64Cu enrichment using the Szilard-Chalmers effect – The influence of γ-dose

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ABSTRACT

Cu is an important trace metal which plays a role in many biological processes. The radioisotope 64Cu is often used to study such processes. Furthermore, 64Cu finds applications in cancer diagnostics as well as therapy. For all of these applications 64Cu having high specific activity is needed. 64Cu can be produced in cyclotrons or in nuclear reactors. In this paper we study the effect of gamma dose on the production of 64Cu according to the Szilard-Chalmers reaction using Cu(II)-phthalocyanine as a target. For this purpose, irradiations were performed in the nuclear reactor of the Delft University of Technology using a novel irradiation facility helping to limit the dose produced by gammas present in the reactor pool. The obtained 64Cu activity yield was in general above 60% in accordance to the theoretical expected value. An increase in gamma dose has no significant influence on the obtained activity yield but increases the loss of Cu from Cu(II)-phthalocyanine up to 0.9% and hence decreases the specific activity that can be obtained. However, without optimisation, when reducing the gamma dose specific activities in the order of 30 TBq/g can be achieved.

1. Introduction

Cu is a trace element important in numerous biological processes like neurotransmitter synthesis and iron metabolism (Kim et al., 2014), but it is also involved in tumour angiogenesis and neurodegenerative diseases (Laura et al., 2013). 64Cu is a radioactive isotope of Cu with a half-life of 12.7 h, decaying either by electron capture (44%) and β+ (17.5%) to stable 64Ni or by β− (38.5%) to stable 64Zn (Laboratory, 2000). Because of these decay characteristics 64Cu can be used in imaging i.e. in positron emission tomography (PET) and/or in radionuclide therapy (Chakravarty et al., 2016; Laura et al., 2013). For these applications a specific activity of at least 1 TBq/g is needed (Kim et al., 2014).

64Cu can either be produced by proton activation in a cyclotron or by neutron activation in a nuclear reactor. In proton activation nickel targets are bombarded with a beam of 15.5 MeV protons (McCarthy et al., 1997), allowing for the 63Ni(p,n)64Cu reaction to take place. After irradiation 64Cu has to be separated from the Ni target. The advantage of this method is that non-carrier added 64Cu is produced. However, special targets and cooling are needed (Le et al., 2009). In neutron activation either Zn or Cu targets can be used. The 64Zn(n,p)64Cu reaction needs fast neutrons to take place, which are available in irradiation facilities situated in the core of nuclear reactors (McCarthy et al., 1997). In those facilities often only small volumes can be irradiated, making this production less likely to meet regular clinical demands (Chakravarty et al., 2016). Furthermore, long lived 65Zn is co-produced as waste. For the 63Cu(n,γ)64Cu reaction thermal neutrons are needed. Although many reactors exist where this production route can be implemented, the specific activity of the produced 64Cu is low. Using enriched targets and high flux reactors leads to higher specific activity, but increases costs. Moreover, as natural Cu contains 69.18% 63Cu (Laboratory, 2000), using enriched targets only allows for a factor of maximum 1.5 increase in SA.

The problem of low specific activity 64Cu can be overcome by utilising the recoil effect that occurs during (n,γ) reactions. In 1934 L. Szilard and T.A. Chalmers (Szilard and Chalmers, 1934a, b) already showed that enriched 125I can be produced using natural targets and chemical effects due to nuclear transformations. During neutron activation the energy of the excited state is distributed between the emitted prompt gamma and the newly formed product nucleus. (Fig. 1). The recoil energy of the product nucleus can be calculated using the following equation: $E_r = \frac{537}{M} E_{\gamma}$, in which $E_r$ is the recoil energy of the product nucleus in eV, $E_{\gamma}$ the average energy of the prompt gamma in MeV and $M$ the mass of the recoiling atom in u. If the recoil energy is

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higher than the energy of chemical bonds (typically in the order of eV (Song and Le, 2017)) the product nucleus will be released from its chemical environment (Yoshihara and Sekine, 2011). Provided that the recoiling atoms and the target are in different chemical form, the enriched isotope can easily be collected.

