Influence of Nuclear Structure on the Fusion Below the Classical Barrier

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The investigations of mechanisms influencing the fusion cross sections near the threshold have been continued. [1] Fusion excitation functions extending from a few μb up to several 100 mb have been measured for the systems $^{40}Ar+^{112,116,122}Sn$, $^{86}Kr+^{70,76}Ge$, $^{86}Kr+^{92,100}Mo$, and $^{86}Kr+^{99,102,104}Rn$.

The experimental method, briefly described in Ref. 1, involves the combination of a velocity filter and activation methods. Particular emphasis is put on a comparative study of the various systems, in order to isolate nuclear structure effects, if any. A recent study [2] has shown that the trends of the fusion cross sections below threshold (in the 10 to 100 mb range) could be well described in the Hill-Wheeler picture if the inverted oscillator frequency $\hbar\omega$ was adjusted to the data. This frequency was then found to increase smoothly with the reduced radius of the projectile target system, indicating a correlation of the apparent subbarrier transparency with the strength of the proximity potential [3] between the two nuclei.

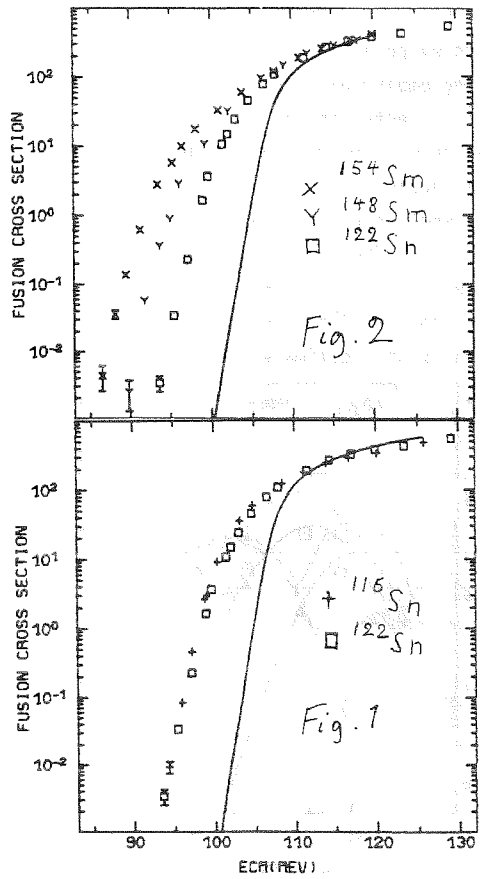
Our data, extending down to a few μb cross sections, now show that a clear correlation exists between 'subbarrier' fusion and the ground state properties of the nuclei involved. This is illustrated in Figs. 1 - 2. In Fig. 1 we compare the excitation functions for the fusion of ^{40}Ar with ^{116}Sn (crosses) and ^{122}Sn (squares). The full curve represents the rigid spheres approximation to fusion of $^{40}Ar+^{122}Sn$ using a potential very close (1.5 %) to the proximity potential and calculating the subbarrier transmission with the WKB method. The comparison of the $^{40}Ar+^{122}Sn$ data with this reference curve suggests the following characterization: Around and above the 200 mb level, the 'geometrical' range, the data are well described by the calculation. Below the 1 mb level, the experimental deexcitation function is characterized by a quasiexponential slope that is very similar to the one calculated, except that it is shifted downward by about 8 MeV. This energy range could be characterized as dominated by 'pure' tunnelling giving the 'true' subbarrier transparency. Between 1 mb up to just above 100 mb there is a third range that is characterized by a varying slope and that is not reproduced by a two-spheres calculation. These characteristics are not *quan*titatively changed if 6 neutrons are removed from the tin isotope, i.e. for ^{116}Sn (crosses). In the figure we have downshifted the data for ^{116}Sn by the difference in fusion barrier heights predicted by the proximity potential. [3]

Figure 2 shows that this conclusion is modified if the Sn isotopes are replaced by the Sm isotopes. Again the data are downshifted relative to the ^{122}Sn data (squares) by the fusion barrier differences calculated with the proximity potential. Whereas the data for the various isotopes tend to converge in the 'geometrical' range, it is obvious that for the deformed nucleus ^{154}Sm, a strong influence of the static deformation is seen. But even ^{148}Sm, which is basically a 'spheric' nucleus, differs markedly from the tin isotopes: Its character as transitional nucleus becomes apparent.

A preliminary evaluation of the ^{86}Kr data shows that even for this more massive projectile a correlation with the collective character of the nuclei involved (i.e. the Ru isotope series) subsists.

Although it is too early to draw quantitative conclusions from these observations it seems clear that the surface modes (quadrupole and octupole) of the colliding nuclei play an important role as doorways to fusion. [4]

[1] W. Reisdorf et al., GSI Sc. Report 1980, p. 2
[2] U. Jahnke et al., Phys. Rev. Lett. 48 (1982) 17
[3] J. Blocki et al., Ann Phys. (N.Y.) 105 (1977) 477
[4] H. Esbensen, Nucl. Phys. A352 (1981) 147



Asymmetric Angular Distributions for Symmetric Fragmentation in $^{56}Fe + ^{208}Pb$ collisions

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Binary reaction products from the interaction of ^{208}Pb-ions with targets of ^{26}Mg through ^{58}Fe and ^{64}Ni have previously been studied[1] with a large position sensitive ring counter. When analyzed on the basis of fusion models the cross sections for mass equilibrated fragments demonstrated that deformations induced at contact influence fusion of the heaviest systems in a significant and characteristic way. In particular, for the heaviest systems it was observed that there is a way from the entrance channel to a symmetric exit channel that bypasses the slow process of compound nucleus formation.[1] In a schematic model of nuclear coalescence and reseparation[2] this type of reaction is predicted for trajectories that pass over the "conditional saddle" for capture in the entrance channel but do not pass over the "true saddle" for fusion.

