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Basic Studies of a Gas-Jet-Coupled Ion Source
for On-Line Isotope Separation

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Basic Studies of a Gas-Jet-Coupled Ion Source
for On-Line Isotope Separation*

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Abstract

A hollow-cathode ion source was used in a gas-jet-coupled configuration to produce ion beams of fission products transported to it from a ^{252}Cf fission source. Solid aerosols of NaCl and Ag were used effectively as activity carriers in the gas-jet system. Flat-plate skimmers provided an effective coupling of the ion source to the gas jet. Ge(Li) spectrometric measurements of the activity deposited on an ion-beam collector relative to that deposited on a pre-skimmer collector were used to obtain separation efficiencies ranging from 0.1% to >1% for Sr, Y, Tc, Te, Cs, Ba, Ce, Pr, Nd and Sm. The use of CCl_4 as a support gas resulted in a significant enhancement of the alkaline-earth and rare-earth separation efficiencies.

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1. Introduction

An on-line isotope separator facility is being developed at the Idaho National Engineering Laboratory. The facility is based on the gas-jet transport of fission-product activity from mg size ^{252}Cf sources into the ion source of an ISOLDE-1 type¹⁾ electromagnetic mass separator.* Our dispersion magnet is, however, a 90° mass analyzer. Although there are several test reactors in the immediate area of the laboratory, their use as a neutron source for on-line study of fission-product radioactivity is limited. We chose, therefore, to utilize large californium sources as our source of fission products. Cf-252 offers some attractive features. Principally, when compared to $^{235}\text{U}(n,f)$, ^{252}Cf spontaneous fission provides a significantly higher relative yield of rare-earth and valley-element activity. In addition, the large californium sources can be located in a laboratory-scale shielded cell²⁾ in close proximity to our mass separator and to a radiochemistry laboratory. The chemistry laboratory is equipped with fast chemical separation systems for complementary fission-product studies.

The purpose of this paper is to report on our efforts to develop a gas-jet-coupled ion source for use in our ISOL facility. We have followed with interest similar development activities in other laboratories³⁻¹¹⁾ and have chosen the following direction for our ion-source development effort. Because of its larger discharge chamber volume, low pressure operation, generally low operating temperature, and the very limited success with coupling it to a gas-jet system^{3,5,7)}, we did not attempt

* Nucletec S. A., Geneva, Switzerland.

to use the standard oscillating electron or Nielsen type ion source¹²⁾. Instead we chose to implement a version of the Sidenius hollow-cathode ion source^{13,14)} which has the attractive features of high pressure and high temperature (>2000°C) operation, high ionization efficiency and a small discharge volume. Limited success with direct adaptations of this ion source in a gas-jet-coupled configuration has been reported by others^{4,5)}. Moltz et al.^{10,11)} have demonstrated more recently the utility of this type of ion source for the production of a relatively broad range of elements transported by the gas-jet technique from a reaction chamber in a cyclotron beam. Although Zirnheld et al.^{8,9)} have demonstrated the use of a gas-jet-coupled hollow-cathode ion source for production of a broad range of fission products, his version of ion source has an inherently low efficiency which is unsuitable to our requirements. The use of an electron bombardment heated cathode, similar to that of Mazumdar et al.⁶⁾ is a feature we hope to incorporate in our future work. This feature has the potential of very high temperature operation and long cathode lifetime.

Parallel to our ion source development effort has been the development of an aerosol-loaded gas-jet transport system. This effort centered on the use of solid aerosols. Solid aerosols, as compared to liquid aerosols, are potentially more compatible with the temperature and pressure gradients encountered in coupling to an ion source. In addition, the production of solid aerosols by the evaporation/sublimation method in a heated tube¹⁵⁾ are capable of stable, reproducible long-term operation. The use of NaCl aerosols produced in this way for efficient transport of radioactivity in a gas-jet system was first demonstrated by Ghiorso et al.¹⁶⁾.

The use of NaCl aerosols in gas-jet-coupled ion-source systems has been reported previously^{5,6,10}).

