CERN LIBRARIES, GENEVA

CM-P00065831

CERN-PRE-78-077

HIGH-TEMPERATURE CARBIDE TARGETS FOR FAST ON-LINE MASS

SEPARATION OF ALKALI AND NOBLE GAS ELEMENTS

L.C. Carraz, S. Sundell and H.L. Ravn

CERN, Geneva, Switzerland

M. Skarestad and L. Westgaard

CERN, Geneva, Switzerland

and

Department of Chemistry, University of Oslo, Blindern, Norway

ABSTRACT

Targets of vanadium, lanthanum and uranium carbides kept at temperatures around 2000°C have been used at the ISOLDE on-line isotope separator at the CERN 600 MeV Synchro-cyclotron for production of Ar, Xe, Rb, Cs and Fr isotopes. The target materials are prepared either by sintering finely divided powders or by impregnation of graphite cloth with the respective target elements. Isotopic-production yields peak at 10^8 - 10^9 atoms/s, and the distribution of time delays between production and measurement extend well below 1 s. The measured delay is treated in the framework of simple diffusion theory and a relation between observed and decay-loss-corrected yields has been derived. Approximate formation cross-sections have been deduced and compared with empirical systematics. In the course of the development work the following new nuclides were identified: ^{32}Ar ($T_{1/2} \sim 75$ ms), ^{46}Ar (9 ± 1 s), ^{201}Fr (48 ± 15 ms), ^{202}Fr (0.35 ± 0.02 s) and ^{230}Fr (19 ± 1 s).

Geneva - 16 May 1978

(Submitted to Nuclear Instruments and Methods)

1. INTRODUCTION

The extensive use, in recent years, of on-line electromagnetic isotope separators for the study of new phenomena encountered in far unstable nuclei has required the development of new techniques, which allow a continuous release and subsequent ionization of nuclear reaction products formed in a target situated in a particle beam. These techniques depend strongly on the properties of the bombarding particles. For the ISOLDE separator on line with the CERN 600 MeV proton Synchro-cyclotron, target and ion-source arrangements providing isotopic beams of about 40 elements have been developed¹⁻⁸⁾.

The crucial condition for the application of these techniques to nuclides with half-lives in the subsecond region is a rapid escape of the products from the target and ion-source unit. Particularly fast targets are foils, powders or other forms of solids with large surface-to-volume ratio, kept at temperatures near their melting points. Diffusion to and evaporation from the surface, as well as any subsequent adsorption/desorption steps encountered in the confined space of the target container and ion source, are in general the rate-determining processes⁹⁾. Only few such targets have been used in the past since prediction of their properties, and therefore the design of appropriate systems, has been difficult owing to the lack of diffusion parameters and surface desorption energies for systems of interest for nuclear studies. The available data mainly referred to the release of fission products and actinides from different nuclear reactor fuel composites. This situation has been rapidly changing over recent years and a series of systematic studies⁹⁻¹⁷⁾ of the release properties of a variety of refractory metals and compounds have recently been published. Improved theoretical understanding of the transfer mechanisms and new experimental data have prepared a more solid ground for the choice of target matrix for the production of many elements. With techniques that allow target and ion-source temperatures in excess of 2000°C and the preparation of stable refractory matrices with a very short solid-state diffusion path, target systems with delay-time components in the vicinity of 1 s have been designed. On-line studies of extreme short-lived nuclei produced from U₂O₈-graphite composites^{18,19)} ThO₂²⁰⁾, CeO₂²⁰⁾, Nb^{8,21)}, and Ta⁸⁾ show the strength of this approach.

Uranium carbide was a natural choice as a target matrix exhibiting short delay-times, as it has long been known that a variety of fission-produced elements are more or less rapidly released from graphite matrices containing uranium carbide^{10,11)}. In the on-line experiments referred to above^{18,19)} where the uranium-oxide and graphite composites are operated in the temperature region 1300-1900°C, the target most likely consists of uranium carbide, since the conversion of oxide to carbide occurs at a temperature of 1500-2000°C²²⁾.

We report in this paper on the use of different carbide-containing matrices as targets for fast on-line mass separation and study of rare gases and alkali metals. In Section 2 the choice of the target matrices on the basis of thermodynamic data is discussed and some of the properties of the target materials are reported. In Section 3 we describe the experimental procedures. The on-line results in terms of production yields, delay-times, and cross-sections are discussed in Section 4. The range of short-lived nuclei which have been made accessible by this target concept is illustrated in Section 5, where half-lives of some new noble-gas and alkali isotopes are given. Finally in Section 6 it is pointed out how different refractory carbides open up the fast on-line mass separation of other elements.