In an attempt to search for evidence for the coexistence[2] of three distinct reaction channels in an "overcritical" system with $(Z^2/A)_{eff.}>33$ (complete fusion-fission, capture-fission, and deep inelastic scattering) and to assess the associated reaction times, we have examined the $^{56}Fe + ^{208}Pb$ system, $(Z^2/A)_{eff.}=38.58$, with two complementary experimental techniques. Differential cross sections $d^3\sigma/dZd\Theta dE$ were measured at E/B=1.2 in the range of centre of mass angles 20^0 through 120^0 for projectile-like fragments and symmetric fragments up to Z≈60 with a position-sensitive ΔE,E - ionization chamber. After integration over all Q-values<-10 MeV, these data are shown in Fig. 1. In addition, angular distributions were measured with an off-line X-ray activation technique between 5^0 and 178^0 in the centre of mass system at E/B=1.1, 1.2, 1.3, and 1.6 for 38≤Z≤83. For all values of Z (except for $(Z_1 + Z_2)/2$) we observe angular distributions that are asymmetric around 90^0. As an example, Fig. 2 shows angular distributions $d^2\sigma/dZd\Theta_{cm}$ for Z=50 and Z=56 at E/B=1.2. The forward-backward asymmetries are most pronounced at the lowest bombarding energy and decrease gradually with increasing bombarding energy. This shows that mass equilibration can occur in less time than required for half a revolution of the system at all bombarding energies and precludes an interpretation of the angular distributions in terms of the standard fission theory of Halpern and Strutinsky.

For that theory to hold it is required that the fissioning system makes many revolutions as it slowly decays over the barrier.

We are presently combining the data taken at E/B=1.2 from both experiments in order to construct a complete set of cross sections in the Z-Θ_{cm} plane. So far, the data do not seem to call for a distinction of different reaction channels. This indicates that the time scales for inelastic scattering and symmetric fragmentation processes narrow considerably for very heavy systems.

[1] H. Sann et al., Phys. Rev. Lett. 47, 1248 (1981) and R. Bock et al., Preprint GSI-81-35 (1981), to appear in Nucl. Phys. A.

[2] W. J. Swiatecki, Report LBL-12642 (1981), to appear in Nucl. Phys. A.

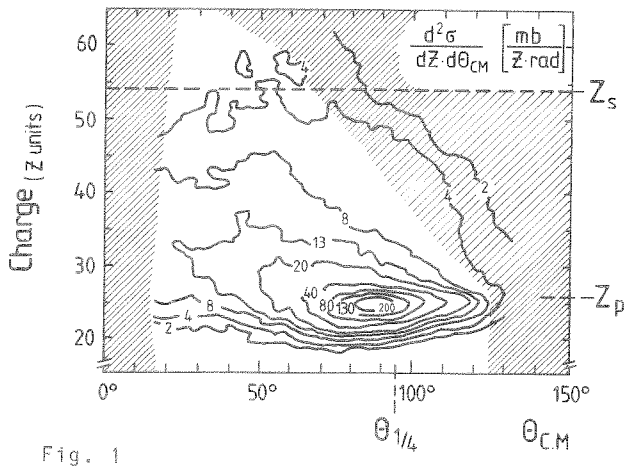

Fig. 1

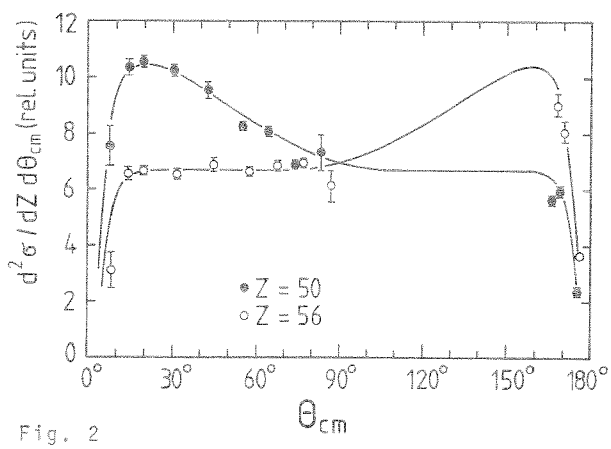

Fig. 2

Charge-, Mass-, and Energy Distributions in the Reaction of 7.5 MeV/u ^{238}U-ions with ^{197}Au

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The measurements of differential cross sections for target-like products formed in the collision of 7.5 MeV/u ^{238}U-ions with ^{197}Au using a stacked foil technique and radiochemical separations[1] were completed. For sufficiently long-lived isotopes of the elements Hf, Ta, Ir, Pt, Au, Hg, and Tl differential cross sections

$$\int_{R_1}^{R_2}\int_{\Theta_1}^{\Theta_2} (\delta^4\sigma/\delta Z\delta A\delta R\delta\Theta)\, dR d\Theta$$

have been evaluated (R = range in mylar, Θ = laboratory angle). In a separate experiment, cross sections $\delta^2\sigma/\delta Z\delta A$ integrated over all angles Θ and ranges R were measured for elements with $69 \leq Z \leq 83$. By combining the results from both experiments, the primary fragment distribution can be obtained[2].

The gross features of the reaction are visualized by velocity diagrams for a product in the vicinity of the target (^{194}Au, fig. 1) and one far removed from the target (^{178m}Ta, fig. 2). In both figures, given catcher foils are indicated by their limiting laboratory angles and velocities. The numbers within the quadrangular fields representing a given catcher foil denote the associated differential cross section. For ^{194}Au, fig. 1 shows a pronounced peak in the cross section at small Q-values and near the grazing angle together with a remarkably long tail towards very large Q-values and forward angles. For residual isotopes further and further removed from the entrance channel, the cross section is increasingly concentrated at forward angles and at larger Q-values (fig. 2). These observations clearly indicate a deflection function typical for systems where the Coulomb repulsion dominates over the attractive nuclear potential.

The fact that a given residual Au isotope is observed with Q-values ranging from 0 to 350 MeV (see fig. 1) corresponding to excitation energies in the associated primary fragments of about 0 to 150 MeV, is interpreted in the following way: in the absence of charged particle evaporation from these neutron-rich species (which is suggested by evaporation code calculations[3]) we observe primary atomic numbers, the mass numbers being modified by neutron evaporation. The Q-value distribution shown in fig. 1 then tells us that ^{194}Au has been formed by neutron evaporation from ^{198}Au through ^{207}Au, with the most neutron-rich mass numbers being associated with the highest excitation energies. This indicates that the N/Z-equilibration in the ^{238}U + ^{197}Au reaction occurs by a shift of the centroid of the peak cross section in the N/Z-plane from the entrance channel neutron number to values near N ≈ 123, which corresponds to the minimum of the potential energy surface of the combined system for Z = 79.

