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MIXTURES OF MOLTEN METALS AND GRAPHITE AS TARGETS FOR ON-LINE MASS SEPARATORS

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Abstract

Targets consisting of fine-grained powders of graphite and metals which do not form thermostable carbides have been investigated. The targets were heated to temperatures well above the melting point of the respective metals. A target of Pt powder mixed with graphite powder is found to be suitable for production of neutron-deficient Au isotopes, when irradiated with 910 MeV ^3He projectiles. The yields of neutron-deficient Cd isotopes from a target of Sn powder mixed with graphite powder are also reported.

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1 INTRODUCTION

Refractory carbides have been extensively used as target materials for on-line mass separators [1]–[5]. Owing to their thermal stability and favourable diffusion properties, they provide efficient and stable production of short-lived isotopes of a large number of volatile elements formed through nuclear reactions.

The release of volatile nuclear-reaction products from solid-state targets is generally more rapid than the release from molten-target materials, as shown in refs. [6,7] which also contain a general discussion of the release kinetics. The diffusion velocity is higher in liquid materials than in solid-state materials, but if large amounts of target material are used, the transport length is considerably larger in a liquid material (0.5–1 cm) than in a porous, solid-state matrix (1–5 μm). An additional delay arises because the surfaces of liquid metal targets may easily be covered with a thin solid oxide layer, through which the volatile products have to diffuse before they evaporate. This hindrance for the release cannot be compensated for by the more rapid transport inside liquid material.

In order to obtain improved yields for the most exotic and short-lived nuclides, faster targets are needed. We have therefore tested the performance of targets that consist of fine drops of non-carbide-forming metals suspended in a porous graphite-powder matrix. Our choices were guided by the need for efficient production systems for neutron-deficient Au and Cd. The development of Au beams was motivated by the systematic studies of ground-state properties in this mass region by on-line collinear laser spectroscopy [8], and a faster system for Cd was needed for experiments with ^{97}Cd .

2 EXPERIMENTAL

The ISOLDE mass separators [9,10] are situated at the CERN Synchro-cyclotron (SC), which can deliver a 910 MeV ^3He beam or a 600 MeV proton beam of 2 μA and 3 μA intensity, respectively. In the experiments reported here, the targets were irradiated with the 910 MeV ^3He beam. Owing to the long range of these particles, large amounts of materials (10–300 g/cm²) may be used as targets. The target material is contained in a tantalum cylinder, 20 cm long and 1.9 cm in diameter (fig. 1). In addition to the heating provided by the irradiating ^3He beam, the target may be heated by passing an electrical current through the Ta cylinder. Volatile nuclear-reaction products are released from the target material and diffuse via the transfer line into the ion source. The whole target-ion-source unit is at a potential of 60 kV, and the ions are extracted out of the ion source and accelerated through this potential. Subsequently, the ions pass through the separator magnet, where they are separated according to their mass/charge ratio.

2.1 The platinum/graphite target

The best yields of neutron-deficient nuclei are obtained through spallation reactions, and the optimum conditions are obtained with a target close to the desired product element. The reaction yields of neutron-deficient Au isotopes from slightly heavier elements, e.g. Hg, Tl, Pb, and Bi, are high, but these elements or their chemical compounds are not suitable as target materials, owing to their too high volatility. It was therefore decided to use a target of Pt, which has one proton less than Au, and use a ($^3\text{He}, \text{pxn}$) reaction.

Off-line release studies have shown that mixtures of powders of platinum-like metals and graphite at temperatures around 1900 °C release volatile nuclear-reaction products rapidly [6]. The metal powders must be mixed with graphite in order to prevent sintering. For elements such as Rb, Be, Sr, As, and Se the delay is of the order of seconds.

The Pt/graphite mixture and the Ir/graphite mixture showed an almost identical release behaviour. However, Pt/graphite cannot be heated above 1900 °C because the Pt vapour pressure then exceeds 10^{-3} Torr. The phase diagram of the Pt/graphite system [11] shows that at 1900 °C, Pt will be molten, and the target will be in the form of graphite powder with liquid Pt.

The target material was prepared by thoroughly mixing similar volumes of Pt powder and graphite powder. The particle sizes were of the order of 1–5 μm . The target material was shielded from contact with the Ta cylinder by means of an inner graphite tube, and outgassed at 1900 °C. The target contained a total of 11.9 g Pt, 24 g C, and the thickness was 7.93 g Pt/cm².

The ion-source used was of the ISOLDE hot-plasma type [12], with an operation temperature of 1900 °C. The ionization efficiency of this source for Au was measured off-line by introducing 60 μg of stable Au in the empty Ta container of the target and ion-source system shown in fig. 1. The Au slowly leaked into the ion-source as the container was heated. By integration of the resulting Au ion-beam, an efficiency of $43 \pm 10\%$ was obtained.

