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METHODS FOR PRODUCTION OF INTENSE BEAMS OF UNSTABLE NUCLEI: NEW DEVELOPMENTS AT ISOLDE

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ABSTRACT

An overview of progress in the target and ion source techniques at the ISOLDE on-line mass separator is given. The production of high intensity beams of mass-separated radioactive nuclei by bombardment of targets with 600 MeV protons and 910 MeV ^{3}He from the CERN synchro-cyclotron is discussed. Off-line tests performed in order to clearify the release properties of different target materials are described. The targets are metal powders or foils, alloys, carbides, oxides, intermetallic compounds or molten metals. The influence of reactive gases on the release rates and progress in ion-source techniques are also discussed. Recent on-line tests are described in detail, and systems are suggested for the production of elements which are not yet available as primary products in on-line mass separators.

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1. INTRODUCTION

On-line isotope separation [1] is a technique in which the ion source of an electromagnetic mass-separator is coupled directly to an accelerator target so that a continuous beam of unstable nuclei emerges. Beams of this kind have a wide range of applications in nuclear, atomic and solid state pysics and have played an important role in extending our knowledge of nuclei far from stability, see e.g. the reviews [2]. The present paper describes progress in the experimental techniques for making such beams at the ISOLDE facility coupled to CERN's 600 MeV synchro-cyclotron (SC).

The reason why high-energy protons are especially useful as projectiles for on-line mass separation is their long range, which permits the use of very thick targets, up to 300 g/cm^2. This leads to production yields for the most abundant products as high as 10^{11} atoms per second per µA of incident proton beam. The selective release and subsequent ionization of submicroscopic amounts of products in the presence of the bulk of target material is a major challenge to the experimentalist and one that can only be met through a combination of techniques from high temperature physics, metallurgy and chemistry. The main part of the present paper describes progress in laboratory tests of new target systems (Section 4), in the development of ion sources (Section 5) and in on-line tests of completed prototype units (Section 6). We begin by a brief summary of the main features of the existing installation.

2. The ISOLDE facility and target system

A schematic view of the SC-ISOLDE mass separator facility [3] is given in fig. 1. The 600 MeV proton beam (with an intensity up to 4 µA) or 910 MeV ^{3}He beam from the SC irradiates a target situated on one of the two mass separator stations. The nuclear reaction products formed in the up to 170 g/cm^2 thick targets are thermalized within the solid or liquid target matrix. Subsequently they evaporate from the bulk material into the vacuum and diffuse to a suitable ion source. The target and the ion source are kept at 60 kV potential, and the ions are extracted from the source and accelerated through this potential. After mass separation the beams of radioactive nuclei are directed to the experiments via a beam line system.

The target and ion-source system is thus of crucial importance since it determines entirely by its performance (number of elements, purity, speed, yield, emittance) the quality and intensity of ISOLDEs beams. Today, methods have been developed to make radioactive ion beams of the 66 elements shown in fig. 2, the intensities of which can be found in the beam catalouge [3].

In the following, we briefly describe the basic principles of targets for on-line mass-separation. An earlier overview of ISOLDE systems was given by Ravn in 1979 [4]. On-line performance of a number of other systems has also been described in several articles [5 -9].

2.1 The target and ion source principles

Spallation, fragmentation and fission processes produce a wide range of both neutron-deficient and neutron-rich nuclei. With a proton intensity of $I = 1\ \mu A$ the production rate in a target of $N = 10^{24}$ atoms/cm^2 becomes $A = I*N*\sigma = 10^{11}$ atoms/s for a nuclide with a formation cross-section of $\sigma = 10$ mb. The general problem is now one of mass transfer, i. e. to continously separate the produced 10^{11} nuclei from the 10^{24} target atoms, and subsequently convert them into an ion beam. The different steps of this process are schematically illustrated in fig. 3 together with the losses from various effects. These determine the overall yield, which is the most important single parameter of the system. Each of these steps which are described briefly below can be studied separately off-line by means of radioactive tracer methods.

Target material and product release.

The target materialmust be chosen so that it will release the element of interest, and so that the nuclear reaction cross-section is favourable. This can almost only be achieved by heating a low vapour-pressure target material to a temperature at which the reaction products of interest are released by diffusion, desorption and occasionally also by chemical processes.

Target to source transfer

The chemical affinity of the released species to the walls of the target container and the transfer tube that leads to the ion source plays an important role since it may cause losses by adsorption. By control of the temperature of this line chemical selectivity may be achieved by means of selective condensation or adsorption.

Ionization

No universal ion source exists for on-line mass separators. The ion source to be combined with the target is optimized with respect to efficiency, selectivity and construction material for the element of interest only.

Decay losses

The losses caused by the decay during the mass transfer process play a more important role for the short-lived nuclei than the trivial losses through leaks in the system. This delay loss can be theoretically described in terms of diffusion and desorption processes often dominated by one specific transfer process. For a detailed discussion we refer to previous papers [4,10]. So far, the fastest systems developed are based on solids in form of foils, wires and powders kept at high temperature. Usually the delay in such systems is described by the distribution p(t) in the values of the delay times t measured from the instance the nuclide is formed until its detection. This p(t), which most often is a sum of exponential functions, allows us to derive the relation between the observed yield Y_0 and and the decay-loss corrected yield Y. For a nuclide with mean lifetime T_m much shorter than the average delay time the yield is given by,

$$Y_0/Y = 3(\mu_s T_m)^{1/2}/\pi ,$$

where μ_s is in this case a delay constant characteristic of a target. where the delay is governed by diffusion in the grains of a powder. The release from a molten target which is characterized by one single exponential function [11], is then

$$Y_0/Y = \mu/(\mu + T_m^{-1}) = \mu T_m \quad \text{(for small } T_m\text{)}$$

The fact that the yield decreases only with the square root of the mean life for a solid powder target and linearly for a liquid system, illustrates that high temperature, solid-state targets are most favourable for production of short-lived nuclei. A direct comparison of two target systems is difficult due to the complexity of the measured delay distributions for solid targets. However, one can always define a "release half-time" for any system, i. e. the time at which 50 % of the activity of a particular element has been released. For liquid targets the release half-time is at best 30 s, for the best solid targets it is less than 1 s.

3. OFF-LINE TESTS OF MATERIALS

Most of the materials used for targets at ISOLDE are refractory elements or compounds.

A target material must satisfy the following conditions:

1. A high formation cross-section for the nuclides of interest.
2. A high release rate (short delay time) in continuous on-line operation for the element of interest with, preferably, element-selective release.
3. High-temperature stability with low vapour pressure ($\leq 10^{-3}$ torr) at the operating temperature.
4. High specific density.
5. Inertness against undesired high-temperature chemical reactions, i.e. with the target container material.
6. Resistance to radiation damage.

The material tests discussed in the present paper only represent an overwiew. Further details can be found in refs. [10,12].

A number of pure elements and chemical compounds which fulfill conditions 1-6 reasonably well and therefore are suitable as target materials have been found. Some of these are frequently in use, others have not yet been tested on-line. They can be grouped into different categories, and in the following, we will discuss their release properties based on the off-line radiotracer tests. We define the release as the fraction of the product left in the samples as function of heating time.

3.1 Metal powders

Powders of refractory metals are among the most often used type of targets at ISOLDE. Already in 1965, Andersen et. al. [13]. described off-line tests of the release of reaction products from this type of materials. More extensive studies have been reported by Carraz et. al. [10] and Hoff et. al. [12].

As an example, we show in fig. 4 the release of Hf and lanthanide elements from rhenium powder under various conditions. In fig. 4a, the release observed when the powder is heated to 1900 °C is shown. There is no observable release of Hf, and the lanthanide elements are liberated relatively slowly. Pure Re metal powder is thus not interesting as a target material for lanthanide production.

In fig. 4b, the release of these elements from the same matrix is shown, but now under the influence of 5×10^{-2} torr partial pressure of CF_4. There is hardly any change for the lanthanides elements, but now there is an observable liberation of Hf due to the formation of volatile HfF_4 and HfF_3. From fig. 3 it can be seen that the improvement obtained by the addition of reactive gases is rather limited in this case, and that the unfavourable release properties of Re are mainly due to slow diffusion inside the grains. Since the addition of a reactive gas may have fast and direct influence only at the surface of the matrix, one concludes that surface desorption is of importance only for Hf and not for the lanthanides. It is likely that the strong sintering of the Re powder is the explanation for this behaviour.

An improvement in this case can therefore only be expected by minimizing the sintering. In the case of Re powder, this is possible by dilution of the metal with graphite powder. Rhenium does not form thermostable carbides, and the two elements act like a mixture of two metals [14]. The release of nuclear reaction products from a mixture of equal volumes of Re-powder and graphite powder is shown in fig. 4c. From this matrix, the lanthanide elements are liberated according to their volatility, Yb being the fastest one and Lu the slowest one. Hf stays in the material under these conditions. When the rhenium/graphite mixture is exposed to 5×10^{-4} torr of CF_4, all these elements leave the target relatively rapidly (fig. 4d). This series of experiments demonstrates that pure rhenium metal powder cannot be used in targets, while a rhenium/graphite mixture is useful when exposed to a CF_4 atmosphere. The delay in the Re/graphite system is mainly due to surface desorption, which is strongly diminished by CF_4 addition. It should be noted that the effect of gas addition is much larger for the Re/graphite matrix than for the pure metal, although the applied CF_4 pressure is 30 times lower.

The Re/graphite system is thus a promising production target for Hf, when used in combination with CF_4. A similar method has been used by the ISOCELE group [15] who produced mass separated beams of HfF_3^+-ions from a LuF_3 target exposed to CF_4. LuF_3 is maybe too volatile for use as an ISOLDE target.

A similar example is shown in fig. 5, i.e. the properties of a mixture of equal volumes of ruthenium powder and graphite powder. A number of volatile elements is rapidly released also without gas addition, while Y and Zr stay in the target. If a partial pressure of 5×10^{-4} torr CF_4 is applied, a very rapid release also of Y is obtained, while no important improvement is seen for Zr. Their different behaviour is probably due to different stability of their carbides.

3.2 Powder target versus foil targets - an important distinction

As discussed by Carraz et. al. [10], two processes govern the release of nuclear reaction products from a target matrix: the in-grain diffusion and the surface desorption. In an extensive study of these phenomena, Beyer and Novgorodov [16] obtained results of great importance for the target development work. They found that for the desorption from the surface of a Ta foil, the most volatile elements leave the surface at the highest rate. For example:

Rb > Sr > Y >> Zr, and Cs > Ba > lanthanides >> Hf.

For the diffusion inside the foil, however, the mobility of the same elements is almost the opposite, i.e.:

Y > Zr > Sr > Rb, and lanthanides > Ba > Hf > Cs.

Since the total surface in a powder target is much larger than in a foil target (roughly a factor of 100), one may expect their release properties to be different. In chapter 4, we will illustrate this phenomenon with on-line tests.

In fig. 6, the release of reaction products from Ti foil is shown. It seems to be very promising as target material for production of neutron deficient Ca and Sc. Although Ca is more volatile than Sc, the latter element is released more rapidly, illustrating that the mobility inside a Ti foil is higher for Sc than for Ca. This observation is in accordance with results of Beyer and Novgorodov [17].

The pure metals constitute the most "clean" type of target material used at ISOLDE, and are therefore preferable in combination with sensitive ion-sources, i.e. hot plasma ion-sources and negative surface ionizers. The former is sensitive to the gas pressure caused by outgassing of the target, the latter to surface contamination by impurities from the target.

It has beem particularly difficult to construct a purely metallic fission target, in which uranium or thorium would have to be used in relatively large amounts. The pure metal powders are not useful because of sintering or corrosive behaviour. This problem has been solved by using a mixture of thorium and niobium powders in the ratio Nb:Th = 10:1. Thorium foil is an even better solution, but is presently not available at a reasonable cost.

3.3 Carbides

All refractory metals, exept the platinum-like elements, form stable, refractory carbides, and some of these compounds are among the most used targets at ISOLDE. Their release properties were investigated by Carraz et al. [10]. and the details are not repeated here. One particularly interesting feature is the influence of CF_4-addition to uranium carbide, which is shown in fig. 7. As can be seen, the release of lanthanide elements is considerably improved through the gas addition, while the elements forming more stable carbides are not much influenced.

Uranium carbide is a very frequently used target at ISOLDE, especially in combination with positive surface ionizers. With negative surface ionizers and hot plasma ion-sources, this material is less favourable due to its release of CO.

Not all carbides are suitable as target materials. For example, the carbides of the group IV elements (Ti, Zr, Hf) liberate the reaction products far too slowly [12].

3.4 Oxides

Some refractory oxides are also useful as target materials. ZrO_2, HfO_2, ThO_2 [18] and Ta_2O_5 react only moderately with Ta, and can be used as target materials without severe material problems. Cerium oxide, CeO_2, was tested on-line by Hansen et. al. [18] but reacts rapidly with Ta. The release of nuclear reaction products from fresh samples is in some cases relatively rapid [12], but on-line experiments with ThO_2 have shown [19] that the liberation rate decreses after some hours application due to sintering.

It can be seen in fig. 8 that the release from CaO is rapid for some elements, but CaO is reduced on Ta surfaces at high temperature, and the material was therefore considered unsuitable for use in ISOLDE. However, it was found that a Re foil could be used to efficiently protect the CaO from reaction with Ta . Additionally, the degree of sintering at 1600 °C turned out to be extremely low. Even after more than 10 hours of heating, the material was still a fluffy powder.

Using this simple protection, CaO and also the other group II oxides MgO and BaO have been shown to work as target materials [20]. Results from on-line tests are shown in chapter 4. Attempts have also been made to use CaO as a diluent to prevent the sintering of other oxides, which may then become more favourable as targets.

One particularly interesting possibility is the formation of gaseous oxides from oxide targets. Examples of release of carbon in the form of CO from ZrO_2 and Ta_2O_5 are shown in fig. 9.

3.5 Borides

We have observed that CaB_6 has sufficiently good high temperature properties to be applied as target material and that the elements Ar, Cl, K, Na, and F are released from CaB_6 at 1800°C at a rate only slightly lower than from CaO. This was confirmed in an on-line test where a high impurity level in the boride used prevented a proper assessment of this othervise interesting type of material.

3.6 Molten metals

The molten metal targets have been used since the early days at ISOLDE [21], and are still the most efficient production systems for a number of elements. Like pure solid metal targets they are particularily clean and stable during prolonged heating and irradiation periods. One of the most interesting molten metal targets would be uranium or thorium. The containment of molten U and Th is extremely difficult, and only dilute solutions of Th in lanthanum could be used on-line. Recently, off-line tests with an yttrium container for U have shown a useful lifetime. However; the delay of molten metal targets is relatively long compared to the fastest solid state targets.

3.6 Intermetallic compounds

Some low-melting metals, particularly Ge and Sn, form refractory intermetallic compounds with the group IV metals Zr and Hf. Some of these compounds have favourable release properties, and attemps have been made to use them as target materials. Their stability at high temperature have to be further clarified before they can be used on-line.

4 ION SOURCES

The choice of ion source to be combined with the above mentioned targets has so far mainly been dictated by efficiency and secondly by selectivity. A number of other properties like energy spread and emittance have largely been neglected, but are now under study [22]. 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 Sidenius [23] and is the subject of a number of conference proceedings [24,25]. High efficiency sources dedicated to on-line mass separation are much rarer and these have been reviewed by Kirchner [26]. Further references can be found in the EMIS series of conference proceedings [27-29] and recent work of Gill and Piotrowski [30] and Nitschke [31].

Ions of many elements and a number of molecules can be produced in a surface ionization process. An atom of low ionization potential (W) can be ionized by contact with a surface of high work function (Φ) which is hot enough to thermally desorb the ions. The efficiency (ε) of this process is given by the Saha-Langmuir formula:

$$\varepsilon = n_+/(n_+ + n_0) = \left\{1 + C*e^{(W-\Phi)/kT}\right\}^{-1}$$

Where n_+ = flux of ions leaving the surface

n_0 = flux of neutrals leaving the surface

C = statistical constant

W = first ionization potential of the atom

Φ = electron work function of the surface k = Boltzman's konstant

T = absolute temperature

Several systems which operate along these lines have been developed at ISOLDE for ionization of alkali elements (Group I), alkaline earths (GroupII), the elements Al, Ga, In and Tl (Group III), Sc, Y, and the lanthanides (Group IIIb)

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. This process is described by an expression similar to the above mentioned Saha-Langmuir formula. An ion source based on this principle has been developed at ISOLDE [6] which produce negative ions of elements with an electron affinity larger than 2.5eV. The major advantage of this source is the 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. A large variety of such sources are used at on-line mass separators.The plasma is generated by accelerating electrons from a heated cathode into a low pressure gas usually confined in a cylindrical volume with one or two electrodes kept at an anode potential. In order to confine the electrons the assembly is situated in an axial magnetic field. The ionization mechanism is rather universal and has little preference for any particular element. A chemical selectivity therefore has to be achieved outside the ion source. 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 [32]. The 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 exist in several versions depending on the transfer-line temperature to be used. The target material is kept in a cylindrical tantalum oven, 20 cm long and 2 cm in diameter. The released products are transferred by molecular flow in vacuum to the ion source via a tubular line.

