In-Target Yields for Radioactive Ion Beam (RIB) Production with EURISOL

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Abstract. EURISOL DS (European Isotope Separation On-Line - Design Study) project is the European common effort in planning a next generation RIB factory able to deliver secondary beams up to 10¹³ pps at energies up to 150 MeV u⁻¹. The proposed schematic layout of the facility is based on four target stations, three direct targets of 100kW of beam power and one multi-MW (MMW) target two stages assembly. Being produced via spallation the RIBs produced in the direct targets are mainly proton rich. While in the multi-MW target high intensity RIBs of neutron rich isotopes are produced by fission in actinide targets placed in the fast neutron spectrum given by a liquid metal spallation source. The purpose of this paper is to summarize a work carried out within Task 11 "Beam Intensity Calculations": estimation of the in-target yield intensities produced in the various target configurations. Benchmark studies were performed initially in order to verify the accurate description of the spallation models used by the MCNPX2.5.0 code and to choose the best options to be used for the present work requirements. Numerous calculations using MCNPX2.5.0 combined with the evolution code CINDER'90 were carried out to assess the performance of the direct targets. The production rates in the case of the MMW-fission targets were obtained with a given and fixed geometry (optimized to reach 10¹⁵ fission s⁻¹). Only fissile material, moderator and reflector were free.

1. Introduction

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. In EURISOL [1], four target stations are foreseen, three direct targets of 100 kW of beam power and one multi-MW target assembly, all driven by a high-power particle accelerator (~1 GeV). 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, whereas high energy spallation reactions will take place in the direct targets leading to proton-rich nuclei. A possible layout is given in FIG. 1.

Within this project a task were dedicated to the estimates of ion beam intensities. This paper reports the in-target yields obtained with several possible targets. It contains three main parts: benchmarking, direct target and fission target. The first part aims at defining which spallation models should be used and how well suited they are for EURISOL. In the second one numerous calculations were performed due to many possible configurations for the direct targets (material, dimensions, beam energy, etc.). The third one is devoted to the fission (or MMW) target. In this case the complicated geometry is fixed, but fissile material remains a free parameter, as moderator and reflector.



FIG.1. EURISOL facility layout (when EURISOL-DS project started in 2005)

Once obtained these yields should be used with extraction efficiencies to provide the realistic beam intensities.

2. Benchmarking

Although some valuable experience is already available from presently operating ISOL facilities world wide, the design of a new generation RIB factory requires specific and validated modelling tools. Then, the first step was to benchmark the model(s) possibly used on residue and neutron production with experimental data.

In MCNPX2.5.0 [2] we can use 10 spallation models (or to be more precise, model combinations, namely Intra-Nuclear Cascade model \oplus Deexcitation model). We have begun this benchmark study using mass distribution data of reaction products obtained at GSI [3] in inverse kinematics (one example in FIG. 2). This step allowed us a first selection among the 10 models; in this way the first insight of the quality of the models was obtained. Then experimental mass distributions for some elements, which are interesting as RIBs will be also compared to model calculations. These data [3] have been obtained for an equivalent 0.8 or 1.0 GeV proton beam, which is approximately the proposed projectile energy. We note that in realistic thick targets the proton beam will be slowed down and some secondary particles are produced. Therefore, the residual nuclei production at lower energies is also important. For this reason, we also compared some excitation function calculations with the associated data obtained with γ -spectroscopy [4], [5] to test the models in a wide projectile energy range.

We tried also to benchmark on thick target, but very few data exist. Fortunately the only ones which are available coincide with the proton beam energy and target material proposed for EURISOL, namely around 1 GeV for the energy and fissile material (ex.: U) or heavy nuclei (ex.: Pb) for the target. The first set of data, mass distributions of 5 elements, was obtained from ISOLDE [6] (E_p =1.0 and 1.4 GeV; target materials are ThC_x and UC_x). The second set contains the specific activities of 28 radionuclides in different places along the thick target, from a dedicated experiment done at Dubna [7] ($E_{projectile}$ =660 MeV; target material is natural Pb). Quantitatively the MCNPX predictions are often within a factor of 2. See FIG. 3.



FIG. 2. Mass distribution of the reaction products from $^{238}U(1A.GeV)+p$. GSI Data [3] are in black and model calculations are in red for the ten model combinations available in MCNPX2.5.0. Intra-Nuclear Cascades used are INCL4, Isabel and Bertini (the three first lines), and combined to various evaporation/fission models, which are Abla, Dresner/RAL and Dresner/ORNL (the three columns). The last graph (bottom left) is the stand-alone package CEM2k.



FIG. 3. On the left side, mass distribution for Xe element obtained at ISOLDE [6] with the reaction p(1.4 GeV)+UCx. On the right side, specific activities of ¹⁸³Re obtained with the reaction p(660 MeV)+Pb at Dubna [7]. The yellow rectangle indicates a too poor statistic in this region. More explanations on the figures are in the text.

