

## RADIATION SAFETY WITH HIGH POWER OPERATION OF EURISOL

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*The European Community has launched the design study for a next generation RIB facility able to increase, by a few orders of magnitude, the exotic beam intensity and availability in Europe. Forty institutes and laboratories within Europe, North America and Asia are taking part in this consortium, named EURISOL DS project (European Isotope Separation On Line Design Study). In EURISOL, four target stations are foreseen, three direct targets of approximately 100 kW of beam power and one multi-MW (MMW) target assembly, all driven by a high-power particle accelerator. In this MMW station, high-intensity RIBs of neutron-rich isotopes will be obtained by inducing fission in several actinide targets surrounding a liquid metal spallation neutron source.*

*The envisaged increase in RIB intensities at EURISOL means a drastic increase of the radioactive inventory and corresponding radioprotection related issues. Safety aspects of the future RIB production targets (aiming at a few  $\sim 10^{15}$  fissions/s) will become decisive in limiting the beam intensities, in selecting the production method and materials, and in the final cost of the facility. New technical challenges arise that will in most cases also critically affect the safety approval procedures. The handling and disposal of open radioactive high intensity RIB production targets (e.g. UCx, ThCx) including liquid Hg converter target is yet unexplored. Equally, containment of gaseous radioactivity and its migration will be crucial in the same context.*

*The progress made so far on most of these issues within Task 2 “multi-MW target” and Task 5 “Safety and Radioprotection” will be summarized.*

### I. INTRODUCTION

The EURISOL DS project<sup>1</sup> aims at a design study of the ‘next-generation’ Radioactive Ion Beam (RIB) facility based on the ISOL method, which will extend and amplify, beyond the year 2015, the work presently being carried out at the first generation RIB facilities world wide, in the fields of nuclear physics, nuclear astrophysics and fundamental interactions. The scientific case for EURISOL includes<sup>2</sup>: (a) the study of atomic nuclei under extreme and so-far unexplored conditions of composition, rotational angular velocity (or spin), density and temperature; (b) the investigation of the nucleosynthesis

of heavy elements in the Universe, an important part of nuclear astrophysics; (c) study the properties of the fundamental interactions governing the Universe, and in particular of the violation of some of their symmetries; (d) potential applications of RIBs in solid-state physics and in nuclear medicine. These cases require a ‘next generation’ infrastructure such as the proposed EURISOL facility, with intensities several orders of magnitude higher than those presently available or planned, allowing the study of hitherto completely unexplored regions of the Chart of the Nuclei.

The main components of the proposed facility are<sup>1</sup>: a driver accelerator, a target/ion-source assembly, a mass-selection system, and a post-accelerator. As shown in Fig. 1, the proposed ISOL facility would use (a) three 100 kW proton beams on thick solid targets to produce RIBs directly via spallation-fragmentation, and (b) a liquid metal 4 MW proton-to-neutron converter, similar to an intense spallation neutron source such as SNS<sup>3</sup>, to generate high neutron fluxes, which would then produce RIBs by fission in secondary actinide targets. An alternative windowless liquid mercury-jet ‘converter’ target to generate the neutrons was also proposed for this multi-MW target station<sup>4,5</sup>.

The purpose of this article is two fold:

- a) Summarizing the work carried out within Task 2, with special attention to the coupled neutronics of the mercury proton-to-neutron converter and the fission targets. The overall performance of the system, which will sustain fast neutron fluxes of the order of  $10^{15}$  n/cm<sup>2</sup>/s, is evaluated, together with the production of radio-nuclides in the actinide targets, showing that the targeted  $10^{15}$  fissions/s can be achieved;
- b) Equally, presenting the progress made on safety and radioprotection (Task 5). Some particular emphasis will be given to the shielding and activation studies of the Multi Megawatt target (MMW, liquid Hg surrounded by UCx). In this context, the benchmarking utility of physics models within high energy Monte Carlo transport codes will be underlined.

