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USE OF REFRACTORY OXIDES, CARBIDES AND BORIDES AS TARGETS FOR ON-LINE MASS SEPARATION

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ABSTRACT

Ion beams of a number of elements have been produced at the CERN-ISOLDE facility by 600 MeV proton bombardment of thick targets consisting of a variety of metal oxides, carbides and borides. The nuclear reaction products are released from the target material at high temperature and continuously transferred to the ion-source of the ISOLDE on-line mass-separator. The materials studied are CaB_6, ScC_2, LaC_2, TaC, ThC_2, UC_2, MgO, CaO, BaO, and ThO_2. The obtained beam intensities are of the elements He, C, N, Ne, Mg, Al, Cl, Ar, Ca, Br, Kr, I, Xe, Ba, At, Rn. The intensities are high enough to facilitate the study of hitherto unknown nuclei.

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1. INTRODUCTION

The development of targets has been an important part of the work at the ISOLDE on-line mass-separator at the CERN synchro-cyclotron (SC). In other papers in these proceedings [1,2] we have described the facility, the ion-source systems, and the development of targets consisting of pure metals. Different chemical compounds are also important as ISOLDE targets, and in the present paper the investigation of oxide-, carbide-, and boride-targets is described.

Oxides and carbides were not important as target materials in the early days of ISOLDE, but gained increasing importance when the upgrading of the facility around 1975 made it possible to heat targets up to about 2200 ^{o}C. In the present paper, we present a survey of the most recent progress in the development of these types of targets, as well as the more recently introduced borides. Details about the construction and function of the separator can be found in [1,2] and are not repeated here. Results from off-line studies of the release of nuclear reaction products are not described, unless they have not been published previously. The results of off-line tests are described in refs. [3,4].

Already in 1958 Cowan and Orth [5] showed that some fission products are rapidly released from graphite, and in 1968 a target of uranium impregnated on a graphite cylinder was successfully used at the OSIRIS fission product mass separator in Studsvik [6]. This system was further developed to a uranium impregnated graphite cloth target five years later [7]. At OSIRIS the target is kept at 1600 ^{o}C and liberates a large number of elements.

Rapid release of some elements from ceramic matrices of ThO_2 and CeO_2 was demonstrated in on-line experiments by Hansen

et al. [8] in 1973. Borides have not been extensively used in high temperature targets.

2. THE TARGET SYSTEMS

The criteria for selection of target materials are described in ref. [2]. All materials must be thoroughly tested before they are used on-line.

2.1 Carbides

Off-line tests of different types of uranium carbide targets in order to prepare their use at ISOLDE were performed by Carraz et al. [3] around 1975. The results were promising, and showed that the release of many elements was rapid, with release-half-times (i.e. 50 % released) of about 1 min for the most volatile elements already at 1600 ^{o}C. On-line tests at higher temperatures revealed release half-times of some seconds [9]. Consequently, carbides have been frequently utilized as target materials at ISOLDE for a number of years, in particular in combination with positive surface ionizers. The production of alkali elements (Li, Na, K, Rb, Cs, Fr) and most of the group 3A elements (Ga, In, Tl) from uranium carbide was reported by Carraz et al. [9] and Bjørnstad et al. [10].

The carbide targets used at ISOLDE are always made with an excess of carbon. Normally, the oxide of the target element is mixed with graphite powder, and the material is converted to carbide during a heating period when CO is released and pumped off. The amount of excess graphite depends on the use of the target. The graphite-rich targets have the most favourable delay

properties. Therefore, if the target is to be used for production of short-lived nuclei, the graphite excess is large (e.g. U:C = 1:10) in order to minimize the delay of the reaction products. If the target is intended for production of more long-lived nuclei (e.g. sources for other laboratories), the delay is uncritical, and one can use less graphite excess and more dense material. In that case, the target material may be produced by pressing the powder to tablets by means of a conventional pharmaceutical tablet machine.

Many different carbides have been tested off-line for the release of nuclear reaction products [3,4]. Most of them, but not all, have favourable release properties. Most refractory carbides show a stable high-temperature performance, and the rate of sintering seems slow compared to most other powder materials.

The carbides of the rare earth elements have turned out to be rather unfavourable from a technical viewpoint, exhibiting a prolonged outgassing and a substantial evaporation loss of target material even at low temperature. We interpret this as a consequence of the production method; reduction of oxides. The rare earth elements occur in stable, volatile molecular compounds like LaC and ScO at high temperatures, and thermodynamic data [11,12] indicate that these compounds are responsible for the loss of material. These carbides should therefore not be produced through the reduction of oxides, but through direct reaction between the metal and graphite.

There are two main disadvantages with carbides. Firstly, they often contain a relatively high amount of volatile impurities. Secondly, leakage oxygen leads to the formation of CO in the target, and in turn to a slow but problematic transport of carbon to critical surfaces in the ion-source system. Some car-

bides also have a corrosive behaviour in contact with the Ta target cylinder, and must be kept in graphite containers.

The carbon transport makes carbides unfit for use in combination with the negative LaB_6 surface ionizer. The work function of this surface is very sensitive to deposits, and in combination with carbide targets it has an unreliable performance. If carbides are to be used in combination with plasma ion-sources, the transfer line between target and ion source should be cooled, in order to preserve a high ion-source efficiency which otherwise may be destroyed by the material outgasing from the large target. The positive surface ionizers are less sensitive to this kind of impurities.

2.2 Oxides

Due to the difficulties in combining a negative surface ionizer with a carbide target, ThO_2 has so far been extensively used for the production of Cl, Br, I and At production. This system has been described previously [13] and will also be briefly discussed in the present paper.

Oxide targets are more favourable than carbides with respect to contamination of surfaces, but many of them sinter very strongly and are therefore unfavourable for the production of the most short-livet nuclei. Several oxides also give strong containment corrosion problems and are thus difficult to use. Particular thermal stability, even in contact with the Ta target cylinder, has been observed for ZrO_2, HfO_2 and ThO_2. Off-line tests of these compounds have been published previously [4]. The release of nuclear reaction products is less rapid than from carbides. Additionally, long-time use leads to slower release due to sintering.

The CaO, and probably also the other alkaline earth oxides, are too corrosive to be used as target materials at high temperature in a Ta cylinder, although we have found the release of a number of elements from CaO to be rapid. However, it is possible to protect the Ta cylinder from the material by means of a Re foil. Under these conditions, the alkaline earth oxides behave excellently. They hardly show sintering, even after long-time use at high temperature, and give rapid release of some reaction products. On-line tests of these materials will be described in ch. 3.

2.3 Borides

Generally, borides liberate nuclear reaction products slowly, and the most refractory borides are not suitable for target purposes. We found, however, that calcium boride (CaB_6) was sufficiently rapid, and an attempt was made to use it as target. Unfortunately, the amount of impurities was too high, causing malfunction of the ion source and a rapid breakdown of the system.

3. ON-LINE PERFORMANCE

The production of unstable nuclei from carbides was extensively discussed in the report given at the Zinal conference [10], and the data given there will only be occasionally discussed here.

Since then, carbides and oxides have shown their usefulness in the production of many more elements. In the following, each group of elements is discussed separately, and the measured production yields are given in terms of atoms per second per μA

of incident proton beam. The curves drawn through the measurement points are only for guiding the eye.

3.1 Calcium, barium and radium

All alkaline earth elements except Be and Mg can be efficiently ionized on a hot tungsten surface ionizer, and high temperature carbide targets should therefore be excellent production systems for short-lived isotopes of these elements. Off-line tests indicate a rapid release process [3,4].

In fig. 1, we show the production of Ra from a ThC_2 and a UC_2 target. The high yields obtained here have enabled optical spectroscopy on short-lived Ra isotopes [14].

A measurement of the production of neutron-rich Ca (table 1) and Ba isotopes (fig. 2) from a uranium carbide target has also been performed. The Sr production has not been investigated, but the yields from carbide targets are likely to be high also for this element.

3.2 Aluminium and magnesium

From a target of scandium carbide, combined with a W surface ionizer, it is possible to produce unstable isotopes of Al and Mg (fig. 3). In this case, the target was equipped with a leak of CF_4 which provides a pressure of about 2×10^{-4} torr inside the target cylinder. The effect of CF_4 in this case is not quite clear since the Al production from scandium carbide without CF_4 has not been investigated. A small yield of Mg is also observed for the first time from this system, but this element is not likely to be influenced by CF_4 addition, and is not very efficiently surface ionized.

