

# LARGE-SCALE ISOTOPE PRODUCTION WITH AN INTENSE 100 MEV PROTON BEAM: RECENT TARGET PERFORMANCE EXPERIENCE\*

F. M. Nortier<sup>1</sup>, LANL, Los Alamos, NM 875453, U.S.A.

J. W. Lenz, J.W. Lenz & Associates, Waxahachie, TX 751653, U.S.A.

P. A. Smith, P.A. Smith Concepts & Designs, Los Alamos, NM 875443, U.S.A.

## Abstract

Isotope production targets for proton beams with energies greater than 30 MeV often require a design in which certain target materials must be encapsulated. Performance information of such target capsules under extended bombardment with intense proton beams is not widely published. Final capsule design and choice of capsule material are often based upon in-house operational experience. The 100 MeV Isotope Production Facility (IPF) at Los Alamos National Laboratory employs encapsulated RbCl salt and gallium metal targets for the large-scale production of medical isotopes Sr-82 and Ge-68. For the encapsulation of RbCl targets, EB welded 316 SS and inconel 625 were used in recent routine productions and in test runs, while niobium capsules were used in the bombardment of gallium metal targets. Capsule design, production beam conditions and performance data compiled from routine production runs at LANL over a two- year period are presented. The data show that inconel 625 is superior to 316 stainless steel for encapsulation of RbCl salt targets. On the other hand the data for niobium capsules are inconsistent and somewhat counter intuitive considering the relatively low operating temperatures predicted by detail thermal modeling of the gallium targets.

## INTRODUCTION

A 100 MeV proton beam is used at the IPF [1,2] for the production of radioisotopes used in medicine, science and industry. Extended target irradiations with beam currents of up to 250  $\mu$ A are used in regular large-scale production runs. Most target materials subjected to such high power densities and extreme radiation conditions do not maintain their cooling configuration and must be encapsulated in a durable metal to ensure sustained removal of the heat during extended irradiation. Occasional failure of target capsules during extended irradiation is a common problem. The complex failure mechanisms are not very well understood and they are difficult to control. A systematic approach to understanding and preventing capsule failures is time consuming and expensive and the target designer usually has to rely on intensive modeling, qualitative visual inspections of irradiated targets and target survival data. Factors contributing to such target failures include:

- Internal over pressure due to trapped gas expansion or boiling of the target material.
- Corrosion of the capsule window due to

chemical attack by the target material.

- Erosion of the capsule window due to mechanical interaction with the pulsed beam and turbulent cooling water.
- Work hardening of the window due to mechanical pulsation and thermal cycling.
- Radiation damage to the window.
- Undetected weakness in the capsule window or weld due to material or fabrication deficiencies.

## IRRADIATION CONDITIONS

In order to lower the peak power density on the IPF targets the 25 kW pulsed beam is swept in a circle across the target faces. The beam spot completes approximately 3 revolutions during each beam pulse, resulting in a ring-shaped beam profile. Nominal beam parameters of relevance are listed in Table 1.

Table 1: Beam Parameters

Beam Parameter	Value
Pulse duration	625 $\mu$ s
Average power	25 kW
Spot size	12.5 mm
Sweep radius	12.5 mm
Sweep frequency	5 kHz

The targets [3] are disc shaped and have a nominal diameter of 50 mm. Up to three targets are irradiated simultaneously with nominal incident energies of 90 MeV, 65 MeV and 30 MeV. Targets are separated by spacers to form 5 mm thick cooling channels between the target faces. Typical cooling water velocities flowing over the target faces range between 2 and 5 m/s. The RbCl targets and gallium targets for which survival data are presented here were irradiated typically in a target stack arrangement shown in Fig. 1.

Encapsulated RbCl salt targets are used for the production of Sr-82. The capsule halves are machined either from 316 stainless steel or from inconel 625 with a window thickness of 0.3mm. The salt pucks are prepared by casting and then sealed inside the capsule under vacuum by means of electron beam welding. For the production of Ge-68, gallium metal is encapsulated in niobium. The niobium capsule halves are also fabricated as described above. Pre-welded capsules are filled with liquid gallium through a fill port, which is plugged and sealed under vacuum by electron beam welding.

