The REMIX Project: Second Year Research Activities

G. Pupillo[1], L. Mou[1], J. Esposito[1], L. De Dominicis[1,2], G. Sciacca[1,3], S. Cisternino[1,3], M. Campostrini[1], V. Rigato[1], P. Martini[4], A. Boschi[4], A. Duatti[4], L. Canton[5], F. Barbaro[5,6], L. De Nardo[5,7], L. Meléndez-Alafort[1,8], M. Bello[1,7], A. Fontana[9], A. Colombi[6,9], M.P. Carante[6,9], F. Groppi[10,11], S. Manenti[10,11], M. Colucci[10,11], L. Confalonieri[10,11], F.C. Bolchini[10,11], S. Bortolussi[6,9], U. Anselmi-Tamburini[6,9], E. Cazzola[12], G. Gorgoni[12], E. Nigron[13], Y. Lashko[5,14], F. Haddad[13]

[1]INFN, Laboratori Nazionali di Legnaro, Legnaro (PD), Italy; [2] Dipartimento di Fisica dell'Università di Padova, Padova, Italy; [3]Dipartimento di Ingegneria Industriale dell'Università di Padova, Padova, Italy; [4] Università di Ferrara, Ferrara, Italy, [5] INFN Sezione di Padova, Padova, Italy; [6] Dipartimento di Fisica dell'Università di Pavia, Pavia, Italy; [7] Dipartimento di Fisica e Astronomia dell'Università di Padova, Padova, Italy; [8] Istituto Oncologico Veneto IOV-IRCCS, Padova, Italy; [9] INFN Sezione di Pavia, Pavia, Italy; [10] Dipartimento di XXX dell'Università di Milano, Milano, Italy; [11] INFN Sezione di Milano, Milano, Italy; [12] Ospedale Sacro Cuore Don Calabria, Negrar (VR), Italy; [13] GIP ARRONAX, Saint-Herblain, Nantes, France; [14] Bogolyubov Institute for Theoretical Physics, Kyiv, Ukraine.

INTRODUCTION

REMIX is a three-years project funded by INFN-CSN5 for the years 2021-2023, with the goal of finding possible ^{47}Sc and medical Terbium isotopes (^{149}Tb, ^{152}Tb, ^{155}Tb and ^{161}Tb) production routes by using accelerators. All the radionuclides of interest in the REMIX project, except for the therapeutic ^{161}Tb, can be used to obtain theranostic radiopharmaceuticals, since they emit radiation suitable for both therapeutic and diagnostic purposes.

The project is carried out at the LNL, PD, PV, FE and MI sections of the INFN. It is also supported by the collaboration with the GIP ARRONAX (Saint-Herblain, France), the Sacro Cuore Don Calabria Hospital (SCDCH, Negrar, VR, Italy) and the Istituto Oncologico Veneto (IOV, Padova). It is organized in working packages (WP), as already described in a previous Annual Report 2021 [1]. This report will shortly summarize the main outcomes of the second year of the REMIX project.

TARGET MANUFACTURING AND CHARACTERIZATION

The WP1 is dedicated to the target manufacturing and characterization. Twenty targets for each isotopically enriched Ti (^{49}Ti and ^{50}Ti) were realized [2]. The procedure adopted follows these steps: the cryomilling of the powder to reduce the size up to 10 µm and HIVIPP deposition. The average thickness measured by weighing is 468±110 µg/cm^2 and 638±200 µg/cm^2 for ^{49}Ti and ^{50}Ti, respectively (Fig. 1).
The deposits were analysed with EBS using AN2000 accelerator to obtain the exact amount of Ti deposited. Three spectra for each target have been acquired and the data are under investigation. From the first results the deposits were uniform, and some targets have been used for the nuclear cross section measurements (WP2).

At the end of 2022 the first test on $^{nat}Gd_2O_3$ powder sintering have been started using the TT_Sinter machine in Pavia.

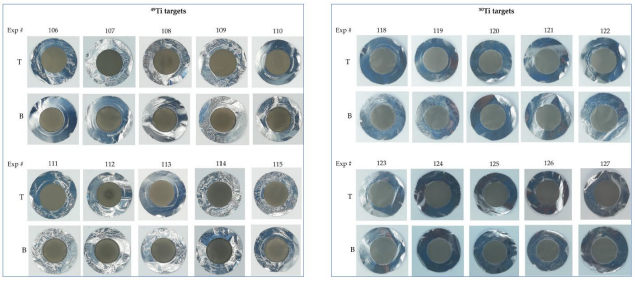

Fig. 1. Enriched ^{49}Ti (left) and ^{50}Ti (right) targets realized with the HIVIPP method for the REMIX project; "T" and "B" labels indicate "top" and "bottom" targets, respectively.

NUCLEAR CROSS SECTION MEASUREMENTS

During the second year of the REMIX project, the $^{49/50}$Ti enriched targets realized in the WP1 were irradiated by using the stacked-foils target technique (WP2). Four irradiation runs with a duration of 1.5 hours each and a constant beam intensity of about 100 nA, were performed at the GIP ARRONAX facility. Six foils of ^{49}Ti were irradiated to obtain the cross section trend of the ^{49}Ti(p,x)^{47}Sc reaction which study started the previous year, and 6 ^{50}Ti foils to start the study of the ^{50}Ti(p,x)^{47}Sc production cross section. The activation of each foil was measured by γ-spectroscopy; the data analysis is currently ongoing. Results will be shared with WP4 and WP5 to find out the best ^{47}Sc production parameters.

