NEW TARGETS FOR ON-LINE MASS SEPARATION OF NUCLEI
FORMED IN 600 MeV PROTON AND 910 MeV ^3He REACTIONS

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ABSTRACT

The performance of the previously reported UC_2 -graphite cloth, VC, La and Pb targets, as well as the newly developed ThC_2 -graphite, UC_2 -graphite, ScC_2 -graphite, and CaB_6 targets is discussed. Production-yield curves for the elements Li, Na, Ar, K, Ga, Rb, In, Cs, Hg, Tl, and Fr are given for 600 MeV protons and 910 MeV ^3He . It is shown that the latter projectile gives higher yields for the neutron-deficient isotopes produced in fragmentation and spallation processes.

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1. INTRODUCTION

In recent years great progress has been made in the fast-target techniques for the production of intense and pure beams of mass-separated radioactivity. Especially, the development of high-temperature targets (1700-2200 °C) has given the field a great push forward and permitted the study of nuclides with half-lives down to milliseconds. Typical examples of these targets are the refractory metal powder targets such as Nb, Ta, and Hf, and also the refractory carbides such as VC, UC₂-graphite cloth and LaC₂-graphite cloth^{1,2}).

The high temperature and the short solid-state diffusion path make it possible to achieve a fast release of the reaction products which is decisive for the yields of short-lived nuclides. The production rates may become very high since the long range of the relativistic bombarding particles allows the use of thick targets. As one link in a long chain of related articles, this paper deals with new target developments which have led to substantial improvements, both with respect to increased intensity of the separated beams and to the number of elements available for study.

In Section 2.3 we give the details of a new method for the preparation of carbide targets. In Section 3 the results of the survey of the produced elements are discussed in terms of the isotopic yield distribution as obtained from 600 MeV proton reactions and, in most cases, also from irradiations with a 910 MeV ³He beam. Finally, in Section 4, we summarize the present status and mention some possibilities for further development.

2. EXPERIMENTAL

2.1 Design of the target and ion-source unit

The ISOLDE target unit is described in detail elsewhere^{3,4}). It consists of a target material container which can be heated to 2400 °C. The evaporated nuclear reaction products are allowed to diffuse to the ion source via a transfer tube, which may be either heated or cooled. A high chemical selectivity is obtained by combining different target materials with positive surface ionization, negative surface ionization, or plasma discharge ionization.

2.2 General target material treatment

To ensure good working conditions of the ion source and the separator, it is important to keep the pressure in the ion source below $\sim 10^{-4}$ Torr. This requires high purity of the materials used in the entire assembly. Furthermore, a sintering and outgassing procedure which lasts for more than 24 h at temperatures of 1500-2000 °C is necessary. This is normally done at an off-line target pump stand, which has quick connectors identical to those of the on-line separator. In some cases the target material alone is reacted in an earlier-described⁵⁾ vacuum oven. Temperature calibration of the target container as a function of the applied current is performed simultaneously with a two-colour optical pyrometer through a glass window in the vacuum chamber. This is a necessary procedure since a thermocouple connected to a high-temperature target lasts for only the first 5-6 hours of on-line conditions.

2.3 Targets

The UC₂-graphite cloth target. The development of this target is described in detail elsewhere^{2,5,6)}. Here we give only a brief reminder that a graphite tube, filled with ~ 380 UC₂-graphite cloth disks of 15 mm diameter, was inserted in a 20 cm long tantalum target container. The graphite tube served the purpose of increasing the lifetime of the Ta container, which otherwise would react with the UC₂-graphite cloth. In recent targets this reaction is delayed by means of two Ta-foil layers inside the Ta container; these prevent direct contact between the UC₂-graphite cloth and the container wall. The target thicknesses were 13-17 g/cm² of U.

The ThC₂-graphite target. A new general method for the production of carbide materials was used in the preparation of this target matrix. Graphite powder and ThO₂ were mixed in a ball mill. From this mixture, 2-3 mm thick tablets with a diameter of 14 mm were formed in a compacting press which applied a pressure of ~ 5 tons. The tablets were placed in a graphite tube and reduced to ThC₂ in the vacuum oven at ~ 2000 °C. The tablets consist of a solid solution of ThC₂ in excess graphite, and have a high porosity due to the voids left after the compacting

procedure and from those created by the reduction process. The advantage of this procedure over the previously described graphite-cloth impregnation²⁾ method is that it allows thicker targets to be made within a given container length. The currently used material has a Th:C molar ratio of 1:3.8; when placed in a standard Ta target container, it allows for Th target thicknesses of ~ 50 g/cm². For practical reasons a 5 cm target of only 12 g/cm² of Th was used in the present experiment.

