EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

PROGRESS IN TARGETS AND ION SOURCES FOR ON-LINE SEPARATORS

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ABSTRACT

The application of on-line mass separation to the study of nuclei far from stability on a broad scale has been made possible by the development of new ion-source and target arrangements. These units often integrated detemine together the elements available as well as their yields and chemical purity.

The paper summarizes recent developments in this field and the material will be discussed under three main headings according to the types of bombarding particles used for the production of far-unstable nuclei.

The selected examples will be given for

i) ion sources and targets for thermal fission;

ii) heavy-ion target and ion-source combinations;

iii) target and ion-source systems for high-energy proton reactions.

The major items of progress include gaseous discharge ion sources and surface ionization sources developed to meet the special requirement of on-line separators. Significant improvements in the delay-time characteristics have been demonstrated with high-temperature targets (T > 2000°C), consisting of refractory metals or chemical compounds. This technique now permits the study of nuclides with very short half-lives.

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1. INTRODUCTION

Like most other fields of physics, the study of nuclei far from stability is strongly dependent on the development of new experimental approaches. A major part of the last years' progress in this field is notably due to new developments in ion sources and targets for producing element selected beams of radioactive ions for on-line mass separation. It is a pleasant, but also somewhat difficult, task to summarize the new experimental techniques since there is no shortage of target and ion-source configurations on which interesting progress can be reported. Because the presentation here is necessarily brief, I will restrict myself to giving selected examples which illustrate what the new techniques are and in what direction further progress seems likely. For discussion purposes the examples are grouped according to the three types of nuclear reactions currently used for the production of the far-unstable nuclei: high-energy proton reactions, fission and heavy-ion reactions.

Before turning to the new techniques I would like, however, briefly to remind you of the basic difficulties and requirements in producing beams of short-lived radioactive nuclei.

2. GENERAL PROBLEMS

In the on-line isotope separation the particle beam intercepts the production target situated directly inside the ion source or attached to it via a tube. By the correct choice of target matrix only the reaction products should be released in a form acceptable to the vacuum requirement of the ion source. This problem has been solved with varying degrees of success for approximately 40 elements. The crucial point, however, is that the production reactions are very complex and form many isotopes of different elements in high yields, whereas the product of interest usually is formed in a very low cross-section. The basic difficulty is therefore to obtain sufficiently high selectivity. The separation in A as performed by the mass separation gives only clean conditions in combination with a chemical separation. Such element-selective target and ion-source systems have only been achieved for about 20 of the elements.

The last important problem for the study of the short-lived nuclei is the delays inherent in the system, i.e. the time between formation of a nuclear reaction product and its collection in front of the measuring equipment. Most on-line target and ion-source systems have average delay periods in the range of tens of seconds and only a few systems are fast enough to operate efficiently in the half-life region of 10-100 msec.

3. TARGET AND ION-SOURCE COMBINATIONS FOR HEAVY-ION REACTIONS

The very short particle range for heavy ions in matter determines strongly the configuration of the target and ion-source arrangement. The effective amount of target material is so small that it can be situated conveniently inside the ion source. The high temperature here will permit a fast diffusion and evaporation of the products. The large linear momentum transfer imparted to the reaction products may be used for a fast transfer of products from an external target into the ion source, as demonstrated by Tarantin et al.[1].

3.1 The PINGIS ion source

At the PINGIS separator in Stockholm Fransson and Af Ugglas have used refractory metal targets situated inside a conventional plasma ion source with circular end extraction[2].

Recently they have further developed this concept to a molten-Bi target contained inside a novel small volume ion source as seen in Fig. 1[3]. The wall of the discharge chamber acts as a cathode heated externally by electron bombardment. The target, which consists of drops of molten Bi separated by 2 Ta tubes, is accommodated in the target end of the discharge chamber which likewise is heated by electron bombardment. This system has mainly been used to produce At and Po in (α,n) and (α,p) reactions, respectively, but it can be extended to a variety of solid and molten targets. Operated as a plasma ion source the arrangement has no chemical selectivity, but say by turning off the anode voltage and the support gas it can be operated in a surface ionization mode, so that the higher selectivity characteristic of this type of ionization can be obtained.

3.2 High-temperature surface ionizers

The surface ionization configuration used at PINGIS resembles closely the ion source developed by Raiko et al.[4]. The principle of this new high-temperature surface ionization source is seen in Fig. 2. The ionization process occurs in the cylindrical cavity of a tungsten rod heated to $\sim 2800^{\circ}C$ by electron bombardment; the ions are extracted from a 0.2 mm orifice. The remarkable property of this ion source is that yields of the rare earth and actinide[5] elements of 50-80% are obtained. This is one to two orders of magnitude more than one would expect from the Saha-Langmuir formula, which describes the surface ionization effect. Raiko and Latuszyński[6] explain this qualitatively by a complex interaction between two effects: the space charge produced in the volume by the ions and electrons flowing from the wall, and the effective extraction orifice for ions due to the penetration of the extraction field into the cavity. The ideas that emerge from this ion-source work have been used by several on-line separator groups.

3.3 The BEMS-2 ion source

The high-temperature surface ionizer used by Karnaukhov et al.[7] in the BEMS-2 separator on-line with the Dubna heavy-ion cyclotron is shown in Fig. 3. The energetic recoils which emerge from the target enter the source, which is kept at 2400°C by electron bombardment, through a 1 μ thin W window. Most of the recoils are stopped in the catcher part at the source front, from where they diffuse out and become ionized with delays of 5-10 sec and a yield of 15-40%. The efficient cooling and the application of a 50 $\mu g/cm^2$ carbon foil as heat screen between the target and the ion source allows the use of low melting targets such as Sn. The latest version of the ion source[8] is provided with a two-position target holder so that target charges can be accomplished during the experiment. This system has mainly shown its strength in the production of very neutron-deficient isotopes of the elements from Cs to Eu[9], where it is one of the most powerful separation methods. Surface ionization in the form used here unfortunately does not provide any appreciable element separation.

3.4 New gaseous discharge ion source

The discharge ion sources used in on-line separation work have usually been taken over from the off-line separators, where the requirement for long lifetime and stable high efficiency operation are less stringent. This situation is rapidly changing and ion sources specially developed to fulfil the requirement of on-line separation are now being developed[10]; we have already seen one example in the PINGIS ion source. Another important example is the ion source shown in Fig. 4, developed by Kirchner and Roeckl[11] for use in the separator on-line with the heavy ion accelerator at GSI in Darmstadt.

Since Dr. Roeckl is going to give a detailed report of their work in a moment I will simply give the few details that are necessary for this discussion. Two constructional features distinguish this source from others. The cathode is a capsule heated internally by electron bombardment. This brings the advantage of long lifetime even in the presence of agressive chemical vapours and a very good definition of the cathode emissive surface. The electron emission is enforced by means of a grid in front of the cathode which gives a good decoupling of the different ion-source operating parameters. This allows the source to be operated with stable high efficiency with pressures down to 2×10^{-5} Torr, which is almost two orders of magnitude below the operation pressure of most other arc-discharge ion sources. The lateral openings allow the heavy-ion beam or the reaction recoils to be admitted to the source. Of course, this source is not limited only to being used in connection with heavy ions; in fact we have at ISOLDE adopted this principle to our proton target and ion-source system.

4. TARGET AND ION-SOURCE SYSTEMS FOR HIGH-ENERGY PROTON REACTIONS

The use of protons in the GeV range to produce nuclei far from stability is experimentally very attractive. The spallation reaction is a most powerful means for forming neutron-deficient nuclei. The formation cross-sections for neutron-rich nuclides from high-energy proton-induced fission is generally much smaller than those of thermal neutron fission. It is, however, a very practical production method for the heavy isotopes outside the fission product region. Taking into account that the present target technology allows the utilization of proton beams of the order of 10^{14} sec^{-1}, also the formation of products in the region of the fission mass peaks becomes quite attractive. The very long range of high-energy protons in matter (typically 100-300 g/cm^2) demands target thicknesses of the same order of magnitude if a reasonable part of the beam is to be utilized. The deleterious effects of the beam are consequently weakened by spreading out over a large volume of material. The choice of target matrix is not only restricted to refractory materials, but allows the use of a wide choice of elements and chemical compounds.