4.1 Positive surface ionization source with Ta or Re ionizer

This source consists, as shown in fig.10, 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 alkali 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.

4.2 High temperature surface ionizer with W cavity

If a W tube heated to 2400 °C is used as illustrated in fig.10, the source will ionize elements with $W < 6.5$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 Kirchner [26]). The ionized elements are Tl, 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 source.

4.3 Negative surface ionization source

The principle and construction of the source is shown in fig. 11. The negative ion emitter onto which the product flow impinges consists of a small LaB_6 pellet ($\Phi = 2.7$ 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 efficiences of up to 40% of 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.

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

For ionization of the noble gases (GROUP 0) the system shown in fig.12 is used. A water cooled copper section is inserted in the transfer line between target and ion source. In this way only gases are admitted to the source which is kept at a temperature of 1000 °C. Note that the cathode in the ISOLDE version is directly heated by the current usually employed to heat the line. The efficiencies achieved for He, Ne, Ar, Kr, Xe, and Rn are 0.5%, 2%, 5%, 15%, 30%, and 40% respectively.

4.5 Plasma discharge ion source with line at 400°C

A source very similar to the one discussed in section 4.4 is shown in fig. 13. The only difference is that the line is kept at an intermediate temperature at 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.

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

In the version shown in fig.14 the line directly ends in the disc shaped cathode and the same current is used to heat both. This construction has the advantage that the less volatile products are transferred to the source at high temperature without encountering any cold walls where they could be lost by adsorption. By means of heat screens the temperature can be maintained at 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 CF_4-addition.

5 ON-LINE TESTS

By connecting the target to a suitable ion-source configuration, it is in many cases possible to obtain beams of a single element in on-line mass separators. Due to the complexity of high energy nuclear reactions where a wide range of nuclei is produced, selectivity is a necessary condition for many experiments.

The chemical selectivity is obtained by one or several of the following means:

1) Selective release from the target
2) Condensation of unwanted species in the line
3) Selective ionization
4) Formation and mass separation of molecular sidebands

In this chapter, we will present on-line tests showing the production of mass separated beams with various degrees of element selectivity. Note that the rates are given in atoms/s per µA protons incident on the target.

5.1 Systems with positive surface ionizers

Beams with high element selectivity are particularly easy to obtain for the alkali elements, by means of a tantalum positive surface ionizer. Pure beams can then be obtained for K, Rb, Cs and Fr. The combination of a uranium carbide target and a positive surface ionizer has been one of the most frequently used target systems at ISOLDE. As an example, we show the production of Cs isotopes from this system in fig. 15. Information about yields of other alkali elements may be found in refs. [5, 9].

When a hot tungsten ionizer is applied in conjunction with the UC target, other elements , like Ga, In, Ca, Sr, Ba, Ra and the lanthanides are ionized with efficiencies of the order of 1 - 60%. The high production rate of neutron-rich In nuclei from a UC target combined with a W ionizer has given access to the region of the doubly magic ^{132}Sn [33], which is of special interest to nuclear physics. The yields of In isotopes from this system are shown in fig. 16.

High yields of neutron rich nuclei of light elements (below K) can also be obtained with targets lighter than U and Th. For instance, the Iridium-graphite system developed by the Orsay group [34] is very powerful for producing light alkali elements when combined with the W ionizer. This material has also been tested on-line at

ISOLDE, and in fig.17 the production of Na is shown. By means of this target material, the Orsay group has reached the 5.5 ms ^{34}Na [35] which is considered to be the heaviest bound isotope of Na.

The different performance of foil targets compared to powder targets was briefly discussed previously in this paper. The surface delay of most lanthanide elements is considerable, and it is therefore possible to achieve relatively good selectivity for the most volatile lanthanide, Yb, if a target of Ta powder is connected to the hot tungsten surface ionizer. Due to the large surface, the other lanthanides are to a large extent retained in the target, and a supression of 2-3 orders of magnitude relative to Yb is obtained. The yields from a Ta powder target are shown in fig. 18.

For some purposes, an unselective lanthanide target may also be useful. This can be achieved by replacing the Ta powder with Ta foil. The total surface in such a target will be lower, and the delay of the less volatile lanthanides correspondingly diminished.

The yields of lanthanide elements from a Ta foil-target with a tungsten surface ionizer are shown in fig. 19. From this system, an almost unselective production of all these elements is obtained. A foil target is, as expected, more favourable for the less volatile elements. On the other hand, the advantage of an element-selective system is lost.

Neutron-deficient isotopes of the alkali elements may be obtained in different ways. For instance, very intense beams of light Rb isotopes are produced with high selectivity by combining a target of Nb powder with a Ta surface ionizer (fig. 20). Another example is the production of neutron- deficient K isotopes from a target of scandium carbide (fig. 21).

5.2 Systems with negative surface ionizers

The negative surface ionizers are more sensitive to surface impurities than the positive ones, and contamination of the LaB_6 surfaces may be a severe problem. It is therefore crucial to use the negative ionizers in combination with "clean" target materials. Vosicki et. al. [6] reported high yields of halogens from a system with a uranium carbide target in combination with a LaB_6 surface ionizer, but the system had an unstable performance, probably due to carbon "poisoning" of the LaB_6 surface [36].

The cleanest condition for a negative surface ionizer is therefore obtained in combination with a pure metal target or an oxide target. ThO_2 has been used with some success [19], but as mentioned previously, this material seems to sinter heavily and therefore turns out to have rather unfavourable release properties. The best results have been obtained with purely metallic targets. One example is the production of neutron deficient Br isotopes from a Nb target and a LaB_6 ionizer (fig. 22).

5.3 Systems with cooled transfer-line and plasma ion-source

For the remaining elements, neither positive nor negative surface ionization provide satisfactory ionization yields. It is then necessary to use plasma ion-sources.

By means of a water-cooled line, highly selective beams of noble gas isotopes have been obtained. One example is presented in fig. 23, which shows the production of Rn isotopes from a ThO_2 target.

As mentioned previously, group II element oxides are particularly favourable as target materials, provided that they are shielded from contact with the Ta target container. With a FEBIAD plasma ion-source and a water-cooled transfer-line, high yields of neutron-deficient Ne isotopes are obtained from MgO. Carbon is also produced from this target, in the form of carbon monoxide, which passes the cold line. A similar process is the production of neutron-deficient Ar isotopes [37], which are obtained from a CaO target (fig. 24).

5.4 Systems with transfer-line at 400°C and plasma ion- source

Relatively selective systems are obtained when targets of the molten metals Ge, Sn and Pb are used, they produce the volatile metals Zn, Ga, Cd and Hg respectively. The target and line temperatures are not high enough to let less volatile elements, target material or impurities pass into the ion source. In fig. 25, we show as an example the production of Zn and Ga isotopes from a target of molten Ge.

5.5 Systems with transfer-line and plasma ion-source kept at 1900°C

The line-heated plasma ion-source described in section 4.6 is designed for the production of elements which will not pass through a cold or a intermediate temperature transfer-line. The main difficulty with this type of ion-source is its sensitivity to contaminations from the target material. Even small quantities of volatile impurities will give substantial stable beams, and reduce the ionization efficiency. The purity requirements for the target materials are therefore extreme.

The most successful application of the hot plasma ion-source has been in combination with a Th/Nb alloy target kept at 1600 °C. The production of Po and At from this target system is shown in fig. 26. The line-heated plasma source will probably be tested in combination with other target materials in the near future.

6 SUMMARY AND OUTLOOK

To give an impression of the state of the art with respect to the ISOLDE targets we show in table 1 a summary of production systems for the different elements. Some of the systems have already been in use for a long time, others only exist as ideas.

The main problem for the target development in the near future, will be to invent methods which provide the necessary element selectivity. Only a limited number of elements are selectively produced in the present systems. For most elements, chemically selective production systems do not exist.

Four fundamentally different ways seem to be possible:

1) Laser ionization. It has been shown [38-41] that two-step laser ionization can provide elementally pure beams. This method is still on the development stage and

there are no immediate plans for using it on the production side of an on-line mass separator.

2) Selective filters. A certain selectivity may probably be achieved using selectively absorbing filters in the transfer line. For instance, quartz should be an efficient catcher for the alkali metals. More systematic research should be performed in this direction.

3) Separation of molecular ions. The formation of molecular ions in an on-line mass separator has already been used [42, 43], but the possibilities are far from being exhausted. In this way, the beams are shifted into another region of the mass spectrum where a disturbing isobaric activity may vanish. For instance, iodine is separated as the molecular ion AlI^+ in the presence of aluminium vapour. Thereby, it is also chemically separated from disturbing isobars of Xe and Cs, which do not form a similar ion.

4) Isobaric mass separation far from stability may be reached by means of the second ISOLDE separator that will be taken into use in 1986 for which it is planned to reach a resolution of $M/\Delta M = 20000$.

Higher efficiencies and longer lifetime may be achieved by means of Electron Cyclotron Resonance (ECR) ion sources optimized for singly charged ions. Due to their fundamental simplicity, plasma ion-sources excited by microwaves now often replace traditional hot-cathode ion-sources for example at accelerators.

The main advantage at ISOLDE for this type of source is that higher yields of singly charged ions can be obtained. Bechtold and Sheikh [44] have shown that this is especially true for low Z gaseous elements. The simple construction of these sources results in very uncritical, stable, economic and long-lived operation. Furthermore this radiofrequency plasma-source seems to have some unused potential for ionization also of less volatile and heavier elements. An off-line test of this source principle is presently being done [45].

We have demonstrated the extensive possibilities for production of intense beams of unstable nuclei at ISOLDE, and have shown the importance of the development work of targets and ion sources for continued progress in research on nuclei far from stability.

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FIGURE CAPTIONS

Fig. 1. Perspective view of the SC-ISOLDE facility. 1) the 600 MeV synchrocyclotron. 2) Proton beam-lines.3) The ISOLDE-2 target area. 4) The ISOLDE-2 analyzing magnets. 5) The ISOLDE-2 experimental areas. 6) The ISOLDE-3 target area. 7) The ISOLDE-3 analyzing magnets. 8) The ISOLDE-3 experimental areas.

Fig. 2. Elements of the periodic system from which beams of unstable nuclei are produced at ISOLDE.

Fig. 3. Schematic presentation of the mass-transfer steps between the target and the ion beam.

Fig. 4. Release of nuclear reaction products from rhenium powder or a rhenium/graphite mixture.

Fig. 5. Release of nuclear reaction products from a ruthenium/graphite mixture. There is no observable release of Y, Zr, and Tc without CF_4 addition.

Fig. 6. Release of nuclear reaction products from Ti foil.

Fig. 7. Release of nuclear reaction products from a UC 1:10 mol matrix, which shows the effect of CF_4 addition.

Fig. 8. Release of Ar from CaO.

Fig. 9. Release of CO from ZrO_2 and Ta_2O_5. The measurement is performed by monitoring the CO vapour pressure over the oxide, admixed with a small amount of graphite powder.

Fig. 10. The target and ion source assembly. The constriction at the end of the transfer line is the positive surface ionizer.

Fig. 11. The Principle of the negative surface ionization source. The molecular flow of product atoms from the target are directed onto the LaB_6 surface. The emitted ions and electrons are extracted by means of the electrode U2. A magnetic field H separates the electrons out of the beam and directs them to the electrode U1.

Fig. 12. Plasma discharge ion-source conncted to the target via a watercooled transfer-line kept at 30°C.

Fig. 13. Plasma discharge ion-source conncted to the target via a transfer-line kept at 400°C.

Fig. 14. High temperature plasma discharge ion-source. The line, the cathode and the discharge chamber are all heated to 1900°C by means of the same current.

Fig. 15. Production yields of Cs isotopes from 600 MeV proton-induced and 910 MeV ^{3}He-induced reactions in UC_2-graphite cloth connected to a positive surface ionizer. The line is only drawn to guide the eye.

Fig. 16. Production yields of In isotopes from 600 MeV proton-induced and 910 MeV ^{3}He-induced reactions in UC_2-graphite cloth connected to a positive surface ionizer. The line is drawn to guide the eye.

Fig. 17. Production yields of Na isotopes from an Orsay type iridium/graphite target connected to a positive surface ionizer. The line is only a guide.

Fig. 18. Production yields of lanthanide isotopes from a Ta powder target. The line is only a guide.

Fig. 19. Production yields of lanthanide isotopes from a Ta foil target connected to a positive surface ionizer. Individual measurement points are not indicated. The line is only a guide.

Fig. 20. Production yields of Rb isotopes from 600 MeV proton-induced spallation in a Nb powder target connected to a positive surface ionizer. The line is only a guide.

Fig. 21. Production yields of K isotopes from 600 MeV proton-induced and 910 MeV ^{3}He-induced spallation in a scandium carbide target connected to a positive surface ionizer. The line is only a guide.

Fig. 22. Production yields of Br isotopes from 600 MeV proton-induced spallation in a Nb powder target combined with a LaB_6 negative surface ionizer. The line is only a guide.

Fig. 23. Production yields of Rn isotopes from 600 MeV proton-induced

reactions on a thick thorium carbide target connected to a plasma discharge ion-source via a water-cooled transfer-line. The yields from a similar ThO_2 target are shown for comparison. The line is only a guide.

Fig. 24. Production yields of Ar isotopes from 600 MeV proton-induced reactions on a target of CaO connected to a plasma-discharge ion-source via a cold transfer-line. The yields from a similar vanadium carbide target are shown for comparison. The line is only a guide.

Fig. 25. Production yields of isotopes of Zn and Ge from 600 MeV proton-induced reactions on a molten Ge target connected to a plasma-discharge ion-source. The line is only a guide.

Fig. 26. Production yields of isotopes of Po and At from 600 MeV proton-induced reactions on a target of Th:Nb = 1:10 equipped with a hot plasma-discharge ion-source. The line is only a guide.

Table 1. Production systems for different elements.

Element	Target material		Ion source	Special techniques	Category
	n.deficient	n.rich			
He	Ta foil		plasma	cold line	A
Li	Ta foil		pos. surface		A
Be	Ta foil		hot plasma		C
B	Ta_2O_5		hot plasma	separate as BO_x^+	D
C	MgO	Ta_2O_5	plasma	cold line, as CO^+	C,D
N	MgO	Ta_2O_5	plasma	cold line	C,D
O	UC_2		plasma	cold line, as CO^+	D
F	Ta foil		hot plasma	as AlF^+ or BaF^+	D
Ne	MgO	ThC_2	plasma	cold line	A
Na	UC_2, Ir, Ta		pos. surf.		A
Mg	Al	Ta foil	hot plasma		D,C
Al	UC_2		pos. surf.		A
Si	Re		hot plasma	CF_4 - addition	D
P					D
S					D
Cl	Ti foil	Ta foil	neg. surf		D,C
Ar	CaO	ThC_2	plasma	cold line	A
K	Sc_3C_4	UC_2, Ta	pos. surf.		A
Ca	Ti foil	UC_2, Ta	pos. surf.		D,B
Sc	Ti foil	Ta foil	pos. surf	CF_4 - addition (?)	D

Ti	Ir foil		plasma	CF_4 - addition	D
V	Ir foil		plasma	CF_4 - addition	D
Cr					D
Mn	Fe_3C	Ta foil	plasma		D,C
Fe					D
Co					D
Ni					D
Cu	Zr_5Ge_3	Ta foil	hot plasma		D,C
Zn	Ge	UC_2	plasma	cooled line	B,C
Ga	Ge	UC_2	pos. surf.		B,A
Ge					D
As	Ru, ZrO_2	ThO_2	hot plasma		D
Se	Ru, ZrO_2	ThO_2	hot plasma		D
Br	Nb	Th foil	neg. surf.		A,C
Kr	SrO, Nb	ThC_2	plasma	cold line	A
Rb	Nb	UC_2	pos. surf.		A
Sr	Zr	UC_2	pos. surf.		D,B
Y	Zr foil	Th foil	pos. surf	CF_4 addition (?)	D
Zr	Ru		hot plasma	CF_4 addition	D
Nb					D
Mo					D
Tc					D
Ru					D
Rh					D
Pd		UC_2	hot plasma		D

Ag		UC_2	hot plasma		C
Cd	Sn	UC_2	plasma	cooled line	A,C
In	Hf_5Sn_3	UC_2	pos. surf,		D,B
Sn	$TeCl_4$	Th foil	hot plasma		C
Sb	TeO_2/KCl	ThO_2	hot plasma		C,D
Te		ThO_2	hot plasma		D
I	$BaZrO_3$, BaO	Th foil	neg. surf.		A
Xe	BaO, LaC_2	ThC_2	plasma	cold line	A
Cs	La	UC_2	pos. surf.		A
Ba	La	UC_2	pos. surf		D,A
La-Lu	Ta foil	Th foil	pos. surf.		B,D
Hf	Re/gr	Th foil	hot plasma	CF_4 addition	D
Ta	Re/gr		hot plasma	CF_4 addition	D
W					D
Re					D
Os					D
Ir					D
Pt					D
Au		UC_2, Th foil	hot plasma		D
Hg	Pb	UC_2	plasma		A,D
Tl		ThC_2, Th foil	pos. surf.		C
Pb		ThC_2, Th foil	hot plasma		C
Bi		ThC_2, Th foil	hot plasma		C
Po		ThC_2, Th foil	hot plasma		C

