STUDIES FOR THE OPTIMIZATION OF RADIOACTIVE BEAMS IN SPIRAL

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Since 2001, the SPIRAL facility, located at the GANIL in Caen (France), successfully delivers beams of radioactive gaseous ions. These are key for studies in nuclear structure, hot nuclear matter and reaction mechanisms, including research in super heavy elements or halo nuclei. To achieve those, stable ions ranging from C to Kr of up to 6kW can potentially irradiate the SPIRAL target, where fragmentation reactions produce a stream of radioisotopes. The unstable species diffuse out of the target core, effuse through the target and transfer lines and, eventually are ionized in an ECRIS ion source. The radioactive ions can then be electromagnetically extracted, mass separated and injected into the CIME cyclotron, which accelerates them up to 25 MeV/A, while performing a fine isobaric separation.
Several of the previous steps are antagonistic, for instance, larger targets accept higher primary beam intensities at the expense of a hindered diffusion and effusion behavior. This article discusses the optimization of SPIRAL targets for exotic noble gas isotope production, based on the simulations with the RIBO Monte Carlo code.

I. INTRODUCTION

The production and use of Radioactive Ion Beams (RIBs) far from the valley of stability has fascinated scientists and has helped them understand the atomic and nuclear properties. The pioneer laboratories struggle to achieve higher intensities of utterly exotic beams that will allow further advances in nuclear physics, astrophysics, solid state physics and other disciplines of science. These efforts involve the optimization of each of the production steps of radioisotopes. In particular, computations are conducted to minimize the decay losses during the extraction of radioisotopes from the target where they are created through nuclear reactions.

In this article, simulations with the Radioactive Ion Beam Optimizer (RIBO) (Ref. 1) scan the importance of several design parameters of the *System for the Production of RAdioactive Ions on-Line* (SPIRAL) target (Ref. 2, 3) for the production of Kr isotopes.

I.A. RIBO: The Radioactive Ion Beam Optimizer

When an isotope is generated inside a powder aggregate, it diffuses across solid matter up to the grain surface and then it effuses through the porous structure until it finds the powder-vacuum interface. Next, it effuses through the vacuum system, following a random walk of collisions with the system walls, sticking on them for some time. Finally, after flowing through a temperature controlled line (that performs chemical selection), the isotope reaches the ion source and, in the lucky cases, it is ionized and extracted before it finds the ion source outlet as a neutral isotope. These processes may consume a considerable amount of time, during which radioactive decay may occur, hereby lowering the number of extracted isotopes.

The task of approaching RIB intensities to the requirements of new physics studies encompasses the engineering effort to maximize all relevant phenomena in beam formation, namely production, diffusion, effusion, ionization, and ion extraction. RIBO was written to meet the demand for a global optimization tool of these closely interlaced factors.

RIBO is Monte Carlo simulation code based on constructive solid geometries (CSG). It integrates diffusion and effusion for variable pressure and conditions, including diffractive reflections, transport through porous media, adsorption, ionization in surface and plasma sources and ionic transport, among others. Latest developments allow setting moving walls and tracking complex trajectories.

I.B. Radioactive Ion Beams at GANIL

I.B.1. GANIL and the SPIRAL project

In conventional ISOL facilities, like ISOLDE or the future projects of EURISOL (Ref. 4), SPES (Ref. 5) and SPIRAL2 (Ref. 6) primary beams of light ions, i.e. protons, neutrons (originated in proton-neutron converters), or deuterons, are used to irradiate targets. These are tailored for the production of a specific set of isotopes, typically through fission and spallation reactions. A different target is required for each physic case.

In turn, the Grand Accelerateur National a Ions Lourds (GANIL), like GSI, MSU or RIKEN, uses inverse kinematics (Ref. 7). In these laboratories the radioisotopes are created by fragmentation of the primary ion projectiles. A broad fan of neutron-rich nuclei can thereby be produced by simply adjusting the ion mass of the primary beam that bombards a same target.