4.1 The ISOLDE-2 target and ion-source system

Most ISOLDE targets work on the basis of molten metals[12,13], where the volatile reaction products are released by an evaporation from the surface of a low-vapour pressure melt kept in a container outside the ion source. Together with targets of refractory ceramics[10], metal powders[14] and molten salts, systems capable of producing the wide range of elements shown in Fig. 5 have been developed. Emphasis in this work has been put on systems with chemical selectivity. In general, the vapour pressure conditions are such that several neighbouring elements are released simultaneously from the melt. However, by different ion-source techniques we have in most cases been able to restore the chemical selectivity. To give you an idea of the technology involved I will give you the salient features of our molten-thorium target just as an illustration.

A schematic view of the target is seen in Fig. 6. The target material is contained in a half-filled Ta cylinder, placed axially in the proton beam. Since we have not been able to find a container material that will withstand thorium at its melting point of 1800°C we have chosen a Th-La alloy that melts at 1100°, thus reducing the corrosion of the Ta container to an acceptable limit. In order to speed up the bulk transport processes the target is heated to 1300°C, well above the melting point of the Th-La alloy, by ohmic losses from a current of typically 500 A. (I may interject here that a 1 μA proton beam deposits approximately 250 W in this kind of target arrangement, so that the maximum obtainable proton beam of 5 μA will deliver about the power needed to keep the target molten. As to an upper limit for the power dissipation, we believe that we can handle a factor of 10 higher.)

Of the elements formed by spallation of Th, only Ra, Fr and Rn are volatile enough to be released from the melt and to be transferred by diffusion through the heated side tube leading to the ion source. If a beam of Fr^+ is wanted, the source is constituted by a 3 mm Ø Ta constriction at the end of the transfer line. The surface ionization effect on the 1000°C hot Ta allows Fr to be ionized with an efficiency of 75%, whereas the ionization efficiency of Ra and Rn is less than 10^{-6}%. In order to make a Ra^+ beam, the Ta surface ionizer is replaced by one made of Re and the temperature is raised to about 2000°C. We have not measured the ionization efficiency now obtained for Ra, but an efficiency of 50% is expected from calculations by the Saha-Langmuir formula.

The Re surface ionization cannot provide a means of separation of Ra from Fr, which is also ionized with high efficiency. In order to restore the chemical selectivity of the target system for Ra, we intend to make use of the specific tendency of the group II elements to form molecular ions in surface ionization. By addition of small amounts of fluoride vapours to the target assembly, it has been possible in test experiments to divert more than 90% of the radioactive Ra nuclides into the RaF^+ side band, which displaces the Ra spectrum 19 mass units away from the Fr spectrum. Pure beams of Rn isotopes, efficiently discriminated against Fr and Ra, have been obtained by lowering the temperature on the transfer line so that Ra and Fr are condensed. The ionization of Rn is done by means of a plasma-type ion source. A very elegant possibility to obtain selectively halogen beams (and in the case discussed here, At) is to make use of the negative surface ionization effect as demonstrated by Venezia et al.[15] and Reeder et al.[16]. The expression which governs the ionization yield in this case in $n_-/n_0 = \alpha\, e^{(A-\Phi)/KT}$ where n_- is the number of negative ions, n_0 the number of neutrals, α a constant, A the electron affinity, K Boltzmann's factor and T absolute temperature. For the halogens the formula predicts yields of 1% on a 2000°C graphite ionizer. Due to technical difficulties only yields of 10^{-4}% have been achieved. The experimental yields of Fr and Ra obtained by irradiating the 30 g/cm^2 Th target with 10^{11} protons/sec is seen in Fig. 7. With a proton beam of 4×10^{13} sec^{-11} and improvements in the target and proton-beam geometry, the yield is expected to rise by a factor of up to 1000. Running at such high intensities begets a series of problems related to repair and exchange of the highly radioactive target and ion-source assemblies. These are therefore constructed as simple compact units, of which four can be accommodated on a turntable near to the separator and interchanged by remote control as shown in Fig. 8.

4.2 The ISOCELE integrated target and ion source

The molten-metal targets are operated at temperatures high enough to give a fast release of the products. On the other hand, the temperature is limited by the onset of massive evaporation of the target itself; this clearly will interfere

with the operation of the ion source. The use of a high current isotope separator will make it possible to work at higher temperatures and also to isolate more elements, as has been demonstrated by Puteaux et al.[17] at the ISOCELE separator on-line with the Orsay synchro-cyclotron.

This group has made use of the relative short range of their proton and ^{3}He beams to accommodate the target directly inside the discharge chamber of a high-intensity ion source shown in Fig. 9. The advantage of this system is that up to 10 mA currents can be handled, which allows the use of target product combinations where the equilibrium vapour pressure of the target material is much higher than can be used in the more generally used ion sources. In fact, a considerable part of the target is lost during the operation of the system. A further advantage is that the target surface can serve as the anode of the ion source, so that it is continuously bombarded by the plasma. Since the delay of the release from a molten metal is most likely in the surface desorption step, the plasma bombardment may serve to reduce the delay.

5. ION SOURCES AND TARGETS FOR THERMAL FISSION

Several chemical compounds of ^{235}U have been used as targets in separators on-line with reactors and neutron generators. The emanating power of uranium stearate was used very early for the fast selective production of the rare gases Kr and Xe[18]. Uranium tetrafluoride has the property of releasing elements in the Zr-I region[19]. The formation of a very pure beams of the alkali metal ions with delay-time components of 5 msec was pioneered by Klapisch et al.[20] in their mass spectrometric studies of fission. Their application of the exceptional fast diffusion of the alkali elements in graphite, in combination with selective surface ionization, has given rise to a number of solutions to the target problem. Cowan and Orth[21,22] showed, in their studies related to the loss of individual fission products from uranium-graphite fuel elements, that a variety of other elements exhibited relatively fast diffusion in graphite at temperatures around 2000°C. I think this principle of the target element being dispersed inside or on the outside of a graphite matrix deserves attention, since it offers many possibilities for improvements and extension to other elements and particles.

5.1 The OSIRIS target and ion source

The OSIRIS on-line isotope separator group at Studsvik has extended this target concept to many more elements and to higher production rates. Perhaps this experiment[23] on-line with a reactor is the most important separator for a large-scale production and study of the fission products. The plasma ion source which contains the target material occupies here the most favourable position on the inside of the reactor shield near to the core where the flux is 4×10^{11} neutrons per cm^2 sec. The target arrangement shown in Fig. 10 contains $^{235}U_3O_8$ as a cover onto

the inner surface of the anode cylinder. A second cylinder made of graphite cloth heated by the plasma to about 1300° serves as a recoil catcher that allows about 18 fission product elements simultaneously to diffuse to the surface and evaporate into the ion source. The group has recently developed a new target arrangement[24], which consists of finely divided $^{235}U_3O_8$ particles distributed onto the fibres of a graphite cloth cylinder. In this way, the effective target amount was increased by a factor of 10 to 3 g of ^{235}U. Still the delay time of only 0.5 sec was retained. The corresponding increase in production rate brought the most abundant beams up to an intensity of 10^8 atoms/sec. The efficiency[25] of the target and ion source varies from 30% down to 0.1% and, as seen from the activity scan in Fig. 11, the system has no appreciable separation from the isobaric precursors.

6. DELAY IN TARGET AND ION-SOURCE SYSTEMS

The analysis of the hold-up inherent in the target and ion-source assembly is important for the development of faster targets, as well as for the measuring of nuclear reaction yields by means of on-line mass separation.

From a number of experimental and theoretical studies[1-7,12,14,26-29], we have today a good understanding of the processes which determine the delay. Usually the delay-time characteristics of a target system are described by the distribution $p(\tau)$ in the values of delay times τ from the instant the nucleus is formed until it is detected. Most often $p(\tau)$ is determined by differentiation of the experimentally measured integral delay curve $A(t)$. The measured count rate A of a shielded long-lived product (half-life $T_{1/2} = \ln 2/\lambda$) sampled at regular intervals Δt ($\ll T_{1/2}$) as a function of time t after switching off the beam, can be expressed as

$$A(t) = \lambda R \, \Delta t \int_t^\infty e^{-\lambda\tau} \, p(\tau) \, d\tau \, , \qquad (1)$$

where R denotes the production rate including the over-all separator efficiency. Typical examples of such curves are shown in Fig. 12 for two different ISOLDE targets. The distribution $p(\tau)$ allows us to derive a release yield, the ratio between the observed yield Y_{obs}, and the true (i.e. decay loss corrected) yield Y. Table 1 summarizes the results for the two target types mentioned. Applied to the recently observed[30] N = Z nuclide ^{74}Rb ($T_{1/2}$ = 60 msec) a release yield of 5% is obtained when produced from our high-temperature Nb target, whereas with our molten-Y target only a yield of 1‰ can be achieved. An interesting possibility to reduce the delay times of liquid targets has been demonstrated by Ansaldo et al.[31] at Princeton. In their system the target liquid was allowed to boil under reflux, such that only the Ar reaction products were allowed to diffuse to the ion source. In this way delay times of the order of 1 sec were achieved.