2. CHOICE AND PROPERTIES OF TARGET COMPOUNDS

In an off-line study⁹⁾ of a series of refractory matrices, consisting of metals and chemical compounds, we have determined the release rate and efficiency for a variety of elements as well as the high-temperature stability of the matrices. It was generally found that the metal powders of the elements in group VB of the periodic table, and the carbides of U, Ce, La, Y, Sc and V, gave the fastest release. Furthermore it was shown, in agreement with the observations of Orth¹¹⁾, that the release rate increased with decreasing concentration of uranium in the graphite samples. This is most likely also the case for other solid solutions of metal carbides in graphite, since anomalously fast ion diffusion is generally observed in near- or non-stoichiometric compounds like glasses and graphite compounds²³⁾. On this basis, together with other relevant data^{24,25)} for refractory materials, we have chosen the target materials which are listed in Table 1. For the production

of Fr isotopes and neutron-rich isotopes of the other alkali elements a target of 10 atoms % uranium-impregnated graphite cloth [U(gr)] was used. The material was prepared in a way similar to that described in another paper²⁶⁾, with the difference that the oxide-covered graphite fibre matrix was heated several hours at 2000°C in order to fully convert the oxide into carbide and form a homogeneous solid solution of carbide in the fibres. The high-temperature stability of this matrix is excellent, and no visible change in the structure could be observed after 250 hours of in-beam service at a temperature of 2000°C. The maximum usable temperature quoted in Table 1 is determined by the melting point of the UC₂-C eutectic. A pure 50 μ UC + UC₂ powder, which because of its density allows a high production rate, albeit at the expense of a longer delay-time⁹⁾, was also investigated on line. It should be mentioned that these uranium-containing target materials are very inflammable and are easily ignited by shocks or when struck with metallic tools.

For the production of the neutron-deficient Xe isotopes a solid solution of LaC₂ in graphite fibres [La(gr)] is chosen from its chemical analogy with the UC₂ cloth. The maximum usable temperature quoted in Table 1 for this material is determined by the partial pressure of La over the matrix, which for a proper operation of the ion source should be kept below 10⁻³ Torr. A characteristic property of this material is that it hydrolyzes very quickly when exposed to moist air. Vanadium carbide powder was chosen as target for the production of the Ar isotopes owing to the short delay-times measured off line for this material. By analogy with the La and U targets, cloths impregnated with V or Sc most probably would constitute faster targets for Ar, but with a considerable loss in target thickness. The maximum usable temperature of VC is determined by the partial pressure of V. Both VC and the UC-UC₂ powder showed a considerable (30 vol %) shrinking due to sintering.

The only container material which can stand the prolonged contact at high temperature with the VC and UC target matrices is graphite, whereas LaC₂ can be contained in tantalum.

3. EXPERIMENTAL TECHNIQUES

3.1 The electromagnetic isotope separator

The new fast carbide targets were tested and used for production runs at the ISOLDE isotope separator facility on-line with the CERN 600 MeV Synchro-cyclotron. The proton beam intensity on the target was usually about 1 μ A, but beams up to 4 μ A will eventually become available. Further details about the separator and the experimental programme are given in other papers^{3-8,27)}.

3.2 Target and ion-source assembly

For the production of the alkali elements the new carbide materials, with or without an inner graphite crucible, simply replaced the molten metals used in the earlier described^{6,7)} target systems, in combination with a surface ionization source. A plasma ion source is required for the ionization of the noble gases. Such ion sources cannot discriminate against the other elements released simultaneously from the target. In order to restore the selectivity for noble gases a copper tube kept at a temperature of about 30°C is inserted in the line between the target and ion source (Fig. 1). All other elements are condensed or adsorbed in this tube and only the noble gases are transmitted. The plasma ion source also shown in Fig. 1 is a modified version of the FEBIAD ion source developed by Kirchner and Roeckl²⁸⁾.

The advantage of this source is its high stability and its capability of operation at very low pressures. In fact, we do not as usual add a supporting gas, since the small quantities of gas released from the cathode and the target suffice to sustain the plasma. Only occasionally is a noble gas from a calibrated gas leak (10^{-6} st. cm³/s) admitted to the source for efficiency measurements and mass calibration purposes. The measured ion-source efficiencies vary between 10% and 30% for the noble gases. The lifetime of the assembly is of the order of 200 h, in which period the Ta target container, which also serves as a heating element, is partially converted into very brittle TaC and eventually fails owing to thermal stress.

3.3 Detection methods

Intensities of the radioactive ion beams were in general measured with standard α , β , and γ spectroscopic equipment in combination with different kinds of extracted-beam-handling and source-collection systems, as described elsewhere⁷⁾. In addition, the high ion-beam intensities now encountered ($> 10^8 \text{ s}^{-1}$) allowed a direct measurement in a Faraday cup connected to a precision electrometer. The integral delay curves were obtained by sampling a radioactive beam at regular intervals after switching off the proton beam and, by means of a small tape transport system, bringing the sources one by one in front of a thin plastic scintillator for beta counting.

4. RESULTS AND DISCUSSION

4.1 Measured yields

Observed yields of Ar, Xe, Rb, Cs, and Fr isotopes, given as atoms per second measured at the collector side of the isotope separator, are summarized in Figs. 2-6. Yields of some additional nuclides are included in Table 2. The yield curves are found to peak at 10^8 - 10^9 atoms/s, assuming a proton beam intensity around 1 μA . In the cases of Fr and Ar the peaks are situated well to the neutron-deficient side of beta stability in accordance with a spallation production mechanism, while the isotopic yield distribution of the fission products Cs and Rb peaks close to the beta stability.

