

Research Article

Target and PADC Track Detectors for Rare Isotope Studies

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A higher yield of rare isotope production methods, for example, isotope separation on-line (ISOL), is expected to be developed for the EURISOL facility. In this paper as a part of the ongoing project, high power-target assembly and passive detector inclusion are given. Theoretical calculations of several configurations were done using Monte Carlo code FLUKA aimed to produce 10^{15} fissions/s on LEU-C_x target. The proposed radioactive ion beam (RIB) production relies on a high-power (4 MW) multibody target; a complete target design is given. Additionally we explore the possibility to employ PADC passive detector as a complementary system for RIB characterization, since these already demonstrated their importance in nuclear interactions phenomenology. In fact, information and recording rare and complex reaction product or short-lived isotope detection is obtained in an integral form through latent track formation. Some technical details on track formation and PADC detector etching conditions complete this study.

1. Introduction

Astrophysical nuclear phenomena that relate to the formation process of the universe and its dynamics with the major outcome, for example, the sun, red giant stars, supernova explosions, and massive stars, among others, can be studied in a much smaller scale. Employing radioactive nuclei interacting with different targets, it is possible to reproduce the extreme physical conditions that accompany the transformation and formation of nuclear matter. Through few radioactive nuclear beam facilities [1] that have been operational for more than two decades, sufficient experimental results were gathered from which a new became evident horizon, arising in the exploration of nuclear phenomena at the extreme conditions of isospin asymmetry, including exotic nuclear behavior study. Facilities that entered operation few years ago with science programs effectively supplied an invaluable experience, providing data for further development [2, 3].

In general terms, facilities with radioactive ion beam production rely mainly on two complementary techniques, namely: in-flight and isotope separation on-line (ISOL). The first type of beam is designed primarily to produce very short-lived radioisotope beams at research centers such as

FAIR (Germany), RIKEN (Japan), and RIBF (USA). The other type is meant to provide high radioactive ion beams (RIBs) at ISAC-TRIUMF (Canada), SPIRAL-2 (France), SPES (Italy), and ISOLDE at CERN. Targets in ISOL applications should operate at high temperature to promote the diffusion of the gas atoms of radioactive isotopes produced in the bulk of the target [4]. So far, ISOL targets have been employed for decades, and today around 3000 radioisotopes are known, and beams of 70 elements have been produced in the energy range, 40 MeV/A up to 1.4 GeV/A. Up-to-date facilities having the capacity to produce intense beams of unstable nuclei will open new fields of nuclear physics research including short-lived alkali and rare earth active isotopes. To cope with these objectives, advanced experimental techniques are required to provide a new experimental platform. The existing ISOL facilities producing low-intensity beams of short-lived near-dripline nuclei suffer from the drawback of having as obstacle the diffusion and effusion time. To explore further the exotic regions and discover unexpected phenomena at neutron dripline, an improvement (at least 100 times better than the existing ones) on beam production is required. The new generation of RIB facilities is conceived in the multi-MW EURISOL-DS

facility (European isotope separation on-line radioactive ion beam facility) aim to reach 10^{15} fissions/s. To reach this goal, also a hybrid technique, relying on a combination of fragmentation, gas stopping, ionization, and reacceleration, has been proposed. It is expected that new exotic beam facilities, in which radioactive nuclides produced by spallation, fission, or fragmentation reactions are, will enter into operation together with new and more sensitive detection techniques. The target geometry and material in this case should be designed to provide sufficient luminosity considering the technical requirements, the spatial assembly, and safety constraints related to the production of intense radioactive nuclear beams (RNB).

To improve the EURISOL detection capability, a complementary system is proposed, since a large set of experimental results have been obtained with passive polyallyldiglycol-carbonate, PADC for short, or $(C_{12}H_{18}O_7)$ detectors in nuclear interaction studies by Barbui et al. [5]. The technique demonstrated their importance in several fields, for example, in galactic cosmic ray (GCR) and studies on the ISS, Pálfalvi et al. [6]. In the next sections, some technical details are given on a new target for the Eurisol facility that will be available in the near future. The last section is devoted to the passive detector applications.