[1] K. Sümmerer et al., GSI Scientific Report 1979, 29

[2] J. Poitou et al., Nucl. Instr. and Meth. 180 (1981) 221

[3] W. Reisdorf, private communication

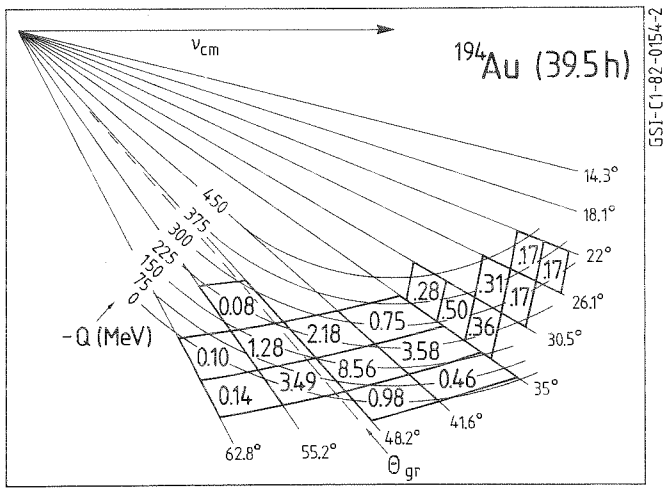

Fig. 1: Velocity diagram for ^{194}Au. The cross sections are given in mb. For further explanation see text.

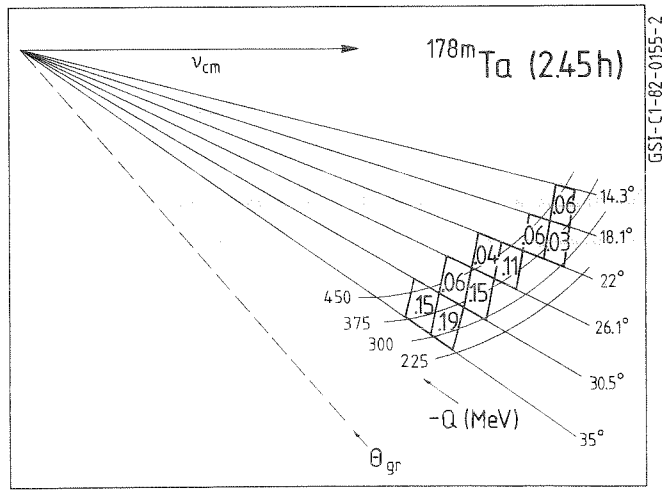

Fig. 2: Velocity diagram for ^{178m}Ta.

Search for Long-Lived Superheavy Elements in the $^{238}U + ^{248}Cm$ Reaction [B]

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First attempts to detect superheavy nuclei produced in bombardments of ^{248}Cm with ^{238}U remained unsuccessful[1]. The sensitivity for detecting superheavy nuclei in these experiments was much less than optimum because of premature failures of the ^{248}Cm-metal targets in the intense ^{238}U beams. After extensive target tests with pulsed electron beams, low-energy heavy ion beams and 7.5 MeV/u ^{238}U beams the original target apparatus[2] was modified to allow cooling of the targets from both sides by nitrogen gas[3].

With the improved target cooling we were able to collect within four days of beam-time an integral particle number of 2×10^{16} on target. The heavy target-like fragments recoiling from the target were stopped in a water-cooled stack of copper foils which was then processed by gas phase and solution chemistry to prepare thin sources of fractions containing the elements Os, Ir, Pt, Au, Hg, Tl, Pb, Bi, Po, and At and eventually their superheavy homologues. These fractions were mounted on thin Ni-foils and are being counted between two opposed surface barrier detectors inside a neutron multiplicity counter. Noble gases like Rn and eventual superheavy species that are volatile at room temperature were condensed at -200°C on a Pd surface faced by a single surface-barrier detector. So far, after 180 days of counting, no fission events were recorded in the gas phase chemistry fractions carrying elements that are volatile above room temperature up to 1020°C. In a solution chemistry fraction containing elements forming anionic bromide complexes one fission event with two fission fragments and two neutrons in coincidence was recorded. In the noble gas fraction during the first two-hours counting interval two fission events were registered. The significance of these events is certainly marginal. However, previous indications for fission events in the noble gas fractions isolated off-line from reaction products of $^{238}U+^{238}U$, Ref. 4, and $^{136}Xe+^{238}U$ collisions[5] strongly suggest that the possible existence of spontaneous fission activity associated with the decay of a gaseous reaction product deserves further study.

Assuming 75% chemical yields for superheavy elements in analogy to their chemical homologues and 6 events or 3 events (compatible with 2 - 1 registered events at 95% confidence level) we obtain the cross section limits given in Fig. 1. The sensitivity now achieved for the regime of life-times in excess of several hours is close to the edge of current detection limits. Therefore, further experiments should focus on shorter-lived species by using fast on-line chemical separations[6] which can be performed with equal sensitivity.

[1] W. Brüchle et al., Scientific Report 1980, GSI 81-2, p. 65 (1981)

[2] J.D. Molitoris, J.M. Nitschke, Report LBL-9725 (1981)

[3] W. Brüchle et al., this report

[4] H. Gäggeler et al., Phys. Rev. Lett. 45, 1824 (1980)

[5] S. Yashita, R.E. Leber, Report LBL-6547 (1977)

[6] H. Dornhöfer et al., this report

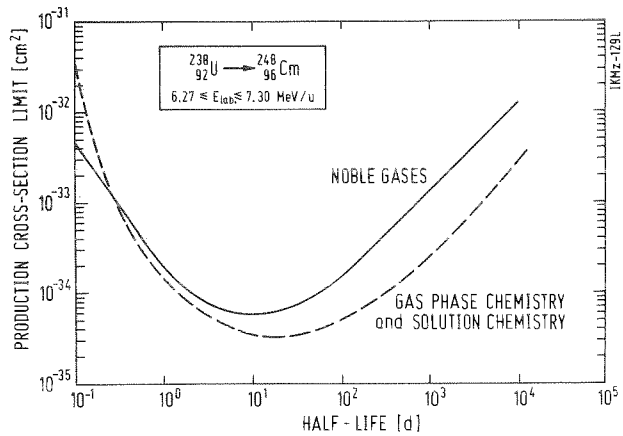

Fig. 1: Upper limits for the production cross sections of superheavy elements in the $^{238}U+^{248}Cm$ reaction.