Yield measurements of Cs isotopes, released from 80 g/cm² uranium carbide powder target, have also been carried out. This target which is ~ 6 times thicker than the corresponding uranium-impregnated graphite cloth target gave as expected higher yields of long-lived isotopes, but the corrosion of the target container which developed after 15-20 hours at 1500-2000°C strongly limited the target lifetime.

The yields given have not been corrected for variation in the over-all system efficiency. This effect may account for a large part of the scatter in the points.

4.2 Delay-times

Experimental delay data are shown for the alkali elements Na, Rb, Cs, Fr, and the noble gases Ar, Xe in Figs. 7-9. In general we observe fast release of the products at target temperatures around 2000°C and qualitatively it is seen that nuclides having half-lives above $\frac{1}{2}$ min will not be affected much by decay losses in the transport process. With reference to the more short-lived nuclides, however, we present here an analysis of the delay data based on the formalism used in Refs. 5-8. A delay function $p(\tau)$ describes the distribution of delay-times. The form of this function depends mainly on the release mechanism(s). By comparing the present delay measurements with off-line measurements⁹⁾ and with results from OSIRIS^{26,29)}, it seems reasonable to assume that solid diffusion is the rate-determining step in the release process for those products that have been measured. According to Ref. 8 we then have

$$p(\tau) = \frac{6\mu}{\pi^2} \sum_{n=1}^{\infty} e^{-n^2\mu\tau}, \quad (1)$$

where $\mu = \pi^2 D/r^2$ is the diffusion parameter, D the diffusion coefficient, and r the radius of the microparticles. The measured count rate per sample of a product with half-life $T_{1/2} (= \ln 2/\lambda)$, as a function of the time t after switching off the proton beam, is then (assuming no parent effects)

$$A(t) = \mu\lambda R \Delta t \int_t^{\infty} e^{-\lambda\tau} p(\tau) d\tau = \mu\lambda R \Delta t \frac{6\mu}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{\lambda + n^2\mu} e^{-t(\lambda+n^2\mu)}, \quad (2)$$

where Δt ($\ll T_{1/2}$) is the sampling time, R the production rate, and μ the over-all system efficiency. If $\mu \gg \lambda$, an assumption which is fulfilled in most of the present cases, the following simplified formula may be used

$$F(t) = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-n^2\mu t}, \quad (3)$$

where $F(t)$ now represents the "fractional activity" defined as the activity per sample after beam-off, relative to the equilibrium activity measured while beam-on.

The diffusion parameter μ is determined by fitting Eqs. (2) or (3) to measured delay points either graphically⁹⁾ or numerically, using, for example, the minimization program MINUIT³⁰⁾. In Table 3 are given the μ -values determined by the delay measurements shown in Figs. 7-9. If we assume the relation

$$\mu = \mu^0 e^{-Q/RT}, \quad (4)$$

where R is the gas constant and T the absolute temperature, it is possible to calculate the activation energy Q for the release process in cases where measurements have been performed at different temperatures. This has been done for Cs, Rb, and Ar, as demonstrated in Figs. 10 and 11. The calculated activation energies are typical for diffusion processes in solids.

Having derived a representative μ -value for a particular product element released from a given target, it is possible to evaluate the release yields^{*)} for different isotopes of that element. The release yield, Y_{obs}/Y , is given by the ratio $A(t)/\mu\lambda R\Delta t$ when $t = 0$ in Eq. (2)

$$\begin{aligned} Y_{\text{obs}}/Y &= \frac{6}{\pi^2} \mu \sum_{n=1}^{\infty} \frac{1}{\lambda + n^2 \mu} \\ &= 3(W \coth W - 1)/W^2 \quad \text{where } W = \pi\sqrt{\lambda/\mu} \\ &= \frac{3}{\pi} \sqrt{\mu/\lambda} \quad \text{when } \lambda > \mu. \end{aligned} \quad (5)$$

where λ is the decay constant of the isotope.

Although the solid diffusion presumably is the time-controlling release mechanism, a contribution to the delay may occur owing to diffusion of the product from the particle surface to the outlet of the ion source. Thus for very short-lived nuclei the following corrected form of Eq. (5) has been suggested^{8,26)}

*) The release yield of a nuclide is defined as the ratio between the observed yield of the nuclide and that which would be obtained if there was no delay in the separation system. This ratio is referred to, by some authors, as the transmission factor or the reduction factor.

$$\frac{Y_{\text{obs}}}{Y} = \left(\frac{3}{\pi} \sqrt{\mu/\lambda} \right) \left(\frac{\nu}{\nu + \lambda} \right) \quad (6)$$

where ν ($\gg \mu$) is a constant characteristic for the effusion of the product out of the target container. The parameter ν can be estimated by means of the following equation^{2,9)}

$$\nu = \frac{A_s}{V} \sqrt{\frac{RT}{2\pi M}}, \quad (7)$$

where A_s is the outlet area of the ion source, V is the volume of the target container, M the atomic weight of the product, R the gas constant and T the absolute temperature^{*}). For the present target arrangement Eq. (7) gives $\nu = 23 \text{ s}^{-1}$ for $M = 100$ and temperature 2050°C , which implies that the correction term in Eq. (6) should become important for nuclides with half-lives below $\sim 0.06 \text{ s}$. The decay-loss corrections quoted in Figs. 2-6 have been carried out according to Eq. (6). It is then assumed that the alkali elements Rb, Cs and the noble gases Ar, Xe effuse at least as fast as Fr.

