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RECENT DEVELOPMENT OF HIGH TEMPERATURE METAL TARGETS FOR ISOLDE

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ABSTRACT

The improved target and ion-source system to be used at the SC-ISOLDE on-line mass separators is presented. Tests of a number of refractory metals with respect to their use as target materials are described. The materials discussed are pure metals and mixtures of graphite and platinum-like metals. Rapid release of a number of nuclear reaction products from powders and foils of Ti and Th is shown. The release properties of Re and Ir are described. The enhanced release of refractory elements obtained from these metals by means of reaction with CF_4 are related to the importance of in-grain diffusion versus surface desorption. The difference in performance between powder and foil targets is discussed.

On-line tests of some of these materials combined with new high-temperature plasma-discharge type ion-sources are described and potential use of yet untested systems is discussed.

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1. INTRODUCTION

Pure metals are among the most successful target materials for on-line mass separators. In particular, solid targets of refractory metals are excellent for this kind of use since they release no impurities which may interfere with the functioning of an ion source. In the present paper, we will describe the latest development of ISOLDE ion sources and their combination with a number of new metallic target materials and outline the current and potential performance of the new systems.

The ISOLDE mass separators¹⁻²⁾ are situated at the CERN synchro-cyclotron (SC), which provides a 910 MeV ^3He beam and a 600 MeV proton beam which may reach an intensity of 4 microampere. Due to the long range of these projectiles, large amounts of target materials 10-300 g/cm² may be used at ISOLDE. The intensities of the secondary beams of unstable nuclei reaches 10^{11} atoms/s per microampere of incident proton beam.

2. THE ISOLDE TARGET AND ION SOURCE SYSTEM.

The ISOLDE target and ion source assembly used for the experiments reported in this paper is shown in fig.1. The target material is usually contained in a Ta cylinder, 20 cm long and 1.9 cm in diameter. The target is resistively heated by leading a high current through it in addition to the heat supplied by the irradiating beam. Volatile nuclear reaction products are released from the target material and diffuse via a transfer line into the ion source. Target and ion source is mounted on vacuum feed throughs situated in an watercooled base plate and enclosed by an aluminum bell jar. The unit is coupled to the acceleration chamber of the separator by means of quick connectors and a pumping port which can be sealed off with a valve. The target and ion source form a selfcontained unit specifically optimized for each element or group of elements. Transport to and from the separator is done remotely by means of an industrial robot as shown in fig.2.

The choice of ion source to be combined with a large number of different target materials has so far mainly been dictated by efficiency and secondly by its capability of selective ionization. A number of other properties like energy spread and emittance have largely been neglected, but are presently under study³⁾. The widespread use both in basic research and industry of stable heavy-ion beams has lead to the development of a large variety of ion sources, unfortunately with little emphasis on efficiency. Most of our inspiration for development of sources comes from work in this field which is reviewed by Freeman and Sidenius⁴⁾ and is the subject of a number of conference proceedings⁵⁻⁶⁾. High efficiency sources dedicated to on-line mass separation are much rarer and these have been reviewed by Kirchner⁷⁾.

Ions of many elements and of a number of molecules can be produced in a positive surface ionization process. Several systems which operate along these lines have been developed at ISOLDE for ionization of alkali elements (Group I), alkaline earths (Group II), the elements Al, Ga, In and Tl (Group III), Sc, Y, and the lanthanides (Group IIIb) ¹⁾. The inverse process to positive surface ionization occurs when an atom with a high electron affinity gains an electron by impinging on a surface with a low electron work function. An ion source based on this principle has been developed at ISOLDE ⁸⁾ producing negative ions of elements with an electron affinity larger than 2.5 eV. The major advantage of this source is the high efficiency and selectivity with which ion beams of the halogen (Group VII) elements can be produced.

The remaining elements will have to be ionized in a plasma discharge. Many different versions of such sources are used at on-line mass separators. The plasma-discharge ion-source currently in use at ISOLDE is based on the Forced Electron Beam Induced Arc Discharge (FEBIAD) concept originally developed by Kirchner ⁹⁾. Its principle is that electrons from an indirectly heated disc shaped cathode are accelerated into the anode chamber by means of a grid. The ISOLDE sources based on this principle exist in several versions depending on the transfer-line temperature to be used.

