

Transfer Cross Sections from the Reaction of  $^{48}\text{Ca} + ^{254}\text{Es}$

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We bombarded a  $4.8\text{-}\mu\text{g}/\text{cm}^2$   $^{254}\text{Es}$  target, electrodeposited on a thin beryllium foil, with 266-MeV  $^{48}\text{Ca}$  ions from the SuperHILAC at the Lawrence Berkeley Laboratory. This energy is about 11% above the Coulomb barrier, which generally provides the maximum transfer cross sections for heavy actinides.

The reaction products recoiling from the einsteinium target were collected on a  $6.3\text{ mg}/\text{cm}^2$  thick copper catcher foil. Following each of two short ( $\sim 2$  h) bombardments, the catcher foil was transported to a laboratory for separation of the actinide products before isotopic analysis by alpha spectroscopy with surface-barrier detectors. Figure 1 shows the measured cross sections for the actinide products.

The steady decrease in cross section for each transferred proton resembles the results from heavier systems like  $^{238}\text{U} + ^{248}\text{Cm}^1$ , and it does not show the relative enhanced yields for two proton transfer products as observed with lighter projectiles like  $^{18}\text{O}$  and  $^{22}\text{Ne}^2$ .

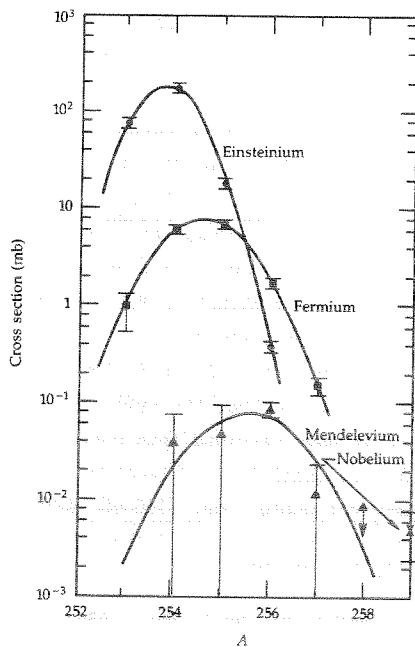


Fig.1: Measured cross sections for isotopes of Es, Fm, Md and No produced in the bombardment of  $^{254}\text{Es}$  with 266-MeV  $^{48}\text{Ca}$  ions.

Fig.2 shows a comparison of cross sections for the production of (a) Fm isotopes ( $\Delta Z=1$ ), and (b) Md ( $\Delta Z=2$ ) and No ( $\Delta Z=3$ ) isotopes in the bombardment of  $^{254}\text{Es}$  with 98 MeV  $^{18}\text{O}$ , 121 and 126 MeV  $^{22}\text{Ne}$ , and 266 MeV  $^{48}\text{Ca}$ . Although the Fm cross sections from all the irradiations are comparable, the Md cross sections are more than one order of magnitude lower for the  $^{48}\text{Ca}$  bombardment. We may gain a better qualitative understanding for this from considerations where the level density of the final system is related to the product excitation energy,  $E^*$ , as do Hoffman and Hoffman<sup>3</sup>. They described the cross section as being roughly proportional to  $\exp(-c E^*)$ . However, differences in transfer probabilities and the kind of transfer mechanism for projectiles like  $^{18}\text{O}$ ,  $^{22}\text{Ne}$  and  $^{48}\text{Ca}$  may also play an important role.