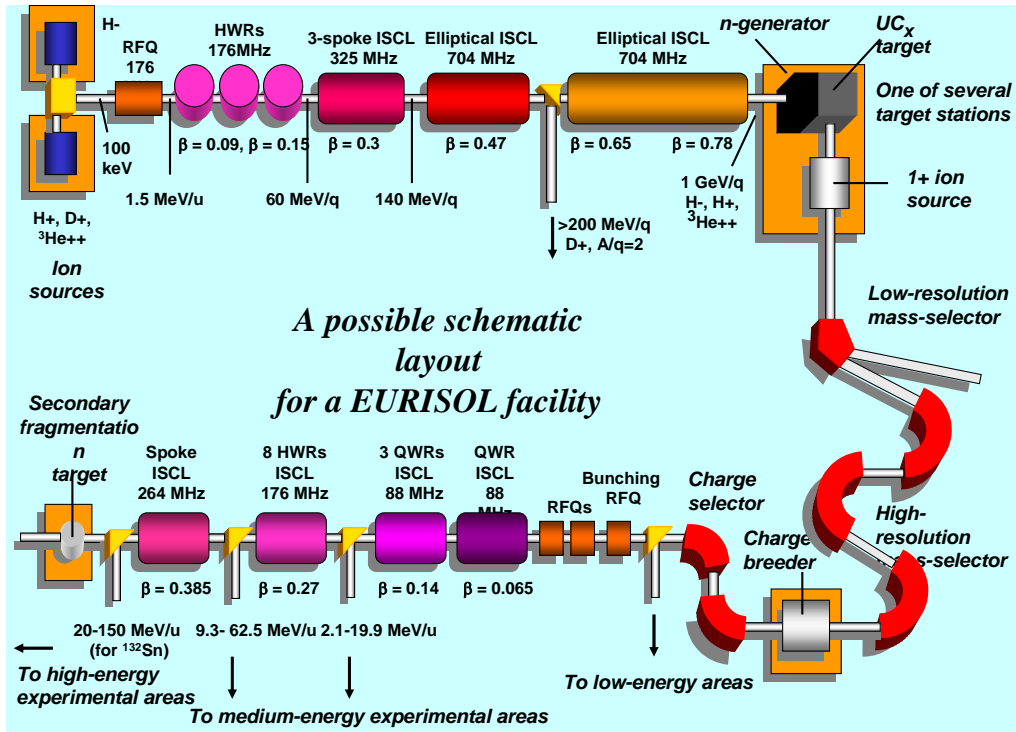


Fig. 1. EURISOL DS schematic layout, presenting the multi-MW target station (three direct targets are not shown in this picture). Source: [www.eurisol.org](http://www.eurisol.org) (November 2007).

## II. NEUTRONIC DESIGN OF THE MMW TARGET

Following the results from the earlier studies<sup>4,5</sup>, performed using the Monte Carlo particle transport code FLUKA<sup>6</sup>, a baseline design was defined. In order to maximise the neutron production, favour a fast-neutron spectrum and confine the charged particles inside the assembly, a 8 cm radius 40 cm long mercury proton-to-neutron converter was proposed, surrounded by fission targets and, possibly, by a neutron reflector (Figure 2). For the latter, beryllium-oxide (BeO) was proposed due to the high albedo of this material and to produce <sup>6</sup>He via (n,α) reactions in <sup>9</sup>Be for neutrino physics (β-beams)<sup>1</sup>.

The neutron flux distribution in the baseline design is rather isotropic a few cm away from the centre of maximum production (from 0 to 10 cm from the impact point), as elaborated in Ref. 4. The flux in the fission target is  $\sim 10^{14}$  n/cm<sup>2</sup>/s per MW of primary beam, similar to those of conventional nuclear reactors. These flux levels are more than sufficient to produce the aimed  $\sim 10^{15}$  fissions/s with reasonable fission target volumes and using an acceptable beam power.

Several calculations were carried out to assess the performance of fission target materials for the baseline design. The use of uranium-carbide was analysed and compared to thorium-oxide, for the same target densities, the latter producing one order of magnitude lower fission rates.

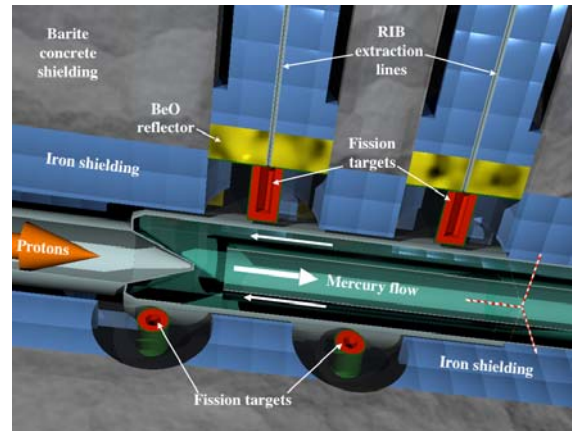


Fig. 2. Schematic representation of the newest baseline MMW target design.

To increase the release efficiency of the fission target by enhancing diffusion and effusion, porous graphite-based fission targets were proposed, similar to those foreseen for the Munich Fission Fragment Accelerator<sup>7</sup> (MAFF). Table 1 summarizes the results of the analysis, for a 105 cm<sup>3</sup>, 1.6 g/cm<sup>3</sup> target, with 8 g of fissile material for 160 g of graphite and 4 MW of proton beam.

The detailed isotopic distribution of the fission fragments may be observed in Fig. 3, allowing the prediction of in-target RIB intensities for specific isotopes. These distributions show the nature of the isotopes produced by fission: these lie on the unstable,

neutron-rich area of the chart of nuclides ( $\beta^-$  emitters), ranging from manganese to terbium. The use of depleted uranium carbide or thorium oxide entails a reduction in the production of asymmetric fission fragments ( $32 < Z < 42$  and  $50 < Z < 58$ ); thus, the presence of  $^{235}\text{U}$  is advantageous for the production of elements such as krypton or tin, major references in the physics case for EURISOL<sup>2</sup>.