2. Fission Target System

A new fission target for EURISOL-DS is presented with a description of its geometry and technical characteristics [7]. The definition of the most specific characteristics of the target is based on the results obtained with Monte Carlo code: FLUKA [8]. The calculations consider the entire system (fission target, neutron converter, reflector, extraction beam lines, shielding, experimental rooms, etc.), aimed to produce 10^{15} fissions/s on a multiple-body target. In Eurisol-DS, neutrons inducing the fission reactions are provided by a proton beam of 1 GeV and up to 4 mA impinging on a mercury converter. The multibody target (six individual and independent target modules), gives the best ratio fission to neutrons produced by the Hg converter (Figure 1).

2.1. Design. The fissile material best geometry is a set of piled-up discs of uranium carbide enriched with 2% enrichment of ^{235}U (or low-enrichment uranium LEU- C_x). This resulted as the outcome after several geometries explored in order to define the shape and composition of uranium target, taking into account the mechanical and space constraints (Figure 2) [9].

So far, LEU- C_x targets envisioned for EURISOL did undergo preliminary test at the IRIS facility of PNPI with pulsed protons from the Gatchina synchrocyclotron (with $0.1\ \mu\text{A}$, 1 GeV) producing fragmentation, fission and spallation reactions. During these tests, some neutron-rich isotopes from a solid tantalum converter have been produced [10]. Similar UC_x target is under the R & D program at radioactive ion beam facilities so that for the EURISOL-DS target design it was possible to rely on experimental data which were advantageously included [11]. The resulting

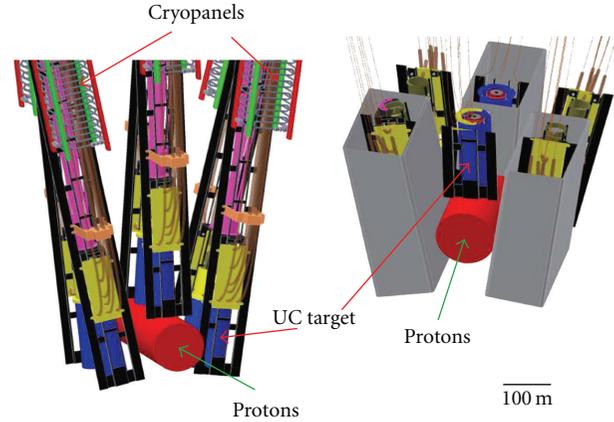


FIGURE 1: The multibody LEU- C_x target schematic layout showing details on its geometry. On the left, the six-module tilted target system is given; the other shows the squared external container (in gray) [8].

Eurisol multibody fission target geometry has a cylindrical tantalum container with 86 UC discs; its external diameter OD = 35 mm and internal diameter ID = 8 mm and of 1 mm thickness, each corresponding to a volume of $1.55\ \text{cm}^3$; the whole system is water cooled and insulated from the ground by beryllium oxide insulator. The entire system is enclosed in a concrete shielding. Systematic calculations indicated that inclining the extraction lines resulted in the best arrangement, favoring the increase in the fission yield due to minimizing the distance between mercury converter and each individual target module. Moreover the mercury converter is surrounded by a graphite reflector to slow down the neutron energy maximizing the neutron flux on the fission targets. The shape of the containers was also studied, evaluating cylindrical and squared containers. A schematic layout is given in Figure 1.

The most interesting results obtained are related to the total energy deposited on targets. The configuration including a squared full-target container with 1 cm of water coat and without using additional moderator is sufficient to keep the system in temperature-controlled steady state. In all cases, the total energy deposited was lower for the squared containers without using additional moderator material compared to other materials.

