

Optimization of the production process of gadolinium oxide targets for nuclear medicine by spark plasma sintering

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ABSTRACT

Terbium-155 (¹⁵⁵Tb) radionuclide is in the recent years under the spotlight of the nuclear medicine scientific community, leading an excellent candidate for the theranostic approach due to its useful nuclear properties. However, terbium radionuclides application is extremely challenging because of their limited availability in enough amounts mainly for supply issues. Developing suitable and reliable solid targets is a technological issue in the cyclotron-based production of medical radionuclides. The present study investigates the Spark Plasma Sintering (SPS) technique to manufacture coin-shaped gadolinium oxide (Gd₂O₃) targets for ¹⁵⁵Tb production by low-energy (i.e. medical) cyclotrons. A key feature of this approach is the multilayer configuration, consisting of a niobium backing (disk Ø 23.5 mm, 99.9% purity), a platinum foil (25 µm thick, 99.99+% purity), and a sintered pellet of ^{nat}Gd₂O₃ starting from powder (particle size <10 µm). Different temperatures and sintering configurations were tested to optimize the process. Scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and X-ray diffraction (XRD) were used to analyze the powder, pellets, and targets both from morphological and microstructure features. The main outcomes emerged demonstrate the feasibility of manufacturing coin-shaped Gd₂O₃ targets via the SPS technique, thus underscore its potential to enable routine ¹⁵⁵Tb production through the ¹⁵⁵Gd(p,n)¹⁵⁵Tb reaction route with medical cyclotrons.

1. Introduction

Nowadays, radiopharmaceuticals are widely used for the diagnosis and treatment of both oncological and non-oncological diseases [1]. A radiopharmaceutical is a drug containing a radionuclide, used either for the diagnosis or treatment of various medical conditions, depending on the type of radiation emitted by the radionuclide. [2]. For imaging purposes, radionuclides emitting γ or β⁺ radiation are suitable for single-photon emission computed tomography (SPECT) and for positron emission tomography (PET), respectively. On the other hand, the radionuclides that emit β[−], auger-electrons, or α radiations can effectively destroy pathological cells [3,4]. In the last twenty years, a new frontier in nuclear medicine has emerged, with the so-called “theranostic approach”, a well-established strategy that combines diagnostic imaging and therapy by using either the same molecule or closely related

analogues. Theranostics enables a personalized medicine approach, allowing patients to receive targeted treatments while avoiding unnecessary and expensive therapies [5–7].

A notable example of the theranostic radionuclides is provided by terbium isotopes (Tb): ¹⁵²Tb (β⁺ emitter), ¹⁵⁵Tb (γ emitter), ¹⁴⁹Tb (α emitter), and ¹⁶¹Tb (β[−] emitter). The interest in terbium-based radiopharmaceuticals has increased over the past decade, as highlighted in recent reviews [8,9]. In particular, the physical properties of ¹⁵⁵Tb make it well-suited for SPECT imaging. Its chemical characteristics enable its use as a diagnostic counterpart to therapeutic radiolanthanides or pseudo-radiolanthanides, such as ¹⁷⁷Lu and ⁹⁰Y. Furthermore, ¹⁵⁵Tb represents an ideal diagnostic match for the α-emitter ¹⁴⁹Tb or the β[−]-emitter ¹⁶¹Tb, paving the way for a true theranostic approach. Albeit several methods for ¹⁵⁵Tb supply have been reported in the literature [10,11], achieving the necessary quality and quantity for medical

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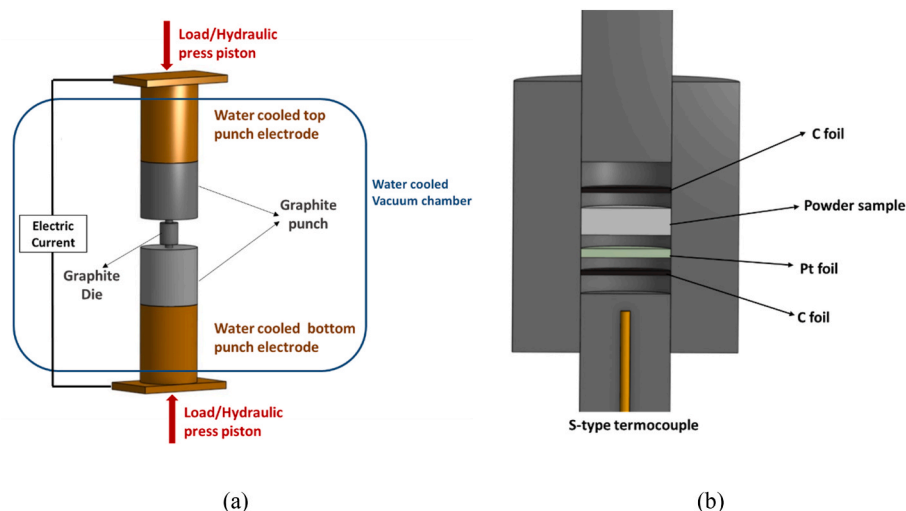


Fig. 1. (a) Scheme of the SPS apparatus; (b) focus on the assembled graphite structure for Gd_2O_3 powder sintering.

applications remains however challenging. One potential solution is the production of ^{155}Tb via proton irradiation of highly enriched ^{155}Gd and ^{156}Gd oxide targets, as demonstrated by Favaretto [12] and Dellepiane [13]. The $^{155}\text{Gd}(p,n)^{155}\text{Tb}$ reaction can be successfully exploited using medical cyclotrons, while the $^{156}\text{Gd}(p,2n)^{155}\text{Tb}$ reaction is feasible using both research and commercial cyclotrons. While the second reaction route generally leads to a higher production yield, the first one is instead characterized by a higher Radio Nuclidic Purity (RNP). In both cases, the impurity produced is ^{156}Tb , an unwanted radionuclide which half-life is comparable to that of ^{155}Tb (5.35 days). Its production can be minimized by selecting a suitable energy range. Regarding the $^{155}\text{Gd}(p,n)^{155}\text{Tb}$ reaction, it was estimated that an ultra-pure ^{155}Gd target (>98% with <2% of ^{156}Gd) is required to reach an acceptable RNP and Dose Increase (DI) that is safe for clinical use [14]. The enrichment level of ^{155}Gd currently available commercially is not sufficiently high. This requirement would substantially increase material costs, also given that the natural abundance of ^{155}Gd is 14.8%. However, the study of ^{155}Tb production using a medical cyclotron is still an R&D issue. Further, in vivo dosimetric studies are required to better understand the importance of ^{155}Tb , so that a scenario involving diffuse worldwide production of ^{155}Tb could offer a much more favorable economic return for companies producing enriched material [14].

One of the key technical issues for ^{155}Tb production is establishing a reliable ^{155}Gd target manufacturing process, along with the radiochemical separation process. Cyclotron-based radioisotope production is a multidisciplinary process that develops in several stages [15], including target fabrication, cyclotron irradiation, radiochemical separation and purification, quality control, and labeling of molecules to obtain the final radiopharmaceutical product. The cycle often ends up with the recovery of costly isotope-enriched materials following the radiochemical process. One of the most critical technological challenges in cyclotron-based radioisotope production is the design and preparation of a suitable target [16]. In this work, the main objective was to study a target configuration compatible with irradiation in an ACSI TR19/300 cyclotron and suitable for easy dissolution in a hot cell, by using remote manipulators, thus easing the future routine production. In general, the composition and thickness of the target material are determined by the selected nuclear reaction to maximize the radionuclide production efficiency. Since the target material is often isotopically enriched and therefore expensive, minimizing material losses is essential to ensure economically sustainable production. The support material must be appropriately selected based on its thermo-mechanical characteristics, to withstand the thermal power deposited on the target during irradiation, and its chemical inertia, to avoid contamination in

the final product due to the dissolution process. Consequently, the most suitable target production technique depends mainly on the physical and chemical properties of the starting material, its commercially available forms, and the material of the support plate.

