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HEAVY ISOTOPE PRODUCTION BY MULTINUCLEON TRANSFER REACTIONS WITH ^{254}Es*

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Summary

Fast automated on-line and quasi-on-line radiochemical techniques are applied to search for new isotopes, to measure their decay characteristics and to study the cross sections of the heaviest most neutron-rich actinide isotopes in reactions of ^{16,18}O and ^{22}Ne projectiles with ^{254}Es as a target. The measured yields for isotopes up to lawrencium-260 are three or more orders of magnitude higher than in any other reaction used so far. A comparison with data for similar transfers from ^{248}Cm targets is made. Transfer cross sections are extrapolated for the production of unknown, neutron-rich isotopes of elements 101 through 105, and the unique potential of ^{254}Es as a target to make these exotic nuclei accessible is demonstrated.

1. Introduction

One major effort in nuclear chemistry always was, and still is, to extend our knowledge beyond the present limits of known nuclei to learn what finally determines the limits of stability for a nucleus. From the chemist's point of view one is especially interested in investigating the chemical properties of the very heavy elements and to find the end of the Periodic Table of Elements.

Compound nucleus reactions were previously used to establish the present limits of known actinide and transactinide isotopes [1 - 5]. Detailed studies of many interesting properties and the production of still heavier nuclides are prohibited by losses from the relatively high excitation energy

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of the compound nucleus and the severe fission competition in the de-excitation process, or because of the limited number of available target and projectile combinations. Short spontaneous fission (sf) half-lives for the most neutron-rich nuclei make it impossible to produce nuclides heavier than ^{257}Fm in multiple neutron capture reactions [6].

However, systematic studies of transfer reactions with ^{248}Cm targets and light projectiles like ^{16,18}O and ^{20,22}Ne have demonstrated that the problem of high excitation energies and subsequent losses due to fission competition can be partially avoided in these reactions [7].

We report on measured yields of neutron-rich actinide nuclides from transfer reactions of light projectiles with ^{254}Es as a target. This target isotope, a 270-day α-emitter with $Z = 99$, is the heaviest nuclide that can be produced in the multimicrogram quantities necessary for heavy ion bombardments. Besides classical radiochemical separations we have applied a variety of newly developed fast chemical and physical techniques. Helium jets loaded with KCl clusters were coupled to an automated chemistry apparatus or to devices with rotating catcher wheels to measure products with half-lives as short as 0.6 s. Our results demonstrate that cold, multinucleon transfer reactions with ^{254}Es as a target offer a unique possibility to access and investigate new neutron-rich actinide nuclides.

2. Experimental techniques

Our experiments were performed with 101 MeV ^{16}O, 98 MeV ^{18}O and 127 MeV ^{22}Ne ions at the Lawrence Berkeley Laboratory's (LBLs) 88-inch cyclotron and the UNILAC accelerator at GSI. In-target projectile energies were determined using a surface-barrier detector. The particle flux was typically kept between 1×10^{12} s^{-1} and 2×10^{12} s^{-1}. Einsteinium oxide targets with a diameter of 3 mm which contained 24.2 and 27.5 μg cm^{-2} ^{254}Es (isotopic purity, 96.5% and 92%) were produced by electrodeposition on 2.4 mg cm^{-2} beryllium foils. These were mounted in target chambers designed either to catch products with a recoil angle of not more than 45° in two 2.8 mg cm^{-2} gold catcher foils, or to transport the activity by means of a helium jet containing KCl clusters to an Automated Rapid Chemistry Apparatus (ARCA) or a Rotating wheel Multidetector Apparatus (ROMA) [8].

Chemical fractions of fermium through nobelium were separated from gold catcher foils within 1 - 2 h. After separating the gold on an anion exchange column a No^{2+} fraction was separated on a column with di(2-ethylhexyl) orthophosphoric acid (HDEHP) as a stationary phase. Fermium and mendelevium were separated on a cation exchange column with α-hydroxyisobutyric acid (α-HIB) as a complexing agent.