In 1986 E.L. Hetherington et al. (1986) reported that they could achieve specific activities up to 40 TBq/g Cu using 250 mg Cu (II)-phthalocyanine targets utilising the Szilard-Chalmers effect for an irradiation of 12 h with a neutron flux of $5\times10^{17}$ n/m$^2$s. Longer irradiations resulted in lower specific activities. These irradiations were performed in the high flux Australian Reactor (HIFAR), a heavy water nuclear reactor. They concluded that using the Szilard-Chalmers method is an effective way to produce $^{64}$Cu in nuclear reactors with a moderate thermal neutron flux, however no explanation was given for the decrease in specific activity. This decrease can possibly be attributed to the destruction of the target due to gamma radiation present in the reactor pool, leading to non-active Cu being released. However, the effect of the gamma dose on the specific activity of $^{64}$Cu using this target or similar has not been researched so far.

The objective of the present paper is to investigate the influence of the gamma dose on the specific activity of $^{64}$Cu using Cu(II)-phthalocyanine as a target and a special irradiation facility allowing for decreasing the gamma dose with little loss of neutron flux. Cu(II)-phthalocyanine is chosen because of its known chemical stability (Shapkin et al., 1977).

2. Method

The applied method was based on the procedure described by Hetherington et al. (1986). Samples of 1, 5, 10, 20 and 100 mg of Cu (II)-phthalocyanine (dye content 9, Sigma Aldrich, 11 w% Cu) were irradiated in different facilities having different characteristics at the Hoger Onderwijs Reactor at the Delft University of Technology which we simply numbered as 1 and 2. We also used a special lead shielded facility allowing to change the gamma dose. This facility was denoted with FB3, 6 and 9, where 3, 6 and 9 refer to the cm of lead used. The neutron fluxes and the gamma dose of all facilities are given in Table 1.

After irradiation the samples were opened and dissolved in 2 mL 97% H$_2$SO$_4$ (J.K. Baker). This solution was slowly transferred into 20 mL MilliQ water (in house Millipore MilliQ system). 12 mL of 20% NH$_4$OH (J.K. Baker) was added to the Cu(II)-phthalocyanine solution to neutralise the pH. The neutralised solution was transferred onto a 1 g acetic acid buffer conditioned Chelex 100 (100–200 mesh, sodium form, Sigma- Aldrich). A 1 M pH 5 acetic acid buffer was used to condition the Chelex. After all Cu(II)-phthalocyanine solution was passed over the column, the column was rinsed with 8 mL of MilliQ water. Then 16 mL of 1 M HCl (stock solution was made by diluting 30% HCl (Sigma Aldrich)) was passed over the column to remove the trapped Cu-ions. Again, the column was rinsed with 8 mL of MilliQ water. All fractions were collected in separate PE counting vials (Wheatman). Also, the Chelex was transferred to a counting vial. All fractions were measured by gamma spectroscopy (Wallac$^\text{TM}$ gamma counter, PerkinElmer). The Cu concentration of all liquid fractions was determined by elemental analysis using ICP-OES (Optima 4300, PerkinElmer). Using this data the
The neutron fluxes for the different irradiation facilities of the Hoger Onderwijs Reactor Delft that were used in this work with their corresponding gamma dose rates. The gamma dose was actually a combination of neutron and gamma dose since the two cannot be measured independently. To be able to compare the different facilities and the impact of the gammas, the gamma dose was normalised relative to the thermal neutron flux.

| Name of Irradiation facility | Thermal neutron flux [n/\text{m}^2\text{s}] | Epithermal neutron flux [n/\text{m}^2\text{s}] | Fast neutron flux [n/\text{m}^2\text{s}] | Gamma dose rate [\text{Gy}/\text{s}] | \(\gamma\) Dose per thermal neutron [\text{Gy}/\text{n}] |
|-------------------------------|------------------------------------------|------------------------------------------|------------------------------------------|---------------------------------|-------------------------------|
| FB3                           | \(6.22 \times 10^{16}\)                  | \(1.44 \times 10^{15}\)                  | \(4.39 \times 10^{15}\)                  | 29.33                           | 0.469                          |
| FB6                           | \(5.64 \times 10^{16}\)                  | \(1.04 \times 10^{15}\)                  | \(3.28 \times 10^{15}\)                  | 19.47                           | 0.359                          |
| FB9                           | \(3.88 \times 10^{16}\)                  | \(6.00 \times 10^{14}\)                  | \(1.94 \times 10^{15}\)                  | 11.61                           | 0.299                          |
| FB1 (a pneumatic facility)    | \(3.11 \times 10^{16}\)                  | \(7.20 \times 10^{14}\)                  | \(2.60 \times 10^{15}\)                  | 62.65                           | 2.01                           |
| 2                             | \(2.30 \times 10^{17}\)                  | \(1.74 \times 10^{16}\)                  | \(7.91 \times 10^{16}\)                  | 485.8                           | 2.11                           |
| 60Co source                   | –                                        | –                                        | –                                        | –                               | –                              |