Presented in this paper are the main features and results of our experimental studies pertaining to the use of solid aerosols in a gas-jet-coupled ion source. The experimental setup we have used is described in detail in section 2. In section 3 we discuss the measurements we have made to optimize the transport of fission-product activities and the production of ion beams in a gas-jet-coupled ion source. A brief summary is given of our aerosol transport studies with an emphasis on those features which specifically relate to a capillary/skimmer/ion-source coupled system. A more extensive description is given in section 3 of the experiments we have done to measure elemental separation efficiencies for the gas-jet-coupled ion source as a function of several system parameters: aerosol type, support gas, arc power and anode construction materials. Conclusions drawn from this work are given in section 4.

2. Experimental Setup

A schematic illustration of the experimental arrangement for our studies is shown in fig. 1. Briefly, the system consists of a small ^{252}Cf fission source, an aerosol generator and gas-jet transport system, a skimmer chamber, a Roots pumping system, an ion source and a beam formation/collection system. No mass dispersion stage was used for mass analysis of the ion beam.

2.1 Fission Source

An approximately $7\text{ }\mu\text{g}$ ^{252}Cf oxide deposit on a platinum disc was the source of fission products for this work. The source, fabricated at the Oak Ridge National Laboratory, is mounted on the top cover plate of the aluminum source chamber. An approximately $1 \times 10^{-3}\text{ mg/cm}^2$ nickel foil is placed over the californium. The nickel window transmits the energetic fission fragments, but is a reasonable barrier to the low-energy recoil californium atoms. Alpha contamination of the source chamber and the remote collection apparatus is, therefore, minimized. The inner volume of the source chamber is approximately 300 cm^3 . For optimum thermalization of the fission fragments in this volume of helium the chamber is maintained at a pressure of approximately 2×10^5 Pascals (2 atm).

2.2 Gas-jet Transport Apparatus

The gas-jet transport apparatus consists of a regulated helium gas supply, an aerosol generator, connecting tubing, the source chamber, a teflon capillary, a skimmer chamber and a high capacity pumping system. A brief description of specific features of this system is described below.

In our work we have explored the use of solid aerosols of NaCl, AgCl, Ag, Cd, Te produced by evaporation/sublimation of the respective materials in a mullite* ceramic tube heated with a tube furnace. NaCl aerosols produced at a temperature of 600°C were used for the majority of the ion-source tests reported here. Ag aerosols produced at a temperature of 1150°C were used for a few tests.

A 23 m delay line connected the aerosol generator to the source chamber. The purpose of this approximately 0.6 cm ID PVC line was to tailor the aerosol size distribution which enters the source chamber. The delay line effectively removes aerosols greater than 5×10^{-6} m and less than 5×10^{-9} m in size. Aerosols in this size range are efficiently transported through the Teflon capillary tube which connects the source chamber to the skimmer chamber. The approximately 5 m long transport capillary has a 0.86 mm ID and a 1.42 mm OD.

The skimmer chamber was built with features which enabled it to be used for aerosol transport studies, gas-jet-coupled ion source test operation and future installation on the mass separator. Axial alignment of the capillary with respect to the skimmers was achieved by means of the X-Y translation stage which supported a sliding shaft feedthrough. The distance from the exit end of the capillary to the first skimmer was controlled by the Z translator which was attached to the steel capillary holder. The steel capillary holder was radially constrained by the shaft feedthrough. A gate valve in the skimmer chamber wall common to the

* The chemical composition of mullite is $3 \text{ Al}_2\text{O}_3 \times 2 \text{ Si O}_2$. Tubes were obtained from McDanel Refractory Porcelain Co., Beaver Falls, Penn. 15010.

first skimmer could be closed to isolate the ion source discharge chamber from the skimmer chamber. A foil collector and vacuum interlock were incorporated in the skimmer chamber to permit direct collection of the aerosol and attached radioactivity in front of skimmer number 1. This capability provided the flexibility necessary for both transport efficiency measurements and gas-jet-coupled ion source separation efficiency experiments. Helium gas expanding from the capillary into the skimmer chamber was pumped away with a ROOTS* pumping system.