TABLE I. Fission rates for the MAFF targets (fiss/s).

Reflector material	Graphite + Nat. U	Graphite + $^{235}\text{U}$	Graphite + $^{232}\text{Th}$
Vacuum	$1.4 \times 10^{12}$	$3.2 \times 10^{13}$	$4.6 \times 10^{11}$
Water	$3.3 \times 10^{12}$	$2.3 \times 10^{14}$	$4.4 \times 10^{11}$
Be-Oxide	$1.7 \times 10^{12}$	$6.1 \times 10^{13}$	$4.4 \times 10^{11}$
Iron	$1.5 \times 10^{12}$	$3.7 \times 10^{13}$	$4.7 \times 10^{11}$

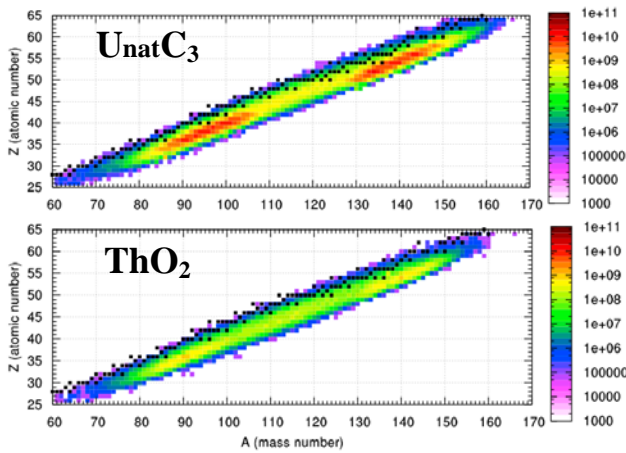


Fig. 3 In-target fission fragment distribution (nuclei/cm<sup>3</sup>/s per MW of primary beam) as a function of atomic number (Z) and mass number (A), for two actinide targets. Stable isotopes are represented by black squares.

Moreover, a relevant benefit of the large fission densities in uranium-carbides is the possibility to investigate the lower end of the so-called *terra incognita*, neutron-rich isotopes of neodymium and above (e.g.  $^{157}\text{Nd}$ ,  $^{159}\text{Pm}$ ,  $^{162}\text{Sm}$ ,  $^{163}\text{Eu}$ ,  $^{166}\text{Gd}$ ,  $^{167}\text{Tb}$  etc.), hitherto unexplored.

### III. ENGINEERING DESIGN AND INTEGRATION OF THE MMW TARGET STATION

A key parameter in the design of the MMW target is the power distribution, determining the maximum beam intensity that the system can withstand, which is in turn correlated with the fission rates. A detailed engineering study, including the design of the liquid metal flow and beam window is presented in Ref. 8.

With the present configuration, the maximum power density reaches  $\sim 2 \text{ kW/cm}^2/\text{MW}$  of beam, as presented in Figure 3. This corresponds to a mercury temperature of

180 °C, well below the boiling point. Concerning the beam window, by optimizing its thickness and the liquid metal flow, the maximum Von-Mises stress was reduced to  $\sim 135 \text{ MPa}$  and the maximum temperature to 200 °C.

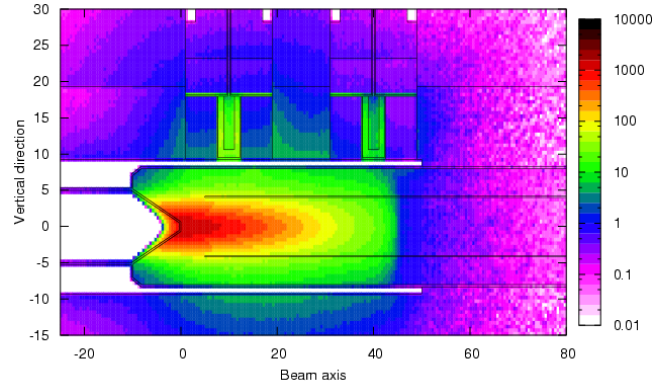


Fig. 3. Energy deposition ( $\text{W/cm}^3/\text{MW}$  of proton beam) in the MMW target assembly.

Due to the extreme operating conditions of the beam window in terms of mechanical stresses and radiation damage, a windowless transverse film target was also developed<sup>8</sup>. This design would allow for larger power densities in the liquid metal without the handicap of an aging beam window to be replaced every few months. The experimental validation of the concept is being carried out using a mercury loop the Institute of Physics at the University of Latvia. The film behavior and flow stability seem compatible with the EURISOL requirements, although further experiments are required to test nominal flow rates.

