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Target preparation of RbCl on a copper substrate by sedimentation method for the cyclotron production of no-carrier-added ^{85}Sr for endotherapy

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Abstract. ^{85}Sr was produced via the $^{85}Rb(p,n)^{85}Sr$ reaction. Rubidium chloride deposition on copper substrate was carried out via the sedimentation method in order to produce strontium-85. Optimum conditions were achieved as a result of several repeated experiments with different amount of ethyl cellulose (EC) and acetone. 520 mg of RbCl, 208 mg of EC 4 mL of acetone were used to prepare a layer of enriched rubidium chloride of 11.69 cm^2 area and 62.2 mg/cm^2 thickness. Target quality control was done by a SEM photomicrograph and a thermal shock test. The deposited target was irradiated at a 20 μA current and a 15 MeV proton beam for 30 min. No degradation was observed. The RbCl surface following bombardment was white, except the central area that was light brown and also without any crack or peeling off.

Key words: rubidium chloride • target • sedimentation • radionuclide

Introduction

^{85}Sr is an important radionuclide having a 64.8 d half-life, which has plentiful importance in nuclear medicine fields such as endotherapy. Strontium-85 was used as a bone pain palliation radiopharmacy. It plays a useful role in bone imaging of metastatic breast cancer [2, 3]. Also skeletal survey of bone metastatic tumors was carried out with ^{18}F and ^{85}Sr using scintillation camera and whole-body scanner [12].

^{85}Sr production via the $^{85}Rb(p,n)^{85}Sr$ reaction is regarded as the most interest desirable reaction for its high yield and low impurity production. Also the $^{nat}Rb(p,x)^{85}Sr$ reaction is used for ^{85}Sr production. To prepare rubidium target, [^{85}Rb] RbCl or $^{85}RbCO_3$ were employed as the target material. Various rubidium targetry were designed and manufactured by the researchers. Some of these were considered and compared to find economic, simple and practical method to design an endurable target in more intense beam. Thin RbCl targets (100 mg/cm^2) were prepared by pressing uniformly distributed target material in a special windowed holder made from aluminum [4]. For preparing sample of rubidium chloride in the Kandil *et al.* work, the powder was pressed on an aluminum foil of 10 μm thickness and 13 mm diameter under a pressure of 6 ton/cm^2 for 10–15 min. Samples of thickness 18–22 mg/cm^2 were obtained. Each sample was covered with a 10 μm thick Al foils [5, 6]. In another targetry method, encapsulated RbCl salt targets are used for the production strontium

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radioisotope. The capsule halves are machined either from 316 stainless steel or from Inconel 625 with a window thickness of 0.3 mm. The salt pucks are prepared by casting and then sealed inside the capsule under vacuum by means of electron beam welding [10]. To obtain thin targets for irradiation, layers of fine powder RbCl were prepared via sedimentation technique, too. The powder was sedimented on an aluminum backing (15 mm in dia., 0.1 mm thick) and covered with a 0.01 mm thick Al foil [11]. Thin samples were prepared by means of a special sedimentation method. A thin copper foil (25 μm) was used as the backing [7].

The aim of this work was to prepare a target material by coating of RbCl via sedimentation method on a pure copper substrate that has sufficient stability at high-power beam bombardment.

Materials and methods

High purity (> 99%) RbCl powder (Aldrich Chemical Company) was used as a target material to prepare rubidium target.

According to codes ALICE/ASH [1], TALYS-1.0 [8, 9] and the experimental data that have been reported by Levkovskij [9] and Kastleiner [7], the proton entrance energy should be less than 13 MeV to get full benefit of excitation function and to avoid the formation of radionuclides impurities. Physical thickness of rubidium chloride layer is chosen as for a given beam/target angle geometry to provide a light-particle exit energy of about 3 MeV. According to SRIM code [17], the thickness has to be 1472 μm for 90° geometry. To minimize the thickness of the rubidium chloride layer and to increase heat transfer, a 6° geometry is preferred, in which a 147.2 μm RbCl layer is recommended [15].

Target preparation

To prepare the target, a particular device made of Teflon (PTFE) was constructed. It consists of two plates of 19 × 10 cm^2 surface and 3 cm height. The upper plate contains an elliptical window of 11.69 cm^2 (same as the copper substrate). The copper substrate was placed between these two plates, the upper part is fitted on it with six supporting pins and it is sealed by an *O*-ring fitted-window [14, 16]. The window geometrical shape determines the actual target coating area. To achieve the desired thickness (147.2), 480 mg RbCl is required. But in order to avoid proton beam interaction with Cu backing and ^{65}Zn production, rubidium chloride deposit should be 158 μm. RbCl of 520 mg is needed to achieve this thickness.

