

N. Trautmann

Institut für Kernchemie der Universität Mainz, D-65 Mainz, Germany

Abstract

A survey is given on the progress of fast chemical separation procedures during the last few years. Fast, discontinuous separation techniques are illustrated by a procedure for niobium. The use of such techniques for the chemical characterization of the heaviest known elements is described. Other rapid separation methods from aqueous solutions are summarized. The application of the high speed liquid chromatography to the separation of chemically similar elements is outlined. The use of the gas jet recoil transport method for nuclear reaction products and its combination with a continuous solvent extraction technique and with a thermochromatographic separation is presented. Different separation methods in the gas phase are briefly discussed and the attachment of a thermochromatographic technique to an on-line mass separator is shown.

1. Introduction

Among various techniques rapid chemical separations are a useful tool in studying short-lived nuclei. The requirements on the rapid separations depend considerably on the particular case. For the detection of a nuclide through a characteristic radiation, a rather unselective procedure may be sufficient. On the other hand, for detailed decay studies even a very selective chemical separation may not be sufficient due to interferences from accompanying isotopes. Here the difficulty can be overcome by combining a chemical step with a mass separation.

An important aspect which must be taken into account in working out chemical separation methods is continuous or discontinuous performance. As long as a relatively small number of experiments give the desired information discontinuous procedures are to be preferred. When such an experiment would have to be repeated too often, a continuous process delivering a steady source of short-lived activity is more appropriate, but such procedures are, in general, harder to accomplish.

In the following survey some fast chemical methods will be described which are representative of the progress in this field, and their use will be illustrated by a few examples. Of course, it is not feasible to cover all the brilliant techniques that have been developed during the past few years in a paper such as this.

Those who want to have a more comprehensive overview are referred to review articles on this subject^{1,2,3,4}).

For the convenience of the author most of the examples, but not all, have been selected from work done at the Institut für Kernchemie in Mainz. Since a TRIGA pulsing reactor is the major facility of this institute the application of rapid chemical separations is mainly demonstrated for the study of neutron-rich nuclei. However, the techniques are also applicable directly or after slight modifications to other regions of the periodic table.

2. Discontinuous separation procedures

Most of the discontinuous separations are performed from aqueous solutions by transferring either the desired or the accompanying elements into a second phase which can be gaseous, liquid or solid. The advantages of wet chemistry are the availability of a large variety of well-known classical procedures. Whether such a method can be accomplished on a short time scale depend on a fast completion of the chemical reactions involved and on techniques enabling fast phase separations. The transfer into a solid phase can be considerably accelerated by replacing the time-consuming precipitation step by a heterogeneous exchange reaction⁵). In solvent extraction the transport from one liquid phase into another may be fast in most cases, but the separation of phases is the slowest step. This can be overcome by using either H-centrifuges⁶) or layers of quasi-solid solvents prepared by fixing the organic phase on fine-grained plastic carriers through which the solution is filtered quickly⁷). The transport from a liquid into a gaseous phase can be performed very rapidly with high yields; as it was demonstrated, for instance, for the volatilization of hydrides by nascent hydrogen⁸).

Normally, a combination of several separation steps based on such techniques is required for the selective isolation of an element. An example for this is the separation of niobium from fission product mixtures by its sorption on glass from strong nitric acid⁹). Figure 1 shows the apparatus and the time schedule for this procedure.

A solution of ²³⁵U or ²³⁹Pu in 0.1 N nitric acid containing SO₂ for the reduction of the halogens and tartaric acid for complexing antimony is sealed in a polystyrene capsule and irradiated with a neutron pulse. After the pulse the capsule is transported in a pneumatic system to the apparatus where it is smashed by impact on the wall. The transit time through the 5 m pipe is about 100 msec. The halogens and silver are removed from the solution by exchange with two preformed silver chloride layers. Then the layers are washed with dilute nitric acid. The filtrates are collected in concentrated nitric acid and the resulting solution is passed through two fiber glass filters which adsorb niobium, presumably in a cation exchange reaction. After washing with 10 N nitric acid the filters are transferred by pressure to Ge(Li)-detectors placed in a shielded position. Counting of the sample is started 2.2 sec after the end of irradiation. With the time schedule given in Figure 1 a niobium yield of about 25 % has been achieved.

This separation method in combination with high resolution γ -ray spectroscopy enabled the identification of neutron-rich nuclides of niobium⁹) up to 0.8 sec ¹⁰⁴Nb.

Separations on a time scale of seconds cannot be performed by hand but have to be operated automatically. This is done by an electronic programmer which controls the whole sequence and from which signals at predetermined times are sent to magnetic valves delivering, for instance, pressure to small pistons moving teflon stopcocks.