An adaptation of manually performed ion exchange and liquid–liquid extraction chromatographic separations [9, 10] to a microprocessor controlled ARCA allowed the separation of mendelevium through lawrencium

fractions within 5 - 15 min [11]. The chemistry was performed on-line in a cyclic mode of operation. Reaction products carried by the jet clusters over a distance of about 50 m into a chemistry laboratory were collected for some time on a glass filter frit. Then the gas jet was disconnected for the time a dilute solution of HCl was passed through the frit in order to dissolve the products and deliver them to a small column filled with HDEHP on an inert support. The flow of all solutions was maintained by three independently controlled high-pressure liquid chromatography pumps with pump-heads made from an inert material (Kel-F®). Separated No^{2+} was eluted from the first column in 0.03 N HCl and further cleaned with 4 N HCl on a cation exchange column. After the elution in 4 N HCl from the HDEHP column a mendelevium and lawrencium separation was performed on a cation exchange column with α-HIB. These fractions were assayed for α-particle and sf activities.

To study even shorter lived isotopes reaction products were continuously transported into our ROMA [8] within one second of production and were deposited there on 70 $\mu g\ cm^{-2}$ polypropylene foils which were stepwise rotated between 15 pairs of surface-barrier detectors to measure α- and sf-decays with typical collection-measuring cycle times of 4 - 60 s. Total transport and deposition efficiencies for the jet system were determined to be 13% for the experiments with ^{22}Ne and 50% with ^{18}O as a projectile. The efficiencies were obtained by normalizing cross sections for longer lived species like ^{255}Fm to results from catcher foil experiments where chemical yields (typically 80%) were known. For ^{16}O a 50% efficiency was assumed.

3. Results

Formation cross sections for the transeinsteinium isotopes are shown in Fig. 1 for (a) 101 MeV ^{16}O, 98 MeV ^{18}O and (b) 127 MeV ^{22}Ne as projectiles. We have also included data points for ^{259}Md and the newly found isotope ^{260}Md from a very recent experiment of 108 MeV ^{18}O and 126 MeV ^{22}Ne on ^{254}Es [12, 13] despite a somewhat higher energy in the ^{18}O experiment. These samples were prepared by mass separation after the end of the bombardment. All curves in Fig. 1 are gaussian with a variance of $\sigma^2 = 0.914\ u^2$ (FWHM = 2.25 u). Of most interest are the high absolute cross sections, especially in comparison with other possible reactions forming the same product. Peak cross sections of about 1 mb for mendelevium compare with only about 1 μb in $^{18}O + ^{249}Cf$ [14], 100 nb in $^{238}U + ^{248}Cm$ [15] and 10 nb in ^{18}O, $^{22}Ne + ^{248}Cm$ [7] reactions. The neutron-rich isotopes ^{259}No and ^{260}Lr are produced with 3.5 μb and 1.1 μb respectively in the $^{22}Ne + ^{254}Es$ transfer reactions, while reactions like $^{248}Cm(^{18}O, \alpha 3n)^{259}No$ and $^{248}Cm(^{15}N, 3n)^{260}Lr$ give cross sections of about 27 nb [4] and 2 nb [1] respectively. These larger cross sections with ^{254}Es as a target can easily be understood when one bears in mind that for systems with light projectiles and heavy targets at