The fastest fraction of the exotic atoms diffuses out of the target and effuses through the vacuum system towards the ion source without decaying into more stable nuclei. In GANIL, an Electron Cyclotron Resonance Ion Source (ECRIS) of the type NANOGAN-III (Ref. 8) is used to produce ions (q/m = 0.09-0.40), which are then extracted (7 – 34 kV), preselected ($\Delta m/m = 4x10^{-3}$) and injected into the *Cyclotron for Isotopes of Mean Energy* (CIME). Most of the RIB purity is achieved by CIME (K = 265, B*ρ = 2.344 Tm), which acts as an efficient spectrometer for fast ions ($\Delta m/m \sim 5 \times 10^{-4}$)

SPIRAL was funded in 1993 and it began to produce RIBs by September 2001. The facility is run by GANIL with the support of various French National and European institutions.

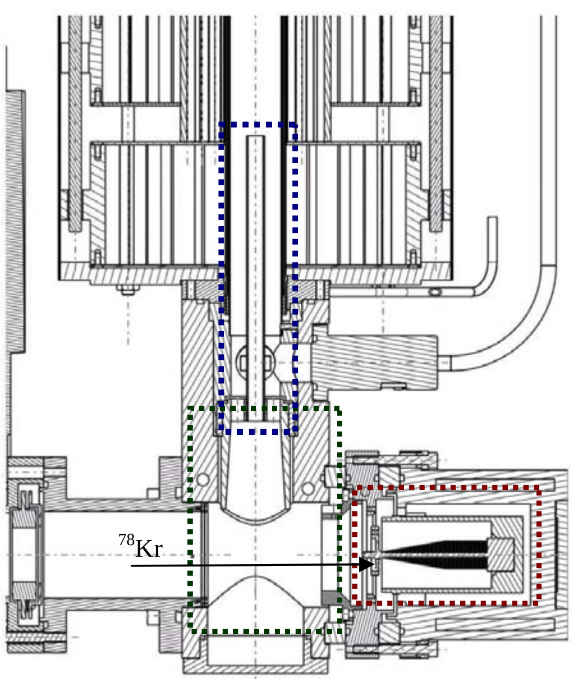

Fig. 1. The SPIRAL target and ion source assembly. The target (right bottom), transfer line (middle bottom) and connecting tubes (center) are in dashed boxes. Source: GANIL

I.B.2. The SPIRAL target

The SPIRAL target and ion source system is located inside a well shielded cave beneath ground level. The bombarding ion beam would produce a very sharp heating depth-pattern, related to the Bragg peak of its range. In order to mitigate the uneven heat deposition, the target has a conical front-end that helps to distribute the production of isotopes within various pellets. Moreover, the beam is rotated around the beam axis with an offset of 5.5 mm to favor transverse heat uniformity. In addition, external Ohmic heat is fed through a central rod. The target length was chosen to allow stopping primary beams and products from nitrogen to kripton.

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TABLE I. Some parameters of SPIRAL target

1	Carbon grain size	1 micron
2	Diameter of target pills (after cone)	12.6 mm
3	Conical full angle of first pills	12.5^{o}
4	Thickness of a pill	0.5 mm
5	Distance between pills	0.8 mm
6	Total number of pills	53
7	Temperature in target area	2273 K
8	Temperature in the triangular plug	1000 K
9	Temperature in the transfer line	350 K

Fig. 1. sketches the SPIRAL target with 55 graphite aggregate (1 micron) pellets, of excellent thermal properties, low atomic number and high sublimation temperature. Indeed, the heat robustness of the target was proved in the tests carried out at Louvain-la-Neuve. As for the ion production and transport, the coupling between the NANOGAN-III ECRIS and CIME was successfully tested at SIRa prior to installation.

Lots of efforts have already been gathered to achieve the present design (Ref. 9, 10 and 11). However, more exotic and heavier isotopes demand further optimization of several parameters to improve the extraction efficiency. For example, the ^{78}Kr primary beam has a range of 1.87 mm in carbon, which means that only 4 out of 55 pellets are used in the production of radioisotopes. An analysis needs to be carried out to determine to which extend a smaller target would help the release of heavy isotopes and what other modifications could be made to increase the yield. Similarly the powder grain size and the elements of the transfer line should be closely looked at.