Some of the ion sources are quite sensitive to running conditions and the high ionization efficiencies quoted in this paper can only be obtained under clean conditions. The positive surface ionizers are the least sensitive ones and can be used in connection with a variety of target materials. The ionization efficiency is relatively insensitive to impurities from the target material.

The most sensitive type of ion source is the negative surface ionizer. The ionizer surface is LaB₆, with a work function around 2.7 eV when it is clean, but contamination of this surface tends to increase the work function and thereby decrease the ionization efficiency. For example, carbide targets lead to a slowly growing carbon contamination on the ionizer ¹⁰⁾, probably arising from carbon transport due to CO formed by a rest gas pressure of oxygen. The plasma ion-sources are normally shielded from the outgassing of the target by a temperature controlled transfer-line, preventing the non-volatile impurities to diffuse from the target to the ion source. In this way, a chemical selectivity is achieved as well for elements e.g. Cd, Hg and the noble gases. However, in order to obtain beams of less volatile elements, the line must be kept at relatively high temperature. Under these conditions, the choice of target materials is very restricted. A too high outgassing rate from the target material will cause intolerable conditions for running the ion source in a stable and efficient mode. In a system like ISOLDE, which utilizes large amounts of target material, the purity of the target is of crucial importance. Solid state targets of refractory metals are the purest materials available for use in on-line mass-separators.

2.1 Positive surface ionization source with Ta or Re ionizer

This source consists, as shown in fig.1, of a constriction at the end of the transfer tube which is

heated resistively by a current of 120-400 A. The ionization effect takes place on the interior surface of the tubular constriction.

With a constriction made of tantalum kept at a temperature of 1000 °C the alkalis K, Rb, Cs, and Fr are selectively ionized with an efficiency of about 90 %. If the constriction is lined with a Re foil and heated to 1700 °C, it also ionizes the alkaline earths Ba and Ra with 10 to 50 % efficiency.

2.2 High temperature surface ionizer with W cavity

If a W tube heated to 2400 °C is used as the ionizer shown in fig.1, the source will ionize elements with $W < 6.5\text{ eV}$. In this case also other ionization phenomena contribute to a somewhat higher ionization efficiency than given by the Saha-Langmuir formula (see the discussion by Kirchner⁷⁾). The ionized elements are Ti, the lanthanides, In, Y, Sr, Ga, Sc, Ca, Al, Na and Li. The efficiencies for this group of elements vary from 5 to 50 %. A number of molecules are also efficiently ionized by the different versions of the surface-ionization source.

2.3 Negative surface ionization source

The principle and construction of the source is shown in fig. 3. The negative ion emitter onto which the product flow impinges consists of a small LaB_6 pellet ($F = 2.7\text{ eV}$), situated in the constriction of the transfer line. In order to assure an adequate pumping speed of the target material, an annular shaped opening is left around the pellet through which about 50% of the product atoms escape without hitting the ionizer. The abundantly emitted electrons are deflected onto a separately powered electrode by means of a permanent magnetic field. Ionization efficiencies of 1-40% for Cl, Br, I and At have been obtained. An as yet unused potential of this source is the ionization of volatile chemical compounds with high electron affinity.

2.4 Plasma discharge ion sources with cooled line at 30°C

For ionization of the noble gases (GROUP 0) the system shown in fig.4 is used. A watercooled copper section is inserted in the transfer line between target and ion source. In this way only elements or compounds which are gaseous at room-temperature are admitted to the source which is kept at a temperature of 1000 °C. Note that the cathode in the ISOLDE version is ohmically heated. The efficiencies achieved for He, Ne, Ar, Kr, Xe, and Rn are 0.5%, 2%, 5%, 15%, 30%, and 40% respectively.

