PROPAGATION OF RADIOACTIVE CONTAMINANTS ALONG THE ISOLDE BEAMLINE

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Abstract

Several Radioactive Ion Beam (RIB) facilities are in construction worldwide or undergoing upgrade. To ensure safe management of radioactive contamination, RIB vacuum systems are completely hermetical, with collection and storage of the effluents before controlled release. The layout of new RIB vacuum systems would be designed more efficiently with a better knowledge of the contaminant's propagation along the machine. At CERN, an experimental analysis of the propagation of radioactive species along the beam-line of ISOLDE was carried out during the last beam-run. The Monte-Carlo code Molflow+ was then used to interpret the results. We observe that radioactive rare gases are the major source of contaminant propagation. Propagation times of the order of some seconds were calculated and benchmarked with measurements for ^{6}He performed on beta scintillators on-line. The blocking effect of magnet separators and beam optics elements was partially confirmed.

INTRODUCTION

Since the first production of a radioactive ion beam (RIB) at the Niels-Bohr Institute in 1951 and the first ISOLDE facility in 1967 at CERN, RIB facilities have flourished worldwide, fostering nuclear research with exotic nuclei production. Several new projects [1], including an intensity upgrade of ISOLDE are bound to see the light of day in the coming years.

ISOLDE [2] produces RIBs by irradiation of a thick target with the pulsed 1.4GeV, 2μA average current proton beam from the PS Booster accelerator. Exotic nuclei generated by the collision diffuse and effuse through a tiny transfer line to the ion source, where ~10% of them are selectively ionized. Isotope separation is then achieved in either of two on-line separators. The General Purpose Separator (GPS) consists of a 70º bending magnet and an electrostatic switchyard, allowing selection of three mass separated beams. The High Resolution Separator (HRS) consists of two (90º and 60º) bending magnets plus focussing-defocussing and correction electrostatic multi-poles. The HRS is followed by a Radio Frequency Quadrupole Cooler and Buncher. The two separator lines converge into a common beam distribution system at the main switchyard, conveying high purity isotope beams to several experimental stations.

Most of the nuclei produced in the target are not ionized [3]; the volatile species diffuse via the transfer line across the ion source and through the beam-line. These are believed to be the cause of the contamination of the vacuum of RIB machines. To avoid contaminants dispersion along the beam-line, vacuum is achieved by turbomolecular pumps, connected via a totally hermetical common backing manifold to a set of primary pumps [4]. These convey the effluent gases to a gas storage system, which confines radioactive gas for controlled decay before release to the atmosphere via the nuclear ventilation system. By this scheme, all vacuum effluents from target to experimental beam-lines flow together into a common storage, thus mixing highly contaminated with less contaminated gas.

In order to gain knowledge on the distribution of neutral contaminants along the beam-line, we have sampled contamination with filters installed along the vacuum system, which were then analysed by gamma spectroscopy. In parallel, we have carried out a Monte-Carlo simulation of the diffusion of rare gases from the target along the beam-line. This article presents the approach and preliminary results obtained on the GPS branch of Isolde.

SAMPLING OF RADIOACTIVE SPECIES

Hybrid filters were installed at the exhaust of each turbomolecular pump during the machine shutdown. They consist in two filtering disks, one of 150μm thickness in hydrophilic mixed cellulose ester with 3 μm pores, the other one in carbon impregnated cotton fibre, clamped together by two stainless steel nets and installed in the thickness of a ISO-KF standard o-ring supporting ring. The turbomolecular pumps being isolated both at the inlet and at the outlet by valves, installation did not require venting of the beam-line nor of the backing manifold. The filters were kept in place for 4 months, seeing 9 different targets and more than 30 beams. The 3 filters installed on the turbomolecular pumps of the GPS branch were then removed after a 40h beam stop and analysed within 7 days in gamma spectroscopy with a high-purity Germanium detector. Pump GPS21 is installed upstream of the separator magnet and switchyard, while GPS22 and GPS23 are both installed at the same longitudinal coordinate right downstream of it. Radioactive species appearing in the 3 filters and relative activity are presented in Figure 1.

All species, besides ^{194}Au, are long-lived isotopes, of half-lives of the order of days. The gold isotope ^{194}Au is progeny of the very long-lived ^{194}Hg, in secular equilibrium. With the exception of the antimony isotopes, all species are issued from rare gas parents (radon, xenon and krypton) or from mercury (^{185}Os). Indeed, the specific

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activity found in each filter is the result of a decay occurring in the zone below the turbo and is therefore proportional to the integrated flow of the parent species through that particular turbo-pump during the operation run period. This fact confirms that the contamination of the vacuum system is the consequence of the propagation of rare gases and volatile species.