2.2. Materials' Selection. Due to radiation dose determination, around the mercury converter, beryllium oxide and graphite as a reflector was devised to weaken the neutrons flux; it is expected that these neutral particles leaving the region around the fission target could reach levels outside the security-guideline-recommended values. Calculations show the highest target fission yield using beryllium oxide reflector reaching up to $6.5 \cdot 10^{14}$ fissions/mA; that is, the number of fissions will be beyond the main designed operational parameter when the beam current reaches 4 mA on target. However, even reaching the main objective of producing 10^{15} fissions/s, the high toxicity of beryllium oxide suggests to evaluate the convenience of using graphite. In fact, similar

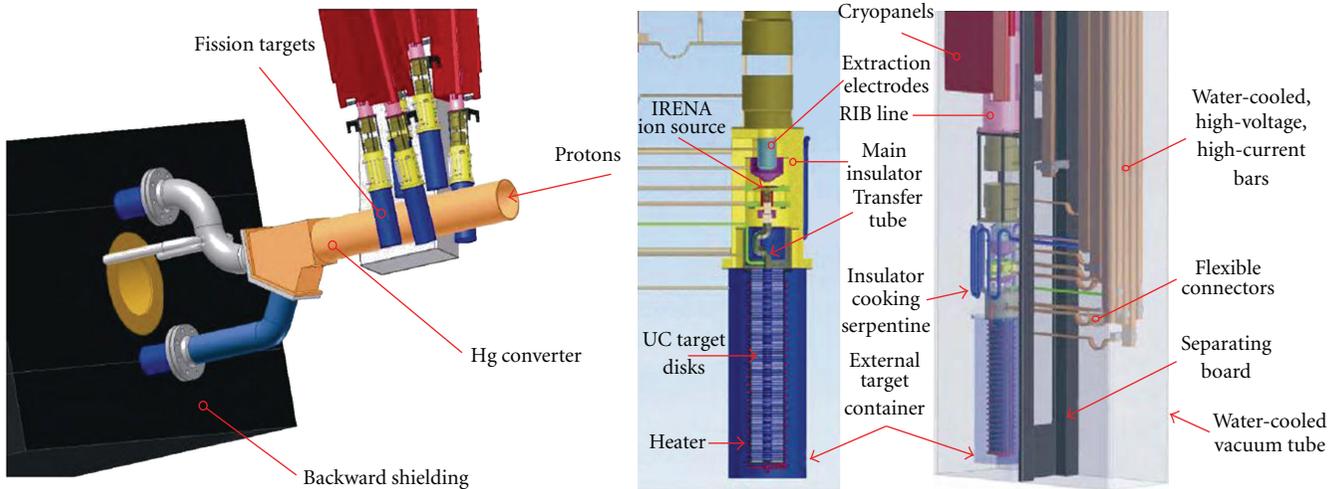


FIGURE 2: Mechanical and material geometry in the final target setup. On the left are the target details.

yields are predicted by simulation using graphite, inducing considering it as the most convenient reflector material. The additional advantage is the graphite commercial availability and its low cost being a priority during material selection. For both materials, a considerable amount of energy is absorbed by the reflector too, mainly due to charged particles escaping from the multi-MW converter. A reflector made of graphite has another advantage: total energy calculated on this reflector substance was 128 kW/mA, using just one target: such result suggests an improvement when it comes to heat dissipation.

On the optimization of the general system, that is, dimensions and geometry of reflector and moderator, the following observations are drawn.

- (1) Beryllium oxide as reflector gives the higher fission rate for all materials considered.
- (2) The second best target fission yield was obtained using graphite as a reflector.
- (3) The fission yield, obtained using air instead of water in the region defined for moderator material, is comparable with the fission yield using water, with the advantage of reduced risk related to high-voltage electric discharge.
- (4) Beryllium oxide is the best reflector material, but graphite was selected for showing similar performance and being a more practical solution and much less expensive.
- (5) The study shows that the fission yield is not affected by the use of the moderator material, consequently suggest to remove the moderator from the design. On the other hand, the results show that the reflector material plays a relevant role to reach the high fission rate and the safety conditions required for the Eurisol facility.

