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Electrochemical co-deposition of Ni-Gd₂O₃ for composite thin targets preparation: production of ¹⁵⁵Tb as a case study

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Abstract

In the last years, ¹⁵⁵Tb has attracted enormous interest due to its potential role in theranostics in nuclear medicine. To estimate its production yield, the aim of this study was to develop a method to prepare thin Gd-enriched-containing targets aimed at the ¹⁵⁵Gd(d, 2n)¹⁵⁵Tb nuclear cross section measurement. To this end, the electrochemical co-deposition method has been chosen to manufacture Ni-Gd₂O₃ composite targets. Several process parameters that have an impact on the deposit quality, have been investigated to increase the incorporation of Gd mass (up to 3 mg). To validate the concept, seven targets made by natural Gd were irradiated with deuteron beams at the GIP ARRONAX facility cyclotron, with an energy range ranging from 8 MeV to 30 MeV to extract the cross section values by using the stacked-foils method. Results obtained turned out to have great consistency with existing published data thus validating the proposed method. Therefore, an alternative target manufacturing concept aimed at cross section measurement is presented in this work.

Key works: electrochemical co-deposition, gadolinium target, cross section measurement, terbium production, stacked-foil technique

1. Introduction

Theranostics, a new medical approach combining therapy and diagnostic, has become one of the core keywords in recent years in nuclear medicine. Applications of radionuclides have evolved with the development of theranostics, among which the application of Terbium radioisotopes has been mentioned in several articles (Cavaier et al., 2017; Cleeren, 2021; M  ller et al., 2018, 2012). As an emitter of Auger electrons and gamma rays, ¹⁵⁵Tb (half-life: 5.32 d) can be applied in Auger therapy as well as single photon emission tomography (SPECT) diagnosis (M  ller et al., 2014; Nica, 2019; Webster et al., 2019). However, applications of ¹⁵⁵Tb are in short supply due to its limited availability. Nowadays, it is mainly produced by high-energy spallation reactions followed by mass separation, which requires rare equipment and high cost (Naskar and Lahiri, 2021). Therefore other reaction routes, involving gadolinium targets coupled to light-charged particle beams, are being taken into account to get an increase of ¹⁵⁵Tb supply (Duchemin et al., 2016; Formento-Cavaier et al., 2020; Naskar and Lahiri, 2021; T  rk  nyi et al., 2014). Enriched Gadolinium targets make it possible through the following reaction routes involving either proton or deuteron beams: ¹⁵⁵Gd(p, n)¹⁵⁵Tb (threshold energy: 1.61 MeV) or ¹⁵⁵Gd(d, 2n)¹⁵⁵Tb (threshold energy: 3.88 MeV) (Koning et al., 2019). To evaluate the optimal accelerated beam energy ranges, produced contaminants as well as the production yield, reaction cross sections must be determined, first. To this end, high quality and thin Gd-enriched targets are required.

The electrodeposition method to prepare thin targets is often used, as it can be performed with a low cost and simple installation system. In case of Gd target preparation, a critical issue is the redox potential of Gd³⁺/Gd which is very low ($E = -2.28$ V/SHE) in acid solution (Lide,

2004). A strong competition with water decomposition is therefore occurring, and good adherence of Gd deposits present in aqueous solution are unattainable under the hydrogen evolution reaction (HER). To overcome such a problem caused, several Gd deposits tests were carried out in organic mediums (Gucik et al., 2020; Kovács et al., 2016; Small et al., 2016), however, they resulted either with too small area ($<1.1 \text{ cm}^2$) or too fine ($<1 \text{ }\mu\text{m}$). In addition, these experiments have strict restrictions on moisture content, which makes them complicated to master. Molten salt mediums were also studied (Caravaca et al., 2007), but this experiment had also many constraints (high temperature, pure argon environment ...). It is noteworthy to recall that other methods for thin target preparation, such as High energy Vibrational Powders Plating method (HIVIPP) (Skliarova et al., 2020) or Magnetic Sputtering (MS) (Skliarova et al., 2019) are available. However, the former method needs a set-up under vacuum and takes considerable time, and the latter requires control of air pressure and temperature.

