A technical study to economise the amount of zinc used in the production of radiogallium

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Abstract

For the production of radiogallium, the targets were prepared in two forms, namely, electroplated metal and pressed oxide. The target holder was selected from Cu-metal as a circular disk. The experimental yields of ^{66,67,68}Ga produced from both irradiated ^{nat}ZnO and zinc metal targets are given and compared with the estimated yields as well as with the previously reported values. The ZnO target developed in this work appears to be more convenient and economical for local production of short-lived radiogallium, e. g. ^{66}Ga and ^{68}Ga.

Keywords: Radiogallium, ^{nat}Zn and ^{nat}ZnO-targets, electrodeposition, pressing technique, radiogallium production yield

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1. Introduction

Radiogallium (^{66}Ga, ^{67}Ga and ^{68}Ga) are widely used in nuclear medicine, commonly as a trivalent citrate compound for imaging, also as valuable agents in the detection and localization of certain neoplasms and inflammatory lesion [1-3]. The decay data of those gallium radioisotopes are given in Table 1 [4]. They are produced by irradiations of enriched Zn isotopes by proton at various cyclotrons. Furthermore, bombardment of natural Zn with protons of sufficient energies also resulted in the formation of those radionuclides [5-8].

The cyclotron target is generally prepared by electrodeposition of zinc on different metal backings, namely, gold, copper and steel [9-13]. In preliminary work we demonstrated the electroplating of acid-dissolved Zn in aqueous solutions. However, the quality was unacceptable due to the complex compounds formed between the bath residuals and the plated metal. Adjustments in the plating current density, plating bath temperature, and solvent washes of plated Zn did not remedy the poor quality and degradation of material, resulting in bead formation. To complicate matters, the formation of free radicals from the plating solution created a hydrogenation reaction at the cathode which caused it to become embrittled. In other cases, a small amount of an adequate surfactant in the electrodeposition bath results in beneficial quality change of the deposits, namely the homogeneity [14,15].

In the present work we concentrated on the simple Zn-targetry and thereby saving the amount of zinc used. The experimental yields of ^{66,67,68}Ga in irradiated targets were investigated.

2. Experimental

2.1 Targetary

2.1.1 Chemicals and reagents

High purity Zn-metal (99.97%, Koch-light laboratories Ltd., England); potassium bicarbonate, hydrochloric acid, obtained from Sisco research laboratories PVT-LTD Mumbai, India.

2.1.2 Preparation of Zn-targets via electrodeposition

Zinc targets used for this purpose were prepared by electrodeposition using a cell that correlates with an electrodeposition cell given in the literature [9,16]. Firstly, pure metallic zinc (1-2 g) was transformed to zinc sulfate. The electrodeposition of Zn on target holder was carried

out at constant current intensity and continual stirring of the electrolyte. The aqueous solution of zinc sulfate (pH 3.5-4.5) was made in distilled water (50 mL). A copper cathode and a platinum anode were used. The electrolytic cell takes 50 mL of solution. The cell voltage was adjusted to 6 V and the current density to 35 mA/cm^2 [9]. The applied current decreased during the electrodeposition process due to the decrease in the conductivity of the solution as the zinc layer grew. Also the pH of the solution decreased; therefore, it is necessary to neutralize it by adding drops of concentrated solution of ammonia. A layer of 200-300 mg of zinc was obtained.

2.1.3 Preparation of ZnO- target via pressing technique

Pure metallic zinc (0.3 g) was transformed to zinc sulfate that was consequently transformed to zinc carbonate. To avoid the releasing of gas during an irradiation process, zinc carbonate was heated to a temperature above 400 ^{o}C, whereby it was converted to ZnO. The loss in the weight corresponds to the carbon dioxide released.

2.2 Design of the target holder for pressing technique

The substrate selected was Cu-metal due to its good thermal conductivity. It is designed as a circular disk fitted in the irradiation chamber in the cyclotron radioisotope beam line. The thickness of the Cu-disk is 3 mm with a central circular groove of 5 mm radius, and 2.7 mm depth (Fig. 1), in which the ZnO powder was pressed under a stress of 1 ton/cm^2 and then covered by a Havar foil of 10 μm thickness. The covering foil was fixed by a metal ring to avoid contamination during the irradiation resulting from distraction of the pressed powder. The ring had a small cut in its circumference to facilitate the handling.

The irradiated surface area is an important parameter which helps in the enhancement of thermal conductivity for the ZnO-target. The irradiated area and area density in the present work for the ZnO-target were 0.785 cm^2 and 382 mg/cm^2, respectively. By increasing the target area and consequently decreasing the area density, it is possible to get a thin ZnO-target. The thin target is the boundary between the cover foil and the target holder. If the target is thin this would lead to a good heat removal.

2.3 Irradiation and yield measurements

Both the ^{nat}Zn metal and ^{nat}ZnO targets were subjected to increasing beam current tests. The irradiations were performed at the MGC-20E cyclotron in Debrecen, Hungary, at beam current

of 1, 5, 10, 12 and 15 μA of the incident proton energy 16 MeV for one hour. The targets were cooled by water jet from the back and by circulating helium at the front.