3.3 Noble gases

The alkaline earth oxides (kept in a Re foil for protection) have been used recently for the production of noble gases in combination with a FEBIAD plasma ion source. The transfer line has been equipped with a water cooling in order to efficiently remove less volatile elements and ensure the chemical selectivity of the system.

The yields of the noble gases Ne, Ar, and Xe from targets of MgO, CaO, and BaO, respectively, are shown in figs. 4-6. Obviously, the production of Kr from SrO, a system which has not been tested, is feasible. The Ar yields from a vanadium carbide target are given for comparison in fig. 5, and are clearly inferior to those obtained from CaO.

Similar types of targets with ThO_2 or ThC_2 as target material have been used for the production of Rn and neutron-rich Kr and Xe and of Rn. These yields are shown in figs. 7-9. The two targets are equally good for the production of noble gases. The performance of these targets is illustrated by the discovery of two new Rn isotopes, ^{227}Rn ($T_{\frac{1}{2}}$ = 23 s) and ^{228}Rn ($T_{\frac{1}{2}}$ not yet measured).

3.4 Halogens

Since carbide targets have been shown to cause an unstable ion-source performance, and Th and U metals give containment problems, it has been difficult to find a satisfactory fission target material for use with the negative ionization method. The production of Cl, Br, I, and At from ThO_2 in combination with a negative ionizer is shown in figs. 10-13. So far, this target material is the best that could be found for this application, but the results obtained with it indicates that the sintering problems are severe [13].

It was shown by Carraz et al. [3] that barium zirconate ($BaZrO_3$) is suitable as target material with respect to release and thermochemical properties, and the production of neutron deficient iodine isotopes from this material is given in fig. 14. Although the delay in the target system is considerable (which can be seen from the relative yields of ^{115}I (1.5 m) and ^{116}I (2.2s)), this material is so far the best ISOLDE production target for neutron deficient I isotopes.

CONCLUSION

We have demonstrated that refractory carbides and oxides are useful production target for a number of elements. By means of appropriate shielding in the target container, it has been possible to apply several new materials in ISOLDE targets. In particular, the alkaline earth oxides have turned out to have a much better performance than previously expected.

Further use of oxides and carbides are planned in the near future. For instance, Ta_2O_5 is a potentially strong target material for the production of carbon as CO. When used in combination with a hot plasma ion-source, a number of other molecular oxide ions, e.g. SeO^+, are also likely to occur. Development work along these lines will also be pursued. The possibilities of carbides and borides are far from being exhausted, but further progress with these materials will necessitate the combination with a hot plasma ion-source. Tests involving the addition of reactive gases are planned.

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Table 1; Production yields of Ca isotopes from a uranium carbide target combined with a W surface ionizer

Mass	Yield (at/s)
49	$2.5 \cdot 10^5$
50	$2.4 \cdot 10^4$
52	$1.0 \cdot 10^2$

FIGURE CAPTIONS

Fig. 1. Production yields of Ra isotopes from 600 MeV proton-induced reactions in a ThC_2 and a UC_2 target.

Fig. 2. Production yields of Ba isotopes from 600 MeV proton-induced reactions in a UC_2 target.

Fig. 3. Production yields of Na, Mg and Al from 600 MeV proton-induced reactions in a ScC_2 target equipped with a calibrated leak of CF_4 providing a pressure of 2×10^{-4} torr in the target cylinder.

Fig. 4. Production yields from 600 MeV proton-induced reactions in a MgO target connected to a plasma ion-source via a water-cooled transfer-line.

Fig. 5. Production yields of Ar isotopes from 600 MeV proton-induced reactions in targets of CaO and VC connected to a plasma ion-source via a water-cooled transfer-line.

Fig. 6. Production yields of Xe isotopes from 600 MeV proton-induced reactions in a target of BaO.

Fig. 7. Production yields of Rn isotopes from 600 MeV proton-induced reactions in targets of ThO_2 and ThC_2 connected to a plasma ion-source via a water-cooled transfer-line.

Fig. 8. Production yields of Kr isotopes from 600 MeV proton-induced reactions in a target of ThC_2 connected to a plasma ion-source via a water-cooled transfer-line.

Fig. 9. Production yields of Xe isotopes from 600 MeV proton-induced reactions in targets of ThO_2 and ThC_2 connected to plasma ion-sources via water-cooled transfer-lines.

Fig. 10. Production yields of Cl isotopes from 600 MeV proton-induced reactions in a 40 g/cm^2 target of ThO_2 connected to a LaB_6 negative ionizer.

Fig. 11. Production yields of Br isotopes from 600 MeV proton-induced reactions in a 40 g/cm^2 target of ThO_2 connected to a LaB_6 negative ionizer.

Fig. 12. Production yields of I isotopes from 600 MeV proton-induced reactions in a 40 g/cm^2 target of ThO_2 connected to a LaB_6 negative ionizer.

Fig. 13. Production yields of At isotopes from 600 MeV proton-induced reactions in a 40 g/cm^2 target of ThO_2 connected to a LaB_6 negative ionizer.

Fig. 14. Production yields of I isotopes from 600 MeV proton-induced reactions in a target of $BaZrO_3$ connected to a LaB_6 negative surface ionizer.

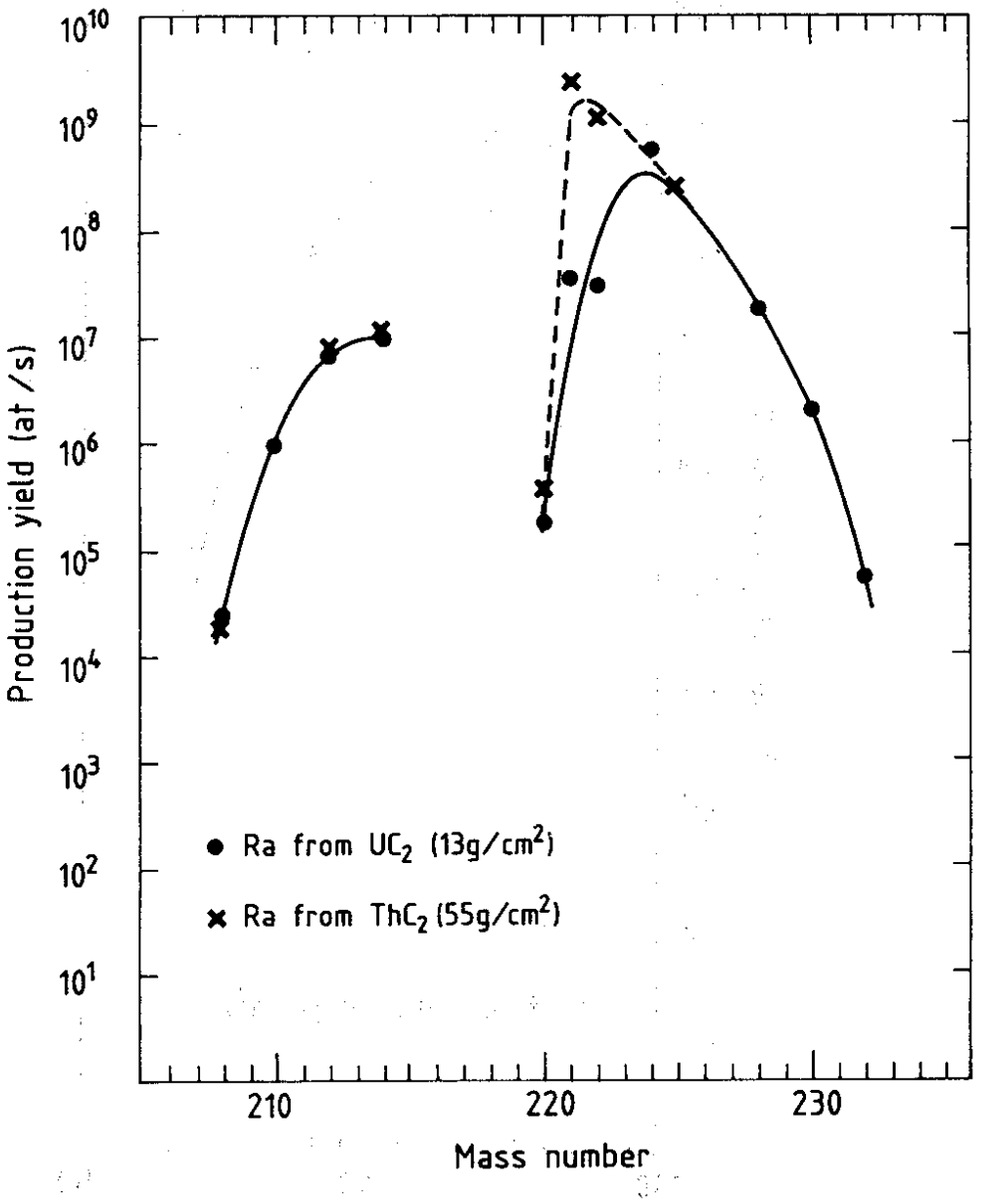
Production yield (at/s)
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
210
220
230
Mass number
● Ra from UC_2 (13g/cm^2)
× Ra from ThC_2 (55g/cm^2)