At the National Institute for Nuclear Physics – Legnaro National Laboratories (INFN-LNL), the Field Assisted Sintering Technology Spark Plasma Sintering (FAST SPS) technique is used for manufacturing targets of different materials to estimate the production yield and RNPs using low-energy cyclotrons [17–19]. Compared to conventional sintering processes, SPS works with lower sintering temperatures, higher heating rates, and shorter sintering times. A further advantage of this technique, considering the expensive isotopically enriched materials commonly used in this field, is the negligible loss of the starting material. SPS also enables the joining of similar or dissimilar materials without filler [20–23]. The sintering of Gd_2O_3 powder by SPS has been described in a few works in the literature [24,25] for different technological applications. The hydraulic pressing of Gd_2O_3 powder has been already used for the production of ^{155}Tb , the sintering approach has been considered by the authors to obtain more robust Gd_2O_3 pellets. The Gd_2O_3 nanopowder was successfully sintered by SPS into a pellet with a thickness of about 300 μm , while the same sintering parameters guarantee pellets with a thickness of about 600 μm when using micropowder. These first natural Gd_2O_3 pellets were encapsulated in an Al-holder for the irradiation with a medical cyclotron. According to our preliminary study, the micrometric powder seems to be closer to the enriched one. The production of thin pellets (lower than 600 μm) starting with Gd_2O_3 micrometric powder is not a trivial task, and investigations are needed. Additionally, for easy handling of the target, the coin-shaped design is more favorable [26].

In this work, the authors have developed and set up the Gd_2O_3 target manufacturing technology, enabling production via the $^{155}\text{Gd}(p,n)^{155}\text{Tb}$ reaction route using the medical cyclotron at the IRCCS Sacro Cuore Don Calabria Hospital (SCDCH, Negrar di Valpolicella, Verona, Italy). The new configuration includes the sintering of Gd_2O_3 powder to create a pellet and its adhesion to a backing disc. The most challenging part is to obtain a mechanically robust Gd_2O_3 pellet with a thickness of about 200–150 μm starting from micropowder. This thickness is considered optimal to exploit the proton energy range with the maximum cross-section to maximize the production of the ^{155}Tb and minimize the production of ^{156}Tb as reported in Ref. [14]. The sintering parameters were optimized using the natural powder to refine the entire process before applying it to the expensive enriched powder. Each target was characterized both in terms of morphology and microstructure (e.g., Scanning Electron Microscopy (SEM) and X-Ray Diffraction spectroscopy (XRD)), with the

Table 1
SPS process parameters of different target production steps.

	Temperature [°C]	Pressure on sample [MPa]	Dwell time [min]	Heating rate [°C/min]
Gd ₂ O ₃ /Pt pellet system	1200-1300- 1500	48	5	100
Backing Pt/ Nb	1000	11	3	200
Target Gd ₂ O ₃ / Pt – Pt/Nb	900	48	5	100

aim of determining the optimal set of sintering parameters. This work paves the way for the future use of enriched ¹⁵⁵Gd[Gd₂O₃] material to allow efficient production of ¹⁵⁵Tb, and it was performed in the framework of the project APHRODITE-155, part of the LARAMED program at INFN-LNL. The project aims to compare the production routes for ¹⁵⁵Tb using low-energy and high-energy cyclotrons, starting from ¹⁵⁵Gd targets and ¹⁵⁹Tb (for indirect production), respectively [27].

2. Materials and methods

The thickness of the foils and the pellets were measured using a Mitutoyo micrometre screw gauge (0-25 mm, 0.001 mm). A KERN ADJ 200-4 analytical balance (Max 220 g, d = 0.0001 g) was used for weighting. The Gd₂O₃ powder with a purity of 99.9% Rare Earth Oxide (REO) and a particle size of <10 µm, and the platinum foils (purity 99.95%, thickness 25 ± 10% µm) were purchased from Alfa Aesar. Niobium discs (purity 99.9%, diameter = 23.5 mm and thickness = 1.5 mm) were purchased from Goodfellow.

The enriched ¹⁵⁵Gd[Gd₂O₃], with 90.20 ± 0.10% enrichment level, was purchased by ISOFLEX.

2.1. Pellet production with SPS process

The Gd₂O₃ powder was sintered with the SPS process by using the TT_Sinter machine, which was designed and assembled by INFN Pavia division and University of Pavia [18,28]. The powder particles were placed into a graphite structure composed of a punch and die appropriately designed for the realization of a 10 mm diameter pellet. A graphite felt is used around the graphite die to shield against thermal radiation and help evenly distribute heat during the sintering process. The process occurs in a vacuum of about 1×10⁻² mbar, and the temperature is measured by a S-type thermocouple placed in a hole in the lower graphite punch (Fig. 1b). In order to obtain high-quality sintered pellets, both novel and previously reported configurations have been investigated [29,30].

Two graphite foils (thickness 20 µm) were placed beneath the Pt foil and above the powder to prevent them from sticking to the graphite die after sintering. The schematic sketch of the experimental setup is shown in Fig. 1b. The adhesion of the Pt foil to the Gd₂O₃ powder during sintering was performed at temperatures of 1200, 1300, 1500 °C, under a constant pressure of 48 MPa. The heating rate was 100 °C/min, with a dwell time of 5 min (Table 1). The temperature range was selected based on literature data [24,25,31], while also considering the material's melting points (T_m (Gd₂O₃) = 2420 °C; T_m (Pt) = 1768 °C) and the Gd₂O₃ phase transformation occurring at 1250 °C.

Once determined the optimal parameters, a simultaneous sintering of three pellets in series was tested by using graphite spacers, as demonstrated in a recent work by Cisternino et al. [18]. At the end of the