2.3 Ion Source/Skimmer Arrangement

Two versions of a Sidenius hollow-cathode ion source¹⁴⁾ were used in our studies. The first version was a direct utilization of a DANFYSIK model 911A hollow-cathode ion source and associated source magnet housing together with a single skimmer (orifice diameter of 1.5 mm). For this adaptation the evaporator oven and gas injector tube of the commercial ion source were replaced by an anode extension tube of tantalum. In addition we used a specially fabricated base flange which included radial ports for installation of electrical feedthroughs and radial ports which permitted both the introduction of support gas into the hollow anode and the measurement of the pressure in that region. The distance from the skimmer to the discharge region in this early version was approximately 10.4 cm.

The second version of a gas-jet-coupled ion source, which we used for the majority of our tests, is illustrated in fig. 2. This version incorporated two flat-plate skimmers and significant modifications to

* Leybold-Heraeus pumps; WS-1000, DK-200.

or replacement of many of the components of the DANFYSIK ion source. The filament capsule assembly and the terminal tube from the commercial source were retained. To better match the divergence properties of the aerosol stream transmitted through the skimmers, the anode piece and extension tube were replaced by a shortened component. Internal radial dimensions of the anode were made larger than those in the commercial source. These dimensions were set to transmit approximately 45% of the aerosols from a stream with a 3° half-angle divergence at the capillary into the discharge region of the ion source. In addition, the projection of the anode in the discharge region was changed to essentially duplicate that in fig. 2 of ref. 14. A longer anode insulator was used. This modification helped to minimize electrical breakdown of the anode insulator because of conductive plateout. The ion source standoff support was also shortened to accommodate the shortened anode structure. Also, the source magnet vacuum housing was replaced by a shortened version as shown in fig. 2. An external magnet was used to cancel the magnetic field due to current flowing in the filament. Support gas was fed directly into the hollow anode as shown in fig. 2.

Two flat-plate skimmers were positioned in the base flange as shown in fig. 2. Both skimmers were 1 mm thick and they were separated by a distance of 14.6 mm. Orifice dimensions were 1.5 mm and 3.0 mm, respectively, for the first and second skimmers. The distance from the first skimmer to the discharge region was approximately 5.5 cm in this shortened version of the ion source.

The interskimmer region could be pumped on through the bypass line and the pressure could be monitored in this region. Although the bypass arrangement did not allow true differential pumping across the first skimmer, it did enable us to control the pressure in the interskimmer region and in the ion source for small capillary/first skimmer separations (< 5 mm). Earlier tests with the single skimmer/ion-source arrangement without bypass pumping indicated significant pressure buildup in the ion source for capillary/skimmer separations of < 15 mm. The bypass arrangement permitted us to support the ion-source discharge on gas fed through the support line (bypass fully open) or in helium transmitted through the skimmers (bypass partially open).

The selection of flat plate skimmers was based on tests made with a gas-jet system at Chalk River¹⁷⁾. We have also made calculations to verify that turbulence in the vicinity of a flat skimmer negligibly affects the heavy aerosol stream passing through the skimmer.

The use of a side-jet¹⁷⁾ to improve the pressure differential across a single skimmer was also tested. The pressure reduction achieved was not sufficient to warrant its use considering the additional gas load it placed on our pumping system.

2.4 Beam-Formation and Collection System

The ion-beam formation system, illustrated in fig. 1, extracts ions from the ion source at ground potential and accelerates and focuses them to a collector held at a negative voltage. In contrast to this, with the mass separator, ions are extracted from an ion source at a high positive voltage and accelerated to ground potential. However, the experimental arrangement of fig. 1 reproduces the extraction field

of the mass separator because the extractor cone is identical for both. Hence tests made with the arrangement of fig. 1 should be a good indication of performance of the gas-jet-coupled ion source when it is installed on the mass separator.