Ranges of 1.4 MeV/u Uranium Projectiles in Solids

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We have described previously a method for measuring the mean range R_p and the longitudinal range straggling $\sigma_{\parallel}$ of ^{238}U projectiles in solids. [1] The uranium ions are implanted into a stack of self-supporting foils and the number of ^{238}U atoms that were stopped in each of the foils is obtained by thermal neutron activation analysis. In order to get the best target uniformity they were produced by evaporation at the GSI target laboratory.
The measured R_p is a projection of the integral path length R_{int} in the beam direction. R_{int} can be calculated from R_p by the LSS theory. [2] The R_{int} of 1.4 MeV/u uranium ions in C, Al, Ti, Ni, Nb, Pd, and Sn are shown in Fig. 1 and compared to the values obtained according to the Northcliffe and Schilling tables [3], the Littmark and Ziegler tables [4], the LSS theory [2], and to a semi-empirical approach of Mukherji and Srivastava [5]. The R_{int} values are larger than the R_p ones by 0.2 % in C and up to 3 % in Sn. The Z_2-oscillations in the stopping powers cause the minimum and maximum of the R_{int} at $Z_2 \approx 22$ and 28, respectively.
The measured longitudinal range straggling values due to the stopping process and to target inhomogenieties σ_{rt}, are shown in Fig. 2 together with the range straggling values according to the LSS theory [2] and to the Littmark and Ziegler tables [4]. It is seen that the tables as well as the theory reproduce fairly well the measured results. The range straggling of fission fragments in gases [6] is about 40 % larger than the LSS values while in rolled solids targets [7] the values are about 80 % larger. It is concluded that the target inhomogenieties in evaporated targets are significantly lower than in rolled targets.

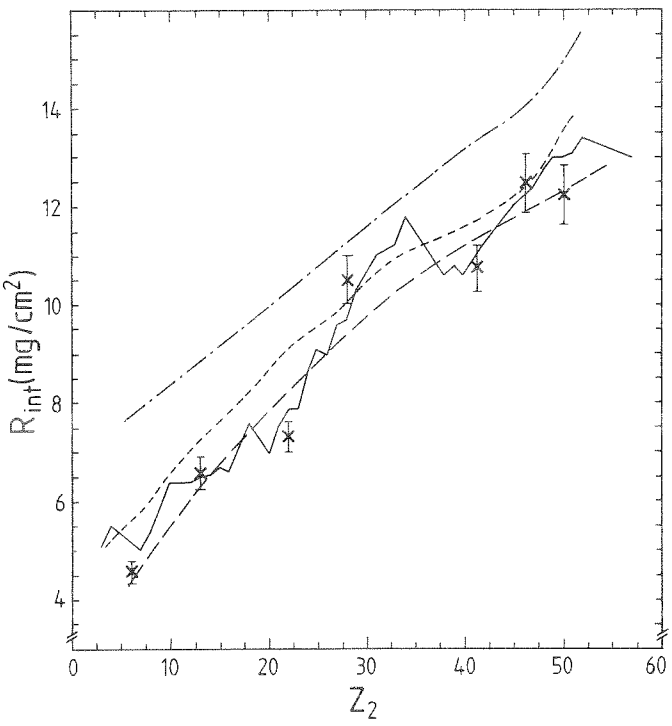

Fig. 1: Integrated mean path length of 1.4 MeV/u ^{238}U ions in solids

x - this work
– – – Northcliffe and Schilling [3]
– · – · – LSS theory [2]
– – – – Mukherji and Srivastava [5]
——— Littmark and Ziegler [4]

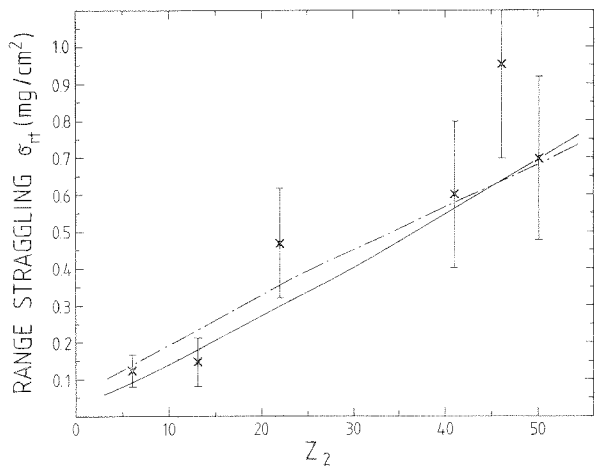

Fig. 2: Longitudinal range straggling of 1.4 MeV/u ^{238}U projectiles in solids

x - this work
– · – · – LSS theory [2]
——— Littmark and Ziegler [4]

[1] Y. Laichter, H. Geissel, M. Schädel, P. Armbruster, GSI-81-2, p. 138

[2] J. Lindhard, M. Scharff and H.E. Schiøtt, Mat. Fys. Medd. Dan. Vid. Selsk. 33, (1963) No. 14

[3] L.C. Northcliffe, R.F. Schilling, Nuclear Data Tables 7A (1970) 233

[4] Y. Littmark, J.F. Ziegler, Vol. 6 of The Stopping and Ranges of Ions in Matter, Perg. Press (1980)

[5] S. Mukherji and B.K. Srivastava, Phys. Rev. C6 (1974) 3708

[6] M. Pickering and J.M. Alexander, Phys. Rev. C6 (1972) 332

[7] Y. Laichter and N.H. Shafrir, Nucl. Phys. A371 (1981) 45

Further Tests of Gd/Cm-Targets with High-Intensity ^{238}U-Beams

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Hitherto, the limited stability of metallic ^{248}Cm targets on molybdenum substrates during heavy-ion bombardments has limited the sensitivity in the search for superheavy elements in the ^{238}U+^{248}Cm reaction[1,2]. Test bombardments in 1980 where alternative substrate materials were used[2] indicated that integral particle numbers of 5-7 x 10^{14} lead to the same failure mechanisms as observed before[1]. These tests also indicated a large reduction in thermal conductivity or a problem in transferring heat across the Cm-substrate interface to the fast flowing N_2 cooling gas on the substrate side. The heat transfer problems could not be duplicated with a pulsed electron beam. Thus, it is possible that radiation damage[3] specific to heavy-ion bombardments plays an important role in the target failure mechanisms.

In 1981 we modified the target system in order to investigate the effect of an improved cooling of the target. The latter was achieved by i) reversed mounting of the target, so that the Gd/Cm metal deposit was directly cooled by N_2 cooling gas, and ii) by mounting the target into a newly designed "diffuser" resulting in equal cooling of both sides of the target. In the second case an additional Mo-window separated the target and the cooling gas from the recoil chamber where the reaction products are stopped. The initial velocity of quasi-elastic and deep-inelastic target-like fragments is sufficiently high to penetrate this additional window. The results are summarized in the Table. Target failures were observed at beam integrals of both 2.5 x 10^{15} particles for Gd targets on Mo and Nb substrates when only the Gd deposit was cooled. At a beam integral of 6.3 x 10^{15} particles the test of a doubly-cooled Gd-Mo target had to be stopped due to lack of beam-time without target failure. Such an integral particle number had never been achieved on a single target before.