1. M. Schädel et al., Phys. Rev. Letter **48**, 852 (1982)
2. M. Schädel et al., GSI Scientific Report 1982, p. 67 (1983), and contribution to this report
3. D. C. Hoffman and M. M. Hoffman, Report LA-UR-82-824 (1983)

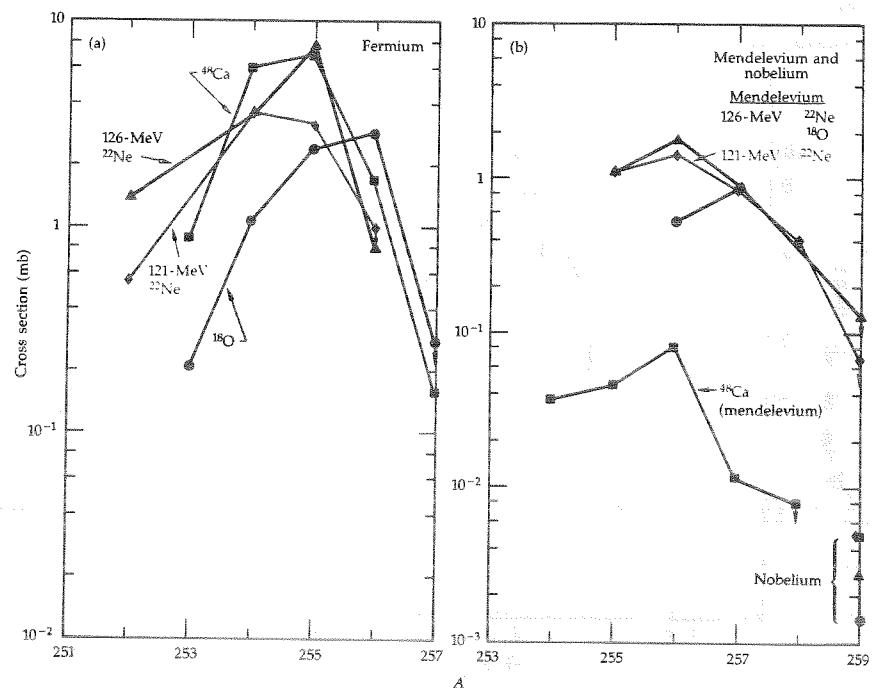


Fig.2: (a) Comparison of cross sections for the production of Fm isotopes in the bombardment of  $^{254}\text{Es}$  with 98-MeV  $^{18}\text{O}$  (dot), 121-MeV  $^{22}\text{Ne}$  (diamond), 126-MeV  $^{22}\text{Ne}$  (triangle), and 266-MeV  $^{48}\text{Ca}$  (square). (b) Production of Md and No isotopes.

Equilibration of the Neutron to Proton Ratio for Heavy Asymmetric Systems in Heavy Ion Collisions

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The pending question whether N/Z-equilibration between complex nuclei is caused by a collective mode of high frequency analogous to the E1 giant resonance<sup>1</sup> or whether this equilibration results from the statistical exchange of independent nucleons<sup>2</sup> has attracted much theoretical interest. The models available at present differ by 1. the choice of the collective variables assuming hereby correlated motion of neutrons and protons or, conversely, independent particle propagation, 2. the transport equations, 3. the transport coefficients, and 4. the details of the collision dynamics.

We have confronted new experimental data on nuclide distributions in damped collisions of  $^{208}\text{Pb}$  with  $^{74}\text{Ge}$  and of  $^{86}\text{Kr}$  with  $^{197}\text{Au}$  measured<sup>3</sup> with a large area position-sensitive ionisation chamber and published radiochemical data for  $^{132}\text{Xe} + ^{197}\text{Au}$  - collisions<sup>2</sup> with the predictions of three models. In the case of the  $^{208}\text{Pb} + ^{74}\text{Ge}$  and  $^{86}\text{Kr} + ^{197}\text{Au}$  data we have folded the theoretical distributions i) with resolution functions to take into account the finite Z- and A-resolution of the detection system and ii) with the particle evaporation pattern. The resulting theoretical (secondary) distributions are then compared to the uncorrected data. For the  $^{132}\text{Xe} + ^{197}\text{Au}$  system the published data are representative of the primary distributions and are thus compared directly to the theoretical predictions.