At	ThC_2, Th foil	neg. surf.		C
Rn	ThC_2	plasma	cold line	A
Fr	ThC_2	pos. surf.		A
Ra	ThC2	pos. surf.		A
Ac				D
Th				D
Pa				D
U				D

A) Satisfactory yields, good element selectivity

B) Satisfactory yields, insufficient element selectivity

C) Primary on-line beam observed

D) Project

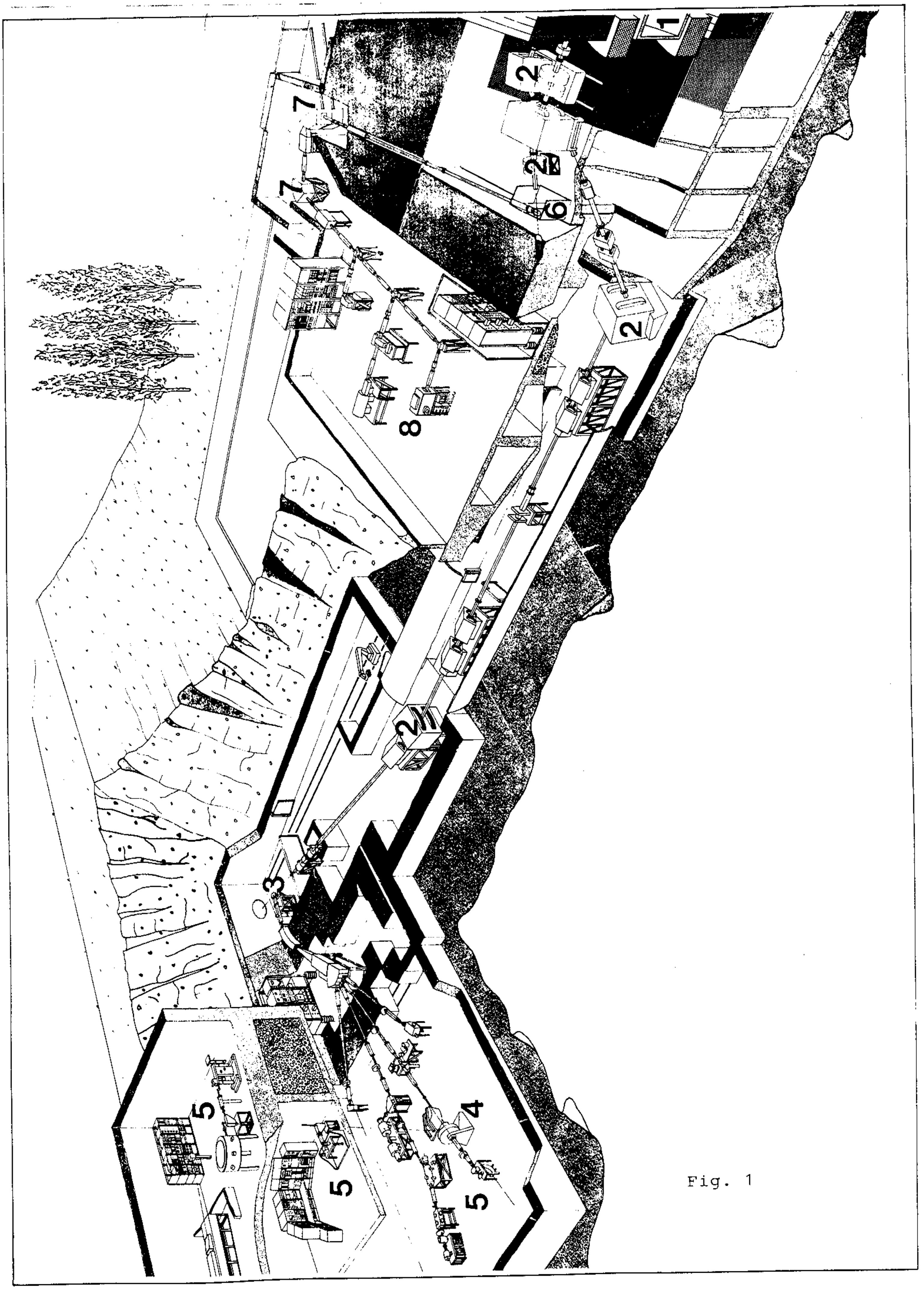


Fig. 1

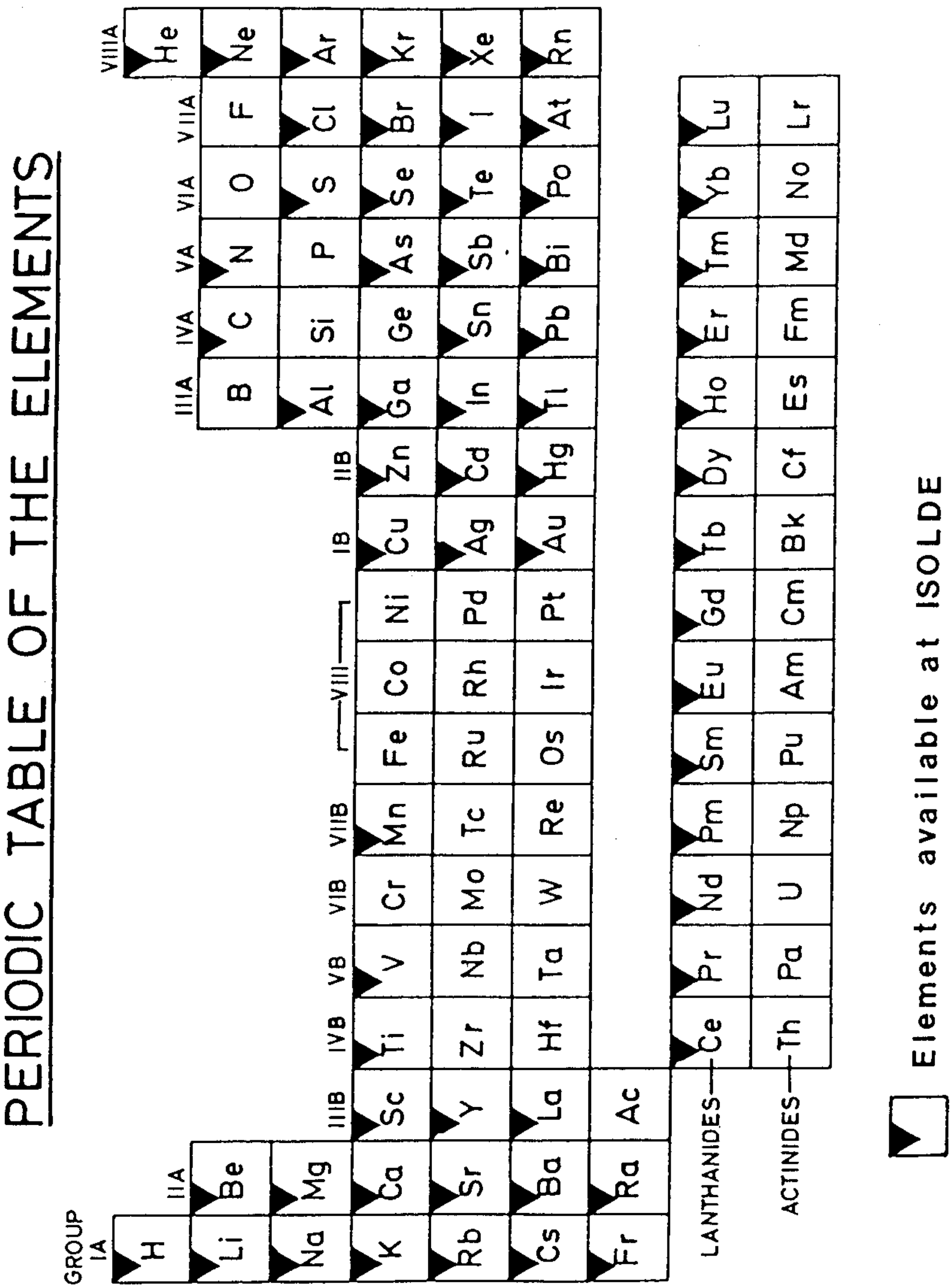
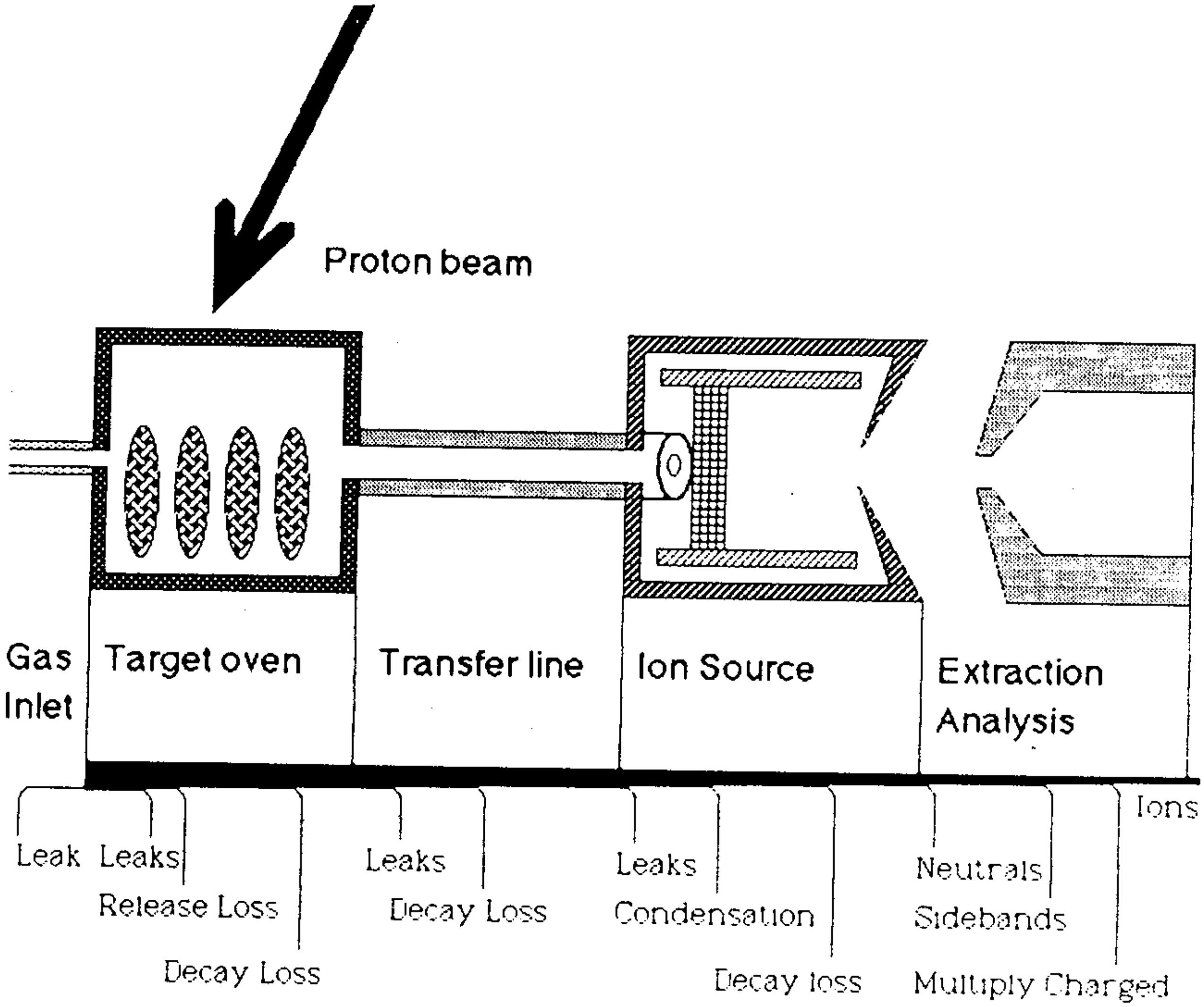

PERIODIC TABLE OF THE ELEMENTS
GROUP IA
IIA
IIIB
IVB
VB
VIB
VIIB
VIII
IB
IIB
IIIA
IVA
VA
VIA
VIIA
VIIIA
H
He
Li
Be
B
C
N
O
F
Ne
Na
Mg
Al
Si
P
S
Cl
Ar
K
Ca
Sc
Ti
V
Cr
Mn
Fe
Co
Ni
Cu
Zn
Ga
Ge
As
Se
Br
Kr
Rb
Sr
Y
Zr
Nb
Mo
Tc
Ru
Rh
Pd
Ag
Cd
In
Sn
Sb
Te
I
Xe
Cs
Ba
La
Hf
Ta
W
Re
Os
Ir
Pt
Au
Hg
Tl
Pb
Bi
Po
At
Rn
Fr
Ra
Ac
LANTHANIDES
Ce
Pr
Nd
Pm
Sm
Eu
Gd
Tb
Dy
Ho
Er
Tm
Yb
Lu
ACTINIDES
Th
Pa
U
Np
Pu
Am
Cm
Bk
Cf
Es
Fm
Md
No
Lr
Elements available at ISOLDE

Fig. 2



Proton beam
Gas Inlet
Target oven
Transfer line
Ion Source
Extraction Analysis
Leak
Leaks
Release Loss
Decay Loss
Leaks
Decay Loss
Leaks
Condensation
Decay loss
Neutrals
Sidebands
Multiply Charged
Ions

Fig. 3

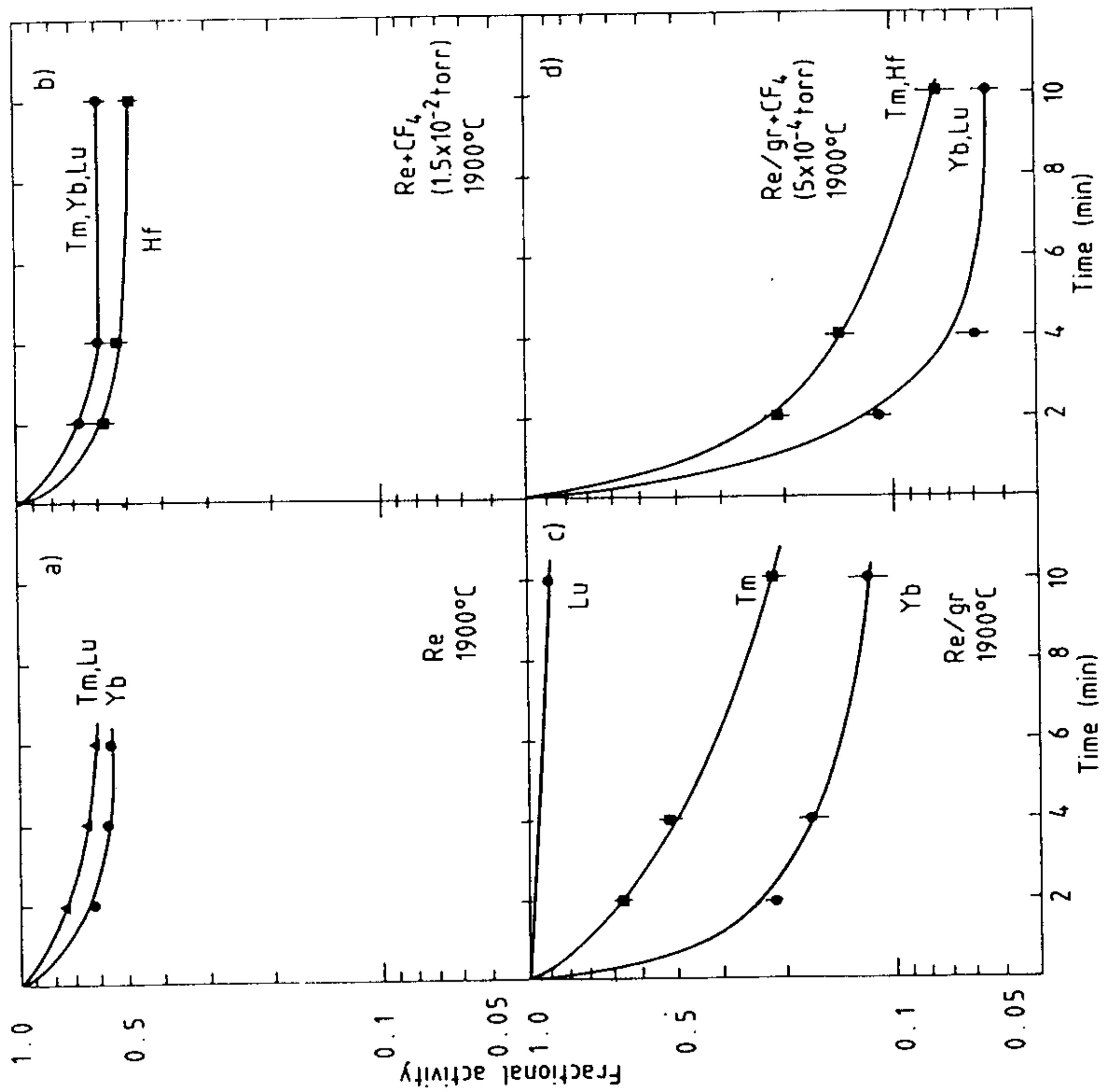

a)
Tm,Lu
Yb
Re
1900°C
b)
Tm,Yb,Lu
Hf
Re+CF4
(1.5x10-2 torr)
1900°C
c)
Lu
Tm
Yb
Re/gr
1900°C
d)
Re/gr+CF4
(5x10-4 torr)
1900°C
Tm,Hf
Yb,Lu
Fractional activity
1.0
0.5
0.1
0.05
1.0
0.5
0.1
0.05
Time (min)
2
4
6
8
10
Time (min)
2
4
6
8
10