The success with the double-sided cooling was persuasive enough to resume ^{238}U-ion bombardments of ^{248}Cm-targets on Mo-substrates and to repeat the search for long-lived superheavy elements using radiochemical techniques[4]. In four days of allotted beam time an integral of ~2 x 10^{16} particles was accumulated on two targets. This is considered close to the edge of current technological limits.

Table: Further Gd- and Cm-metal target tests

Substrate (mg/cm^2)	Metal deposit (mg/cm^2)	Beam integral ($x10^{15}$ particles)	Comments[a]
Mo (4.5)	Gd (4.5)	2.6	Reversed mounting. Maximum substrate temp. 760°C at $2.1x10^{11}$ p/s; failed at 530°C.
Nb (3.3)	Gd (3.9)	2.5	Reversed mounting. Max. substrate temp. 720°C at $1.4x10^{11}$ p/s; failed at 700°C.
Mo (4.4)	Gd (4.0)	6.3	Double-sided cooling. Max. window temp. 688°C at $1.6x10^{11}$ p/s; no target failure.
Mo (4.6)	Cm (5.3)	6.6	Double-sided cooling. Temp. not monitored. 3 big cracks in the direction of the N_2-flow.
Mo (4.7)	Cm (4.2)	12.8	Double-sided cooling. Temp. not monitored. No target failure.

[a] In all cases the cooling gas flow was 1.2 l/s N_2 at 1.3 - 1.4 bar.

[1] M. Schädel et al., Scientific Report 1979, GSI-80-3, p. 63 (1980)

[2] R.W. Lougheed et al., Report UCRL-84604 (1980)

[3] I. Maor, Report GSI-81-9 (1981)

[4] M. Schädel et al., this report

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KINEMATIC GAS JET CHAMBER FOR DAMPED HEAVY-ION REACTIONS

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For gas jet experiments with high-intensity heavy-ion beams (10-100pnA Pb or U) a gas-cooled double window and a target wheel rotating inside the gas chamber were built previously[1]. With this system we achieved only low gas jet efficiencies for Hg detection both in irradiations of HgS and Pb with Xe ions and in off-line gas jet tracer experiments. There it was shown[2] that most of the ^{203}Hg tracer activity was thrown on the chamber walls due to gas turbulences caused by the rotating wheel.

Therefore we altered the design[1] of the target and gas chamber by inserting a conical foil downstream of the wheel (fig. 1). Now turbulences due to the wheel rotation are confined to the target chamber, which is separated from the thermalization chamber by the foil (8 µm Al). It is supported by twelve copper wires (Ø 1 mm), which are soldered in between two rings, thus defining the conical shape of the foil. Reaction products to be thermalized first have to penetrate this foil, which is fixed and sealed by glue. The pressures in both the target and the thermalization chamber have to be equalized in order to avoid rupture of the large area conical foil. The geometry of the cone and of the thermalization chamber has to be adapted to the kinematics of the reaction under study, here U + U[3]. The chamber shown in fig. 1 has an angular acceptance for reaction products from 10° to 60°. The angular distributions for the damped reaction U + U fall essentially into this interval[4].

The primary ion beam is stopped in a water-cooled copper block at the downstream end of the wire support. The transport gas is fed into the chamber through a 90 mm² annular slit (Ø 60 mm, width 0.5 mm) and sucked into the capillary through a nearly exponential funnel, so as to reduce gas turbulences and losses of activity to the walls.

The target wheel chamber is scavenged by a gas (here He because of its low stopping power for reaction products), which is kept at the same pressure as the thermalization chamber (2 bar for U + U). The energy loss of 10 MeV/u ^{238}U ions in the gas-cooled window (2 x 3.5 mg/cm² Mo, 1.15 mg/cm² N_2 of 1.3 bar) and in the He (2 bar, 0.32 mg/cm²) before the target amounts to 3 MeV/u.

The thermalization chamber has a maximum length and diameter of 12 and 10 cm, respectively, and a volume of 650 cm³. The oven of the gas chemical separation stage[2] was coupled to the Ar chamber by a 25 m long capillary of 1 mm inner diameter. The Ar flow rate was limited by the hot oven to 10 bar cm³ s^{-1}, which means a maximum transport time of 130 s for 2 bar Ar in the gas chamber. The cooling water for the beam stopper is led through copper tubes inside the gas chamber. The vertical tubes are squeezed to oval shape in order to diminish losses by implantation of reaction products.

Our design of the target chamber and the thermalization chamber, separated from each other by a supported conical foil, may be called "kinematical chamber" as its geometry accounts for the reaction kinematics, in our case for the U + U reaction.

The gas-cooled double window together with the rotating target wheel (16 targets of 45 x 15 mm² size) allow gas jet experiments on damped reactions with intense heavy-ion beams (e.g. > 10 pnA ^{238}U).

[1] G. Dersch et al., annual report 1980, GSI 81-2, p. 194
[2] N. Greulich et al., this report
[3] H. Dornhöfer et al., this report
[4] H. Gäggeler et al., Nucl.Instr. and Meth. 188 (1981) 367

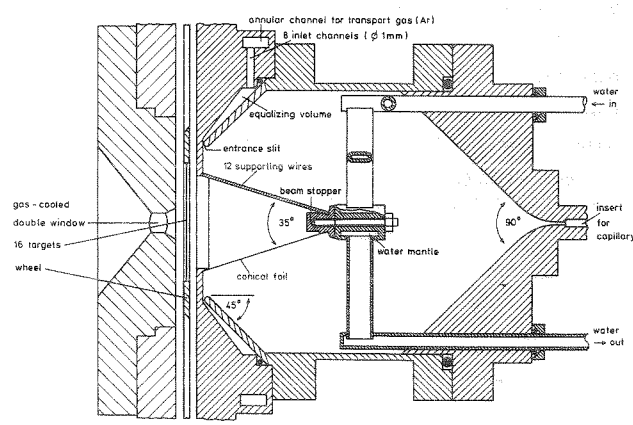

Fig. 1: Cross section through the "kinematical" thermalization chamber for the U + U reaction.

