Surface Analysis of Nb Influenced by Liquid Ga at Estimated Temperature of ⁶⁸Ge Production Environment

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1. Introduction

⁶⁸Ga has attracted great attention in medical research such as position emission tomography (PET) because it decays mainly by positron emission and has short halflife (~68 min) [1]. Therefore, the production of ⁶⁸Ge, long-lived (half-life: ~288 days) parent radioisotope of ⁶⁸Ga, becomes one of the most important issue. An encapsulation should be required when the natural Ga target is used for the production of ${\rm ^{68}Ge}$ by proton irradiation, because the natural Ga has relatively low melting point (~30°C) and therefore is easily liquefied by the temperature increase resulted from the bombardment of energetic protons. According to Noriter et al.'s report, they estimated the maximum temperature as high as 148°C by simulation with the irradiation condition of 29.5 MeV in proton energy and 250 μ A in beam current [2]. In this research, we carried out the heating experiment of Nb, which is the best candidate material for the encapsulation, in the liquid Ga at the estimated temperature of ⁶⁸Ge production environment, and the effect of liquid Ga on the morphology and chemical composition of Nb was investigated.

2. Methods and Results

2.1 Material Preparation

Ga (Strategic Metal Investments, 99.9%) was melted by heating at temperature slightly above its melting point (~40°C), and then 2 mL of Ga was transferred to each vial (Thermo Scientific Reacti-VialTM #13223). Nb (Goodfellow, thickness: 0.1 mm, 99.9%) foils were cut into size of 5 mm x 25 mm and dipped into liquid Ga in vials.

2.2 Heating Experiment

After the preparation of vials containing liquid Ga and a piece of Nb foil, they were put on the heating device (Reacti-Therm Heating/Stirring Module, Thermo Scientific). Heating temperature was 120°C and heating time was varied from 1 day to 14 days. After the heating experiment, Nb foils were took out, and then carefully washed several times in order to remove the residual Ga on Nb foil surfaces.

2.3 Surface Morphology

Surfaces of Nb foils were observed using field emission scanning electron microscope (FESEM, Hitachi S-4800). The amount of the adhesion material monotonically increased as the heating time increased (Fig. 1). When the heating time increased up to 3 days, the adhesion material in the form of either nanoparticles or aggregates was hardly found on the Nb foil surface. After the heating more than 5 days, some nanoparticles were found on the Nb foil surface, and then the number of nanoparticles were increased as the heating time increased. Some aggregates were also found when the heating time increased to 14 days (see arrowed area in Fig. 1f).



Fig. 1. FESEM images of a pristine Nb foil (a) and Nb foils heated in the liquid Ga for 1 day (b), 3 (c), 5 (d), 9 (e), and 14 days (f).

2.4 EDS analysis

In order to identify materials which adhered on the metal foil surfaces, they were analyzed by using energy dispersive X-ray spectroscopy (EDS). Nb foil exhibited the peaks of an element that composes pristine foil, the peaks of Ga whose intensities were much smaller than that of the Nb peaks, and a small oxygen (O) peak which possibly came from the Nb foil surface oxidized naturally or by the heating.



Fig. 2. EDS peak of Nb foil heated in the liquid Ga.

2.5 XRD and XPS analysis

X-ray diffraction (XRD) peaks were centered at 38.44° , 55.59° , and 69.66° , corresponding to (110), (200), and (211) planes of pure Nb, respectively (Fig. 3). However, no XRD peaks of Ga were found. This result indicated that the heating in the liquid Ga at 120° C for 2 weeks did not affect the Nb foil and the amount of Ga which adhered to the Nb foil was too small to be detected by XRD.



Fig. 3. XRD peaks of Nb foil heated in the liquid Ga.

The chemical composition change of the Nb foil surface was investigated using X-ray photoelectron spectroscopy (XPS). XPS survey spectra of Nb foils before and after heating in the liquid Ga are shown in Fig. 4. XPS survery spectra of pristine Nb exhibited characteristic peaks of Nb, C 1s peak which possibly came from the surface contaminant, and O peaks which probably resulted from the natural oxidation. However, only small 3d peaks of Nb remained among characteristic peaks of Nb, and Ga peaks appeared after the heating in the liquid Ga.



Fig. 4. XPS survey spectra of Nb foils before and after the heating in the liquid Ga.

3. Conclusions

The effect of liquid Ga on Nb foils at the temperature higher than the melting point of Ga was investigated. After the heating of Nb foils in the liquid Ga at 120°C, only a little amount of Ga remained on the Nb foil surfaces after several times of washing. In the view point of the separation of Ga from the Nb foils, Nb showed good property. In addition, Ga was just physically attached on the Nb Surfaces without any chemical bonding, and thus the Nb foil itself was not affected by the liquid Ga.

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