It should be noted that, in the above procedure for calculations of release yields, the target matrix has been taken to consist of spherical microparticles of uniform size. In the case of vanadium carbide powder this assumption may be justified, although a certain distribution in the particle sizes is known to exist⁹⁾. In the cases of impregnated graphite cloth, a geometrical model of graphite fibres of cylindrical cross-section seems more appropriate. This choice leads, however, to calculated release yields that differ only slightly (less than 10%) from those based on the spherical model. Therefore, the delay function given in Eq. (1) has been assumed for all targets.

The major source of error in calculated release yields is due to non-uniformity in the porous target structure and to possible mixtures of several diffusion components. Such effects may, for example, explain the apparent slow release component(s) in many of the delay curves, not properly accounted for by Eq. (1). For this reason,

^{*}) Since Eq. (7) does not take into account the conductance of the complex structure of the target matrix, the containers, and the interconnecting tube, it only gives an upper limit.

the yields given by Eq. (5) should be taken as upper limits. From off-line experiments⁹⁾, it is estimated that the real yields could be 20-30% lower for products in the half-life region 0.1 s to 1 min. For products of shorter half-lives (down to ~ 10 ms) the factor may increase to about two, as indicated by measurements on short-lived Fr isotopes³¹⁾.

The delay properties of different targets can be compared on the basis of Eq. (5), and in Fig. 12 this is done for Cs released from uranium targets of different compositions. It is evident that the uranium-impregnated graphite cloth target exhibits the most favourable delay properties.

4.3 Cross-sections

A detailed discussion of the spallation and fission reactions, in terms of isotopic mass yields, charge dispersions, and charge distributions, is outside the scope of this article. Still, it may be of interest to deduce approximate formation cross-sections from the observed yields, in particular towards the extreme ends of the yield curves, as such information is not readily obtained by conventional, more precise, methods. Although individual points may be uncertain, with perhaps as much as a factor of two due to over-all efficiency variations (Section 4.1), such errors should average out to give correctly the distribution over an extended mass region.

Since our experiments have not been optimized for cross-section measurements, a number of interfering effects have to be considered⁷⁾. Absolute reaction rates are difficult to measure in on-line experiments like those performed at ISOLDE, and the yields normally have to be related to selected cross-sections³²⁻³⁴⁾ determined by other methods. The observed yields of short-lived products have been corrected for decay loss in the target according to Eq. (5), with uncertainties depending upon half-lives as estimated in conjunction with the discussion of release yields (Section 4.2).

The formation cross-sections are in principle cumulative, except where the product is shielded by stable nuclides. However, for nuclides on the wings of the isotopic yield distribution the parent contribution is unimportant, as the yields

of precursors become small, and -- in the case of Rb and Cs -- also closer to beta stability the yields can be taken to be largely primary, because the isotopes of neighbouring elements are released from the target with delay times that are short⁹⁾ compared with the half-lives for decay. For Ar, Xe, and Fr isotopes with $A < 222$, near the top of the yield distribution, yields are mainly cumulative, since the precursors stay in the target or the cooled line.

Although our targets are not thick in terms of energy spread of the primary protons ($\Delta E < 5\%$), secondary reactions induced by cascade and evaporation particles are clearly important. Looking first at the spallation distributions, we may safely expect -- since relatively few of the cascade particles have energies approaching the full energy of 600 MeV -- that the neutron-deficient wings of the Ar, Xe, and Fr yield curves in Figs. 2, 3 and 6 are not significantly affected by secondary reactions. Also, the nuclides ^{37}Ar , ^{120}Xe , and ^{212}Fr , used for normalization to thin-target cross-sections, are sufficiently far removed from the respective targets that their production in secondary reactions, and thus the normalization error, should be rather small⁷⁾. The main source of error in the left part of the spallation-yield curves -- which are the most interesting -- therefore lies in the decay-loss correction discussed above. The experimental distributions are compared in Figs. 2, 3 and 6 with calculations according to the empirical systematics of Rudstam³⁵⁾.

In the fission of uranium, thick-target effects become more important. Because of the low fission barrier both cascade and evaporation particles may induce fission and contribute to the yields of neutron-rich Rb and Cs isotopes, thus lifting the observed isotopic yield curves above the ideal thin-target cross-section distribution. Fortunately, the normalization to thin-target data can be made with the nuclides ^{86}Rb and ^{132}Cs , for which the production cross-sections go rapidly down below 100 MeV^{34,36)}. Consequently, evaporation particles ($E < 20$ MeV) do not contribute and in an estimate similar to that of Hagebø and Lund for Pb fission³⁷⁾, we arrive at a conservative upper limit of 25% contribution from secondaries. This is then the normalization error in the effective fission-yield curves for Rb and Cs

(Figs. 4 and 5, right-hand scale). Also shown in the figures are calculated yield distributions based on charge-dispersion and mass-distribution studies of fission of uranium with 570 and 450 MeV protons^{38,39}.