The elements immediately below Pt are refractory. Thus, a target consisting of Pt and graphite irradiated with ^3He will only release isotopes of Hg and Au in the mass region higher than 175. The cross-sections for the production of Hg isotopes through ($^3\text{He},\text{xn}$) reactions are 1–2 orders of magnitude smaller than the cross-sections for the production of their Au isobars through ($^3\text{He},\text{pxn}$) reactions. Therefore, beams containing predominantly Au isotopes with a small admixture of Hg isobars were expected.

The measured yields of Au and Hg isotopes are given in fig. 2. The intensities are given in terms of atoms per second and particle μA of the 910 MeV ^3He -beam incident on the target. High yields of Au isotopes are obtained, with a maximum of 1.6×10^8 atoms/s obtained for ^{192}Au . This is 50 times more than what is obtained at the ISOCELE separator, which uses the Pt(p,xn)Au reaction at a proton energy of 200 MeV [13].

The delay properties of Au in the target was also measured, using the method described in refs. [6,7]. This measurement was performed by monitoring the activity at mass-190 as a function of time after switching off the ^3He beam. A rapidly decreasing curve indicates a fast release of Au from the target. The results, corrected for the decay of ^{190}Au , are shown in fig. 3. The release curve can be qualitatively decomposed into one short and one long exponential components. The characteristic half-times, i.e. the time elapsed before the activity reaches 50% of the start value, are 40 s and 12 min, respectively (see ref. [6] for details). The long component accounts for more than 90% of the total amount of Au produced in the target. Since the mass-190 beam includes a $\sim 2\%$ component of the shorter-lived ^{190}Hg isotope, the short delay component may be due to ^{190}Hg and not to ^{190}Au . This is in qualitative agreement with the observation [6] that molten-metal targets always display single-exponential delay functions and that the delay half-time of Hg from molten Pb is ~ 30 s [6].

A delay of the order of minutes is not satisfactory when short-lived Au isotopes shall be studied, but it seems difficult to improve these conditions for Au. During the above-mentioned Au efficiency measurement, we found that the delay was of the order of several minutes, even in an empty Ta container. The crucial source of the delay therefore seems to be the slow desorption rate of Au from surfaces in the container, rather than a slow release from the Pt/graphite target. This observation is consistent with desorption measurements and theoretical calculations of adsorption enthalpies [12,13], which show

that Au is strongly adsorbed to metal surfaces and is released much more slowly than the other noble metals Ag and Cu.

Table 1 gives other selected yields from the Pt/graphite target system. As expected, the target also functions well for elements which are not refractory and do not form thermostable carbides. The elements forming stable carbides, e.g. the rare earths, are strongly delayed in the target, and the yields are down by 4–5 orders of magnitude compared with pure metal targets, even for nuclides with half-lives of several hours.

2.2 The tin/graphite target

The material for the Sn/graphite target was prepared in a slightly different way, by mixing graphite powder with SnO_2 and pressing the mixture to pellets. Subsequently, the material was heated to a temperature of 1100 °C where the SnO_2 is reduced by graphite to metallic Sn. In this way, a porous graphite matrix containing molten tin drops was obtained. The target was connected to the ion source through a transfer line kept at a temperature of 500–600 °C, which allows Cd to pass, but stops less volatile elements. The mechanical construction of the system is shown in fig. 4. The target thickness was 20 g Sn/cm^2 . The target material was contained in an internal graphite cylinder, and contained a total of 31 g Sn and 20 g C. The target operation temperature was around 1000 °C.

The production yields of Cd isotopes are presented in fig. 5. The Cd yields are slightly different from those obtained from a pure Sn target irradiated with 600 MeV protons [14]. For the more long-lived Cd isotopes near the maximum, the yields from the Sn/graphite target are a factor of 5–10 lower, while for the short-lived Cd isotopes, the yields from the two targets are almost identical. The delay of Cd was measured by monitoring the decreasing activity of the 5.5 min ^{102}Cd after switching off the ^3He beam. The delay curve, corrected for decay, is shown in fig. 6. The characteristic half-time of the delay curve is of the order of 30 s, which is a considerable improvement over the 225 s observed from a traditional liquid-Sn target [6]. However, the introduction of graphite lowers the Sn density of the target by a factor of 6, and the production yields are therefore not improved.

2.3 Other possible target systems for the ^3He beam

The Hg contamination and the slow release of the Au may both be overcome by replacing the Pt/graphite with Ir/graphite, which may be operated at a temperature of ~ 2300 °C. Judging from the results obtained with Au production from the Pt/graphite target, it seems very likely that a target of rhodium and graphite irradiated with ^3He will provide good yields of neutron-deficient Ag isotopes. Silver will be much more rapidly released than Au [13], and the Rh/graphite target should therefore be particularly favourable for Ag. It is also possible that this target may be an efficient one for the production of neutron-deficient Pd isotopes.

Similarly, it is also very likely that neutron-deficient Zn and Cu can be produced efficiently from a target consisting of a mixture of graphite powder and nickel carbide irradiated with ^3He .

3 CONCLUSION

Fine drops of liquid Pt metal suspended in a graphite powder matrix is a suitable target for the production of mass-separated Au isotopes. The delay is of the order of several minutes. The conditions are thus not quite satisfactory for the most short-lived Au

isotopes, i.e. those with half-lives in the second range, but Pt/graphite probably provides the optimum conditions for Au production.