A RbCl thick layer was deposited on an elliptical copper substrate by means of sedimentation method. In this method it was expected that suspension of very fine rubidium chloride powder in water-free acetone would be obtained by mixing and stirring. But the rubidium chloride powder was settled immediately after the stir ending. It seems rubidium chloride grains are too large and too heavy to be suspended. To remedy the problem, the purchased rubidium chloride powder was grinded. 10 ml of acetone was added to 520 mg of the grinded rubidium chloride and then the mixture was stirred for several minutes. The prepared mixture was dried at 50°C using an oven. After that, 208 mg of EC powder was added to the heated grinded powder and the mixture was grinded again. Finally, 4 ml of acetone was added to the EC and rubidium chloride fine-grinded mixture and the prepared suspension solution was immediately loaded into the cylinder of the upper disk. A plate made of Teflon that includes a small hole covered the window. The solution evaporated slowly through the hole at room temperature after about 24 h. The system window must be covered to prevent the solvent (acetone) from quick evaporation and poor adhesion on the target material on the copper substrate.

An unclean surface of the Cu backing may cause blistering, cracking, gas pits and peeling off of the RbCl layer. The substrate was cleaned with a sand-paper (grade 1000) and washed with water, then a mixture of alkali cleaning powders was used to remove oil contaminators. Finally, the surface was cleaned with acetone.

This refined procedure is a result of several repeated experiments with different amounts of EC and acetone. Thermal shock and annealing were performed to study adhesion of the samples. The coated targets with RbCl were examined in morphology by a scanning electron microscopy (SEM) technique.

Irradiation

The coated natural RbCl was introduced into a target holder and bombarded with 15 MeV protons at a current of 20 μA for 30 min. The Agricultural, Medical and Industrial Research School (AMIRS) employs a Cyclone-30 (IBA, Belgium). To protect the target material from reaching excessively high temperatures, a jet of cooling water flows during the irradiation across the back of copper substrate in direct contact with it, so the heat is removed efficiently from the backing. In order to improve the thermal conductivity, the copper substrate includes some grooves in the back. No direct cooling is used over the front of the deposited target.

Results and discussion

Target preparation

With the purpose of optimizing rubidium chloride coating, the experimental situations were examined as follows:

Adhesion of agent amount

Adhesion is an important factor for adhering and coating among physical properties of the samples. Insufficient amount of EC causes poor adhesiveness of deposited rubidium chloride layer. On the other hand, their over abundance reduces thermal conductivity of the target. To optimize the quantity for maximum adhesion and thermostability of RbCl layer, the samples were examined with different quantities of EC with respect to RbCl. According to the target quality control, such as homogeneity, morphology, visual appearance of the

Table 1. Influence of EC amount with 4 ml acetone on 520 mg of RbCl

W (EC) (mg)	*W* (EC)/*W* (RbCl) (%)	t_d [a] (mg·cm^{-2})	Adhesion	Comments
78	15	51.1	unfavorable	rough, porous
130	25	55.6	unfavorable	rough, porous
156	30	57.8	tolerable	rough
182	35	60.0	tolerable	reflective, smooth
208	40	62.2	excellent	reflective, smooth
234	45	64.4	excellent	reflective, smooth
260	50	66.7	excellent	smooth

[a] t_d – calculated thickness of the deposit on Cu backing.

Table 2. Influence of amount of acetone on 520 mg of RbCl

W (EC)/*W* (RbCl) (%)	Acetone (mL)	Adhesion
	3	tolerable
45	4	excellent
	6	excellent
	3	tolerable
40	4	excellent
	6	excellent
	3	unfavorable
35	4	tolerable
	6	tolerable
	3	unfavorable
30	4	tolerable
	6	tolerable

rubidium chloride layer and thermal shock test, the optimum amount of EC was about 40–45% of rubidium chloride (Table 1).

Solvent amount

Solvent amount affects adhesion of the target, too. Fewer amount of solvent results in fast evaporating, hence undesired adhesion. In order to achieve the required tenacity, a different amount of acetone was applied to prepare the suspension (Table 2).

Target quality control

Homogeneity of the RbCl layer, which may affect the production rate of ^{85}Sr, was determined by standard deviation of the layer thickness measured at several spots by a micrometer, while the morphology by SEM (see Fig. 1 for the best coating).