7. CONCLUDING REMARKS

I have presented a set of selected examples on the design of targets and ion sources for on-line mass separation. My list is far from complete and many more examples of current interest could be quoted. I hope, however, that the few examples discussed here illustrate and demonstrate that a new wave of progress is in the making and that there are strong indications that further research in this field will pay off in the future.

It is interesting to note that as in most technologies no single solution is the best possible choice in all situations and as far as the connection of element-selective targets to an isotope separator is concerned many interesting problems are awaiting their solution. I think this is a vigorous new field that should present a great challenge to ion-source builders and users.

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Table 1

Delay-time results

Target	Release-determining mechanism	Delay-time distribution $p(\tau)$	Release efficiency Y_{obs}/Y
Porous solid with spherical particles	Diffusion in the particles	$\frac{6D}{r^2} \sum_{n=1}^{\infty} e^{-n^2\pi^2 D\tau/r^2}$	$\frac{3}{r}\left(\frac{D}{\lambda}\right)^{1/2} \times \frac{\nu}{\lambda + \nu}$
Molten metals	Desorption from the liquid surface?	$\lambda^* e^{-\lambda^*\tau}$	$\frac{\lambda^*}{\lambda + \lambda^*}$

D : diffusion coefficient

r : particle radius

ν : delay constant, depending on the volume of the ion source and target

λ : decay constant

λ^* : delay constant for a liquid target

Figure captions

Fig. 1 : Schematic view of the PINGIS molten-Bi target.

Fig. 2 : High-temperature surface ionizer and its power supply scheme. 1 - Heating shield and electron reflector; 2 - cathode of the ionizer; 3 - Ionizer and vaporizer (a - Re foil, b - ionizer, c - vaporizer chamber, d - vaporizer); 4 - cathode of the vaporizer; 5 - movable rod; 6 - support and cathode supply leads.

Fig. 3 : Schematic view of the BEMS-2 ion source with external target; 1 - W ionizer; 2 - recoil catcher; 3 - filaments; 4 - entrance foil; 5 - Ta ring; 6 - thermal shields; 8 - Ta ring; 9 - extraction electrode; 10 - target holder.

Fig. 4 : The FEBIAD ion source and its power supply scheme. a - Outlet plate; b - anode with radial holes for target connection and heavy ion beam; c - grid for primary electron extraction; d - capsule cathode, heated internally by electron bombardment.

Fig. 5 : Periodic table of the elements.

Fig. 6 : The ISOLDE-2 target and ion-source system.

Fig. 7 : Yields of Fr and Ra from a molten Th target.

Fig. 8 : The automatic coupling of the ISOLDE target and ion-source unit.

Fig. 9 : Schematic view of the ISOLDE integrated target and ion source.

Fig. 10 : The OSIRIS target and ion source.

Fig. 11 : Activity of samples collected by scanning across the collector chamber of the OSIRIS separator. The broken curve in the figure is the mass yield curve of thermal-neutron induced fission of ^{235}U.

Fig. 12 : Integral delay curves normalized to one. The filled circles denote the delay of ^{84}Rb from a molten Y-La alloy at 1300°C. The open circles denote the delay of ^{84}Rb from a Nb powder target at 2200°C.

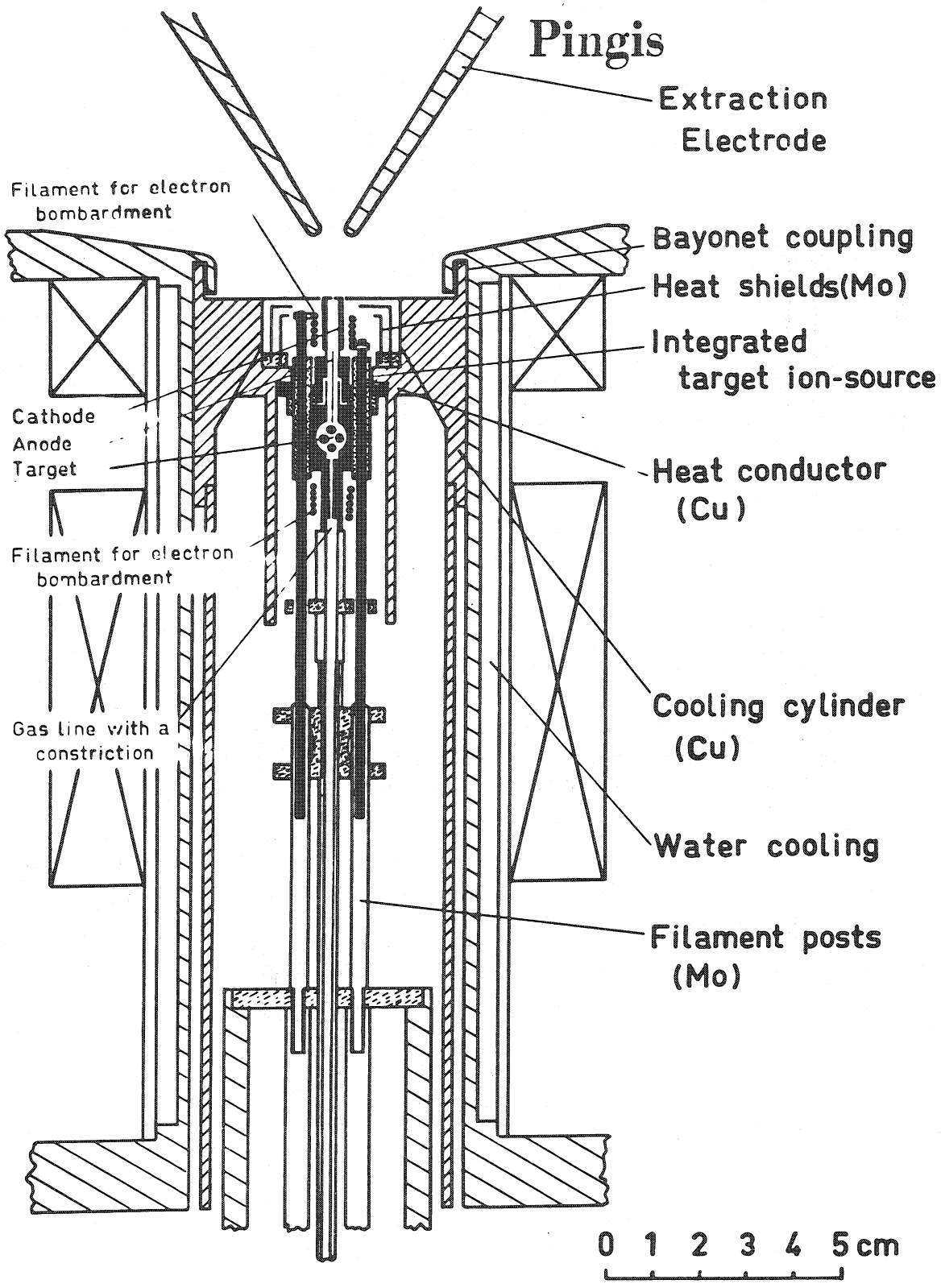

Pingis
Extraction Electrode
Filament for electron bombardment
Bayonet coupling
Heat shields(Mo)
Integrated target ion-source
Cathode
Anode
Target
Heat conductor (Cu)
Filament for electron bombardment
Cooling cylinder (Cu)
Gas line with a constriction
Water cooling
Filament posts (Mo)
0 1 2 3 4 5 cm