The different elements of the MMW target assembly have been integrated in a compact layout, schematized in Fig. 2. Six fission targets are foreseen, coupled with independent ion sources and vertical RIB extraction lines. These extraction lines will be merged outside the bunker and before the isotopic separator to provide very intense RIBs.

The proton-to-neutron converter will be placed in a rail system to allow for simple removal and insertion, and to damp possible vibrations induced by a pulsed proton beam. The fission targets and their auxiliary elements will be inserted and removed from the top of the bunker. The assembly will be surrounded by iron shielding and several meters of concrete to minimize particle leakage. The next chapter presents the results of the detailed shielding and activation studies performed within Task 5 of EURISOL DS.

### IV. SHIELDING AND ACTIVATION STUDIES

The shielding calculations were done using various multi-particle reaction and transport codes as FLUKA<sup>6</sup>, MCNPX<sup>9</sup> and PHITS<sup>10</sup>. For activation studies the

CINDER'90 material evolution code<sup>11</sup> was employed together with MCNPX. As a rule, for each particular observable some code benchmarking was performed in order to assess the predictive power of the physics models and evaluate uncertainties of the design parameters.

#### IV.A. Benchmarking of the transport codes

Since most of the benchmarks were already reported in Ref. 12, here we only illustrate the importance of the code validation procedure depending on the observables of interest. For example, neutron production is best reproduced using the INCL4+ABLA model combination (Fig. 4, red and blue histograms). The same is valid for residual nuclei yields (Fig. 5, red histogram). On the other hand, in order to predict correctly the tritium production at high energies one has to use the ISABEL+RAL model combination (Fig. 6, violet line).

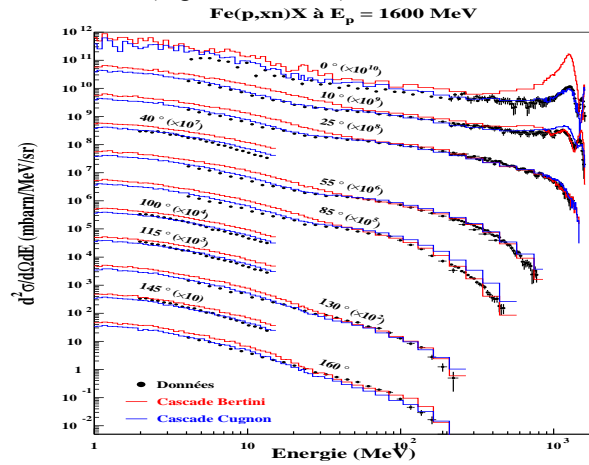


Fig. 4. Double differential cross sections of neutron production from p(1.6GeV)+Fe: data are compared with 2 different physics model combinations within MCNPX. 1GeV p+Pb

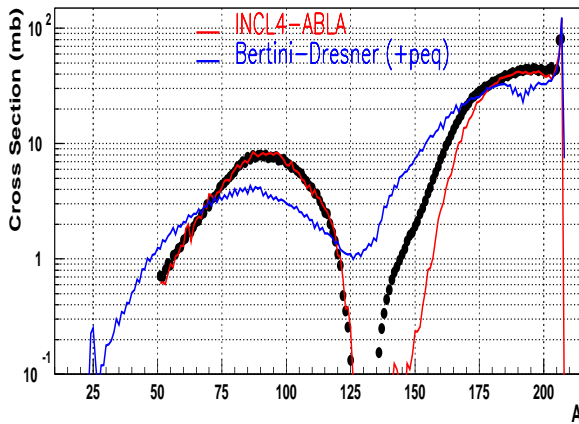


Fig. 5. Production cross section of spallation residual nuclei from Pb(1GeV/u)+H: data are compared with MCNPX code predictions.

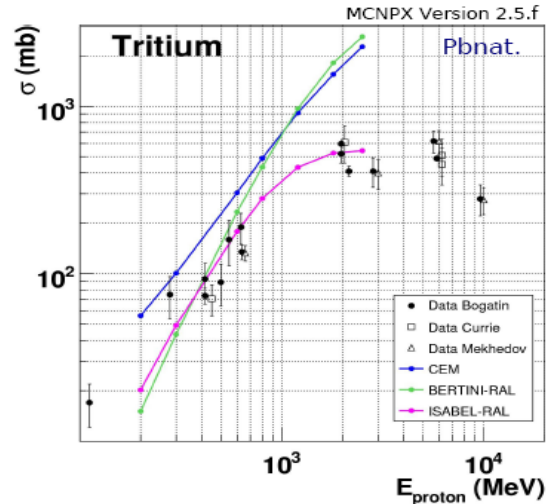


Fig. 6. Tritium production cross section from p( $E_p$ )+Pb: data are compared with MCNPX code predictions.

