Large discrepancies in the excitation function data of the $^{68}\text{Zn}(p,x)^{64}\text{Cu}$ reaction: a possible explanation

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Abstract. The excitation function of the $^{68}\text{Zn}(p,x)^{64}\text{Cu}$ reaction was investigated in an attempt to clarify a serious discrepancy in the recently published data. New measurements based on both a weak $\gamma$-line of 1345.8 keV (0.47%) as well as the 511 keV annihilation radiation were performed after radiochemically separating the Cu from the Zn target matrix. In the case of the 511 keV measurements, the method of decay-curve analysis was employed as the annihilation radiation is not specific for a particular radionuclide. The results from the two methods were found to be in excellent agreement. Simulations were also performed to test the method of 511 keV decay-curve analysis for the effects of possible intruder contaminants.

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1. Introduction

Recently, $^{64}\text{Cu}$ has been one of the radioisotopes investigated for use in internal radiotherapy. The decay properties of $^{64}\text{Cu}$ (I$_{\beta^+}$ = 9%, I$_{\beta^-}$ = 40% and I$_{EC}$ = 41%) make it a promising candidate for endo-radiotherapy with the added bonus of assessing the degree of uptake of the $^{64}\text{Cu}$-labelled therapeutic agent, as well as continuous monitoring thereof in the area around the tumour, using PET.

Surprisingly, recent studies of the $^{68}\text{Zn}(p,x)^{64}\text{Cu}$ excitation function (which has been investigated as a promising reaction for routine production) have yielded very different results. The latest measured cross sections differ by more than a factor of 3, leading to completely different conclusions by the respective experimental teams as to the viability of this reaction for routine $^{64}\text{Cu}$ production.

The large disagreement between the excitation function data of Levkovskij [1] and Hilgers et al. [2] (by a factor of between 3 and 4) is shown in figure 1. This disagreement has been further investigated in the present study. Although Levkovskij does not mention the experimental details of his measurements, it is believed to be based on a conventional stacked-foil experiment, using the only viable $\gamma$-line of 1345.8 keV (0.47%) emitted in the decay of $^{64}\text{Cu}$. In contrast, Hilgers et al. first radiochemically separated the Cu from the Zn target matrix after irradiation of their foil stacks, followed by a decay-curve analysis of the 511 keV annihilation radiation emitted by their sources.

During several experimental runs at iThemba LABS, new data sets were obtained for $p+^{68}\text{Zn}$ on highly enriched target foils (> 98% $^{68}\text{Zn}$) by utilizing both of the above-mentioned methods. As shown in figure 1, the measurements based on the rather weak 1345.8 keV $\gamma$-line and the 511 keV annihilation radiation are in good agreement. Clearly, both data sets also support the values of Levkovskij. Here we speculate as to the possible reasons for the discrepancy and whether it can possibly be related to the method of decay-curve analysis.
2. Decay-curve analysis

A typical example of a 511 keV decay curve obtained at iThemba LABS is shown in figure 2. After chemical separation, liquid sources (10 ml of eluate sealed in serum vials) as well as dry sources (prepared by evaporating a fraction of the eluate to dryness in Teflon point-source holders) were counted repeatedly. The method of counting the 511 keV annihilation $\gamma$-rays was adopted from the work of Kawade et al. [3], utilizing an HPGe detector and a set of acrylic $\beta^+$ source absorbers.

Three $\beta^+$ emitting radioisotopes were identified from their 511 keV as well as other respective decay emissions. These are $^{64}$Cu, traces of $^{65}$Zn which were not completely removed by the chemistry and a relatively small $^{61}$Cu component produced from small amounts of $^{64}$Zn (< 1%) and $^{66}$Zn (< 1%) impurities present in the enriched $^{68}$Zn target foils. The dashed curve in figure 2 is a non-linear regression fit through the measured data of the following decay function:

$$ C(t) = a_0 + \sum_{i=1}^{n} a_i \exp(-\lambda_i t) $$

(1)

Figure 2. A typical decay curve of one of the sources prepared to measure the excitation function of $^{64}$Cu. The solid symbols are measured data points while the dashed curve is a non-linear regression fit of Equation (1) through the data (see text). Note that the symbol size exceeds the error bars.
where $C(t)$ is the count rate at a time $t$ after the end of bombardment (EOB), $a_i$ is the individual EOB count rate from the decay of radioisotope $i$ with decay constant $\lambda_i$ and $a_0$ is a background count-rate term. In this particular case, $n = 3$ (i.e. terms for $^{61}$Cu, $^{64}$Cu and $^{65}$Zn are provided for). Equation (1) was added to the User Defined Function Set of the well known TABLECURVE software package [4] in order to perform the regression fits.