Fig. 4

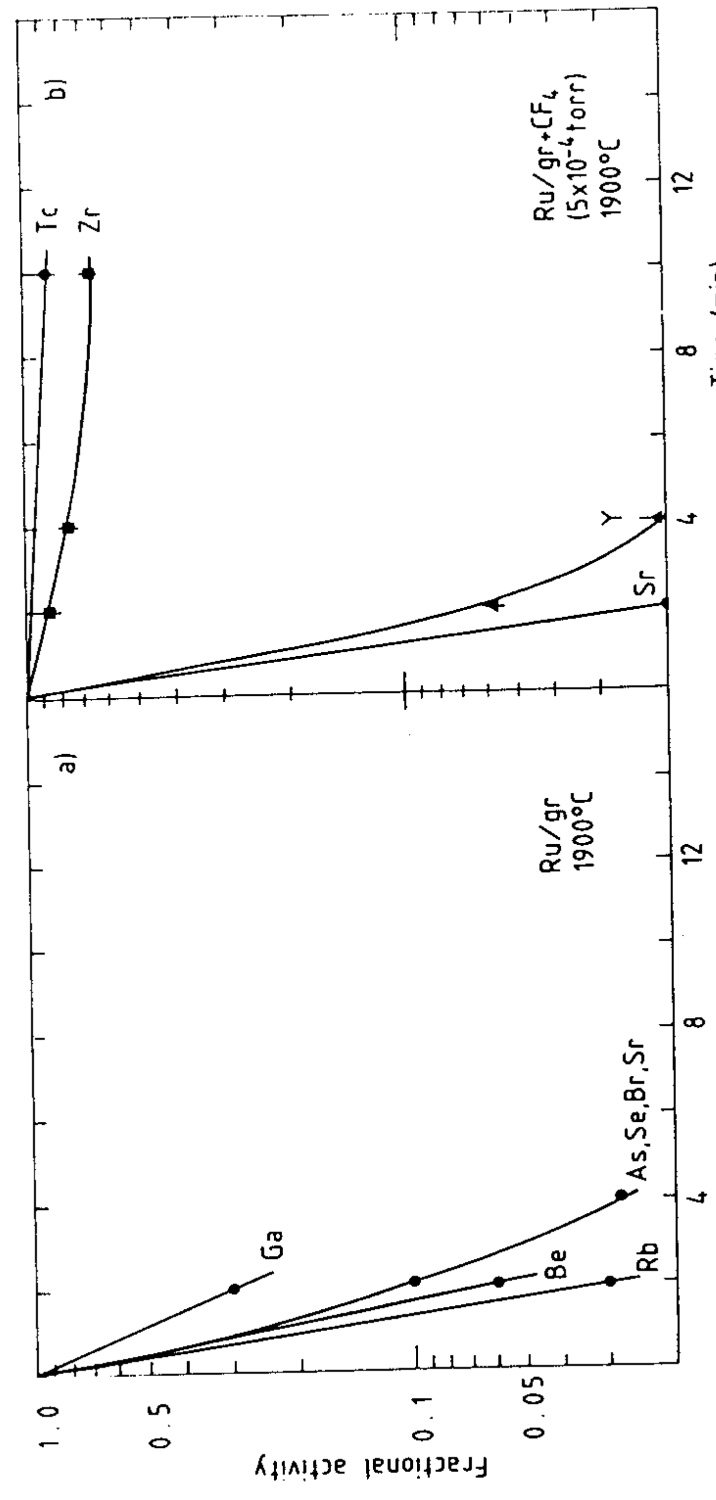

a)
Ru/gr
1900°C
b)
Ru/gr+CF₄
(5x10⁻⁴torr)
1900°C
Ga
Be
Rb
As,Se,Br,Sr
Tc
Zr
Y
Sr
Time (min)
4
8
12
Fractional activity
1.0
0.5
0.1
0.05

Fig. 5

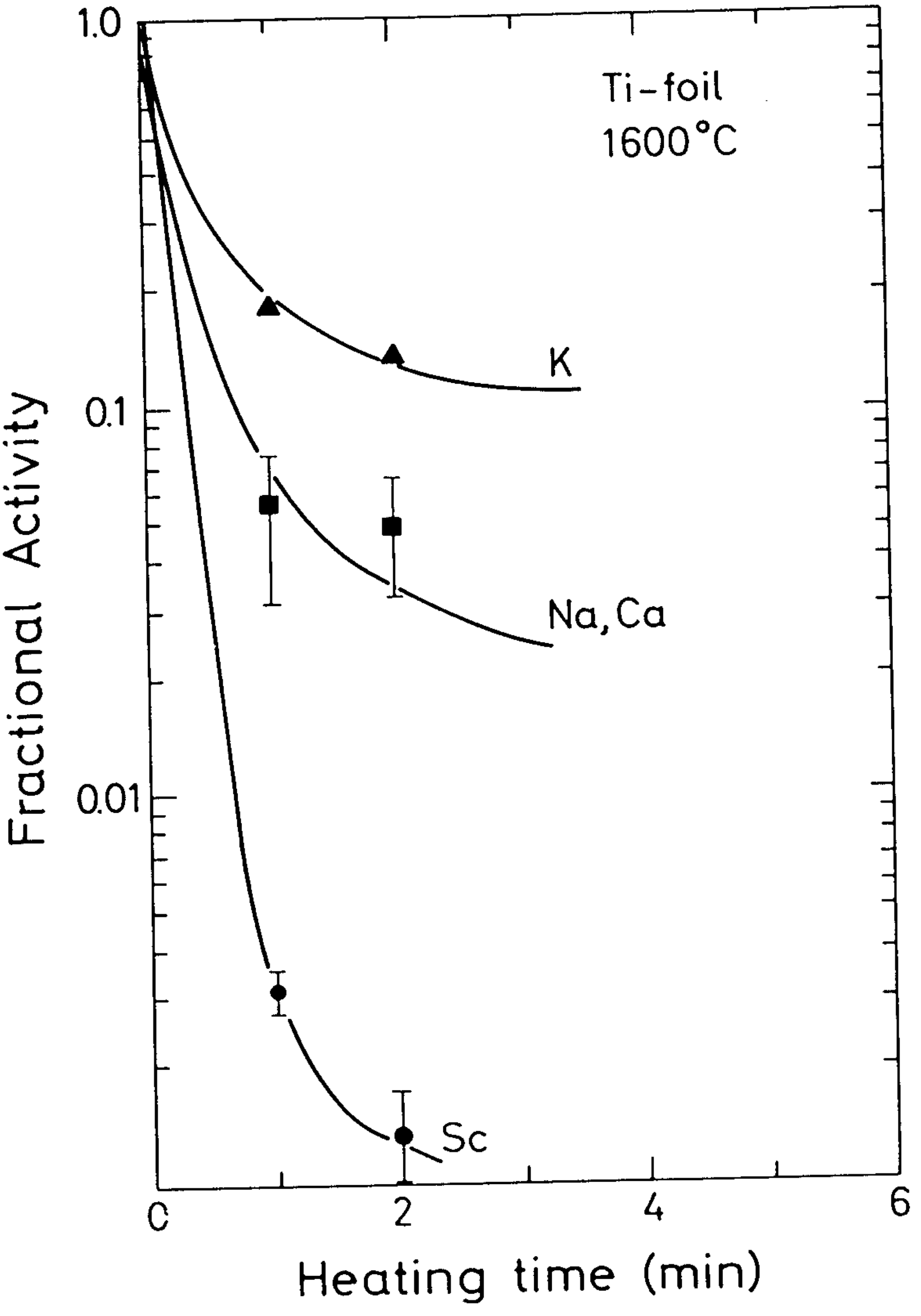
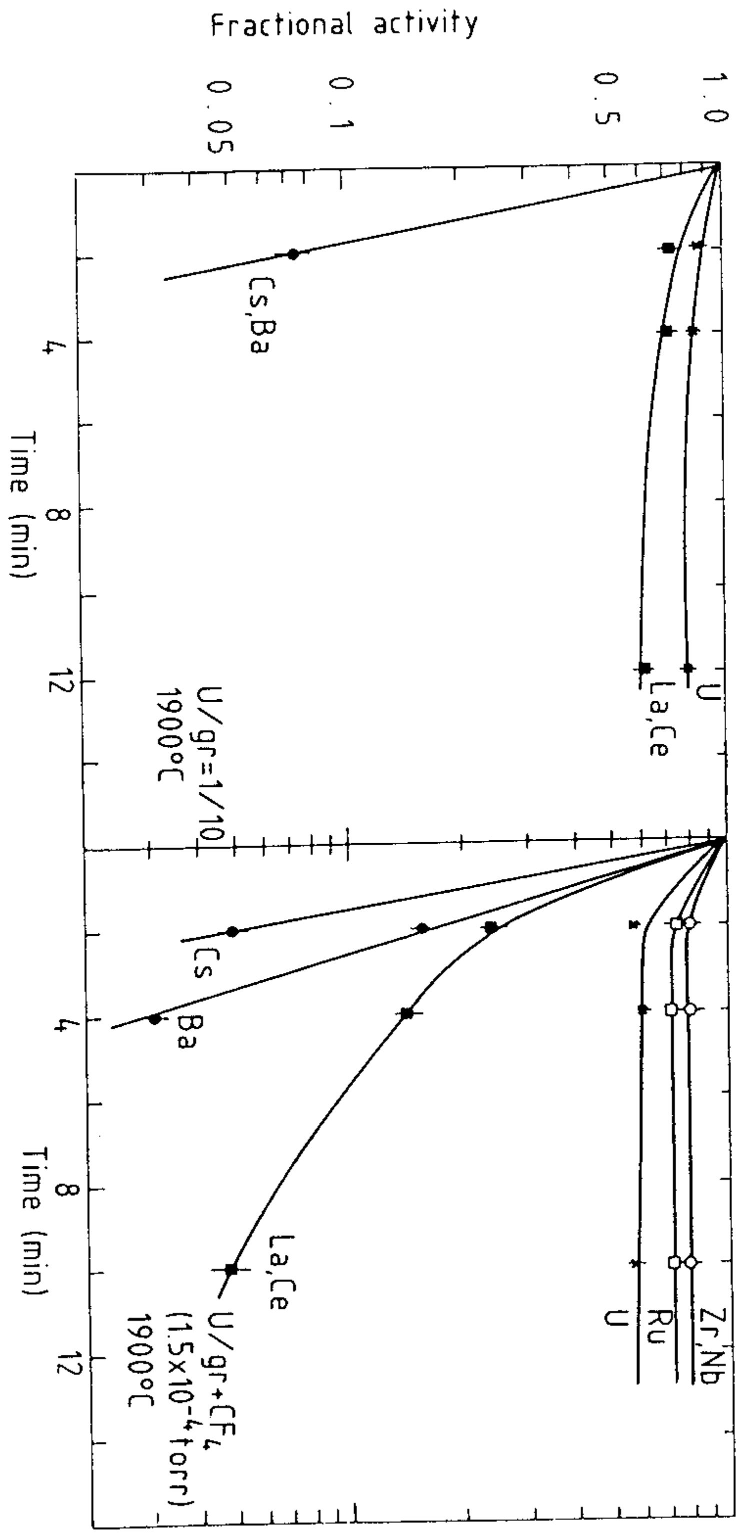

Ti-foil
1600°C
K
Na, Ca
Sc
1.0
0.1
0.01
Fractional Activity
0
2
4
6
Heating time (min)

Fig. 6



Fractional activity
1.0
0.5
0.1
0.05
Time (min)
4
8
12
Cs,Ba
U
La,Ce
U/gr=1/10
1900°C
Zr,Nb
Ru
U
Cs
Ba
La,Ce
U/gr+CF4
(1.5x10-4 torr)
1900°C
Time (min)
4
8
12

Fig. 7

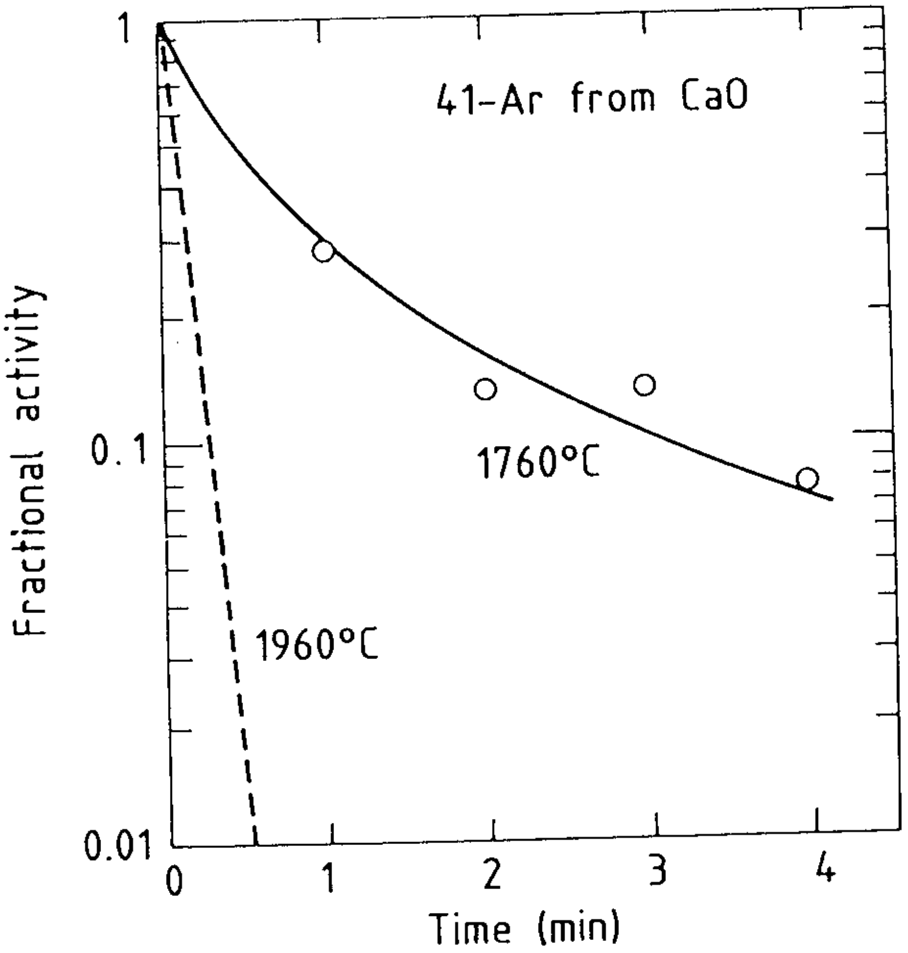

41-Ar from CaO
1760°C
1960°C
Fractional activity
1
0.1
0.01
Time (min)
0
1
2
3
4

Fig. 8

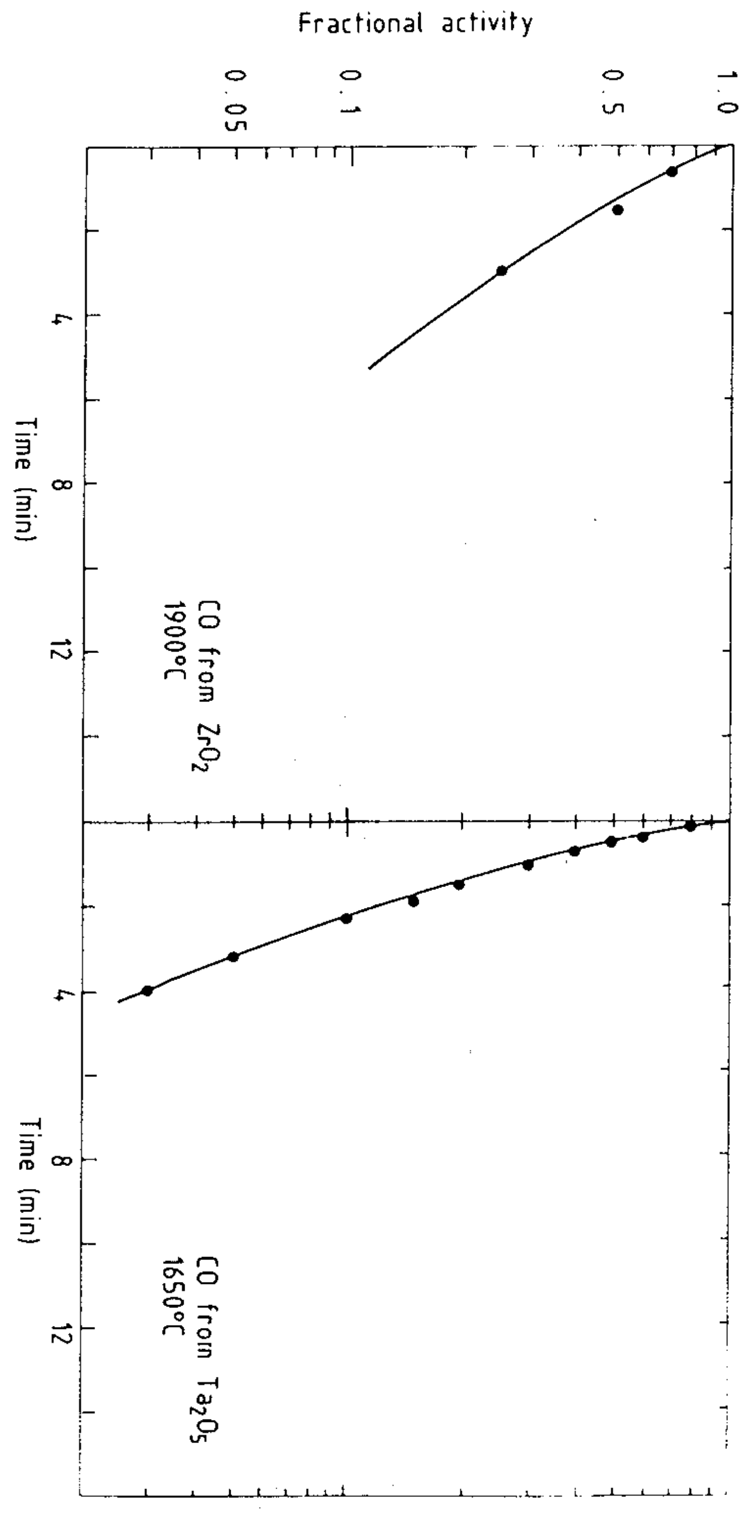

Fractional activity
1.0
0.5
0.1
0.05
Time (min)
4
8
12
CO from ZrO_2
1900°C
Time (min)
4
8
12
CO from Ta_2O_5
1650°C