#### IV.B. Shielding of the MMW Target Station

For the preliminary shielding design, the reference MMW target station presented in Section II was used as a source. Both steel and ordinary concrete (2.2g/cm<sup>3</sup>) were combined as materials to protect against the prompt radiation at full power, namely 4MW of 1GeV proton beam interacting with the liquid Hg target, surrounded by a fission target. The aim in this case is to ensure a dose rate lower than 1μSv/h outside the shielding structures. We note that the preliminary shielding design was already reported in Ref. 13, where FLUKA was used for modelling. Here we repeated nearly identical calculations using the MCNPX code in order to cross-check the final results. Both weighted-window and exponential transform methods were tried as variance reduction techniques. Our results are summarized in Fig. 7, where two-dimensional dose rate map is plotted. After making the projections at forward and perpendicular directions, detailed dose rate attenuation curves are obtained as a function of the shield thickness (see Fig. 8).

In brief, our MCNPX calculations confirm the FLUKA results: after ~2 m thick steel blocks one still has to add ~9 m and ~8 m thick concrete walls at 0° and 90° respectively in order to ensure the dose rates below 1μSv/h. At the backward direction the necessary thickness of the concrete is ~6 m.

In order to minimize the shielding costs, some of the concrete could be replaced by earth layers. For this purpose detailed earth and ground water activation calculations are being performed, including the ground water transport modeling (to be reported elsewhere in the near future).



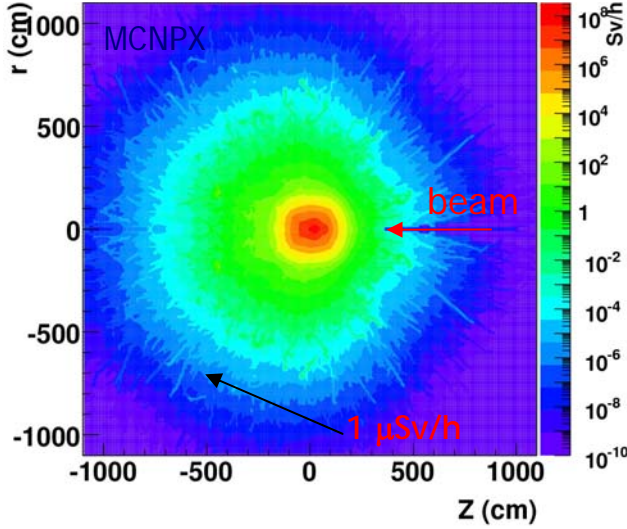


Fig. 7. Two dimensional dose rate map for the shielding design of the MMW target station of EURISOL.

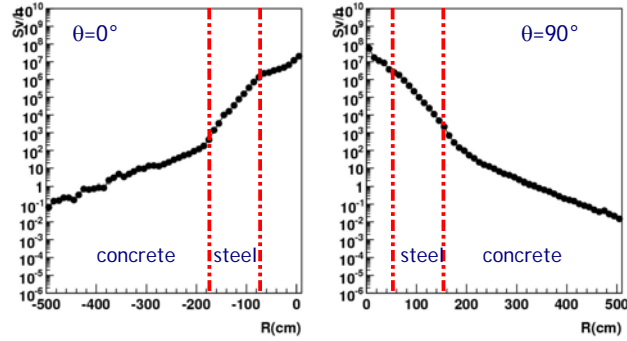


Fig. 8. Forward (on the left) and perpendicular (on the right) projections of the dose rate map presented in Fig. 7.

#### IV.C. Activity Inventory of the MMW Target Station

For the activation calculations we assume variable operational conditions for different activated components, and they will be detailed for each case. The MCNPX code coupled to CINDER'90 was used in this analysis.

##### IV.B.1. Liquid Hg Target-Converter

In the case of a liquid mercury target, we consider the most conservative irradiation conditions, namely 1 GeV protons at nominal 4 MW power for 40 years. The resulting activity in the target active volume (16 cm diameter and 50 cm long; ~10 litres) is presented in Fig. 9.

First, we note that the total activity of the EURISOL mercury target is comparable to the activity of the used nuclear fuel at a research reactor [14], typically of a few tens MW of thermal power. In addition, some alpha emitters are produced in the case of this spallation target (e.g.  $^{148}\text{Gd}$ ). Finally, although the total activity is nearly

model-independent within MCNPX (within a factor of 2), some individual nuclide production is very sensitive to the choice of input parameters (e.g.  $^3\text{H}$  – factor of 10,  $^{148}\text{Gd}$  – factor of 4).