Fig. 3. The specific activity (SA, calculated at the End of Bombardment) as function of the integrated \(\gamma\) dose. Cu(II)-phthalocyanine mass is 20 ± 0.2 mg. Error bars correspond to experimental uncertainty based on \(n = 3\), except for the 0.359 Gy/n that received 700 kGy. There the error bars are based on \(n = 8\). The samples receiving 0.472 Gy/n, 0.359 Gy/n and 0.299 Gy/n were irradiated in a lead shielded facility, while the samples receiving 2.01 Gy/n and 2.11 Gy/n had no shielding.

The yield of the reaction was determined according to: \(\text{yield} = \frac{\text{CPM}_{\mu \text{C}}}{\sum \text{CPM}_{\mu \text{C}}}\)

with \(\text{CPM}_{\mu \text{C}}\) the activity measured in the HCl fractions that contain the freed \(^{64}\text{Cu}\) and \(\sum \text{CPM}_{\mu \text{C}}\) the sum of the activity of all the fractions. The enrichment factor of the Szilard-Chalmers method over normal neutron activation was calculated using: \(\text{EF} = \frac{\text{SA}_{\mu \text{C}}}{\text{SA}_{\text{SC}}}\), with \(\text{SA}_{\mu \text{C}} = \frac{\text{A}_{\text{Cu}}}{\text{A}_{\text{HCl}} \cdot \text{V}_{\text{HCl}} \cdot \text{m}_{\text{Cu}}}\) and \(\text{SA}_{\text{SC}} = \frac{\text{A}_{\text{produced}}}{\text{m}_{\text{Cu}}}\).

3. Results and discussion

Cu(II)-phthalocyanine is irradiated with neutrons at the Reactor Institute Delft in the facilities shown in Table 1. Samples receiving a 0.299 Gy/n, 0.359 Gy/n or 0.472 Gy/n dose are irradiated in the lead shielded facility. Samples receiving a 2.01 Gy/n or 2.11 Gy/n dose are irradiated in non-shielded facilities. After irradiation bond ruptured \(\text{Cu}\) and thereby lowering the yield obtained. Furthermore, a pH of 1 for the 0.1 M HCl solution is essential to be able to remove \(^{64}\text{Cu}\) from the Chelex.

3.1. Influence of the gamma dose on yield and SA

Since the samples receive simultaneously a gamma and a neutron dose irrespective of the facility, we have performed irradiations in a \(^{60}\text{Co}\) source to determine only the effect of gamma radiation.

Fig. 2A shows clearly that the integrated radiation dose received by the samples influences the amount of \(\text{Cu}\) that is released from the Cu(II)-phthalocyanine complex. Although there is always a small percentage of \(\text{Cu}\) that is freed, even if irradiations do not take place, an evident increase of \(\text{Cu}\)-release is observed as the integrated radiation dose increases. The irradiations performed in the \(^{60}\text{Co}\) source show that the gamma dose is most likely responsible for the loss of \(\text{Cu}\) since the released amount of \(\text{Cu}\) is comparable to samples that received a comparable integrated radiation dose constituted of both a gamma dose and a neutron dose. Only the non-irradiated, non-heated sample is an outlier with its 0.37 ± 0.07% Cu loss.