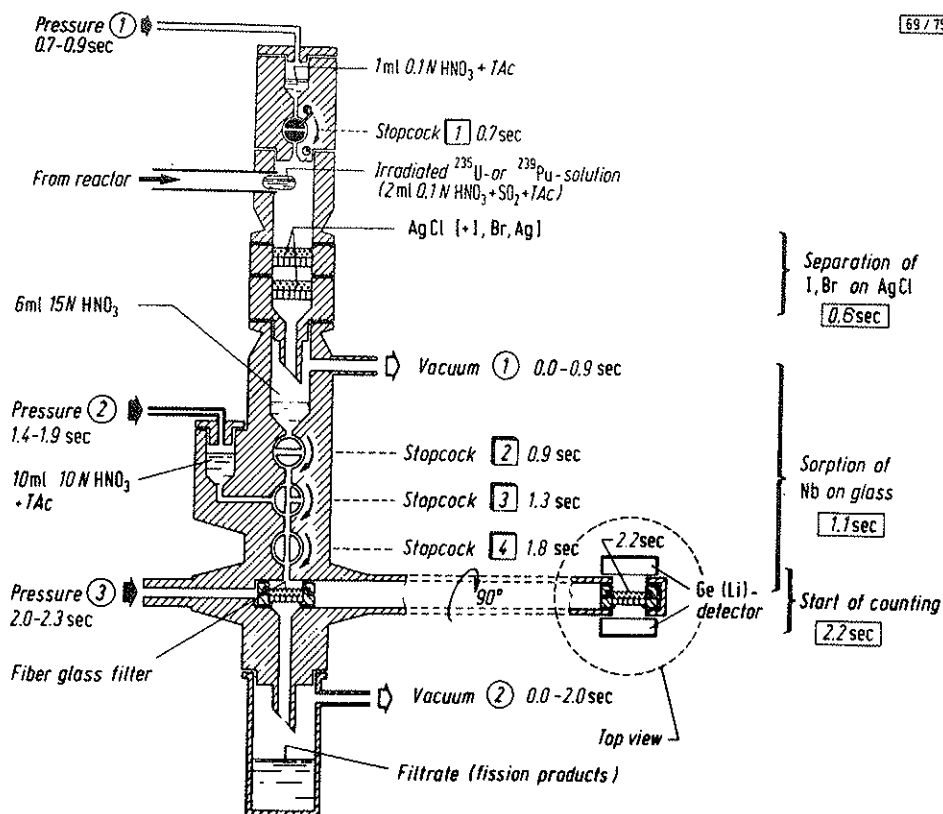


Figure 1.

Apparatus and time schedule for the rapid separation of niobium from fission products⁹⁾. At the right-hand side the mean-time for each separation step is indicated

Table 1

Rapid discontinuous separation procedures from aqueous solutions

Technique	Element [production]	Procedure	Nuclide/Half-life	Reference
Solvent extraction	Zr [U+n]	TBP/7.5 N HNO ₃	¹⁰¹ Zr 2.0 sec	(10)
	Mo [U, Pu+n]	Amyl alcohol/NH ₄ SCN	¹⁰⁷ Mo 3.5 sec	(11)
	Tc [Pu, Cf+n]	AsPH ₄ Cl/0.1 N HNO ₃	¹¹⁰ Tc 1.0 sec	(12)
	Ru [Pu, Cf+n]	Petrolether/5 N HClO ₄	¹¹² Ru 3.6 sec	(13)
Ion exchange resins	Y [U+n]	Cation resin/1 M α-HIB	⁹⁷ Y 1.5 sec	(14)
	Ce [U+n]	Anion resin/PbO ₂ /9 N HNO ₃	¹⁵⁰ Ce 3.4 sec	(15)
Sorption	Nb [Pu, Cf+n]	Glass/10 N HNO ₃	¹⁰⁴ Nb 0.8 sec	(9)
Exchange with solids	Ag [Cf+n]	AgCl/Ag ⁺	¹¹⁸ Ag 4.0 sec	(16)
	I [U+n]	AgI/I ⁻	¹⁴⁰ I 0.8 sec	(17)
Volatilization	As [U+n]	AsH ₃ from HCl+Zn	⁸⁶ As 0.9 sec	(18)
	Se [U+n]	SeH ₂ from HCl+Zn	⁸⁸ Se 1.4 sec	(19)
	Sb [U, Pu+n]	SbH ₃ from HCl+Zn	¹³⁶ Sb 0.8 sec	(20)
	Te [U, Pu+n]	TeH ₂ from HCl+Zn	¹³⁷ Te 3.5 sec	(21)
	Sn [U, Pu+n]	SnH ₄ from HCl+NaBH ₄	¹³² Sn 39 sec	(22)

Separation procedures for other elements in addition to niobium and using the same experimental arrangement as described above have been applied to a number of short-lived fission products by the Mainz group. In Table 1 some of the rapid separations are summarized.

The first column gives the technique, the second one the element and the production mode and the third, the separation procedure. To indicate the efficiency of a particular technique, the shortest-lived nuclide, observed so far, is listed in the next column.

By using solvent extraction procedures zirconium, molybdenum, technetium and ruthenium have been separated from complex fission product mixtures within a few seconds.

Yttrium and cerium can be isolated from the other fission products on ion exchange layers; the former on a cation resin and the latter on an anion resin.

The separation of niobium by sorption on glass has been described in detail. Isotopic exchange has been used for the isolation of silver as well as for iodine.

The volatilization of hydrides by nascent hydrogen generated by zinc powder in concentrated hydrochloric acid or sodium borohydride in HCl is an excellent technique for the separation of arsenic, selenium, antimony, tellurium and tin.

A more sophisticated automated chemistry apparatus was built by Ghiorso et al. for the chemical characterization of the heaviest known elements and applied to the solution chemistry of element 104 by a Livermore-Berkeley group²³).

For the studies of the chemical properties of element 104 the isotope 65 sec $^{261}104$ produced at the Berkeley Super HILAC by the reaction $^{248}\text{Cm}(^{10}\text{B},5n)$ was used.