In this work, an alternative way called the co-deposition method to obtain Gd containing targets has been therefore explored. This method, which emerged in the 1920s and was improved in the 1960s, has been extensively studied to develop composite materials (Lee et al., 2007; Low et al., 2006; Parida et al., 2011). The basic principle of this method is to mechanically transport and physically embed target particles mixed in a bath to the growing deposited layer (Walsh and Ponce de Leon, 2014). Using this method, Gd atoms are trapped in a metal deposit and avoid most of the difficulties associated with direct Gd electrodeposition. The main advantages of this method are the simple experimental conditions and the target manufacturing duration which is short (30-60 min).

Main goals of this work are to obtain $\text{Ni-}^{155}\text{Gd}_2\text{O}_3$ composite targets via the co-deposition method and to get measurement of the cross sections by using these targets. Gd_2O_3 has been selected in this experiment because the enriched ^{155}Gd isotope is easily available in this form. Nickel has been selected as the electroplating matrix metal because the experimental conditions are well known and it is possible to perform it in basic conditions, in which Gd_2O_3 remains stable. In addition, due to its insolubility in alkaline solution, the Gd_2O_3 powder can be easily recycled, which greatly improves the material utilization rate which is of particular importance when isotope-enriched materials are used. In this work, proof of concept experiments have been performed using natural gadolinium, due to the high cost of enriched Gd. Influences of main process parameters (i.e., applied potential, concentration of Gd_2O_3 and stirring speed) on the deposit quality have been studied to validate the optimal experimental procedure. To carry out the experimental measurement, seven of the obtained targets were then irradiated with deuteron beams at the GIP ARRONAX cyclotron, in Nantes (Haddad et al., 2008) in an energy range ranging from 8 MeV up to 30 MeV. The production cross sections have thus been measured and compared with the experimental data available in the literature or world databases (e.g. EXFOR).

2. Materials and methods

2.1 Electrochemical co-deposition study and target preparation

To obtain the $\text{Ni-Gd}_2\text{O}_3$ composite deposits, the following electrolyte was prepared: 0.5 mol/L $(\text{NH}_4)_2\text{SO}_4$ + 0.5 mol/L NH_4Cl + 0.14 mol/L $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ + 1 mol/L NH_3 . These compositions and concentration settings referred to the work of Skitał et al. (2019). The final pH of the electrolyte was fixed at 9.8, adjusted by 5 mol/L of NaOH. Insoluble Gd_2O_3 under powder form (less than 230 mesh size) in basic solution was then added to the solution with a

mass varying from 0.1 g to 6.5 g. All chemical products mentioned were provided by Sigma Aldrich. The stirring magnet was used to ensure that the Gd_2O_3 particles are distributed as evenly as possible in the solution. The stirring speed was set at 100, 300, 400, 600 and 800 rpm.

The co-deposition was carried out in a homemade Teflon cylinder cell containing 35 mL of the electrolyte solution (Fig. 1). A hole with a diameter of 10 mm was on the side of the cylinder to place the working electrode. A sealing film and a stainless steel plate were used to ensure the tightness of the cell and electric connection. For the co-deposition study, the working electrode was a $2.5\text{ cm} \times 2.5\text{ cm}$ gold foil with a thickness of $50\text{ }\mu\text{m}$ provided by Goodfellow (99.95 % purity). The auxiliary electrode was made of a platinum rod having a diameter of 1 mm. A reference electrode Ag/AgCl/sat KCl was also used during the process. The distance between the working electrode and the auxiliary electrode was set at $1.0 \pm 0.1\text{ cm}$. Experiments of chronopotentiometry and voltammetry were carried out by PGP201 Voltalab Potentiostat/galvanostat. The scanning potential was from -0.1 V/NHE to -1.5 V/NHE for 5 cycles, the scanning speed was 0.05 V/s , and the solution stirring speed was 800 rpm. The applied potential of co-deposition varied from -1.30 V/NHE to -1.04 V/NHE according to voltammetry results. Each experiment was carried out at 30°C and lasted for 1 hour.

Surface morphology and Gd_2O_3 distribution were observed by JEOL JSM 7100F scanning electron microscope (SEM) with a magnification of $100\times$, $1000\times$ and $5000\times$ at 15 kV acceleration voltage. Energy-dispersive X-ray spectroscopy (EDX) coupled to the SEM was also used for investigating the surface chemical composition. The dimension of the region analysis was about $1.1\text{ mm} \times 0.8\text{ mm}$ at a magnification of $100\times$. The measuring time was 60 s.