Good agreement between experiment and theory is observed for the marginal and conditional variances for the quantal model<sup>4</sup> of Hofmann et al.. Here, however, the frequency-dependent transport coefficients were not calculated microscopically but were assumed to be constants and were fixed empirically by the asymptotic properties of the system. Also, the transfer of energy from the relative motion to the intrinsic degrees of freedom is parametrized empirically. In addition, the phonon energy of the isovector giant resonance is treated as a constant fixed by the experimental data. Some of these apparent deficiencies were removed in the model of Grégoire et al.<sup>5</sup> who generalized the transport equation of Hofmann to several degrees of freedom. However, the phonon energies for the N/Z-mode are still determined phenomenologically. Fig. 1 depicts for the  $^{86}\text{Kr} + ^{197}\text{Au}$  system the agreement between the (secondary) theoretical results and the experimental data. As a second example we show in Fig. 2 a comparison of the charge widths at fixed primary mass asym-

metry for  $^{132}\text{Xe} + ^{197}\text{Au}$  with the model of Gross and Hartmann<sup>6</sup> which treat on a microscopic level the stochastic exchange of independent particles. Their transport equation is a Fokker-Planck equation with overdamped motion. We have used here a version where dynamical deformations of the colliding nuclei are considered. This enhances nucleon exchange in the entrance channel. All three models, even though quite different in the inherent physical assumptions, seem to reproduce the experimental variances at least qualitatively. Thus, our attempts to analyse data with existing dynamical models in their present stage of development have not succeeded to discriminate between quantal versus statistical fluctuations.

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References

1. M. Berlinger et al., Z. Phys. **A291**, 133 (1979)
2. J.V. Kratz et al., Nucl. Phys. **A357**, 437 (1981)
3. R. Lucas, Thesis, Orsay (1983)
4. H. Hofmann et al., Z. Phys. **A293**, 229 (1979)
5. C. Grégoire et al., Phys. Lett. **99B**, 17 (1981)
6. D.H.E. Gross et al., Phys. Rev. **C24**, 25 (1981) and Z. Phys. **A299**, 63 (1981)

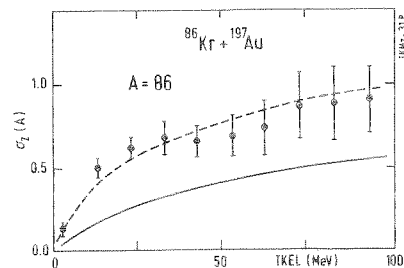


Fig.1: Comparison of the (secondary) charge widths at fixed mass for Kr + Au with the prediction of the Grégoire model<sup>5</sup>. The dashed line results from the primary widths (solid line) after correction for particle evaporation and limited Z and A resolution.

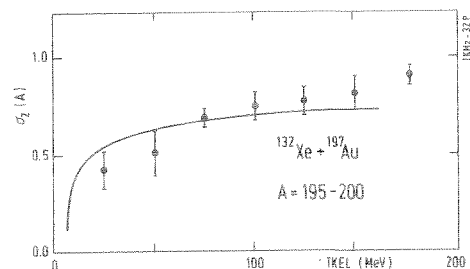


Fig.2: Charge widths at fixed primary mass for Xe + Au and the prediction of the Gross-Hartmann model<sup>6</sup> with dynamical deformations of the colliding nuclei.

Angular Distributions for Symmetric Fragmentation in  $^{50}\text{Ti} + ^{208}\text{Pb}$  Collisions