2.5 Plasma discharge ion source with line at 400°C

A source very similar to the one discussed in section 2.4 is shown in fig. 5. The only difference is that the line is kept at an intermediate temperature of about 200 - 400 °C in order to still transmit

relatively volatile elements, but by condensation prevent vapours from the target to reach the source. This is achieved simply by giving the stainless steel line

a small heat-transfer contact-area through which the heat is conducted to the watercooled mounting plate. The source is mainly used for the elements Zn, Cd and Hg (Group IIb), for which efficiencies of 10 to 60% are obtained.

2.6 Plasma discharge ion source with high temperature line at 1900 °C

In the version shown in fig.6 the line directly ends in the disc shaped cathode and the same current is used to heat the line, the cathode and the body of the ion source. This construction has the advantage that the less volatile products are transferred to the source at high temperature without encountering any cold surfaces where they could be lost by adsorption. By means of heat screening the temperature obtained is 1900 °C. This source is intended to ionize the less volatile elements from the Groups Ib, III, IV, and V and also to be used in conjunction with CF₄-addition.

3. OFF-LINE RELEASE TESTS

In this chapter, we demonstrate the release of nuclear reaction products from a number of different metals. The experiments have been done by irradiating a sample in the SC beam. The radioactive tracers formed in the material are used to investigate how large a fraction of a particular tracer element is left in the sample as function of time at a given temperature. A rapidly decreasing release curve therefore indicates a useful target material. These experiments are described in detail in refs. 11-12. The first studies of metal powders were reported by Carraz et al.¹¹⁾, who found a variety of different release properties. For instance, the release of Yb was shown to be rapid from Hf powder, somewhat slower from Ta powder and very slow (with less than 50% of the activity released after 1 hour) from W powder. In fact Hf is a very suitable target material for Yb. Unfortunately, sufficient amounts with the required purity could not be obtained at reasonable cost, and it has never been used on-line. A satisfactory Yb production is, however, obtained from Ta powder (sect.4.2). Similar results were obtained for Rb, Sr and Y with powders of pure Nb and Mo¹¹⁾. Niobium powder has been frequently used at ISOLDE, particularly for production of Br, Kr and Rb.

The release of nuclear reaction products from solid refractory materials is governed by two processes; (1) the in-grain diffusion inside the crystals and (2) the desorption from the surface of the crystals.

It is important for the target development work to know which one of these processes is the main source of delay for a particular combination of target material and evaporating reaction product. This problem was extensively studied for Ta foil by Beyer and Novgorodov¹³⁻¹⁴⁾, who found that in many

cases the surface desorption determines the release rate.

If the surface desorption is the dominating process, improved release properties may be obtained by diminishing the total surface of the target. This can be done by means of a target consisting of foils instead of powder. Foil targets will also be superior to powder targets with respect to sintering. However, some metals are not readily available as thin foils.

3.1 Release properties of rhenium

In fig. 7, the liberation of Hf and three lanthanide elements from Re powder is shown. As can be seen from fig. 7a, the release from pure Re powder is slow, and this system is clearly not applicable for the production of short-lived nuclides. In order to investigate if the delay is due to in-grain diffusion or to surface desorption, a release study was performed with controlled addition of CF_4 gas (fig. 7b). A certain improvement was obtained for Hf, but not for the lanthanide elements. This result shows that the delay is mainly due to slow in-grain diffusion or long diffusion paths, since it has been shown¹⁵⁾ that CF_4 improves the surface desorption rate of rare earth elements due to formation of volatile fluorides.

The slow in-grain diffusion probably arises from long diffusion paths due to excessive grain-growth as a result of sintering. Consequently, similar experiments were performed on a mixture of Re powder and graphite powder (equal volumes). Re does not form a refractory, stable carbide, and the admixture of graphite powder as a dilution agent should therefore efficiently prevent sintering. The release from the Re/graphite mixture is shown in fig. 7c, and the release properties have clearly improved. If the release test is performed at a partial pressure of 5×10^{-4} torr CF_4 , which is acceptable to the ISOLDE isotope separators, all the investigated elements, including Hf, are rapidly released. The moderate reaction rate of Re metal with fluorine-containing gases¹⁶⁾ at high temperatures assures its performance as target material also in the presence of CF_4 . Similar results have been obtained also for platinum-like metals¹⁷⁾.