Targets, once characterized, were irradiated for cross section measurements, and after that, dissolved in 10 mL of 12 mol/L HCl for 4 hrs at room temperature. 5 mL of the dissolved solution was taken for the measurement of activity with our High Purity Germanium (HPGe) detector, another 5 mL solution was used for chemical composite analysis with our Thermo Scientific iCAP 6000 Series Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES) to determine precisely the Gd atomic contents.

2.2 Targets irradiations

The ARRONAX cyclotron can accelerate both positive ions (H^+ , He^{++}) and negative ions (H^- , D^-). Protons and alpha particles can be accelerated up to 70 MeV whereas deuteron beams can be accelerated only up to 34 MeV. The irradiation process was carried out at the ARRONAX cyclotron in a typical configuration for stacked-foil experiments (Blessing et al., 1995). In these experiments, monitor foils were used to determine the beam flux and degrader foils to adapt the energy of the beam impinging consecutive targets. Titanium foils and nickel foils with a thickness of $15\text{ }\mu\text{m}$ and $20\text{ }\mu\text{m}$ respectively were used as monitors; Aluminum foils with a thickness of $200\text{--}800\text{ }\mu\text{m}$ were used as beam energy degraders. All foils mentioned were provided by Goodfellow (99.99 % purity). Foils were placed in air 6 cm downstream the beam output window, made of $50\text{ }\mu\text{m}$ thick Kapton foil. Seven Ni- Gd_2O_3 composite targets were irradiated in five different experiments (target n°1 and n°2 were irradiated together, target n°4 and n°7 were irradiated together) for 30 min or 60 min by deuteron with an energy range from 8 MeV to 30 MeV. The schematic diagram of one stack

for one particular experiment is illustrated in Fig. 2 as an example. The beam parameters used for the irradiation of each target are presented in Table 1. Mean beam intensity was calculated according to the flux measured by monitors. For each experiment, energy in the middle of the thickness of each foil was determined using SRIM software (Ziegler et al., 2010) and taking into account the beam energy uncertainty (± 0.25 MeV) and energy straggling. Since Gd₂O₃ particles were uniformly distributed in targets (Fig. 5 (e)), in the SRIM simulation, the target was regarded as to be composed of Gd, O, and Ni three components. The number of atoms of each component and the density of the target were determined according to the ICP-AES analysis results.

2.3 Cross section calculation

For a thin target with uniformly distributed Gd₂O₃, the production cross section for the ^{nat}Gd(d, 2n)¹⁵⁵Tb reaction route at energy E , $\sigma(E)$ (mb), can be calculated according to Eq. (1):

$$\sigma(E) = \frac{Act(E) \cdot A}{I \cdot N_A \cdot \rho \cdot \delta x \cdot (1 - e^{-\lambda t})} \quad (1)$$

Where $Act(E)$ (Bq) is the activity of ¹⁵⁵Tb nuclei produced at energy E , A (g.mol⁻¹) is the molar mass of the target nuclei, I (s⁻¹) is the number of projectile per second hitting the target, N_A is the Avogadro number (at. mol⁻¹), ρ is the target material bulk density (g.cm⁻³), δx is the thickness of the target (cm), λ (s⁻¹) is the decay constant of ¹⁵⁵Tb and finally, t is the irradiation time. The total Gd atoms in the deposit was calculated according to ICP-AES results obtained from each irradiated target after its dissolution.

For titanium monitors and nickel monitors, using Eq. (1), the number of projectile per second I' of titanium foils or nickel foils were determined by using the reference cross section $\sigma'(E')$ of the reaction ^{nat}Ti(d, x)⁴⁸V (Hermanne et al., 2018) or the reaction ^{nat}Ni(d, x)^{58(m)}Co (Amjed et al., 2013) respectively. The monitor atomic number N' was determined by its mass, its atomic mass and the Avogadro constant. The average value of monitors' number of projectile per second I'' was then calculated. Since monitor foils have the same area as targets and are located just behind the target foils, I'' is considered equal to I . It can be deduced Eq. (2).

$$\sigma(E) = \frac{Act(E) \cdot A}{I'' \cdot N_A \cdot \rho \cdot \delta x \cdot (1 - e^{-\lambda t})} \quad (2)$$

Since all parameters in Eq. (2) are independent, estimation of cross section uncertainties is based on combined standard uncertainty estimating in ISO guide (Iso and OIML, 1995), as the positive square root of the summation of squares of each parameter (Eq.(3)).