5. NEW ISOTOPES

In the course of and as a first result of the development work described in the present paper, five hitherto unobserved nuclides were identified and their half-lives measured: ^{32}Ar (~ 75 ms), ^{46}Ar (9 ± 1 s), ^{201}Fr (48 ± 15 ms), ^{202}Fr (0.35 ± 0.02 s), and ^{230}Fr (19 ± 1 s).

The $T_Z = -2$ nuclide ^{32}Ar was identified⁴⁰⁾ through its beta-delayed proton decay. The proton spectrum (Fig. 13) consists of a single line, which is interpreted as being due to the proton decay of the 0^+ ($T = 2$) analogue state in ^{32}Cl . The observed half-life for ^{32}Ar indicates a branching ratio of $\sim 17\%$ for the super-allowed $0^+ \rightarrow 0^+$ ($T = 2$) beta transition.

The observed half-life of ^{46}Ar , together with preliminary gamma-spectroscopic data⁴¹⁾, is consistent with a structure of ^{46}Ar similar to that of ^{44}Ar , with most of its beta strength concentrated on a single transition ($\log ft \sim 4$) to a high-lying 1^+ level in the K daughter.

The nuclides ^{201}Fr and ^{202}Fr were identified³¹⁾ through their predominant alpha decay. A further study of the alpha energies and intensities of the parents and daughters from which the alpha branching ratios of the daughters have been deduced is published in Ref. 31.

The half-life of ^{230}Fr , unambiguously determined⁴²⁾ by following the decay of the X-rays emitted by the Ra daughter (Fig. 14), turns out to be somewhat shorter than the 30 s predicted⁴³⁾ by the gross beta theory. The experiments on Fr also gave an improved value for the half-life of ^{229}Fr ⁷⁾: $T_{1/2} = 48 \pm 2$ s.

If we turn now to the prospects of extending the methods described here to produce nuclei even further away from stability, we note first the very steep slope of the Ar, Xe and Fr cross-section curves (Figs. 2, 3, and 6) towards the neutron-deficient side, obviously reflecting the rapid fall in proton binding energy. The

1975 mass-excess predictions⁴⁴⁾ fix the proton drip line at ^{29}Ar , while ^{31}Ar is predicted to be particle unstable against two-proton decay by a popular mass formula (Jänecke-Garvey-Kelson). In the Fr case, four out of eight mass calculations agree to give ^{200}Fr proton unstable. Thus, even under experimental conditions which allow measurement of nuclides with half-lives in the ms range, we can hardly expect to reach further than at most one unit in mass number to the neutron-deficient side.

In the Xe case, the proton drip line is further away, around ^{107}Xe ⁴⁴⁾, and production of at least ^{112}Xe and ^{111}Xe seems to be within reach. (They are, however, predicted to be two-proton unstable by the Beiner-Lombard-Mas mass calculation.) The beta half-lives of these nuclides are estimated from strength-function systematics to be 1 s and 0.1-1 s, respectively. It should be remarked here that in the on-line isotope separator at the GSI heavy-ion accelerator the neutron-deficient Xe isotopes (e.g. ^{114}Xe) are produced⁴⁵⁾ in yields comparable to those obtained at ISOLDE. Towards the neutron-rich side, neither cross-section nor half-lives should prevent a further extension of the yield curves for spallation-produced Fr isotopes. In the Ar case, however, ^{46}Ar is already the heaviest isotope that can be produced in a primary reaction from the vanadium target. The production of Rb and Cs fission products (Figs. 4 and 5) has allowed on-line mass measurements⁴⁶⁾ to be pushed out to the last known isotopes (^{99}Rb and ^{147}Cs); and although the cross-sections are drastically falling and half-lives go down into the 100 ms region, the neutron drip line is still far away, and with sensitive detection methods it should be possible here to pursue experiments still further removed from stability.

6. PERSPECTIVES

The new targets have immediately been taken into use for investigations of the exotic nuclei that have now become available for detailed experiments. High-energy proton reactions are generally considered to be a powerful method for the production of neutron-deficient nuclides through the spallation reaction. In certain cases it has also been used for the production of neutron-rich nuclides, as for example the heavy Ra, Fr, Rn and Ar isotopes, where no other practical method

of production exists. In addition it is worth while again to emphasize the high yields of neutron-rich fission product nuclei. Programmes for extending the measurements of nuclidic masses⁴⁶⁾, spins, and moments to neutron-rich nuclei⁴⁷⁾, as well as delayed-neutron spectroscopy, have already started⁴⁸⁾.

Also, the success of the UC, LaC₂ and VC targets points the way to analogous systems giving other product elements both in connection with the proton beam and the soon available ³He beam. Other possible compounds are given in Section 2, and will be considered for on-line use. In addition, new ion-source developments are expected to widen the range of possible product elements from each target (see Ref. 9). It is already now certain that the refractory carbide targets represent a valuable addition to our arsenal of on-line targets and will help us to probe still further into the properties of far unstable nuclei.

Acknowledgements

It is a pleasure to acknowledge the important contributions the many members of the ISOLDE Collaboration have made to various phases of these experiments. The contributions of O.C. Jonsson, who prepared the target materials and assisted before and during the experiments, are gratefully acknowledged.

Two of us (M.S. and L.W.) are indebted to the Norwegian Research Council for Science and the Humanities for financial support.