Additionally, from results it could be observed that the best target performance is reached with a squared *full-target*

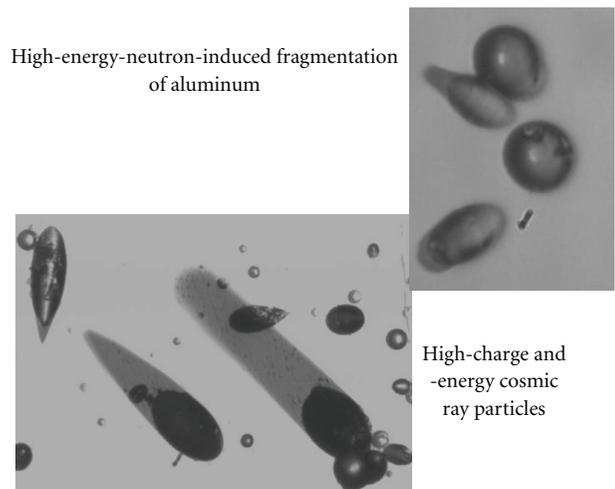


FIGURE 3: Picture of PADC detector after 15 hours etching in 6 N NaOH aqueous solution at 70°C developing the latent tracks of different charge particles (TASTRAK, TASL Co. Ltd., Bristol, UK).

container, due to the availability of a higher volume. This geometry in particular allows to assemble the target components and all the services in two separated regions. Furthermore, in order to achieve the best geometry for beam extraction, the *target container* is tilted few degrees, respect to the center line as shown in Figure 1 (on the right). A slotted support holds 86 discs in a carousel of 200 mm height and 35 mm diameter with graphite separator in between since LEU-C_x target dwelling in tantalum and graphite container offered a configuration with the best performance.

As mentioned, several materials functioning as reflector were evaluated for the multitarget system, and the energy deposited on reflector as well as the energy deposited in the supporting parts was simulated to provide temperature gradient and material thermal stress. The resulting values indicate that eliminating the reflector material and using

