PRODUCTION OF ^{67}Ga BY IRRADIATION OF NATURAL ZINC WITH DEUTERONS IN NRCAM CYCLOTRON

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This paper investigates the production of ^{67}Ga by irradiation of natural Zinc with deuterons of different energies from 8 to 15 MeV. The measured thin-target yield for the production of ^{67}Ga reached a maximum of 1.18 μCi/μAh per mg/cm^2 at 9 MeV. This optimum deuteron energy was confirmed by results from the ALICE code to be 9 MeV for thin target. For this optimum energy, the impurities produced by interfering reactions were identified experimentally and found to be mostly short-lived compared to ^{67} Ga. For commercial production of ^{67}Ga, natural Zinc was irradiated with 12 to 15 MeV, deuterons.

1 Introduction

Gallium-67, known as one of the cyclotron radioisotopes used for the detection and localization of tumors and inflammatory lesions, is normally produced by irradiation of enriched Zinc-68 with protons or alpha particles bombardment of natural copper.[1,2]

The production of ^{67}Ga from enriched ^{68}Zn had been planned by utilizing the 30 MeV cyclotron at karadj city. However, due to the high prices of enriched elements, some research was carried out to investigate the production of ^{67}Ga through irradiation of natural Zinc. In this paper the results of irradiation of natural Zinc by 8-15 MeV deuterons is presented.

2 Experimental Procedure

2.1 Target Preparation

Natural Zinc was first electrodeposited on copper backing target. Copper was chosen as the substrate due to its suitable heat conduction. In this process, $ZnCl_2$ was dissolved in demineralized water and few drops of Hydrazine Hydride was added to depolarized the solution. The obtained opaque solution become transparent by adding few drops of 6N HCl. The PH level of the solution was tried to be kept about 3. The prepared solution was then poured in a cell specially designed for the simultaneously electroplating of 4 targets. Within the cell, the Pt anode electrode is placed in the center of the 4 targets. The current used was about 100 mAmps. The elongated ellipsoid target's area plated were about 12.3 cm^2.

Through exploiting the computer program codes , "RANGE" and "TRIM" the range and stopping power of deuterons with different energies for Zinc were determined. At thickness of about 10 micron, e.g. for 15 MeV energy incident deuterons the energy loss will be about 250 KeV. Considering the irradiation of target at 6°, coating of 1μm of Zinc will be sufficient to provide 10 μm effective thickness. Taking into account the area and density of plated Zinc, the amount of coated material is equivalent to 8-9 mg. Based on this calculation all prepared targets had the same amount of platted Zinc and hence the same thickness. According to Faraday's law, the electroplating time for 8-9 mg Zing is about 4 minutes.

2.2 Target Irradiation

The Copper-platted-Zinc targets situated on special shuttles were sent through the cyclotron solid target room by rabbit system. The shuttles geometry were designed such that the targets see the beam at an angle of 6°. Then the target is bombarded with the intensity of 10 μAh deuterons. The beam current was measured by current integrator which is connected to Faraday Cup. Afterwards, the target cooling system is shut off and target is guided through to the hot cell by rabbit system.

2.3 Dissolving the Target

The irradiated platted Zinc was dissolved in a mixture of 10 N HCl and 20 μl H_2O_2. Through dissolving the target some copper was also dissolved in the solution. Then few drops of the solution was diluted for gamma spectroscopy.

2.4 Gamma Spectroscopy

In the gamma spectroscopy the relative method was chosen. Taking into account the measured efficiencies for standard sources at different positions of the same geometry, the activities of radioisotopes for the prepared samples were measured by calculating the peak area in the recorded spectrum and using detector efficiency and branching ratio values. ^{67}Ga is produced by both the (d,n) reaction on ^{66}Zn and the (d,2n) reaction on ^{67}Zn. Only the (d, 2n) reaction on ^{66}Zn leads to ^{66}Ga.

3 Results and Discussion

Cross section of reaction was calculated by using the values of activity, beam current and weight of sample. The variation of measured cross section for ^{66}Ga and ^{67}Ga in terms of deuteron energies is shown in Figure 1.