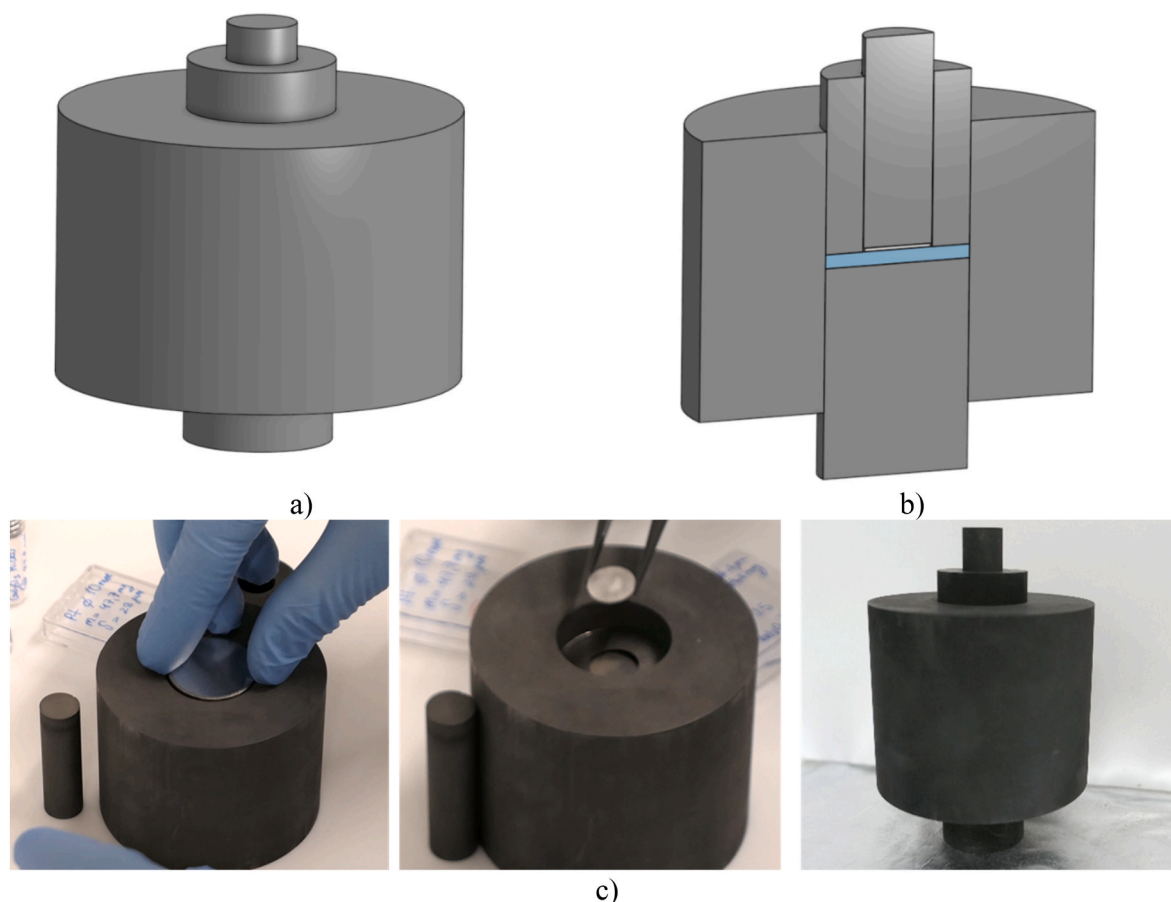


Fig. 2. a) and b) Rendering of the graphite punches; c) pictures of the assembling step for the adhesion of the pellet system to the backing disc.

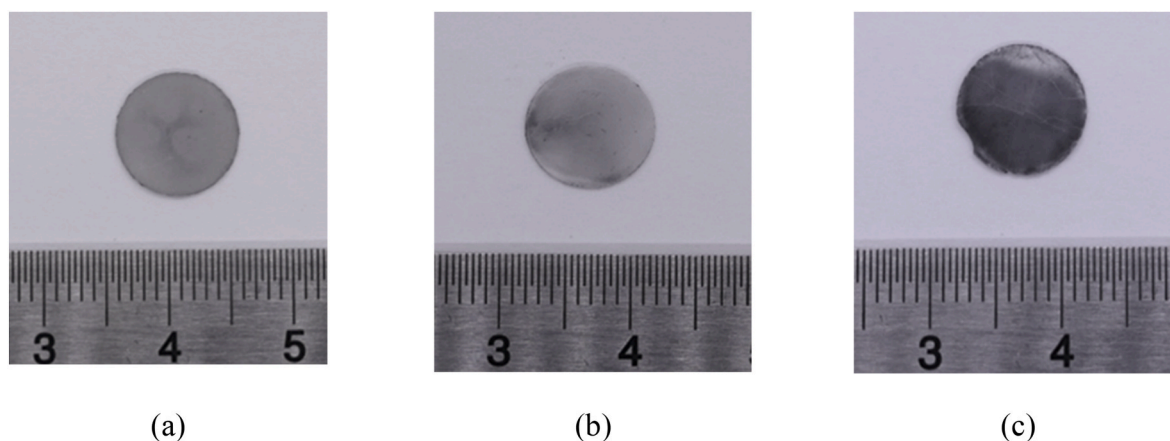


Fig. 3. Picture of HT pellets sintered at 1200 °C (a), 1300 °C (b), and 1500 °C (c).

sintering process, the pellets were washed in an ultrasonic bath for about 5 min in pure ethanol to remove carbon foils and contaminant residue coming from the graphite punch. The uniformity of pellet thickness across the diameter was assessed by measuring the thickness at five different points. Measuring the final pellet weight is essential for calculating the exact amount of material lost during each experiment. This information is crucial for assessing the efficiency of the SPS technique, since the final target will be produced using costly isotopically enriched material.

The theoretical density of Gd_2O_3 declared in the powder datasheet (7.41 g/cm^3) was considered for the calculation of the initial powder mass to obtain precise pellet thicknesses, hereinafter referred to as HT (high thickness, $\sim 300 \mu\text{m}$), MT (medium thickness, $\sim 200 \mu\text{m}$), and LT (low thickness, $\sim 150 \mu\text{m}$). The corresponding starting powder was 174 mg, 116 mg and 87 mg, respectively. The apparent density of each pellet was determined as the total mass divided by its volume, after subtracting the mass of the platinum foil. The relative density, in percentage, was calculated using the theoretical density of gadolinium oxide.

2.2. Coin-shaped target configuration

To manufacture the final target configuration, the pellets were bonded to a suitable backing consisting of Pt foil on a Nb disc. The backing was prepared by adhering the platinum foil to the niobium disc via SPS. Similar to pellets, backing discs can be manufactured in series using graphite spacers.

Tailored graphite components were specifically designed for the adhesion of the pellet to the backing. The configuration consists of a standard die and punch, complemented by a centering ring for accurate pellet alignment. Closure is achieved using a thinner punch, precisely dimensioned to match the pellet diameter, ensuring uniform load distribution across the surface (Fig. 2). The parameter values for the different production steps (pellet fabrication, backing preparation, and final target assembly) are summarized in Table 1.

2.3. Materials characterization

The micro Gd_2O_3 powder was analyzed using scanning electron

microscopy (SEM, Zeiss EVO MA 10) and compared with the isotope-enriched one $^{155}\text{Gd}[\text{Gd}_2\text{O}_3]$ to evaluate differences in particle morphology. Since the powder characteristics may influence the sintering process, this analysis was performed to optimize the SPS parameters before using the enriched powder. On the other hand, nanometric powder was not considered in this study, as it had already been investigated in a previous work by the authors [26].

Pellets and targets were analyzed by SEM in both backscattered electron (BSE) and secondary electron (SE) modes, equipped with energy dispersive X-ray spectrometry (EDS) (Oxford Instruments INCAx-act), to assess the morphological characteristics, thickness, and elemental composition. To investigate the sintering quality and the adhesion between materials, metallurgical interface analyses were performed with tailored sample preparation. Some samples were subjected to hot and cold mounting for comparison using epoxy resins, followed by cutting and standard metallographic preparation (grinding with SiC abrasive papers of 500, 800, and 1200 grit, followed by polishing with cloths using diamond suspension up to $1 \mu\text{m}$). Additionally, some pellets were intentionally fractured without any metallographic preparation. To guarantee optimal electrical conductivity and minimize charging effects, all samples analyzed were subjected to gold sputter coating before SEM observation.

X-ray diffraction (XRD) measurements were carried out on both powders and pellets using a Bruker AXS D8 Advance diffractometer with $\text{CuK}\alpha$ radiation, operating in Bragg-Brentano configuration, within a 2θ range of 10° – 60° (step size: 0.025° , time per step: 1 s).

3. Results and discussion

3.1. Morphological characterization of pellets and targets

The resulting Gd_2O_3 pellets starting from low amount of micro-powder (174–87 mg) are brittle. Thus, the insertion of a platinum foil between the powder and the die during the sintering process was identified as the best solution. This approach improves the mechanical stability of the pellets and allows the sintering of Gd_2O_3 samples of different thicknesses without cracking and/or breaking, even at high temperatures. Moreover, it is useful for further adhesion to the backing.

Table 2
HT pellets sintered at different temperatures.