$$\frac{\Delta\sigma(E)}{\sigma(E)} = \sqrt{\left(\frac{\Delta Act(E)}{Act(E)}\right)^2 + \left(\frac{\Delta I''(E)}{I''(E)}\right)^2 + \left(\frac{\Delta M}{M}\right)^2} \quad (3)$$

The half-life and main γ rays (Nudat 2.8 database, 2021) of ¹⁵⁵Tb, ⁴⁸V and ^{58(m)}Co used in this work are presented in Table 2. As indicated in Table 2, the gamma ray energy of ¹⁵⁵Tb that we have chosen is 180.08 keV, which is very close to that of ^{198m}Au ($T_{1/2} = 2.272$ d, $E = 180.31$ keV, $I_\gamma = 0.04\%$) produced by the gold substrate. This is the reason why the targets were dissolved after irradiation to separate the deposit and the gold substrate. For each target, 5 mL

of dissolved solution sample was measured using an HPGe detector 20 h after EOB. Solid titanium monitor foils were measured by the same detector 19 days after EOB (Nigron et al., 2021) to allow the full decay of ^{48}Sc ($T_{1/2} = 43.67$ h, $E = 983.526$ keV, $I_{\gamma} = 100.1\%$) and avoid interferences. Similarly, solid nickel monitor foils were measured 2 months after EOB (Hermanne et al., 2013) to avoid interferences of ^{56}Ni ($T_{1/2} = 6.075$ d, $E = 811.85$ keV, $I_{\gamma} = 86.0\%$). All targets and foils were counted with an HPGe detector for 24 hrs. The distance between the sample and the detector was 0 cm for liquid samples (contact) and 19 cm for solid samples. The dead time for liquid samples was between 5 % to 1 % and close to 0 % for solid samples. The Fitzpeak software (Fitzgerald et al., 2009) had been used to determine the EOB activity of ^{155}Tb (produced in composite targets), ^{48}V (produced in titanium monitors) and $^{58(m)}\text{Co}$ (produced in nickel monitors). Energy and efficiency calibrations were carried out with a multi-gamma liquid source and a ^{152}Eu sealed source in the same geometry as the irradiated liquid samples and solid samples respectively.

3. Results and discussion

3.1 Optimization of co-deposition parameters

The main purpose of this work was to obtain a composite target with a high Gd atoms content and the best efficiency thanks to Gd initial amount. From this point of view, the effect of different parameters was studied including applied potential, initial Gd_2O_3 loading quantity and stirring speed.

3.1.1. Applied potential effects

To choose the potential ranges for a good Ni deposit, voltammetry curves with 5 cycles were performed. All curves of each cycle have the same behavior and Fig. 3 shows the voltammetry curves associated with the fifth cycle of solutions of Ni at 0.14 mol/L and pH equal to 9.8 with 0.8 g or 6.5 g Gd_2O_3 and without Gd_2O_3 loading. The solution was stirred by a magnet rod at 300 rpm. An oxidation peak can be observed between -0.4 V/NHE and -0.5 V/NHE, which may be explained by the formation of $\text{Ni}(\text{OH})_2$ on the surface of the electrode according to Milosev et al (2007). In the range -1.1 V/NHE to -1.3 V/NHE, the current density is between -10 mA/cm² and -25 mA/cm². The reduction process has also a peak near -1.1 V/NHE, which is probably due to the reduction from $\text{Ni}(\text{OH})_2$ to Ni (Milošev and Kosec, 2007). When the voltage is more negative than -1.3 V/NHE, small fluctuations appear on the curve, which may explain the formation of dendritic structures on the deposits in this voltage range (Fig. 5).

Applied potential effects on average current density, deposit thickness, deposited Gd mass and Gd content fraction are summarized in Table 3. Each of the co-deposition processes lasted 1 hour, with a stirring speed of 300 rpm and a loading of Gd_2O_3 of 6.5 g. Table 3 shows that when the applied potential is low enough (-1.04 V/NHE), the Gd content in the composite is the highest (8.5 ± 0.6 at.%). However, due to the lower current density (-1.9 ± 0.4 mA/cm²), the thickness of the deposit is limited (3.1 ± 0.4 μm), as a result, the total Gd content is limited (only 1.5 mg of Gd were trapped in the deposit in these conditions). As the applied potential becomes more and more negative, the deposit kinetics increases, although the atomic ratio of Gd in the deposit remains relatively low (around 4 %). In this case, the total Gd quantity trapped in the deposit increases due to the growing thickness of the deposit.