\*Work supported by DOE Office of Nuclear Energy

<sup>1</sup>meiring@lanl.gov

## OBSERVATIONS

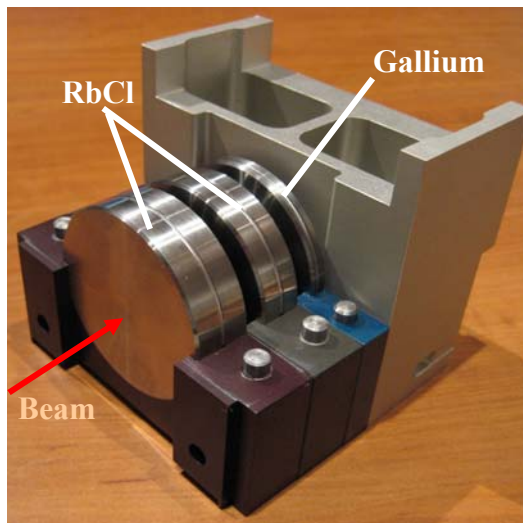


Figure 1: Example of a target stack used for the production of Sr-82 and Ge-68.

A total of 98 RbCl targets were irradiated in extended productions and test runs. Of these 54 targets were encapsulated in 316 stainless steel and 44 in inconel 625. Beam currents used on the stainless steel capsules varied between 100  $\mu\text{A}$  and 200  $\mu\text{A}$ , while the beam currents on inconel capsules varied between 200  $\mu\text{A}$  and 220  $\mu\text{A}$ . By comparison the beam currents used in the irradiation of 38 niobium encapsulated gallium targets varied between 100  $\mu\text{A}$  and 250  $\mu\text{A}$ .

Thermal analyses of RbCl targets under bombardment at a maximum beam current of 250  $\mu\text{A}$  predict a peak temperature in the target of close to, but not exceeding the boiling point of the RbCl salt. By contrast the peak temperature in a gallium target at 250  $\mu\text{A}$  does not challenge the material boiling point at all. This is due to a higher thermal conductivity and a higher boiling point of the gallium over that of RbCl salt. While an accurate prediction of the effects from all the thermal processes occurring in a fully- or partially molten target is difficult, a comparison of predicted temperature distribution patterns with corresponding discoloration patterns on the faces of actual irradiated targets show very good agreement. Such positive visual feedback provides increased confidence in the predictability of our models describing all the coupled mechanisms that participate in establishing the steady state temperature distribution in a partially molten target under bombardment. An example of such a comparison is shown in Fig.2.

## DISCUSSION

The survival data for RbCl targets collected over a period of about 2 years are summarized in Fig. 3. For these targets the occurrence of a catastrophic failure event

could be detected promptly by a distinctive change in the conductivity of the cooling water. Each blue bar in the figure indicates the integrated beam value after which the particular target was confirmed to have survived. A red bar indicates the integrated beam value at which catastrophic failure occurred. Similarly, the survival data for the gallium targets are summarized in Fig. 4. Catastrophic failure of a gallium target was not detectable by a change in cooling water conductivity. Therefore, the irradiation of some of the gallium targets were interrupted from time to time for inspections in order to establish interim points of confirmation of the target status. As in the case of the RbCl targets a blue bar or blue part of a bar also indicates the integrated beam value after which a particular gallium target was confirmed to have survived. By contrast a red part of a particular bar indicates the integrated beam range over which a confirmed failure occurred.