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In studies of angular distributions for fragments with about half the mass of the combined system it was observed for the heaviest systems like  $^{208}\text{Pb} + ^{58}\text{Fe}$  /1/ that there is a way from the entrance channel to a symmetric exit channel that bypasses the slow process of compound nucleus formation. In an attempt to search for angular momentum limits for the coexistence of complete fusion-fission, fast-fission, and deep-inelastic scattering, the angular distributions for symmetric fragmentations had been reexamined in the  $^{56}\text{Fe} + ^{208}\text{Pb}$  system /2/ at  $E/B = 1.1, 1.2, 1.3,$  and  $1.6$  using catcher foil techniques and off-line KX-ray spectroscopy. For all values of  $Z$  ( except for  $(Z_1 + Z_2)/2$  ) angular distributions were observed that were asymmetric around  $\theta_{\text{cm}} = 90^\circ$ . The distributions were forward peaked for  $Z \leq (Z_1 + Z_2)/2$  and backward peaked for the heavy complements /2/. The degree of forward-backward asymmetry and the mean anisotropies were largest at the smallest energy ( $\ell_{\text{crit}} = 35 \hbar$  in a sharp cutoff approximation) and decreased as the bombarding energy increased /2/. This might be seen as evidence for a dynamical hindrance for compound nucleus formation even below  $\ell_{(B-f=0)} = 37 \hbar$  and might help localizing the value of  $x_{\text{cliff}}$  /3/. Then, in a less critical system one might expect to see true compound nucleus fission at low  $\ell$  values and a change to forward-backward asymmetric angular distributions at higher angular momenta.

We examined the system  $^{50}\text{Ti} + ^{208}\text{Pb}$  at energies  $E/B = 1.06$  and  $1.16$  using the same technique as mentioned above. Relative cross sections for  $40 \leq Z \leq 83$  between  $10^\circ$  and  $178^\circ$  in the center-of-mass system were measured. The results at  $E/B = 1.06$  for two elements ( $Z=47, 54$ ) near the symmetric charge split  $Z=52$  are depicted in Figs.1,2. The solid curves are calculations in terms of the standard fission theory /4/. Within error limits, the distributions seem to be symmetric around  $90^\circ$ . When calculating the maximum number of partial waves contributing to the fusion cross section /1/ in a triangular sharp cutoff approximation, we arrive at a value of  $K_o^2 = 25$ , which is much lower than expected for a compound nucleus reaction. Larger anisotropies than expected from saddle point shapes of the rotating liquid drop model have been reported even for lighter systems /5,6/.

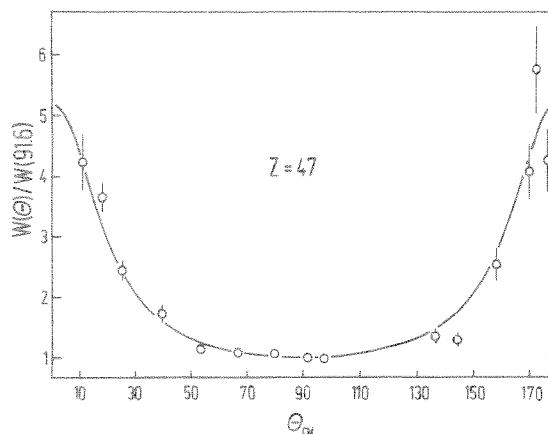


Fig 1 : Angular distribution for  $Z=47$  in the reaction  $^{50}\text{Ti} + ^{208}\text{Pb}$  at  $E/A = 5.0$  MeV/A. The cross sections  $d^2\sigma/dZd\theta$  are normalized to the one which is closest to  $90^\circ$ .

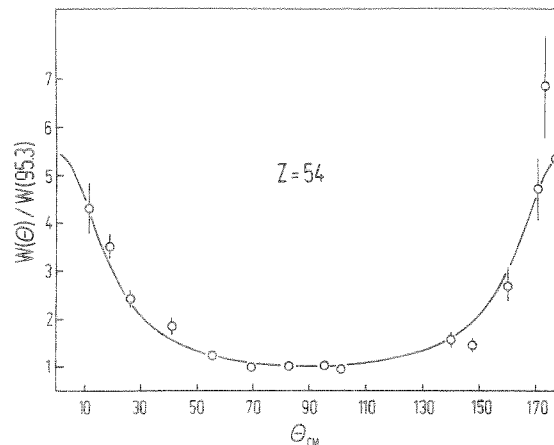


Fig 2 : Angular distribution for  $Z=54$  in the reaction  $^{50}\text{Ti} + ^{208}\text{Pb}$  at  $E/A = 5.0$  MeV/A.