Beam focus is achieved by use of a three-electrode cylindrical Einzel lens. The extractor cone is physically attached to the first cylindrical element. The design of the Einzel lens is based on that used by Scaife et al.¹⁸⁾ in a focused ion-beam sputtering system. The 5.1 cm long focus element has an internal diameter of 6.1 cm and a gap separation of 0.64 cm from the adjacent cylindrical elements. Object and image distances to the focus element midplane were 16.5 cm and approximately 22.0 cm, respectively.

Operational properties of the lens were determined from the diagrams given in ref. 19. Typical operational parameters for the extractor/lens system were -12.0 kV on the extractor element and on the element closest to the collector with -2.0 kV on the focus element.

A cylindrical pocket collector, operated at -2 kV, is used to stop and collect the focused ion beam. The collector is approximately 3 cm deep and has a 1.6 cm inner diameter. Reasonable ion-beam current measurements are possible with the collector. More importantly, it allows the collection of both the direct impinging ions and the sputtered atoms on aluminum foil which lines the collector pocket. Consideration of the collection of sputtered atoms that have been previously deposited by direct collection is important when operating with total ion beam currents of approximately 200 μ A and for collection times of approximately 30 minutes. Both parameters are representative of our experimental conditions.

3. Measurements and Results

3.1 Aerosol Transport Studies

We have conducted several experiments related to solid aerosol production, aerosol characterization, and aerosol transport through a capillary and a capillary/skimmer arrangement. The emphasis of these studies was to develop an aerosol-loaded gas-jet-transport system which was capable of stable, long-term operation, and of efficient transport through a long capillary into a skimmer/ion-source arrangement. Only those aspects of these studies which are most related to optimizing the net efficiency of a gas-jet-coupled ion source are presented here.

Using only the gas-jet transport part of the system illustrated in fig. 1, we have measured the collection efficiencies for transport of fission-product activity from the ^{252}Cf source to the pre-skimmer collector. These measurements were made as a function of tube-furnace temperature and helium flow rate through the tube for aerosols of NaCl, Ag, AgCl, Cd and Te. Each collection efficiency value was based on a comparison of the gross beta-gamma activity measured for a foil placed in the aerosol stream exiting the capillary to the activity deposited on a foil placed directly over the ^{252}Cf fission source. Gross beta-gamma activity measurements were made with a gas-filled ionization chamber. Maximum collection efficiencies of approximately 70% for NaCl, approximately 60% for AgCl, Ag and Te and approximately 40% for Cd were obtained. The actual activity transport efficiencies are larger than these collection efficiencies because no rare-gas activity is collected on the pre-skimmer collector foil, whereas, the foil placed above the fission source stops

and collects all of the fission fragments emitted by the source. Therefore, the transport efficiency for NaCl is estimated to be greater than 90% for a collection efficiency of 70%.

Measurements of the temperature and flow-rate dependence of the collection efficiencies were made to determine the conditions for optimum activity transport and minimal aerosol mass transport. For the NaCl and Ag aerosols used in the present gas-jet-coupled ion source experiments these conditions were: aerosol production temperature (600°C for NaCl, 1150°C for Ag), helium flow rate (approximately 40 cm³/s at STP), and a ²⁵²Cf source chamber pressure of approximately 2 x 10⁵ Pascals (2 atm). At these operating conditions the activity transport was stable and reproducible for the series of experiments with the ion source.

Consideration of the angular divergence of the aerosol stream as it exits from the capillary is of key importance to optimizing transmission of the aerosol stream through the skimmers into the ion-source discharge region. A divergence angle of approximately 3° was determined for NaCl aerosols exiting in a helium gas stream from a 0.86 mm ID capillary. This angle was derived from measurements of the diameter of the aerosol deposit on a collection foil as a function of distance of the foil from the capillary. Measurements which were made of the angular divergence of the aerosol stream transmitted through the first skimmer also indicated an angular divergence of approximately 3° half-angle. The design of the capillary/double-skimmer/ion-source arrangement shown in fig. 2 is based on 100% transmission of a 3° aerosol stream through both skimmers into the rear of the anode piece.