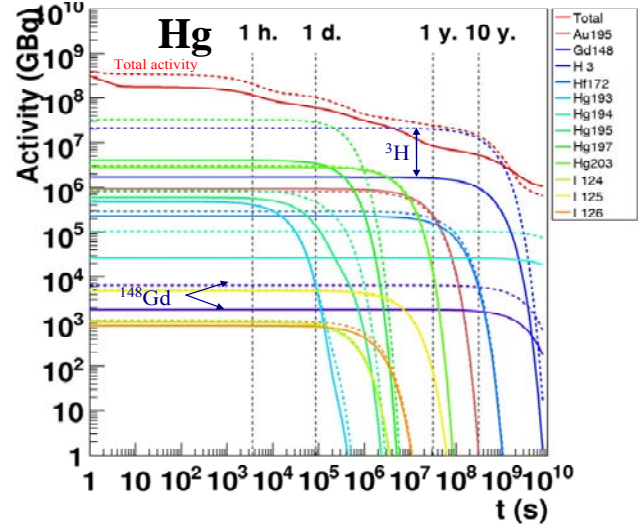


Fig. 9. Residual activity of the MMW target-converter (liquid mercury) as a function of cooling time. Individual radionuclide activities are also presented. Solid and dotted lines are the margins obtained using different models within MCNPX.

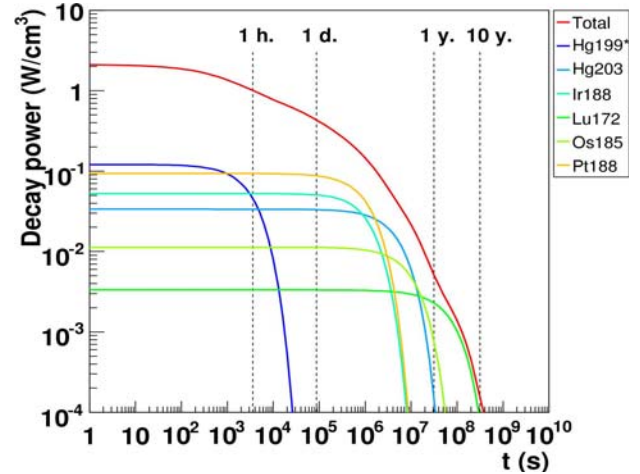


Fig. 10. Decay heat of the mercury target as a function of cooling time. The irradiation time was 5000 h at 4 MW primary beam power.

The decay heat and gamma emission of the mercury target has also been estimated using CINDER'90, as shown in Fig. 10, for 5000 hours of irradiation (annual operating time of the installation after which maintenance operations around the target can take place) at 4 MW beam power. The averaged decay power will be about 0.4 W/cm<sup>3</sup> one day after shutdown or 3.4 kW for the whole “active” target volume. The nuclides giving the main

contribution to the decay heat are also plotted in detail. About  $3 \times 10^{11}$  photons per second and  $\text{cm}^{-3}$  are emitted in the MeV energy range and after 1 day of cooling.

#### IV.B.2. U- and Th-based RIB Production Targets

Concerning the fission targets, we should note separately that here we examine “big” RIB production targets, namely 8 targets with volume  $\sim 1$  litre each. This configuration was a result of our earlier design study<sup>4</sup>. Again we assume conservative irradiation conditions, namely irradiation by 1 GeV protons at nominal 4 MW power for 40 years. The resulting activity in the target active volume of  $\sim 8$  liters is presented in Fig. 11.

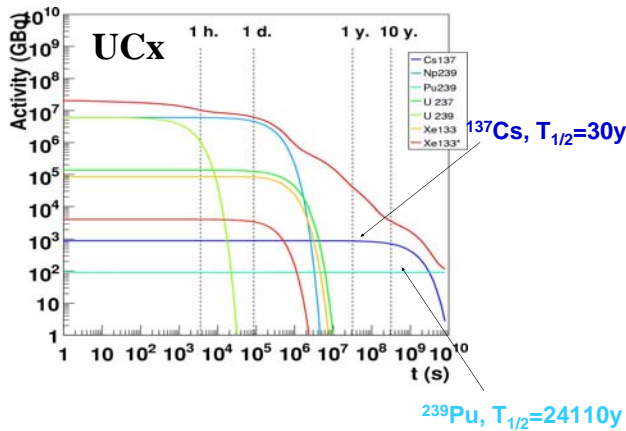


Fig. 11. Residual activity of the MMW fission fragment production target (UCx, based on natural uranium) as a function of cooling time. Individual radionuclide activities are also presented.

We note separately that the total effective density of the production target, made of (Heavy Metal)+(Carbon), is  $3.0 \text{ g/cm}^3$ , where the total HM mass is of the order of  $\sim 13$  kg. Here we compare three types of HM, namely natural uranium, highly enriched uranium (93%) and thorium.

TABLE II. Actinide production in natural uranium fission target with the total mass initially loaded of  $^{\text{nat}}\text{U} \sim 13 \text{ kg}$ .

Irradiation time (days)	Mass of $^{239}\text{Pu}$ (g)	Mass of $^{236}\text{U}$ (g)	Mass of $^{237}\text{Np}$ (g)
30	5.5	0.2	0.1
90	17	0.6	0.4
120	23	0.8	0.5
208	39	1.4	0.9

As shown in Fig. 11, in the case of natural uranium,, the major contributors to the total activity in the long run are  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$ . Indeed, as it is presented in Table 2,

the production of  $^{239}\text{Pu}$  is as high as  $\sim 40$  g per year taking into account  $\sim 70$  % duty cycle of the system.

