Ni target development for the TULIP project

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Abstract. The TULIP project (https://anr.fr/Projet-ANR-18-CE31-0023) aims to produce radioactive ion beams of short-lived neutron-deficient isotopes using fusion-evaporation nuclear reactions in an Isotope Separator On Line (ISOL) system. A Ni target was chosen to produce Rb and Sn isotopes, and was bombarded, respectively, by Ne and Cr primary beams at energies close to the Coulomb barrier. Owing to the Target Ion Source System (TISS) configuration, the operational TISS temperature had to be close to 1300°C. Several tests were performed to determine a configuration able to cope with the constraints related to the TISS, target and beam. This article presents a brief description of the TULIP principles and objectives, and the development of the Ni target.

1 Introduction

The SPIRAL1 (Système de Production d’Ions Radioactifs Accélérés en Ligne, phase 1) isotope separator on line (ISOL) facility, installed at GANIL (Grand Accélérateur National d’Ions Lourds) has been producing radioactive ion beams (RIB) for 20 years. RIB, mainly from gaseous elements, have been produced using an electron cyclotron resonance ion source. Within the framework of an extended authorisation given to GANIL, a far greater variety of other RIB can be produced through combinations of current target materials and primary beams.

Since 2010, new TISS have been developed to enlarge the panel of RIB offered to GANIL users. The TULIP project started in 2015, addresses the production of short-lived neutron deficient isotopes. Its operational principle is quite different from mainstream TISS as it favours efficient atom-to-ion transformation (AIT) over in-target production rates. In the case of Rb+ RIB production, which is the first case studied within the project, the target is a 4 µm thick Ni foil impinged with a Ne ion beam (4.5 MeV/A, ~1.5 pµA). At this energy in-target production is relatively low (~10^6 pps for ^76Rb for example). However, the compact TISS design allows to operate with greater AIT efficiency. Overall, the global RIB production should be high enough to satisfy user requirements, especially for short-lived isotopes.

The TULIP TISS is briefly presented before a more detailed description of the trials performed to obtain a satisfactory Ni target.

2 TULIP target constraints

TULIP essentially consists of a Ni target and a graphite catcher enclosed in a graphite cavity (Fig. 1). The initial aim was to maintain the cavity and the catcher at a temperature of 1573 K through ohmic heating, produced by high current circulating in the cavity walls and the target. Considering the case of a Ne beam on a Ni target, evaporated residuals, including Rb species, continue their post-production path through the Ni target up to the graphite catcher where they stop within a few µm. These radioactive atoms diffuse out of the catcher, effuse in the cavity, are surface ionized and finally accelerated to form an ion beam.

3 First target heating tests.

The cavity was maintained at a temperature of 1620 K for 8 hours with a vacuum pressure between 10^-6 and 10^-7 mbar. The target’s appearance progressively changed: The foil shrank and tore, and the surface changed from glossy to mat (Fig. 2). A SEM (Scanning Electron Microscope) observation (Fig. 3) revealed Ni crystals and fusion at the torn edges. At 1620 K pure Ni does not fuse but when exposed to carbon the fusion temperature drops to 1599 K [1] for 2% content by weight. Consequently, the cavity temperature must be limited to 1573 K.

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Comparable observations were made with different versions of the TULIP TISS during experiments aimed at not only characterizing the target material, but the overall TISS performances. The lifetime of the target was indeed longer at low temperatures. For temperatures of 1573 K and beyond, the maximum lifetime of the target was observed to be 8 h.

A more systematic study of the out-of-beam target behaviour, using progressive heat was performed. Each Ni foil was placed in a graphite oven and after each (i) heating period \( \Delta t \) (see Table 1), the total length of the foil was measured. A \( \sim \)2×2 mm\(^2\) sample was then taken from the foil and the average size of the grains measured.

### Table 1. Heating test; duration, temperature, pressure versus sample number \( i \), with observed relative shrinking and grain sizes. The calculated “annealing magnitude” (see text) is given in column 5.