More information, details and references can be found in [8].

These models were also benchmarked on neutron emission, since residues in the case of the fission target option in Eurisol are produced via neutron-induced fission, neutrons coming from spallation reactions in a Hg convector (see section 4.). On this observable, with thin or thick target, all models give comparable good results. This study is developed in [9].

Taking into account the above conclusions including the benchmarks on residue and neutron production from thin and thick targets, the use of Isabel-Abla or INCL4-Abla within MCNPX 2.5.0 is recommended, and sometimes CEM2k also if one combines quality and running time.

3. Direct target

In order to estimate the in-target RIB yields in direct targets we had to study 320 different configurations of cylindrical targets, whose characteristics are summarized in TABLE I. The choice of the beam spot (Gaussian profile) $\sigma = R/3$ came from a previous study [10, 11]. As σ values cannot be smaller than 3 mm [10], the minimal target radius R was automatically equal to 9 mm. The other radii have been chosen to successively increase the target volume by a factor 2. Then, we have considered different target lengths. According to the ongoing EURISOL_DS, we have selected E values ranging from 0.5 to 2 GeV. Finally, as EURISOL targets are designed to stand up to 100 kW, we have fixed P to this maximal value.

Target material	Al_2O_3	SiC	Pb (molten)	Та	UC ₃	
ρ [g.cm ⁻³]	2.0	3.2	11.4	12.5	2.418	
R [mm]	9.0 - 12.7 - 18.0 - 25.5					
L [cm]	50 - 75 -	32 - 48 -	9-18-	8-16-	40 - 60 -	
	100 - 125	64 - 80	27 - 36	24 - 32	80 - 100	
Beam particles	Protons					
P [kW]	100					
E [GeV]	0.5 - 1.0 - 1.5 - 2.0					
σ [mm]	R/3					

TABLE I: PARAMETERS USED FOR THE DIRECT TARGET IN-TARGET YIELD CALCULATIONS



FIG. 4. Charge number distribution of the production rates for the nuclei produced inside Al_2O_3 , Pb, SiC, Ta and UC₃ targets on the left and Pb, Ta and UC₃ targets on the right ($R = 18 \text{ mm}, M = \pi.\rho.R^2.L = 2 \text{ kg}$, spallation model = INCL4/ABLA)

To get the production rates, CINDER'90 [12], an evolution code, has been coupled to MCNPX. This code calculates the residues due to low energy neutrons, not given by MCNPX, and takes into account the decay of all isotopes. Since from our benchmarks (see section 2.), INCL4 and Isabel give similar results and CEM2k is a fast running code, we used 2 models that are INCL4/Abla and CEM2k.

Attempting to "optimize" our targets, according to elements or isotopes studied, we started by plotting mass and charge distribution for all targets and the two extreme energies. To simplify this work, radii (18mm) and masses (~2kg) of the targets were fixed. An example is given on FIG. 4. Then bi-dimensional graphs were plotted to get optimal lengths and radii (FIG. 5).



FIG. 5. ${}^{92}Kr$ (target = UC₃, E = 0.5 GeV) on the left and ${}^{180}Hg$ (target = Pb, E = 1 GeV) production rate per incident proton $[10^{-6} \text{ s}^{-1}]$ as a function of target length L and radius R (model = INCL4/ABLA).

Isotope	Material	Length (cm)	Beam energy (GeV)	In-target rate (at/s)
¹¹ Li	Al ₂ O ₃	~75	1	1.8 10 ⁹
⁷ Be	SiC	~48	0.5	$1.0 \ 10^{13}$
¹¹ Be	Al ₂ O ₃	~50-75	1	$1.2 \ 10^{11}$
¹² Be	Al ₂ O ₃	~50-75	1	$3.0 \ 10^{10}$
¹⁸ Ne	SiC	~48-64	1	8.3 10 ¹⁰
²⁵ Ne	SiC	~32-48	1	3.2 10 ⁹
²⁰ Mg	SiC	~64	1	$2.4 \ 10^{10}$
⁷² Ni	UC ₃	~40	0.5	$7.6 \ 10^{10}$
⁸¹ Ga	UC ₃	~40	0.5	$1.9 \ 10^{10}$
⁹² Kr	UC ₃	~40	0.5	8.9 10 ¹¹
¹³² Sn	UC ₃	~40	0.5	$2.9 \ 10^{11}$
²⁰⁶ Hg	Pb	~18	0.5	$2.9 \ 10^{11}$
¹⁸⁰ Hg	Pb	~27	1	$1.5 \ 10^{10}$
²⁰⁵ Fr	UC ₃	~(40-60)	1	1.8 10 ⁹

TABLE II: IN-TARGET PRODUCTION RATES FOR SOME ISOTOPES WITH DIRECT TARGETS USING INCL4/ABLA

Finally we summarize in TABLE II optimal configurations for some isotopes and give the estimated production rate. It must be stressed that only in-target yields have been calculated, no extraction efficiencies have been taken into account. All yields obtained here should be multiplied by the effusion/diffusion efficiencies to get realistic ion beam intensities.