3.2 Separation-Efficiency Experiments

We have conducted a series of experiments to measure the separation efficiencies for both versions of the gas-jet-coupled ion source. For a typical experiment, the determination of the separation efficiency entailed the following:

- (1) establishing the conditions for optimum activity transport through the capillary,
- (2) collecting aerosols and activity on the pre-skimmer collector (see fig. 1),
- (3) measuring the activity with a gas-filled ionization chamber or with a Ge(Li) detector,
- (4) establishing stable operating conditions for the gas-jet-coupled ion source,
- (5) collecting all ion beams formed in the ion-beam collector (see fig. 1)
- (6) measuring [in the same geometry as (3)] the activity with a gas-filled ionization chamber or with a Ge(Li) detector.

Separation efficiency is defined here as the relative activity measured on the ion-beam collector foil divided by the relative activity measured on the pre-skimmer collector foil. Defined in this way, the separation efficiency includes the effects of losses in the skimmer arrangement, in the ion source and in the beam formation system.

Only a few separation efficiency experiments were done with the first version of the gas-jet-coupled ion source. These experiments were characterized by the use of NaCl aerosols, an argon support gas and the measurement of the gross beta-gamma activity with a gas-filled ionization chamber. A gross fission-product separation efficiency of $< 0.01\%$ was estimated for this version of the ion-source system.

An extensive series of tests were made with the second version of the gas-jet coupled ion source (see fig. 2). The objective of these experiments was to measure elemental separation efficiencies as a function of several parameters: (1) aerosol type (NaCl, Ag), (2) support gas (He, Ar, CCl_4), (3) arc power and (4) anode material (Ta, Mo). These experiments were characterized by the use of a calibrated Ge(Li) detector in the determination of the relative isotopic activities deposited on the pre-skimmer and ion-beam collector foils. Elemental separation efficiencies were subsequently obtained from the relative isotopic activities.

An example of the system operating conditions for one experiment in this series is given in Table 1. The quoted parameter values for the helium flow rate, for the various chamber pressures and for the filament power are representative of all of the experiments. Experiments with NaCl and Ag were also run for a capillary/first skimmer separation of 7 mm. With the capillary at the 7 mm position the interskimmer pressure was reduced to approximately 29 Pascals (220 mT). For some experiments the arc discharge was supported only by the He gas delivered to the anode region via the capillary skimmer coupling. This was accomplished by partially closing the bypass valve to raise the interskimmer region pressure to 47-57 Pascals (360-440 mT).

A typical elemental efficiency experiment was set up to optimize detection of fission-product activities with half-lives of approximately 10 minutes. This was done by collecting the activity on the pre-skimmer and ion-beam collector foils for 30 min., waiting for 5 min., and then measuring gamma-ray spectra from the sources for three sequential 10 min. time periods. The gamma-ray spectra were measured with a large volume, calibrated Ge(Li) detector with the source placed on the detector face. All sources were counted, therefore, in a fixed geometry.

Isotopic separation efficiencies were derived from the analyses of the gamma-ray spectra for the pre-skimmer and ion-beam collector foils. Peak areas and energies were obtained by fitting each gamma-ray peak with a single gaussian function using the GAUSS V algorithm²⁰⁾ implemented on an interactive computer. Identification of the presence of a specific fission-product activity was based principally on the correlation of up to three observed peak energies to corresponding known energies^{21,22)} of the most intense lines from a fission-product with a half-life compatible with the collection/count time sequence. An isotopic separation efficiency based on a single gamma-ray energy was computed, therefore, as the ratio of the peak area for the ion beam foil spectrum, to the corresponding peak area for the pre-skimmer foil spectrum. The "best estimate" of an isotopic separation efficiency was obtained as the average of up to three of the single gamma-ray based quantities for a specific radionuclide. Similarly, estimates of the elemental separation efficiencies were obtained by averaging the "best-estimate" isotopic efficiencies where possible.