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Outgoing time at 473 K</th>
<th>Heating temperature</th>
<th>Heating time</th>
<th>( \sum \lambda_i \Delta t_i )</th>
<th>Relative shrinking</th>
<th>Average grain size</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>hours</td>
<td>K</td>
<td>hours</td>
<td></td>
<td></td>
<td>µm</td>
</tr>
<tr>
<td>1</td>
<td>18.6</td>
<td>1079</td>
<td>2</td>
<td>3.4E-05</td>
<td>0</td>
<td>7.58</td>
</tr>
<tr>
<td>2</td>
<td>15</td>
<td>1122</td>
<td>2</td>
<td>1.7E-04</td>
<td>0</td>
<td>8.51</td>
</tr>
<tr>
<td>3</td>
<td>16.6</td>
<td>1243</td>
<td>2</td>
<td>3.8E-03</td>
<td>0</td>
<td>9.13</td>
</tr>
<tr>
<td>4</td>
<td>15.5</td>
<td>1319</td>
<td>2</td>
<td>2.7E-02</td>
<td>0.63</td>
<td>16.4</td>
</tr>
<tr>
<td>5</td>
<td>15.4</td>
<td>1468</td>
<td>2</td>
<td>2.3E-01</td>
<td>1.39</td>
<td>25.1</td>
</tr>
<tr>
<td>6</td>
<td>16</td>
<td>1558</td>
<td>2</td>
<td>1.3E+00</td>
<td>2.86</td>
<td>38.1</td>
</tr>
<tr>
<td>7</td>
<td>3</td>
<td>1566</td>
<td>15</td>
<td>1.8E+01</td>
<td>4.19</td>
<td>45.2</td>
</tr>
</tbody>
</table>

From the observed correlation between relative shrinking and grain size (Fig. 4), the shrinking process was expected to saturate for a heating time just beyond 15 h at a temperature close to 1573 K. This time was estimated by considering the material change was essentially due to the temperature induced movement of Ni atoms. To compare results, an “annealing magnitude” was defined as being the product of \( \Delta t_i \) and the kinetic rate \( \lambda_i \), where \( \lambda_i \) depends on the temperature and on the activation energy for atomic displacement, according to the Arrhenius law [2]. According to the fit of relative shrinking versus \( \sum \lambda_i \Delta t_i \), a maximal shrinkage should be obtained for \( \sum \lambda_i \Delta t_i \) higher than 5.

### 4 An alternative to a pure Ni

Adding another metal to Ni was considered, the intention being to reduce the mobility and evaporation rate of Ni atoms. Instead of using an alloy, a layer of a metal was considered. To avoid the production of isobaric contamination from nuclear reactions with the primary beam, the atomic mass of the added metal should be distant from that of Ni, and so excludes the transition metals of the fourth period in the Mendeleev classification. If the added metal is part of the sixth period, nuclear reactions with the primary beam can lead to alpha emitting nuclei, which then requires specific safety authorization at SPIRAL1. We therefore favored a metal from the fifth period.

To favor nuclear reactions of the Ne primary beam on mainly Ni nuclei, the relative abundance of the added metal must be minimal. The temperature induced phase
changes in the alloy, due to beam wobbling, had to be avoided to limit mechanical stress.

The final choice is based on the two following observations. At a given temperature, the mobility (or self-diffusion) of metallic atoms in their own solid matrix decreases when their fusion temperature increases, as illustrated in Fig. 5.

![Fig. 5. Self-diffusion versus fusion temperature for metals of the fourth, fifth and sixth periods. Diffusion coefficients are extracted from [3].](image)

Secondly, the fusion temperature and lattice parameter or density of the material are strongly related (Fig. 6).

![Fig. 6. Fusion temperature of metals of the fourth (4P), fifth (5P) and sixth (6P) periods versus their lattice parameter. Crystallographic structure is indicated; bcc for body centered cubic and fcc for face centered cubic. Data extracted from [3].](image)

We can say that the atomic mobility depends on the ways offered to the atoms to move. And that the diffusion coefficient of Ni in a metal having a higher fusion temperature than Ni is lower than the Ni self-diffusion coefficient. For example, at a temperature of 1573 K, the diffusion coefficient of Ni in Mo is estimated to \(10^{-12}\) cm\(^2\)/s, compared to the self-diffusion coefficient of Ni of \(10^{-9}\) cm\(^2\)/s. Finally, within the fifth period of the Mendeleev classification, Mo seems to be the best candidate. Regarding the behavior with respect to temperature, the binary phase diagram given in ref. [4] indicates that for up to 20% in weight of Mo present in Ni-Mo alloy, the fusion temperature remains higher than 1693 K, and this fulfills the initial operational aim of 1573 K.

A 4 µm thick Ni foil was then covered with a layer of 0.09 µm of natural Mo on each face and was tested over 30 h at 1523 K. The relative shrinkage observed was lower than 1% and the relative mass evaporation was 0.8%, within the aim of 1% for a duration sufficient for a first irradiation test.

### 5 Mo-Ni-Mo sandwich target

Our observations of longer lived targets suggests that a thicker Mo covering is an appropriate choice. A 300 ± 6 nm thick layer was deposited on each face by KERDRY (https://kerdry.com) using the cathodic sputtering method [5]. The relative Mo weight was equal to 17%, corresponding to a fusion temperature of 1693 K.