More details concerning this work can be read in [13].

4. Fission target

FIG. 6 shows the target set-up as model build by MC code. The geometry model represents the last design variant, MAFF-like, able to accommodate 30kW load heat. The production targets are cylinders with internal axial holes, having a volume of 181 cm³ (R_{ext} = 1.75 cm, R_{int} =0.4 cm, H=20 cm).



FIG 6. Geometry model used in MCNPX simulations. Left: XY cross section cut through the model in the plan Z=0; Right: YZ cross section cut through the model in the plan X=0. Zones of the model are indicated in the graphs.

A homogenized material of fissile-graphite compound ($\rho = 1.88 \text{ g cm}^{-3}$) with mass rate 1/20 and the actinide mass of 15 g was accounted in all calculations performed. For all configurations studied based on Uranium the moderator material was water while the reflector material was beryllium oxide. Only in case of Thorium used as fissile the iron was used in the model for both moderator and reflector materials. MCNPX2.5.0 was used in simulations.

Derivation of the fission yields was based mainly on recommended tabular yields for the fission products given in [14]. These recommended yields include many nuclides which fission by neutrons at three energies: T for thermal energies (0.0253eV), F for fast energies (2 MeV) and H for 14 MeV for high energy. The yields were calculated using the formula:

$$Yield = T \int_{0}^{ET} \sigma_f(E) \Phi(E) dE + F \int_{ET}^{EF} \sigma_f(E) \Phi(E) dE + H \int_{EF}^{1GeV} \sigma_f(E) \Phi(E) dE$$
(2)

where the integral limits were chosen as follow: ET = 5 keV and EF = 5 MeV.

In the analysis six target assemblies configurations were accounted: five cases based on Uranium compounds with 235 U percentages of: 99.99, 20, 3, 0.072 (natural Uranium) and 0.02 (depleted Uranium) as well as 232 Th.

The results presented are normalized to 1 mA intensity proton beam on the target to allow easily the desired scaling to other values.

Configuration	²³⁵ U*	²³⁸ U	Total	²³⁵ U [%]
99.99% ²³⁵ U	5.7689E+14		5.7689E+14	100
20% ²³⁵ U	1.3823E+14	1.2021E+12	1.3943E+14	99.14
3% ²³⁵ U	2.1712E+13	1.3931E+12	2.3106E+13	93.97
Unat	5.2449E+12	1.4172E+12	6.6621E+12	78.73
U _{dep}	1.4591E+12	1.4223E+12	2.8814E+12	50.64
²³² Th	5.2366E+11		5.2366E+11	

²³²Th for Th case

TABLE III: FISSION RATES FOR ALL TARGETS [FISSIONS $\rm s^{-1}~mA^{-1}]$ AND PERCENTAGE OF THE $^{235}\rm{U}$ CONTRIBUTION.

TABLE III shows clearly that for all accounted Uranium mixtures more than half of the total fission rates are due to fission of the ²³⁵U component. The number of fissions (~10¹⁵/s) required by the baseline EURISOL project is thus met for the most favourable producing system (²³⁵U case), since a 4 MW target corresponds to 4mA beam. The use of other Uranium mixtures or ²³²Th is possible although it may reduce the magnitude of yields but yet it extends the yields range over a larger area of interests.

As a matter of example FIG. 7 shows the distribution for Kr and Sn isotopes for the six studied target configurations. These elements are two of the most relevant ones for RIB production at EURISOL as indicated in [13]. The differences in the yields between the six systems follow the fission rate trend previously discussed.



FIG. 7. Fission yields isotopic distributions for two selected products. Left: Kr and right Sn

All calculated isotopic distributions display an almost centred-Gauss like shape (as in the FIG. 7) that shows that the contribution of the ²³⁵U in the mixture is dominant comparatively with ²³⁸U whose fragment distribution is biased towards the neutron-rich wing of the distribution. ²³²Th and ²³⁸U present a slightly extend further to the ²³⁵U fragment distribution to lighter and heavier mass. This explains the enhanced production yield in the range (72 < Z < 78) for Ni produced from ²³²Th that reach values even higher than those obtained for 20% ²³⁵U configuration.

5. Conclusion

After benchmarking the available spallation models in the transport code MCNPX2.5.0 for the EURISOL purpose, methodologies have been developed to estimate the in-target fission yields arising from both direct targets and converter-fission targets assembly of the EURISOL facility. This study provides quantitative estimates of the fission yields for a variety of isotopic distributions come out from different target systems. Obtained results represent helpful information for the beam intensity predictions at the future EURISOL facility and using effusion/diffusion efficiencies realistic beam intensities will be provided.

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6. References

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