Figure 2 shows the experimental arrangement. The recoil products attached on NaCl-aerosols in He-gas are deposited on polypropylene rabbits (2) which are pneumatically delivered (3) to a turntable (4). The rabbits are either shuttled to a second turntable (4a) for direct α -counting or moved to a station (5a) where the recoil products are dissolved. The solution is put on the top of the column (6) consisting of 0.25 F trioctylmethylammonium chloride (Aliquat-336) in ortho-xylene and supported on an inactive fluorocarbon powder. Actinides, lanthanides and yttrium as well as the alkali and alkaline earth elements are eluted from the column with 12 M HCl due to the weak complex formation while Zr, Hf and presumably element 104 form strong anionic chloride complexes in concentrated hydrochloric acid and are maintained on the column. These elements can be subsequently eluted with 6 M HCl. The eluted fractions are dried (7) and their α -ray spectra are measured with 50 mm² surface-barrier detectors (9). The time, energy and sample identification of each α -event is stored on magnetic tape. Counting is started about 2-3 min after the end of irradiation. Because only a few atoms are produced per bombardment it is necessary to repeat the separations many times. For this reason, the experimental apparatus and data storage are fully automated and controlled by a PDP-9 or -15 computer, allowing a cyclic discontinuous running of the system. In preliminary experiments six events attributable to $^{261}104$ decay were found. Their distribution indicates that the chemical behaviour of element 104 is much more like that of hafnium and different from that of heavy actinide elements.

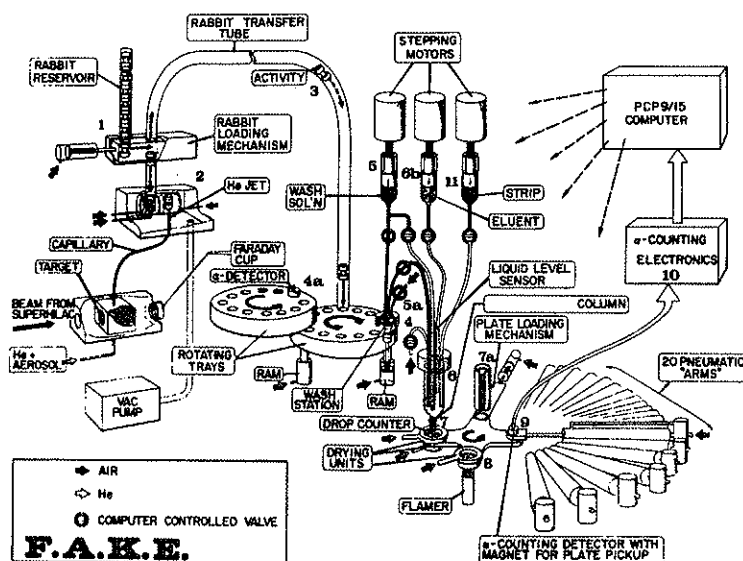


Figure 2.

Automated chemistry apparatus for solution chemistry of element 104²³)

Another problem is the fast separation of chemically very similar elements like the lanthanides or the heavy actinides. This may be solved with the high speed liquid column chromatography, a technique that rivals gas chromatography in speed and elegance and which has been applied heretofore mostly to organic compounds. Recent developments have been directed towards its use in radiochemical separations^{24,25}).

The improvement in the performance of high speed liquid chromatography can be largely attributed to the availability of high efficiency column packings consisting either of pellicular materials, 30 μm in diameter, with a solid glass core and a thin crust or of small porous particles with diameters of 5-10 μm . The high speed liquid chromatography is not limited to adsorption but can also be extended to ion exchange or solvent extraction. The application of the latter to the fast separation of some lanthanides and yttrium²⁶) is shown in Figure 3.

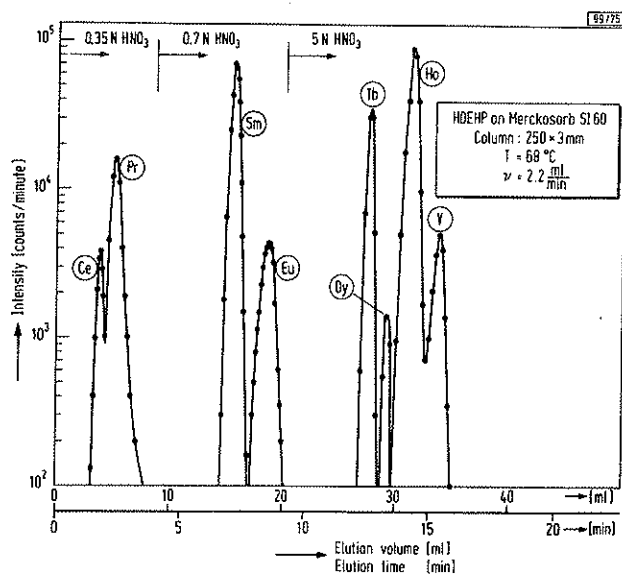


Figure 3.