Fig. 9

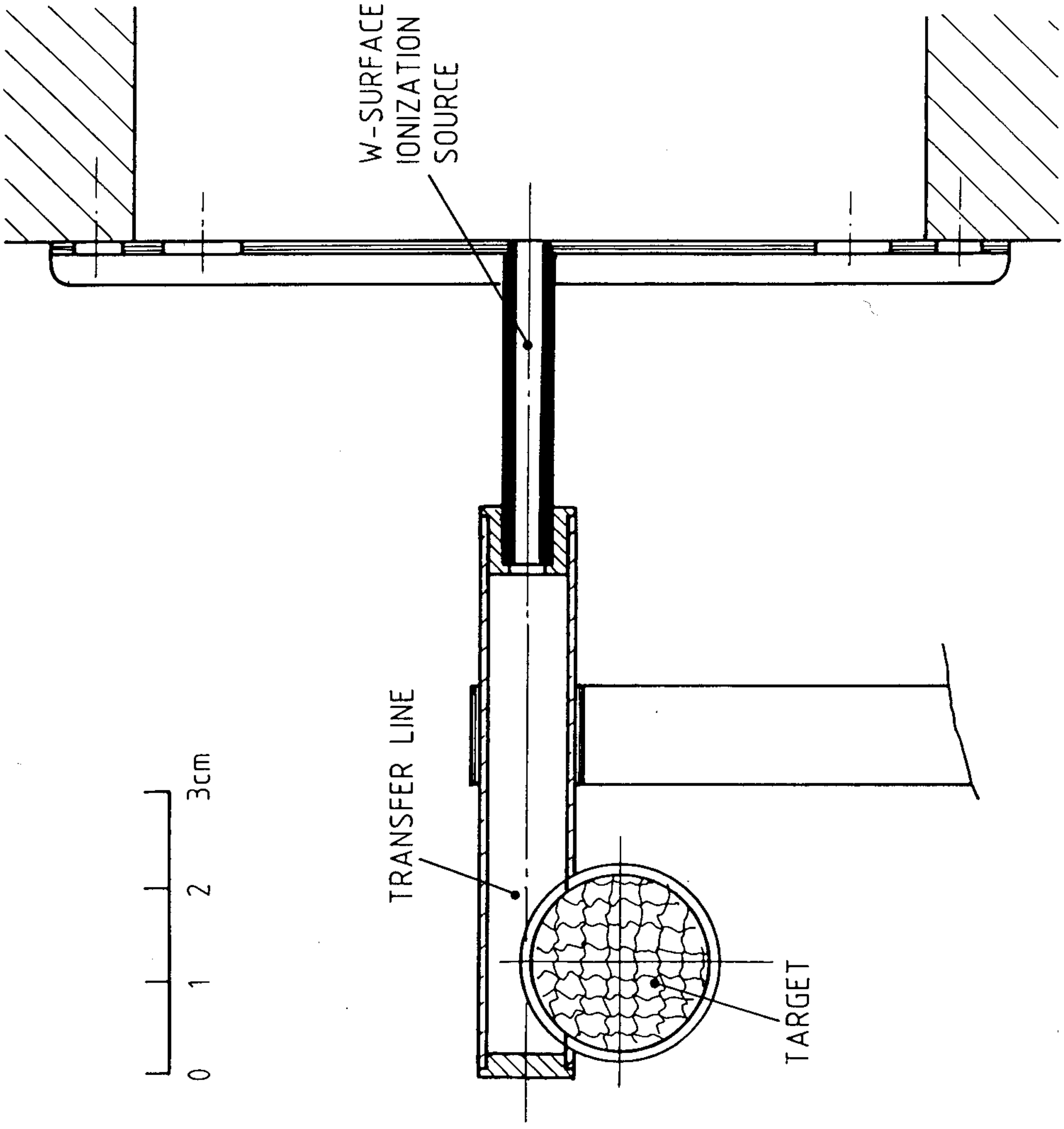


Fig. 10

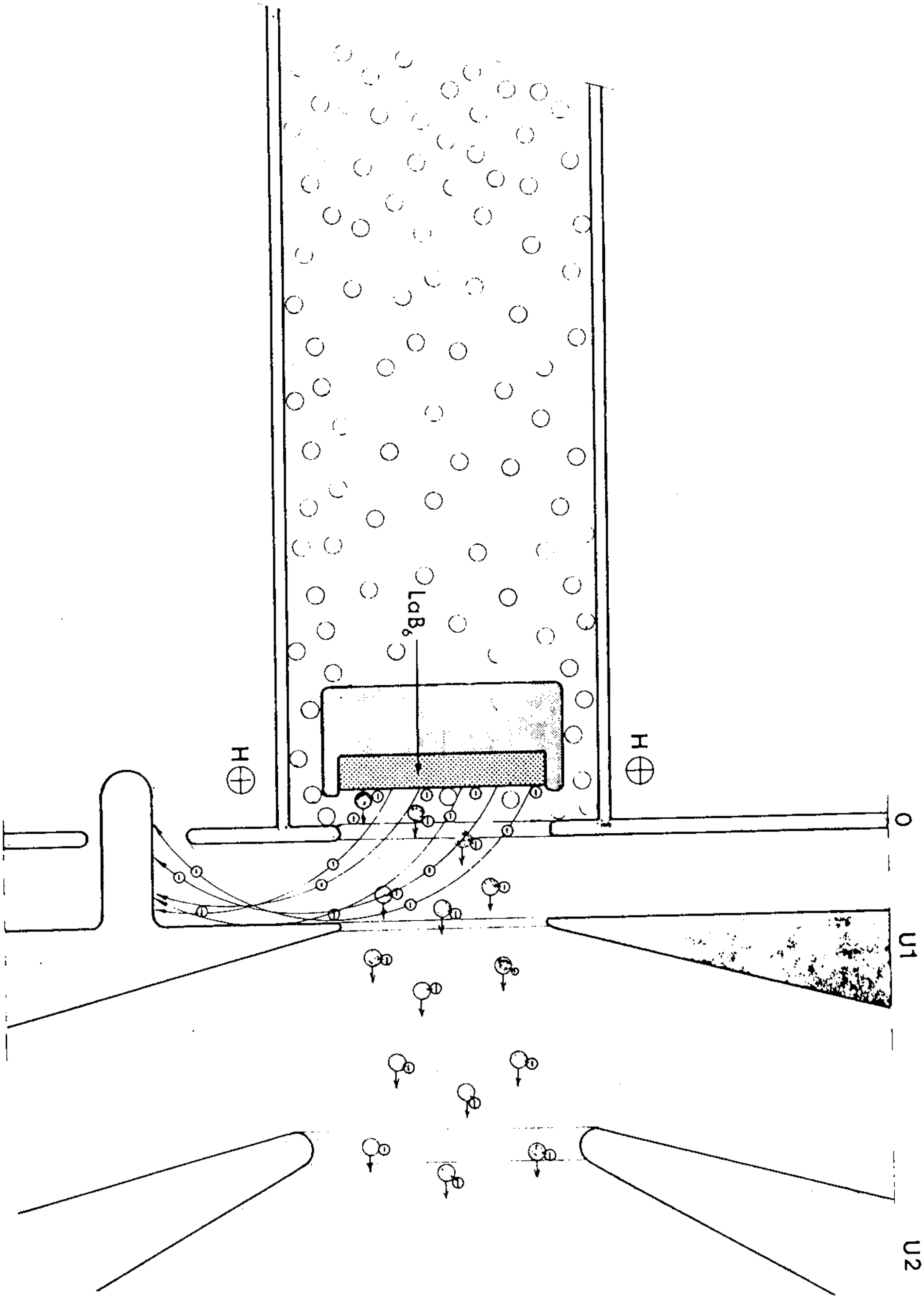


Fig. 11

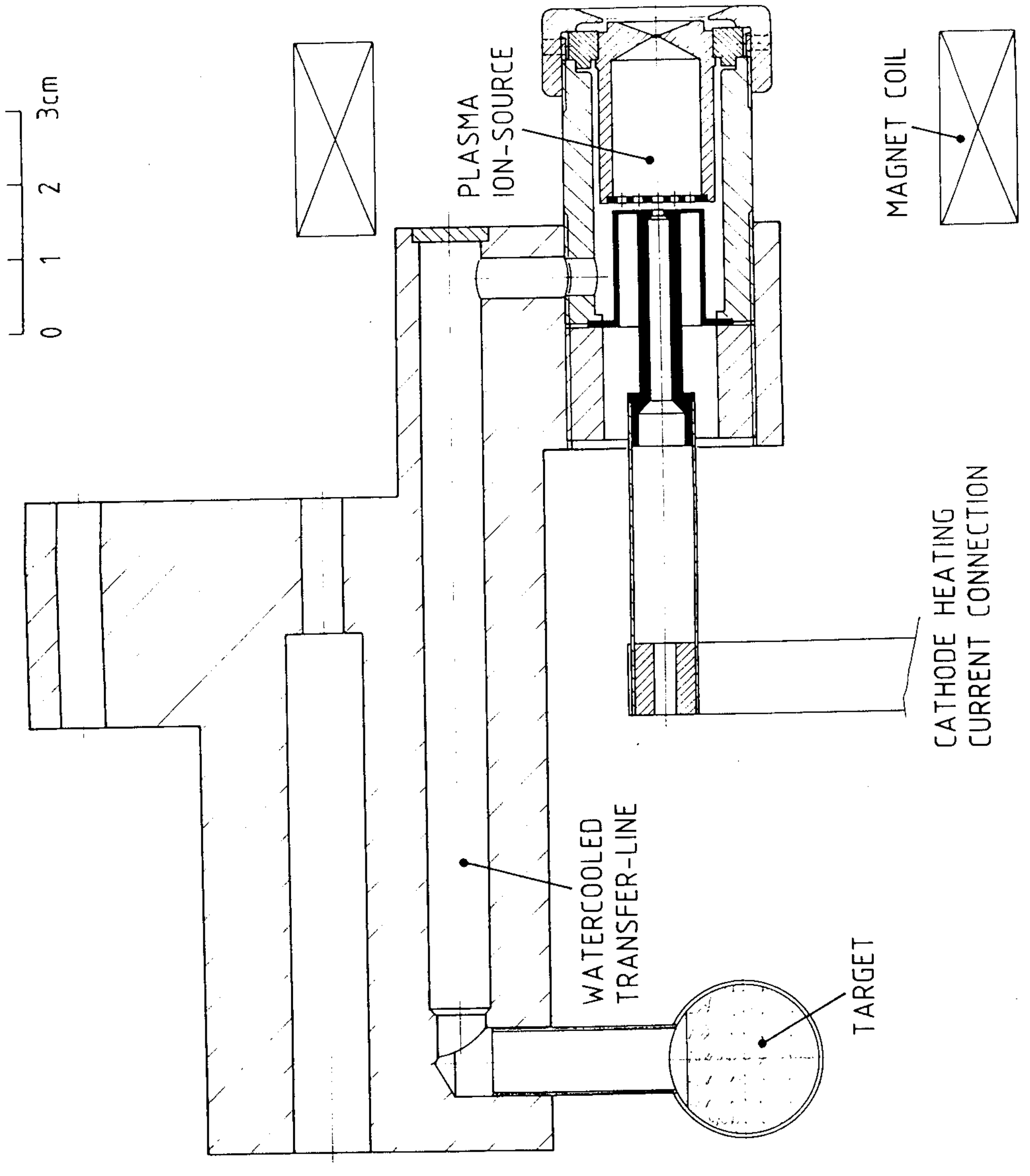

0
1
2
3cm
PLASMA ION-SOURCE
MAGNET COIL
CATHODE HEATING CURRENT CONNECTION
WATERCOOLED TRANSFER-LINE
TARGET

Fig. 12

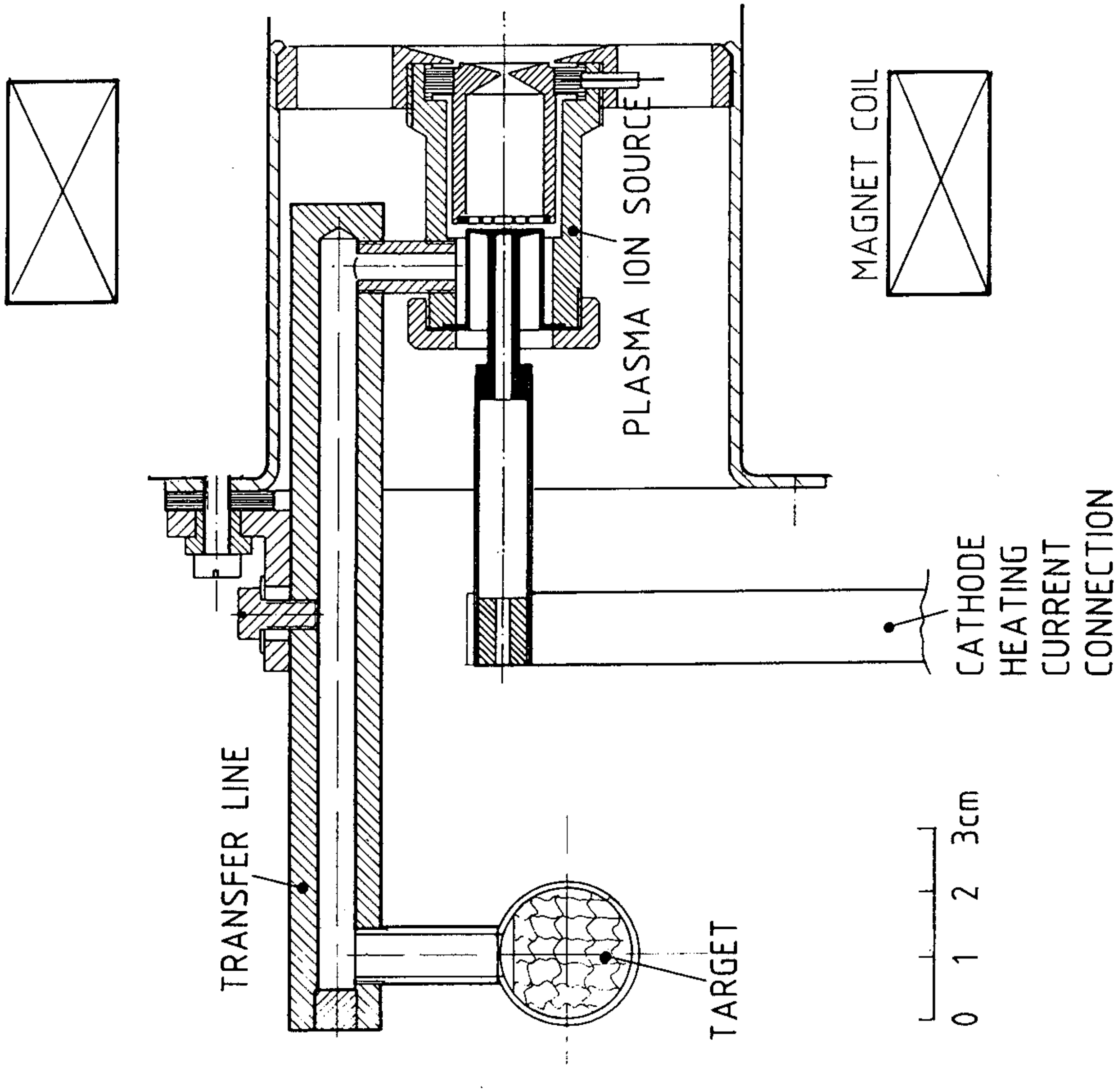

MAGNET COIL
PLASMA ION SOURCE
CATHODE
HEATING
CURRENT
CONNECTION
TRANSFER LINE
TARGET
0
1
2
3cm

Fig. 13

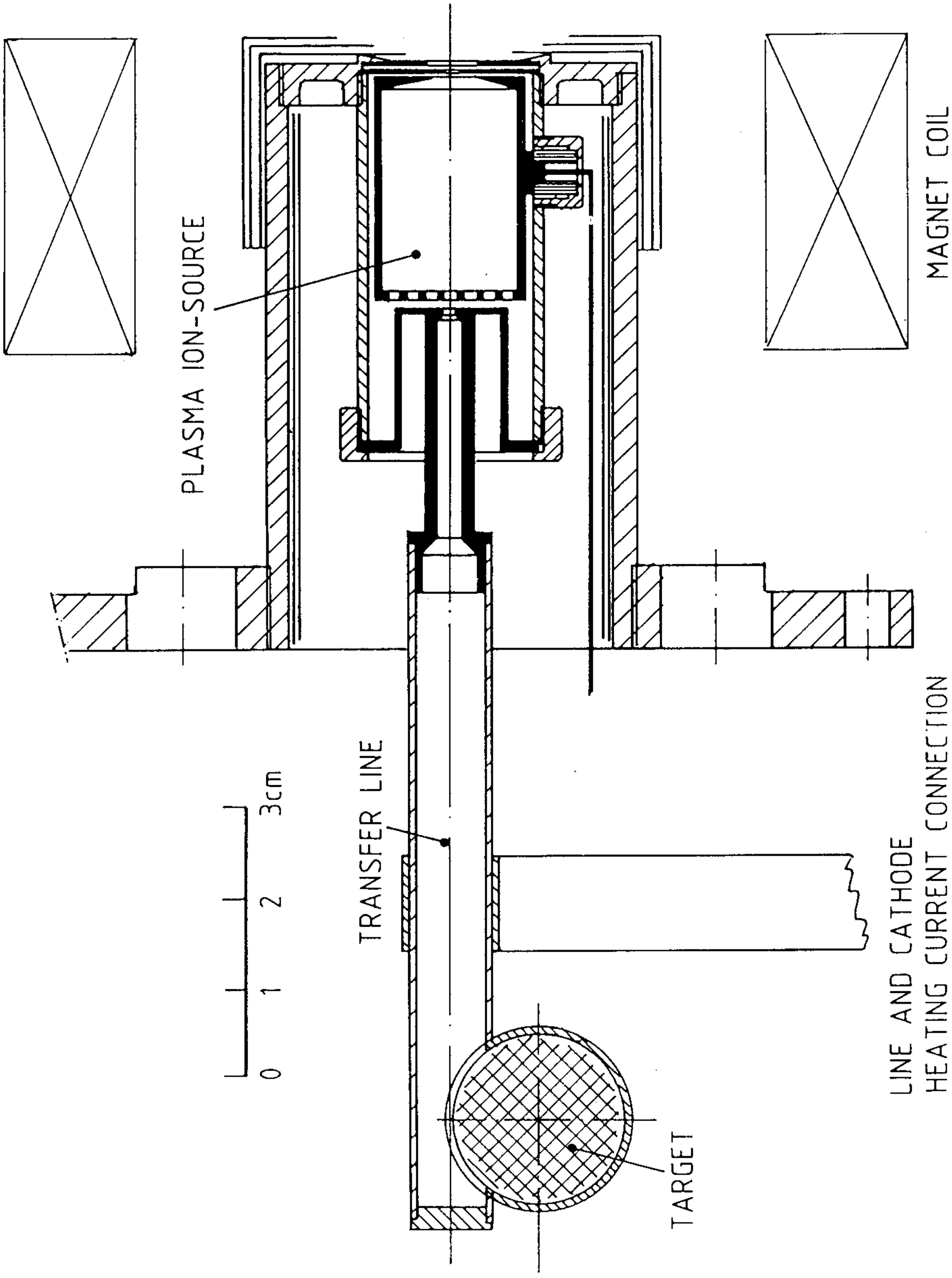

PLASMA ION-SOURCE
MAGNET COIL
TRANSFER LINE
LINE AND CATHODE HEATING CURRENT CONNECTION
TARGET
0
1
2
3cm

Fig. 14

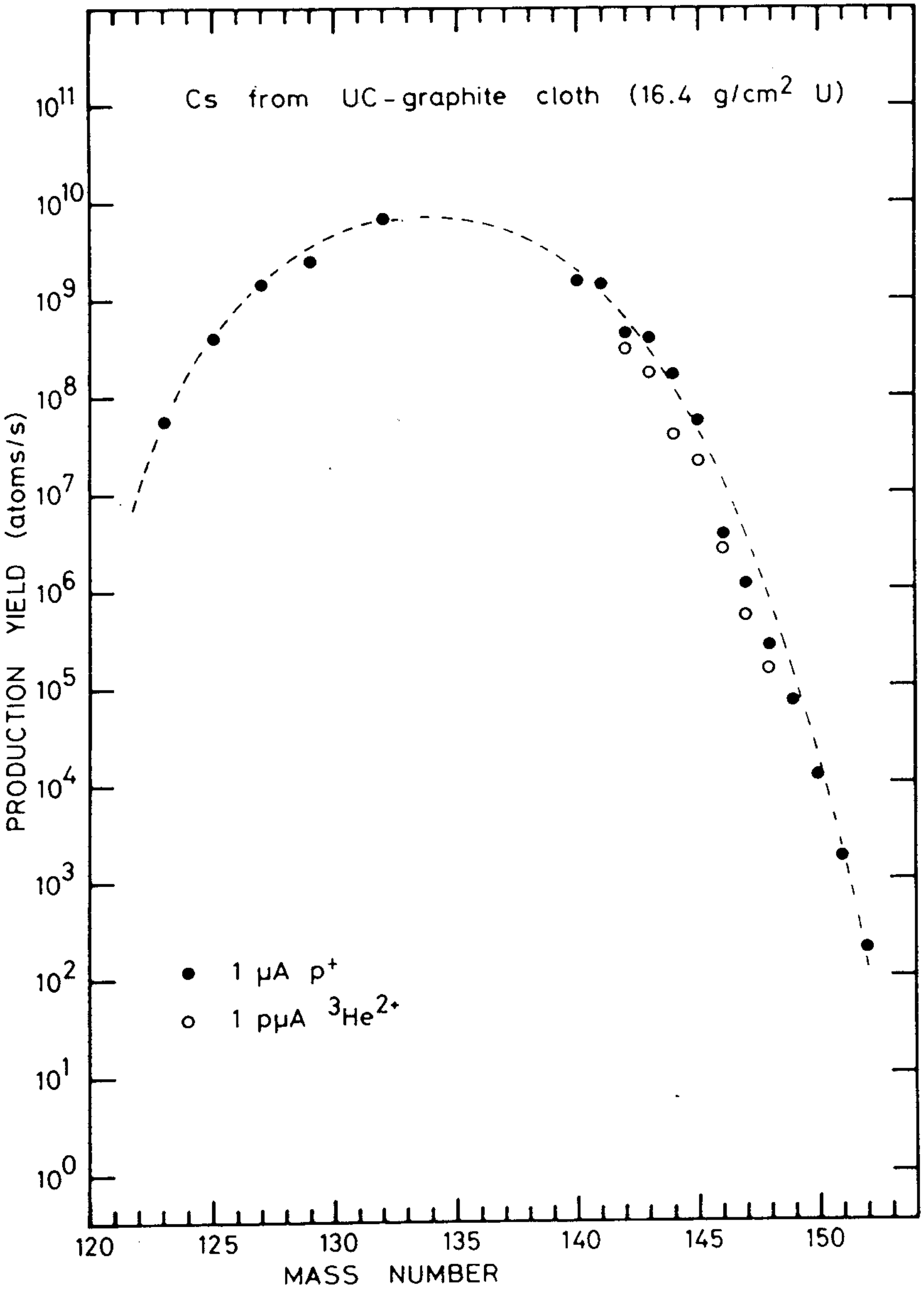