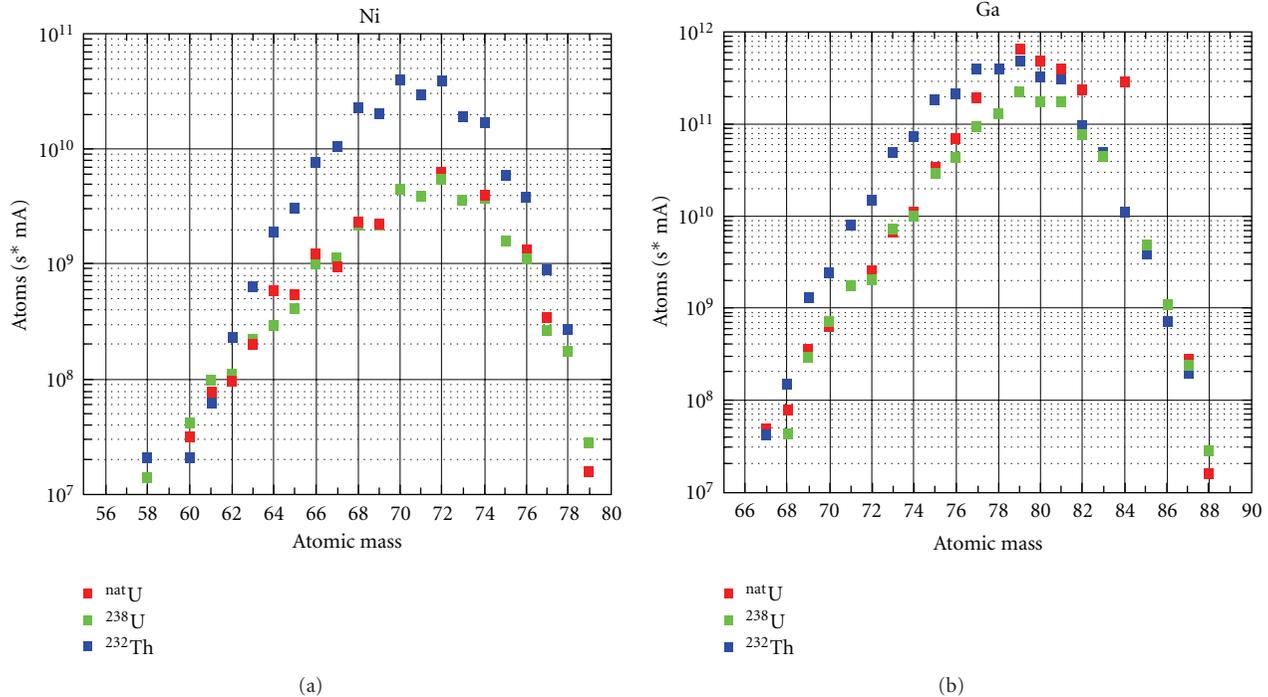


FIGURE 4: Nickel and gallium isotope production yields calculated for ^{235}U , ^{238}U and ^{232}Th EURISOL-type target materials. Ref.: CERN-BE-Note-2010-002.

only concrete shielding, the total energy released on such region is higher than using graphite as reflector. Furthermore it was determined that the specific power of $5 \text{ kW/cm}^3/\text{mA}$ observed at the center of the target varies with a gradient of 5 times higher at 25 mm of distance on a radial path.

2.3. Safety Considerations. Restriction on uranium enrichment imposed to employ LEU- C_x targets limiting it at 2%; however, several other enrichments were considered to determine its influence on the whole system oriented to produce the highest fission rate. No relevant differences were observed, and as expected, results disclosed lower contribution on ^{238}U to the fission rate. The radiation-induced damage in structural and fuel material has also been considered; an evaluating program (Target Prototypes Irradiation) is in progress for EURISOL (TARPIPE) at Injector 1 of the Paul Scherrer Institute. During the preliminary tests, samples of metal, carbide, and oxide compounds were irradiated in order to reach several displacements per atom (dpa) corresponding to 3 weeks of target operation at nominal EURISOL parameters. Visual and microscopic observation of the material before and after irradiation allows assessing sintering and irradiation effects at the operational temperatures. Concerning the fission product diffusion new sub micrometric and nanometric target materials are under development, to evaluate the material high-temperature stability and performance under intense irradiation. Another problem addressed is the radioactive waste disposal produced onto the target, the liquid Hg converter target, and its auxiliary systems, which as estimated will reach

approximately 10^9 GBq. This implies that specification of *hot* material handling and prevention of release accidents should be comparable to those applied in nuclear power industry. At the end of EURISOL operations, the irradiated liquid mercury (~ 20 tons) has to be treated as highly radioactive waste. In the case of volatile radioactive species, a cryotrap system was already tested to freeze gaseous radioactivity in a closed environment.

3. PADC: A Complementary Solid State Nuclear Track Detector

The PADC detectors have been a matter of interest in the few last decades. They can detect many charged particles having a linear energy transfer (LET) above a certain threshold and provide information about their energy and charge. The particles by ionization produce damage zones called latent tracks in the detector material of few nm in diameter which can be blown up by chemical treatment to be visible through optical microscope as seen in Figure 3. The ionization-specific density along the particle path follows the Bragg curve, providing information on the incident nuclei through etched tracks [12].