Separation of several lanthanides and yttrium by means of high speed liquid chromatography using di(2-ethylhexyl)-orthophosphoric acid (HDEHP) as stationary and nitric acid as mobile phase²⁶

Di(2-ethylhexyl)-orthophosphoric acid (HDEHP) adsorbed on silica gel particles with a diameter of 10 μm is used as stationary phase in a column of 250 mm length and 3 mm diameter and nitric acid of different normality as the mobile phase. To decrease the time required for a complete separation the properties of the mobile phase are changed with time so that the distribution coefficients of the more strongly sorbed species are reduced during the later stages of the separation; i.e. gradient elution is used. As it is seen from Figure 3 seven lanthanides and yttrium can be separated within 20 min and the gaps between the elution peaks are large enough for a complete separation of all the lanthanides with the described conditions. Compared to the hitherto fastest procedure this is an improvement of about one order of magnitude.

Also some attempts were made to isolate short-lived single lanthanides from fission product mixtures by combining the high speed liquid chromatography with an electrophoretic method²⁷) with which

the lanthanides as a group were separated from the remaining products. Without optimization of the technique this could be achieved in 3 min²⁸).

3. Continuous separation procedures

The main line of development in the field of rapid separations for detailed studies of short-lived nuclides is concentrated now on continuous separation procedures. In an on-line operation the target is permanently bombarded and the reaction products from the target are transported either to a detector or to a chemical system with subsequent radiochemical separations and measurements of the chemically purified reaction products. For the transport of nuclear reaction products one has to distinguish between volatile and non-volatile species. Volatile species may escape from a thick target by a diffusion-evaporation process or may be formed in an appropriate stopping gas in which nuclear reaction products recoiling out of a thin target are slowed down.

For non-volatile species the only way of getting the activity out of the target area is by nuclear recoil. The recoil atoms are thermalized in a non-reactive gas and are transported in a gas-jet to the desired position.

The gas jet recoil transport method has found wide application for rapid and efficient transportation of nuclear reaction products from the production site with its high radiation environment to a low back-ground area^{3,29}). It is generally recognized that large clusters are necessary to carry the radioactivity long distances through narrow capillaries. Mostly, helium is used as the carrier gas mixed with some additives to form clusters to which the recoil atoms are attached. In accelerator experiments the large cluster carriers are produced by the interaction of low-molecular weight impurities in the helium with the intense ionizing radiation of the beam. A high-yield gas jet transport system for fission products with the absence of a strong ionizing atmosphere can be achieved by adding liquids to the transport gas and then irradiating the mixture with an intense UV radiation or by employing gases from a gas reservoir at temperature and pressure above the critical point. Here the aerosols are formed while the gases expand in the throttle valves of the supply bottle.

Some of the various "recipes" given for an effective transport of fission products in the gas-jet are summarized in Table 2 and compared with the transport efficiency of pure helium. With pure helium gas no activity is transported. With the combined action of additives and UV-light a significant increase in transported activity could be observed in the order water, trichlorethylene, ethanol and carbon tetrachloride³⁰). Using a La Mer aerosol-generator for the production of oil clusters together with a He-jet transport system 70-90% overall efficiencies for fission products could be reached³¹). Successful transport of radioactivity was also achieved with carbon dioxide, ethane and especially with ethylene^{32,33}) yielding an efficiency of 70 %, even in mixtures with helium or nitrogen. Recently it has also been shown that massive clusters are not required in a He-jet system operated at liquid air temperatures³⁴). For most gas-jet systems only small differences in transport efficiency between chemical elements were found. However, there are some indications that in at least a few cases it may be possible to achieve a partial separation in the course of the thermal-

ization and transportation process leading to ultra-fast chemical separations³⁵).

Table 2

Average transport efficiencies of fission products for different gas-jet systems

Gas	Additive	Average transport efficiency (%)
"Pure" Helium	-	<0.1
Helium	Water	21
Helium	Trichlorethylene	29
Helium	Ethanol	32
Helium	Carbon tetrachloride	50
Helium	Oil	75

Carbon dioxide	-	30
Ethane	-	55
Ethylene	-	70
"Cold" Helium	-	40-70

A great deal of attention has been devoted to exploring the possibility of performing chemistry at the end of the gas-jet system. For combinations with chemical separation procedures the observation was important that the yields remained almost unchanged if the reaction products were collected on a foil in a vacuum chamber or if the low-pressure end of the capillary was kept at atmospheric pressure³⁶. The advantages of such a system are obvious. There is no need for any expensive pumping arrangement and the rapid chemical separations can be run completely on-line.

Continuous separation procedures from aqueous solutions have recently been accomplished with an on-line operating solvent extraction system³⁷) consisting of several H-centrifuges (SISAK-system). In these specially designed continuous flow centrifuges perfect phase separation takes place, i.e. no measurable amounts of one phase occur in the other. The combination of this new technique with a gas jet transport system³⁸) is outlined in Figure 4 for the isolation of trivalent lanthanides from fission products.

The fission fragments are produced by bombarding an ^{235}U target with thermal neutrons and are thermalized in a 1:1.4 mixture of ethylene and nitrogen. After the transport through a 7 m long, 1 mm i.d. polyethylene capillary the gas mixture enters a static mixer where it is mixed with nitric acid of pH 1.4 heated up to about 90°C to take advantage of the enhanced dissolution of the fission products from the clusters at higher temperatures. The gas-liquid mixture is fed into a degassing unit to remove noble gases together with the $\text{C}_2\text{H}_4\text{-N}_2$ carrier gas. In a second mixer the solution is contacted with a solution of HDEHP in Shellisol T. The trivalent lanthanides as well as other elements like Zr, Nb, Mo etc. are extracted into the organic phase. The phases are then separated in the first centrifuge (C1). By contacting the organic phase with an oxidizing solution La, Pr and other trivalent lanthanides are stripped from the organic phase whereas Ce as Ce(IV) and the other extracted elements remain in the HDEHP-solution. After phase separation in the second centrifuge (C2) the aqueous phase is pumped to the detector cell and the measurements are carried out directly on the liquid phase. The delay time between production and measurement is estimated to amount to about 10 sec caused by a hold-up time of 3 sec in each centrif-

uge and the delay in the gas-jet system and the tubes.