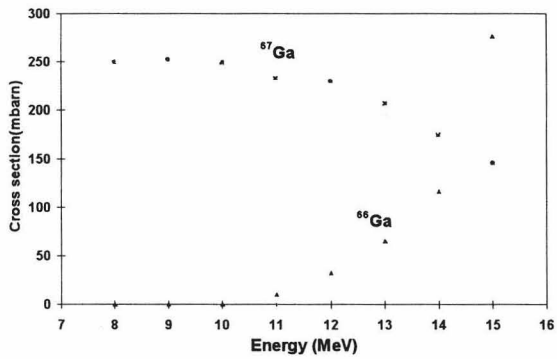

Figure. 1. Excitation function for the production of ^{66}Ga and ^{67}Ga by deuteron bombardment of natural Zinc.

The results for production yield of ^{67}Ga for 8-15 MeV deuteron energy indicated that for thin target the optimum deuteron energy is about 9 MeV.

3.1 Theoretical Calculations of Cross Section

Alice-91 code[5] for calculation of cross section was employed in this work. The interaction of protons and deuterons with natural zinc were investigated. Natural zinc consisted of five isotopes with different abundance as follows: ^{64}Zn (48.6 %), ^{66}Zn (27.9 %), ^{67}Zn (4.1 %), ^{68}Zn (18.8 %) and ^{70}Zn (0.6 %).

The calculation was performed for isotope (A, Z, N) and its recoils until (A-4, Z-4, N) and (A-4, Z, N-4). The calculated results are presented for protons and deuterons in Figures 2 and 3 respectively.

As can be seen from Figure 2 the energy increments of 0.2 MeV used for protons in the energy range of 15-30 MeV.

Although at proton energy of 24 - 25 MeV cross sections for production of ^{67}Ga and ^{66}Ga are maximum and minimum respectively, the percentage of ^{66}Ga in final product is quite higher than USP limitation. Therefore proton irradiation is not recommended for medical applications. In the case of deuteron irradiation as indicated in Figure 3, the optimum deuteron energy for production of ^{67}Ga is about 9 MeV. At this energy contribution of ^{66}Ga is negligible.

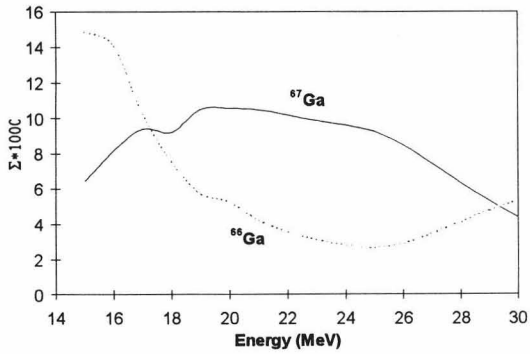

Figure 2. Cross section of ^{66}Ga and ^{67}Ga by proton bombardment of natural Zinc (ALICE code)

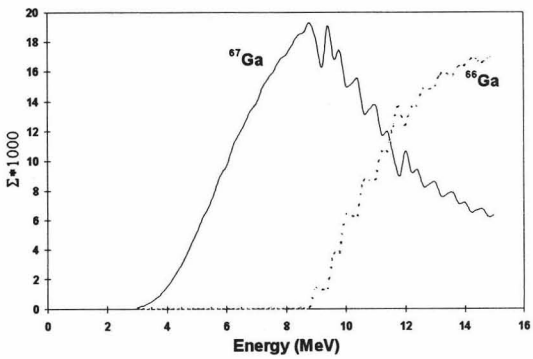

Figure. 3. Cross section of ^{66}Ga and ^{67}Ga by deuteron bombardment of natural zinc (ALICE code)

3.2 The ^{67}Ga Yield in Thick Targets & Commercial Productions

Taking into accounts the measured cross-sections for 8-15 MeV deuterons, and energy loss of deuteron for different thicknesses, one can calculate the production yield for thick targets[3]. The obtained values are presented in Table 1.

Table 1. Thick targets yields

Energy (MeV)	Yield of ^{67}Ga μCi/μAh	Yield of ^{66}Ga μCi/μAh
8	37.57	0
9	65.13	0
10	98.07	0
11	135.37	24.06
12	177.07	38.66
13	222.32	122.85
14	267.42	301.35
15	310.23	737.5

As can be seen from Table 1 our thick target yield values are lower than those reported by Steyn and Meyer [4]. At 12 MeV deuteron energy, for example they obtained 218.9 μ Ci/μAh, while our calculation value is 177.07 μCi/μAh. This could be due to non uniformity of electroplated target and error in beam current reading.