Depending on the chemical treatment (solvent composition, concentration, temperature, and time), the LET threshold can be varied in a large range from about $5 \text{ keV}/\mu\text{m}$ making it possible to selectively analyze the particle tracks. This phenomenon supports the initiative that PADC detectors could be advantageously employed also to detect and identify rare radioactive isotopes and their reaction products in the

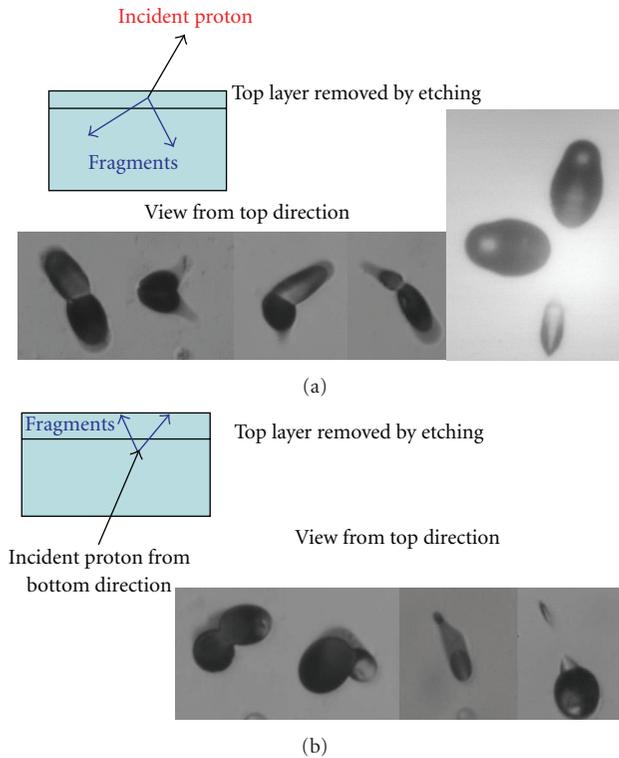


FIGURE 5: (a) Etched particle tracks originated from one fragmentation process releasing two or three particles from the point of the reaction into different directions (incident particle from the top). (b) Etched particle tracks originated from one fragmentation process releasing two or three particles from the point of the reaction into different directions (incident particle from the bottom).

frame of the ongoing EURISOL project. In EURISOL-target, the product yields have typical values as given in Table 1, with a Gaussian isotope distribution as per in Figure 4.

Illustrations of a simple high-energy-induced fragmentation of the constituents (Carbon and Oxygen) of the PADC detector material are given in Figures 5(a) and 5(b). The number and shape of the released particle tracks, as seen from the top view direction of the detector, depend on the original direction of the incident proton and the removed layer.

The mechanism of etched track evolution has been described elsewhere by Somogyi [12], and here we recall it briefly through a schematic representation given in Figure 6 on which the observed etched tracks appear of different size (diameter), shape, and position on the surface, revealing the complexity of the PADC's detector analysis.

Following Figure 6 (lower right), we consider an impinging charged particle " n " leaving a latent track shown with broken line. At the point "O", a complex reaction occurs and the emitted charged fragments take the assumed direction α , 1, 2, and 3, producing by energy loss a latent track again. Depending on the reaction origin "O", two or more track cone shapes arise during chemical etching as shown at lower left side of Figure 6, cone walls might be concave or convex depending on the etching side.

Sajó-Bohus et al. 2005 [13] developed a multietching approach to establish the correspondence of tracks to incident particles or to identify rare isotopes. At each etching step, track information leads to LET parameter determination; this method could be employed in addressing important question on radioactive beam characterization, quality, yield, decay rate, and identification.