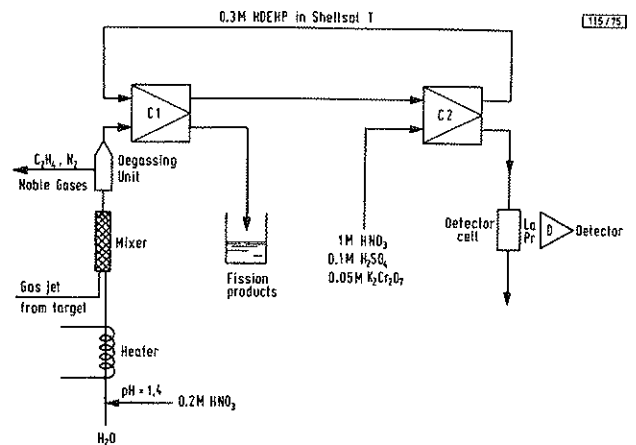


Figure 4.

Experimental set-up for the continuous separation of trivalent lanthanides from fission products by connecting a gas jet recoil technique with the fast chemical on-line system SISAK³⁹)

A γ -ray spectrum of the trivalent lanthanide fraction³⁹) is shown in Figure 5. This spectrum was measured on-line during 25 min. Most of the γ -rays can be attributed to neutron-rich lanthanum and praseodymium nuclides and their daughter products. The shortest-lived nuclide identified hereby is ^{150}Pr with a half-life of 6.2 sec.

For even shorter-lived activities the delay time of the SISAK-system has to be decreased. This can be achieved with an improved version of the H-centrifuge, the volume of which is reduced by one order of magnitude compared to the older one. The first runs with the new system, just performed, look very promising.

Continuous separation methods in the gas phase have been used for a long time by workers at Dubna for separating short-lived transactinide nuclides via volatile chlorides^{40,41}). The activities were produced using thin targets in a heavy ion cyclotron. The atoms recoiling from the target were collected in flowing nitrogen and then mixed with a chlorinating agent downstream. The passage of the activities through a thermochromatographic column produced distinct absorption zones from which the properties of the chlorides of new elements were estimated.

Another approach, in which the recoil atoms are transported over several meters by a gas-jet and then brought into contact with a reactive agent to form volatile species³²) is outlined in Figure 6.

The physico-chemical data on bromides of fission products suggest that the bromides might be suitable for a separation of neutron-rich nuclides in a thermochromatographic column. The fission products of ^{235}U are fed through a 7 m capillary into a reaction chamber in which the gas-jet (ethylene-nitrogen mixture or pure ethane) is mixed with nitrogen gas loaded with bromine by bubbling through liquid bromine. For an efficient bromination the reaction

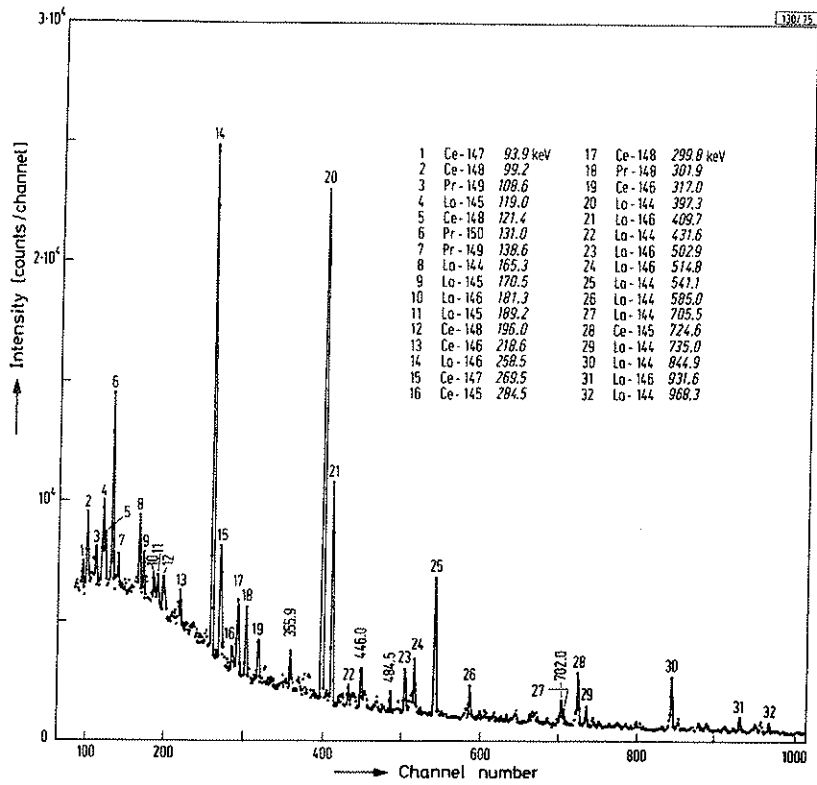


Figure 5.