For commercial production, the thick targets are prepared by electroplating of Zn with thickness of 80-100 μm and the targets are bombarded for 6 h at an average beam current of 100 μA.

The bombarding energy is 12 MeV . It is found that the yield, during routine production usually varies between about 70-90 percent of the expected value, due to diffusion out of ^{67}Ga.

About 36h after the end of bombardment the Zn layer is dissolved and chemistry process is started. The chemistry procedure was the same as used by winkel[6] for $GaCl_3$ production. The $GaCl_3$ must be converted to Ga-Citrate, to be suitable for injection. Therefore the method of Neirinckx and Van der Merwe[7] is used which yields a carries-free and iron-free product.

In reference time usually after one half-life of ^{67}Ga from end of irradiation the ^{66}Ga content has dropped to limit of USP.

Since half-lives of ^{68}Ga, ^{65}Ga and ^{64}Ga (68.3 m, 15.2 m and 2.6 m, respectively) are very short as compared to ^{66}Ga and ^{67}Ga, they do not pose any difficulties in the presenting of ^{67}Ga process.

After bombardment ,replating and chemistry the final solution contains only ^{66}Ga as a radioisotopic impurity. ^{65}Zn, which normally is formed from copper impurities, was not detectable even after a 600 μAh run.

The resultant Ga-Citrate still is not suitable to be used for patients. Quality control and sterilization should be carried out. The quality control is consisted of several analytical techniques such as gamma spectroscopy, for radionuclide impurity determination, chromatography for measuring the percentage of citrate and polarography to identify the impurities present in solution such as Ga, Cu and Zn (see Figure 4). After dispensing of Ga-Citrate into vials, the sterilization will be done in autoclave for 30 minutes at 120°C.

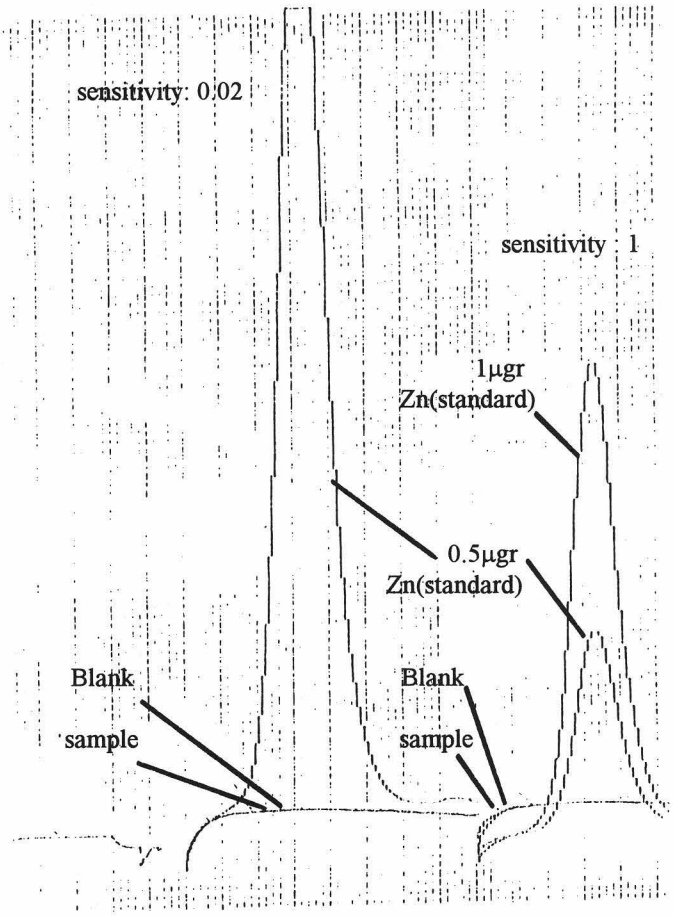

Figure. 4. The polarography results of Zinc

Acknowledgments

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