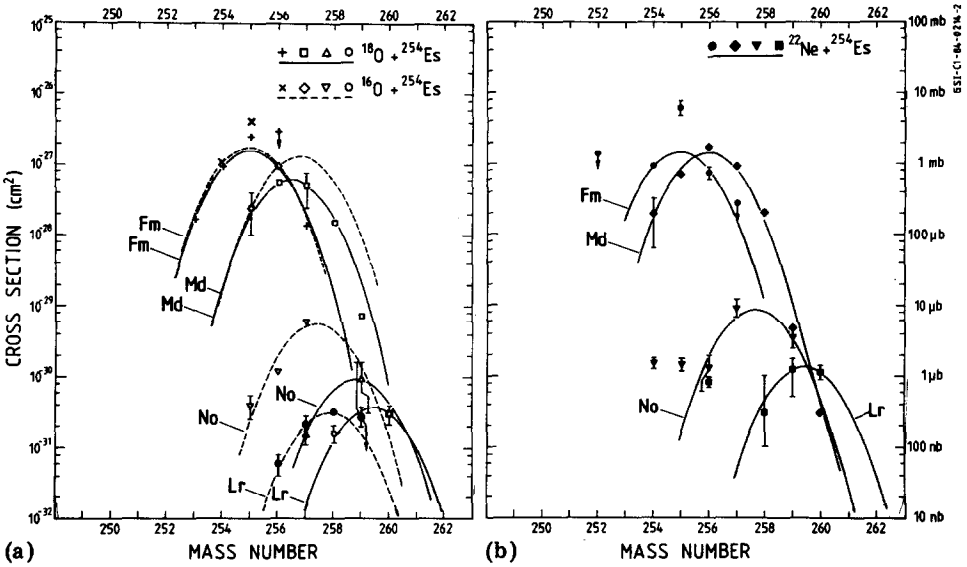

Fig. 1. Isotopic distributions measured for (a) 101 MeV ^{16}O and 98 MeV ^{18}O and (b) 127 MeV ^{22}Ne on ^{254}Es.

barrier energies (i) about 50% of the total reaction cross section appears in transfer channels [16], (ii) transfer products heavier than the target are formed predominantly, and most importantly, (iii) because of the differences in ground-state Q values, Q_{gg}, these primary fragments are formed with very low excitation energies (not more than 10 MeV), so that in most cases the observed products are directly formed or they arise from the one-neutron evaporation channel [14, 15]. This results in a much higher survival probability against fission in the de-excitation process, compared with the three or four neutrons evaporated after compound nucleus formation or from fragments formed in transfer reactions between two very heavy partners like $^{238}U + ^{238}U$ [17] or $^{238}U + ^{248}Cm$ [15].

A systematic feature, which is most pronounced with ^{18}O as a projectile (see Fig. 1) are equally high cross sections for the 1p(Fm)- and the 2p(Md)-transfer followed by a large gap of two to three orders of magnitude and then again only slightly higher cross sections for the 3p(No)- over the 4p(Lr)-transfer. Such a relative enhancement of products formed in transfers of even numbers of protons can also be seen in product yields from $^{18}O + ^{248}Cm$ [7] and from lighter targets like ^{181}Ta and ^{208}Pb [16], clearly indicating a structural effect in the light projectile which is possibly due to differences in the Q_{gg} values. Since one may also speculate about a diproton transfer, it is somewhat surprising that the data do not show any evidence for an intense α-transfer channel. No such strong "odd–even effect" is observable in reactions between two heavy nuclei like $^{238}U + ^{238}U$ [17] or $^{238}U + ^{248}Cm$ [15].

It is surprising to see in our experiments that the difference in neutron number for ^{16}O and ^{18}O projectiles is not fully reflected in the peak position