Fig. 1

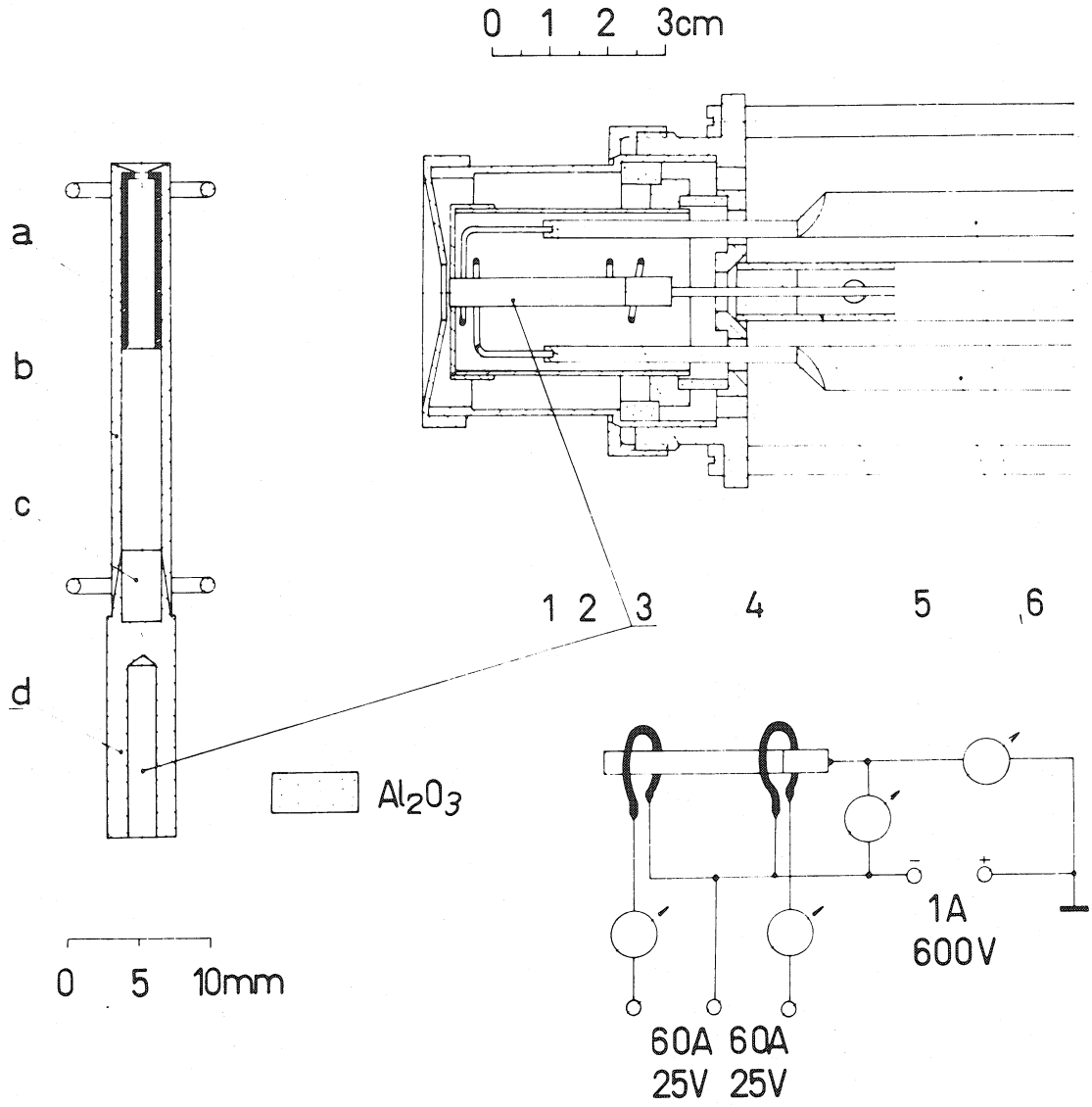


Fig. 2

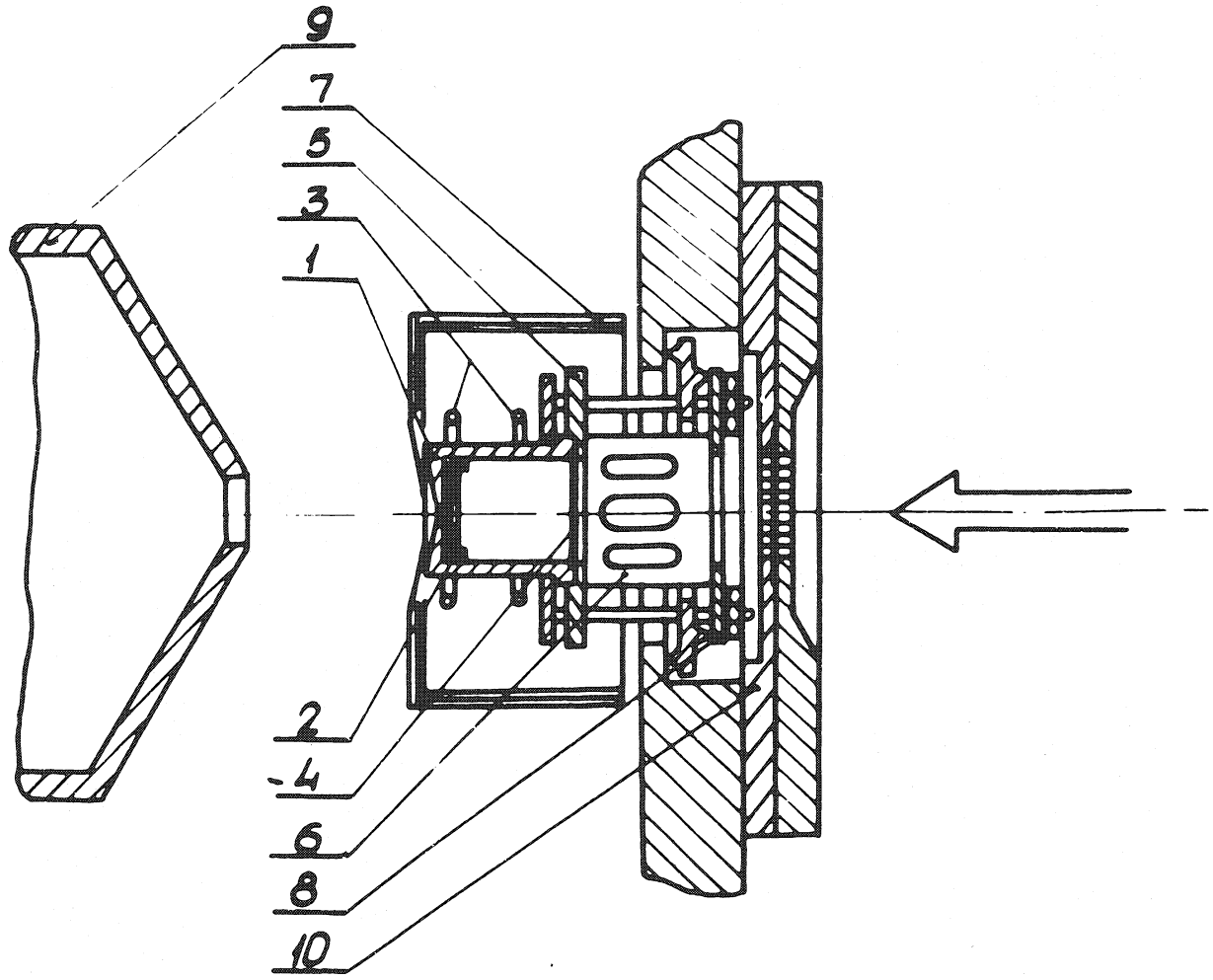


Fig. 3

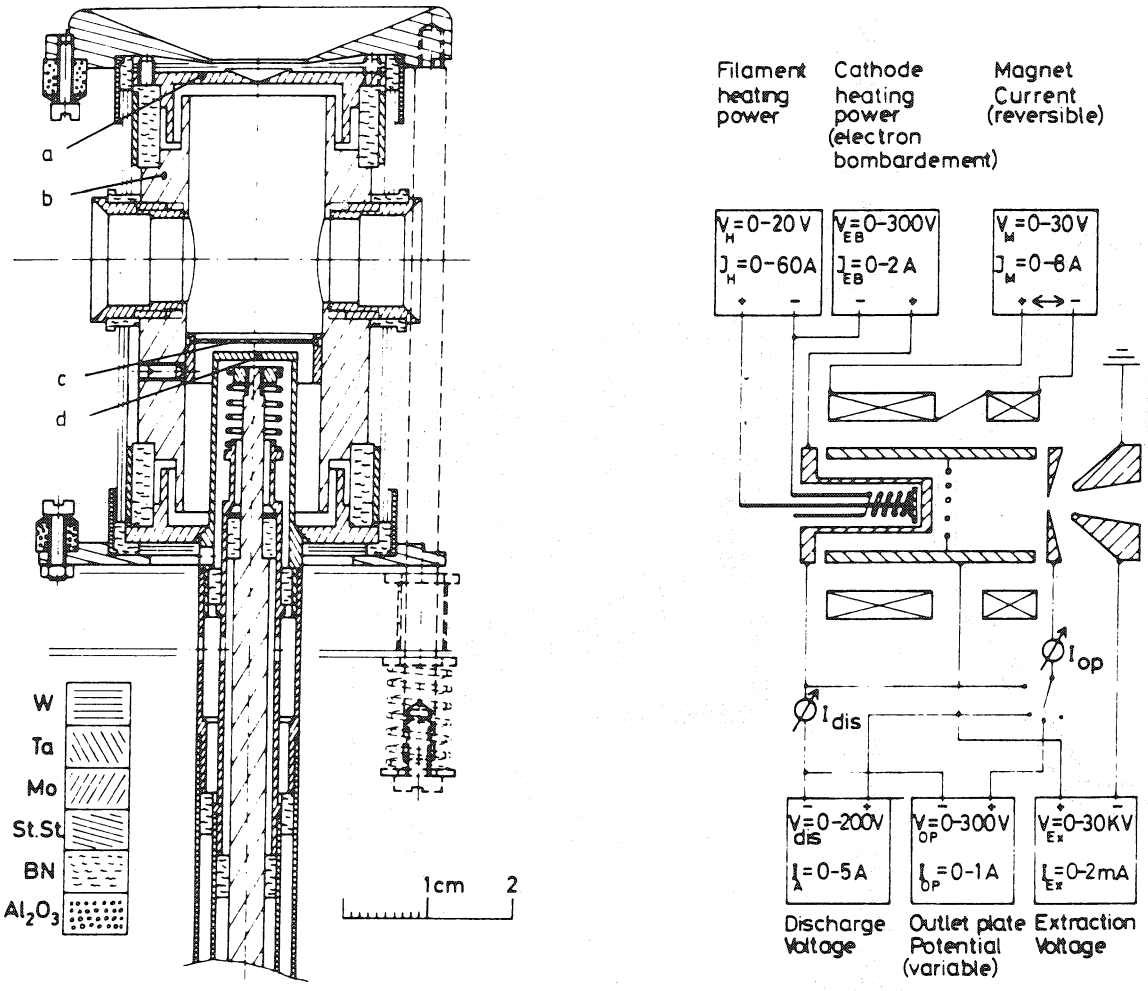
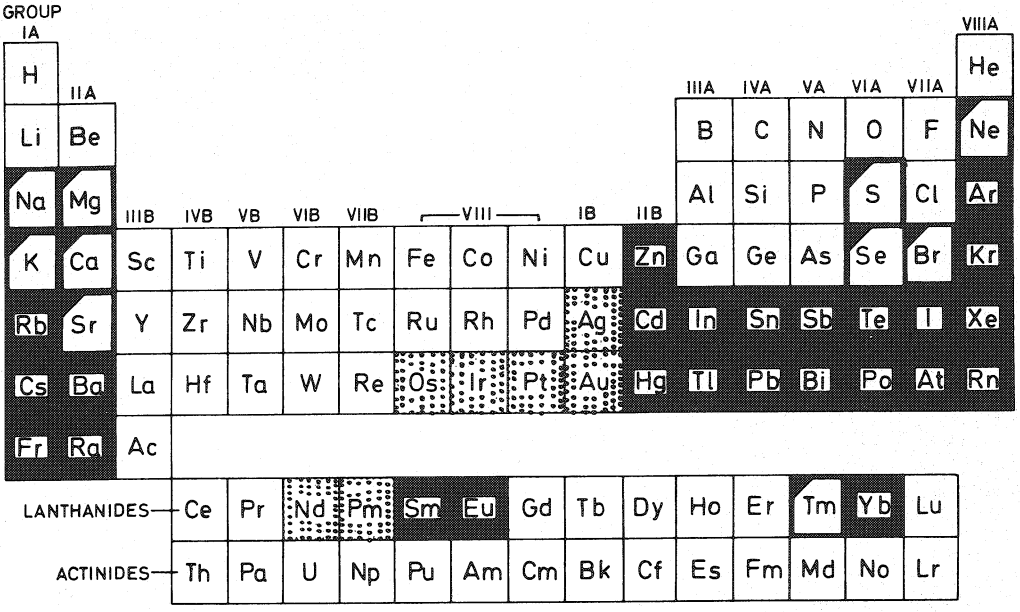

a
b
c
d
W
Ta
Mo
St.St
BN
Al_2O_3
1cm
2
Filament heating power
Cathode heating power (electron bombardement)
Magnet Current (reversible)
V_H=0-20 V
J_H=0-60A
V_{EB}=0-300V
J_{EB}=0-2 A
V_M=0-30V
J_M=0-8A
I_{op}
I_{dis}
V_{dis}=0-200V
I_A=0-5A
V_{OP}=0-300V
I_{OP}=0-1A
V_{Ex}=0-30KV
I_{Ex}=0-2mA
Discharge Voltage
Outlet plate Potential (variable)
Extraction Voltage