γ -ray spectrum of neutron-rich lanthanum and praseodymium nuclides and their daughter products³⁹⁾ from thermal-neutron induced fission of ^{235}U , obtained on-line with the technique outlined in Figure 4

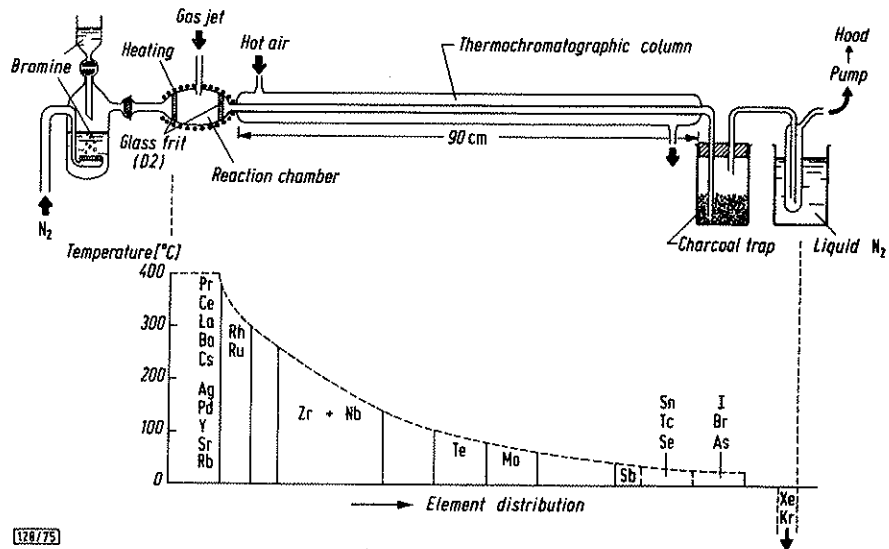


Figure 6.

Schematic diagram of the apparatus for continuous separation of fission product bromides by combination of a gas-jet transport system with a thermochromatographic column³²⁾

chamber is heated to 400°C. To prolong the contact time of the clusters with the reactive gas a glass frit is put between chamber and thermochromatographic column. The volatile bromides are swept by the gas through a 5 mm (i.d.) open tubular column. A temperature gradient is established over the thermochromatographic column through heating with hot air. At the exit of the column the gas passes through a charcoal trap at room temperature and then through another trap, held at the temperature of liquid nitrogen

The distribution of the various fission products along the column and in the traps is shown in the lower part of Figure 6. The bromides of the alkali, alkaline earth and the lanthanide elements as well as those of yttrium, palladium and silver are non-volatile under the described conditions and remain in the reaction chamber. Ru and Rh are found at the hot end of the column. Zr and Nb are moderately volatile and Mo and Te are found in the colder part of the column. Sb, Sn, Tc, Se go partly to the cold end of the column and are also found in the charcoal trap together with As, Br, I. The noble gases are collected in the last trap. An underlying background of non-volatile fission products is also observed in the column caused by clusters which were not destroyed and have passed the glass frit. The contaminations disappear by heating the reaction chamber to 700°C and filling it with quartz wool.

The delay-time between production and deposition of short-lived nuclides was found to amount to 3-4 sec. Among other things this set-up was used to measure the delayed-neutron spectrum of 1.7 sec ^{135}Sb as discussed by K.-L. Kratz et al. in their contribution to this conference, indicating the short time scale achieved in on-line separations in the gaseous phase in conjunction with a gas-jet system.

Drawbacks of the thermochromatographic method are the relatively poor resolution; e.g. the deposition zones are rather broad with some overlap between adjacent elements, and the shift in the deposition zone depending on the half-life of the nuclides. This can be circumvented by substituting the temperature gradient by a sharp step to a lower temperature where the volatile compound can be deposited on a small spot with the disadvantage of a decreasing selectivity. The selectivity is improved by adding a second reactive gas at the deposition spot through which the desired or the undesired elements are transported further downstream. An alternative to this method is the replacement of the adsorption chromatography through the chemisorption. For this approach suitable wall materials which form stable complexes with the volatile compounds at certain temperatures are required. Both methods have been investigated by the Darmstadt-group in detail⁴²⁾.

Figure 7 shows an example for the separation of fission products using the reactions with different wall materials⁴³⁾. This separation was carried out with fission product chlorides in a temperature gradient from 700°C to 25°C. The wall was coated with BaCl_2 , NaCl , KCl and CsCl . Due to the formation of different complexes with distinct deposition temperatures, elements like Zr and Nb, which are difficult to separate by adsorption chromatography are well resolved. In principle a high selectivity can be achieved by using several different wall materials so that at each zone of coating one element is fixed.

In some cases a combination of fast chemical separation methods with on-line isotope separators

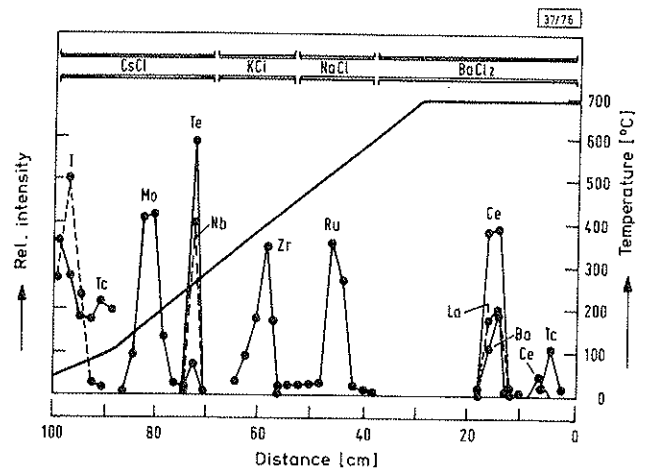


Figure 7.