The VC target. Pure VC powder was compressed into tablets and loaded into a graphite-lined Ta target container, as described for ThC₂ in the previous section. After outgassing at 1700 °C, the density of the tablets was measured to be 4 g/cm³; this corresponds to 70% of the specific density of pure VC. The target thickness was 62.3 g/cm² of V.

The ScC₂-graphite target. The ScC₂-graphite target was prepared in the same way as the ThC₂; it had an Sc:C molar ratio of 1:3.8, which allowed for an Sc target thickness of 12.8 g/cm².

The CaB₆ target. Commercially available CaB₆ powder was loaded into the graphite-clad Ta target container and compressed manually to about 50% of the specific density of pure CaB₆. The Ca target thickness obtained was 7.5 g/cm².

The Pb and La targets. These two targets, described elsewhere³⁾, are here referred to only for the comparison of Hg and Cs production rates from ³He and protons. The target thicknesses were 140 g/cm² of La and 170 g/cm² of Pb.

2.4 Beams from the CERN Synchro-cyclotron (SC)

The beams available at ISOLDE from the CERN SC are ~ 4 μ A 600 MeV protons, ~ 0.5 p μ A *) 910 MeV ³He²⁺, and ~ 0.1 p μ A 86 MeV/u ¹²C⁴⁺. Other heavy ions such as ¹⁵N⁵⁺, ¹⁶O⁶⁺, and ²⁰Ne⁶⁺ have been accelerated at the synchro-cyclotron, but their relatively low intensity makes them less interesting at present. Only those results obtained by the first two beams mentioned above will be given here. The use of ¹²C⁴⁺ as a production projectile has been rather extensively studied, and the results will be published in a forthcoming paper⁷⁾.

*) 1 p μ A = 6.24×10^{12} particles/s.

2.5 The production yield measurements

In this article the performance of the different targets will be presented only by the measured production yields. Their release properties are discussed in another paper⁷⁾.

The production yield is defined here as the measured intensity of the mass-separated beams in the collector tank. The yields were determined by in-beam beta-decay measurements at saturation for the very short-lived products. For the long-lived products, collection and successive singles counting or multiscaling of the beta activity were used in order to determine the correct component from the complex decay curves. The most abundant radioactive ion beams were measured directly in a Faraday cup. In order to average short-time variation in the bombarding particle flux and in the separation and detection efficiency, it was found necessary to determine the intensity ratio between two adjacent masses rather than the absolute value. The method will be outlined more closely in a forthcoming paper⁷⁾.

For the various measurements, the ion beam was brought into a low-background counting area and collected on an aluminized mylar tape for transport of the activity. The beta particles were counted in a thick efficiency-calibrated 4π particle scintillator. For gamma rays, a 17% Ge(Li) detector was used. Charged particles were recorded by silicon surface barrier detectors. The delayed neutrons were measured with eight ^3He proportional counter tubes embedded symmetrically in paraffin-wax, yielding a detection efficiency of 14%. The P_n values used were taken from other papers⁸⁻¹¹⁾.

Intercalibrated (identical) Faraday cups were used to check the beam transmission in the beam line.

The yields are calculated by the usual formula:

$$Y = \frac{S}{\epsilon_d \epsilon_t I_B [1 - \exp(-\lambda t_{\text{coll}})] [\exp(-\lambda t_{\text{decay}})] \frac{1}{\lambda} [1 - \exp(-\lambda t_{\text{count}})]},$$

where

S = counts in time t_{count} ,
 ϵ_d = detector efficiency,
 ϵ_t = beam line transmission (70-80%),
 I_B = branching ratio,
 t_{coll} = collection time,
 t_{decay} = waiting time,
 t_{count} = counting time.

3. RESULTS AND DISCUSSIONS

In this section the production yields for the various elements from the different target matrices are discussed. The target and ion source temperatures as well as the target thicknesses are summarized in table 1.

3.1 Li, Na, K, Rb, Cs, and Fr from UC_2 -graphite cloth

The production yields for the alkali elements are shown in figs. 1-6. Among the alkalis, Li is the most difficult to ionize; and for fast release, the temperature of both the target and the ion source must be high. Even at a line temperature of 2500 °C, the Li-production yield increases linearly with the temperature.