II. METHODOLOGY AND RESULTS

Simulations have been run for five alternative configurations derived from the current SPIRAL target. Each case has been simulated in four batches of 10000 particles each, requiring no more than 3 hours/run in a 5-year-old P-IV laptop running Linux. Average figures have been obtained with a precision better than 1.3 %.

The input file that describes the geometry has 127 regions that relate to a total of 148 quadrics by means of Boolean expressions. The isotopes are uniformly born in the first four (or 10, for Ar) pills. Effusion through the powder is carried out with a special module, which samples a number of semi-analogue random tracks and then builds up macro-tracks. The 'real' value for the micro-step depends on the microscopic structure of the compressed powder, but its impact in the total effusion path is low. In this case, a starting value of 15 micron with fully open powder porosity has been assumed. Both hypotheses have been reviewed (sections II.B and II.C).

The temperatures of the surfaces are adjusted to the values of table I. Collisions are assumed fully diffusive. The injection tube to the ion source is separated from the rest of the transfer line by a grid of 50% transparency. It has not been necessary to implement with detail this grid because RIBO can handle semi-transparent walls. Residual pressure has been set to zero.

II.A. Standard target configuration

The standard configuration for SPIRAL target is the one introduced in section 1.B.II. The probability density for the birth of particles in the pills is uniform due to the conical shape and the rotation of the beam around the target axis. RIBO was forced to generate particles within the first four pills.

The output file contains a table that shows for each i-j combinations which percentage of the particles was born in cell *i* and died off at cell *j*. In our case we observe that 100% of the particles end at the entrance of the ion source (as expected in absence of other termination statements), and that 17.2% were born in the first pill, 23.3% in the second, 31.04% in the third and 27.4% in the fourth. Thus, it is confirmed that the production scales to the pill size (cone effect), except for the fourth pellet, which is partly (30%) beyond the Bragg Peak reach of the ^{78}Kr primary beam.

The total number of collisions in the vacuum system is low, about 2410, with less than 10% of those occurring in the target-container assembly. This figure has no relevance for noble gases like Kr or Ar, but it could be important for isotopes that had a big chemical affinity with the walls of the system.

Due to rediffusion into the compressed powder, particles spend some time in each of the carbon pellets, including all those that do not contribute to the isotope generation. Each of these times is lower than 0.3% of the total with a slight tendency for higher times in the pills at the back end of the target. In any case, the total time spent in the target container is relatively low, less than 30%, the rest takes place in the transfer line, which has a big pumping volume.

The previous two points imply that changes affecting only the target may have a very small effect in the effusion profile, because the transfer line is the one to cumulate most of the effusion delay.

Table 2 contains a summary of the average effusion parameters for this target and for its variants.

RIBO computes the statistical momenta up to 5th degree for the release times of simulated events. From this numbers it calculates the coefficients (t_1=14.9, t_2=240.7 ms) for an analytical fit with two exponentials or the effusion release efficiency (the fitting errors of order 3 and for are lower than 3.8 and 2.5 %, respectively):

$$E(t) = \left(1 - e^{-t/t_1}\right) \cdot e^{-t/t_2} \qquad (1)$$

The effusive release fraction derived from eq. 1 for ^{71}Kr (64 ms) is above 10%. The final yield is obtained by multiplying this number by the absolute generation, the diffusion release fraction and by the ionization efficiency.

TABLE 2. Effusion parameters: average effusion time, collisions, distance traveled out of pellets and in the pellets, and effusion release efficiency for ^{71}Kr (64ms).

target	<t> [ms]	<nc>	<d> [m]	<dp> [m]	^{71}kr E [%]
II.A	256.6	2410	93.59	0.117	12.6
II.B	256.4	2411	93.59	0.113	12.6
II.C	259.1	2434	94.57	0.119	12.11
II.D	258.5	2333	94.11	0.115	10.9
II.E	250.5	2252	88.55	0.061	11.2
II.F	82.5	720	30.15	0.051	28.4

II.B. Influence of the microstructure in the pellets

The carbon pellets are made of 1 micron grains. Powder with 4 micron grains is also available. In order to assess the impact on the effusion release of shifting to a larger grain size, the sampling micro-step has been multiplied by accordingly (15→60 micron).