In the case of RbCl targets relatively frequent failures were observed for stainless steel capsules. Four out of six failures occurred at an integrated beam value of approximately 10 mAh. For the inconel capsules no failure was observed up till now despite the fact that a large percentage of the inconel capsules have been subjected to significantly longer bombardments. The observed failures in niobium encapsulated gallium targets are random and show no correlation with integrated beam value. The present data suggest that one may expect a niobium capsule to fail at any integrated beam value between zero and 150 mAh. While a wide variety of beam currents were used in the irradiations, it is important to note that no correlation between failure rate and beam current was observed for any of the data sets. In the case of stainless steel capsules there is a noteworthy indication that the failures correlated with the duration of irradiation rather than with the integrated beam value. As stated above, no correlation with beam current value is discernable. It is concluded that the capsule failures included in the body of data presented here must still be driven mainly by parameters associated with the control of the target fabrication process.

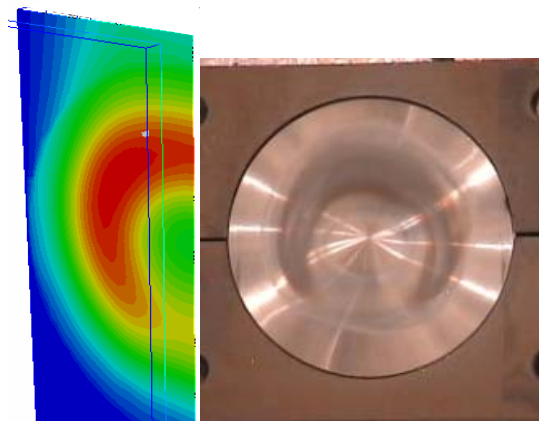


Figure 2: Comparison of an expected temperature distribution pattern with real target discoloration pattern on the face of a gallium target.

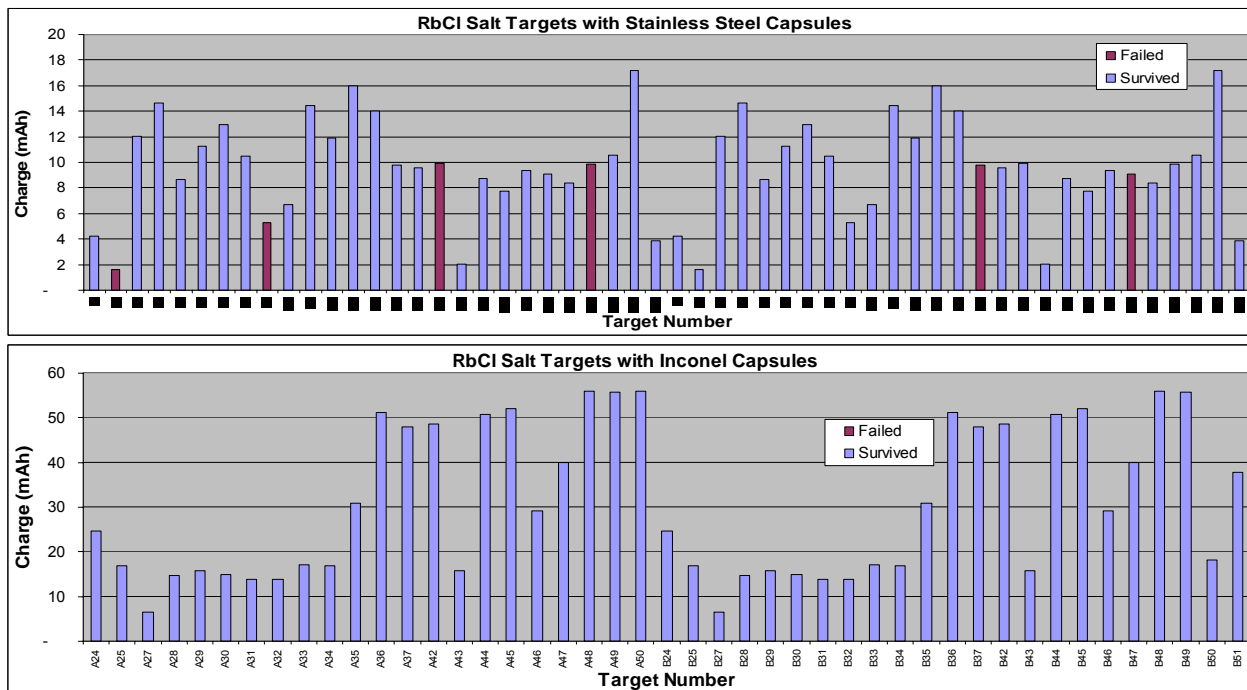


Figure 3: Survival data for RbCl targets encapsulated in stainless steel and inconel capsules.