For the fragmentation products (Li and Na) there is a trend towards higher production yields with ^3He than with the protons (figs. 1 and 2). This trend appears to be washed out at K (fig. 3), and in the fission product region (Rb and Cs) it is not present at all [figs. 4 and 5, where for comparison the isotopic yields from thermal neutron-induced fission¹²⁾ are also shown]. The measured yields for ^3He are here even somewhat lower than those for protons.

In the region of spallation products (Fr) the mass distribution with ^3He appears flatter than for protons, and the yields for the most neutron-deficient isotopes have even higher absolute values (fig. 6). This variation is illustrated in fig. 7, where the ratio $Y(^3\text{He})/Y(p)$ is plotted versus the mass number. A substantial gain in production yield could be achieved for the lightest isotopes with ^3He instead of protons as the bombarding projectile, assuming the same beam intensity. The

relative effect on the production yields of the two projectiles can be tentatively understood in terms of higher energy deposition and transfer of angular momentum by the ^3He than by protons.

3.2 Ga, In, and Tl from UC_2 -graphite cloth

As the first ionization potential for Ga, In, and Tl is around 6 eV, they can be ionized with efficiencies of the order of 10% in a tungsten surface-ionization source. The resulting yield curves for Ga, In, and Tl are given in figs. 8-10, respectively.

With this ion source, Ga can be produced almost radiochemically pure (contamination factors $\ll 10^{-3}$). For the heaviest isotopes ($A \leq 80$), Rb and Sr may appear as contaminants, but the measurements can always be designed to favour Ga because of the big difference in half-life.

The production yields for $A > 78$ have not yet been properly measured, but a neutron mass scan, presented in fig. 8, showed activity up to $A = 85$.

The problem with contaminant alkalis is more pronounced in the In production, especially for $A \geq 124$, where Cs (and some Ba) isotopes are produced in relatively large amounts. The yields for $A > 130$ are not measured, although nuclear spectroscopy experiments have been successfully performed on $A = 132$ ¹³). Since Ga and In are fission products, they are not expected (in accordance with the results presented above for Rb and Cs) to be produced in larger amounts by the ^3He beam than by the proton beam. For Tl isotopes, produced in deep spallation, one would expect the ^3He projectiles to be better than the protons. The experimental results given as the ratio $Y(^3\text{He})/Y(p)$ in fig. 11 reveal that the isotopes with $A < 187$ do have yields from ^3He bombardment that are 10-100 times higher than the corresponding proton-induced yields.

3.3 Tl and Fr from ThC_2 -graphite

When comparing U and Th as targets for the production of Tl and Fr, the latter matrix, as shown in figs. 12 and 13, gives higher absolute yields, which is to be expected because of its closer proximity to the products of interest.

The yield curve given in fig. 12 from ^3He -induced reactions shows a higher maximum and decreases more slowly on the neutron-deficient side than the corresponding proton curve. The ratio of these two curves is shown in fig. 14. The yield curve maxima are, as expected, somewhat higher from Th than from U, but a longer delay time in the former matrix tends to reduce the gain on the very neutron-deficient side, where the delay-time losses become more and more important owing to the shorter half-lives.

3.4 Ar from VC

In order to avoid a strong decomposition of the VC compound, the highest target temperature that can be used is $\sim 1900^\circ\text{C}$. The target is connected to a cooled line ($< 100^\circ\text{C}$) allowing only noble gases to diffuse to the FEBIAD-plasma ion source²⁾. The earlier published²⁾ yield curve for protons has been revised and is shown in fig. 15. The production of Ar by ^3He , compared to production by protons, gives higher yields for the neutron-deficient nuclides, and similar or somewhat lower yields for the neutron-rich ones. This effect is likewise ascribed to the higher total energy available in the reacting system.

3.5 K from ScC_2 -graphite

The carbide of Sc is relatively stable up to $\sim 1900^\circ\text{C}$. At this temperature the fast diffusion and release of K are ensured. Again the ^3He -induced yields are higher than the corresponding proton-induced ones (fig. 16). No systematic production of K has been attempted from a VC target, so a direct comparison between the two matrices is not possible.

According to spallation cross-section calculations using the formula of Silberberg and Tsao¹⁴⁾, the production of neutron-deficient Ar isotopes would be favoured by a factor of 3 to 10 when using Sc as a target element instead of V, assuming normalized conditions.

Also, a direct comparison of the two matrices is not possible here, because no attempt has been made to produce Ar from the Sc target.