Thermochromatographic separation of fission products by reaction with different wall materials⁴³⁾

is very desirable, e.g. if the target-ion source system processes many elements. At the Studsvik OSIRIS separator, a thermochromatographic system working at low pressures has been attached to the collector chamber⁴⁴⁾. The system is shown in Figure 8.

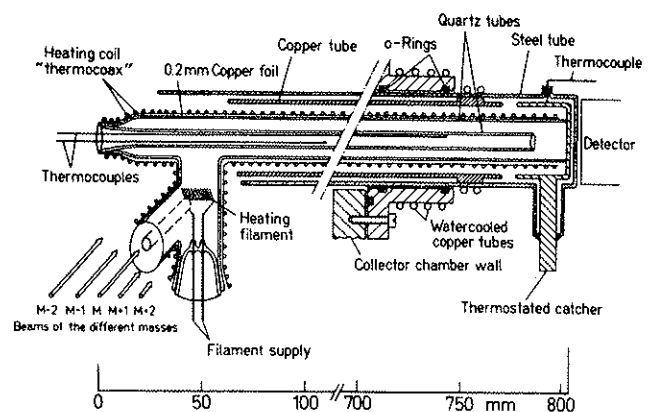


Figure 8.

Schematic picture of a thermochromatographic separation apparatus attached to the collector chamber of an isotope separator⁴⁴⁾

It consists of two concentric quartz tubes (length 800 mm, dia 20 mm and 10 mm, respectively) the outer one surrounded by heating coils. A copper foil between the quartz and the coils smoothes the heat from the coils. A steel tube surrounding the coils can be moved axially through a hole in the collector chamber and vacuum sealing is achieved by means of two O-rings. One end of the thermochromatographic apparatus is provided with a narrow entrance tube which allows a selected ion beam to enter and to hit a collector. The choice of the collector material depends on whether or not a chemical reaction between the material and the incoming beam is desired. In the simplest case the collector is an

inert material, for instance, tungsten and the ions, after being stopped and neutralized, rapidly diffuse to the surface and evaporate. The other end of the apparatus contains a catcher whose temperature can be adjusted in the range from -100°C to 350°C. This arrangement is suitable only for elements with a moderate deposition temperature (~400°C). It was applied to the separation of short-lived Cd and Zn isotopes in the mass chains 119-128 and 75-80, respectively^{4,5}). Pure sources of these nuclides, except for the daughter activities growing in have been prepared. Furthermore halogen isotopes with half-lives down to 1 sec have been successfully separated from accompanying isobaric contaminations^{4,4}).

Instead of an inert material one may use a collector material in which the incoming atoms form volatile compounds by chemical reactions.

4. Closing remarks

To conclude, one can say that rapid chemical separations combined with high-resolution radiation spectroscopy offers one important approach for studying nuclides far from the region of β -stability.

The advantages of the chemical methods are selectivity and sensitivity. Chemical procedures show an ideal resolution with regard to the atomic number and enable the detection of even a few atoms in a bulk of interfering activities. In the past, the main restriction in the application of rapid chemical separation techniques has been seen in the time scale. Here considerable progress has been made by developing fast discontinuous and continuous radiochemical separations as outlined in this paper. The future trends may be mainly directed towards the improvement of existing and development of new continuous procedures. For instance, the continuous solvent extraction with the centrifuges is not only limited to the separation of lanthanides but can be extended to almost all elements. The investigations of fast separations in the gas phase are far from being completed and will probably be continued for further years.

