methods, which are summarized in Table 4. All count rates (A_{β^+}, $A_{511\text{ keV}}$ and $A_{\text{k-X-ray}}$) refer to values at EOB and were normalised for the efficiency of the detector used as well as for the fluorescence yield in case of X-rays. The results on I_{β^+} obtained *via* various methods are summarized in Table 5. Each value is based on three independent measurements and each deviation is the square root of a quadratic sum of the standard deviation and the systematic uncertainty. Due to somewhat larger uncertainty in beta counting, in the averaging process, we gave double weight to γ-ray and X-ray spectroscopic measurements. Thus, using a weighting factor of (1 : 2 : 2) to the three measurements listed in Table 5, a value of $(58.2 \pm 1.9)\%$ for I_{β^+} of ^{76}Br was obtained. This is close to the I_{β^+} value of $(56.0 \pm 1.4)\%$ given in the evaluated data file [2].

It should be mentioned that detailed investigations on the decay of ^{76}Br were carried out [11] using scintillation detectors both for β^+ spectrum analysis and γ-ray spectroscopy. For X-ray counting a gas proportional counter was used. The radionuclide was produced *via* the ^{75}As$(\alpha, 3n)^{76}$Br reaction at 50 MeV and was radiochemically separated from the non-isotopic impurities. The sample contained a high proportion of ^{77}Br but its contribution could be taken into account by decay curve analysis. The experimentally determined value [11] for β^+ branching in ^{76}Br was given as $(62.5 \pm 7.5)\%$. Later studies concentrated on γ-ray spectrometric measurements using Ge(Li) detectors (for references to original works see [2]). In the latest evaluation, the I_{β^+} value has been reduced to 56%, mainly on the basis of adjustments of γ-ray intensities. The experimental techniques used in the present work were superior to those employed earlier: the ^{76}Br-sample used was free from all radionuclidic impurities and the X-rays were analysed spectroscopically using a Si(Li) detector. Our result may therefore be considered as more accurate; however, it is close to the evaluated value.

Similar to ^{64}Cu and ^{76}Br, each ^{124}I sample was also counted using different detectors. The I_{β^+} value obtained *via* a ratio of the annihilation peak intensity to the Te X-ray intensity has already been reported [3]. In the present work we concentrated on validation of the annihilation peak measurement by $\gamma\gamma$-coincidence counting (as described in Sect. 3.1) and on the determination of the ratio of the beta count rate to the absolute activity of ^{124}I. The latter was obtained *via* γ-ray spectroscopy using the γ-rays of energies 603 keV ($I_\gamma = 0.61$) and 1691 keV ($I_\gamma = 0.104$) as well as *via* X-ray spectroscopy using the K_α X-rays of energy 27.4 keV ($I_{\text{X-ray}} = 0.475$). From positron counting of three independently prepared samples we obtained a value of $(22.1 \pm 0.5)\%$ for the I_{β^+} of ^{124}I. This value is in excellent agreement with the value of $(22.0 \pm 0.5)\%$ determined by us previously from γ-ray spectroscopic investigations [3] as well as with the value of $(21.6 \pm 0.4)\%$ reported by Woods *et al.* [12] on the basis of 4π $\beta\gamma$-coincidence measurements on a mass separated ^{124}I sample.

3.4 Implications of precise nuclear data and conclusions

Due to quantitative nature of positron emission tomography, a proper knowledge of positron branching in the decay of the used radionuclide is mandatory. Though for a radionuclide like ^{76}Br, with a high β^+ branching of about 58%, a small uncertainty in the positron emission intensity is not very critical, for ^{124}I and ^{64}Cu, with relatively low positron emission rates, an accurate knowledge of the intensities is most essential. The I_{β^+} of ^{124}I was determined accurately a few years ago [3] and the present measurement done using complementary techniques confirms that value. Regarding ^{64}Cu, our measurement should provide a more reliable value.

For positron emitters with a few associated γ-rays of well-known intensities, *e.g.* ^{76}Br and ^{124}I, the absolute radioactivity can be measured *via* γ-ray spectroscopy. This is especially valuable while measuring nuclear reaction cross sections. In the case of ^{64}Cu, which does not emit any prominent γ-ray, the determination of absolute activity can be done *via* beta counting, $\gamma\gamma$-coincidence counting, analysis of the 511 keV annihilation radiation in a γ-ray spectrum, or *via* X-ray spectrometry. All those methods are, however, not radionuclide specific, unless a chemical separation is done and the half-life of ^{64}Cu is carefully analysed. In recent years, we reported cross sections for the formation of ^{64}Cu *via* the reactions ^{64}Ni$(p, n)^{64}$Cu [13], ^{68}Zn$(p, \alpha n)^{64}$Cu, ^{66}Zn$(d, \alpha)^{64}$Cu and $^{\text{nat}}$Zn$(d, x)^{64}$Cu [4] using enriched target material (in the first three cases), chemical separation and annihilation radiation characterisation. The I_{β^+} was adopted as 19.0%. In view of the new I_{β^+} value of 17.8%, the reported cross section values need to be normalised. The reaction ^{64}Ni$(p, n)^{64}$Cu on highly enriched ^{64}Ni (cf. [13]) has become the method of choice for production at a small-

sized cyclotron and thick target yields under production conditions have been reported [14, 15]. The normalisation of cross sections with respect to the new I_{β^+} value would now lead to theoretical yields of ^{64}Cu about 7% higher than the previous values. This enhancement is, however, within the limits of experimental uncertainties of measured cross sections.

A few authors have measured cross sections of the ^{68}Zn$(p, \alpha n)^{64}$Cu, ^{66}Zn$(p, 2pn)^{64}$Cu, ^{nat}Zn$(p, x)^{64}$Cu and ^{nat}Zn$(d, x)^{64}$Cu reactions using the weak 1346 keV γ-ray [16–19]; they mostly adopted the value of I_γ as 0.47%. There are considerable discrepancies between the data measured by us on zinc target isotopes using the annihilation radiation and by other groups using the 1346 keV γ-ray. In view of the new value of $I_\gamma = 0.54\%$ determined in this work, all the cross sections reported in the literature [16–19] should be decreased by 15%. Thus an increase of ^{64}Cu formation cross sections measured by us [4] by 7% and a decrease of cross sections measured by others [16–19] by 15% would lessen some of the discrepancies, but certainly not remove them. The present work, on the other hand, dispels one suspicion that the discrepancies could be due to use of faulty decay data.

The above conclusion is supported by a recent study by Szelecsényi *et al.* [16] who determined the ^{68}Zn$(p, \alpha n)^{64}$Cu reaction cross sections using enriched target material, chemical separation and γ-ray spectroscopic analysis of both the annihilation radiation and the weak 1346 keV γ-ray. The two results were found to be in agreement within the limits of experimental uncertainties. In contrast, in cases where natural targets were used, and particularly when no chemical separation was done, the analysis of the 1346 keV γ-ray may be associated with large uncertainties. On the other hand, the radiochemical method may also entail considerable uncertainties in the determination of separation yield. Apparently, in order to solve discrepancies in the ^{64}Cu formation cross sections, some more careful measurements and evaluations are necessary.

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