- /1/ R. Bock et al., Nucl. Phys. **A388**, (1982) 334
- /2/ G. Guarino et al., GSI Annual Report 1981
- /3/ S. Bjørnholm and W. J. Swiatecki, Nucl. Phys. **A391**, (1982) 471
- /4/ R. Vandenbosch, J.R. Huizenga, Nuclear Fission ( Academic Press 1973 )
- /5/ H. Rossner et al., Phys. Rev. **C27**, (1983) 2666
- /6/ M.B. Tsang et al., Phys. Rev. **C28**, (1983) 747

„Fragmentation“ in the system  $^{197}\text{Au} + ^{197}\text{Au}$  at energies  $\leq 15 \text{ MeV/u}$ <sup>B</sup>

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Medium-heavy fragments (MHF in the mass range 10 to 40) are observed in many different reactions, e.g. in proton- or heavy-ion induced reactions at relativistic energies as well as in systems like  $^{129}\text{Xe} + ^{122}\text{Sn}$  or  $^{92}\text{Mo} + ^{92}\text{Mo}$  at about  $10\text{-}15 \text{ MeV/u}$ <sup>1-4</sup>. At present, it is not clear whether "fragmentation" is a general decay property of hot heavy nuclei which accounts for the formation of MHF in all the above mentioned reactions or whether there are several different mechanisms producing fragments in the same mass range.

We therefore started studies of the fragmentation of  $^{197}\text{Au}$  nuclei by different projectiles. Gold was chosen as target because it is much heavier than the MHF we are interested in. At CERN, a bombardment with  $^{20}\text{Ne}$  at  $49 \text{ MeV/u}$  is planned. At the UNILAC  $^{197}\text{Au}$  at energies  $\leq 15 \text{ MeV/u}$  was used as a projectile. The highest excitation energy per  $^{197}\text{Au}$  nucleus in both systems is of the same order of magnitude ( $\sim 500 \text{ MeV}$ ) but the light projectile is faster than the Fermi velocity of the bound nucleons while the heavy projectile is slower. Thus in the heavy system thermalization of the excitation energy becomes conceivable. Therefore one might expect a rather dramatic change in the reaction mechanism. An indication of such a change is indicated by the behaviour of the excitation function for the formation of e.g.  $^{24}\text{Na}$  from  $^{197}\text{Au} + ^{197}\text{Au}$  which we measured at  $E_{\text{lab}} = 9, 11, 13$  and  $15 \text{ MeV/u}$ . We calculated the ratio of the  $^{24}\text{Na}$  cross section to the total reaction cross section, hereby using the classical formula  $\sigma_r = \pi R^2(1 - V_C/E_{\text{CM}})$ . This ratio is shown in the figure and compared to that for formation of  $^{24}\text{Na}$  from  $^{197}\text{Au}$  and  $^{208}\text{Pb}$  in proton<sup>1</sup> and pion<sup>5</sup> bombardments. With the slow projectile, the fraction of the  $^{24}\text{Na}$  cross section is larger by two orders of magnitude.

For the  $^{197}\text{Au} + ^{197}\text{Au}$  reaction, it seems unlikely that the MHF originate from the first step of binary, deeply inelastic collisions: This is demonstrated by the distribution of projected ranges of  $^{24}\text{Na}$  which we measured at  $15 \text{ MeV/u}$  at lab angles  $\theta_{\text{lab}} \leq 45^\circ$ . It shows a maximum at the velocity of the center of mass. If one calculates the possible velocities of  $^{24}\text{Na}$ -nuclei with the assumption that they are emitted in binary reactions with energies equal to or larger than the Viola energy one finds that in the vicinity of the velocity of the center of mass there should not appear any light fragments at all. Furthermore, the widths of the element distributions of binary HI reactions can be estimated from a

universal, empirical formula found by Wollersheim et al.<sup>6</sup>. Application of this formula to our system shows that the widths are by far too small for such reactions to account for the large yields of  $^{24}\text{Na}$ .