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An On-Line Chemistry for the Separation of Superheavy Elements from Heavy Ion Reactions

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Direct searches for long-lived superheavy elements (SHE) have yielded upper limits for their production cross sections[1]. A lack of sensitive measurements for short-lived SHE prompted the present experiment, which is directed towards a fast chemical separation of superheavy elements from spontaneously fissioning actinides produced in the damped collision process. The separation is based on the predicted volatility of the elements around Z=114. We have bombarded 6.9 mg/cm^2 U foils with a $1.2 \cdot 10^{11}$ p/s U beam with a projectile energy of 7.0-5.6 MeV/u within the target. The reaction products were transported with a gas-jet system to a thermochromatographic oven. The gas-jet system is described in detail elsewhere[2]. The clusters were stopped in a quartz wool plug at a temperature of about 1000°C from where the volatile elements were continuously evaporated and transported along a heated quartz tube. This tube ended in front of a cooled copper wheel onto which the transported elements were condensed. The wheel was turned stepwise after collection times of 200 s to bring the deposited activity in front of surface barrier detectors. A similar wheel system has been used before[3]. We have measured the transportation efficiency to the oven to be 50±7%. In two subsequent experiments the activity was collected on the cooled wheel, first without, and thereafter with heating of the quartz tube. In Fig. 1 α-spectra measured in the third of four detectors are compared. We attribute the strong α-lines observed in the first experiment (Fig. 1a) to the elements Th, Ra and their daughter products, which are produced with high yields in this reaction. In Fig. 1b the strong α-lines are not present. This is because these elements are not volatile at the temperatures used and they are retained in the oven. The α-lines observed in Fig. 1b can be attributed to the volatile reaction products Bi, Po and At. During a 5 hour experiment using chemical separation the integrated U beam on target was $8 \cdot 10^{14}$p. No fissioning activity was measured.

From this we deduce an upper limit for the production cross section of about 2 nb for SHE having a half-live in the region from 10^2 to 10^4 s.

[1] G. Herrmann, Proc. of the Int. Conf. on Nuclei far from Stability Helsingør 1981, CERN Rep. 81-09 Vol. II, p. 772 (1981)

[2] D. Hirdes et al., this report

[3] R.-D. von Dincklage et al., Nucl. Instr. and Meth. 176, 529 (1980)

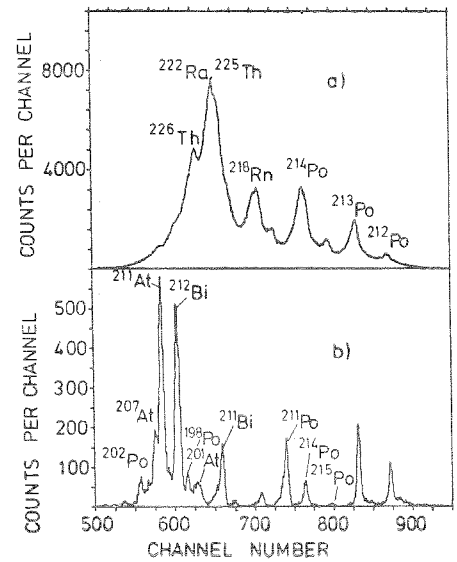

Fig. 1: Alpha spectra of U+U reaction products measured 600 to 800 s after a 200 s collection period.
a) Quartz wool plug removed, activity adherent to clusters collected on the wheel ("without chemistry")
b) with quartz plug, oven and quartz tube heated ("with chemistry")

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Gaschemical Preparation of Samples for α- and sf-Measurements

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The measurement of α-spectra and spontaneous fission events requires extreme thin and homogeneous sources which can be produced by gaschemical methods. The nuclides are volatilized in the elemental state and condensed on thin metal or carbon foils. Well suited for nearly quantitative condensations are oxide-free metals with high solution enthalpies, for example, Ag, Au, Pd, Pt or easily reduceable metals like Ni[1]. The gaschemical method described here constitutes the final step of a gas-phase separation of superheavy elements in the reaction $^{238}U+^{248}Cm$, see Ref.[2].

The apparatus, shown in Fig. 1, was tested with the isotopes ^{203}Hg and ^{212}Pb as representatives for metals volatile at lower or higher temperatures. Elemental ^{203}Hg and ^{212}Pb (1) were deposited in a quartz tube (inner diamter: 6 mm) filled with quartz powder (2). With a furnace (3) a constant temperature was kept along the whole length of the tube. A quartz capillary (4) (inner diameter: 1,5 mm; length: 30 mm) focussed the gas stream before it reached a cooled foil (5). To prevent deposition, the capillary has to be heated separately. The carrier gas stream was diverted symmetrically around the foil, leaving the apparatus through four channels in the aluminium holder (6). Nickel foils with a thickness of 500 µg/cm^2 (≈0.6 µ) (diameter: 20 mm) covered with 30 µg/cm^2 (≈0.02 µ) palladium were used as substrates for the deposition of the elements.

The experimental conditions for the simultaneous volatilization and deposition of mercury and lead were investigated. Mercury could be volatilized and transported in a pure argon stream of 0.1 l/min at 580 °C, whereas the volatilization of lead was only possible with an admixture of at least 15 % hydrogen to the carrier gas argon and at a temperature of 920 °C. The distance between the end of the capillary and the surface of the foil must be 1 mm. At this distance sufficient centering of the activity occurred without destroying the foil. The temperature of the foil, controlled by a thermocouple, is the most important experimental parameter. Lead is still deposited at a foil temperature of 50 °C, whereas for mercury less than -10 °C are necessary; on the other side, the foil could not be kept at -10 °C under the conditions for the lead volatilization. Lead could be deposited with yields of 85 % at a column temperature of 920 °C; here the mercury yield was only 57 %. At 600 °C and in a pure argon stream the yield of mercury increased to 77 %.

Volatile reaction products in the bombardment of ^{248}Cm with ^{238}U were separated gas-chemically and adsorbed on i) quartz powder (volatile elements from 20 °C to 1060 °C and ii) SiO_2/Ag-powder (for even more volatile elements). From these fractions, two samples were prepared: i) Both fractions were heated up to 600 °C in an argon stream of 0.1 l/min for 15 min and the volatilized elements were collected on a nickel foil kept at -10 °C. ii) The fraction on the quartz powder was further heated up to 900 °C in an Ar/H_2-stream for 30 min and the less volatile elements were deposited on a second nickel foil. Both sources yielded α-spectra mainly with Po and At nuclides which show good resolution. Table 1 gives the overall yields of some elements on the two foils as determined by γ-spectroscopy. They exceed 50 % in the worst case and are nearly quantitative for most of these elements.