Summarized in Table 2 is a list of the fission-product isotopes whose activities were identified in the deposits on the ion-beam collector foil. Identification of the rare-earth activities was confirmed by experiments in which gamma-ray spectra were measured for chemically purified sources. The chemically purified sources were prepared by using standard chemical techniques to isolate the rare-earth fraction from the gross fission-product activities deposited on the ion-beam collector foil.

The column labelled "element separated" includes reasonable guesses of the elemental ion beams producing the observed activities. An isotope with a precursor half-life less than approximately 10 s was considered to be ionized. This judgement is based on an estimated holdup time in the ^{252}Cf source chamber of approximately 15 s, a time period sufficient for decay of many of the short-lived precursor activities in the source chamber. Only the longer-lived activities are transported into the ion source. For example, 10.7 min ^{142}Ba which has 1.7 s ^{142}Cs as a precursor, is certainly formed as a primary ion beam with the system. However, 18.0 min ^{104}Tc which has 1.6 min. ^{104}Mo as its precursor, could have been delivered to the beam collector as an ion beam or it could have been produced at the collector by decay of ^{104}Mo which could have been formed as an ion beam.

In addition, because no mass dispersion was used, we did not have sufficient information to determine whether the activity collected as an ion beam was either singly or multiply-charged. Similarly we could not distinguish between single atom or molecular ion beams. Experiments

are planned in which such a decomposition will be determined with our mass separator.

In Table 3 we present estimates of absolute elemental separation efficiencies determined from a selected set of experiments with the gas-jet-coupled ion source. Estimated uncertainties in these quantities range from approximately 30% for the Cs and Ba elements to approximately 50% for the remaining elements. These results are given to indicate the general magnitude of separation efficiencies observed in most experiments under a variety of operating conditions.

The dependence of the elemental separation efficiency on the arc power was investigated for both the Ta anode and the Mo anode cases with the arc supported by argon and activity transported on NaCl aerosols. No observable change in separating efficiencies was noted in the experiments with the Ta anode when the arc power was varied from 70 watts to 180 watts. However, as indicated in Table 3 for the Mo anode case an increase was observed in the separation efficiencies for Ba, Ce, Ce/Pr, Pr and Nd as the arc power level was increased from 60 watts to 170 watts. The increase in arc power results in a higher temperature anode surface. Hence the increased separation efficiencies could be indicative of the role of surface ionization in the formation of ion beams of these elements. It is puzzling that the efficiency for Cs did not also increase. However, of the group of elements given in the table, Cs has the lowest ionization potential and the probability for its ionization by surface ionization could already be maximum at the lower power operation.

We also investigated the use of different support gases, namely He, Ar and CCl_4 , to maintain the arc. When Ar is used to support the discharge, the dominant mechanism for ionization of atoms is collisions with electrons in the discharge. When He is used as the support gas Penning ionization²³⁾ could become an important mechanism for ionization of the neutral atoms. The use of CCl_4 as a support gas introduces the possibility of enhancing the ion-beam production of the rare earths and the more refractory elements such as Zr, Mo and Tc by chemical synthesis in the ion source²³⁾.

Our experiments with He as a support gas as compared to those with Ar indicated essentially no change in separation efficiency for all isotopes listed in Table 2.

However, substantial increases in the separation efficiencies of the rare-earth and alkaline-earth elements was observed when CCl_4 was used as the support gas. A comparison of the absolute elemental separation efficiencies obtained from the Ar and CCl_4 experiments is given in Table 3. An indication of the chemical enhancement of the separation efficiencies by use of CCl_4 is given in Table 4. The observed enhancement is particularly significant for the experiments with the Mo anode. Because of the difference in the arc power between the Ta and Mo anode cases, it is difficult to determine whether the higher chemical enhancements for the Mo case are attributable to more effective chemical synthesis or to differences in physical properties of Ta and Mo. Also, because of the inherent limitation of no mass analysis with our experimental test arrangement, we could not determine whether the increased

yield with CCl_4 is due to formation of molecular or single-atom ion beams. Experiments will be conducted in the future to sort out these effects.