3.2 Tantalum powder and tantalum foil

The release of rare earth elements, Hf, Ba, and Cs from Ta has been extensively studied by Beyer and Novgorodov¹³⁻¹⁴⁾, who found that the in-grain diffusion is most rapid for the lanthanides, followed by Ba, Hf, and finally Cs. For the surface desorption, the sequence is nearly the reverse, with Cs being fastest, followed by Ba, lanthanides and Hf. For the elements Rb, Sr, Y, and Zr, similar results are found. Thus, in a target of Ta powder, the rare earth elements will be delayed compared to Cs and Ba, due to the large total surface. Consequently, in a target of Ta foil, the volatile rare earth elements will be released more rapidly, because the total surface is diminished. Both systems have been

tried on-line, and the results are described in section 4.2.

Beyer and Novgorodov¹⁸⁻¹⁹⁾ also investigated different refractory metals close to Ta for the production of lanthanides, and found that, Hf possesses the most rapid diffusion and desorption properties for these elements, while W had much less favourable release rates. The fact that W can be heated to higher temperatures cannot compensate for this difference.

Therefore, the best target system for production of neutron-deficient isotopes of rare earth elements is a Hf foil target. Similarly, the best system for production of neutron-deficient Y and Sr is a target of Zr foils or plates²⁰⁾.

3.3 Titanium powder and titanium foil

Experiments by Carraz et al.¹¹⁾ showed that Ti powder is a poor target material because of rapid sintering even at 1500 °C. Since Ca and Sc form stable, refractory carbides and oxides, they are hardly released from any carbide or oxide target system. In a Ti foil target at 1600 °C, sintering should not occur, and no thermostable, non-volatile compounds of the product elements are formed.

The release of Sc, Ca, and K from Ti foil is shown in fig. 8. Sc is most rapid, followed by Ca and K, reflecting the difference of in-grain mobility. These results are in accordance with previous measurements²⁰⁻²¹⁾. The off-line experiments indicate that Ti foil is a promising target system for Ca and Sc, and that the sintering problems are avoided. It would be an advantage, however, to avoid Ta surfaces in the system, since we have observed much slower release of Sc from a Ti foil packed in a Ta foil.

4. ON-LINE TESTS

4.1 The iridium/graphite system

A mixture of iridium metal and graphite has for a long time been used with success by the Orsay group²²⁾. This system behaves similarly to the Re/graphite system discussed in section 3.1.

The Ir/graphite system has been particularly efficient for production of exotic fragmentation products, and the system was therefore tested at ISOLDE for the production of neutron-rich Li and Na isotopes. The yields of Na from this target is shown in fig. 9, and are comparable to those obtained from a uranium/graphite target²³⁾. It is also likely that the Ir/graphite system will produce elements like Be, Al, and Sc when CF₄ is added to it, but this is yet to be verified.

The off-line tests indicate that Re/gr is a more favourable system than Ir/gr¹²⁾, and it can also be heated to higher temperatures. On-line tests of Re/gr targets are planned.

4.2 Tantalum metal systems

The Ta powder target has been used at ISOLDE for a long time. Off-line tests¹¹⁾ showed that the only rapidly released element was Yb. As the most volatile one of the lanthanides, Yb can be produced from Ta powder with a reasonable degree of selectivity. The surface delay of a metal atom on a metal surface roughly follows the same dependence as the vapour pressure. The more refractory lanthanide elements are therefore more strongly delayed on the surface in the target.

A selective production of Yb is therefore observed from a Ta powder target connected to a W surface ionizer (fig. 10). Also some other rare earth activities are observed, but the yields are suppressed by 2-3 orders of magnitude compared to Yb.

Since the selectivity of the Ta powder target was based on the high surface delay, one should expect that a drastic decrease in the total surface would increase the yields of the other rare earth elements as discussed in ref. 6. By replacing the Ta powder with Ta foil, the total surface in the target is diminished by a factor of roughly 100; the exact figure is hard to estimate accurately. The yields of rare earth elements from such a target is shown in fig.11. Apart from the replacement of Ta powder with rolls of Ta foil, the conditions are identical.