Results (second row of table 2) show that this parameter hardly makes any change to the total effusion efficiency. However, it is expected that *diffusion* gets worse because the diffusion time constant grows as the square power of the diameter of the grains, as observed in (Ref. 3)

It should be noted that for non-noble gases (desorption takes place in each collision), a looser structure can be very beneficial because the desorption time upon each collision may dominate the release.

II.C. Importance of the pellet surface finish.

The surface of the pellet may be less porous than the inner body, due to the superficial stress applied during machining. If that were the case, particles would have a harder time finding the path out of the pill, but in equal proportion, they would be less likely to make excursions back into the pills from the outer volume. In overall, surface close porosity could be beneficial in a case like SPIRAL, where only 4 pills are producing particles because the remaining 49 are potential sources of time losses. However, particles may globally effuse faster if they can cross through the pills instead of having to a path around them (this is especially true in very tight structures).

Various phenomena are counteracting their actions. In order to have a first order estimation for the combined effect of all of them, RIBO was run with 50% permeability in the surface of the pills. The results, shown in the third row of Table 2, are again very insensitive to powder related characteristics. For non-noble gases, this statement would not hold.

II.D. Influence of the triangular plug

One of the questions for future target improvements is whether the triangular plug at the front-end of the container is a deterrent for effusion since it stays in the way of the isotopes exiting the target container. This piece provides Ohmic heating to the target. If it were removed, the temperature in the target could drop by 200 K, thus requiring external heating.

The RIBO input geometry was modified by rendering obsolete the regions that describe the plug, and simulations were run with 2073 K in the target. The results, always shown in Table 2, are discouraging as no benefit is observed for an eventual removal of the triangular plug.

II.E. Shortening the target length

Due to the short range of their corresponding primary beams, neither Kr, nor Ar production need more than a fraction of the target. In the output files from the previous simulations it is remarked how particles spend time in and around the unproductive section of the target. It is a good principle to reduce the volume (effusion) and total surface (sticking) of the target as long as this is compatible with a good pumping speed and isotope production. However, for noble gases and for the models used in the simulations, there is no evidence that smaller targets would increase the effusion efficiency, as shown in table 2.

II.F. Changing the coupling ion-source transfer line

Two technical solutions are used for the ion source entrance (Ref. 8).

In the present design, the 50% transparent grid and the ion source are linked by a pipe of 34 mm of diameter and 139 mm long. This pipe hosts a smaller tube (d = 26 mm) coaxially. Isotopes flow in the space between the two concentric pipes.

In an alternative design, the inner pipe is smaller (d=10 mm), and particles can flow through its interior as well as through the space between the small and the big pipe. The 50% transparent grid does not block the inlet to the smaller pipe. The pipes are longer, 190 mm instead of 139, however, the conductance is much better in this case ($C \sim R^3/L$). Therefore, the effusion release should be much faster in this case.

Simulations (last row of table 2) confirm these predictions. The drawback of this configuration comes from the ion source efficiency, which is lower than with the present design.

II.G. Release of Ar isotopes

The methodology is the same as the one used for Kr, except that particle generation is restricted to the first ten pellets. Simulations show that with the present target ^{31}Ar could be extracted (effusion only) with an efficiency of about 4%, while the effusion release fraction for ^{32}Ar would be close to 30%.

All the analysis made for Kr is extensible to Ar.

III. CONCLUSIONS

Simulations performed with the Radioactive Ion Beam Optimizer show that the effusive release of noble gases is fast, and mainly depends of the design of the transfer line. A dedicated shorter target is not needed and will not help in the release. The same is true for the removal of the triangular heating plug in front of the conical end.

Thicker powder grains will only reduce the diffusion process and are highly unadvised as long as the fine grains withstand the heating without sintering.

More studies of the transfer line are needed to evaluate the trade off between effusion and ionization efficiency.

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