Results of INTEFF_TOTEM Project: magneTron sputtering cyclOtron TargEt Manufacturing

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INTRODUCTION

INTEFF_TOTEM was a Proof-of-Concept project in the framework of "Innovazione tecnologica dalla fisica fondamentale" (INTEFF) program promoted by INFN Technology Transfer Committee (CNTT). This project was funded by the Ministry of Economic Development (MISE) for the period from February 2021 to August 2022.

The project aim was the enhancement of the INFN patent no. WO 2019/053570 "Method for obtaining a solid target for radiopharmaceuticals production", developed within the framework of the LARAMED project at LNL. A technological R&D program to improve the deposition efficiency of some materials (e.g., Cr, ZnO) on different substrates (metal and non-metal) with Magnetron Sputtering (MS) technique was carried out. The goal was to limit, as much as technically achievable, the losses of the material to deposit, to make this technique also attractive for the expensive isotopically enriched materials. Such materials are typically used to manufacture cyclotron solid targets for the production of medical radionuclides. The initial estimated Technology Readiness Level (TRL) of this patent was 4. At the end of the project, to evaluate the developed technology some targets were irradiated with the proton beam of the medical cyclotron available at the S. Cuore Don Calabria Hospital (SCDCH) in Negrar, (VR). Thus, with the performed improvement we have achieved a TRL of 6.

Moreover, a technological development of another INFN patent held by LARAMED group (no. WO 2019/220224 A1, "A method and a target for the production of ^{67}Cu") is strictly related to this project. The first step towards the manufacturing of a ZnO multilayer target for ^{67}Cu production with a high energy cyclotron (e.g., the BEST 70p at LNL) was studied using both MS and Spark Plasma Sintering (SPS) techniques.

The INTEFF_TOTEM project was carried out in the framework of the LARAMED target development activities [1] and in collaboration with the Surface Technologies and Superconductivity Service at the LNL.

PROJECT RESULTS

This project was organized in six Work Packages (WP). A brief description of the activities and related results obtained for each WP are hereafter reported.

The first task of the project was dedicated to the development of the recovering shield for the sputtering processes which could give a solution for the main planar MS drawbacks **(WP1. New MS configuration and material recovery)**. A new grounded shield was firstly designed and investigated under the METRICS project [2]. Such shield allows to keep the plasma generated in a controlled volume and collect all the material which is usually lost during the deposition.

A first target of 70 μm ^{nat}Cr on a Nb coin was manufactured by using the recovering shield in the framework of TOTEM project. Exploiting the results obtained in the WP1 for planar MS configuration, it was possible to prepare cylindrical Cr cathode, which then was used in **WP5**.

However, the main goal of the project was the design, realization, and test of a new magnetron configuration, the so called inverted (IM) or hallow cylindrical magnetron. These tasks were carried out within the **WP2 (Inverted magnetron system)**, **WP4 (Mechanical design and realization)** and **WP5 (IM deposition tests)**. The details are described in [3].

To find out the working condition of the IM prototype, more than 30 deposition tests were performed using a Cu cylindrical cathode with the sample holder in different orientations (**realized in WP4).** As a result of **WP2,** an optimal configuration with horizontally placed sample holder was found and a 60 μm Cu deposition was carried out onto Nb backing.

Within the **WP5**, the reactive sputtering depositions of ^{nat}ZnO over Nb backing starting with Zn cylindrical target were performed. Up to now, a thickness of maximum 30 μm was reached and two ZnO targets were manufactured and tested under the proton beam. The absence of Zn contaminants in the deposits, was proved by X-Ray Diffraction analysis and more details can be found in [4].

Within the **WP3 (ZnO pellet realization with SPS)** the SPS technique was investigated as an alternative method to produce solid targets with negligible material losses. First ZnO pellets starting from ZnO nano and micro powder were realized by SPS using the TT_SINTER machine present at the INFN Pavia section [5,6]. The pellets obtained using slow heating/cooling rates in some temperatures ranges (e.g., 20 °C/min) and starting from ZnO micro powder showed good mechanical strength. Carbon contamination coming from the graphite press was unavoidable. Further studies (within the new CUPRUM_TTD CSN5-INFN project) will be performed to understand if this could be an issue for the radiochemistry dissolution step. Moreover, the first target system composed by ZnO pellet attached to Au/Nb backing was manufactured using SPS.