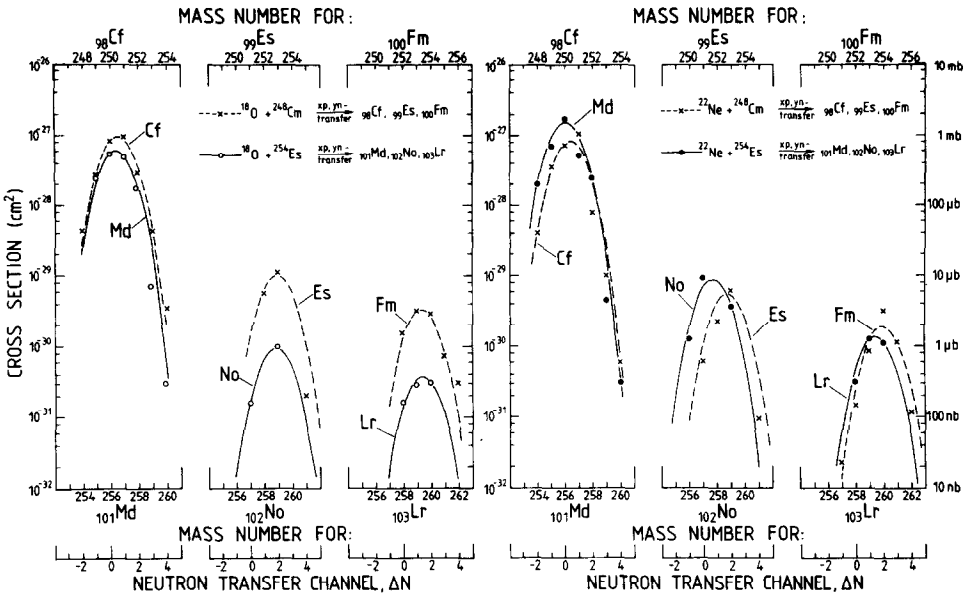

Fig. 2. Comparison of measured isotopic cross sections for two-, three- and four-proton and x-neutron (ΔN) transfer products from (a) ^{248}Cm and ^{254}Es as targets and ^{18}O and (b) ^{22}Ne as projectiles. Data for ^{248}Cm targets are from Ref. 7.

of the isotope distributions for 1p(Fm)- and 2p(Md)-transfer products as was observed earlier with ^{248}Cm targets [7] while for heavier products it is again observed. Because of the similarity of Q_{gg} values for similar transfers from ^{248}Cm and ^{254}Es targets one would not expect such a pronounced difference. In order to gain additional insights into the reaction mechanism and to base extrapolations of cross-sections on firmer grounds, comparison with yield data from other reactions can be made by correlating cross sections for isotopes which are formed in equal transfer channels, (xp, yn), [15]. This is shown in Fig. 2 where cross sections are plotted for two-, three- and four-proton and x-neutron (ΔN) transfer products from the reactions of the two projectiles ^{18}O (a) and ^{22}Ne (b) with ^{248}Cm [7] and ^{254}Es as targets. Positive ΔN values characterize neutron transfer from the projectile to the target. The heights and positions of the isotope distributions for elements near the target Z match very well for similar transfers from the different targets. However, for 3p(Es, No) and 4p(Fm, Lr) transfers, the situation is not so simple. The ^{22}Ne systems match fairly well, but the ^{18}O yields are different by an order of magnitude. This cannot be explained by small differences in the incident projectile energy, because in all reactions the energy was chosen high enough above the barrier to be in a region where cross sections do not change drastically with small changes in energy [14]. A calculation of the maximum excitation energy, $E^*_{max} = Q_{gg} + (E_i - E_f)$, with E_i being the incident energy and E_f being the Coulomb energy in the two fragment exit channel, does not show any significant differences in energies

to account for the observed effect. An explanation has to await further studies. As a result, we find that simple extrapolations from one reaction system to another which in some cases lead to surprisingly good predictions may not always do so.

We base our predictions for cross sections of yet unknown heavy actinide nuclides on the isotope distributions fitted to the measured data. The results are given in Table 1. These predictions can be uncertain by about one order of magnitude. A comparison between the predicted cross section for ^{260}Md based on data from our first experiments and the measured cross sections from a very recent experiment [12, 13] (numbers given in parentheses in Table 1) may indicate the accuracy of these predictions. These cross sections should be high enough to permit not only the identification of these isotopes but also the investigation of their decay properties, especially if the proposed large 40 μg einsteinium target [18] should become available. For isotopes of elements 104 and 105, at the maxima of the isotope distributions around mass numbers 262 and 264, cross sections between 1 and 10 nb can be expected. Increasing α-decay half-lives for these more neutron-rich isotopes may also provide the chemist with the means to study the onset of atomic relativistic effects which may cause deviations of chemical properties in these high Z elements.

TABLE 1

Extrapolated cross sections for heavy, neutron-rich actinides in ^{18}O + ^{254}Es and ^{22}Ne + ^{254}Es reactions

Isotope	*Cross section* (nb)	
	$^{18}O + {}^{254}Es$	$^{22}Ne + {}^{254}Es$
Md-260	740 (320)	240 (298)
261	9	2
No-260	490	470
261	85	22
262	5	0.3
Lr-261	100	380
262	12	43
263	0.5	2

For a comparison measured data for ^{260}Md are given in parentheses.

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