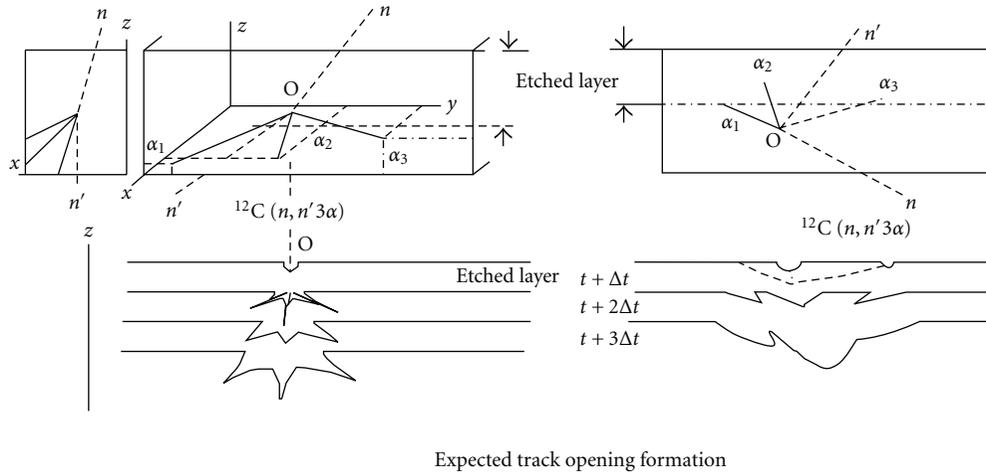
During the past years, we collected a set of tracks induced by large variety of charged particles including high-charge and-energy (HZE) cosmic rays and its interpretation with the purpose to form a large database related to particle identification. A set of pictures of similar tracks form a "gallery" that is interpreted on PADC's tracks obtained by calibration; these were obtained exposing different sets of detectors to high-energy neutron reference fields, low and high Z accelerated ions. A sizable set of PADCs were exposed inside the Service Module Zvezda on the ISS during different missions to further extend the detection capability to include complex high-energy reaction products. In order to deduce their mass, energy, and range, the LET was carefully determined and was between 7 and 1000 keV/ μm . Track analysis was made by an improved version of the VIRGINIA image analyzer, developed by the Research Institute for Technical Physics and Material Science [14] of the Hungarian Academy of Sciences.

This program enables the operator to verify the acceptance of a track as a member of a specified "class" based on measurable track parameters as shape, eccentricity, areas or perimeter, having the option to introduce other selective parameters for track classification which enables particle identification following Yadav [15]. Further, Sajó-Bohus et al. [13], devised a procedure to reduce uncertainty in particles identification of registered tracks originated from a mixed field.

For instance, the following etched tracks of cosmic ray origin can be identified by PADC.

- (i) Protons which can be directly detected if their energy is below ~ 20 MeV.
- (ii) High-energy neutrons can be observed through p , C, and O recoils and fragmentation products as α and p , d and t together with scattered ions (e.g., Be).
- (iii) Target fragments of short range, mostly induced by high-energy protons within the detector material.
- (iv) Low Z particles (e.g., C, O, Ne, Mg, Si) of Galactic Cosmic Rays (GCR).
- (v) Fe-group and ultrahigh atomic number and energetic (UHZE) particles: these penetrate several detector sheets leaving well-defined tracks; their identification, for a selected energy range, is possible following Günther et al. [16].
- (vi) Projectile GCR fragmentation; these reaction products have high energy and LET and induce tracks that are of nearly cylindrical shape.

An interesting result is given by recoil of C and O target atoms; they were discriminated since analyzed tracks of minor axis values follow a kind of saddle distribution where the first peak is around $7.2 \mu\text{m}$ and FWHM of $1 \mu\text{m}$, and the



Expected track opening formation

FIGURE 6: Schematic track evolution geometry of the three alpha particles after different etching times. Compare the left sketch to the picture of the three alpha tracks shown on the right side of Figure 5(a).

TABLE 1: In-target production yield for typical nuclei and delivered intensities.