Fig. 1

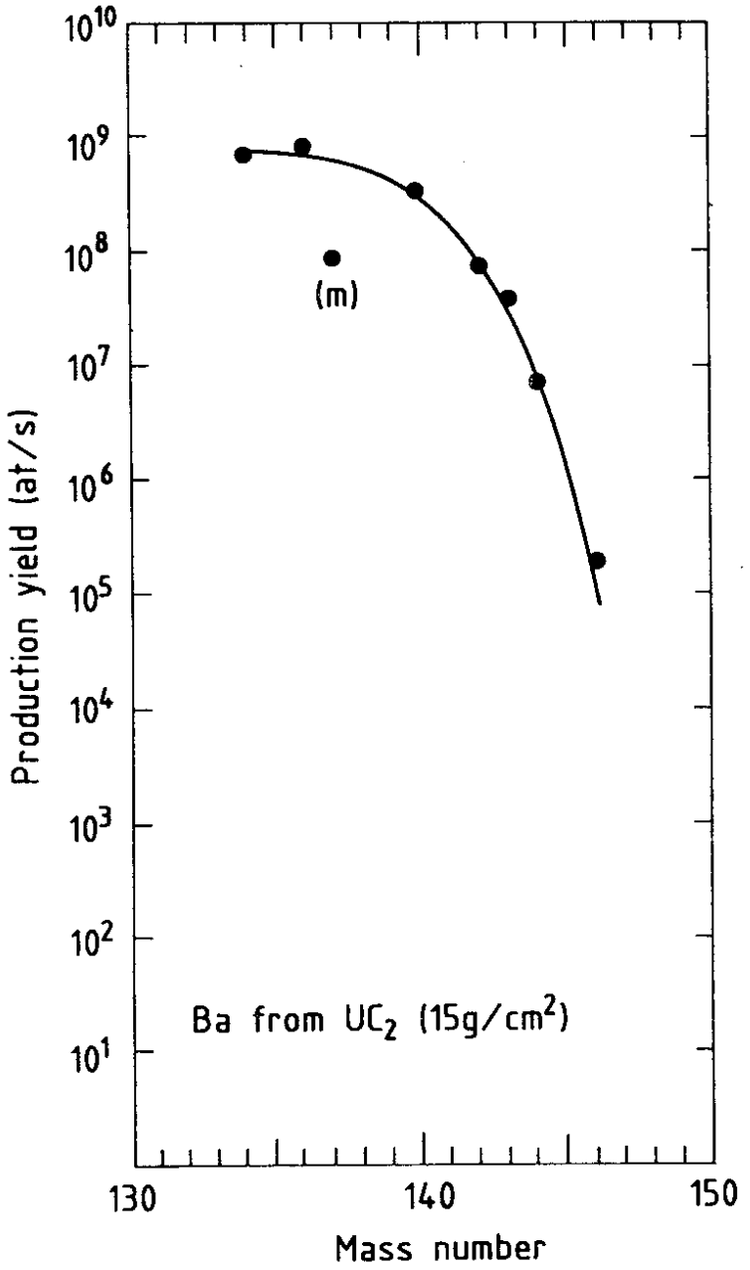
Production yield (at/s)
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
(m)
Ba from UC_2 (15g/cm^2)
130
140
150
Mass number

Fig. 2

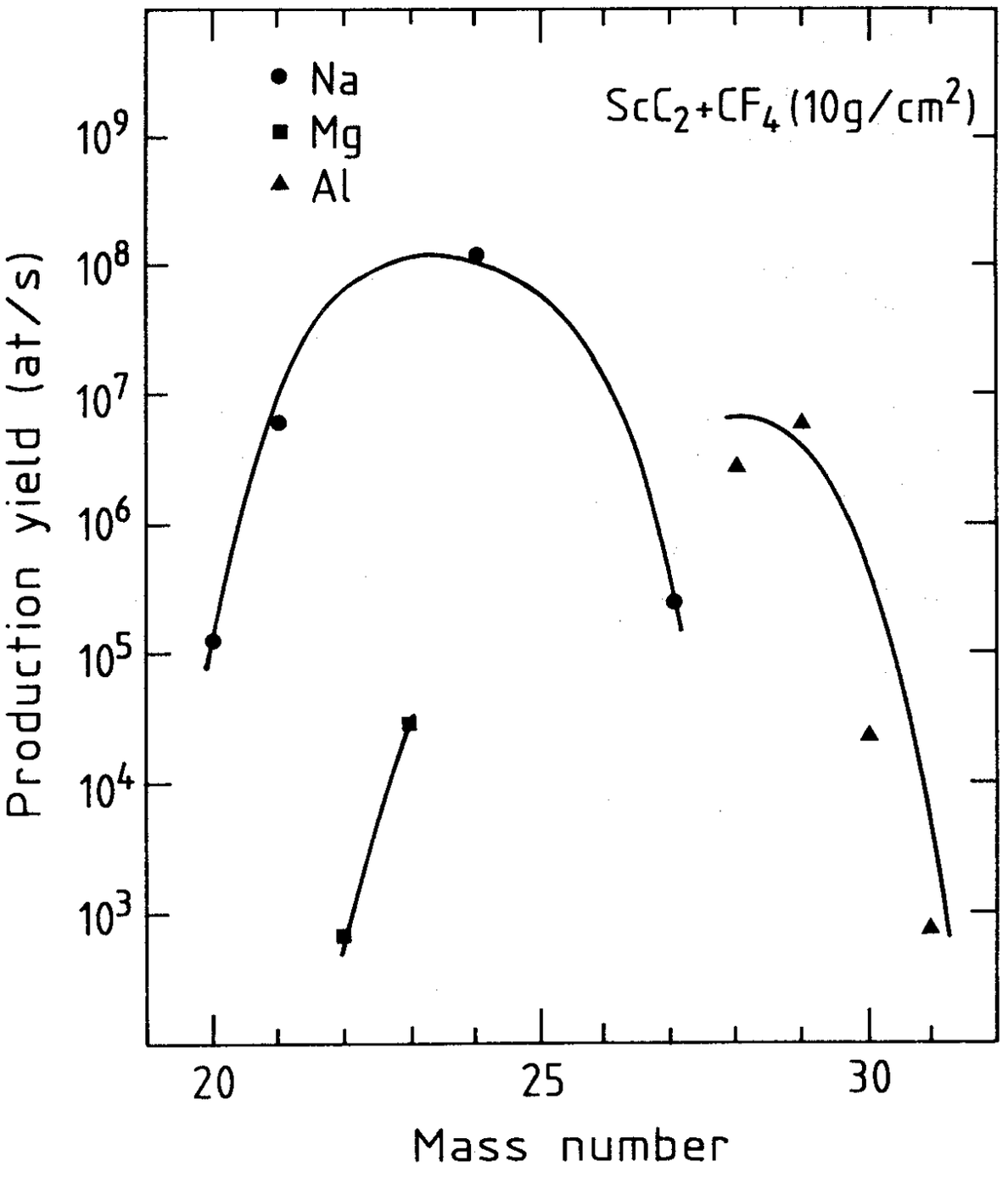
Na
Mg
Al
ScC_2+CF_4 (10g/cm^2)
Production yield (at/s)
10^9
10^8
10^7
10^6
10^5
10^4
10^3
20
25
30
Mass number