For the production of neutron-deficient Cd isotopes, the Sn/graphite target functions in a satisfactory way, and has more favourable release properties than the liquid-Sn target, but the production yields of short-lived Cd isotopes are not improved, because of the low target material density.

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Table 1: Yields from the Pt/graphite target used in combination with a hot plasma-discharge ion-source

${}^6\text{He}$	1.7×10^7	${}^{74}\text{Ga}$	4.9×10^4
${}^8\text{Li}$	4.4×10^6	${}^{71}\text{Se}$	5.5×10^3
${}^{20}\text{F}^+$	4.2×10^3	${}^{77\text{m}}\text{Se}$	2.0×10^5
${}^{22}\text{F}^+$	4.9×10^2	${}^{74}\text{Br}$	3.9×10^4
${}^9\text{Be}{}^{20}\text{F}^+$	2.4×10^4	${}^{74\text{m}}\text{Br}$	5.5×10^4
${}^9\text{Be}{}^{21}\text{F}^+$	1.3×10^4	${}^{77}\text{Br}$	2.3×10^6
${}^9\text{Be}{}^{22}\text{F}^+$	2.0×10^3	${}^{77\text{m}}\text{Br}$	6.8×10^6
${}^{27}\text{Al}{}^{22}\text{F}^+$	1.1×10^3	${}^{78}\text{Br}$	1.2×10^7
${}^{19}\text{Ne}$	2.5×10^3	${}^{77}\text{Kr}$	1.8×10^6
${}^{24}\text{Na}$	1.1×10^6	${}^{104}\text{Ag}$	3.7×10^6
${}^{25}\text{Na}$	1.2×10^6	${}^{105\text{m}}\text{Ag}$	3.5×10^6
${}^{27}\text{Mg}$	1.5×10^5	${}^{112}\text{Ag}$	4.2×10^6
${}^{40}\text{Cl}$	2.2×10^4	${}^{105}\text{Cd}$	1.3×10^6
${}^{39}\text{Cl}{}^{138}\text{Ba}^+$	2.5×10^5	${}^{111\text{m}}\text{Cd}$	7.6×10^6
${}^{40}\text{Cl}{}^{138}\text{Ba}^+$	1.1×10^5	${}^{111}\text{In}$	2.2×10^7
${}^{41}\text{Cl}{}^{138}\text{Ba}^+$	4.4×10^4	${}^{112}\text{In}$	1.3×10^6
${}^{42}\text{Cl}{}^{138}\text{Ba}^+$	1.2×10^4	${}^{112\text{m}}\text{In}$	2.2×10^6
${}^{43}\text{K}$	1.3×10^6	${}^{111}\text{Sb}$	3.7×10^3
${}^{45}\text{K}$	4.5×10^5	${}^{112}\text{Sb}$	3.7×10^4
${}^{57}\text{Mn}$	1.0×10^3	${}^{118\text{m}}\text{Sb}$	2.2×10^6
${}^{62}\text{Cu}$	1.2×10^4	${}^{118\text{m}}\text{I}$	1.0×10^6
${}^{66}\text{Cu}$	1.8×10^6	${}^{123}\text{Xe}$	4.7×10^6
${}^{63}\text{Zn}$	3.8×10^5	${}^{123}\text{Cs}$	4.6×10^6
${}^{71}\text{Zn}$	3.2×10^5	${}^{123\text{m}}\text{Cs}$	4.1×10^6
${}^{71\text{m}}\text{Zn}$	2.9×10^6	${}^{157}\text{Dy}$	8.2×10^5
${}^{72}\text{Zn}$	3.0×10^5	${}^{157}\text{Tm}$	1.6×10^4
${}^{74}\text{Zn}$	1.1×10^5	${}^{157}\text{Yb}$	1.4×10^3

Figure captions

- Fig. 1** Plasma-discharge ion-source connected to the target via a transfer line kept at 1900 °C. The line, the cathode, and the discharge chamber are all heated to 1900 °C by means of the same current.
- Fig. 2** Production yields of Au and Hg isotopes from a Pt/graphite target (7.9 g Pt/cm²) at 1900 °C, irradiated with 1 particle μ A of 910 MeV ³He beam. The lines are only to guide the eye.
- Fig. 3** Activities of ¹⁹⁰Au samples produced after ³He-beam cut-off (delay curve), corrected for decay losses; Pt/graphite target at 1900 °C.
- Fig. 4** Plasma-discharge ion-source connected to the target via a transfer line of 500–600 °C.
- Fig. 5** Production yields of Cd isotopes from a Sn/graphite target (20 g Sn/cm²) at 1000 °C, irradiated with 1 particle μ A of 910 MeV ³He beam. The lines are only to guide the eye.
- Fig. 6** Activities of ¹⁰²Cd samples produced after ³He-beam cut-off (delay curve, corrected for decay losses; Sn/graphite target at 1000 °C.

Fig. 1

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Fig. 6