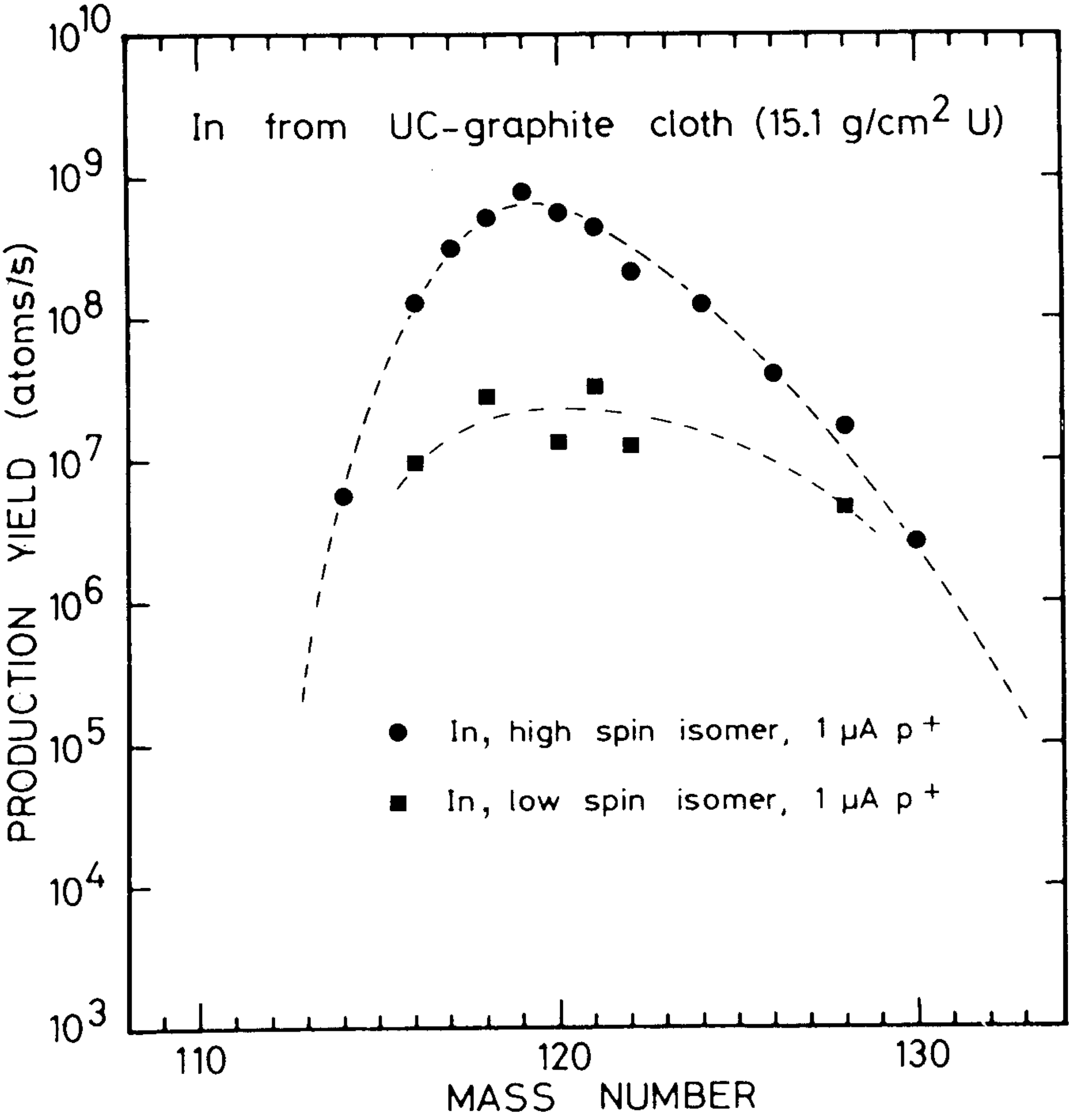

Cs from UC-graphite cloth (16.4 g/cm^2 U)
PRODUCTION YIELD (atoms/s)
10^{11}
10^{10}
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
10^0
● 1 μA p^+
○ 1 pμA $^3He^{2+}$
120
125
130
135
140
145
150
MASS NUMBER

Fig. 15



In from UC-graphite cloth (15.1 g/cm^2 U)
PRODUCTION YIELD (atoms/s)
10^{10}
10^9
10^8
10^7
10^6
10^5
10^4
10^3
● In, high spin isomer, 1 µA p$^+$
■ In, low spin isomer, 1 µA p$^+$
110
120
130
MASS NUMBER

Fig. 16

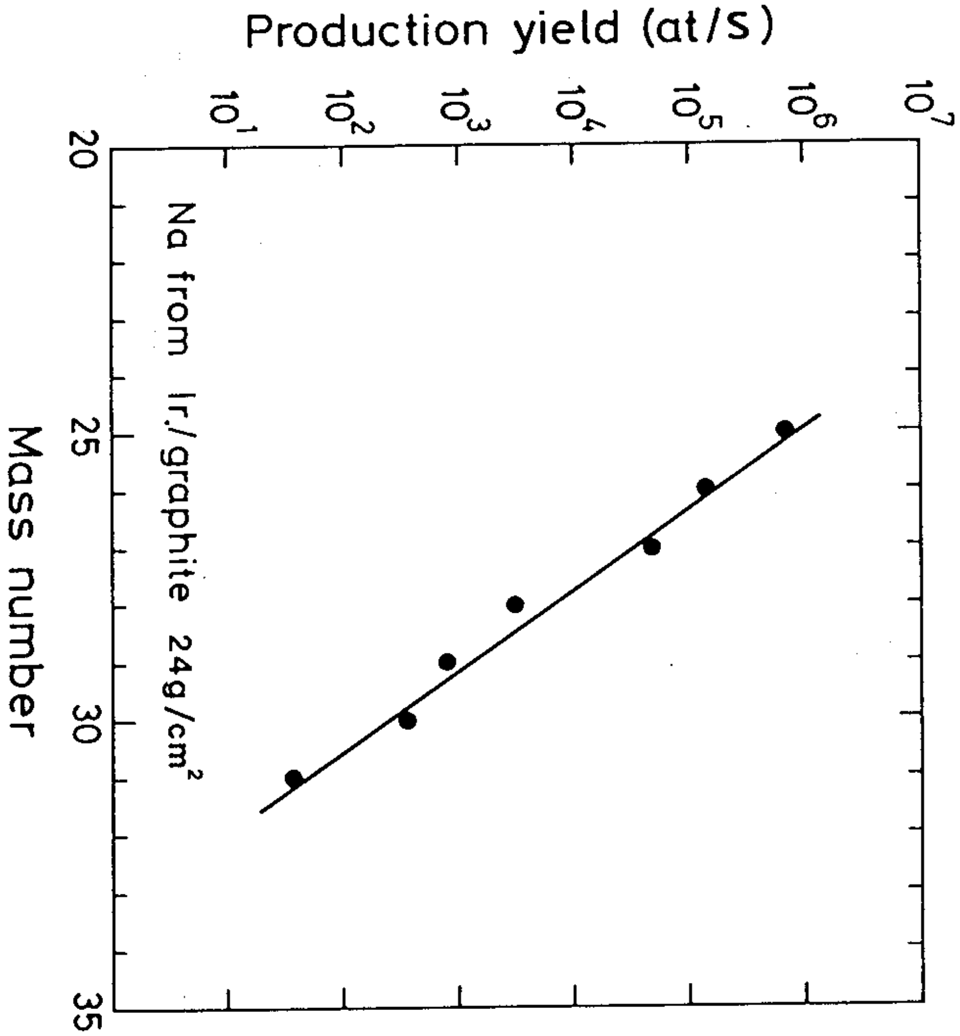

Production yield (at/s)
10^1
10^2
10^3
10^4
10^5
10^6
10^7
20
25
30
35
Mass number
Na from Ir/graphite 24g/cm²