[1] B. Eichler, ZfK Rossendorf Report 374 (1978)
[2] M. Schädel et al., this Scientific Report

Table 1
Gaschemical yields of some products from $^{238}U+^{248}Cm$

Element	Ni/Pd-foil 600 °C %	Ni/Pd-foil 900 °C %	Total %
Zn	0	100	100
Br	0	90	90
Cd	30	70	100
Sn	21	36	57
I	2	69	71
Hg	70	0	70
Pb	0	100	100
Bi	0	100	100

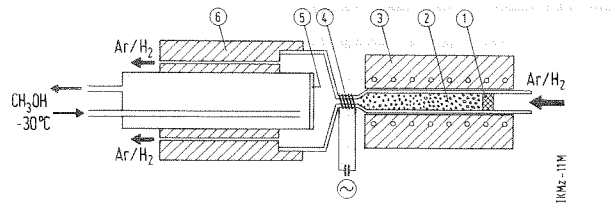

Fig. 1 Experimental set-up

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On-line Gas-phase Chemistry for Polonium

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So far searches for superheavy elements (SHE) from heavy ion reactions have mainly been carried out by off-line chemical techniques which allow the isolation of longer-lived species ($T_{1/2} \geq 3$ h). In order to extend the covered time regime towards shorter half-lives we have developed an on-line chemical method which should enable the separation of species with half-lives down to about one second. The set-up is described in another contribution to this annual report[1]. For test experiments one should try to use a "typical" representative for SHE. Several estimates show that the physical and chemical properties of polonium should be very similar to those of the predicted SHE around Z=114. Therefore, the studies were performed with the short-lived isotope ^{211m}Po ($T_{1/2}$=25.5 s).This nuclide was produced at the Göttingen cyclotron in the reaction ^{208}Pb(α,1n)^{211m}Po (E_α=30 MeV). The recoiling products were transported with a He-jet, loaded with KCl-aerosols, along a teflon capillary (ϕ_i=1 mm) to a quartz-tube inserted in an array of three ovens. At the position of the maximum temperature (1100 °C) a quartz wool plug was placed as collector for the transported products. Species volatile at this temperature such as polonium were further transported through the tube wich was kept at a variable temperature by means of the second oven. The outlet of the quartz tube was placed in front (distance 1 mm) of a water-cooled copper wheel.

From time to time, this wheel was turned to surface barrier detectors in order to measure the amount of ^{211m}Po deposited on the wheel. In a first series of experiments the α-activity of ^{211m}Po at the collector was measured as a function of the temperature of the second oven for i) an unpacked quartz column (ϕ_i=3 mm) and ii) quartz columns in which several metal foils were inserted. As an example, Fig. 1 shows the measured ^{211m}Po activity at the collector site with a 10 cm long silver foil in the column as a function of the temperature of the foil. The gas flow rate was about 500 ml/min NTP He. Below 800 °C the silver foil increasingly hinders the transmission of ^{211m}Po through the column. Table 1 summarizes the measured temperatures for 50 % transmission of ^{211m}Po through SiO_2, Pt, Au, Ag, Cu and Pd columns. At these temperatures the retention time is equal to the half-life of ^{211m}Po (25.5 s). From the metal foils investigated, Pd is the best absorber for Po since no transmission was observed up to 1100 °C. This observation is in agreement with theoretical estimates of metal-metal interaction energies[2]. For the system Po/Pd an additional experiment was performed at a fixed temperature of 1000 °C and varying lengths of the Pd-foils. The results demonstrate that already an 8 mm long Pd-cylinder leads to a decrease of the ^{211m}Po transmission (at T=1000 °C) by a factor of two. These experiments with polonium show the striking effect of metal-metal interactions which can be applied to the separation of carrier-free elements in gas-phase systems.

[1] H. Dornhöfer et al., this Scientific Report
[2] B. Eichler, ZfK Rossendorf Report 396 (1979)

Table 1

Temperatures for 50 % transmission of ^{211m}Po

Material	Length (cm)	Temperature (°C)
SiO_2	15	360 ± 50
Pt	7	650 ±100
Au	10	860 ± 50
Ag	10	560 ± 50
Cu	10	620 ±100
Pd	10	>1100

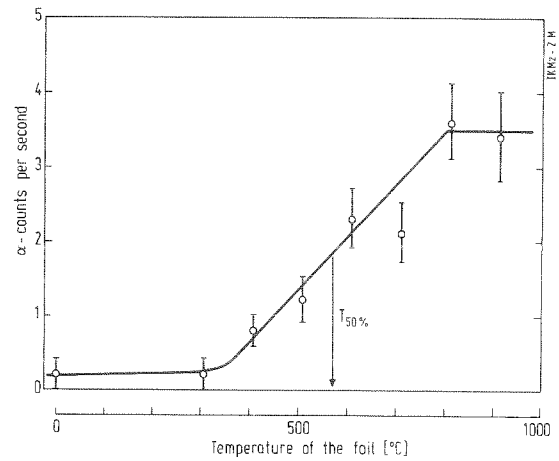

Fig. 1 Transmission of ^{211m}Po through a silver cylinder in a quartz tube as a function of the temperature

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Gaschromatographic Separations of Volatile Lanthanide Complexes

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The applicability of gaschromatographic methods for fast separations of actinides can be explored with lanthanides due to their similar chemical and physical properties. Lanthanide compounds volatile in the temperature region between 150 °C and 600 °C exist in form of ligand complexes with, e.g., hexafluoro-acetylacetone (HHFA) and tri-n-butylphosphate (TBP)[1] or Lewis-complexes with aluminumtrichloride[2]. In this work, first results of gaschromatographic separations of lanthanide complexes are described.

The lanthanide tracers were either produced by activation or by fission of ^{235}U with subsequent separation of the lanthanides by thermochromatography. The experimental set-up is shown in Fig. 1 with the gaschromatograph (1) as the main part. Anhydrous lanthanide-chlorides (2) were placed in a furnace (3) and heated up to 800 °C. A stream of dry hydrogen chloride (0.4 l/min) was fed into the column, consisting of a 4 m long quartz-tube with 4 mm inner diameter filled with quartz powder (64 mesh/cm^2). The separation-procedure was started by transferring a boat filled with $AlCl_3$ (4) in the furnace position. After passing the column, the separated complexes were catched in a cooled (-30 °C) trap (5) placed in front of a shielded Ge(Li)-detector (6). Integral chromatograms were measured in the multispectra mode. The complexes of the lanthanides with HHFA/TBP were synthesized with a solvent extraction method[1] and the resulting cyclohexane solution was directly syringed into the injector of the gaschromatograph. In these experiments, an 1 m long glass column with 2.4 mm inner diameter, filled with Chromosorb G (80-100 mesh) was used. The carrier gas was pure nitrogen.