Results of the thermal shock tests are given in Table 3. No crack formation or peeling off was observed with the RbCl layers at 250°C for 1 h, indicating a good adhesion for the purpose. The results also indicate that the target can resist 250°C, without any crack or peeling off.

Irradiation

The thick deposit of ^{85}RbCl was bombarded in this study. No target material loss was observed at a current beam of up to 20 μA.

Production of strontium-85 with this method gives the opportunity to apply a high beam power of about 300 W (20 μA) provided that the target material can withstand this current beam and demonstrate stability. This also makes the use of target design that is developed to spread the beam over a greater surface (about 11.69 cm^2) than powder targets (about 1–1.3 cm^2), and using a slanted target (6°) that allows to apply a thinner layer of target material, while providing the same effective target thickness, both of which improve thermal conductivity into the copper backing and allowing to use a high beam current.

In addition to the mentioned method, the target preparation was carried out by applying cellulose nitrate instead of EC, as reported by Rösch *et al.* [13]. A number of tests was done with different cellulose nitrate quantity. Although, the cellulose nitrate amount was increased twice in relation to rubidium chloride, the desired tenacity of target that has sufficient stability at high-power beam bombardments was not obtained. Due to the increased thickness and target surface in

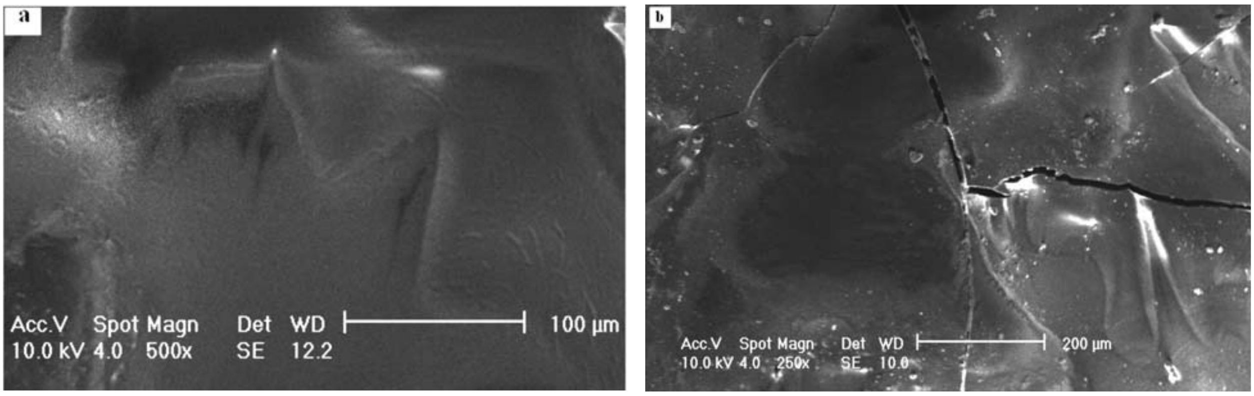

Fig. 1. SEM of rubidium chloride deposit on the Cu backing: a – for 520 mg of RbCl, 208 mg of EC, and 4 mL of acetone suspensions (no crack was observed); b – respectively for 480 mg, 170 mg and 6 mL (some cracks on the target were observed).

Table 3. Thermal shock test[a]

W(EC)/W(RbCl) (%)	Adhesion[b]	150°C	200°C	250°C	300°C	350°C
45	excellent	stable, white	stable	stable, tan	stable, brown	unstable
40	excellent	stable, white	stable	stable, tan	stable, brown	unstable
35	excellent	stable, white	stable, tan	unstable	unstable	unstable, dander
30	tolerable	stable, white	unstable	unstable	unstable, dander	unstable, dander

[a] The heating of the target for 60 min followed by submersion of the hot target in cold water (8°C).
[b] Adhesion at room temperature.

this work, suitable adhesion by using cellulose nitrate as a replacement for EC was not achieved.

Conclusion

In summary, ^{85}Sr was produced by irradiation of a RbCl thick deposit target that was prepared by means of the sedimentation method and optimum parameters were obtained. The target was irradiated up to a 20 μA current with 15 MeV and no degradation was observed. Improvement of cooling system that contains a jet of water-cooling and a circulating flow of chilled helium in front of the target would improve the thermal conductivity, and so this is expected to allow using a higher beam current up to 40 μA.

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