Sintering temperature [°C]	Starting powder mass [mg]	Gd_2O_3 pellet mass [mg]	Lost mass [mg]	Thickness Gd_2O_3 [μm] /[mg/cm^2]	Apparent density [g/cm^3]	Relative density [%]
1200	173.3	164.0	9.3	310/208.9	6.74	90.9
1300	178.5	172.4	6.1	308/219.5	7.13	96.2
1500	172.9	157.2	15.7	299/200.3	6.70	90.4

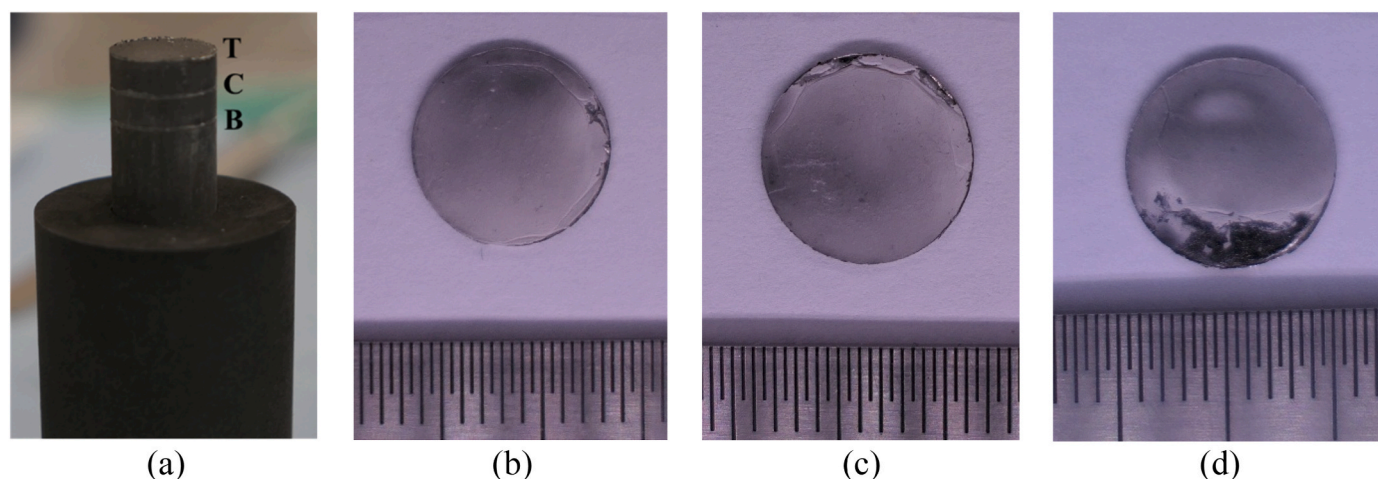


Fig. 4. (a) Configuration of Gd_2O_3 pellets sintered in series at 1300 °C: (b) top, (c) center, and (d) bottom.

Table 3

Pellets sintered at 1300 °C with different starting powder.

	N. of experiments	Starting powder mass [mg]	Final Gd_2O_3 mass [mg]	Mass lost [mg]	Thickness Gd_2O_3 [μm]/[mg/ cm^2]	Apparent density [g/ cm^3]	Relative density [%]
HT	(n = 5)	174.6 ± 4.4	168.8 ± 3.2	5.8 ± 4.1	333.8 ± 20.8 /215.5 $\pm$ 4.0	6.5 ± 0.5	87.2 ± 6.3
MT	(n = 5)	116.4 ± 0.9	115.8 ± 3.6	2.0 ± 0.9	266.0 ± 11.4 /147.5 $\pm$ 4.6	5.6 ± 0.3	75.0 ± 3.9
LT	(n = 22)	86.4 ± 2.0	84.5 ± 2.2	1.9 ± 0.8	209.9 ± 22.8 /107.7 $\pm$ 2.8	5.2 ± 0.5	69.9 ± 7.1

The effect of sintering temperature was first investigated on the HT pellets, starting from an initial powder mass of approximately 174 mg. Fig. 3 shows the HT Gd_2O_3 pellet sintered at (a) 1200 °C, (b) 1300 °C, and (c) 1500 °C, respectively. The pellet sintered at 1500 °C appeared darker and more brittle, with distinct cracks, compared to those obtained at 1200 °C and 1300 °C. A similar change in the color of sintered stabilized zirconia has been reported in the literature [32]. This suggests that the observed changes could be attributed to a deviation from stoichiometry due to oxygen loss, as proposed in Ref. [33]. This alteration could impact on the optical and dielectric properties of the material; however, it does not pose significant issues for the purpose described in this work. In Table 2, the resulting characteristics of the HT sintered pellets at different temperatures are reported.



Fig. 5. Photograph of Gd_2O_3 targets produced with SPS.

The observed differences in the pellet mass between samples are due to manual operation during the weighing of the starting powder. Despite literature reporting that increasing the sintering temperature leads to higher density, the highest relative density is achieved at 1300 °C. Additionally, at 1500 °C, the highest powder loss is observed. Based on these preliminary results, 1300 °C was thus selected as the sintering temperature for the following experiment. This temperature was also used to produce the pellets in series, while keeping the other process parameters unchanged. However, as can be observed from Fig. 4, the pellets manufactured in series show scarce reproducibility and uniformity, probably due to the low powder mass (LT pellets) and assembly operation.

According to the recent nuclear physics cross-section measurements, it has been confirmed that the optimal energy range for the production of ^{155}Tb by proton beam is 10.5–8 MeV [14]. This energy interval corresponds to a thickness of about 108 mg/ cm^2 (LT~150 μm) of ^{155}Gd [Gd_2O_3]. This result has been determined by using the SRIM-2013 code [34] considering the proton energy loss through Gd_2O_3 . Consequently, low amount of starting powder was used to achieve the desired thickness (LT). No significant differences were observed among the various pellets (HT, MT and LT) sintered one by one, keeping unchanged 1300 °C as the maximum temperature. They all appeared intact and robust. However, as also shown in Table 3, the density of the final pellet appears to decrease, reducing the amount of powder. A lower initial powder quantity per unit volume could result in fewer interparticle contacts, which are crucial for effective densification. The reduced contact area hinders the formation of necks between particles during the sintering process, leading to a final structure with lower density and more residual pores [35]. Furthermore, a reduced powder quantity tends to distribute less uniformly within the dies, creating areas with loosely packed particles. This uneven distribution probably causes microstructural variations, ultimately increasing the porosity of the final sintered pellet.

Once the production of LT pellets was optimized, the final target was manufactured by adhering the pellets to the backing. Niobium was

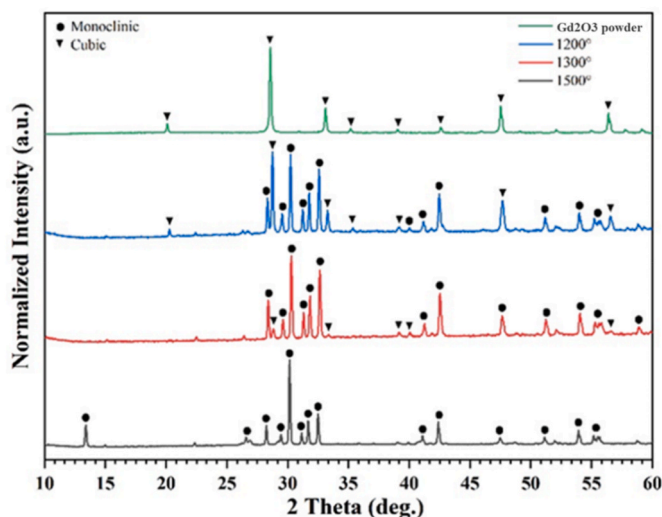


Fig. 6. Normalized XRD patterns of natural powder and HT pellets sintered at 1200 °C, 1300 °C, and 1500 °C.