2.3.1 Target inspection

The electroplated ^{nat}Zn metal target remained unaffected by irradiations up to 15 μA. The ^{nat}ZnO target as shown in Fig. 1 was irradiated by beam currents starting from 1μA upwards. A visual inspection showed that the target remains stable without cracks or spot burn even when the current was raised to 5 μA. At 10 μA small cracks start to appear. At 12 and 15 μA irradiation, it changed to pale yellow but still maintained its stable state. At currents over 15 μA the target was damaged but it still remained in its groove.

The targets could not be measured directly due to high activity of the Cu activation products. The target material zinc was therefore dissolved in 7 M HCl. A 1-10 % fraction of the dissolved irradiated target was then taken and the activity measured with standard high resolution γ-ray spectrometer consisting of a HPGe detector coupled to a multi-channel analyzer. The peak area analysis was done using the software Gamma-vision (Version 5.1, EG & G ORTEC). The fractions were measured at different and large distances to avoid coincidence losses. Detector efficiencies for different measuring distances (0, 5 10, 20 and 50 cm) were carefully determined using different standard sources of ^{133}Ba, ^{60}Co, ^{137}Cs, ^{22}Na and ^{154,152}Eu.

3. Results and discussion

The present work was mainly directed towards preparation of ZnO-target by the pressing technique and studying the circumstances, such as preparation of the powder target, holder design, irradiation and handling of the target holder after irradiation. Also investigation of experimental yields of ^{66,67,68}Ga in both irradiated Zn metal target and ZnO-target was the aim.

Experimental work showed that about 300 mg of Zn metal was needed in the preparation of the ZnO-target, which is much lower than the amount of 1500 mg, used in the Zn-target preparation.

3.1 Radiogallium yields

The present experimental yields of ^{66,67,68}Ga produced in pressed zinc oxide irradiated with 16 MeV protons of beam currents of 1, 5, 10, 12 and 15 μA are shown in Fig. 2. The activity increases linearly as the beam current increases. The values for ^{68}Ga at 12 and 15 μA are, however,

too high, possibly due to large counting errors in strong samples because of the very weak gamma ray intensity (3%).

Table 2 gives the irradiation conditions and the experimental yields of ^{66,67,68}Ga at EOB for both targets (Zn-target and ZnO-target) as well as the calculated yields given recently by Aslam et al [8], after normalization to natural abundance of zinc. As expected from reports by Szelecsényi et al. [17], and Aslam et al. [8], in this work the measured yields are lower than the calculated ones. However, the experimental yield loss of ^{67}Ga is too high as compared to that of ^{66,68}Ga with respect to the calculated yield. It is known that ^{67}Ga comes from 16 MeV proton activation of ^{67,68}Zn, with their abundances of 4.1 and 18.7 %, respectively. Hence, ^{68}Zn contributes strongly to its formation via the ^{68}Zn(p,2n)^{67}Ga reaction, whose threshold is, however, rather high. Thus a small uncertainty in the proton energy could lead to extreme loss in its activity.

Moreover the ZnO-target showed ^{66,67,68}Ga yields 16 to 20 % lower than those from the Zn-target. This is due to the loss of some proton energy by absorption in the oxygen content and due to the presence of lesser number of Zn atoms in ZnO than in metallic Zn. There is an agreement between the present yields from the ^{nat}Zn-target and the reported yields (Table 3) [1,18,19].

The activities of ^{66,67}Ga in the ZnO-target could be raised by an irradiation for 20 h of 15 μA beam current to about 30 GBq of ^{66}Ga and about 700 MBq of ^{67}Ga. The ^{68}Ga yield reaches saturation (about 25 GBq) after an irradiation of 2 h. Thus sufficient quantities of radiogallium could be produced using the above described target.

4. Conclusion

The two targets, i. e. Zn-metal and ZnO, are prepared by consuming 1.5 and 0.3 g of Zn-metal, respectively. This is due to the low electroplating yield in the case of the metal target. In industrial scale production of ^{67}Ga it is not a problem because the electrolytic bath residue is reprocessed. Furthermore, the target is subjected to irradiation at several hundred μA which leads to large amounts of ^{67}Ga and makes the process economical. At a laboratory scale, however, for the quick production of ^{66}Ga or ^{68}Ga, the simple targetry of ZnO-target leads to quantities of ^{66,68}Ga sufficient for practical applications. The experimental yields of radiogallium in the case of ZnO-target are 16 to 20 % smaller than those from the Zn-target. Nonetheless, the preparation of the ZnO-target is much easier and more economical. Thus the ZnO-target developed in this work could

be conveniently used for local production of short-lived radiogallium (e. g. ^{66}Ga and ^{68}Ga) but it is not a complete alternative to the Zn-metal target used for large scale production of ^{67}Ga.

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Figures Caption

Fig. 1. Cu-holder containing the ZnO-target, Havar foil cover and steel ring.

Fig. 2.Radiogallium yields, produced in the ^{nat}ZnO-target irradiated at 16 MeV proton energy for one hour, against the beam current (a) ^{66}Ga, (b) ^{67}Ga and (c) ^{68}Ga.