Fig. 4

PERIODIC TABLE OF THE ELEMENTS



Separated at ISOLDE

Within reach with present technique

Studied as decay product

Fig. 5

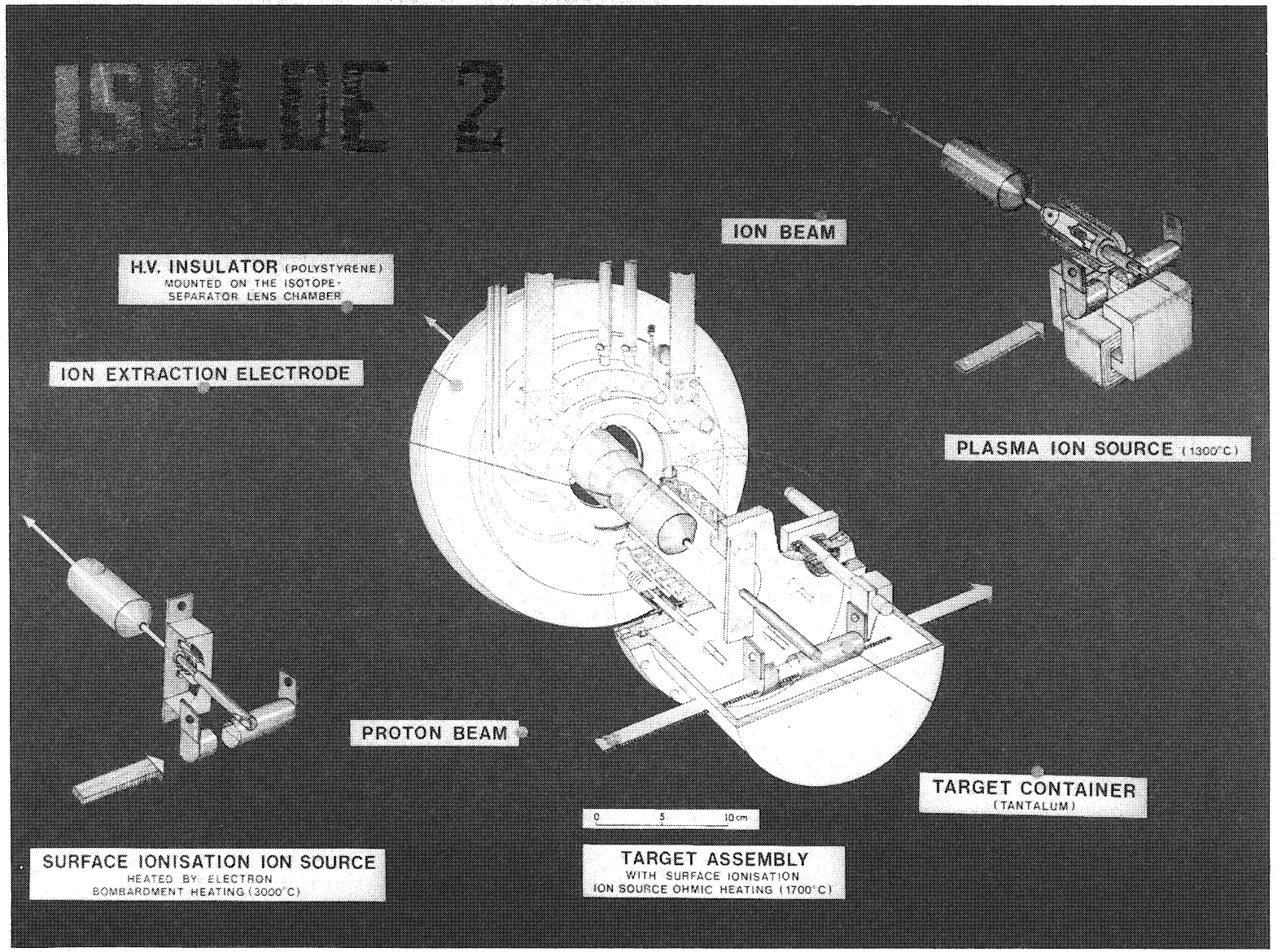

ISOLDE 2
ION BEAM
H.V. INSULATOR (POLYSTYRENE)
MOUNTED ON THE ISOTOPE-
SEPARATOR LENS CHAMBER
ION EXTRACTION ELECTRODE
PLASMA ION SOURCE (1300°C)
PROTON BEAM
TARGET CONTAINER
(TANTALUM)
0 5 10 cm
SURFACE IONISATION ION SOURCE
HEATED BY ELECTRON
BOMBARDMENT HEATING (3000°C)
TARGET ASSEMBLY
WITH SURFACE IONISATION
ION SOURCE OHMIC HEATING (1700°C)

Fig. 6

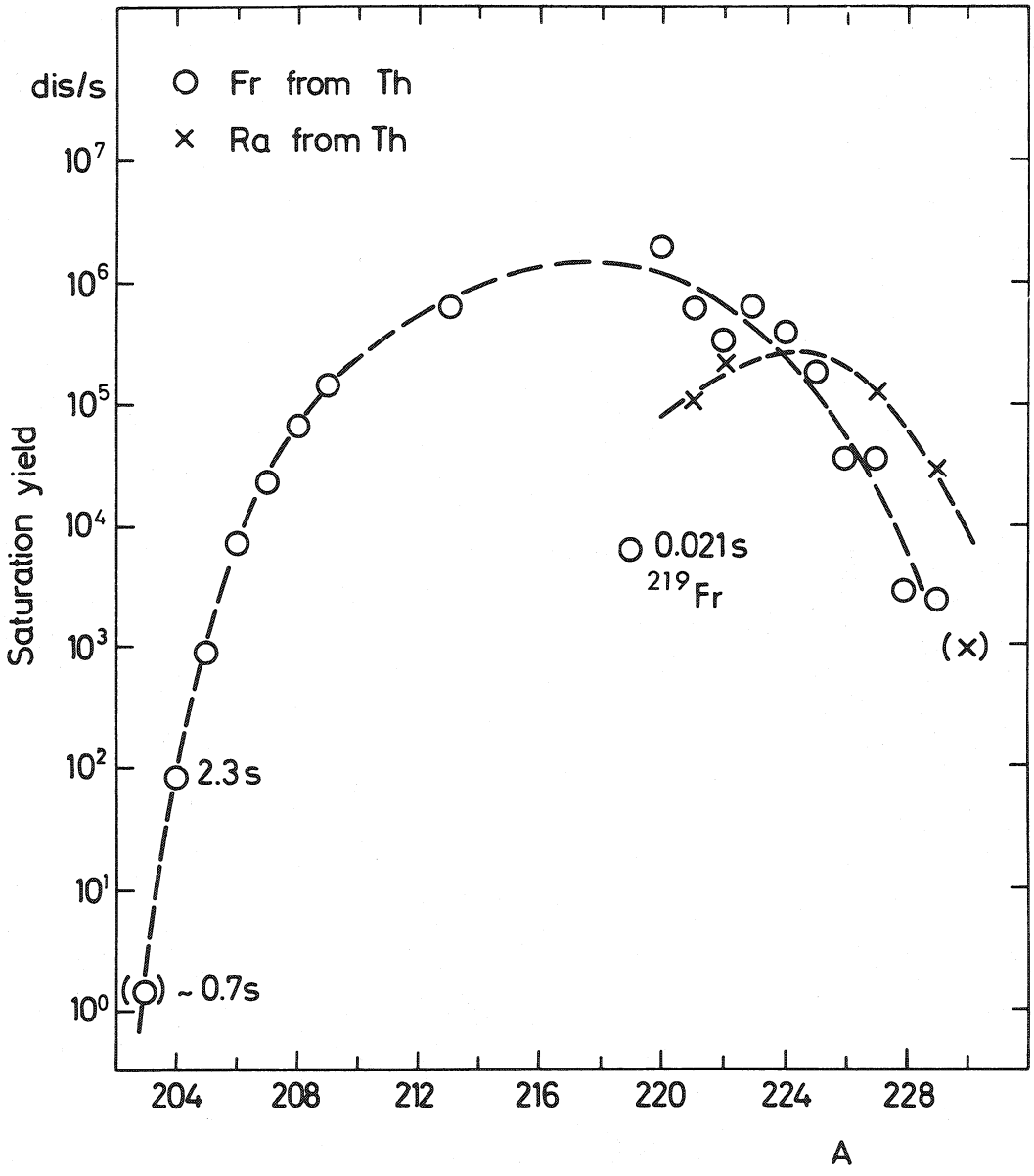

dis/s
O Fr from Th
× Ra from Th
Saturation yield
10^7
10^6
10^5
10^4
10^3
10^2
10^1
10^0
O 0.021s
^{219}Fr
(×)
O 2.3s
(O) ~ 0.7s
204
208
212
216
220
224
228
A