selected as the backing material due to its high melting temperature ($T_m = 2468$ °C) and sufficient thermal conductivity ($K = 53.7$ W m⁻¹ K⁻¹). Platinum was chosen due to chemical inertness in acidic conditions, thus allowing the dissolution of the Gd₂O₃ pellet after irradiation without introducing contamination, and also because it was already incorporated into the pellet system. The target has been designed to fit perfectly with the geometry of the solid target station of the ACSI TR19/300 medical cyclotron (13–19 MeV) at the IRCCS SCDCH. This cyclotron target station could hold a coin with a maximum thickness of 2 mm and a diameter of 24 mm, and it enables direct helium gas cooling at the front side and water cooling from the back side of the target. The proton beam is perpendicular to the target with a diameter of 10 mm. As demonstrated in a previous work [19], this new coin-like target system allows an easy handling by telemanipulators inside a conventional hot cell, thereby minimizing the operator's radiation exposure. Moreover, the target configuration is perfectly compatible with the dissolution module employed during the radiochemical process [36], while maintaining high chemical inertness and minimizing metal contamination. A representative example of a gadolinium oxide target produced using the SPS technique is shown in Fig. 5. The data collected from seven targets ($n = 7$) point out an increase in pellet density because of the adhesion process. Specifically, the relative density of the pellets calculated after the adhesion is $77.8 \pm 4.0\%$, compared to $70.0 \pm 3.1\%$ prior to adhesion. The increase in density suggests that the adhesion process induces an additional sintering effect on the pellets. It should be noteworthy to highlight that, for sintered pellets used as solid targets for medical radionuclide production, achieving a relative density of 100% is not a strict requirement. Moreover, lower relative density may be advantageous with respect to the subsequent chemical processing, as it can ease and speed dissolution of the irradiated target material. Nevertheless, the pellet density remains a key parameter, as it directly determines the effective target thickness required to achieve the desired radionuclide production yield.

3.2. XRD characterization

The XRD patterns of HT pellets sintered at different temperatures, along with the original natural Gd₂O₃ powders, are shown in Fig. 6.

At room temperature, Gd₂O₃ crystallizes into the cubic crystal structure. Around 1250 °C, it transforms into the monoclinic phase, while temperatures of about 2200 °C or higher, it changes into the hexagonal phase. The transformation of the cubic phase to monoclinic was found to be irreversible [37]. The Gd₂O₃ powders confirmed the

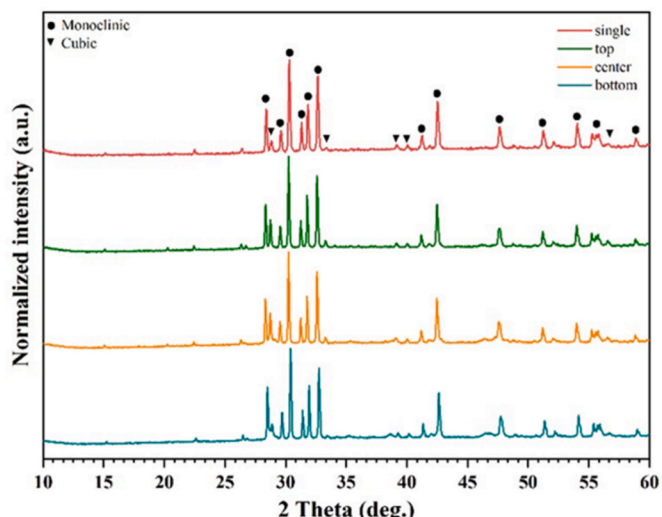


Fig. 7. Normalized XRD patterns of HT pellets sintered in series and in single-mode at 1300 °C.

presence of the cubic phase, with the three most prominent peaks observed at $2\theta = 28.55^\circ$, 33.09° , and 47.51° . The pellets sintered at 1200 °C and 1300 °C showed an incomplete phase transitions, containing a mixture of cubic and monoclinic structures, consistent with previously reported in the literature [25]. Whereas, at 1500 °C, the XRD pattern revealed a predominant monoclinic phase in the sintered pellet.

Moreover, the XRD analysis (Fig. 7) of the three HT pellets sintered in series showed no significant differences in their diffractograms, confirming that the same phase composition was consistently maintained. The observation of the coexistence of both cubic and monoclinic phases at 1200 °C and 1300 °C, despite the theoretical transition temperature being 1250 °C, may be related to the current electrical distribution inside the volume of the SPS setup. As reported by Tamburini et al. [22], the electric current distribution is primarily influenced by the die geometry, as well as the thermal and electrical conductivities for both the die and sample materials. The authors demonstrated that, in the case of a non-conductive sample, no current flows through the sample itself. Instead, the current is primarily carried by the surrounding graphite matrix, resulting in a relatively low gradient from the inner to the outer part of the matrix. In contrast, for the conductive sample, the highest current density is observed along the lateral surface of the sample. Moving outward into the graphite matrix, the current density drops sharply until it reaches a minimum. Furthermore, a significant radial distribution of the current within the conducting sample indicates that the current does not flow uniformly but follows a more complex and non-linear path. In the present study, the use of the platinum foil may have induced current deflection and localized heating effects. Since the temperature distribution is directly linked to the current flow, the difference in electrical resistivity between the platinum foil and the graphite components leads to non-uniform heating, with areas near the foil reaching higher temperatures than the inner parts of the powder.

The XRD analysis of the LT pellets showed no significant differences compared to the HT pellets. Both the cubic and monoclinic phases are present. This suggests that the difference in thickness does not appreciably affect the crystalline structure of the pellets.

It is worth noting to highlight that the crystallographic phase of Gd₂O₃ does not affect the production yield of ¹⁵⁵Tb. Nevertheless, the precise knowledge of target properties, such as density, is essential to predict radioisotope yield performance under the beam irradiation and to estimate the time needed for the radiochemical dissolution process.

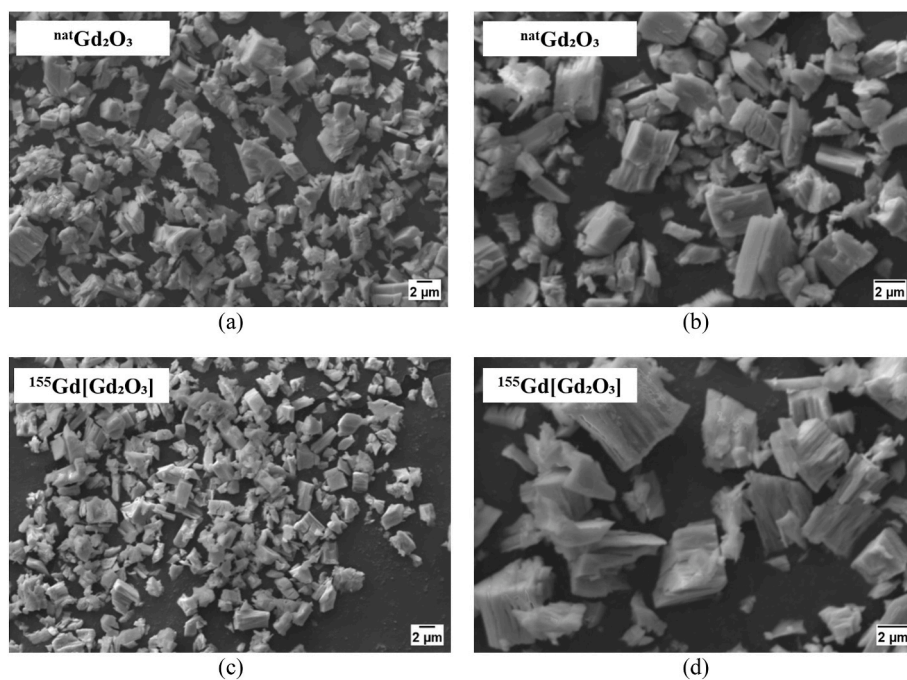


Fig. 8. SEM micrographs at different magnifications of:(a and b) natural gadolinium oxide powders and (c and d) isotopically enriched 155-gadolinium oxide powders.