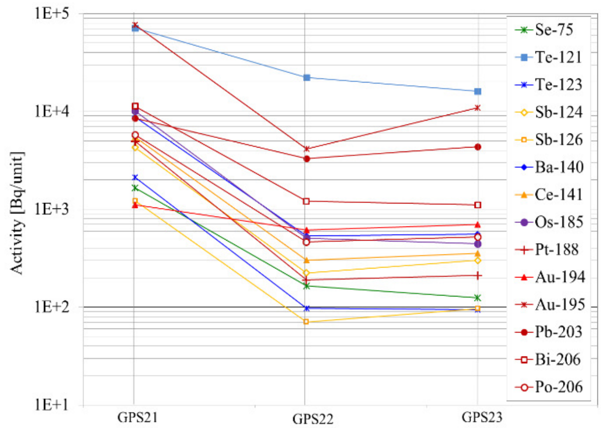

Figure 1: Activity [Bq/units] measured on the 3 filters installed on GPS turbopumps. Green: progeny of Kr. Blue: progeny of Xe. Red: progeny of Rn. Purple: progeny of Hg.

It is difficult to extract information from the absolute value of activity: indeed, this depends strongly on the species-specific yield for every particular target having seen proton beam during the reference period. It is possible however to infer information on the propagation of parent species. The activity detected on the filters features a decrease along the beam-line. Successive filters thus collect only 4% to 10% of the activity seen on the preceding one. Some species however seem to propagate more efficiently beyond the separator magnet: these are ^{121}Te (27%), ^{194}Au (59%) and ^{203}Pb (45%).

We may argue that different effects are acting to hinder the propagation of radioactive rare gas isotopes. On one hand, beam optics elements like the separators magnets and electrostatic switchyard and the related beam-line bends, act as obstacles to the free effusion of gases down the line. On the other hand, the population of short lived parent species decreases naturally by radioactive decay. To evaluate one effect against the other, simulations were performed.

MONTE-CARLO SIMULATION

The Test-Particle Monte-Carlo simulation code Molflow+ was developed in the 90s for calculation of pressure in molecular flow regime [5]. The code is constantly under development at CERN. Time-resolution was recently implemented and benchmarked with literature and experiment [6]. In Molflow+, the vacuum chamber walls are described by an assembly of planar polygons or facets. The geometry can be imported from CAD programs in STL format. The ISOLDE 3D model was thus reconstructed in Autodesk Inventor by the analysis of the existing drawings of single segments and the 3D CATIA (Dassault Systèmes) integration model.

Once the geometry is imported, a gas source is described as a rate of gas release from a facet, while a pump or gas sink is described by converting its pumping speed into a sticking probability multiplied by molecule impingement rate.

After the particles are created, a ray-tracing algorithm determines the collision locations, takes into account the radioactive decay and calculates the pressure and density. The process is described in detail in a CERN technical document [7].

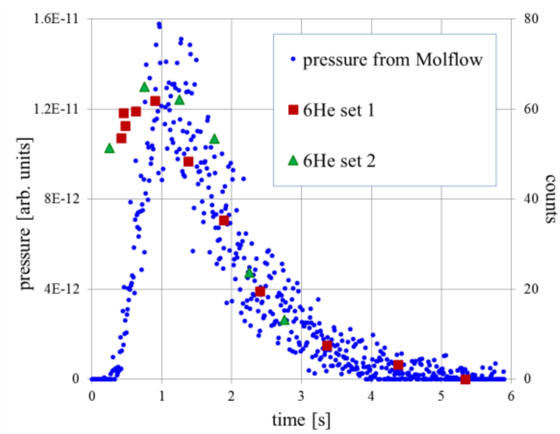

Figure 2: Benchmarking dynamical Molflow+ with the beta scintillators, for ^{6}He. Units are arbitrary.

Dynamical Molflow+ calculation on the propagation time of ^{6}He was compared with the beta-scintillator counts at the tapestation downstream of the main switchyard merging GPS and HRS beam. The isotope ^{6}He, featuring a half-life of 0.8s, is the main neutral species generated at the target. Propagation time of a ^{6}He square pulse of 100ms from the exit of the transfer line to the tapestation was simulated and compared with the scintillators measurement triggered by the Booster proton pulse. The results are shown on figure 2. Tapestation data, for two different data sets, are corrected with subtraction of background. The ^{6}He release pulse was not measured here but it is known from previous measurements to have a typical rise time 8-20ms and a fall time 80-120ms. Agreement between the Molflow+ distribution of arrival times and what obtained from the tapestation is excellent, in spite of some "early" counts on the tapestation.