For these two reasons, production of MHF in the first, binary step of a deeply inelastic collision can be ruled out, and other mechanisms must be considered, e.g. sequential break-up of a gold-like heavy fragment or true three body-reactions.

#### References

1. R. Wolfgang et al., Phys. Rev. **103**, 394 (1956)
2. H. H. Gutbrod et al., Nucl. Phys. **A387**, 177c (1982)
3. P. Glässel et al., Z. Phys. **A310**, 189 (1983)
4. S. Gralla et al., GSI Scientific Report 1982, p. 22
5. N. Porile et al., Phys. Rev. **C18**, 2231 (1978)
6. H. J. Wollersheim et al., Phys. Rev. **C25**, 338 (1982)

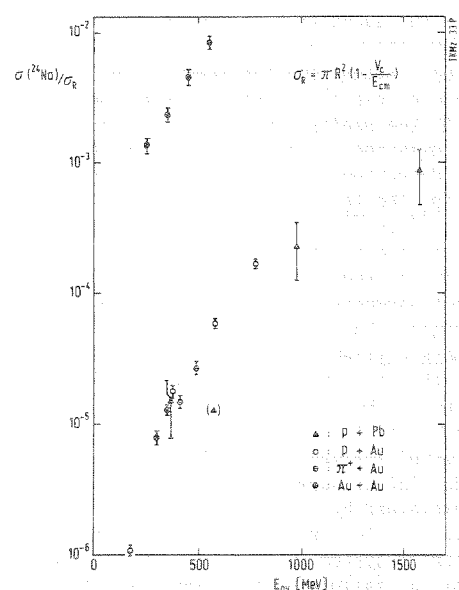


Figure: Ratios of cross sections for the formation of  $^{24}\text{Na}$  to the total reaction cross sections  $\sigma_r = \pi R^2(1 - V_C/E_{\text{CM}})$  versus available energy. The  $^{197}\text{Au} + ^{197}\text{Au}$  data are from this work, those for the proton and pion bombardments from refs. 1 and 5.

Studies on Short-Lived Praseodymium Isotopes at the HELIOS Facility

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The investigations for an efficient mass separation of the lanthanides<sup>1</sup>, which serve as model elements for the heavier actinides, have been continued at the helium jet on-line mass separator facility HELIOS. With  $\text{PbCl}_2$  as cluster material and an integrated skimmer-high temperature ionization source overall efficiency values of a few percent were obtained for these elements. Furthermore decay studies on neutron-rich praseodymium isotopes, produced by thermal neutron-induced fission of  $^{235}\text{U}$ , have been performed by  $\gamma$ -spectroscopic measurements. A fast microprocessor controlled tape system<sup>2,3</sup> was used to transport the mass separated activities from the focal plane of the separator to the detector position.

The  $\beta^-$ -decay of 2.3 min  $^{148}\text{Pr}$  has been studied by  $\gamma$ -singles,  $\gamma$ - $\gamma$ -coincidence and  $\gamma$ - $\gamma$ -angular correlation measurements. In the latter mode  $5 \times 10^6$  coincidence events were collected per angle (at 90, 135, 180, 225, and  $260^\circ$ ) resulting from a total counting time of 40 h. The data analysis has not yet been completed.

With an upgraded  $^{235}\text{U}$ -target (18 mg instead of 6.5 mg) the identification of  $^{152}\text{Pr}$  and studies on its decay properties became possible. In a  $\gamma$ -multiscaling experiment, using the  $\beta$ -gate technique to suppress the background in the  $\gamma$ -ray spectra, the half-life of  $^{152}\text{Pr}$  was determined to be  $3.8 \pm 0.2$  s from the decay of the strongest  $\gamma$ -lines at 164, 285 and 1469 keV (Fig. 1). In this experiment the mass separated activity was collected for 16 s, transported to the Ge(Li)-detectors within  $\sim 2$  s and measured for  $8 \times 2$  s. Altogether 561 cycles were summed up.