In principle, all contributing radioisotopes (to the 511 keV emission) can be determined in this way provided that there is no series decay present. However, one should be concerned about the possible inaccuracy of the method. In order to investigate this further, we performed several simulations by adding a fictitious quantity of activity to the measured data and then re-analyzing the decay curve of Equation (1), first with the correct number of terms (i.e. $n = 4$), thereafter with either one too few (i.e. $n = 3$) or one too many (i.e. $n = 5$) terms. Here we only present two selected cases of those studies. In figure 3, a contaminant with an EOB activity of 10% of that of $^{64}$Cu was added to the measured data and analyzed as if its presence was unknown (i.e. with $n = 3$). The resultant deviation of the extracted $^{64}$Cu activity is plotted as a function of the contaminant half-life. As can be seen in figure 3, when the contaminant has a similar half-life to that of $^{64}$Cu the deviation is 10%, as expected. At larger contaminant half-life values, the deviation is somewhat larger that 10%. Nevertheless, one can say that the deviation is of the same order of magnitude as the level of the intruder contaminant. (Note that at this level of contamination, one could still obtain a good regression fit. Significantly above 10%, however, this was no longer the case as the fit would clearly deteriorate.)

![Figure 3. Percentage deviation of the $^{64}$Cu activity extracted from the decay-curve analysis if a 10% contaminant is introduced (see text) plotted as a function of the contaminant half-life.](image)

In figure 4, the deviation of the intruder activity is plotted as a function of its half-life, in this case using the correct number of terms in Equation (1), i.e. $n = 4$. It is plotted as the ratio of the activity extracted from the regression fit and the actual activity, which should be unity if the analysis is good. While one would expect a problem if the intruder radioisotope has a half-life close to that of $^{64}$Cu, it is surprising that a large deviation is found at intruder half-lives much smaller or much larger than that of $^{64}$Cu, even exceeding an order of magnitude. We found a good extraction from the analysis only at intruder half-life values larger than about 4 times that of $^{64}$Cu. Thus, at half-lives shorter than $\sim 4T_{1/2}$ of $^{64}$Cu, the decay-curve analysis failed to accurately extract the known value of the intruder activity which was deliberately added to simulate the effect of such a contaminant. From this example, one can make the further statement that if $^{64}$Cu would be one of several isotopes in a mixed source, without a sufficiently long experimental time window in which it is the dominant 511 keV emitter, the decay-curve analysis might similarly also fail to extract its activity accurately.
3. Conclusions

Not surprisingly, 511 keV decay-curve analysis can give very spurious results if the number of terms in Equation (1) are either too few or too many, even if the actual fit through the data appears to be good. We believe that the activity of only a very dominant contributing isotope can be extracted accurately for a given window of time. That time window should also be sufficiently long, at least $0.5T_{1/2}$ to $2T_{1/2}$ or more. In our opinion, the decay-curve method can be very unreliable under certain conditions, thus the data analysis should be augmented with information from other available emissions, if possible. We could not determine without doubt whether the discrepancy between the various sets of cross sections of the $^{64}$Cu excitation function could be attributed to failure of the decay-curve method, therefore we want to stress that we do not make such a statement. Our objective here is merely to point out that our simulation studies indicate that 511 keV decay-curve analyses (the radiation of which is in itself not specific for a particular radionuclide) can sometimes fail, therefore it is a possible explanation for discrepancies in such cases. We also do not make the statement that the set of lower values has conclusively been shown to be incorrect. The results of this study, however, support the values of Levkovskij, i.e. the higher values. Lastly, Hilgers et al. made the statement that since $^{64}$Cu decay only yields a weak 1345.8 keV (0.47%) $\gamma$-line, the 511 keV decay-curve analysis was mandatory. Our conclusion is that in spite of the fact that the 1345.8 keV $\gamma$-line is weak, it is nevertheless suitable for the experimental determination of the $^{68}$Zn(p,x)$^{64}$Cu excitation function.

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