Fig. 3

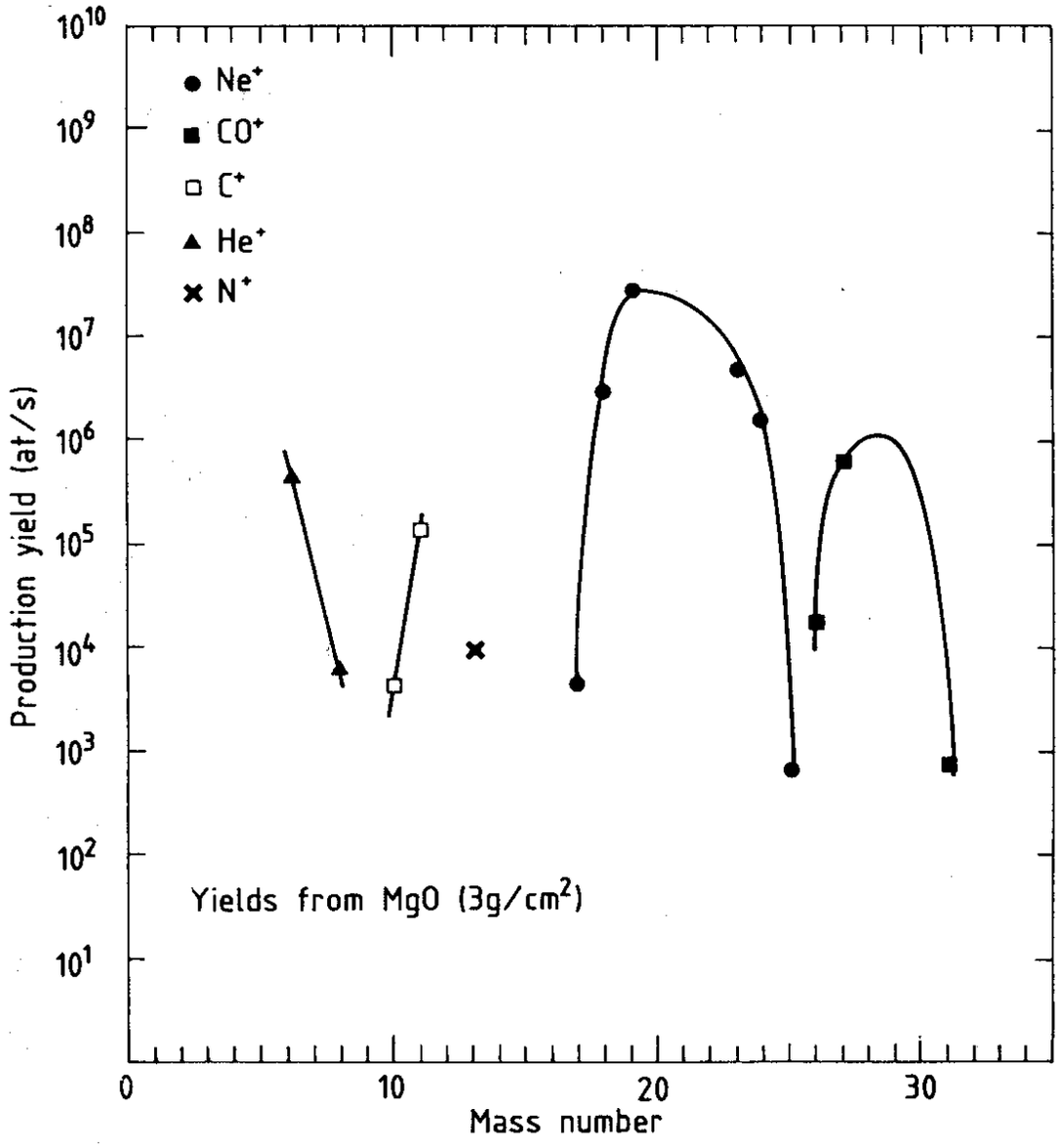
Ne+
CO+
C+
He+
N+
Production yield (at/s)
Mass number
Yields from MgO (3g/cm2)

Fig. 4

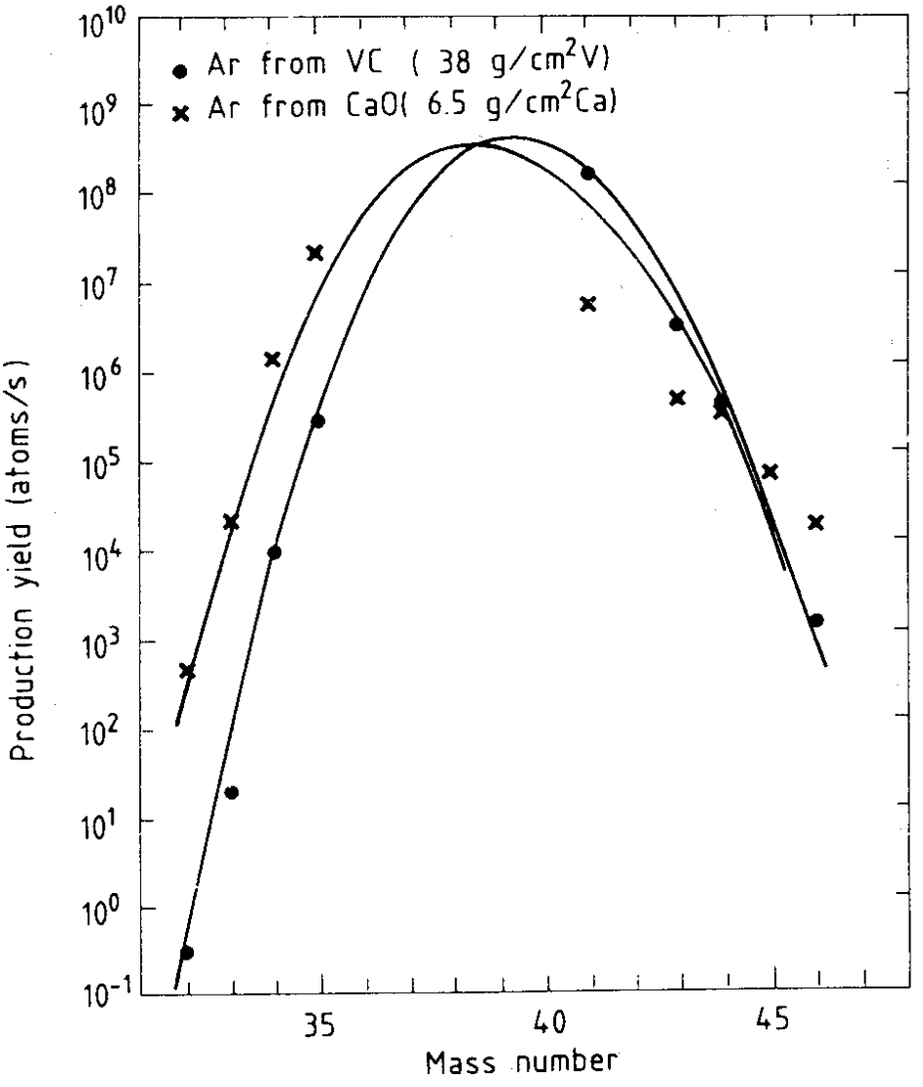
Ar from VC (38 g/cm²V)
Ar from CaO(6.5 g/cm²Ca)
Production yield (atoms/s)
Mass number
10¹⁰
10⁹
10⁸
10⁷
10⁶
10⁵
10⁴
10³
10²
10¹
10⁰
10⁻¹
35
40
45