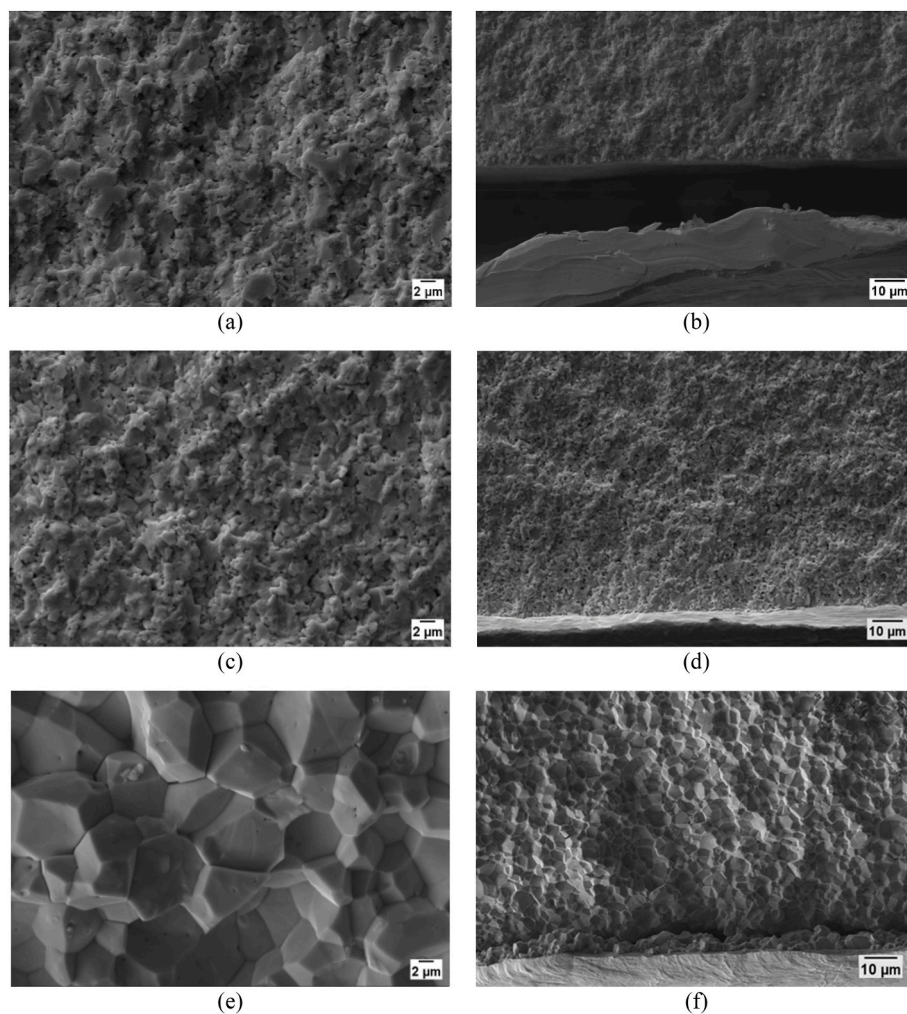


Fig. 9. SEM - SE micrographs at different magnifications of fractured pellets sintered at: 1200 °C (a and b); 1300 °C (c and d), and 1500 °C (e and f).

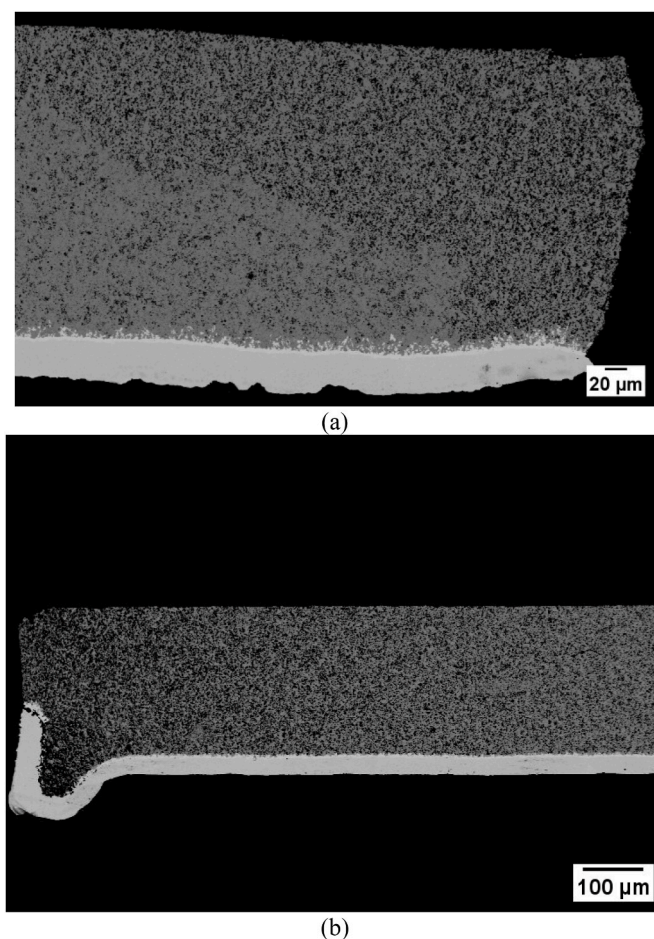


Fig. 10. SEM-BSE micrograph of a 1300 °C HT pellet: (a) hot-mounted and (b) cold-mounted procedures.

3.3. SEM characterization

SEM micrographs demonstrated that the micrometric natural gadolinium oxide powder (Fig. 8a and b) and the isotopically enriched powder (Fig. 8c and d) share similar morphology and particle dimension. The Gd_2O_3 microparticles are irregularly distributed, forming three-dimensional crystalline domains, in accordance with previously reported data in the literature [38]. No quantitative particle size distribution analysis was performed; therefore, the comparison is based on qualitative SEM observations.

To evaluate the effect of different temperatures on the sintering process, cross-sections analyses of the fractured HT pellets were

performed by SEM. The micrographs of the three pellets at 1200 °C, 1300 °C, and 1500 °C, are shown in Fig. 9. From a macroscopic point of view, the HT pellets sintered at 1200 °C and 1300 °C turned out to be similar and well sintered (see Fig. 9(a–d)). However, the platinum foil was detached from the pellet at 1200 °C, indicating weak adhesion. In contrast, as the temperature increased to 1500 °C, noticeable growth of grains is observed (see Fig. 9(e–f)). The temperature was high enough to promote particle coalescence and grain growth, leading to the formation of a larger and more uniform crystalline structure. These observations support the choice of the 1300 °C process as the optimal sintering temperature and are consistent with those reported in the literature, despite different materials. For instance, as reported in Ref. [32], when the sintering temperature increased from 1100 °C to 1400 °C, a dramatic increase in the grain size of fully stabilized zirconia was observed. Moreover, in a recent study of Liang et al. [39], zinc oxide grains gradually grew with increasing sintering temperature from 500 °C to 800 °C during the SPS process. In both studies, the authors highlighted that the increase of the temperature leads to progressive grain growth.