In a second experiment  $7.5 \times 10^6$   $\gamma$ - $\gamma$ -coincidence events from the  $\beta^-$ -decay of 3.8 s  $^{152}\text{Pr}$  have been collected which allowed to establish a partial decay scheme for  $^{152}\text{Pr}$  thus far not yet well known<sup>4</sup>.

<sup>1</sup>M. Brügger et al., GSI Scientific Report 1981, GSI 82-1, p. 225 (1982)

<sup>2</sup>M. Brügger et al., GSI Scientific Report 1982, GSI 83-1, p. 229 (1983)

<sup>3</sup>M. Brügger et al., to be published

<sup>4</sup>J.C. Hill et al., Phys. Rev. C27, 2857 (1983)

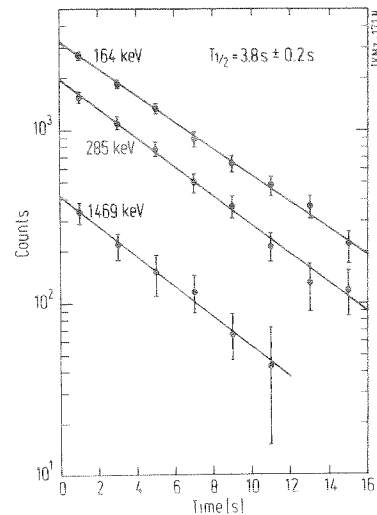


Fig. 1: Decay of the strongest  $\gamma$ -lines of  $^{152}\text{Pr}$

A Search for New Isotopes from Reactions of  $^{16}\text{O}$  and  $^{22}\text{Ne}$  with  $^{254}\text{Es}$

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From previous experiments with  $^{16}\text{O}$  and  $^{22}\text{Ne}$  as a projectile and  $^{254}\text{Es}$  ( $T_{1/2} = 276\text{d}$ ) as a target cross sections for longer lived isotopes of elements Fm to No were obtained<sup>1</sup>. Based on these results, and from a comparison with isotope cross sections from similar transfer channels in reactions with  $^{248}\text{Cm}^2$ , we estimated cross sections between 1  $\mu\text{b}$  and 10 nb for yet unknown neutron-rich isotopes of elements 101 through 104. Expected half-lives for these nuclides range from one second up to a few hours.

We irradiated a 30  $\mu\text{g}/\text{cm}^2$   $^{254}\text{Es}$  target with intense beams ( $\leq 300\text{pA}$ ) of  $^{16}\text{O}$ ,  $^{22}\text{Ne}$  and  $^{16}\text{O}$ . Reaction products, recoiling from the Es target, were stopped in a KCl loaded He gas. Whereas the previous experiments<sup>1</sup> were performed with off-line chemistry methods, now on-line techniques were applied.

In one series of experiments products were rapidly transported ( $\sim 200\text{ms}$ ) to our rotating wheel multidetector apparatus, ROMA<sup>3</sup>. The activity deposited on thin plastic foils was moved stepwise to 15 sequential counting stations for  $\alpha$ - and sf-measurements. The irradiations with 98 and 104 MeV  $^{16}\text{O}$  (2-60 s collection times) gave a rather flat background which tended to obscure weak  $\alpha$ -peaks. However, several not readily identifiable  $\alpha$ -peaks were seen in addition to the one from  $^{260}\text{Lr}$ . In 127 MeV  $^{22}\text{Ne}$  bombardments (2-180 s collection times) the  $\alpha$ -spectra were cleaner, and we clearly observed peaks of  $^{256,257}\text{No}$  and  $^{256,260}\text{Lr}$ . The main  $\alpha$ -group of  $^{259}\text{Lr}$  was obscured by the  $^{256}\text{No}$  peak. In this experiment ROMA was operated in a special mode<sup>3</sup> to enhance the observation of  $\alpha$ -events after the possible decay of  $^{265}\text{105}$ . In an on-line analysis no events were observed that could be assigned as a daughter coming from the decay of  $^{265}\text{105}$ . There was also no indication for  $^{263}\text{105}$  from a 101 MeV  $^{16}\text{O}$  bombardment. In the  $\alpha$ -spectrum from this experiment (Fig.1),  $^{255-257}\text{No}$  and  $^{258-260}\text{Lr}$  can clearly be identified. For final results we have to await the off-line analysis.