Fig. 5

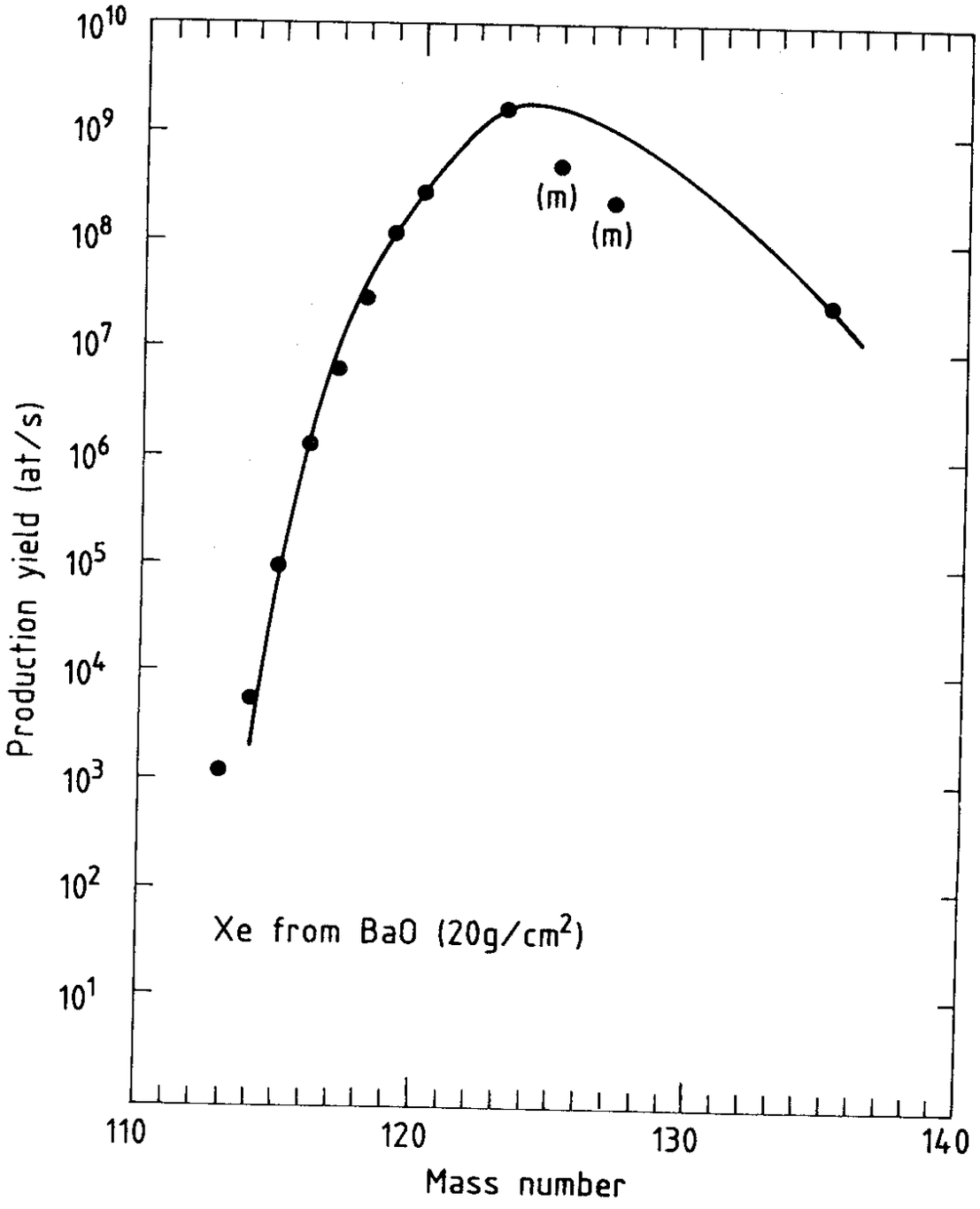
Production yield (at/s)
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
(m)
(m)
Xe from BaO (20g/cm^2)
110
120
130
140
Mass number

Fig. 6

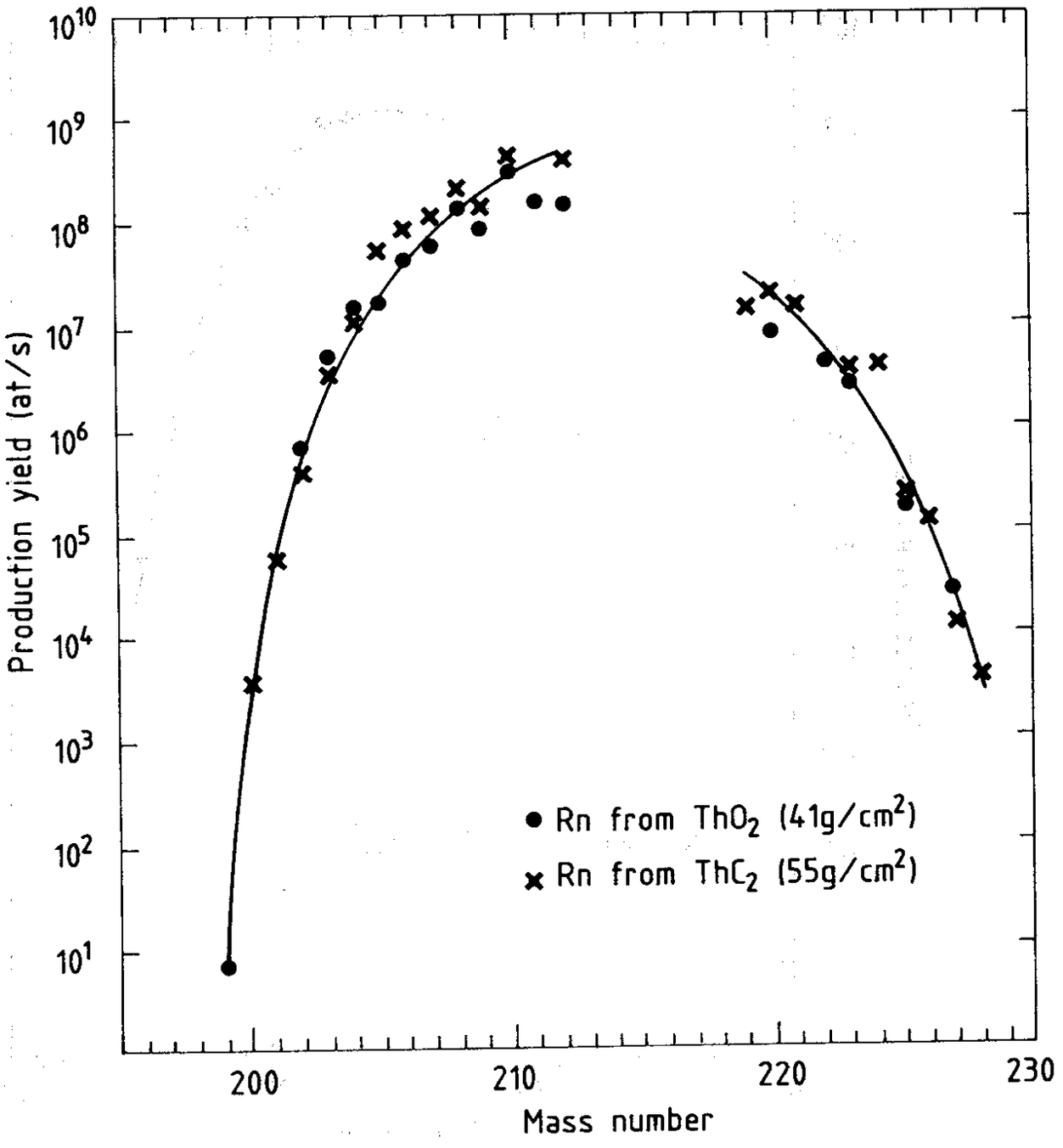
Production yield (at/s)
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
200
210
220
230
Mass number
● Rn from ThO_2 (41g/cm^2)
× Rn from ThC_2 (55g/cm^2)