EDX analysis coupled with SEM were also performed to see deposits morphology. Two kinds of particles with different brightness appear on the surface. The EDX analysis (Fig. 4) found the presence of Gd and Ni particles in the deposits, the darker particles were identified as Ni while the lighter particles were identified as Gd_2O_3 . Fig. 5 presents general morphology studies of four deposits under different potentials (from -1.04 V/NHE to -1.30 V/NHE). As can be seen in (a) and (b) of Fig. 5, with a potential of -1.04 V/NHE, the deposit surface was uneven. This may be because the thickness of the deposit is too thin to smooth the rough surface of the substrate. In addition, under this applied potential condition, the distribution of Gd_2O_3 particles was not uniform, and some Ni matrix was not covered by Gd_2O_3 . When the potential value was set between -1.16 V/NHE and -1.2 V/NHE (c, d, e and f on Fig. 5), the microscopic surface structures of deposits were very similar. Their surfaces were smoother with more uniform Gd_2O_3 trapped in the Ni matrix as compared to the previous conditions. However, when the applied potential was too negative (-1.3 V/NHE), cauliflower-like structures and craters of Ni form on the deposit surface leading to a honeycomb-like overall deposit structure. This morphology is similar to morphologies of Ni or Cu co-deposition in very negative potential (Ćirović et al., 2015; Jović et al., 2006; Nikolić and Popov, 2010). Crater structures may be caused by the formation of hydrogen due to hydrogen reduction reaction (Nikolic et al., 2010). There was very little Gd_2O_3 on the larger cauliflower particles. Gd_2O_3 particles were gathered in the craters. With the formation of dendritic patterns, these deposits exhibit low adhesion and poor uniformity of Gd_2O_3 , which makes them unsuitable as a target.

Considering both the total Gd quantity obtained in the composite target and the quality of the deposit, -1.2 V/NHE was selected as the best applied potential to prepare the composite target for irradiation experiments.

3.1.2. Gd_2O_3 loading quantity effects

The comparison between no loading electrolyte and Gd_2O_3 loading curves (Fig. 3) shows that with the addition of Gd_2O_3 , the current density slightly decreases for the reduction of Ni^{2+} , but the slope of the reduction curve remains unchanged. This means that the reduction of Ni^{2+}/Ni with Gd_2O_3 becomes easier without affecting the reaction mechanism. This may be because the absorption of Gd_2O_3 increases the surface area of the cathode, which in turn provides more nucleation sites for nickel ions (Low et al., 2006). Through comparison between curves (a) and (b) of Fig. 3, it is found that under the same potential, the greater the Gd_2O_3 load, the more negative the current density, which also means that more deposits can be obtained. To investigate the effect of Gd_2O_3 loading, deposit Gd content was analyzed.

Fig. 6 shows the change of the Gd quantity trapped in the deposit as the addition of Gd_2O_3 increased from 0.1 g to 6.5 g. Experiments were repeated 3 times for each Gd_2O_3 loading, Fig. 6 presents mean values and standard deviations associated with each Gd_2O_3 content. The stirring speed was 300 rpm and the applied potential was -1.2 V/NH for all these deposits. The total amount of Gd_2O_3 trapped in the deposit is positively correlated with the amount of Gd_2O_3 in the solution (Fig. 6 (a)). Only 0.4 mg Gd is trapped in the deposit for 0.1 g of Gd_2O_3 loading, while for 6.5 g of Gd_2O_3 loading, the deposit contains 3.0 mg Gd. The effect of Gd_2O_3 loading is more obvious when the amount is smaller: when the loading mass increases from 0.1 g to 2.0 g, the trapped mass increases from 0.4 mg to 2.5 mg. The effect of Gd_2O_3 loading is weakened when its quantity is more than 2.0 g. The change of utilization rate of Gd

with the Gd_2O_3 loading is reported in Fig. 6 (b). When less Gd_2O_3 is added, the ratio of trapped Gd to the incorporated Gd_2O_3 is higher. As the Gd_2O_3 loading increases from 0.1 g to 6.5 g, the incorporation rate of Gd decreases from 0.40 % to 0.04 %. A high incorporation rate is beneficial for cost saving when using enriched Gd oxide. So, a balance must be found to use as little as possible Gd_2O_3 while ensuring that sufficient Gd is trapped in the deposit.