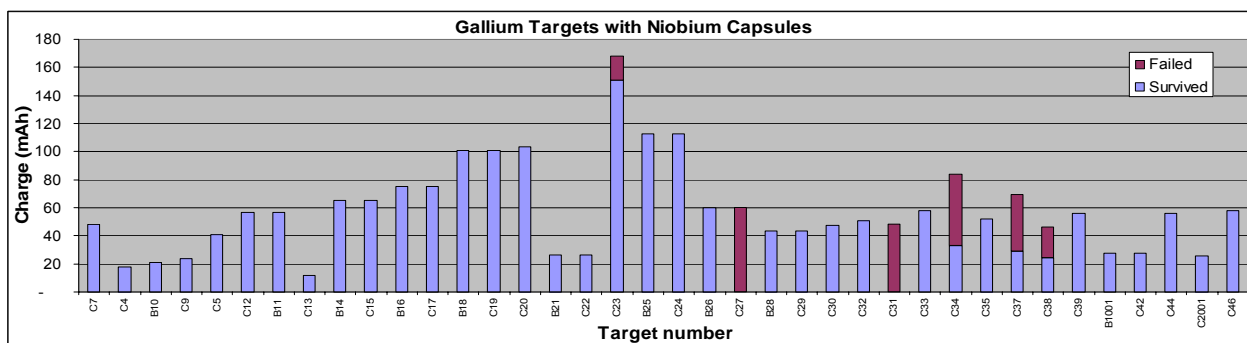


Figure 4: Survival data for gallium targets encapsulated in niobium capsules.

## SUMMARY

Target survival data for encapsulated RbCl and gallium metal targets have been collected for a large number of extended irradiations at a world class 100 MeV isotope production facility. The irradiations include runs for the large-scale production of Sr-82 and Ge-68 as well as several test runs. Beam currents varying between 100  $\mu$ A and 250  $\mu$ A were used. It is commonly acknowledged that, in addition to intensive thermal modeling of targets, isotope production target designers must also rely on practical survival trends to help understand and prevent target failures. Despite this, extensive target survival data are not readily available in the literature. Survival data obtained from almost 150 irradiations are presented here. Clear statistical trends are still not discernable. However, subtle differences in the data for 316 stainless steel, inconel 625 and niobium capsules are useful to help understand the mechanisms that contribute to the onset of a catastrophic target capsule failure in these materials.

## ACKNOWLEDGEMENTS

The authors acknowledge the team members of the LANL Isotope Production Program for their assistance in generating the data. The contributions of the LANSCE Accelerator Operation and Technology Division are also acknowledged. The work is supported by the U.S. Department of Energy's Office of Nuclear Energy.

## REFERENCES

- [1] K. F. Johnson et al, "Commissioning and Initial Operation of the Isotope Production Facility at the Los Alamos Neutron Science Center (LANSCE)". *Proc. European Particle Accelerator Conference 2004, Lucerne Switzerland, July 5-9, 2004*, p. 2816.
- [2] D. R. Phillips et al, "The New 100 MeV Isotope Production Facility at LANL". *Transactions of The American Nuclear Society and The European Nuclear Society*, **TANSAO 89 1-920** (2003) 769.
- [3] F.M. Nortier et al, "High Current Targetry for the 100 MeV Isotope Production Facility", *Transactions of The American Nuclear Society and The European Nuclear Society*, **TANSAO 89 1-920** (2003) 77