The situation is similar using  $^{232}\text{Th}$  (Table 3). In this case the breeding of  $^{233}\text{U}$  takes place resulting in  $\sim 70$  g annual production (after  $^{233}\text{Pa}$  decays into  $^{233}\text{U}$ ).

TABLE III. Actinide production in thorium fission target with the total mass of initially loaded  $^{232}\text{Th} \sim 13 \text{ kg}$ .

Irradiation time (days)	Mass of $^{233}\text{U}$ (g)	Mass of $^{233}\text{Pa}$ (g)	Mass of $^{231}\text{Pa}$ (g)
30	3	7	0.2
90	19	12	0.6
120	28	13	0.7
208	57	13	1.3

TABLE IV. Actinide production in highly enriched uranium target with the total mass of initially loaded  $^{235}\text{U} \sim 200 \text{ g}$ .

Irradiation time (days)	Mass of $^{236}\text{U}$ (g)
30	0.6
90	1.8
120	2.3
208	3.9

It seems that the most attractive situation occurs when the highly enriched uranium target is used as shown in Table 4, i.e. neither additional fissile material is created nor higher minor actinides are produced. This configuration is the closest one to the most recent target design discussed in Section II, so called MAFF target solution.

Of course, in all three cases considered above a very similar activity due to the fission fragments would be created; it simply scales to the total number of fission events, which is designed to be comparable in all cases.

#### IV.D. Activation of the MMW Shielding Structures

For the activation of steel (AFNOR norm and density of  $7.92 \text{ g/cm}^3$ ) one assumed that the lifetime of the installation is 40 years and that it is operated at 2.3MW average power with 1 GeV protons.

The resulting activity as a function of cooling time after the facility is shut down is presented in Fig. 12. As expected, the major contributors in the long run are  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$  and  $^{59,63}\text{Ni}$ .

With identical irradiation conditions as for steel, the activation analysis was performed for concrete (ordinary concrete with the density of  $2.2 \text{ g/cm}^3$ ). Note that for concrete, we took into account the realistic impurities

resulting from an activation analysis at a research reactor. The results are presented in Fig. 13, where one can see that the major contributors in the long run are  $^3\text{H}$ ,  $^{45}\text{Ca}$ ,  $^{55}\text{Fe}$  and  $^{41}\text{Ca}$ .

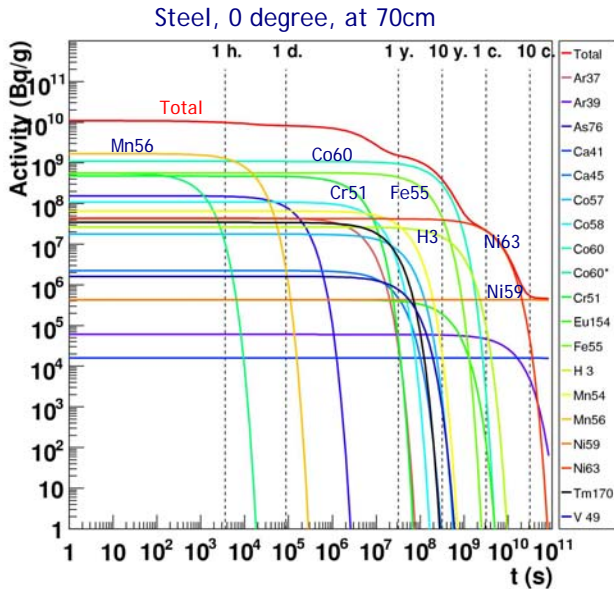


Fig. 12. Residual activity (Bq/g) of the steel shield as a function of cooling time. Both total and individual contributions are presented.

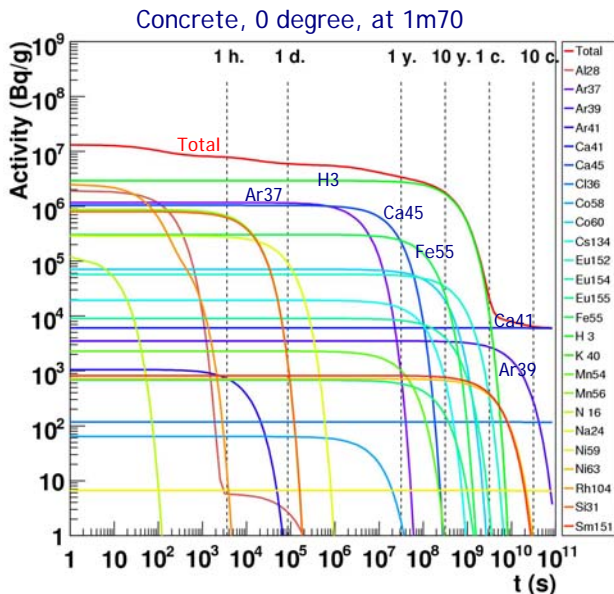


Fig. 13. The residual activity (Bq/g) of the concrete shield as a function of cooling time. Both total and individual contributions are presented.

