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A SUSTAINABLE APPROACH TO COPPER AND GALLIUM RADIOISOTOPE PRODUCTION AND TARGET RECOVERY FROM A SINGLE IRRADIATION OF natZnO TARGET

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Aim/Introduction

Copper radioisotopes are gaining increasing relevance in nuclear medicine due to their versatile decay properties. Among them, ^{61}Cu ($t_{1/2}$ = 3.33 h, β^+= 61%) stands out as a promising PET imaging agent, offering a short half-life which is ideal for matching the pharmacokinetics of many low-molecular-weight radiopharmaceuticals. In this study, we demonstrate that ^{61}Cu can be efficiently produced starting from natural zinc oxide (^{nat}ZnO) targets via the ^{64}Zn(p,α)^{61}Cu nuclear reaction, leveraging the high natural abundance of ^{64}Zn (49.17%) [1]. When irradiating the ^{nat}ZnO targets in a medical cyclotron at 18 MeV, the co-production of gallium radioisotopes (^{66}Ga, ^{67}Ga, and ^{68}Ga) is a significant side process. To optimize resource efficiency, we introduce a sustainable protocol for the dual purification of both copper and gallium fractions from a single irradiation, coupled with a highyield recovery method for the starting ^{nat}ZnO material to allow for subsequent target recycling and processing.

Materials and Methods

^{nat}ZnO pellets (~260 mg) were produced using Spark Plasma Sintering (SPS) and attached on Au/Nb backing supports [2]. Irradiations were performed at the IRCCS Sacro Cuore Don Calabria Hospital on a TR19 cyclotron at 18 MeV, 20 μA for up to 1 hour. ^{61}Cu was isolated via an automated protocol using CU and TK201 resins. Gallium radioisotopes were then selectively recovered from the waste of the first column using a semi-automated two-stage procedure with ZR and DGA resins. Purity and recovery efficiencies were evaluated by ICPMS and γ-spectrometry. Both fractions were tested for radiolabelling with DOTA and DOTATOC Finally, ^{nat}ZnO powder was recovered from the waste through NaOH precipitation and subsequent calcination.

Results

^{nat}ZnO targets were successfully irradiated and processed through an automated separation procedure to isolate copper radionuclides from the target material and co-produced species, achieving a 95% recovery yield (n = 3) and up to 969 MBq of ^{61}Cu. Impurities of zinc, gallium, and cobalt in the copper fraction were below instrumental detection limits. From the waste of the first separation step,

containing gallium and zinc, a secondary purification enabled the isolation of gallium radionuclides with a 91% recovery yield (n = 3), yielding up to 1490 MBq of ^{66}Ga in the final fraction. Both copper and gallium fractions demonstrated excellent labelling performance with DOTA and DOTA-TOC, achieving radiochemical purities > 98% (determined by radio-HPLC) in both cases. The recovery yield of ^{nat}ZnO powder was 84.9 ± 4.7% (n=15), and it was successfully reused for new target fabrication and processing.

Conclusion

This work demonstrates a sustainable approach for the dual production of copper and gallium radioisotopes from a single ^{nat}ZnO irradiation. By integrating efficient radionuclide separation with a target material recovery protocol, this strategy minimizes radioactive and chemical waste, aligning with green chemistry principles and optimizing resource utilization.

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[2] S. Cisternino et al., Ceram. Int. 2025, 51(20), 30693–30707.