The beam intensity to be supported was fixed to 1.5 µA, corresponding to a primary beam power of 150 W, with –30 W deposited in the target. As the target material inevitably transforms from multilayer to alloy due to heating, the emissivity of the target changes and is expected to be intermediate between the Ni (0.19) and the Mo (0.35) values [6]. Assuming the power is evacuated from the target only by radiation, the target temperature ranges from 1190 K to 1390 K. To reduce this temperature span, the target was placed 60 mm upstream from the cavity (Fig. 7), where the temperature induced by the cavity is negligible and estimated to be 640 K according to a simulation performed with the Ansys® software (https://www.ansys.com). To close the cavity, a 2 µm Diamond Like Carbon foil (https://www.micromatter.com) was used.

![Fig. 7. Cross section view of the mechanical layout of the TULIP production system](image)

To test the thermal behavior of the target at on-beam temperatures, expected to be between 1210 K and 1333 K, a test configuration was developed in which the Mo-Ni-Mo target was fixed to the cavity, which served as an oven. The temperature was measured at the center of the target (red Λ in Fig. 8.1) using a disappearing filament pyrometer. The target was heated by mutual radiation from the cavity and the heating electric current flowing through the cavity walls and through the thickness of the target, from right on Fig. 8 up to the exit situated on the left.

The current leaving the cavity is concentrated around the exit (Fig. 8.1), leading to higher temperatures (> 1300 K) and earlier target fusion. After 26 h (Fig. 8.2), the target is no longer connected to the cavity in the exit region, and the hottest points progressively move along the perimeter of the target. After 43 h, the target
foil starts to ripple (Fig. 8.3 and 8.4) and stops rippling after ~65h (Fig. 8.5), when the current no longer has the possibility to flow through the target. Then the target shape stops changing (Fig. 8.6). This evolution and the aperture of the target explain the fluctuations of the measured temperatures and the variations of the heating current, which rose from 300 A to 350 A in order to maintain the temperature within the targeted range.

Fig. 8. Pictures of the Mo-Ni-Mo target versus heating time. The position of the pyrometer filament is indicated in FIG. 8.1. The relative brightness of the different pictures do not allow a comparison between temperatures as the pictures were taken with different camera settings.

After 98 h, the target foil was almost as glossy as it was before heating, suggesting weak crystallization. The flexibility was comparable to the initial one. The evaporated mass was equal to 10% and the relative shrinkage was lower than 1%. With respect to the Ni target requirements for TULIP, the present target seems to fulfil the requirements.

6 Target under 22Ne primary beam

Conserving the mechanical layout as shown in Fig. 7 and the 0.3 µm Mo/4 µm Ni/0.3 µm Mo targeta 22Ne ion beam impinged the target over 9 h at an energy of 4.5 MeV/A and a power up to 111 W (1.12 µA or 7.0 × 10¹² pps). The ion beam was wobbled on a 18 mm diameter with a 9 mm FWHM profile. The beam power density on the target was estimated to be 8 W/cm² at maximum.

After irradiation, tracks on the target revealed a clear misalignment of the beam on the target surface (Fig. 9). In places where the beam misses the target, the temperature is estimated to be 640 K, and the target appearance did not change. On the positions corresponding to the FWHM beam diameter, the maximum beam power density is estimated to be 6 W/cm². The final appearance of the surface is slightly mat, revealing a change in the metal structure. On the rotating beam diameter, where the beam power density is maximum (~8 W/cm²), part of the target has disappeared (the hole in Fig. 9).

At a beam power density of 8 W/cm², the estimated target temperature ranges from 1190 K to 1390 K. This temperature range cannot solely explain the formation of the hole as it is significantly lower than the fusion temperature of 1693 K deduced from the phase diagram given in ref [1]. Since the incident beam characteristics were not well controlled, excessive focusing and greater power density than desired cannot be excluded.

The temperature variations induced by the rotation of the beam are estimated to less than +/- 70 K, superimposed to an average temperature of ~1250 K, assuming a Mo weight percentage of Mo of 17%, an emissivity of 0.22, a heat capacity of 407 J.kg⁻¹.K⁻¹, an alloy mass density of 4.06 × 10⁴ kg/cm², and an incident beam power density of 8 W/cm². With these conditions the operational working point should be in a region of the binary phase diagram where no material structure change occurs. Nevertheless, the operational parameter logbook shows some large beam power variations (from 10 W to 111 W), which could have created undesired mechanical stress provoking the formation of a hole.

From reference [7], the number of atoms sputtered from the Ni surface by ²²Ne⁺ ions should be less than 10, whereas a sputtering rate higher than 2000 is necessary to explain the formation of the hole. The effect of a sputtering created hole can thus be neglected.

Consequently, it is thought that excessive local power densities and beam intensity variations are the most probable contributors to the hole formation.

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