Fig. 7

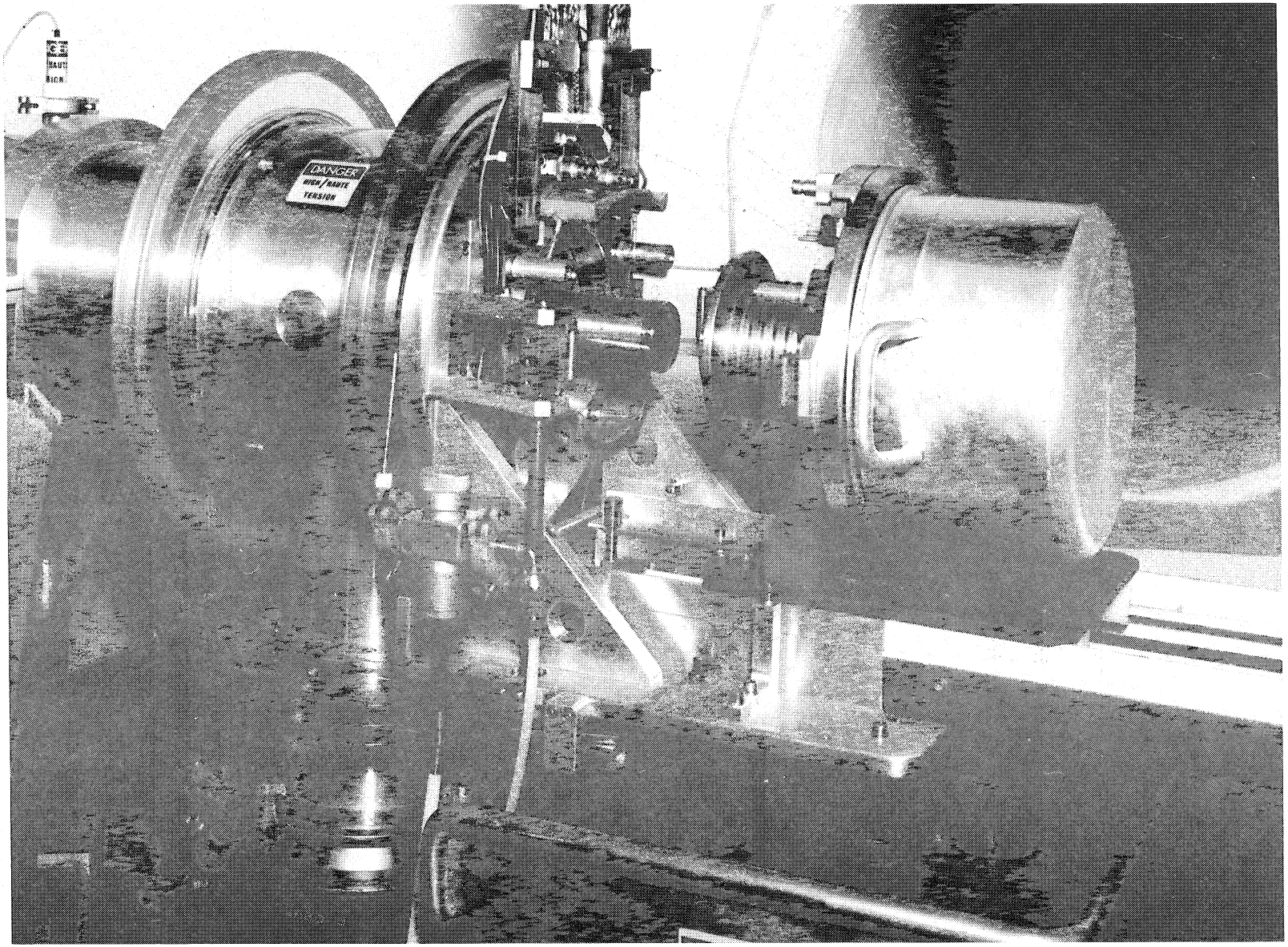


Fig. 8

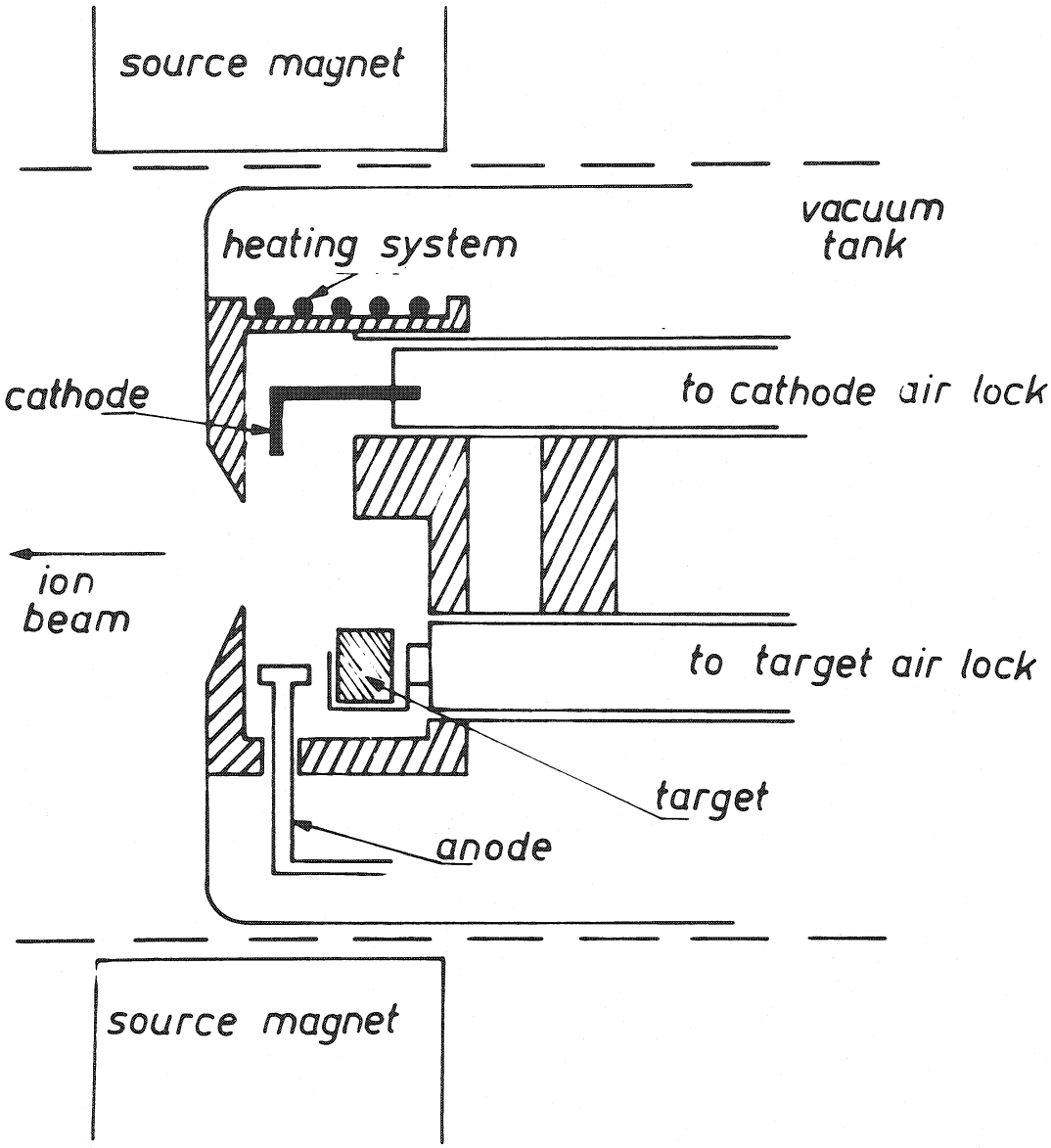

source magnet
vacuum tank
heating system
cathode
to cathode air lock
ion beam
to target air lock
target
anode
source magnet