The dependence of the elemental separation efficiencies on different aerosols in the gas-transport system was investigated in experiments with NaCl and Ag aerosols. The primary objective of these experiments was to determine if the Cl associated with the NaCl aerosol was a contributing factor to the production of alkaline-earth and rare-earth ion beams by chemical synthesis in the ion source. The results of our comparative experiments indicated no significant difference in separation efficiencies for NaCl and Ag. We suspect this observation to be misleading because the experiments with the Ag aerosol were conducted after those with NaCl. Residual buildup of NaCl in the system components could have influenced the Ag experiments outcome.

Our separation efficiency experiments were conducted with both Ta and Mo anodes. As discussed earlier, an increase in the separation efficiencies for Ba, Ce, Ce/Pr, Pr and Nd as a function of arc power was observed for the Mo anode case but not the Ta anode. Separation efficiencies for these elements at a nominal power of 170-180 watts were, however, nearly identical for both anode materials (see Table 3). In addition, a more significant chemical enhancement of the separation efficiencies by use of CCl_4 was observed in experiments with a Mo anode than in those with a Ta anode. The absolute separation efficiencies for both CCl_4 experiments were, however, comparable (see Table 3). Clearly, some interesting trends are observed. The experiments we

conducted were not extensive enough to elucidate the unique benefits of using either anode construction material, but do illustrate the need for additional work.

4. Conclusions

In this paper we have reported on our efforts to develop a gas-jet-coupled ion source for on-line isotope separation. We have shown that solid aerosols of NaCl and Ag can be used to transport, with high efficiency, fission products from a ^{252}Cf fission source into a skimmer/ion source arrangement. We have demonstrated that flat-plate skimmers are suitable for efficient transmission of the aerosols and fission products into the ion-source discharge chamber. However, on the basis of angular divergence measurements for aerosols exiting from the capillary and a comparison of the results of separation efficiency experiments with a long and short anode configuration, we conclude that it is important to match the skimmer-anode geometrical configuration to aerosol divergence. We have demonstrated that a Sidenius hollow-cathode ion source can be used in a gas-jet-coupled configuration for production of ion beams of Sr, Y, Tc, Te, Cs, Ba, Ce, Pr, Nd and Sm with separation efficiencies ranging from 0.1% to greater than 1%.

We have investigated the dependence of the separation efficiencies on several system parameters: aerosol type, support gas, arc power and anode construction materials. No conclusive results could be drawn from a comparison of experiments with NaCl and Ag aerosols. A dramatic enhancement in yield of alkaline-earth and rare-earth elements was observed when CCl_4 was introduced as a support gas. We conclude that

chemical synthesis in the ion source is a promising approach to producing ion beams of these elements and should be investigated further. Our experiments with Ta and Mo anodes indicated a stronger arc-power and chemical-synthesis dependence of the rare-earth and alkaline-earth separation efficiencies for Mo than for Ta. Our experiments were not extensive enough to elucidate the ionization mechanisms involved or the benefits of using either material for an anode. Further investigation is required in this area.

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TABLE 1

Typical parameters for operation of gas-jet-coupled ion source with Ta anode and NaCl aerosols

Capillary/First Skimmer Separation	- 4 mm
Helium Flow rate	- 40 cc/s STP
^{252}Cf Source-chamber pressure	- 2×10^5 Pascals (2 atm)
Skimmer chamber pressure	- 20 Pascals (150 mT)
Interskimmer region pressure	- 39 Pascals (300 mT)
Beam-formation chamber pressure	- 1.3×10^{-4} to 4×10^{-4} Pascals (10^{-6} to 3×10^{-6} mT)
Ion-source support gas	- Argon
Ion-source filament power	- 100 watts (20A/5V)
Ion-source anode power	- 170 watts (2A/85V)
Total ion-beam current	- 180 μA