REFERENCES

- 1) E. Hagebø, Proc. Internat. Conf. on Electromagnetic Isotope Separators and the Techniques of their Applications, Marburg, 1970, Report BMBW-FB K 70-28 (1970) p. 146.
- 2) P. Patzelt, *ibid.* p. 158.
- 3) S. Sundell, P.G. Hansen, B. Jonson, E. Kugler, H.L. Ravn and L. Westgaard, Proc. 8th Internat. EMIS Conf. on Low-Energy Ion Accelerators and Mass Separators, Skövde, 1973 (eds. G. Andersson and G. Holmén) (Chalmers Univ. of Technology and Univ. of Göteborg, Göteborg, Sweden, 1973) p. 102.
- 4) E. Kugler, *ibid.* p. 382.
- 5) H.L. Ravn, S. Sundell and L. Westgaard, *ibid.* p. 432.
- 6) H.L. Ravn, S. Sundell and L. Westgaard, Nuclear Instrum. Methods 123 (1975) 131.
- 7) H.L. Ravn, S. Sundell, L. Westgaard and E. Roeckl, J. Inorg. Nuclear Chem. 37 (1975) 383.
- 8) H.L. Ravn, L.C. Carraz, J. Denimal, E. Kugler, M. Skarestad, S. Sundell and L. Westgaard, Nuclear Instrum. Methods 139 (1976) 267.
- 9) L.C. Carraz, I.R. Haldorsen, H.L. Ravn, M. Skarestad and L. Westgaard, Fast release of nuclear reaction products from refractory matrices, Nuclear Instrum. Methods 148 (1978) 217.
- 10) G.A. Cowan and C.J. Orth, Proc. 2nd Conf. Peaceful Uses Atomic Energy, Geneva, 1958 (United Nations, Geneva, 1958) vol. 7, p. 328.
- 11) C.J. Orth, Nuclear Sci. Engng. 9 (1961) 417.
- 12) M.L. Andersen and B. Scharff, Ark. Fys. 36 (1967) 151.
- 13) E. Latuszyński, K. Zuber, J. Zuber, A. Potempo and W. Żuk, Nuclear Instrum. Methods 120 (1974) 321.
- 14) A. Latuszyński, Nuclear Instrum. Methods 123 (1975) 489.
- 15) G.J. Beyer and A.F. Novgorodov, Akademie der Wissenschaften der DDR, Zentralinstitut für Kernforschung, Rossendorf bei Dresden, ZKF-303, 305, 310, 311 (1976).
- 16) G.J. Beyer, A.F. Novgorodov, O. Knotek, M. Jachim and J. Juschkevitsch, ZKF-307 (1976).
- 17) G.J. Beyer, A.F. Novgorodov and W.D. Fromm, ZKF-309 (1976), to be published in Nuclear Instrum. Methods.
- 18) R. Klapisch, J. Chaumont, C. Philippe, I. Amarel, R. Ferreau, M. Salome and R. Bernas, Nuclear Instrum. Methods 53 (1967) 216.
- 19) S. Borg, I. Bergström, G.B. Holm, B. Rydberg, L.E. De Geer, G. Rudstam, B. Grapengiesser, E. Lund and L. Westgaard, Nuclear Instrum. Methods, 91 (1971) 109.

- 20) F. Hansen, A. Lindahl, O.B. Nielsen and G. Sidenius, Proc. 8th Internat. EMIS Conf. on Low-Energy Ion Accelerators and Mass Separators, Skövde, 1973 (eds. G. Andersson and G. Holmén) (Chalmers Univ. of Technology and Univ. of Göteborg, Göteborg, Sweden, 1973) p. 426.
- 21) J.M. D'Auria, L.C. Carraz, P.G. Hansen, B. Jonson, S. Mattsson, H.L. Ravn, M. Skarestad and L. Westgaard, Phys. Letters 66B (1977) 233.
- 22) K.V. Davidson, W.W. Martin, D.H. Schell, J.M. Taub and J.W. Taylor, Los Alamos Scient. Laboratory Report LA-5005 (1972).
- 23) W. van Gool, Fast ion transport in solids (North Holland, Amsterdam-London, 1967) p. 201.
- 24) E.K. Storms, The refractory carbides (Academic Press, New York, 1967).
- 25) C.A. Stearns and F.J. Kohl, J. Chem. Phys. 54 (1971) 5180.
- 26) G. Rudstam, Nuclear Instrum. Methods 139 (1976) 239.
- 27) Experimental programme at ISOLDE, CERN/SIS-PU 77-10 (1977).
- 28) R. Kirchner and E. Roeckl, Nuclear Instrum. Methods 127 (1975) 307; 131 (1975) 371.
- 29) G. Rudstam, The Swedish Research Council's Laboratory, Studsvik, Research Report LF-65 (1975).
- 30) F. James and M. Roos, CERN Computer 6000 series Program Library, Long-Write-Up D506, D516 (1971).
- 31) L.C. Carraz, E. Hagberg, P.G. Hansen, B. Jonson, S. Mattsson, H.L. Ravn, M. Skarestad and P. Tidemand-Petersson, Fast target technique for studies of alpha decay from very short-lived francium isotopes (to be published).
- 32) A.K. Lavrukhina and S. Rodin, Radiokhimiya 2 (1960) 83.
- 33) G. Friedlander, L. Friedman, B. Gordon and L. Yaffe, Phys. Rev. 129 (1963) 1809.
- 34) G. Rudstam and G. Sørensen, J. Inorg. Nuclear Chem. 28 (1966) 771.
- 35) G. Rudstam, Z. Naturforsch. 21a (1966) 1027.
- 36) B.L. Tracy, J. Chaumont, R. Klapisch, J.M. Nietschke, A.M. Poskanzer, E. Roeckl and C. Thibault, Phys. Rev. C 5 (1972) 222.
- 37) E. Hagebø and T. Lund, J. Inorg. Nuclear Chem. 37 (1975) 1569.
- 38) E. Hagebø, J. Inorg. Nuclear Chem. 29 (1967) 2515.
- 39) J.A. Panontin and N. Sugarman, J. Inorg. Nuclear Chem. 34 (1972) 1485.
- 40) E. Hagberg, P.G. Hansen, J.C. Hardy, A. Huck, B. Jonson, S. Mattsson, H.L. Ravn, P. Tidemand-Petersson and G. Walter, Phys. Rev. Letters 39 (1977) 792.
- 41) A. Huck, G. Klotz, A. Knipper, C. Miehe, H.L. Ravn, C. Richard-Serre and G. Walter, J. Phys. G 4 (1978) L9.