Fig. 9

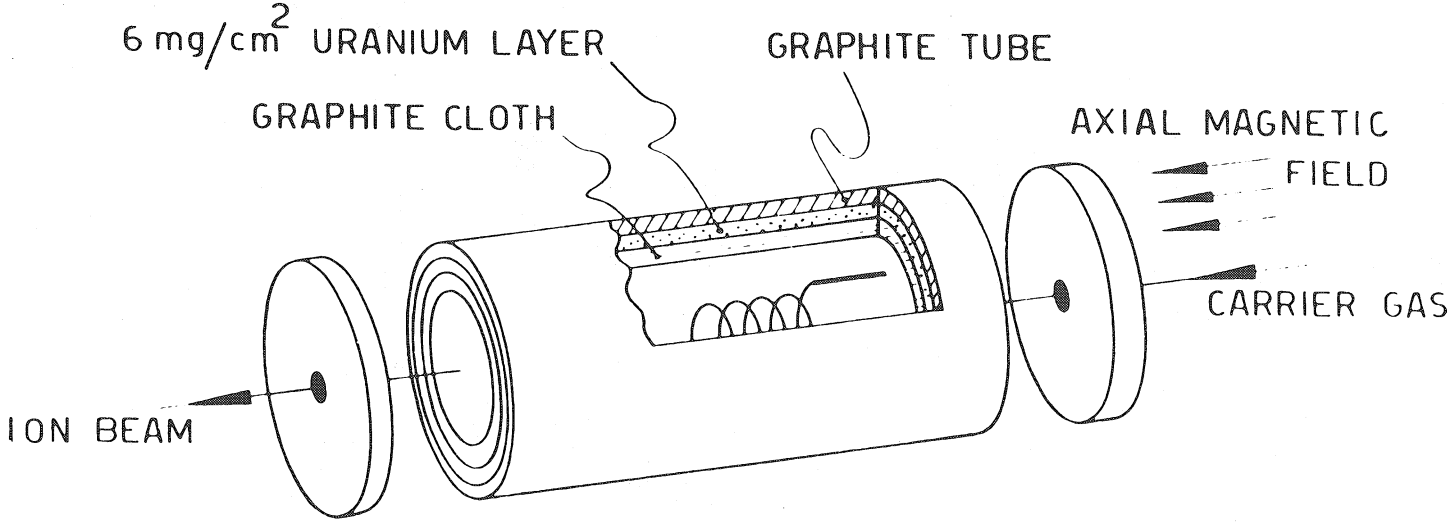

6 mg/cm^2 URANIUM LAYER
GRAPHITE TUBE
GRAPHITE CLOTH
AXIAL MAGNETIC FIELD
CARRIER GAS
ION BEAM
ANODE CYLINDER

Fig. 10

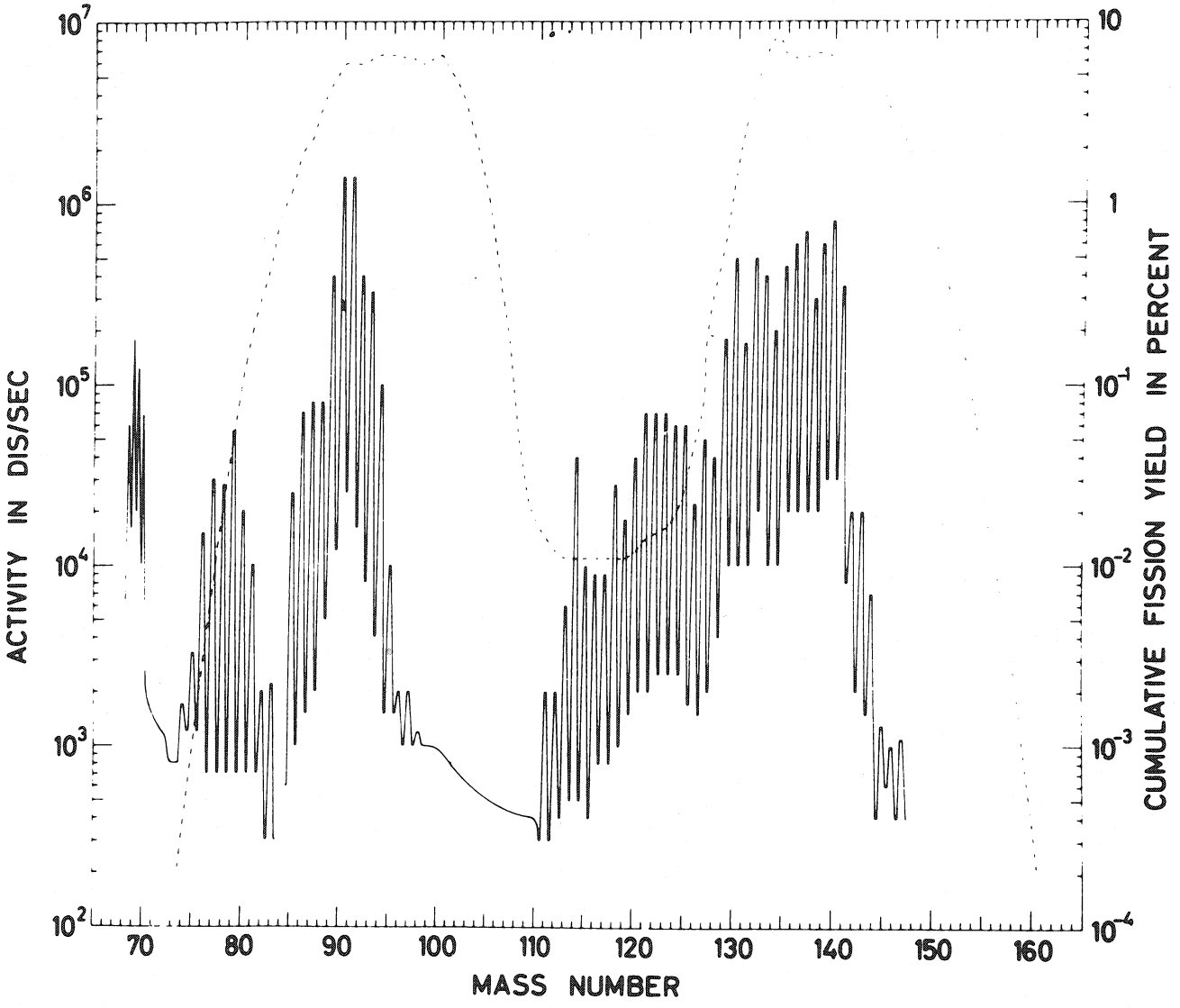

ACTIVITY IN DIS/SEC
CUMULATIVE FISSION YIELD IN PERCENT
MASS NUMBER
10^7
10^6
10^5
10^4
10^3
10^2
10
1
10^{-1}
10^{-2}
10^{-3}
10^{-4}
70
80
90
100
110
120
130
140
150
160

Fig. 11

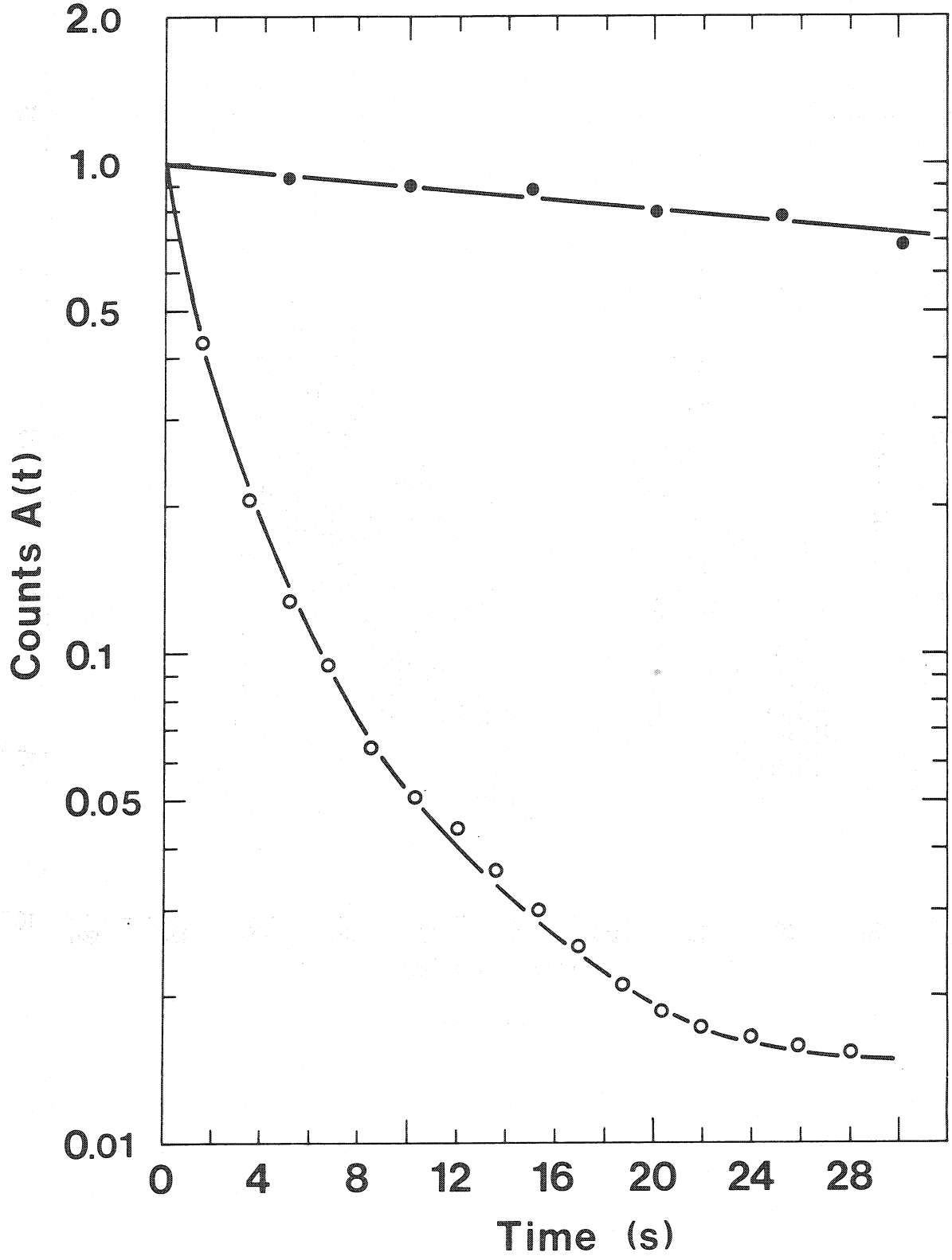


Fig. 12