Fig. 7

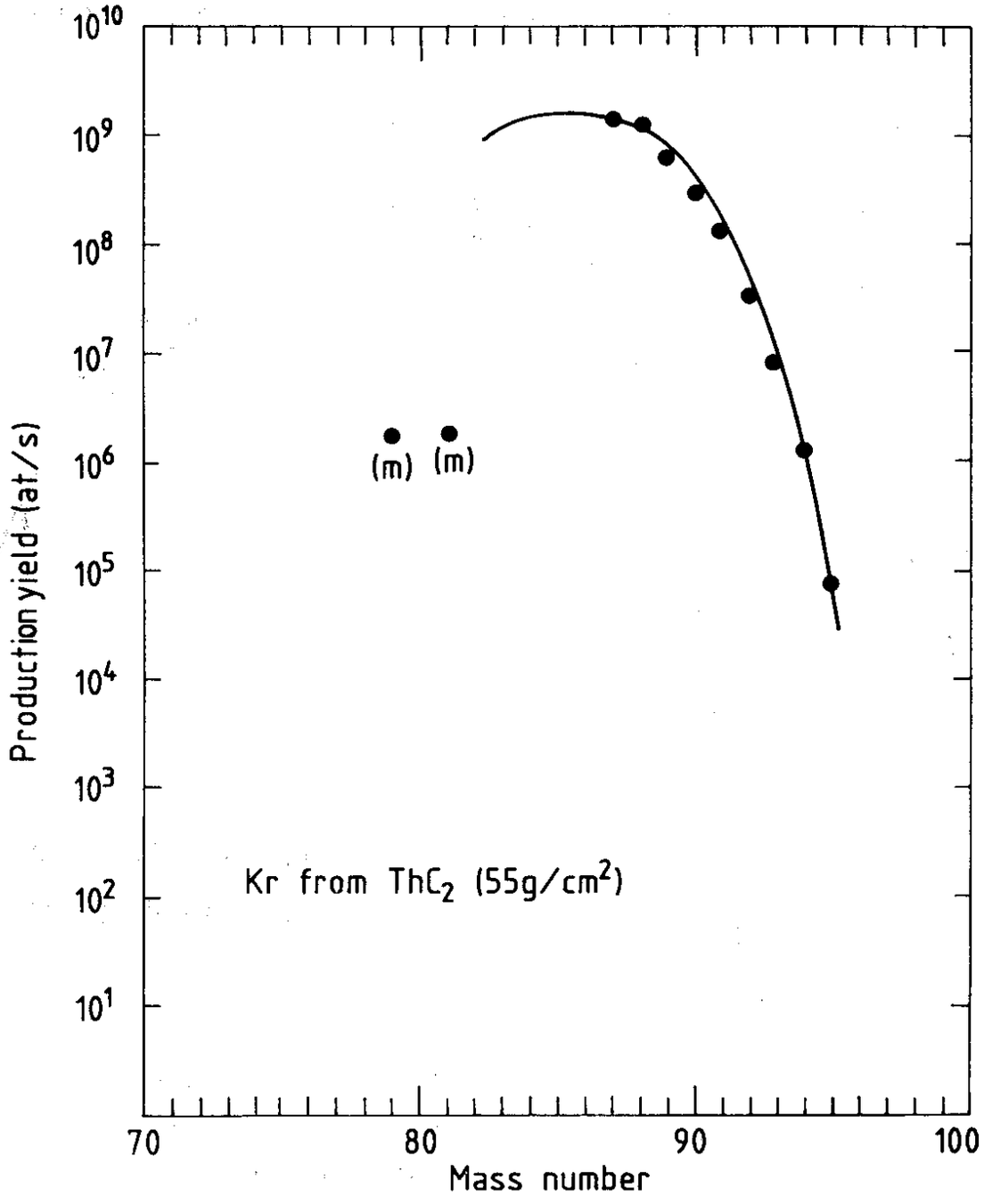
Production yield (at/s)
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
(m) (m)
Kr from ThC_2 (55g/cm^2)
70
80
90
100
Mass number

Fig.8

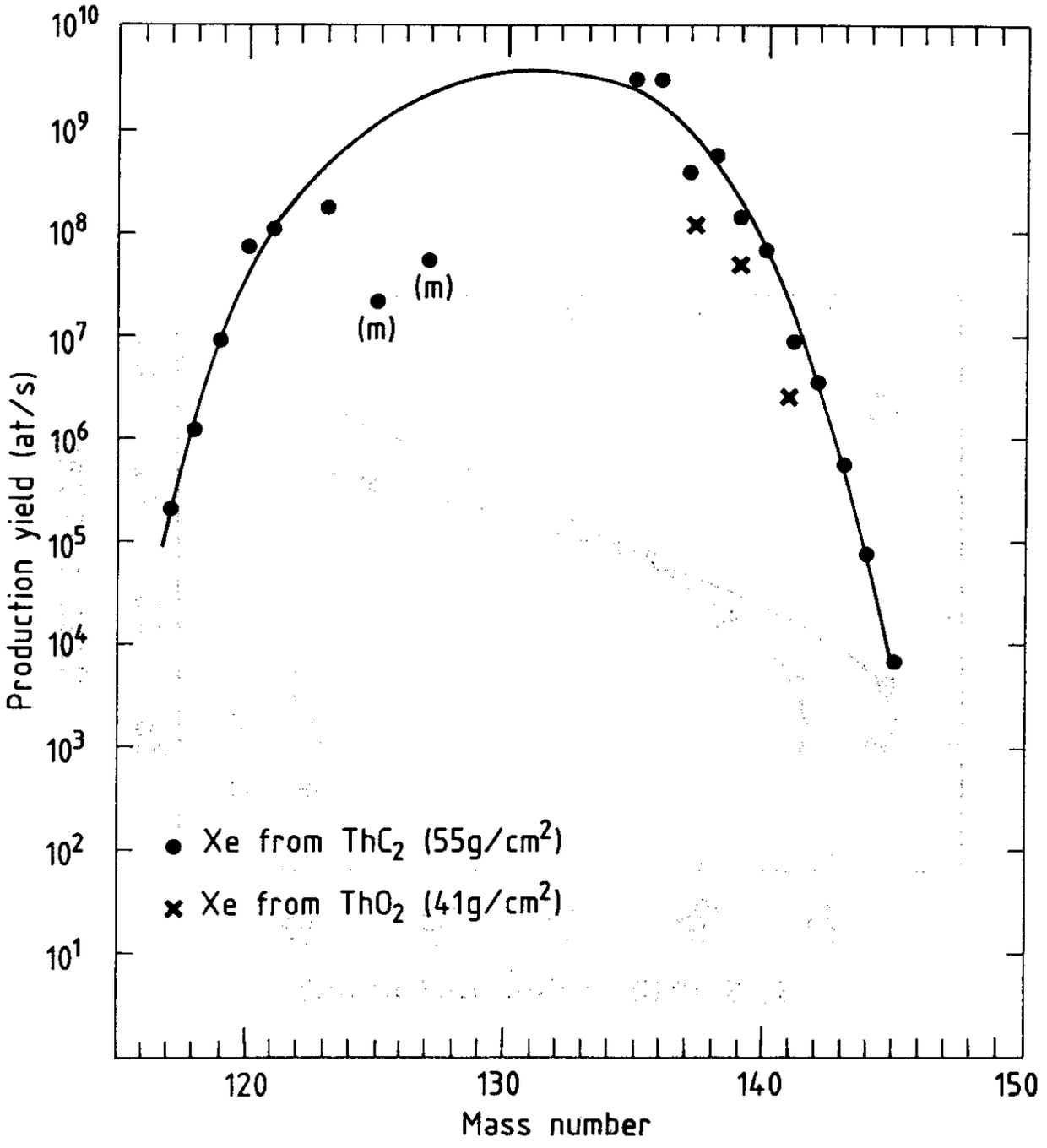
Production yield (at/s)
Mass number
(m)
(m)
Xe from ThC2 (55g/cm2)
Xe from ThO2 (41g/cm2)

Fig. 9

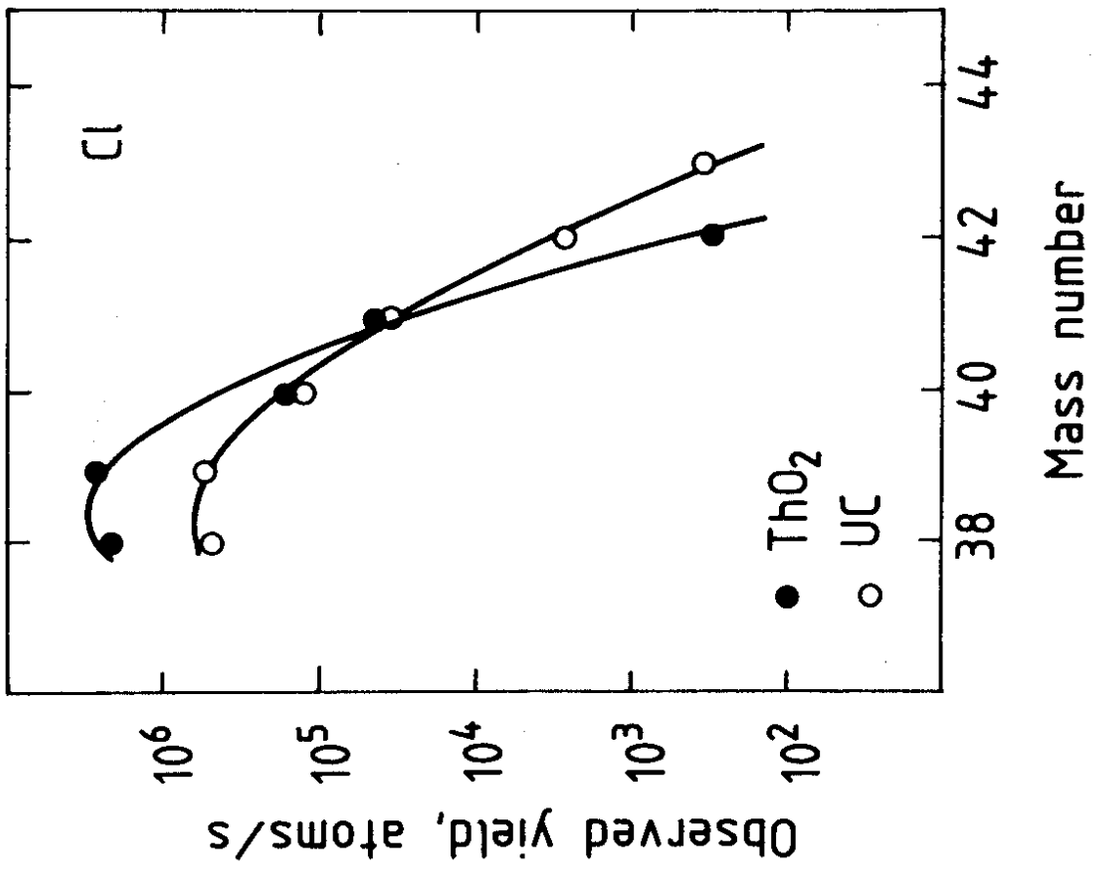
Cl
ThO2
UC
Observed yield, atoms/s
Mass number
10^6
10^5
10^4
10^3
10^2
38
40
42
44