- 42) N. Kaffrell and the GSI-Mainz-Oslo-Warsaw Collaboration, private communication.
- 43) K. Takahashi, M. Yamada and T. Kondoh, Atomic Data and Nuclear Data Tables 12 (1973) 10.
- 44) The 1975 atomic mass-excess prediction, Atomic Data and Nuclear Data Tables 17 (1976) 477.
- 45) R. Kirchner, O. Klepper, G. Nyman, W. Reisdorf, E. Roeckl, D. Schardt, N. Kaffrell, P. Peuser and K. Schneeweiss, Phys. Letters 70B (1977) 50.
- 46) C. Thibault, M. Epherre, G. Audi, H. Wollnik and R. Klapisch, private communication.
- 47) C. Ekström, G. Wannberg and J. Heinemeier, private communication.
- 48) K.L. Kratz, P.G. Hansen and B. Jonson, unpublished ISOLDE document.

Table 1

Characteristics of the target materials

Target material	Symbol used in text	Dimension (μm)	Vapour pressure ^{a)} at 2000°C (Torr)	Maximum temp. (°C)	Target metal thickness (g/cm ²)	Reference	Supplier
UC-impregnated graphite cloth	U(gr)	20	7.6×10^{-6}	2450	14	22, 24	Union Carbide, Geneva
LaC ₂ -impregnated graphite cloth	La(gr)	20	5.3×10^{-4}	2000	7	25	Union Carbide, Geneva
UC + UC ₂	UC	< 50	7.6×10^{-6}	2450	80	24	Ventron, Karlsruhe
VC	VC	< 25	7.6×10^{-4}	2000	38	24	Ventron, Karlsruhe

a) The vapour pressure refers to the partial pressure of the metal atoms over a condensed phase of metal carbide plus excess carbon.

Table 2

Yields of some nuclides released from uranium-impregnated graphite cloth target. The measurements correspond to a proton beam of 1 μ A and a target temperature of 2050°C. A Re surface ionizer at 1700°C was used for ionization.

Nuclide	Half-life	Yield (atoms/s)	Method of measurement
^8Li	844 ms	4.6×10^4	α counting on ^8Be
^{20}Na	446 ms	10	α counting
^{111}In	2.8 d	4×10^5	β^- counting
^{125}In	12 s, 2 s	3×10^6	γ counting on ^{125}Sn
^{127}In	3.6 s, 2 s	4×10^5	γ counting on ^{127}Sb
^{142}Pr	19.2 h	10^7	γ counting
^{147}Nd	10.98 d	5.4×10^6	γ counting
^{149}Pm	53.1 h	4.3×10^5	γ counting
^{150}Pm	27 h	10^5	γ counting
^{151}Pm	28 h	2.1×10^6	γ counting
^{152}Eu	9.3 h	8.3×10^4	γ counting
^{156}Eu	9.4 h	1.3×10^6	γ counting

Table 3

Diffusion parameters and activation energies

Nuclide	Target	Target temp. (°C)	$\mu \times 10^{-2}$ (s $^{-1}$)	Q (kcal/mol)
^{25}Na	U(gr)	2050	15 $\pm$ 1	64 $\pm$ 6
$^{86\text{m}}\text{Rb}$	"	1750	1.9 $\pm$ 0.2	
"	"	1900	7.4 $\pm$ 0.7	
"	"	2050	15 $\pm$ 1	
$^{130\text{m}}\text{Cs}$	"	1750	1.3 $\pm$ 0.1	67 $\pm$ 6
"	"	1900	4.1 $\pm$ 0.4	
"	"	2050	12 $\pm$ 1	
$^{224}\text{Fr}^*)$	"	1900	8.6 $\pm$ 0.8	
^{224}Fr	"	1900	6.5 $\pm$ 0.6	65 $\pm$ 8
$^{130\text{m}}\text{Cs}$	UC	1950	0.23 $\pm$ 0.02	
^{118}Xe	La(gr)	1900	5.7 $\pm$ 1.2	
^{41}Ar	VC	1760	1.4 $\pm$ 0.3	
"	"	1990	7.7 $\pm$ 1.7	
"	"	2100	15 $\pm$ 3	

*) Delay measurement carried out after 290 hours of operation.

