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EXPERIENCE WITH ISOLDE MOLTEN METAL TARGETS AT THE CERN PS-BOOSTER

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ABSTRACT

By moving the ISOLDE mass separators from the 600 MeV Synchrocyclotron (SC) to the 1 GeV PS-Booster Synchrotron [1] the instantaneous energy density of the proton beam went up by 3 orders of magnitude. Sixteen molten metal targets and ion-source units have been used under pulsed proton beam conditions at ISOLDE between 1991 and 1995 and the development of the SC-units to match these requirements is described. Sets of baffles have been designed. Installed in the transfer lines which link the targets and the ion-sources they prevent splashes of the molten metals to flow into the ion sources. The liquid metal containment has been modified so that the electron-beam welds are no longer in contact with the melt. A pyrocarbon disk inserted behind the proton beam entry window reduced its corrosion. Reduction of the energy density of the proton beam obtained by spatial defocusing and time staggered extraction allowed to reduce the shock effect of the pulsed proton beam.

1. Introduction

At ISOLDE [2], radioisotopes are produced by spallation, fragmentation or fission reactions of the target material induced by 1 GeV protons. Typical target materials are refractory metal foils (Ta, Ti, Nb), carbides (Si, U, Th), oxides (Ca, Mg) and molten metals (La, Pb, Sn). They are contained in a 20 cm long, 2 cm diameter Ta-tube of 0.5 mm wall thickness. These containers are heated up to nominal temperature which range from 600 to 2200°C by a DC-current of typically 500 to 1000A. The radionuclides diffuse out of the target material and effuse into the ion-source where they are ionized onto a hot surface, in a plasma or by a resonant laser beam [3,4]. After acceleration to 60kV and mass-separation these radioactive ion-beams are distributed to 15 experimental areas. Under the new pulsed beam conditions at the PS-Booster (3

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10^{13} protons/pulse, beam impact area : 3×4.5 mm^2, pulse width 2.4 μs and repetition rate 0.8 to 0.4 Hz) a number of breakdowns were observed when molten metals were used as targets such as flow of the target melt into the ion source, broken welds and corroded proton beam windows. Effects which were not observed during operation at the SC proton beam (1.5 10^{11} protons/pulse of a width of 30 μs and a repetition rate of 100-300 Hz).

2. Design of molten metal targets

Under a pulsed proton beam, the target container and material are submitted to pulsed thermal stress (typically 10^6 cycles), dilatation waves and corrosion. The observation of target units dismantled in a hot-cell showed that none of the above mentioned effects can be neglected. In this section, the computable parameters such as temperature, thermal stress and vapor pressure and the chosen technical issues to the observed damages are presented.

2.1 Temperature pulse and vapor pressure

The energy deposition from the proton beam has been calculated using the Monte-Carlo programs GEANT and FLUKA [5]. The temperature increase and its time evolution was computed by solving the cylindrical symmetric heat equation. The temperature increase at the center of a Ta-disk after one pulse of 3 10^{13} protons reach 600°C. Its time evolution is shown in figure 1. The vapor pressure was calculated and is shown in figure 2 for the hottest spot of the target material for three representative cases : Proton beam turned off, after a nominal proton pulse and for a defocused proton pulse.

2.2 Thermal stress and tensile strength

The thermal stress in a Ta-disk resulting from a nominal proton pulse is 70 Mpa. It is comparable with the expected tensile strength of Tantalum at 1400°C. Indeed, the observations made on metal foil targets showed that the Ta-containers and metal foil target materials which have been used at temperatures up to 80% of their melting points were strongly deformed.

2.3 Baffles

Cross sections of molten metal containers used at the SC are shown in figure 3. The ion-source of a molten Lanthanum target is located 5 mm above the surface of the melt and was found to be filled up with Lanthanum after very few proton pulses. A special unit was then constructed and showed that a contact was established synchronous with the proton pulses, between the melt and Ta-electrodes which hang between 3 and 10 mm above the surface. This was the first evidence of a proton-beam induced movement in an ISOLDE liquid metal target. Baffles were designed and tested which protected the ion source from the splashes (figure 4). Three successive flat baffles installed under 30 degrees in a 50 mm vertical chimney were sufficient to prevent the liquid Lanthanum splashes generated by the proton pulses (3×10^{13} protons within 2.4 μs) to reach the ion source. A temperature controlled helix was inserted in a 60 mm vertical chimney on the Lead target and tested up to 2/3 of the maximum proton pulse intensity. The temperature controlled helix acts as a condensation loop of the Lead vapors which allows working at a higher melt temperature. Finally a set of