To deeply understand the adhesion to the platinum foil, the HT pellets obtained at 1300 °C were hot-mounted and then cut for metallographic interface analysis. Based on the SEM-BSE images (Fig. 10 a), a clear density gradient is observed across the pellet, the side in contact with platinum appears denser than the edges. This observation suggests that the higher thermal conductivity of platinum foil may induce a temperature gradient during sintering. As a result, the area in contact with platinum heats more quickly and uniformly, promoting the formation of a more compact structure on that side. Taken together, the SEM and XRD findings support the hypothesis that temperature distribution during sintering, even in this small-sized sample, influences both densification and phase formation.

In the hot-mounted samples after the standard preparation process, dark spots were observed along the pellet edges (Fig. 11 a). To verify the origin of these spots, a comparison was made with cold mounting procedure. In this case, no dark spots were observed (Fig. 11 b). However, SEM analysis of the cold-mounted sample confirmed a difference between the edges and the center of the pellet, with the material appearing less sintered towards the outer regions (Fig. 10 b). This suggests that in the hot-mounted sample, the dark spots may be due to resin infiltration into the porous regions of the pellet. Since the calculated density remains unchanged, the structural differences observed are intrinsic to the pellet and not an artifact of the mounting method.

Furthermore, the cross-section analysis revealed a new phase at the $\text{Gd}_2\text{O}_3/\text{Pt}$ interface, indicating the occurrence of a reaction between the initial counterparts. The SEM-EDS mapping was performed along the cross-section to further clarify the elemental distribution, and the results are reported in Fig. 12. The interlayer was found to consist of a new binary Gd–Pt phase devoid of oxygen, resulting from the local reduction of the oxide followed by diffusion and interaction between gadolinium and platinum during the SPS process. Point EDS analyses at the interface

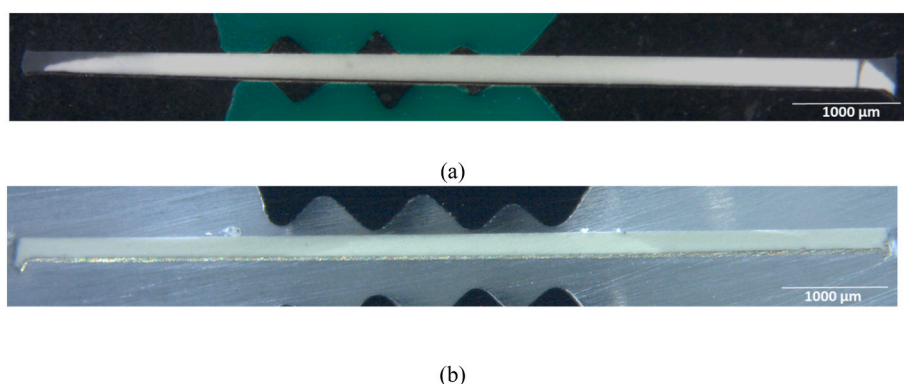


Fig. 11. Optical micrographs comparing (a) hot and (b) cold mounted samples.

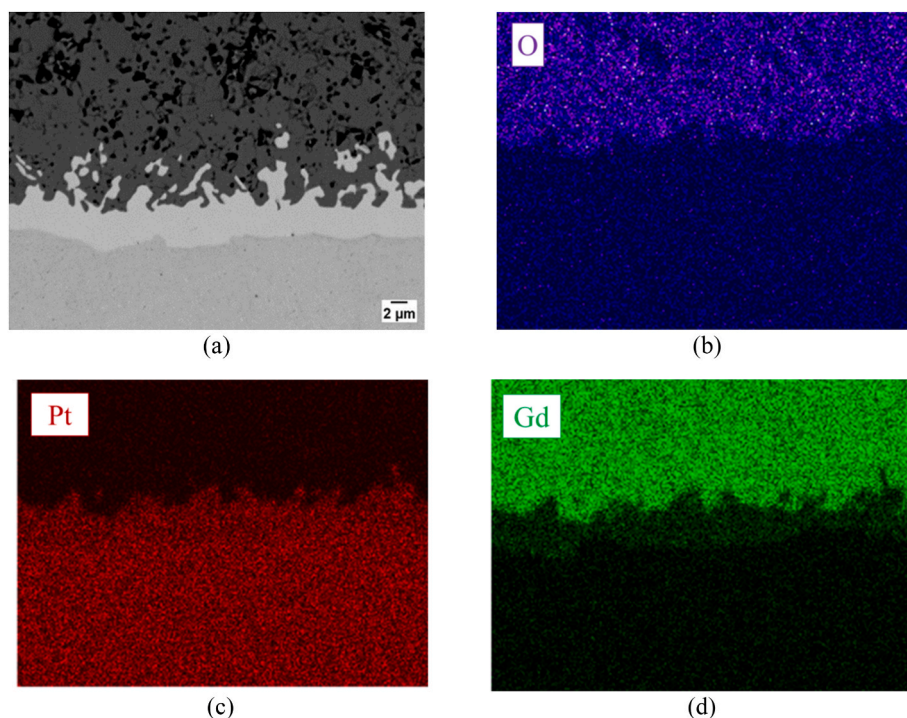


Fig. 12. SEM-EDS elemental mapping performed on the high magnification images of the cross-section of a hot-mounted 1300 °C HT pellet. (a) SEM-BSE micrograph; (b) Oxygen (purple); (c) Platinum (red) and (d) Gadolinium (green). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

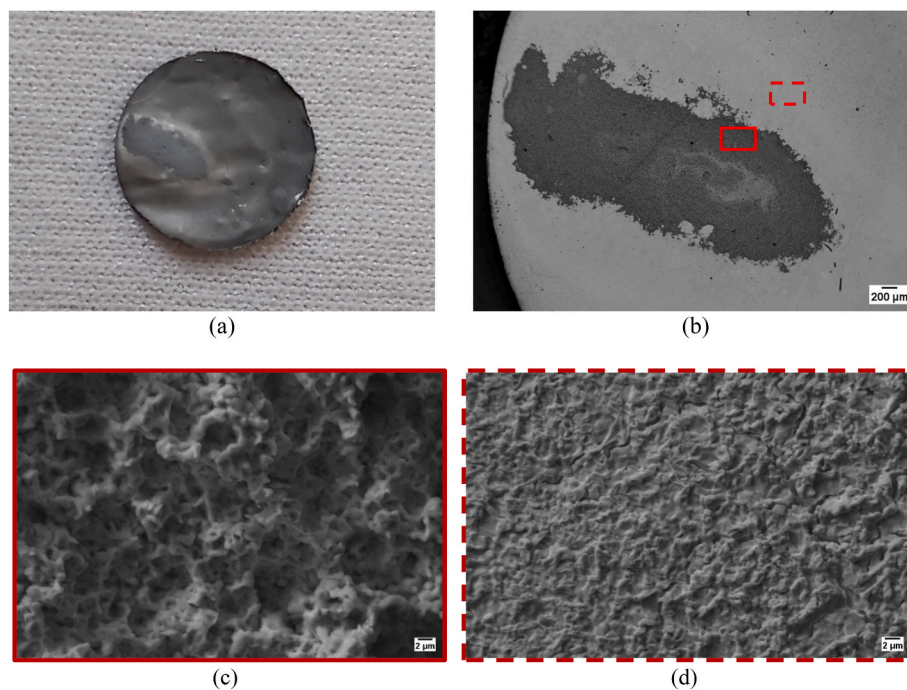


Fig. 13. (a) Photo of platinum foil after the dissolution of Gd_2O_3 pellet (b) SEM-BSE micrograph of Pt foil; (c) magnification of the spot and (d) of the rest of the foil.

confirmed the presence of gadolinium and platinum, with atomic percentages of 19.21% and 80.79%, respectively. This finding raises a concern that some amount of Gd may become trapped in the interlayer.