^{nat}Dy and ^{159}Tb thin targets are available on the market and are being irradiated with the high intensity cyclotron (AVF IBA-C70XP, K=70) of ARRONAX research center in Saint-Herblain, France, to find out the best production parameters for Tb-radionuclides (WP3), in collaboration

with WP4. The γ-spectrometry measurements will be carried out at the LASA lab. During 2022, four irradiation runs have been performed on ^{nat}Dy (two with proton and two with deuteron beams) and ^{159}Tb (two with proton beams) targets. Preliminary results are reported in [3].

NUCLEAR CROSS SECTION MODELLING

^{155}Tb cross section simulations have been performed based on recent measurements obtained with enriched ^{155}Gd-oxide target. The dosimetric impact due to residual ^{156}Gd impurity has been also determined [4]. ^{47}Sc production from enriched Ti targets has been investigated and model parameters have been optimized (WP4) based also on preliminary data measured at ARRONAX by WP2. Optimization techniques based on Genetic Algorithms have been exploited [5].

Nuclear reaction modelling has been performed also for the generator reaction ^{159}Tb(p,5n)^{155}Dy→^{155}Tb (WP4). An accurate description of the cross section has been obtained with a tailored selection of the parameters for compound, preequilibrium and direct reactions. The new curves are in good agreement with the experimental data. These results are the starting point for the calculation of activities and purities of ^{155}Tb and will allow to optimize the timings of the radiochemical separations.

DOSIMETRIC CALCULATIONS

In the framework of WP5, the absorbed dose contributions of different Sc-radioisotopes obtained by proton irradiation of ^{48}Ti target were evaluated using a DOTA-folate conjugate as an example of radiopharmaceutical product and compared to the case of the already considered ^{nat}V(p,x)^{47}Sc reaction [6]. A similar analysis has been performed for a ^{155}Tb-labeled radiopharmaceutical by considering also the contribution of Tb-contaminants co- produced by proton irradiation of Gd target with different enrichment level in ^{155}Gd [4].

TB-155 THICK TARGET YIELD

The purpose of the WP6 is to study the Thick Target Yield (TTY) of the ^{155}Gd(p,n)^{155}Tb nuclear reaction. The irradiation of the thick target (provided by WP1) will be carried out at the Sacro Cuore Don Calabria Hospital with a 19 MeV cyclotron, coherently with the 450 mb peak at around 10 MeV expected by the cross section theoretical estimations and by the recent experimental work by Dellepiane et al. [7]. Planned for 2023, preliminary experiments on natural targets will be performed for dissolution and irradiation tests. Finally, the thick $^{155}Gd_2O_3$ target will be irradiated and the produced activity of ^{155}Tb will be determined through γ-spectrometry measurements.

APPARATUS FOR CROSS SECTION MEASUREMENTS

The WP7 is dedicated to mechanical design and construction of equipment for the conduction of nuclear cross section measurements. In the waiting of LNL beamline operability, during the second year of the REMIX project various components were developed to perform experiments with the GIP ARRONAX beamline (collimator, shown in Fig. 2, and collimator-holder) and the SCDCH cyclotron (target holder for thick yield measurements). All the components were manufactured at the LNL mechanical workshop following the technical drawings produced in the framework of this WP.

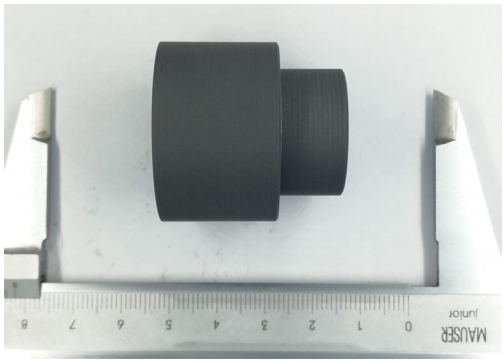

Fig. 2. Collimator in graphite designed and realized at the LNL and installed on the AX3 beamline at ARRONAX for nuclear cross section measurements for ^{47}Sc production.

RESULTS AND DISCUSSION

Results of the REMIX project during 2022 have been achieved without delay, also thanks to the solid network of collaborations and the mutual support in the team. A more detailed description of REMIX major outcomes can be found in specific LNL Annual Reports.

REFERENCES AND FINAL NOTES

Authors would like to thank the LARAMED project and the staff at the ARRONAX and SCDCH for their constant support to these research activities.

[1] G. Pupillo et al., LNL Annual Report 2021 no. 84
[2] S. Cisternino et al., LNL Annual Report 2021 no. 72, 65
[3] M. Colucci et al., This Annual Report
[4] F. Barbaro et al., This Annual Report
[5] Y. Lashko et al, et al., This Annual Report
[6] L. De Nardo et al., This Annual Report
[7] G. Dellepiane et al., Cross section measurement of terbium radioisotopes for an optimized ^{155}Tb production with an 18 MeV medical PET cyclotron, Applied Radiation and Isotopes 184 (2022) 110175, DOI: 10.1016/j.apradiso.2022.110175