Nuclide	In-target [atoms/s]	Release eff [%]	Ionization eff [%]	Rel. Intensity [ions/s]
^{74}Ni	$2.2 \cdot 10^9$	20	7.8	$3.5 \cdot 10^7$
^{81}Ga	$1.0 \cdot 10^{11}$	10	2	$3.8 \cdot 10^9$
^{90}Kr	$4.8 \cdot 10^{13}$	90	80	$3.5 \cdot 10^{13}$
^{132}Sn	$7.2 \cdot 10^{12}$	70	9	$4.5 \cdot 10^{11}$

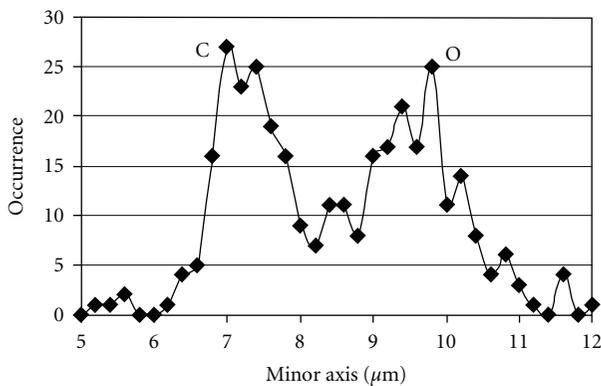


FIGURE 7: Minor axis distribution of recoil Carbon and Oxygen atoms induced by 14 MeV neutrons within the PADC material.

other peak is positioned around $9.7 \mu\text{m}$ and width of $1.3 \mu\text{m}$ as seen in Figure 7.

This is a clear example that shows mass discrimination even considering the track diameter dispersion (for a given energy) since the overlapping tails are less than 5%. This opens the door for further extending this method to higher atomic masses well above $A > 30$, being a region of particular interest in astrophysics or particle physics. It was shown by Kodaira et al. [17] that the technique could be employed to discriminate isotopes of iron with a good resolution. The gap ΔAZ^2 between adjacent isotopes is determined by the relation: $\Delta AZ^2 = [AZ^2 - (A - 1)Z^2]/AZ^2$. The mass distribution is given in Figure 8.

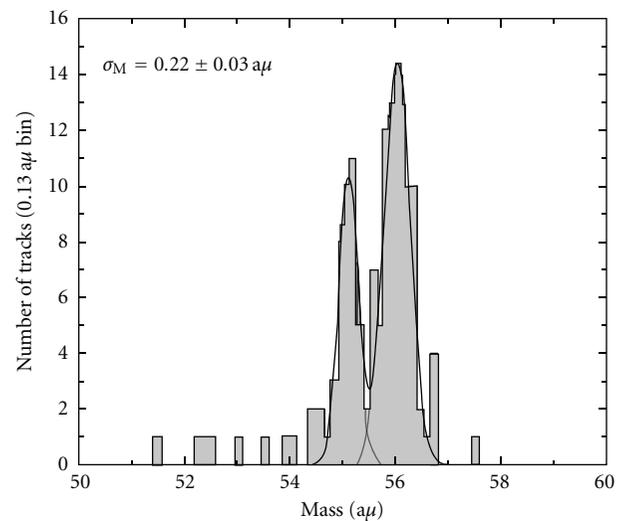


FIGURE 8: The mass distribution of iron isotopes as deduced from track analysis. Adapted from Kodaira et al. [17].

4. Conclusions

The target system for EURIDOS-DS project is a multibody configuration to produce 10^{15} fissions/s on LEU-C_x material; details on moderator reflector and radiation shielding were designed on values obtained by simulation with FLUKA code. Several geometric configurations were taken into account considering the reaction cross-section to select materials for the final system. A preliminary assembly of

the projected target was tested obtaining information on radioisotope type, yield, and radiation damage. These results are under scrutiny to further improve the target system. Having an extensive experience with passive detectors, it was suggested to employ them for beam quality control, to detect radioactive beam or nuclei with short half-life ($t_{1/2} < 1$ s) with particular emphasis on nuclei near the drip line or those with strong isospin asymmetry.

The mentioned target system and the passive detector system are given as a contribution to the EURISOL-DS project.

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