The same metallographic preparation procedure using cold mounting was applied to the LT pellets.

Analyses confirmed that pellets are well sintered and adhered to the metallic foil, despite the reduced starting powder. In the case of reduced

powder quantity, the initial non-uniform spreading of the powder within the die can influence local packing density and affect the contact resistance between the powder and the punches or metallic foil. The presence of multiple low-density regions in thinner pellets is consistent with their lower relative density (Table 3). The density gradient is not limited to the edges but also through the pellet thickness. Moreover, the interlayer between the oxide and the platinum is not homogeneously distributed

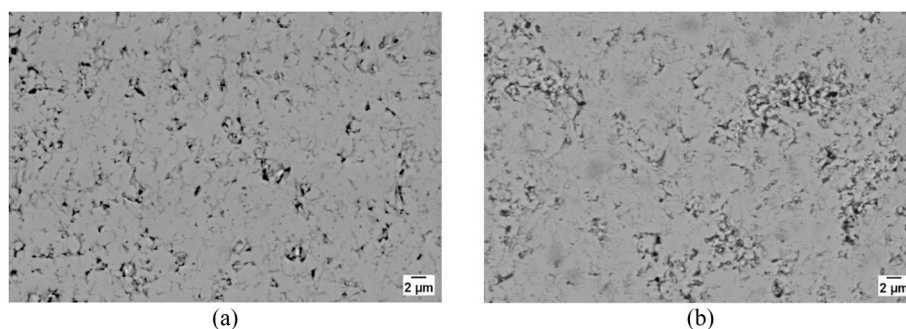


Fig. 14. Cross-section comparison between (a) LT and (b) HT pellets sintered at 1300 °C.

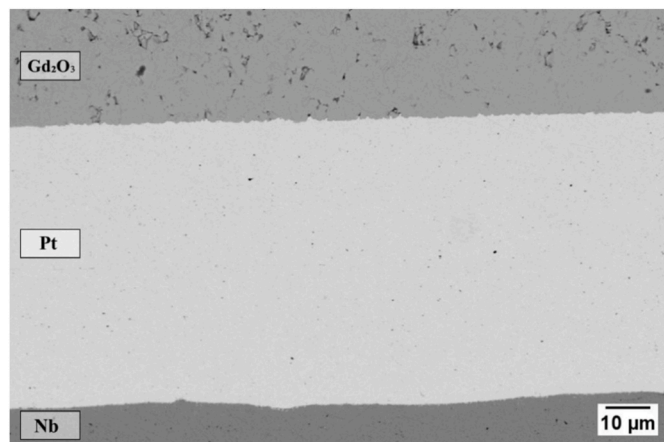


Fig. 15. SEM-BSE of the cold-mounted Gd₂O₃/Pt- Pt/Nb targets cross-section.

along the entire interface but is mainly observed at the pellet edges.

The presence of the interlayer in the LT pellets was further confirmed through Gd₂O₃/Pt system dissolution test in 5 mL of 7 M HNO₃ at 80 °C for approximately 1 h. The remaining platinum foil was analyzed. As illustrated in Fig. 13, a distinct dark spot is observed on the surface of the foil. SEM/EDS analysis confirmed that this dark spot is composed only of gadolinium oxide, while traces of gadolinium and platinum are detected on the rest of the foil. The presence of Gd on the Pt surface was estimated to be at 20 at.%. Investigations on new SPS parameters for the adhesion of Gd₂O₃ and Pt are currently underway to decrease the presence of Gd onto platinum foil after the dissolution step.

Nevertheless, SEM analysis of the cross-section of the cold-mounted LT and HT samples (Fig. 14) appears very similar. Therefore, it can be concluded that, using the same sintering parameters, the initial powder amount does not significantly affect the pellet microstructure.

Finally, a cross-sectional analysis of the targets was carried out to characterize the adhesion between the sintered pellet and the substrate. This evaluation was essential to better understand the interfacial bonding behavior of the different materials involved. As shown in the micrograph (Fig. 15), the adhesion is excellent. No voids or delamination are observed at the Pt/Nb and Gd₂O₃/Pt interfaces, indicating strong bonding. The platinum foils appear joined, forming a single continuous layer. These results highlight one of the key advantages of the spark plasma sintering technique, which is the ability to promote contact between dissimilar materials through localized Joule heating and rapid consolidation under uniaxial pressure. It is worth noting that similarity in coefficients of thermal expansion (CTE) between bonded materials is essential to avoid, at high temperature, strain to failure of the bonded region. From 900 °C onwards, the CTEs of Gd₂O₃ ($\sim 8.0 \times 10^{-6} \text{ K}^{-1}$), Pt ($\sim 9.0 \times 10^{-6} \text{ K}^{-1}$), and Nb ($\sim 7.5 \times 10^{-6} \text{ K}^{-1}$) remain relatively close, indicating good thermo-mechanical compatibility during high-temperature processing [25,40,41]. This matching is also

crucial to ensure the mechanical integrity and optimal performance of the target under proton beam irradiation in the cyclotron. These observations further supported the additional densification effect identified in the pellet after its adhesion to the backing discussed in the previous paragraph. The edges of the pellet appear slightly more compressed, suggesting localized deformation likely caused by the mechanical pressure applied during bonding. Notably, this effect is not reflected in the overall thickness measurements, which remain unchanged.

4. Conclusion

In the present work, a coin-shaped target of Gd₂O₃ for the production of Tb radioisotopes by cyclotron was developed by the SPS technique. We thoroughly investigated the entire manufacturing process, with a focus on the sintering temperature optimization and characterization of the resulting physical, morphological, and microstructural properties of the samples. The use of platinum foil enabled the fabrication of robust and fracture-free pellets with excellent adhesion to the metal backing. Sintering was performed at temperatures of 1200, 1300, 1500 °C, with 1300 °C identified as the optimal temperature, providing the best compromise between having densification, good adhesion to Pt, and minimal material loss. The presence of the platinum foil may alter the current pathway distribution during sintering, creating a temperature gradient within the pellet. This led to enhanced densification on the platinum-facing side, as revealed by SEM, and the coexistence of cubic and monoclinic crystalline phases, as evidenced by XRD analysis. Moreover, reducing the amount of starting powder to achieve a suitable thickness for ¹⁵⁵Tb production by proton irradiation has been demonstrated not to influence the morphology and microstructure of the pellet. Low thickness pellets (Ø 10 mm, starting powder amount 87 mg) were successfully obtained without fracture. Cross-section analysis also revealed the formation of an intermetallic Gd-Pt phase, resulting from local reduction and diffusion during sintering. Overall, these promising results demonstrate the viability of this approach to manufacture a target with enriched ¹⁵⁵Gd[Gd₂O₃] powder, opening promising perspectives for future ¹⁵⁵Tb production via proton irradiation. From the general point of view, this work underlined the versatility of the SPS technique and its ability to sinter even a thin ceramic layer simultaneously attached to a metal foil.

CRediT authorship contribution statement

Gaja Piteo: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation. **Alisa Kotliarenko:** Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation. **Juan Esposito:** Writing – review & editing, Resources. **Petra Martini:** Writing – review & editing, Resources, Project administration, Funding acquisition. **Mirko Pigato:** Writing – review & editing, Formal analysis. **Lucia De Dominicis:**

Writing – review & editing, Investigation. **Gaia Pupillo**: Writing – review & editing, Project administration. **Liliana Mou**: Writing – review & editing, Resources, Project administration. **Sara Cisternino**: Writing – review & editing, Writing – original draft, Validation, Supervision, Methodology, Investigation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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