As expected, the foil target behaves completely differently from the powder target. All lanthanide elements are now produced with good efficiency, and the selective behaviour vanishes completely. Even the most refractory rare earth element, Gd, is observed with good yields. Obviously, it is difficult to perform traditional nuclear spectroscopy on beams from this type of target, but it has turned out to be extremely useful for experiments which do not require selective beams, e.g. colinear optical spectroscopy.

The comparison between the Ta powder and the Ta foil target shows the importance of basic studies in the target development work. Unless it is known that the surface desorption is the main source of delay, it makes no sense to replace the powder with foils.

4.3 Targets around A=45

Attempts have been made to produce neutron-deficient isotopes of Ca from scandium carbide, but the success was rather limited due to the stability of calcium carbide. From a uranium carbide target, which can be heated to much higher temperatures, on-line beams of neutron-rich isotopes of Ti, Sc, and Ca have been observed²⁴⁾ in low yields.

From off-line tests described previously, it is expected that a pure metal target would give good yields of neutron-deficient Ca, and possibly also Sc. A target of Ti foil combined with a W surface ionizer will therefore be tested for the production of Ca and Sc in the near future.

Vanadium foil will not be tested on-line, since we in all respects have found it inferior to Ti foil as production target for Sc and Ca.

4.4. Targets around A=90

Nb powder has for a long time been used as target at ISOLDE for the production of neutron deficient Rb²³), Br⁸), and Kr²⁵). Neutron deficient Sr and Y has not been observed at ISOLDE. For the production of neutron deficient Sr and Y, a Nb powder target will probably not be the most rapid one. From investigations of the release of rare earth elements from Ta¹³⁻¹⁴), it is evident that a foil target should be used for Y production, and Zr is the most favourable target material for both elements²⁰).

5.CONCLUSION

A number of refractory solid metals have been shown to be very suitable as target materials in mass-separators on-line. The liberation of nuclear reaction products depend upon three main factors; (1) in-grain diffusion, (2) surface desorption, (3) the speed of sintering and purity.

If the surface desorption is decisive for the release process, strongly improved yields can be obtained by minimizing the total surface of the target. This has been done by means of foil targets, In metal powder targets, the sintering effect can be diminished through the admixture of graphite powder, unless the metal in question forms a stable carbide, in which case the release properties may be totally different. The further improvement in the release rates which are obtained through the addition of reactive gases may be of great importance for many elements, and on-line tests in these fields are in progress.

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FIGURE CAPTIONS

- Fig. 1. The ISOLDE target and ion source unit equipped with a tubular surface ionization source.
- Fig. 2. An industrial robot is used to transport the ISOLDE target units to and from the target store and the separator.
- Fig. 3. Negative surface ionization source.
1. Permanent magnet	5. Electron suppressor electrode
2. Pole piece for electron suppressor	6. Insulator
3. Transfer line constriction which contains the LaB_6 pellet	7. Mounting plate
4. Product transfer-line	
- Fig. 4. Plasma-discharge ion-source connected to the target via a watercooled transfer-line.
- Fig. 5. Plasma-discharge ion-source connected to the target via a transfer-line kept at 400°C .
- Fig. 6. 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. 7. Release of nuclear reaction products from rhenium powder or a rhenium/graphite mixture.
- Fig. 8. Release of nuclear reaction products from Ti foil.
- Fig. 9. Production yields of Na isotopes from an Orsay type, 13 g/cm^2 iridium/graphite target connected to a positive surface ionizer made of W. The points are normalized to a bombarding proton beam of 1 microampere and the curve is only drawn to guide the eye.
- Fig. 10. Production yields of lanthanide isotopes from a 130 g/cm^2 Ta powder target irradiated with a 1 microampere proton beam. The curve is only a guide.
- Fig. 11. Production yields of lanthanide isotopes from a 122 g/cm^2 Ta foil target connected to a positive W-surface ionizer. Individual measurement points are not indicated.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

0 5 10 cm

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10

Fig. 11