3.6 Ar from CaB₆

Nuclei with $N < Z$ are of particular interest since the isobaric analogue state in the neighbouring nuclei lies within the beta-decay window. The strong demand for more cases with negative isospin projection has led to the development of Ca-based targets which may make it possible to produce elements with $Z < 19$ (i.e. ^{32}Ar , ^{31}Cl , ^{28}S , ^{27}P).

Spallation cross-section calculations gave an enhancement factor of 40 to 200 for the production of ^{32}Ar as compared to V as a target. A study of off-line release, high-temperature stability, and corrosion showed CaB₆ to be suitable. Unfortunately, this choice gives less chance of sulfur release, because CaS is a rather stable compound at high temperatures. In an oxygen-containing Ca compound, however, one could hope for release of sulfur as SO or SO₂.

The on-line experiment revealed some difficulties due to a rather strong and long-lasting outgassing, probably because of impurities in the target material. The operation of the plasma ion-source became difficult since it was connected to the target via an uncooled line. The maximum target temperature was 1500-1600 °C. The two measured yield points of ^{34}Ar and ^{35}Ar plotted in fig. 15 should be considered to represent a lower limit. If one normalizes to the same molar fraction of the target element, the gain in using Ca is a factor of 80 to 90. As expected, S and P were not seen (Ca₃P₂ is also quite stable at high temperatures). Chlorine, which we hoped to produce as CaCl⁺, was not seen, probably because of a too low line temperature.

3.7 Hg from Pb and Cs from La

The yield curves shown in figs. 17 and 18 indicate that ^3He -induced spallation gives higher yields for the neutron-deficient products. The effect is less pronounced in these two systems, where the targets are only two protons away from the product.

4. CONCLUSIONS AND OUTLOOK

All the target materials described here, except (so far) CaB₆, have been used in on-line production of nuclides for studies of nuclear properties. The

potential of these targets to produce several other elements is large and has still not yet been fully examined. For instance, the production of the halogens Cl, Br, and I from the U target has become routine¹⁵⁾. With new developments in the ion-source field and in the chemical evaporation technique^{3,16)}, we expect many more elements to become available as pure beams within the next 3 to 4 years. The oxide-containing materials are especially interesting with respect to the evaporation of some refractory elements that have volatile oxides.

One of the new targets developed at ISOLDE is a TaC₂-graphite target for the production of neutron-rich isotopes of the light noble gases and for a number of other elements. On-line tests show very promising results, and in some experiments TaC₂ will probably replace the U targets.

The results from using the 910 MeV ³He beam are encouraging, and an over-all higher production rate can be established for the spallation and fragmentation products.

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Table 1

Experimental parameters for production of some elements
from UC₂-graphite cloth target

Element	Target temp. (°C)	Ionizer temp. (°C)	Target amount (g/cm ²)
Li	2000	2500	13.6
Na	1800-2000	2000-2200	13.6
K	1800-2000	2000-2200	13.6
Rb	1750-1850	1750-2000	16.4
Cs	1750	2000	16.4
Fr	1800	2000	13.6
Ga	2000	2400	15.1
In	2000	2500	15.1
Tl	2000	2500	15.1

Figure captions

- Fig. 1 : Production yields of Li isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in UC_2 -graphite cloth. The yield of $A = 7$ was measured by ion detection, $A = 8$ by $4\pi\beta$ counting, and $A = 9$ and 11 by delayed-neutron counting (Ref. 8).
- Fig. 2 : Production yields of Na isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in UC_2 -graphite cloth. The yields of $A = 20, 21$, and 25-27 were measured by $4\pi\beta$ counting, $A = 24$ by γ counting, and $A = 27-32$ by detection of delayed neutrons. The P_n values used are based on the new value for ^9Li of $50 \pm 4\%$ (Refs. 8 and 9).
- Fig. 3 : Production yields of K isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in UC_2 -graphite cloth. The yields of $A = 37, 38$, and 44-50 were measured by $4\pi\beta$ counting, $A = 42$ and 43 by γ counting, and $A \geq 49$ by detection of delayed neutrons, for which the P_n values have recently been measured (Ref. 10).
- Fig. 4 : Production yields of Rb isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in UC_2 -graphite cloth. The yields of $A = 79, 81, 86$, and 90-94 were measured by $4\pi\beta$ counting, and $A > 93$ by detection of delayed neutrons. The P_n values for $A \leq 98$ (Ref. 11) are all normalized to the new P_n value of ^9Li (Ref. 8), while those for $A = 99, 100$, and 102 are estimated. We also give the yields for proton-induced spallation in a Nb-powder target (50 g/cm^2) (Ref. 1) and the strongly cumulative yields from thermal neutron-induced fission of ^{235}U (Ref. 12).