Figure captions

Fig. 1 : The high-temperature target and plasma source assembly as mounted in the standard ISOLDE target container.

1. Outer tantalum container and heating element.
2. Inner graphite tube which contains the carbide matrix.
3. Heat screens.
4. Cooled copper transfer line.
5. W/Re thermocouple.
6. Plasma ion source with forced electron beam induced arc discharge.
7. Ion-source magnet.
8. Mass-marker oven and gas inlet.

Fig. 2 : Yields of Ar isotopes. The filled circles are the observed yields at the collector corresponding to a 1 μ A beam of 600 MeV protons impinging on a 38 g/cm² vanadium carbide powder target at 2100°C. The full-drawn curve shows the trend in cross-sections predicted by Rudstam's spallation formula³⁵⁾, using a Gaussian charge distribution. The cross-section scale on the right axis is normalized to 5.5 mb calculated for ³⁷Ar. The observed yields of short-lived isotopes have been corrected for decay losses in the transport process (open squares) according to Eq. (5) in the text.

Fig. 3 : Yields of Xe isotopes. The filled circles are the observed yields at the collector corresponding to a 1 μ A beam of 600 MeV protons impinging on a 7 g/cm² lanthanum-impregnated graphite cloth target at 1900°C. The full-drawn curve shows the trend in cross-sections predicted by Rudstam's spallation formula³⁵⁾, using a Gaussian charge distribution. The curve is normalized to 8.5 mb measured³⁴⁾ for ¹²⁰Xe. The open square represents a decay-corrected yield (see caption to Fig. 2).

Fig. 4 : Yields of Rb isotopes. The circles (open circle for shielded isotope) are the observed yields at the collector corresponding to 1 μ A of 600 MeV protons impinging on a 14 g/cm² uranium-impregnated graphite cloth target at 2050°C. The observed yield of ⁹⁴Rb has been corrected for decay losses (open square) according to Eq. (5) in the text. The cross-section scale is normalized to 8 mb measured³³⁾ for ⁸⁶Rb. The dashed curve is calculated from experimental charge dispersion and mass distribution of fission of uranium with 570 and 450 MeV protons^{38,39)}. A width (FWHM) of the charge-dispersion curves of 2.9 Z-units is assumed for this mass region, and an increase in the A/Z_p ratio (Z_p is the most probable charge) from 2.37 for $A = 78$ to a constant value of 2.42 for $A \geq 90$. The form of the mass distribution is assumed to be that measured for 450 MeV proton fission of uranium.

Fig. 5 : Yields of Cs isotopes (experimental conditions as for Rb, see caption to Fig. 4). In this case the cross-section scale is normalized to 6 mb measured³³⁾ for ¹³²Cs. The dashed curve corresponds to charge dispersion widths of 3.21 Z-units and a constant A/Z_p -value of 2.43.

Fig. 6 : Yields of Fr isotopes (experimental conditions as for Rb, see capture to Fig. 4). The full-drawn curve shows the trend in cross-sections predicted by Rudstam's spallation formula³⁵⁾, using a Gaussian charge distribution. The curve is normalized to 0.2 mb measured³²⁾ for ²¹²Fr.

Fig. 7 : Delay curve for ¹¹⁸Xe released from a lanthanum-impregnated graphite cloth target [La(gr)] at 1900°C. The figure illustrates the technique of delay measurement described in Section 3.3. The curve has been fitted to the experimental points according to Eq. (3) in the text.

Fig. 8 : Normalized delay curves for alkali products released from targets of uranium-impregnated graphite cloth [U(gr)] and uranium carbide powder (UC) at different target temperatures. The curves have been fitted to the experimental points according to Eqs. (2) or (3) in the text.

Fig. 1

- Fig. 9 : Normalized delay curves for ^{41}Ar released from a vanadium carbide powder target at different target temperatures. The curves have been fitted to the experimental points according to Eqs. (2) or (3) in the text.
- Fig. 10 : The diffusion parameter μ for release of Rb and Cs from a uranium-impregnated graphite cloth target as a function of the inverse of the absolute target temperature.
- Fig. 11 : The diffusion parameter μ for release of Ar from a vanadium carbide powder target as a function of the inverse of the absolute target temperature.
- Fig. 12 : Release yields of Cs isotopes from different uranium target matrices. The release yields, Y_{obs}/Y , are given as a function of the half-life of the nuclear reaction product.
- Fig. 13 : Spectrum of protons from ^{32}Cl after beta decay of ^{32}Ar measured with a Si surface-barrier detector. The counts in the low-energy part are due to positrons.
- Fig. 14 : Decay curves for selected K X- and γ -rays from spectra recorded at 20 s intervals after collection of sources at mass positions 230 and 229, respectively.

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 11

Fig. 10

Fig. 12

Fig. 13

Fig. 14