The targets obtained in this project using IM and SPS techniques were tested for thermo-mechanical stability in the specific environment with the SCDCH medical cyclotron within the **WP6 (Irradiation tests)**.

Sputtered targets showed no damages after the irradiation at 19 MeV with currents up to 20 µA after a visual inspection. These results demonstrated a good adhesion between the materials, that guarantees an efficient heat dissipation efficiency during the irradiation. However, the thickness of these targets was too low to obtain high ^{67}Cu yields, thus further investigations are needed to improve the IM configuration and increase the deposited thickness.

Regarding the SPS ZnO pellets, they were irradiated using a dedicated pellet-holder made of two Al parts kept together through permanent magnets (designed within the **WP4**) [5]. This device was necessary to allow the positioning of the pellet (10 mm x 600 µm) in the target station of the cyclotron. The ZnO pellets (in the Al capsule) withstand irradiation currents of 10 µA (energy 19 MeV) without damage. Further irradiation tests will be performed to assess the maximum current sustainable in this configuration.

Finally, the SPS ZnO target system (ZnO/Au/Nb) was irradiated at 19 MeV and 15 µA. The pellet was detached from the backing after its manipulation in the hot-cell, suggesting a poor adhesion between the materials. Indeed, further experiments are necessary to improve this aspect because this configuration would be preferred in respect to the pellet inserted in the holder, because (i) it simplifies the handling of the target before and after the irradiation and (ii) it allows to irradiate the ZnO at 19 MeV exploiting the best energy range for ^{67}Cu production with medical cyclotrons.

The results are shown in Fig.1.

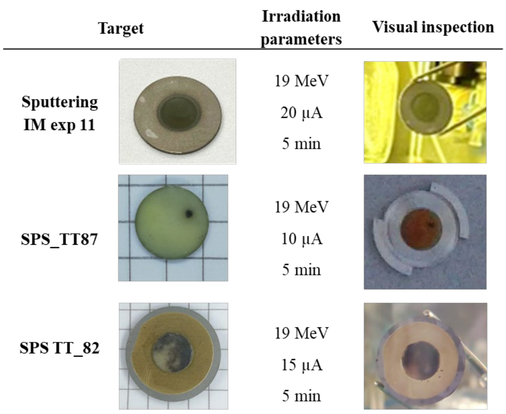

Figure 1. MS and SPS solid targets: before (left) and after (right) the irradiations under the 19 MeV proton beam of the TR19 ASCI cyclotron (Negrar, Verona, Italy).

CONCLUSIONS AND APPLICATION POTENTIALITIES

We have evaluated the performance of the new IM configuration and tested the first targets under the cyclotron beam. In this study it was not possible to calculate the data related to the material utilization amount, since the prototype is not yet optimized in terms of its dimensions. However, in comparison to the previous study [2], no visible losses in the vacuum chamber or outside of the closed working environment has been observed. The cylindrical cathode acted as a self-shield, however the deposition of compounds (as in case of ZnO) could lead to additional losses in the form of powder or flakes that may accumulate on the inactive parts of the cathode.

The preparation of the cathode for any MS configuration demands a significant material volume, unlike other techniques. Therefore, we suggest MS as a potential competitor with other techniques in case of producing multiple targets in a single deposition run, which would minimize the relative material losses. However, it is crucial to validate the ability to deposit a uniform thickness on multiple targets in a single sputtering run in the IM configuration.

Moreover, the first feasibility study on SPS sintering of ZnO for target manufacturing was also evaluated.

In general, since the first target prototypes were tested in a specific environment, the expected deliverables and milestones were reached and the final TRL estimated was 6.

It worth to note that this project has promoted the training of a young researcher through the financing of a dedicated research grant for 12 months.

The activities performed in the project were presented as oral presentations at the international conference "Workshop on targetry and target chemistry" held in August 2022 in Whistler (Canada, BC) and a dedicated paper will be submitted to a peer reviewed journal.

The first results obtained in this project, both on IM and on ZnO target manufactured by SPS method, are the ground for the new INFN project CUPRUM_TTD funded by the INFN CSN5 in 2023-2025, dedicated to the technological development aimed at the production of ^{67}Cu through medical cyclotron.

The main application field of the technology developed in the TOTEM project is the cyclotron-based production of medical radionuclides. However, the new MS configuration could be useful for other application fields, such as aerospace, automotive, fuel cells, solar panels, tube coatings, etc.

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