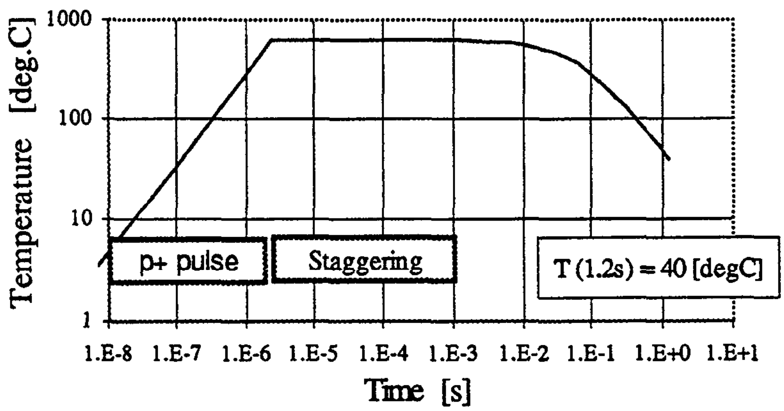

Figure 1. Temperature evolution at the center of a Tantalum disk hit by a 1 GeV proton pulse from the PS-Booster (3 10^{13} protons, beam impact area FWHM : 3×4.5 mm, pulse length 2.4 μs). The time spread of a staggered pulse (≤500 μs between rings) is shown. After a cooling time of 1.2 s the temperature in the center of the disk has increased by 40°C .

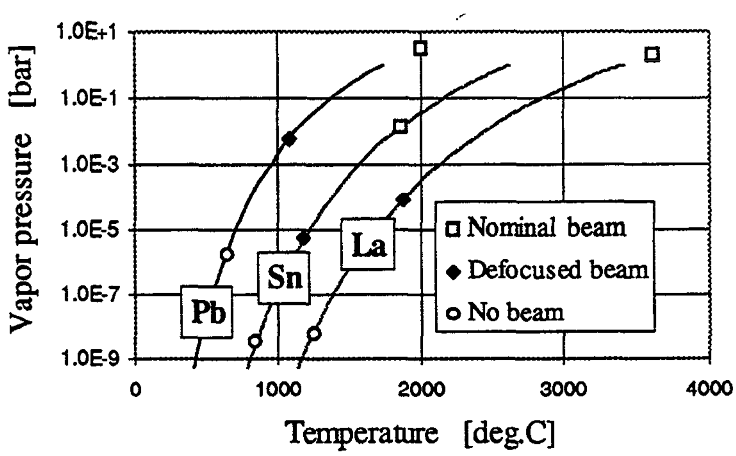

Figure 2. Temperature dependence of the vapor pressure for Lanthanum, Lead and Tin. The Vapor pressure at the typical temperature of the melt (○), the maximum temperature after a nominal proton pulse(□) and the maximum temperature after a defocused proton pulse (◆) are indicated.

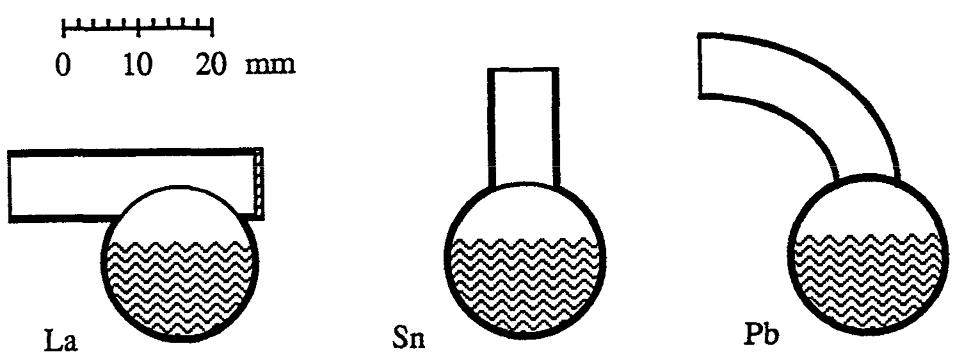

Figure 3. Section of the molten Lanthanum, Tin and Lead target containers successfully used with the SC proton beam. The first part of the transfer lines linking the target to the ion sources are shown.

three temperature controlled Tantalum cones inserted in a 60 mm vertical chimney were successfully tested up to maximum proton beam intensity with a liquid Tin target.