Fig.10

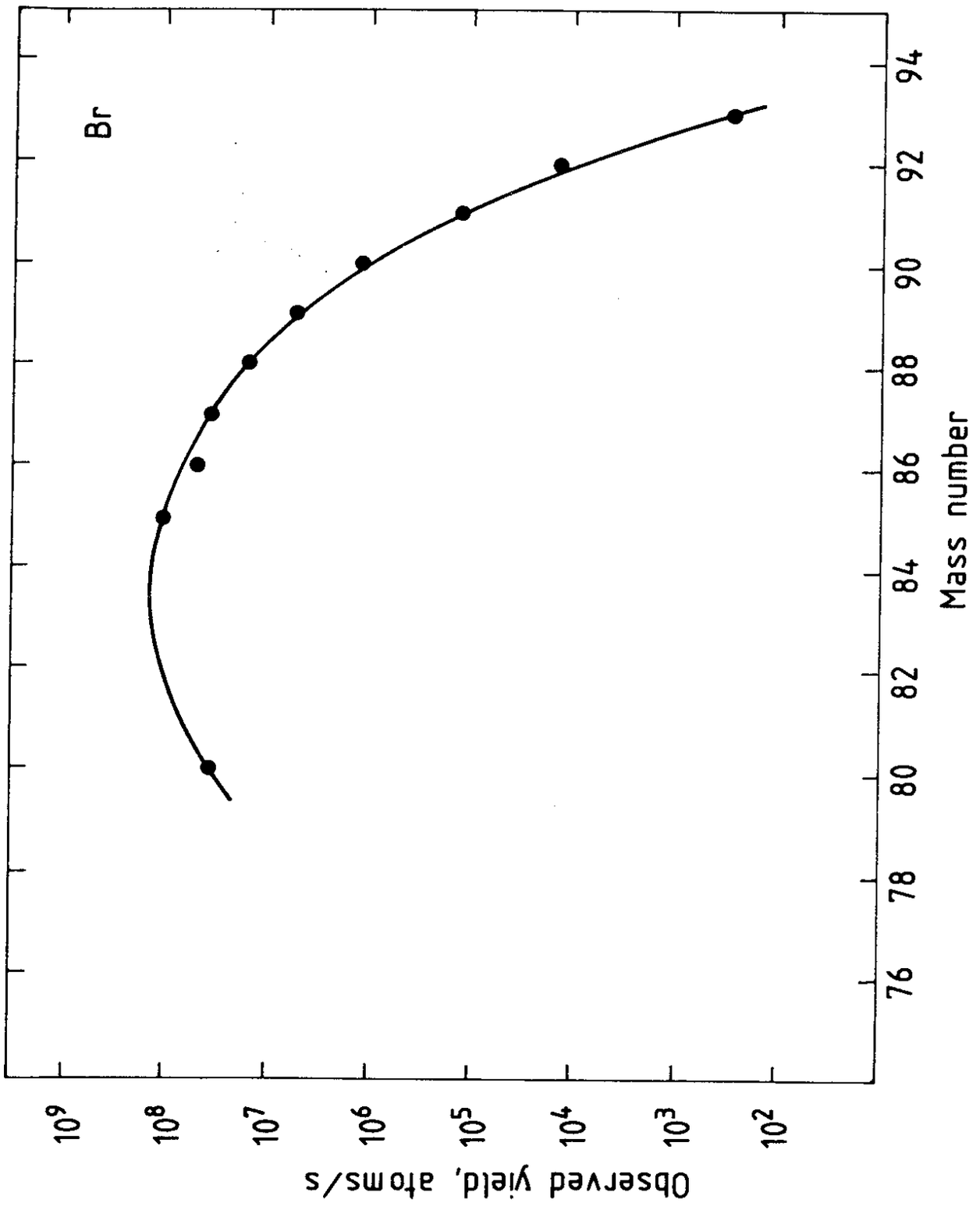
Br
Observed yield, atoms/s
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
76
78
80
82
84
86
88
90
92
94
Mass number

Fig.11

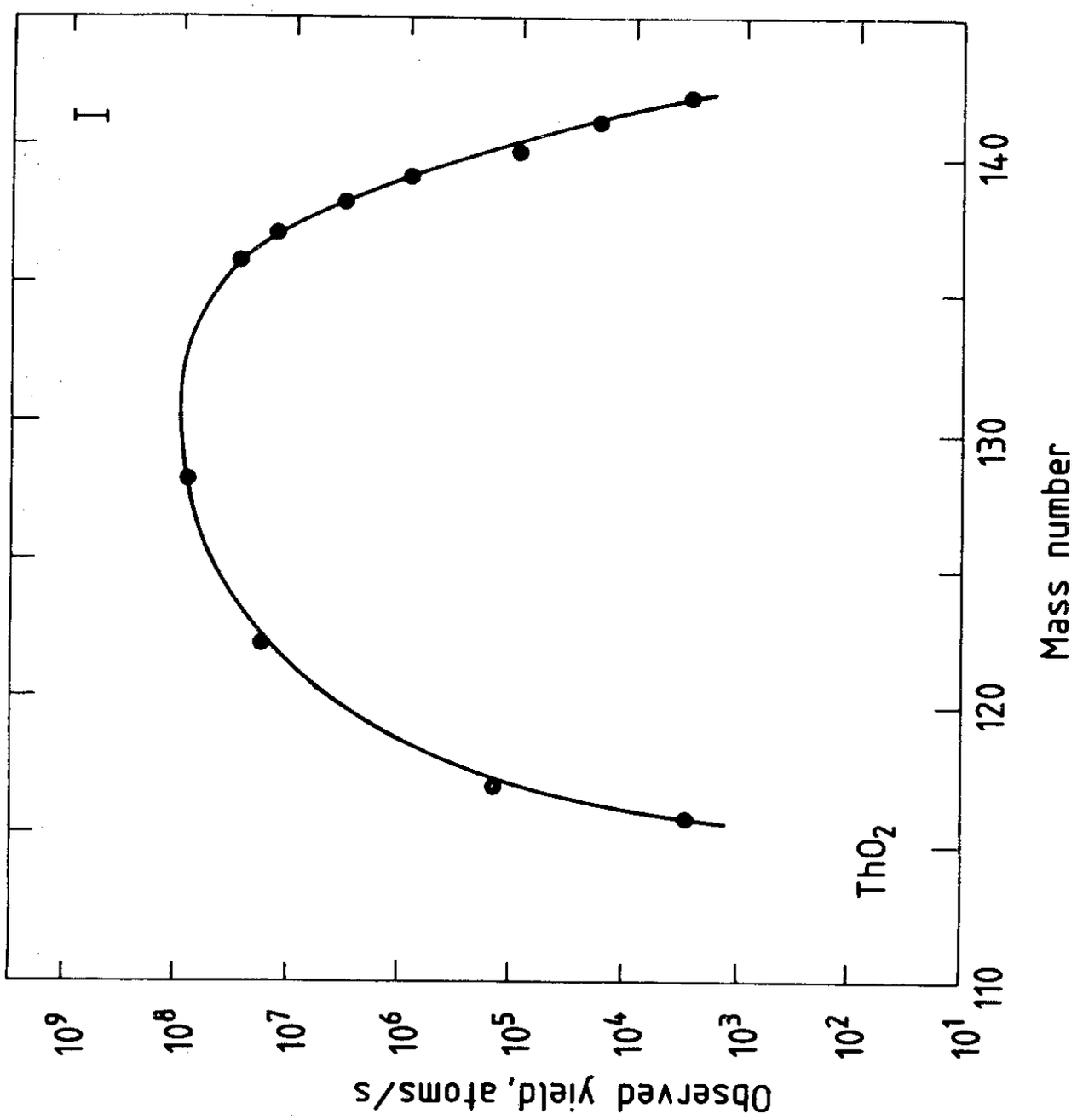
I
ThO_2
Observed yield, atoms/s
Mass number
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
110
120
130
140