The Cu loss naturally has a direct influence on the SA produced. The effect of the integrated \(\gamma\) dose on the SA is given in Fig. 3. According to expectations, the SA decreases when the integrated \(\gamma\) dose received by the samples increases, up to 2000 kGy. There is no obvious explanation for this observation and further experiments are needed to determine whether there is a clear trend and what its cause is. Furthermore, it can be observed that the lead shielded samples, corresponding to 0.299 Gy/n, 0.359 Gy/n and 0.472 Gy/n, have a higher SA compared to the non-shielded samples, corresponding to 2.01 Gy/n and 2.11 Gy/n, when normalised relative to the neutron flux. This indicates that the lead shielding used to reduce the gamma dose during neutron irradiation has a strong influence on the quality of \(^{64}\text{Cu}\) that can be produced. However, the reduction in SA (i.e. factor 2 between the lowest and the highest integrated \(\gamma\)-dose) is not as strong as the \(\text{Cu}\) loss (i.e. factor 5.6 between the lowest and the highest integrated \(\gamma\)-dose) might have suggested. The \(\text{Cu}\) loss depends naturally has a direct influence on the SA produced. The effect of the integrated \(\gamma\) dose on the SA is given in Fig. 3. According to expectations, the SA decreases when the integrated \(\gamma\) dose received by the samples increases, up to 2000 kGy. There is no obvious explanation for this observation and further experiments are needed to determine whether there is a clear trend and what its cause is. Furthermore, it can be observed that the lead shielded samples, corresponding to 0.299 Gy/n, 0.359 Gy/n and 0.472 Gy/n, have a higher SA compared to the non-shielded samples, corresponding to 2.01 Gy/n and 2.11 Gy/n, when normalised relative to the neutron flux. This indicates that the lead shielding used to reduce the gamma dose during neutron irradiation has a strong influence on the quality of \(^{64}\text{Cu}\) that can be produced. However, the reduction in SA (i.e. factor 2 between the lowest and the highest integrated \(\gamma\)-dose) is not as strong as the \(\text{Cu}\) loss (i.e. factor 5.6 between the lowest and the highest integrated \(\gamma\)-dose) might have suggested. The \(\text{Cu}\) loss depends on the activity produced, which depends on the neutron flux and irradiation time. The samples with the highest integrated gamma dose received four times more neutrons compared to the samples receiving the lowest integrated gamma dose. This has a direct influence on the SA and will compensate for the Cu loss to some extent.
The gamma dose does have a small influence on the yield (Fig. 4). The yield of the non-shielded irradiation samples (i.e. 2.01 fGy/n and 2.11 fGy/n) varies between 46% and 56%, while for the other samples it ranges between 58% and 64%. The theoretical yield is calculated to be 60.7%. $^{64}$Cu is known to emit many energetic prompt gammas (Tuli, 1999). If the resulting recoil energy from these prompt gammas is high enough, chemical bonds will break (Yoshihara and Sekine, 2011). The Cu bonds in Cu(II)-phthalocyanine are 6.96 eV (Luo, 2009; Sanderson, 1977), so the minimal energy of the prompt gammas required to break that bond is 904 keV. 60.7% of the prompt gammas emitted during neutron irradiation of Cu are 904 keV or higher (Tuli, 1999). The reduction in yield can be caused by reduced solubility or crosslinking of the Cu(II)-phthalocyanine complexes limiting the amounts that can be extracted. The theoretical maximum of the prompt gammas required to break that bond is 904 keV. 60.7% of the prompt gammas emitted during neutron irradiation of Cu are 904 keV or higher (Tuli, 1999). The reduction in yield can be caused by reduced solubility or crosslinking of the Cu(II)-phthalocyanine complexes limiting the amounts that can be extracted. The theoretical maximum SA (carrier free $^{64}$Cu) that can be reached is $1.4 \times 10^6$ TBq/g. However, there will always be some degree of Cu loss due to interactions within molecules, resulting in a lower SA.

When determining the gamma dose in the different irradiation facilities, the determined dose also contains a contribution due to the neutrons. It is impossible to determine these doses independently. Furthermore, the different irradiation facilities have different neutron fluxes. To compensate for the different neutron fluxes the samples are irradiated for the same integrated neutron flux, producing the same amount of activity. The influence of this irradiation time difference is expected to be minimal as saturation activity is reached at much longer irradiation times. (Supplemental information S1) The $^{64}$Cu yield, SA, and Cu in HCl phase is determined for four different integrated neutron fluxes (Fig. 5).

The $^{64}$Cu yield is not affected by the increase in integrated neutron flux (Fig. 5 A). As the yield is defined as a fraction of the total activity, it is independent from the activity produced. On the other hand, the SA is affected by the increase in integrated neutron flux (Fig. 5 B). It is expected that at some point the saturation SA is reached. With the increase in Cu loss it is more likely for the SA to decrease, unless the Cu loss and fluxes (Fig. 5).
the increase in activity are at equilibrium. Furthermore, a clear difference between the shielded samples (0.473 fGy/n, 0.359 fGy/n and 0.299 fGy/n) and the non-shielded samples (2.01 fGy/n and 2.11 fGy/n) is visible. The non-shielded samples have a lower SA and a higher Cu loss compared to the shielded samples, for the same integrated neutron flux. This difference can clearly only be attributed to the gamma dose received by the samples.

Influence of irradiation time and sample mass on yield and SA.