2.4 *Container*

The strength of the pressure wave in the liquid metal was sufficient to break the welds of the thermocouple pockets which were hanging from the top of the container into the melt and the welds of a stainless steel container. The thermocouple were therefore moved outside the containers. The simulation of the target temperature at equilibrium showed that all liquid metal targets are heated by the conduction (and then convection) of the heat generated by the ohmic losses in the current feed welded on both ends of the container tube. Therefore, an improvement of the mechanical strength of the container was obtained by doubling the wall thickness of the Ta-tube (from 5/10 to 1 mm). In addition, the uniformity of the temperature was increased by placing Ta-heat screens around the container.

2.5 *p^+-beam window and welds*

A metallographic investigation of the tungsten-innert-gas (TIG) welds of Ta containers showed that the grain size is much larger than the material thickness thus explaining the large grain boundary leaks observed on the Ta-container of a molten Lanthanum target. Electron beam welds have a much smaller recrystallisation region and the grain size obtained is small compared to the size of the weld. The proton beam window is now cold crimped into the Ta-tube of the container and then electron-beam welded. The recrystallized part is separated from the melt by 10 mm of cold crimped tube (figure 5).

In a test set-up, a liquid Lead target kept at 700 °C was equipped with an temperature controlled Iron helix. A pyrocarbon disk (thickness 4 mm) was inserted behind the proton-beam window in order to prevent the Lead to corrode the Tantalum. The energy deposition of the proton beam on the window was simulated with a 1 kV electron beam. After 4 days, the window was analyzed by electron beam microscopy which showed a 1 mm deep cavity in the container material at the warmest spot of the container which was not covered by the pyrocarbon disk. Ta-Fe alloy in form of grains and layers were found in the colder regions and on the container surface. It could be concluded that, a pyrocarbon layer is a good protection for the proton beam window, and that helix and container must be made out of the same material.

3. Wave threshold

The environment of the targets used for the production of radioactive ion beam is : High temperature (needed for a rapid diffusion through the bulk material), high voltage, and high radiation rates [6] (average dose per target unit : 1 MGy at 10 cm) which excludes investigations during ion-beam production at least with standard target units. A well known feature of the liquid metal targets is their rather slow release time constants (typically ≥30 s) [7,8] and section 4. This time constant is much larger than the time between two proton pulses (which averages to 2.4 seconds) therefore the intensity fluctuation of an ion beam (half life ≥ 30 s) should be below 6% as observed for Mercury and Cesium isotopes obtained at the SC from molten metal targets of Pb and La respectively. In fact, for low proton beam intensities at the PS-Booster the ion-