TABLE 2

Isotopes identified in He-jet-coupled ion source separation experiments

Isotope	Half-life	Element Separated	Isotope	Half-life	Element Separated
^{92}Sr	2.7 h	Sr	$^{141}\text{Ba}^*$	18.3 m	Cs/Ba
$^{93}\text{Sr}^*$	7.5 m	Sr	$^{142}\text{Ba}^*$	10.7 m	Ba
$^{94}\text{Y}^*$	19. m	Sr/Y	$^{142}\text{La}^*$	92.4 m	Ba
^{101}Mo	14.6 m	Mo	$^{146}\text{Ce}^*$	14.2 m	Ce
^{101}Tc	14.2 m	Mo	$^{146}\text{Pr}^*$	24.2 m	Ce
$^{104}\text{Tc}^*$	18.0 m	Mo/Tc	$^{147}\text{Pr}^*$	12. m	Ce/Pr
$^{105}\text{Tc}^*$	8.0 m	Mo/Tc	$^{149}\text{Pr}^*$	2.3 m	Pr
$^{133\text{m}}\text{Te}^*$	55.4 m	Sn/Sb/Te	^{149}Nd	1.73 h	Pr/Nd
$^{133}\text{Te}^*$	12.5 m	Sn/Sb/Te	$^{151}\text{Nd}^*$	12.4 m	Nd
$^{134}\text{Te}^*$	42. m	Te	$^{152}\text{Nd}^*$	11.5 m	Nd
$^{134}\text{I}^*$	52.6 m	Te/I	^{152}Pm	4.1 m	Nd/Pm
^{138}Xe	14.2 m	Xe/Cs	^{153}Pm	5.4 m	Pm
$^{138}\text{Cs}^*$	32.2 m	Xe/Cs	$^{155}\text{Sm}^*$	22.2 m	Sm
$^{139}\text{Cs}^*$	9.3 m	Xe/Cs	^{157}Sm	83. s	Sm

* Identification of these activities is based on the presence of at least 2-3 of the radionuclide's most intense gamma rays in the collector foil spectra and, for the rare earths, to the presence of these gammas in the radiochemically-separated sources. Identification of the remaining isotopes is tentative.

TABLE 3

Absolute elemental separation efficiencies for He-jet-coupled ion source operated with Ta and Mo anodes, Ar and CCl_4 support gas and NaCl aerosols

Element Separated	Ta Anode Ar ^a	CCl_4 ^b	Ar-1 ^c	Mo Anode Ar-2 ^d	CCl_4 ^e
Sr	0.5	0.8	0.1	0.6	0.3
Sr/Y	0.9	1.4	---	---	1.0
Mo/Tc	0.4	0.3	0.2	0.4	0.2
Te	0.6	1.0	0.5	0.5	1.2
Cs	4.0	3.0	2.5	2.5	1.5
Ba	0.9	1.9	0.3	0.6	1.7
Ce	0.5	1.5	0.2	0.4	1.2
Ce/Pr	0.5	1.6	0.2	0.5	1.4
Pr	0.9	1.0	0.1	0.2	0.9
Nd	0.5	1.1	0.2	0.6	1.6
Sm	0.8	1.9	---	---	1.5

^a Arc power of 150 watts

^b Arc power of 130 watts

^c Arc power of 60 watts

^d Arc power of 170 watts

^e Arc power of 70 watts

TABLE 4

Chemical enhancement of the He-jet-coupled ion source separation efficiencies by use of CCl_4 support gas as compared to use of Ar support gas

Element Separated	Enhancement factor - CCl_4 :Ar	
	Ta Anode ^a	Mo Anode ^b
Sr	>1	3
Te	2	2
Ba	2	6
Ce	3	6
Ce/Pr	3	7
Pr	>1	9
Nd	2	8
Sm	2	>5

^a Arc power of 125-150 watts

^b Arc power of 60-70 watts

Figure Captions

1. Experimental arrangement used to test the gas-jet-coupled ion source system.
2. Detailed drawing of the skimmer/ion-source configuration for the gas-jet-coupled ion source.

ION SOURCE TEST SYSTEM

Figure 1. Experimental arrangement used to test the gas-jet-coupled ion source system.

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GAS-JET-COUPLED ION SOURCE

Figure 2. Detailed drawing of the skimmer/ion-source configuration for the gas-jet-coupled ion source.