Fig. 5 : Production yields of Cs isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in UC_2 -graphite cloth. The yields of $A = 123, 125$, and $140-146$ were measured by $4\pi\beta$ counting, $A = 127, 129$, and 132 by γ counting, and $A = 146-152$ by detection of delayed neutrons. The P_n values are estimated, and the yields normalized to the β measurement at $A = 146$.

Also shown are the yield curve from proton-induced spallation in molten La (120 g/cm^2) (Ref. 1) and the strongly cumulative yields from thermal neutron-induced fission of ^{235}U (Ref. 12).

Fig. 6 : Production yields of Fr isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in UC_2 -graphite cloth. The yields of $A = 202-213$ and $219-221$ were measured by α counting, and $A \geq 222$ by $4\pi\beta$ counting.

Fig. 7 : Ratios of the $^3\text{He}^{2+}$ to proton-induced production yields of Fr isotopes from UC_2 -graphite cloth.

Fig. 8 : Production yields of Ga isotopes from 600 MeV proton-induced reactions in UC_2 -graphite cloth. For $A = 80-82$ and 85 , a mass scan by means of β -delayed neutron detection is shown.

Fig. 9 : Production yields of In isotopes from 600 MeV proton-induced reactions in UC_2 -graphite cloth, all masses measured by the $4\pi\beta$ detector. Also shown are the yields of three Ag isotopes measured simultaneously.

Fig. 10 : Production yields of Tl isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in UC_2 -graphite cloth. The yields of $A = 184-186$ were measured by α detection, and $A \geq 185$ by the $4\pi\beta$ detector (conversion electron detection). The letters g and m along the abscissa mean that the corresponding yield point is from the ground state or the metastable state.

It should be noted that since no corrections have been made for the conversion electron branching (100% used), and a fixed lower discriminator level has been used on the pulse height, the absolute level of the curves was not determined to a better precision than a factor of 2 to 3.

- Fig. 11 : Ratios of the ${}^3\text{He}^{2+}$ to proton-induced production yields of Tl isotopes from UC_2 -graphite cloth.
- Fig. 12 : Production yields of Tl isotopes from 600 MeV proton-induced and 910 MeV ${}^3\text{He}^{2+}$ -induced reactions in ThC_2 -graphite, with target and ion source temperatures of 2000 °C and 2400 °C, respectively. The comments in the text of fig. 10 are also valid here.
- Fig. 13 : Production yields of Fr isotopes from 600 MeV proton-induced reactions in ThC_2 -graphite with target and ion source temperature of 2000 °C. The low temperature on the ion source was used in order to limit the simultaneous ionization of the neighbouring element Ra (in addition, see the text of fig. 6).
- Fig. 14 : Ratios of the ${}^3\text{He}^{2+}$ to proton-induced production yields of Tl isotopes from ThC_2 -graphite. The curve is constructed from the two dashed lines shown in fig. 12, and normalized at $A = 194$. The normalization is done because of uncertainty in the absolute position of the yield curve from ${}^3\text{He}^{2+}$.
- Fig. 15 : Production yields of Ar isotopes from 600 MeV proton-induced and 910 MeV ${}^3\text{He}^{2+}$ -induced reactions in VC. Also shown are the two yield points at $A = 34$ and 35 from proton-induced reactions in CaB_6 . The yield of $A = 32$ was measured by β -delayed proton detection, and the other masses by $4\pi\beta$ detection.

- Fig. 16 : Production yields of K isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in ScC_2 -graphite. The yield of $A = 35$ was determined by in-beam β detection, while the others were measured by the $4\pi\beta$ detector. A tungsten surface ionization source was used at a temperature of 2000 °C.
- Fig. 17 : Production yields of Cs isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in molten La. The yields of $A = 118-137$ were measured by means of the Faraday cup, $A = 116-117$ by the $4\pi\beta$ detector, and $A = 114$ by β -delayed protons.
- Fig. 18 : Production yields of Hg isotopes from 600 MeV proton-induced and 910 MeV $^3\text{He}^{2+}$ -induced reactions in molten Pb. The yields of $A = 186-205$ were measured by means of the $4\pi\beta$ detector, and $A = 177-185$ by α detection.

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