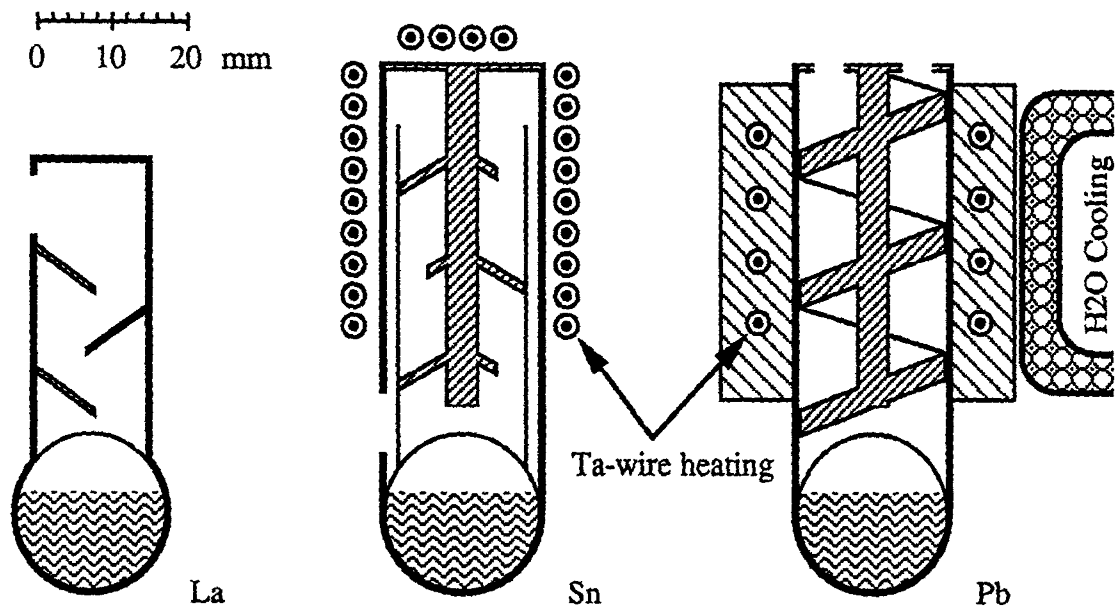

Figure 4. Section of the molten Lanthanum, Lead and Tin target containers designed for operations with a pulsed proton beam. The baffle systems which prevents splashes from liquid metal to reach the ion-source are shown. The chimney of the Tin target can be heated up to 1000°C with a Ta-wire (insulated by Al_2O_3 tube sections). The temperature control (water cooling and regulated Ta-wire heating of the Copper block surrounding the vertical chimney) of the Lead target is designed to operate between 350 and 550 °C.

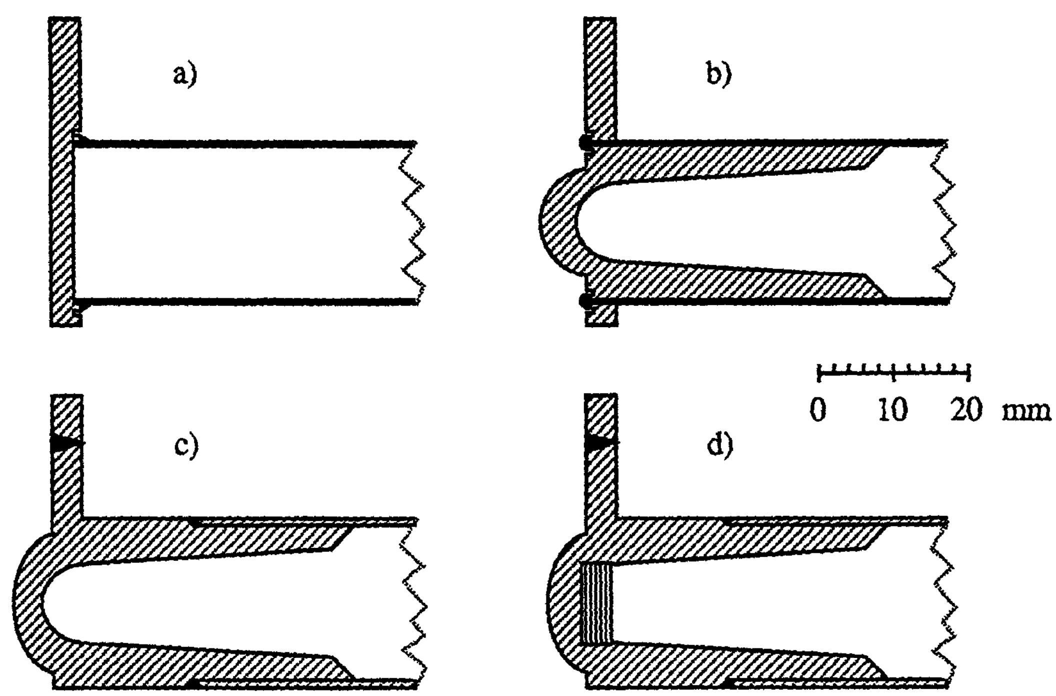

Figure 5. Proton beam windows of the liquid metal containers. a) standard window with TIG-welds and 5/10 mm tube. b) Protected TIG-weld. c) Electron beam weld protected from a direct contact with the melt by a -2/100 tight fit. d) Protected e-beam welds, in addition a 4 mm pyrocarbon disk is tightly fitted under vacuum inside of the proton beam window.

beam intensity is almost constant but large fluctuations of the ion-beam intensities were observed above a given proton pulse intensity. This change in the release mode did coincide with the proton intensity at which contact between the melt and the electrodes has been observed during the "wave test" described in section 2.3. Therefore, a so called "wave threshold", was defined and the smooth or fluctuating behavior of the ion beam intensity is used to monitor the state of the liquid metal. Figure 7 shows a set of successive beam intensity measurements, where smooth behavior is observed below and strongly fluctuating behavior above the threshold.

3.1 Defocalisation test

The energy deposition of the pulsed proton beam heats up the target material and the proton beam window. The target containers which are already at high temperature are therefore submitted to particular high thermal stress. Since the temperature increase is proportional to the energy density, the most natural way to reduce the temperature is to increase the proton beam size. The effect of a two fold increased beam size was investigated by monitoring the ion-beam intensity. We could observe that the wave threshold is 10% higher for a doubled proton beam size (spot size FWHM : 7×11 mm^2) and that the losses in radioactive ion production remains marginal.