Fig. 2 shows a chromatogram obtained with HHFA/TBP-complexes. The measured γ-activity is plotted in relative units against the retention time. The temperature rise was programmed between 100 and 300 °C with a constant rate of 30 °C/min. The separation of the individual lanthanide complexes is rather good, whereas the yields decrease with increasing retention temperature and retention time. The lanthanides appear in a sequence of increasing ionic radii, i.e. decreasing atomic numbers. Similar results were obtained with $AlCl_3$-complexes; here the retention times are, in general, shorter. In another series of experiments, the adsorption enthalpies of Sm-complexes were determined by varying the temperature of the gaschromatograph. The resulting values were -13 kJ/mol for the $AlCl_3$-complex on quartz and -48 kJ/mol for the HHFA/TBP-complex on Chromosorb G. These values are small compared with, e.g., the adsorption enthalpy of ceriumchloride on quartz (-290±49 kJ/mol)[3]. This gives hope for a fast separation of lanthanides and actinides by gaschromatography.

[1] W.C. Butts et al., Anal.Chem. 42, 173 (1970)
[2] D.M. Gruen and H.A. Øye, Inorg.Nucl.Chem.Letters 3, 453 (1967)
[3] U. Hickmann et al., Nucl.Instr.Meth. 174, 507 (1980)

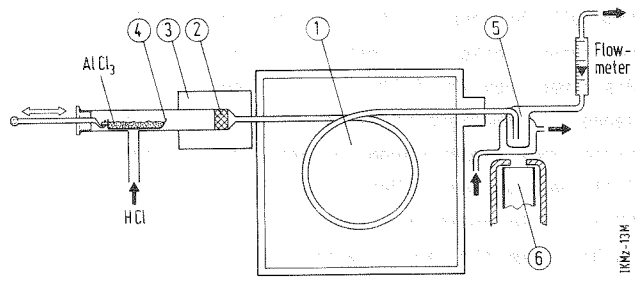

Fig. 1 Gaschromatographic set-up

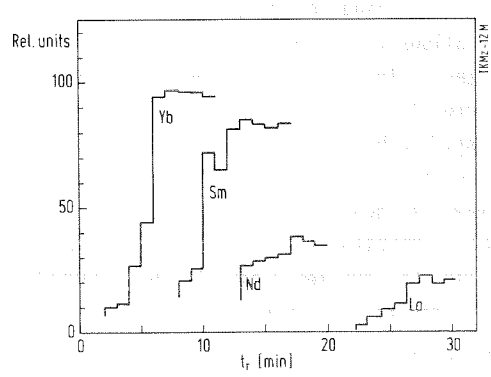

Fig. 2 Gaschromatogram of different lanthanides with HHFA/TBP as complexing agent

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Development of a Fast Automatic High Performance Liquid Chromatography System

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Studies of damped collisions of heavy ions with actinide targets indicate [1,2] that the production of new neutron-rich isotopes of the heaviest actinides (notably Md and No) is within reach. In order for these nuclei to be identified it is necessary to isolate them by fast and specific chemical separations from huge amounts of interferring α-particle and spontaneous fission activities.

We are presently developing a microprocessor-controlled chemistry system which combines three techniques: 1) The reaction products are transported by a KCl-loaded gas jet from the beam line to the chemistry system where they are collected on a quartz filter. The collection time will be equal to the time required for the following chemical separation. 2) The activities are dissolved from the filter and are then subject to subsequent chromatographic separations using high performance liquid chromatography (HPLC) columns and filterbeds as shown schematically in Fig. 1. The first HDEHP column achieves a rough separation of No^{2+} as well as Md^{3+} from the lighter actinides. The No-fraction is then passed through a $BaSO_4$ filter bed to retain Ra and Pb and is then cleaned on a cation exchange column in the HCl system. The Md-fraction from the first HDEHP column, after being passed through an anion exchange filter bed is further separated from other trivalent activities on a cation exchange column with α-hydroxy-isobutyric acid (α-HiB) at pH=4. Absorption of Md^{3+} on a small cation exchange column and subsequent elution with HCl allows to remove the α-HiB. 3) For both the No- and the Md-fraction the effluent from the last column is collected in a rotating titanium crucible, evaporated to dryness, and No and Md are then volatilized[3] from the Ti-surface at 1200°C onto a cooled 1 μm thick Ni-foil which serves as substrate for the counting samples. The sequence involves also the reconditioning of used columns, so that the separation can be repeated many times in order to collect sufficient counting statistics.

All parts of the chromatography system that are exposed to aqueous solutions including the HPLC pump are made from chemically inert materials such as Teflon, KEL-F, glass, and sapphire. The pump achieves a flow-rate of 0.1 to 10 ml/min at a maximum pressure of 35 bar. The components of the system are connected by Teflon tubing of 0.8 mm i.d. and by high-pressure slider-valves. The slider valves are operated pneumatically via 24 V (DC) / 11 W magnetic valves. The system is controlled by a PDP11/03 computer with 4 parallel ports and a serial port connected to a teletype. Each parallel port is lined with a 16 channel driver unit. Each channel supplies 24 V (DC) for operation of a magnetic valve and can be used, in addition, to check certain conditions of the system. An assembler program provides the user with a simple command language and carries out the command sequence operating the complete chemical separation.

Components 1) through 3) have been tested separately, so far. An apparatus for the automatic sample preparation is under construction. Operation of the complete system is anticipated for 1982.

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[1] H. Gäggeler et al., Proc. 4th Int. Conf. on Nuclei far from Stability, Helsingør 1981, CERN 81-09 p. 763

[2] D. Lee et al., Report LA-UR-81-1193 (1981)

[3] C. Frink, Diplomarbeit, Inst. f. Kernchemie, Universität Mainz (1981)

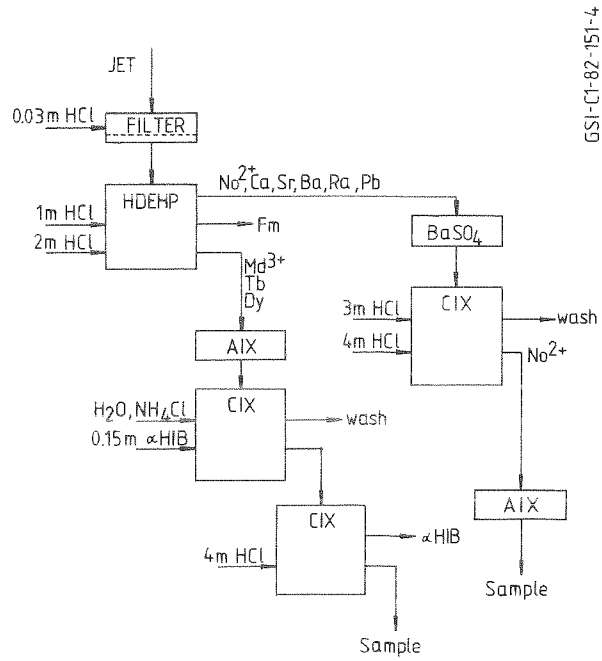

Fig. 1