References

- 1) S. Amiel in L. Yaffe, Nuclear Chemistry, Academic Press, New York and London, p. 252 (1968)
- 2) G. Herrmann and H.O. Denschlag, Ann. Rev. Nucl. Sci., 19, 1 (1969)
- 3) R.D. Macfarlane and W.C. McHarris in J. Cerny, Nuclear Spectroscopy and Reactions, Academic Press, New York and London, p. 243 (1974)
- 4) N. Trautmann and G. Herrmann, J. Radioanal. Chem., 32, 533 (1976)
- 5) W. Eckhardt, G. Herrmann and H.D. Schüssler, Z. Anal. Chem. 226, 71 (1967)
- 6) H. Reinhardt and J. Rydberg, Acta Chem. Scand., 23, 2773 (1969)
- 7) R. Denig, N. Trautmann and G. Herrmann, Z. Anal. Chem., 216, 41 (1966)
- 8) H. Folger, J.-V. Kratz and G. Herrmann, Radiochem. Radioanal. Letters, 1, 185 (1969)
- 9) H. Ahrens, N. Kaffrell, N. Trautmann and G. Herrmann, Phys. Rev., C13 (1976), in press
- 10) N. Trautmann, N. Kaffrell, H.-W. Behlich, H. Folger, G. Herrmann, D. Hübscher and H. Ahrens, Radiochim. Acta, 18, 86 (1972)
- 11) G. Tittel, Annual Report 1974, Institut für Kernchemie der Universität Mainz, p. 50
- 12) N. Trautmann, N. Kaffrell, H. Ahrens and P.F. Dittner, Phys. Rev. C13, 872 (1976)
- 13) G. Franz and G. Herrmann, Inorg. Nucl. Chem. Letters, 11, 857 (1975)
- 14) G. Klein, N. Kaffrell, N. Trautmann and G. Herrmann, Inorg. Nucl. Chem. Letters 11, 511 (1975)
- 15) K.-E. Seyb, Annual Report 1973, Institut für Kernchemie der Universität Mainz, p. 43
- 16) W. Brüchle, Doctoral thesis, Universität Mainz (1976)
- 17) H.D. Schüssler and G. Herrmann, Radiochim. Acta, 18, 123 (1972)
- 18) J.-V. Kratz, H. Franz and G. Herrmann, J. Inorg. Nucl. Chem., 35, 1407 (1973)
- 19) J.-V. Kratz and G. Herrmann, J. Inorg. Nucl. Chem., 32, 3713 (1970)
- 20) W. Rudolph, K.-L. Kratz and G. Herrmann, to be published
- 21) H.D. Schüssler, H. Ahrens, H. Folger, H. Franz, W. Grimm, G. Herrmann, J.-V. Kratz and K.-L. Kratz, Physics and Chemistry of Fission, IAEA Vienna, p. 591 (1969)
- 22) R. Naeumann, H. Folger and H.O. Denschlag, J. Inorg. Nucl. Chem., 34, 1785 (1972)
- 23) E.K. Hulet, J.M. Nitschke, R.W. Loughheed, J.F. Wild, J.H. Landrum and A. Ghiorso, Proc. 4th Int. Transplutonium Element Sympos., Baden-Baden, Sept. 1975, p. 3 (1976)
- 24) D.O. Campbell, J. Inorg. Nucl. Chem. 35, 3911 (1973)
- 25) E.P. Horwitz and C.A.A. Bloomquist, J. Chromat. Science, 12, 11 (1974)
- 26) M. Schädel, N. Trautmann and K.-E. Seyb, Paper presented at the Hauptversammlung der Gesellschaft Deutscher Chemiker, Köln (1975)
- 27) T. Tamai, J. Takada, R. Matsushita and Y. Kiso, Inorg. Nucl. Chem. Letters, 9, 973 (1973)
- 28) M. Schädel and T. Tamai, private communication (1976)
- 29) W.J. Wieseahn, G. Bischoff and J.M. D'Auria, Nucl. Instr. Meth., 124, 221 (1975)
- 30) H.G. Wilhelm, H. Jungclas, H. Wollnik, D.F. Snider, R. Brandt and K.H. Lust, Nucl. Instr. Meth. 115, 419 (1974)
- 31) H. Wollnik, H.G. Wilhelm, G. Röbig and H. Jungclas, Nucl. Instr. Meth. 127, 539 (1975)

- 32) R.J. Silva, N. Trautmann, M. Zendel, H. Ahrens, P.F. Dittner and E. Stender, to be published
- 33) W.J. Wieseahn, J.M. D'Auria, H. Dautet and B.D. Pate, *Can. J. Phys.* 51, 2347 (1973)
- 34) K.-H. Hellmuth, S. Hillebrand, P. Suominen, K. Valli and J. Aystö, Research Report 4/1975, Dept. of Physics, University of Jvaskylä
- 35) K.L. Kosanke, M.D. Edmiston, R.A. Warner and W. C. McHarris, *Nucl. Instr. Meth.* 125, 253 (1975)
- 36) K.L. Kosanke, W. C. McHarris and R.A. Warner *Nucl. Instr. Meth.* 115, 151 (1974)
- 37) P.O. Aronsson, B.E. Johansson, J. Rydberg, G. Skarnemark, J. Aistad, B. Bergersen, E. Kvåle and M. Skarestad, *J. Inorg. Nucl. Chem.* 36, 2397 (1974)
- 38) N. Trautmann, P.O. Aronsson, T. Björnstad, N. Kaffrell, E. Kvåle, M. Skarestad, G. Skarnemark and E. Stender, *Inorg. Nucl. Chem. Letters*, 11, 729 (1975)
- 39) P.O. Aronsson, T. Björnstad, N. Kaffrell, E. Kvåle, G. Skarnemark, E. Stender and N. Trautmann, to be published
- 40) I. Zvara, Yu.T. Chuburkov, R. Tsaletka, T.S. Zvarova, M.R. Shalaevskii and B.V. Shilov, *Atomnaya Energiya* 21, 83 (1966) =*Sov. J. Atom. Energy* 21, 709 (1966)
- 41) I. Zvara, Yu.T. Chuburkov, R. Tsaletka and M.R. Shalaevskii, *Radiokhimiya* 11, 163 (1969) =*Sov. Radiochem.* 11, 161 (1969)
- 42) K. Bächmann, V. Matschoss and S. Tsalas, *Proc. 4th Int. Transplutonium Element Sympos.*, Baden-Baden, Sept. 1975, p. 37 (1976)
- 43) S. Tsalas and K. Bächmann, private communication (1976)
- 44) B. Grapengiesser and G. Rudstam, *Radiochim. Acta* 20, 85 (1973)
- 45) B. Grapengiesser, Research Report LF-59 (1974) Studsvik, Fack