Fig. 12

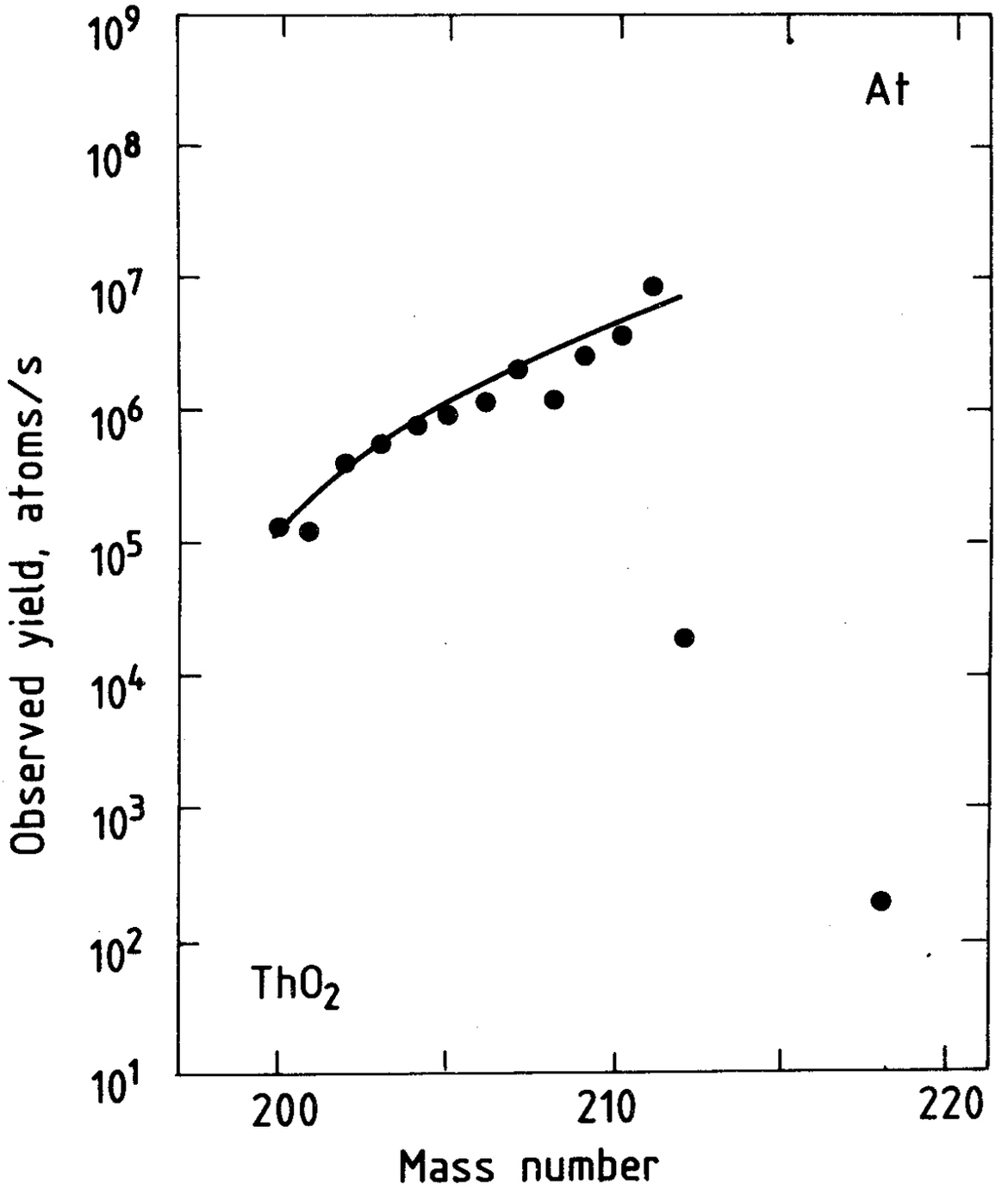
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
Observed yield, atoms/s
At
ThO_2
200
210
220
Mass number

Fig.13

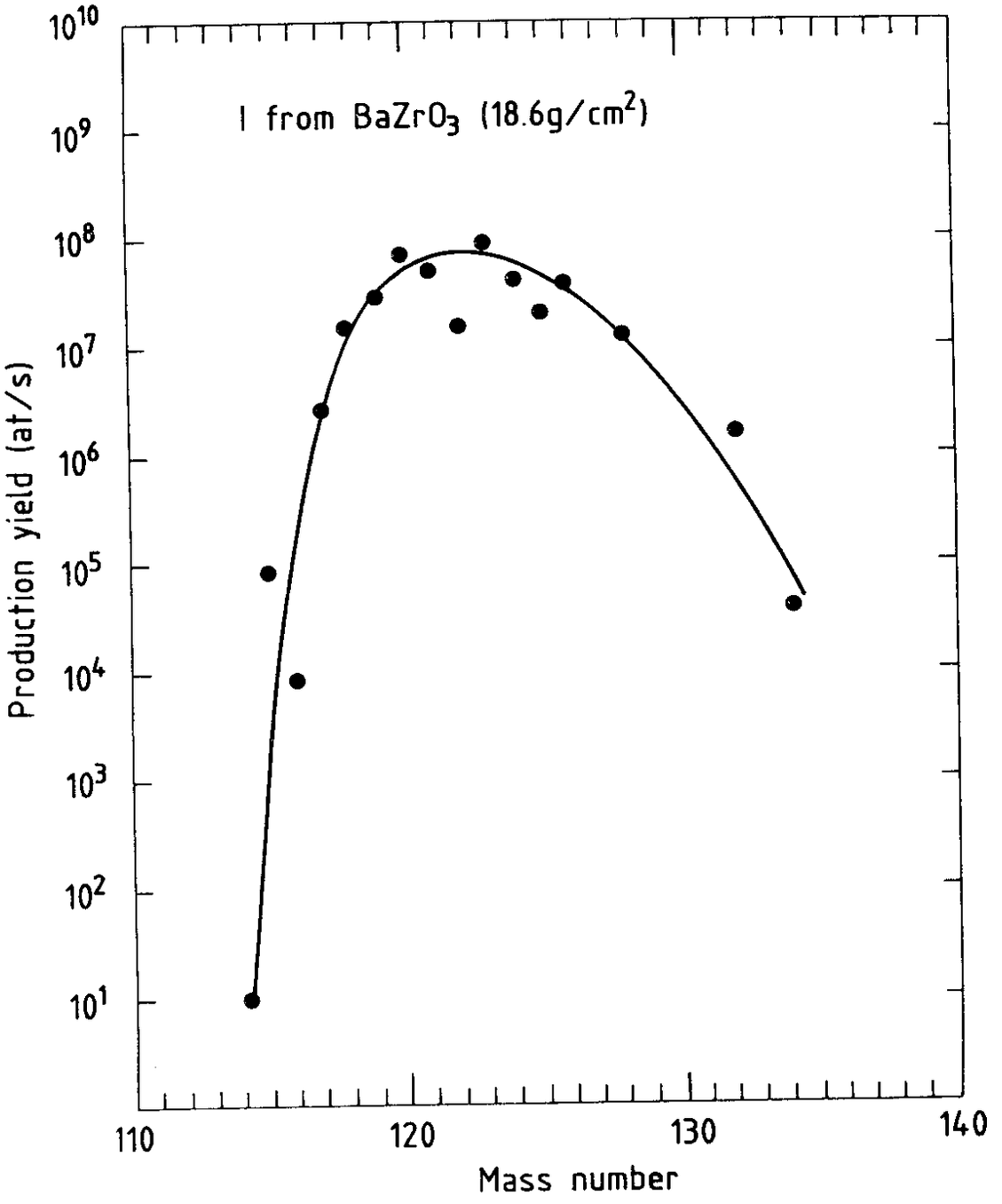
I from $BaZrO_3$ (18.6g/cm^2)
Production yield (at/s)
10^{10}
10^9
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
110
120
130
140
Mass number

Fig. 14