4. Hg-release from a molten Pb target

The radioisotope concentration in a molten metal follows an exponential law. At the SC, the time needed to release half of the Mercury isotopes (release time constant) from a molten lead target was typically 120 s [7]. At the PS-Booster a release time constant of 5 s was measured. This short time constant cannot be explained by its temperature dependence (a temperature above three thousand degree would be needed). The faster release is most probably linked to the violent shaking of the liquid metal after a proton pulse and should improve the production of short lived isotopes.

5. Staggered beam test and Cs-release from a molten La target

The PS-Booster consists of four synchrotrons (Rings) each containing five proton bunches (fifth harmonic). The four rings are extracted at time intervals of 0.6 μs. A "standard" proton pulse from the PS-Booster consists of twenty proton bunches delivered in four groups within 2.4 μs. The speed of sound in liquid metal is typically of the order of 10^4 m/s, therefore the time criteria for the existence of a "shock wave" in a molten metal target is met since the relaxation time for a typical ISOLDE target is of the order of 5 to 10 μs. It is therefore of interest to investigate the effects of a time-staggered extraction of the four acceleration rings on the targets and on the release of radioisotopes. The staggering is obtained by increasing the extraction time intervals between rings.

5.1 Proton beam and experimental set-up

In the staggered extraction mode, only three out of the four rings can be used and the time between two rings can be stretched from its original setting of 0.6 μs up

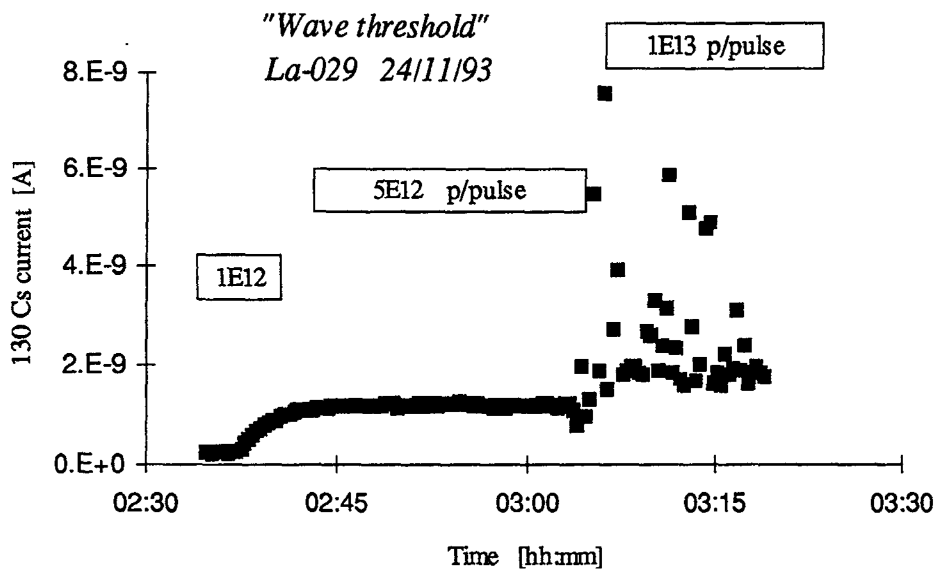

Figure 6. ^{128}Cs ion-beam intensity recorded with successive Faraday cup measurements for different proton pulse intensities. Seven proton pulses were delivered onto the target within the 14.4s PS supercycle. At a proton pulse intensity of 10^{12} and 5×10^{12} protons per pulse the release is smooth, but for a proton pulse intensity of 10^{13} proton/pulse the onset of strong fluctuations of the ion beam intensity are observed.