3.1.3. *Stirring speed effects*

In the process of co-deposition, the capture of Gd_2O_3 particles is a mechanical process where charged $Ni(NH_3)_6^{2+}$ and $Ni(NH_3)_5^{2+}$ ions formed by the complexation of NH_3 and Ni^{2+} could wrap around Gd_2O_3 particles to form clusters. To achieve this, Gd_2O_3 clusters that are in suspension in the electrolyte need to pass through the convection layer, diffusion layer and electrical double layer to finally reach the deposited layer (Low et al., 2006). The effect of stirring is not only to distribute the clusters as evenly as possible in the electrolyte but also to affect the state of the fluid, which will affect the transport of clusters in the convection layer. The effects of stirring speed on the Gd content of Ni- Gd_2O_3 deposits can be seen in Fig. 7. Each point in the figure is the average of three repeated experiments, and the error corresponds to the standard deviation. It can be seen that the Gd content increases when the stirring speed increases from 100 rpm to 300 rpm (from 1.4 mg to 3.0 mg), but it decreases when the stirring speed increases further, from 300 rpm to 800 rpm (from 3.0 mg to 1.8 mg). The low Gd content at lower stirring speed may be explained as a cluster distribution problem: when the stirring speed is too low (100 rpm), the distribution of clusters is not homogeneous, more clusters gather at the bottom of the cell. The low Gd content at high stirring speed may be related to the convection process: when the stirring speed is too high (800 rpm), the electrolyte is in a turbulent state, and the strong hydrodynamic force takes away part of the Gd_2O_3 particles so that they cannot diffuse into the deposited layer.

From these results, the stirring speed was fixed at 300 rpm for the composite target making.

3.2 *Cross section results*

According to parameter analysis, over 50 targets were fabricated to verify the above optimal experimental conditions (an applied potential of -1.2 V/NHE, a stirring speed of 300 rpm and the addition of 6.5 g of Gd_2O_3). Then, seven targets were manufactured with a co-deposition duration varied from 10 to 60 min. These targets were irradiated for cross section measurement.

Gd content in targets and measured cross section values are summarized in Table 4.

Fig. 8 shows the comparison of our measured values and existing data in the literature (Duchemin et al., 2016; Szelecsényi et al., 2016; Tárkányi et al., 2014). The figure evidence that despite a low amount of Gd in the deposit, the measurement results of this work are in good agreement with other data, and the experimental errors of this work are similar to the one in the literature data. This confirms that Ni- Gd_2O_3 composite deposits can be used as targets for the ^{155}Tb cross section measurement, even in the case of small quantity of Gd (0.4 mg, target n° 2 from Table 4). This indicates that the co-deposition method can be applied to manufacture Gd targets. We also see that even a small amount of Gd_2O_3 can be efficient to obtain relevant data which is an advantage for future work on enriched $^{155}Gd_2O_3$ from the point of view of cost.

4. Conclusion

In this study, we have successfully developed a method to make Ni-Gd₂O₃ composite thin targets via electrochemical co-deposition. This method allows manufacturing targets that can be used for production cross section measurements. Considering the surface quality and deposited Gd quantity, the optimal condition for the co-deposition to apply are -1.2 V/NHE potential, with a load of 6.5 g Gd₂O₃ and a stirring speed of the electrolyte equal to 300 rpm. Under these conditions, we can reach a thickness of 14.3 μm after one hour of co-deposition, which contains 3.0 mg of Gd. SEM analysis confirms that the surface of the deposits is flat and the distribution of Gd₂O₃ is uniform.

The feasibility of cross section measurement through composite targets has been confirmed. The measured cross section values show great consistency with the data in the literature. This indicates the possibility to use these technique together with enriched gadolinium material to produce thin targets for cross section measurements. ¹⁵⁵Tb production cross sections induced by deuteron and proton reactions on enriched ¹⁵⁵Gd will be measured as the next step. This also opens the possibility to use this technique for thin target preparation in general when electroplating is difficult.

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Figure Captions

Fig. 1 Schematic diagram of cell structure and electrodes positions used in our experiments: The cell contains 35 mL electrolyte solution. WE refers to the working electrode, AE to the auxiliary electrode, RE to the reference electrode. During the process, a magnetic stirring, a sealing film and a stainless steel plate are also used. Suspended Gd_2O_3 particles (black dots) were evenly distributed in the electrolyte thanks to the stirring. The schematic diagram is not drawn according to the actual scale.