Fig. 17

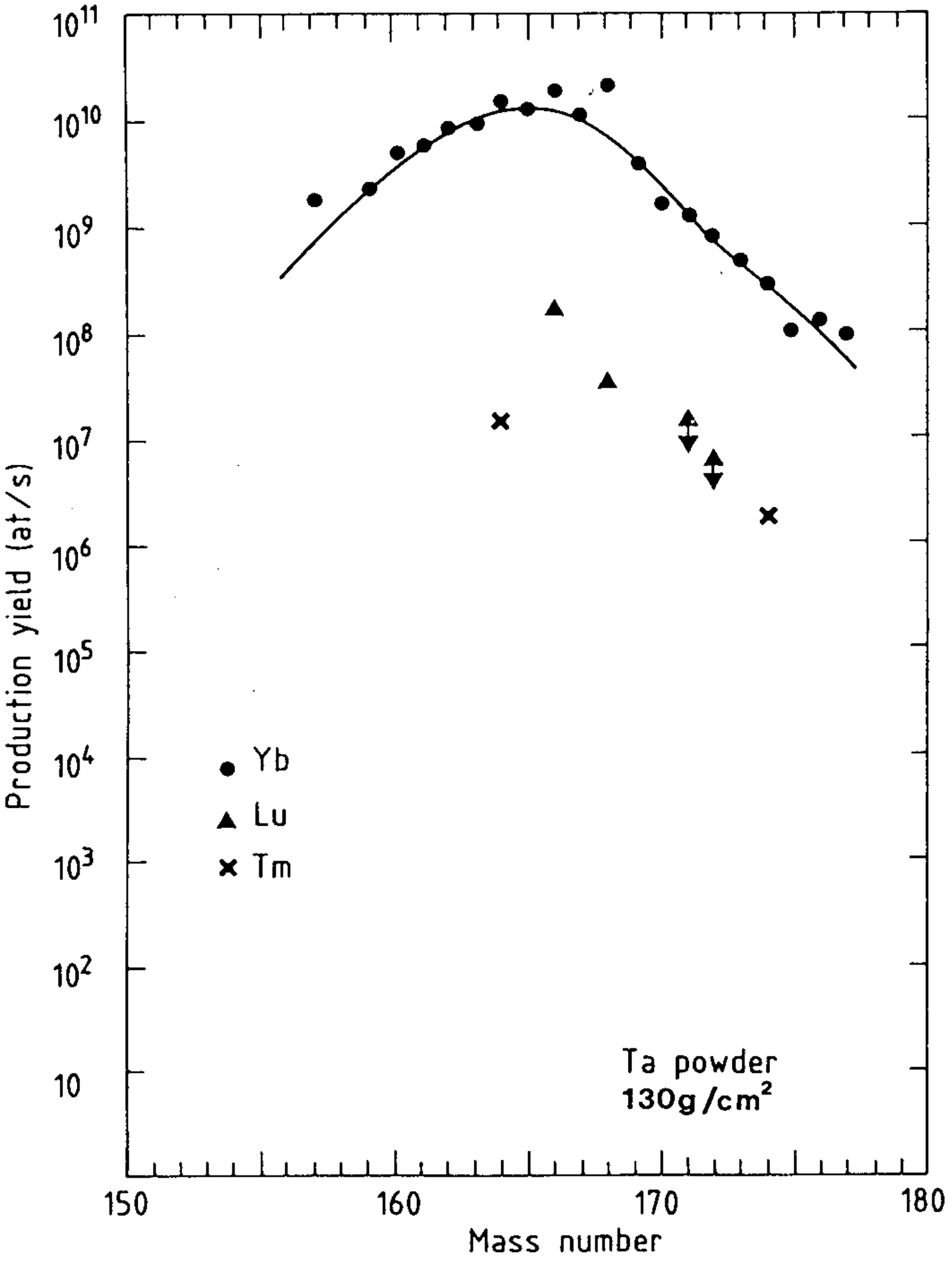

Production yield (at/s)
10
10^2
10^3
10^4
10^5
10^6
10^7
10^8
10^9
10^10
10^11
150
160
170
180
Mass number
● Yb
▲ Lu
× Tm
Ta powder
130g/cm²

Fig. 18

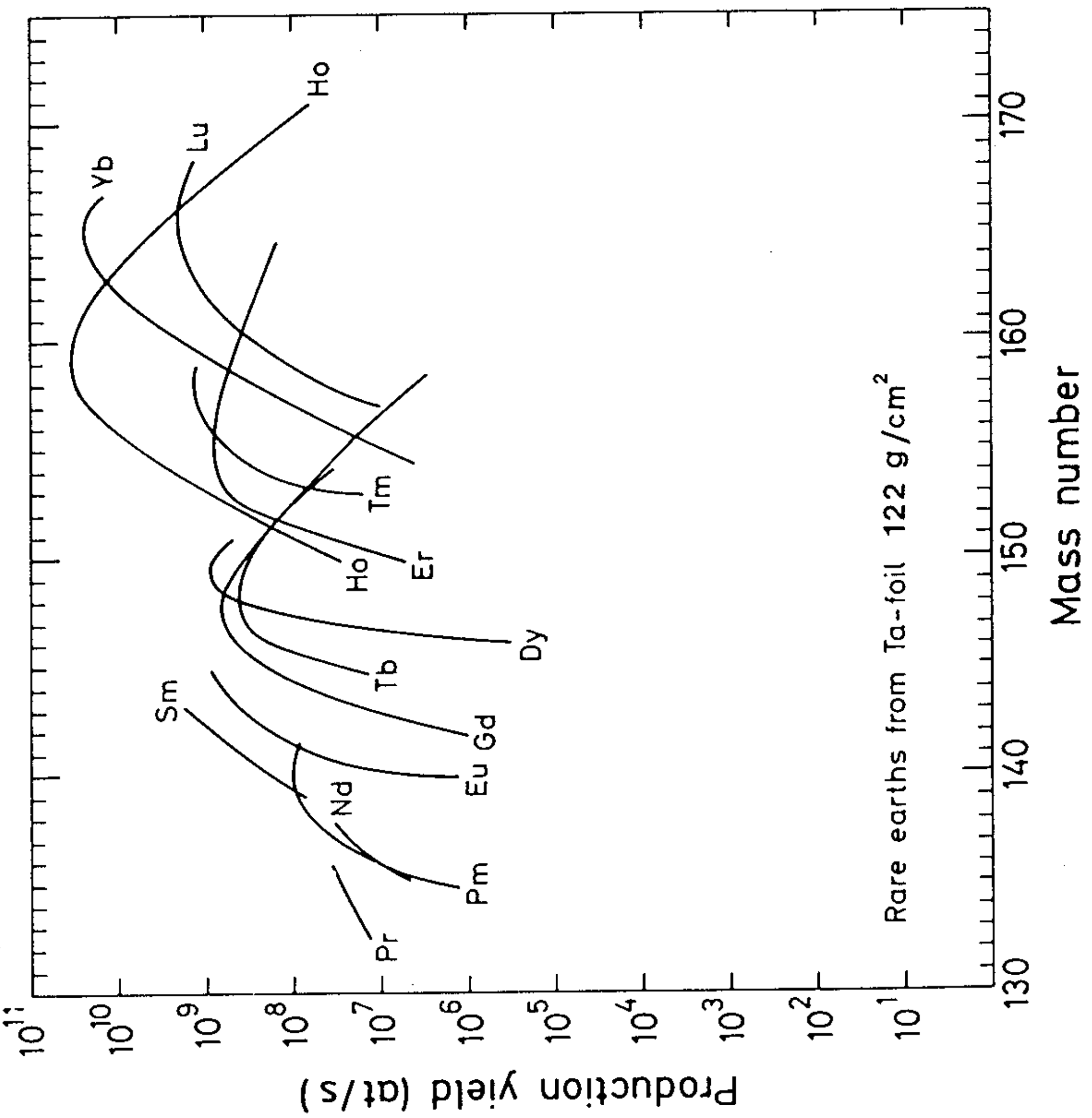

Production yield (at/s)
Mass number
Rare earths from Ta-foil 122 g/cm²
10^11
10^10
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
130
140
150
160
170
Pr
Pm
Nd
Sm
Eu
Gd
Tb
Dy
Ho
Er
Tm
Yb
Lu
Ho

Fig. 19

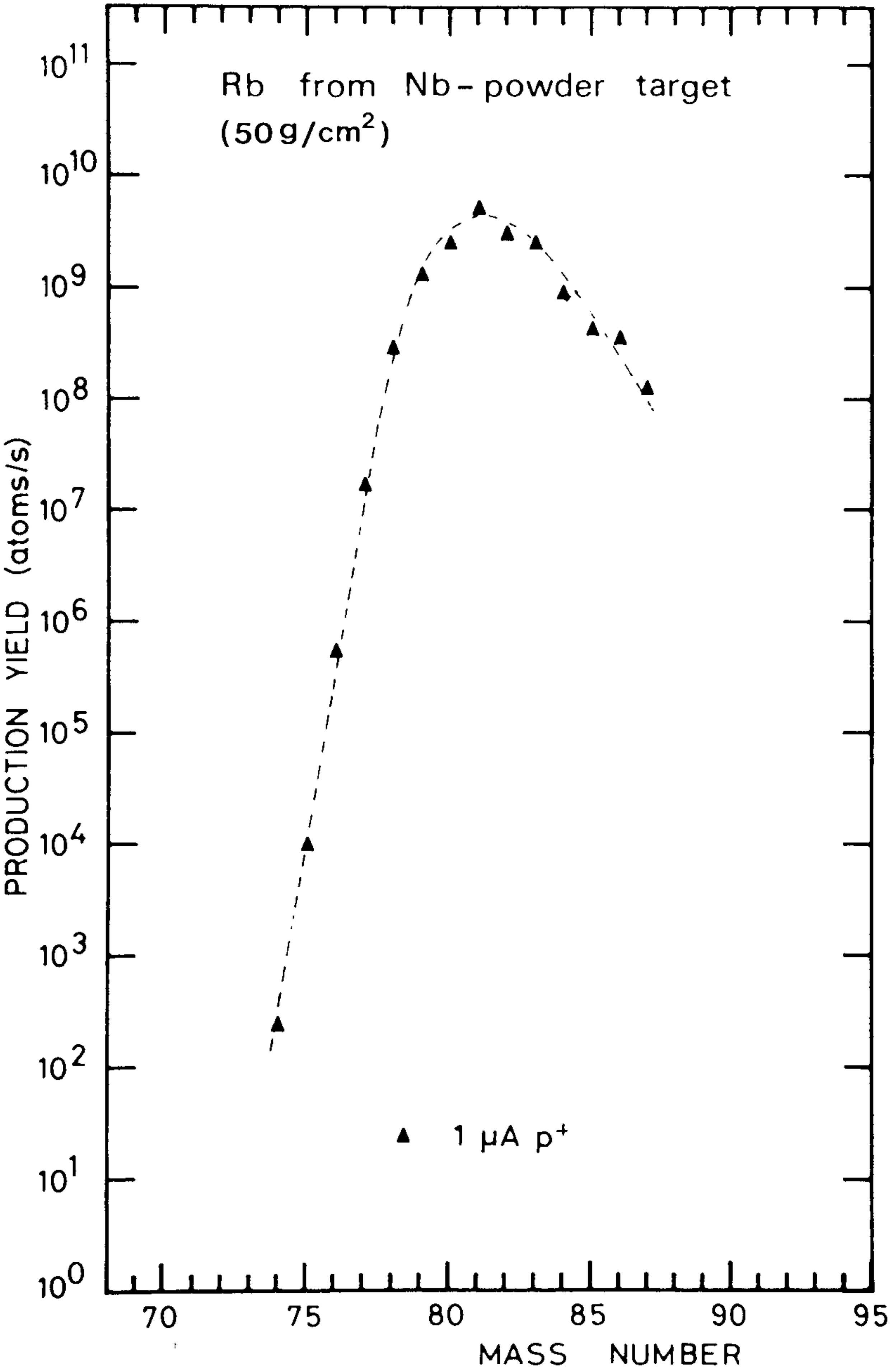
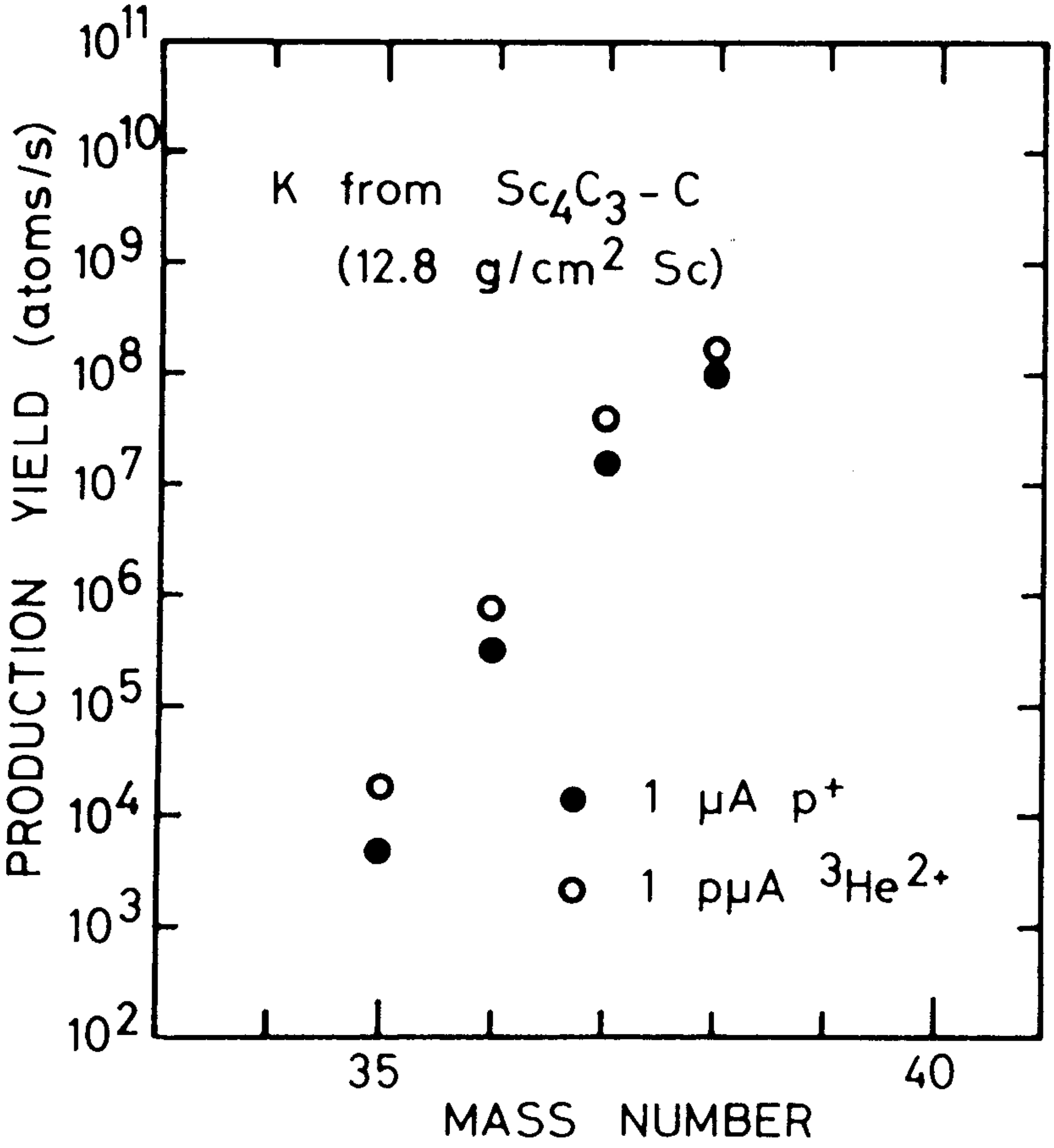

Rb from Nb-powder target
(50 g/cm²)
PRODUCTION YIELD (atoms/s)
10^{0}
10^{1}
10^{2}
10^{3}
10^{4}
10^{5}
10^{6}
10^{7}
10^{8}
10^{9}
10^{10}
10^{11}
70
75
80
85
90
95
MASS NUMBER
▲ 1 μA p⁺

Fig. 20



PRODUCTION YIELD (atoms/s)
10^{11}
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
10^{3}
10^{2}
K from Sc_4C_3 - C
(12.8 g/cm^2 Sc)
● 1 μA p^+
○ 1 pμA $^3He^{2+}$
35
40
MASS NUMBER