Molflow+ was then applied to calculate the distribution of arrival times at the tapestation for the rare gases Xe, Kr and Rn, parents of most of the species found in the filters. Mass was chosen as the standard atomic weight of the element. Figure 3 displays the results. The median of the log-normal fits of the simulated data is taken as a time-of-flight from transfer line to tapestation and presented on Table 1.

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Finally, for each of the 3 parent species we calculated the transmission probability across transparent facets at the entrance of each of the 3 turbomolecular pumps GPS21, GPS22 and GPS23 for an initial arbitrary desorption flux from the entry facet of the transfer line equal to 0.01 mbar·l/s.

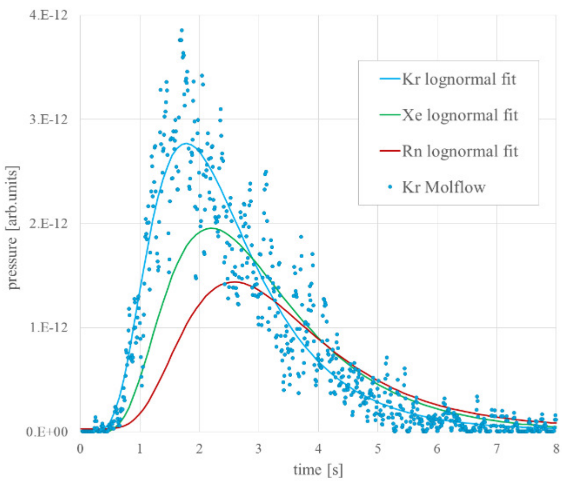

Figure 3: Arrival time distribution calculated with Moflow for Kr, Xe and Rn for a square pulse of gas generated at the transfer line entrance. For simplicity, only the lognormal fit is displayed for Xe and Rn.

Table 1: Transmission Time from Lognormal Fit

Species	Transmission time [s]
Helium	0.87±0.01
Neon	1.40±0.01
Argon	1.76±0.02
Krypton	2.24±0.04
Xenon	2.78±0.08
Radon	3.14±0.05

Ratios of transmission probabilities through GPS22 and GPS23 to GPS21 are compared with filters yields in Table 2, where measured activity on filters GPS22 and GPS23 are averaged out. Propagation of Hg cannot yet be simulated with Molflow+, as we cannot yet account for a finite surface sojourn time.

DISCUSSION

With some exceptions, the transmission probabilities across the GPS separator magnet and switchyard agree qualitatively with what observed from the spectroscopy on the filters. In some cases, the reduction in parent population appears stronger than predicted by pure molecular flow transmission. This occurs in particular when the parent's half-life is much shorter than the typical propagation time of the order of some seconds along the beam-line. Thus, ^{188}Pt, progeny of the 4.4ms half-life ^{196}Rd, is indeed less present after the separator than predicted by Molflow+. However, a short-lived parent does not explain all discrepancies between calculation and measurement. The significant transmission of some of the species found in the filters (^{121}Te, ^{194}Au, ^{195}Au, ^{203}Pb) may be related to the beams produced during the beam-run, with ions being transported across the separator and neutralizing after it. Analysis of this effect is made difficult by the large variety of beams produced at ISOLDE.

Table 2: Filter Yield Ratios

Species	Activity ratio	Rare gas parent	T.P. ratio
^{75}Se	9%	^{75}Kr	10.2
^{121}Te	27%	^{121}Xe	9.7
^{123}Te	4%	^{123}Xe	
^{140}Ba	6%	^{140}Xe	
^{141}Ce	6%	^{141}Xe	
^{188}Pt	4%	^{196}Rn	9.0
^{194}Au	59%	^{198}Rn	
^{195}Au	10%	^{199}Rn	
^{195}Pt	24%	^{199}Rn	
^{203}Pb	45%	^{203}Rn	
^{206}Bi	10%	^{206}Rn	
^{206}Po	9%	^{206}Rn	

CONCLUSION

Our study confirms the validity of a Monte-Carlo approach with the Molflow+ code as a predictive tool for the propagation of radioactive contamination along RIB machines. Propagation time of the order of some seconds between the target and the main switchyard has been calculated and benchmarked with on-line beta scintillator measurements at a tapestation. Reduction of contaminants by the hindering effect of beam optics elements is efficient, but some species pass these elements to a yet unexplained extent.

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