The influence of sample mass is also evaluated as function of irradiation time. The irradiations are performed in the facility having the shortest irradiation time. A possible explanation for this decrease in yield could be that the maximum solubility of Cu(II)-phthalocyanine in sulphuric acid is reached. For the samples up to 20 mg this is unlikely because the vials are shaken thoroughly before transferring the solution into water and no relative increased amount of activity was measured in the Chelex phase. However, for the 100 mg samples this is a likely scenario, as the solution became strongly viscous and relatively more activity was found in the Chelex fraction. Increasing the volume of H₂SO₄ to dissolve the 100 mg sample, increased the yield (49.4 ± 1.3% when using 2 ml H₂SO₄ vs 51.8 ± 1.9% when using 3 ml H₂SO₄) Secondly, the Chelex could be saturated, but it is unlikely since it is in a 2–10 fold excess relative to the amount of Cu ions. Moreover, if the Chelex would be saturated an increased amount of activity would be present in the first 2 fractions collected which was not observed.

The specific activity of ⁶⁴Cu produced increases with the irradiation time and then stabilises for all masses (Fig. 6B). This is expected as with longer irradiation times more atoms get activated and there is only a marginal increase in Cu loss when the samples are irradiated in the lead shielded facility (Fig. 6C). However, these results are contradicted by what is previously reported by E.L. Hetherington et al. (1986). They reported a decrease in SA when increasing the irradiation time. It must be noted that the flux used by E.L. Hetherington et al. was approximately 10 times higher than in this research. This could explain for an important part the differences in the reached specific activity. Furthermore, the shortest irradiation time reported by E.L. Hetherington (Hetherington et al., 1986) was 12 h, while in this research that was 1 h. It is likely that the gamma dose received by E.L. Hetherington’s samples is much higher compared to the lead shielded samples in this research, resulting in the different behaviour in SA between their results and the results presented here. For the 10 mg, 20 mg and 100 mg the percentage of Cu found in the HCl phase is only slightly elevated when irradiated for 5 h at 150 °C compared to the release when also irradiated with gammas. The ⁶⁰Co irradiated samples received a gamma dose of 402 kGy. The samples contained 20 ± 0.3 mg of Cu-phthalocyanine. The experimentally determined standard deviation is based on n = 3.

| Sample Treatment          | Non-treated | ⁶⁰Co | ⁶⁰Co and heating | Heating only |
|---------------------------|-------------|------|-----------------|-------------|
| Cu concentration [mg/l]   | 0.497 ± 0.096 | 0.251 ± 0.046 | 0.217 ± 0.005 | 0.233 ± 0.009 |
| Cu in HCl phase [%]       | 0.371 ± 0.074 | 0.196 ± 0.032 | 0.192 ± 0.024 | 0.164 ± 0.024 |

The enrichment factor (EF) is defined as the factor with which the specific activity is increased using the Szilard-Chalmers method compared to no separation from the target for the same irradiation conditions. An order of magnitude increase is found between the 1 mg and 10 mg samples (Fig. 6B). This can be explained by the fact that for those samples there is also an order of magnitude difference between the SAs produced using the Szilard-Chalmers effect.

Influence of temperature on stability and structure.

During neutron irradiation in the various facilities not only the gamma dose plays a role, but also elevated temperatures can have an effect. To determine whether the different temperatures of the irradiation facilities have an influence on the amount of Cu released from the Cu(II)-phthalocyanine complex two sets of samples are heated up to 150 °C for 5 h, of which one set is irradiated with gammas first in the ⁶⁰Co source to receive a dose of 402 kGy. The results are given in Table 2.

Heating seems to have little effect on the stability of the Cu(II)-phthalocyanine complex. The total thermal energy supplied to the molecules is with $E = \hbar \nu^2 = 5.84 \times 10^{-21} J = 0.036 eV$ at 150 °C not
Number of sentences: 60

Fig. 8. Result from the FTIR measurements of Cu(II)-phthalocyanine samples receiving an integrated neutron dose of 2.9 *10^{21} n/m² and 1052 kGy (blue) or a gamma dose 386 kGy (purple) compared to a control (non – irradiated) (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

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Appendix A. Supplementary data

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CRediT authorship contribution statement

J.L.T.M. Moret: Data curation, Formal analysis, Writing - original draft. T.A. Hardens: Formal analysis. O. van Batenburg: Formal analysis, Supervision. H.T. Wolterbeek: Formal analysis. J.R. van Ommen: Funding acquisition, Supervision. A.G. Denkova: Supervision, Writing - review & editing.

Declaration of competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
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