Fig. 21

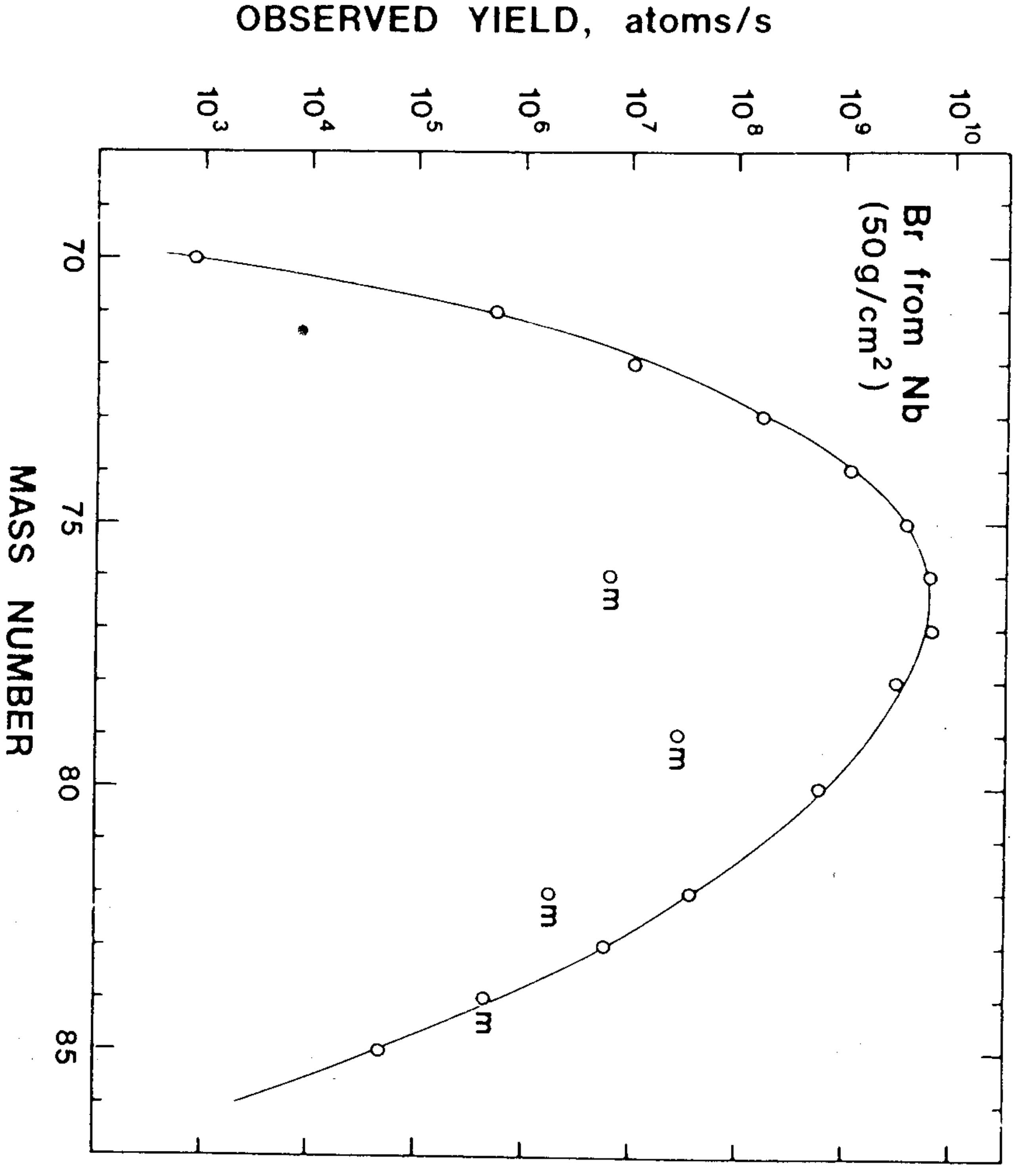
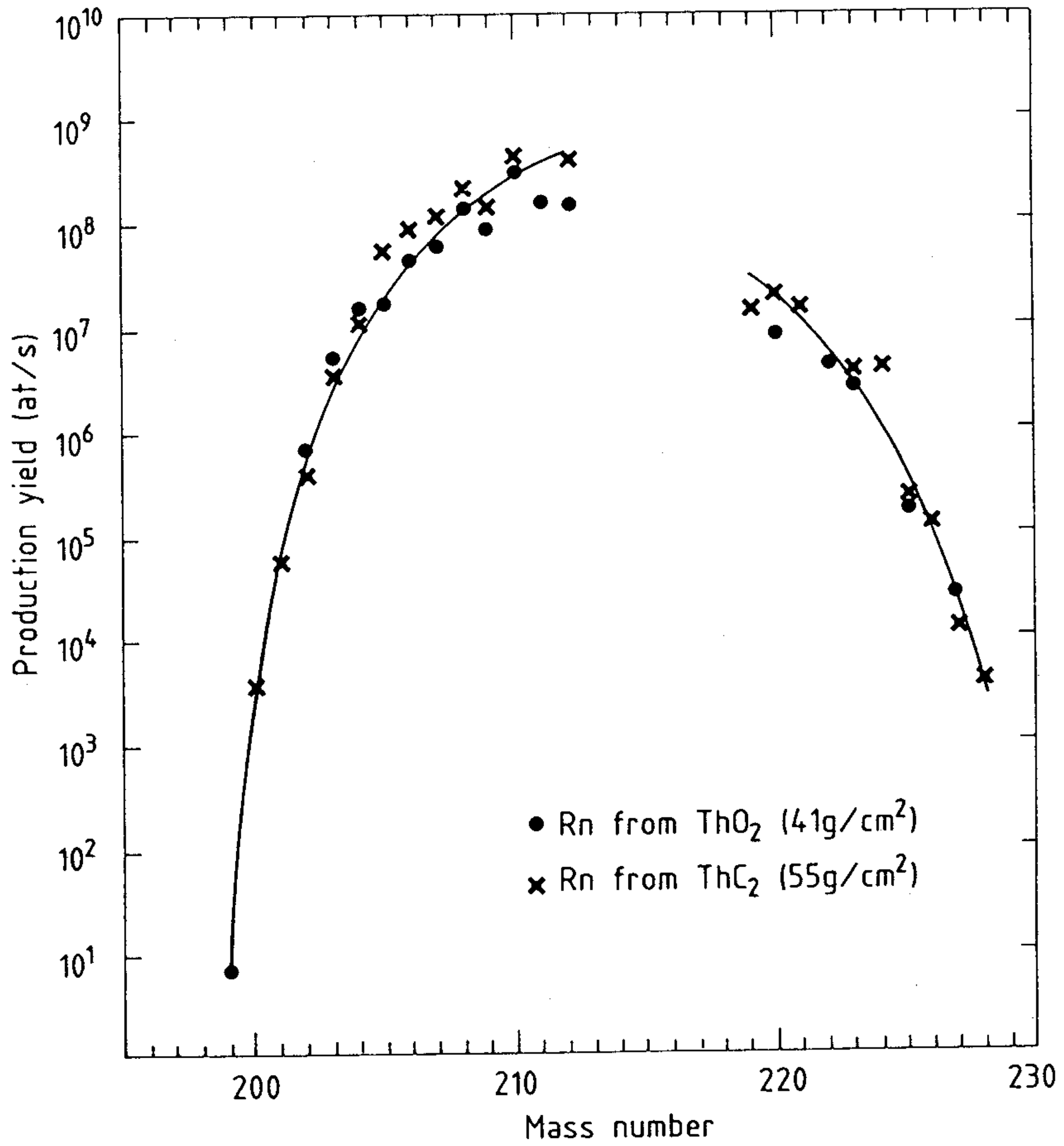

OBSERVED YIELD, atoms/s
10^3
10^4
10^5
10^6
10^7
10^8
10^9
10^10
Br from Nb
(50g/cm^2)
m
m
m
m
70
75
80
85
MASS NUMBER

Fig. 22



10^{10}
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
Production yield (at/s)
200
210
220
230
Mass number
● Rn from ThO_2 (41g/cm^2)
× Rn from ThC_2 (55g/cm^2)

Fig. 23

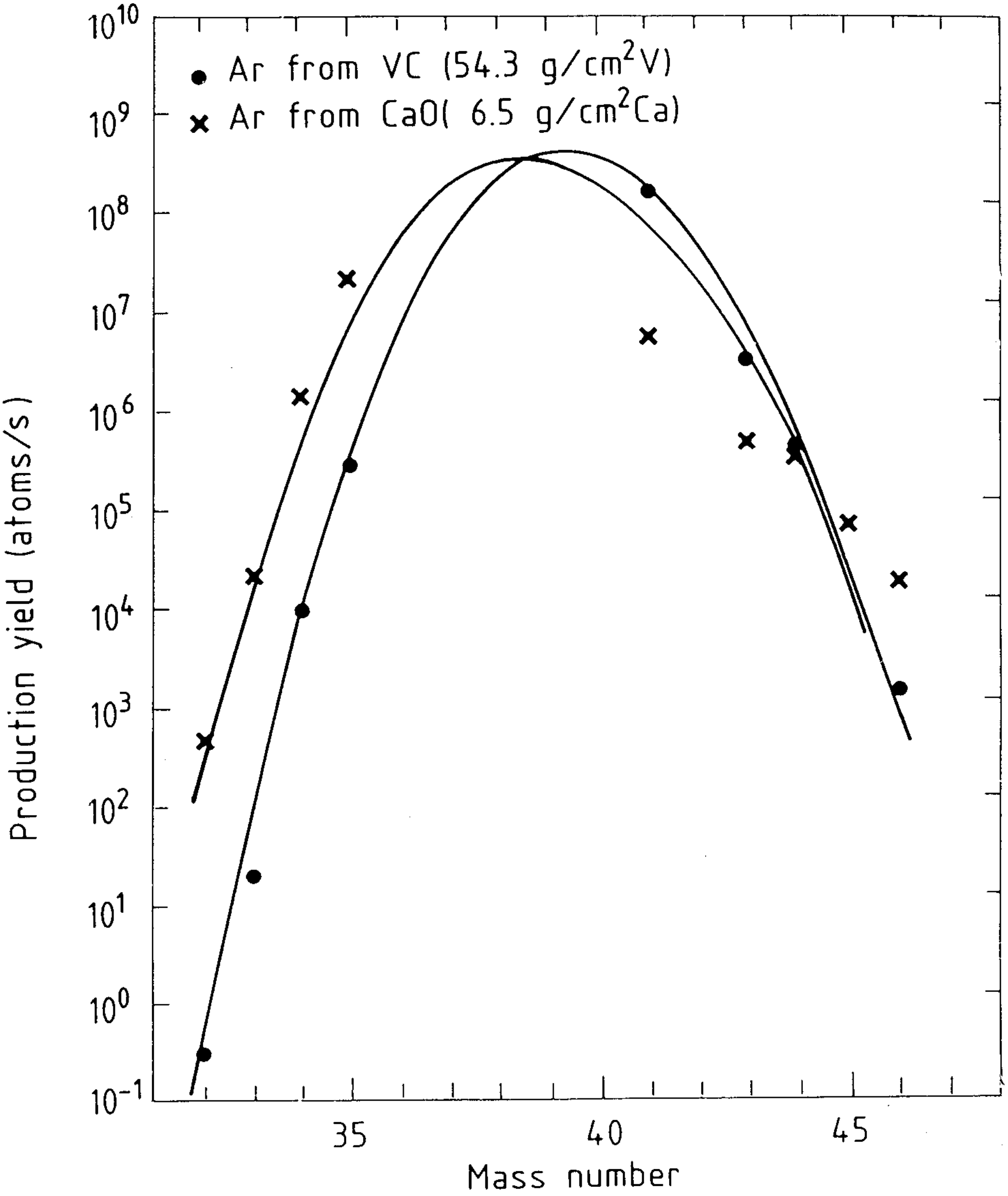
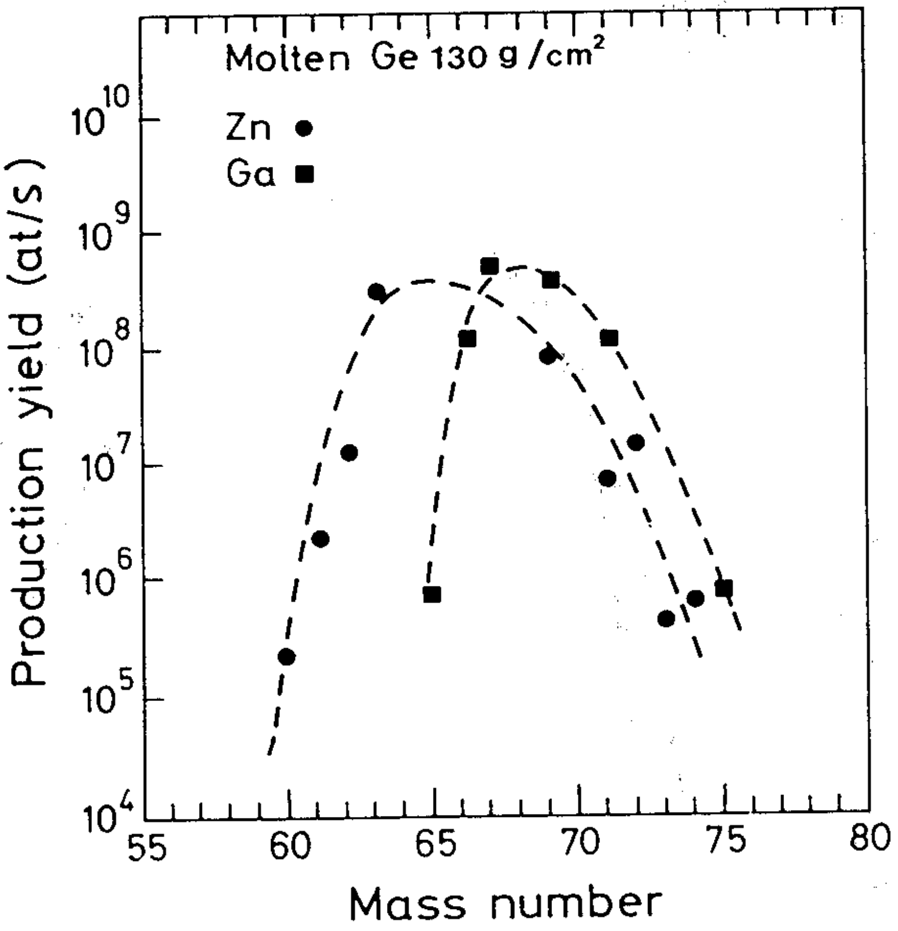

● Ar from VC (54.3 g/cm²V)
× Ar from CaO(6.5 g/cm²Ca)
Production yield (atoms/s)
10¹⁰
10⁹
10⁸
10⁷
10⁶
10⁵
10⁴
10³
10²
10¹
10⁰
10⁻¹
35
40
45
Mass number

Fig. 24



Molten Ge 130 g /cm²
Zn ●
Ga ■
Production yield (at/s)
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
55
60
65
70
75
80
Mass number

Fig. 25

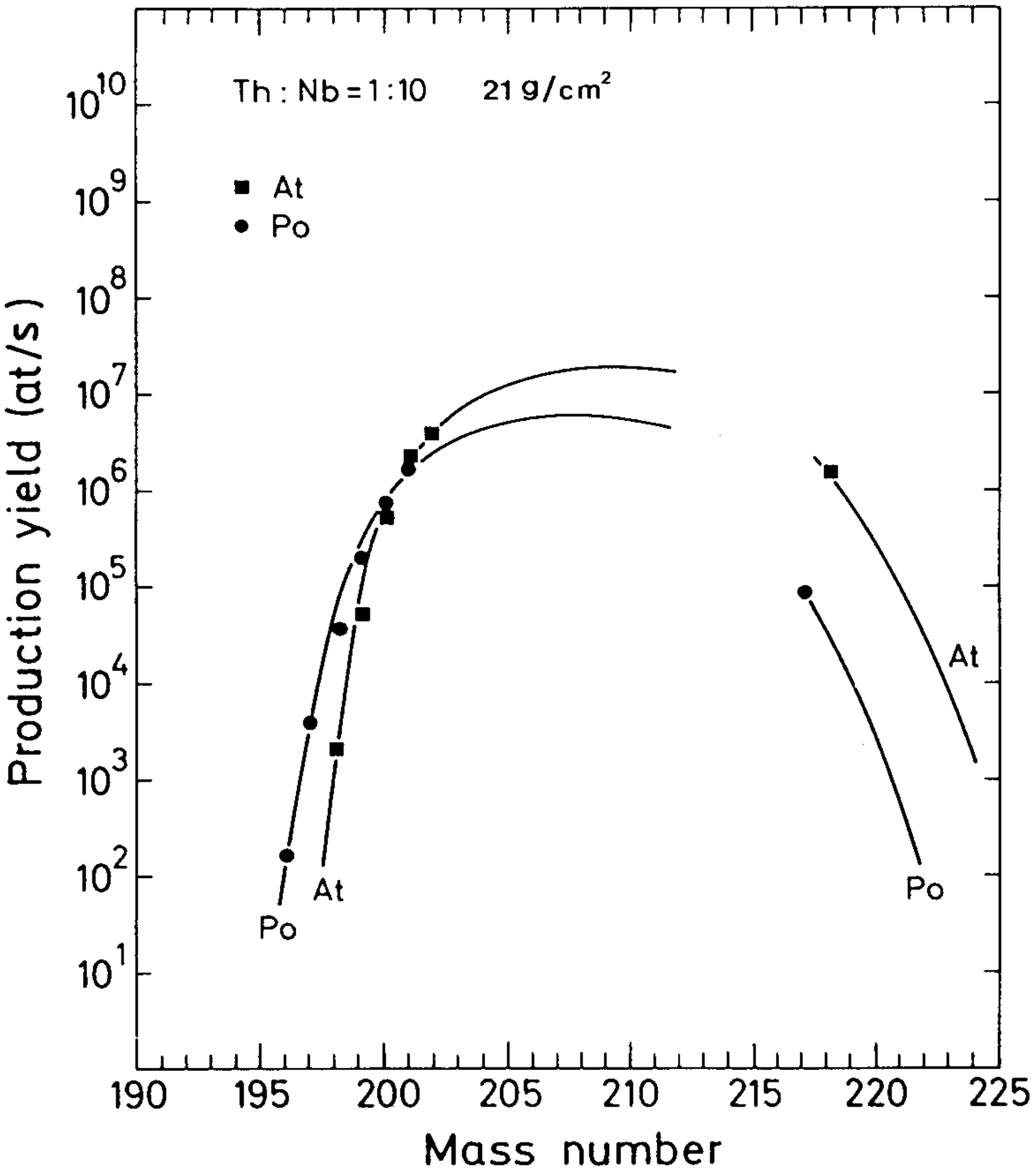

Th : Nb = 1 : 10   21 g/cm²
■ At
● Po
Production yield (at/s)
10¹⁰
10⁹
10⁸
10⁷
10⁶
10⁵
10⁴
10³
10²
10¹
190
195
200
205
210
215
220
225
Mass number
Po
At
At
Po

Fig. 26