Fig. 2 Schematic diagram of the stack composition used for the experiment where we irradiated target $n^{\circ}4$ and $n^{\circ}7$. The size and thickness of the foils are not drawn according to the actual scale.

Fig. 3 Voltammetry curves of Ni with Gd_2O_3 loading (0.8 g Gd_2O_3 for (a) and 6.5 g Gd_2O_3 for (b)) or without Gd_2O_3 loading. Sweep from -0.1 V/NHE to -1.5 V/NHE for 5 cycles with a scanning speed of 0.05 V/s. The curves presented are the curves of the fifth cycle (all curves of each cycle present the same behavior).

Fig. 4 EDX analysis of the chemical composition of the deposits. Applied potential was -1.20 V/NHE, stirring speed was 300 rpm, Gd_2O_3 loading was 6.5g, Ni^{2+} concentration was 0.14 mol/L, pH = 9.8.

Fig. 5 Microscopic images of surface morphology and Gd_2O_3 distribution of the deposits. Loaded Gd_2O_3 was 6.5 g, stirring speed was 300 rpm, applied potential was -1.04 V/NHE (a, b), -1.16 V/NHE (c, d), -1.20 V/NHE (e, f), -1.30 V/NHE (g, h).

Fig. 6 Effects of Gd_2O_3 loading on the quantity of Gd trapped Ni- Gd_2O_3 deposits. (a) mass of Gd in deposits. (b) Percentage of trapped Gd relative to the mass of Gd_2O_3 incorporated. The stirring speed was 300 rpm, the applied potential was -1.2 V/NHE.

Fig. 7 Effects of stirring speed on the Gd content of Ni- Gd_2O_3 deposits. The applied potential was -1.2 V/NHE and the Gd_2O_3 loading was 6.5 g.

Fig. 8 Experimental cross section of $^{nat}Gd(d,x)^{155}Tb$.

Tables

Target number	Mean beam intensity (nA)	Energy (MeV)	Irradiation time (min)
1	60.2 ± 4.7	8.7 ± 0.5	30
2	61.7 ± 4.7	13.2 ± 0.5	60
3	78.7 ± 6.0	14.2 ± 0.4	60
4	53.6 ± 4.0	19.5 ± 0.6	30
5	89.3 ± 6.8	24.0 ± 0.5	60
6	59.5 ± 4.6	24.4 ± 0.3	30
7	53.0 ± 4.1	29.5 ± 0.3	30

Table 1 Beam parameters of each target foil

Radionuclide	Half-life (d)	E _γ (keV)	I _γ (%)
¹⁵⁵ Tb	5.32 (6)	180.08	7.5 (4)
⁴⁸ V	15.9735 (25)	983.525	99.98 (4)
^{58(m)} Co	70.86 (6)	810.7593 (20)	99.450

Table 2 Decay data of detected radionuclides (Nudat 2.8 database, 2021).

Applied potential (V/NHE)	Average current density (mA/cm ²)	Deposit thickness (μm)	Gd quantity (mg)	Gd ratio in the deposit (at.%)
-1.04	-1.9 ± 0.4	3.1 ± 0.4	1.2 ± 0.3	8.5 ± 0.6
-1.16	-9.3 ± 0.6	12.2 ± 0.6	1.8 ± 0.1	3.6 ± 0.1
-1.20	-10.2 ± 0.6	14.3 ± 0.7	3.0 ± 0.4	4.3 ± 0.3
-1.30	-17.3 ± 0.9	26.5 ± 1.8	8.6 ± 0.6	4.5 ± 0.6

Table 3 Applied potential effects on average current density, deposit thickness, deposited Gd mass and Gd content in composites deposits

Target number	Gd content (mg)	Energy (MeV)	Cross section ¹⁵⁵ Tb(mb)
1	3.1	8.7 ± 0.5	24.5 ± 1.9
2	0.4	13.2 ± 0.5	87.8 ± 14.9
3	1.7	14.2 ± 0.4	104.2 ± 14.4
4	2.6	19.5 ± 0.6	208.5 ± 15.7
5	1.8	24.0 ± 0.5	322.7 ± 29.8
6	2.6	24.4 ± 0.3	362.6 ± 27.9

7

2.5

29.5 ± 0.3

304.0 ± 29.7

*Table 4 Irradiated Ni-Gd₂O₃ composite target information and cross section of ^{nat}Gd (d, x)
¹⁵⁵Tb*

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