Finally, in Fig. 14 we plotted the two-dimensional activity profile of the shield surrounding the MMW target station. Note that the proton beam axis is along the  $z$  direction from right to left. This plot represents a cross

view in the  $(r; z)$  plane with a cut in  $x=[-5\text{cm}, 5\text{cm}]$ . The representation at different cooling times helps the characterization and dismantling strategy of the waste for dismantling and final storage. Further studies are in progress, as long as soil and ground water activation are concerned.

Note that final shielding and activation analysis should be performed taking into account also the detailed building geometry and design (e.g., accelerator beam line, radioactive ion source and transport lines, beam dump, etc.).

## V. CONCLUSIONS

In this paper, we briefly summarized the progress made in two tasks of the EURISOL DS project, namely MMW target design (Task 2) and safety and radioprotection (Task 5).

For Task 2, the technical feasibility of a MMW target assembly for EURISOL has been demonstrated by Monte Carlo and finite element calculations as well as by the off-beam experimental tests. The high-energy neutron-induced fission densities aimed for may be achieved with the proposed MMW target baseline design, by using moderate proton beam intensities and reasonable fission target volumes, independently of the actinide composition. A 1 GeV proton beam on a compact mercury proton-to-neutron converter seems favourable to obtain neutron fluxes above  $10^{14}$  n/cm<sup>2</sup>/s/MW of beam, producing more than  $10^{15}$  fissions/s at full primary beam power, and consequently very intense RIBs.

For Task 5, both prompt radiation shielding and activation calculations were finalized in the case of the MMW target station. These results will be further used as a source for the residual dose rate estimates both in normal operation and accidental situations of the facility. They are also indispensable for the nuclear waste characterization in the context of dismantling and the final storage strategy. The use of a highly enriched uranium fission target is favoured in terms of the minimized production of fissile material ( $^{239}\text{Pu}$  or  $^{233}\text{U}$ ) and higher minor actinides.

Finally we note that in most of the cases our predictions were compared to the existing experimental data, leading to a coherent and confident approach for the design study.

## ACKNOWLEDGMENTS

We acknowledge the financial support of the European Commission under the 6th Framework Programme “Research Infrastructure Action- Structuring the European Research Area” EURISOL DS Project Contract no. 515768 RIDS. The EC is not liable for any use that may be made of the information contained herein.

One of the authors (D.R.) thanks the project external partner KAERI and Dr Y.O. Lee in particular for their



kind hospitality during the visiting period at KAERI, where this manuscript was prepared.

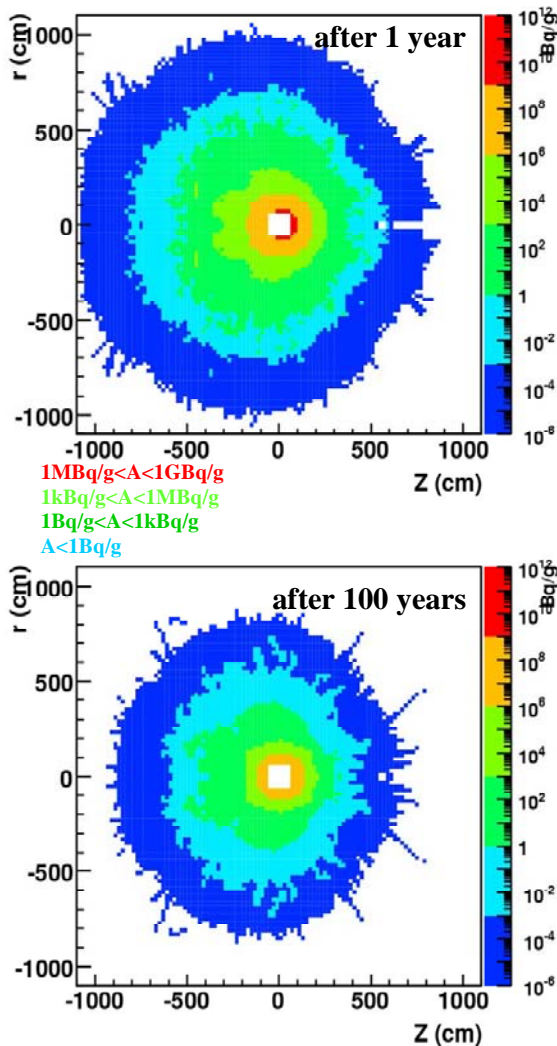


Fig. 14. Activity profile (Bq/g) as a function of shielding (r; z) coordinates of the MMW target station: on the top – after 1 year of cooling, on the bottom – after 100 years cooling.

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