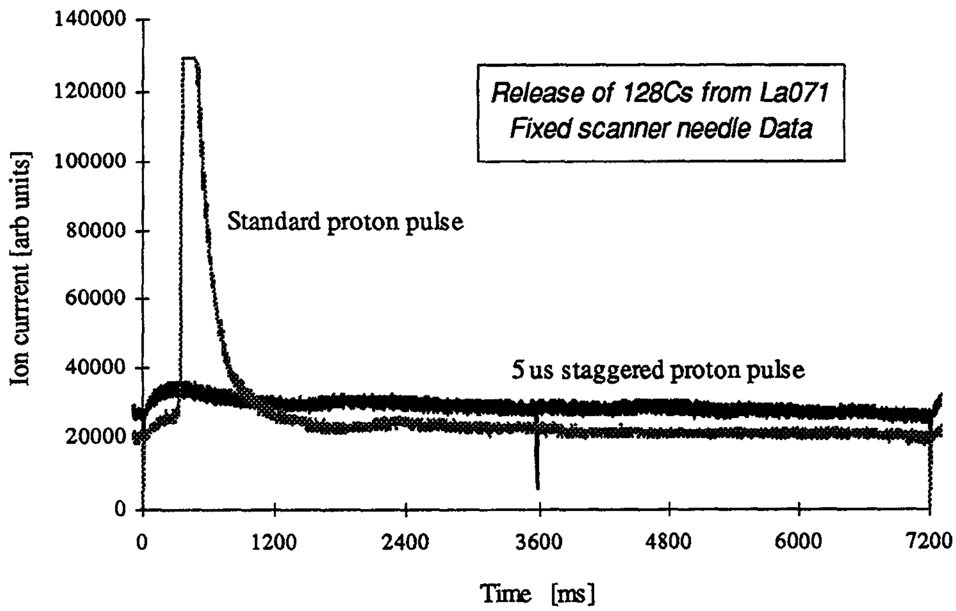

Figure 7. ^{128}Cs ion-beam release from a Liquid Lanthanum target for staggered (5μs between rings) and standard (pulse width of 2.4 μs) PS-Booster proton pulses (1.5 10^{13} protons, FWHM : 7×11 mm). This *"Fixed Scanner Needle"* measurement is an on-line collection of the charge of the ion-beam on a metallic needle, amplification of the signal and readout via a storage oscilloscope.

to 500 μs. Two tests have been performed with a liquid Lanthanum target equipped with a temperature controlled transfer line and a baffle arrangement. During the test, proton pulses were delivered onto the target every 7.2 s.

5.2 Production yield

For a fixed staggering time of 500 μs the proton pulse intensity was increased from 7.5 10^{12} to 2.3 10^{13} proton/pulse. The $^{128}Cs^{+}$ beam intensity has been determined after collection of the ion-beam on an aluminized tape and measurement of its β-decay count rate (Tape Station) and by direct measurement of the charge current of the ion-beam (Faraday Cup or Scanner Needle intercepting the ion beam). The ratio of the ^{128}Cs atoms to the proton pulse intensity was found to be almost constant over the full range. A ≤8 % reduction was observed at maximum beam intensity.

5.3 Influence of the staggering time on the release curve

At a fixed proton pulse intensity of 1.5 10^{13} proton/pulse the staggering time was reduced from 500 to 5 μs and finally brought to the standard settings of 0.6 μs between two rings. The $^{128}Cs^{+}$ beam intensity as function of time (Release shape) was recorded with tape station and scanner needle measurements. Within the precision of these measurements, all release shapes for staggering time higher or equal to 5 μs are identical. Their release time constant do correspond to the SC value [8]. For standard proton beam, the ion beam intensity shows a different behavior. Large ion-beam peaks are recorded on top of the smooth release shape which is observed with staggered proton beam. These peaks show an ion beam intensity 5 to 10 times higher than the smooth part and a FWHM of 0.3 to 0.5 s as shown in figure 8. The rise time (10 to 90%) of these peaks is typically 20 ms. The distribution of the times between the proton pulse and the start of the peak is not constant, half of them do appear 0.3 ±0.2 s after the proton impact but the remaining are "randomly" distributed. In addition, no "closed transfer line" effect could be observed which would have explained the fluctuations by accumulation of the radioactive vapors during temporary obstruction of the transfer line (by splashes) followed by a sudden release. At the end of the test, the proton beam was switched off while recording the intensity of the radioactive ion beam for 150 s. The time constant of the "beam-off" release of the Cesium from Lanthanum was 32.1 s and is in perfect agreement with the SC measurements for a melt temperature of 1250 °C.

6. Conclusion

The liquid Lanthanum and Lead target are delivering ion-beams to the user community with a proton pulse intensity of typically 10^{13} proton/pulse. The containers, ion-sources and proton beam windows are still operational after $2x10^{18}$ protons.

It was shown that for a staggering time greater than the typical relaxation time of 5 μs and for proton pulse intensities up to 2.2 10^{13} proton/pulse the production yield of radioisotopes is not depending on the pulse intensity, and their release time is identical to what was measured with ISOLDE targets at the SC.

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