The carbon foils covering the detectors as a catcher for nuclei from  $\alpha$ -recoils were chemically processed off-line shortly after each of the three beam runs. This was done to search for long-lived daughters such as  $^{257}\text{Fm}$  and  $^{258}\text{Md}$ . Long-term counting is still continuing.

A second series of experiments with the purpose of detecting those new nuclides with half-lives between several minutes and days whose  $\alpha$ -energies would be masked in the on-line counting was carried out with the automated rapid chemistry apparatus, ARCA<sup>4</sup>. The measurements of chemically separated fractions of the elements No, Lr and Md began 7, 17 and 19 min after end of bombardment, respectively. Experiments were carried out with  $^{16}\text{O}$  and  $^{22}\text{Ne}$  as a projectile.

A careful analysis of the large amount of data accumulated is under way to search for unknown isotopes. We will be able to determine cross sections of nearly all of the known heavy isotopes up to  $^{269}\text{Lr}$ , which will provide a large amount of new data for further and better extrapolations. For the time being, extrapolations from preliminary data indicate that the unknown isotopes  $^{260,261}\text{Md}$ ,  $^{260,261}\text{No}$  and  $^{261,262}\text{Lr}$  should be produced with 1.0 to 0.1  $\mu\text{b}$  cross sections. However, alpha emission from these nuclides were not readily observed.

1. M. Schädel et al., Proc. Int. Workshop on Gross Properties of Nuclei and Nuclear Excitations p. 16, XI, Hirschegg (1983), and Proc. Int. Conf. Nucl. Phys., Vol. 1, p. 625, Florence (1983)
2. D. Lee et al., Phys. Rev. C 25, 286 (1982)
3. K. Sümmerer et al., contribution to this report.
4. W. Brüche et al., contribution to this report

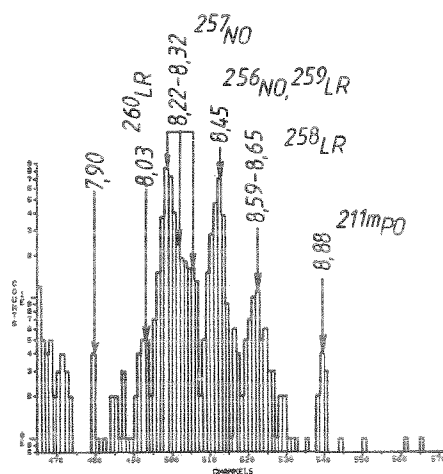


Fig.1:  $\alpha$ -spectrum accumulated during a 12 hour irradiation of  $^{254}\text{Es}$  with 101 MeV  $^{16}\text{O}$ . Data are from the first detector of ROMA<sup>3</sup> taken with a collection time of 4 s.

Attempts to Produce Superheavy Elements in the  $^{48}\text{Ca} + ^{248}\text{Cm}$  Reaction Applying On-Line Chemical Techniques

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Fusion of  $^{48}\text{Ca}$  with  $^{248}\text{Cm}$  to form the compound nucleus  $^{296}116^{100}$  is considered to be one of the most promising reactions for the synthesis of superheavy nuclei. Previous attempts to discover superheavy elements by this reaction failed, however. A possible reason is the relatively high excitation energy, some 40 MeV, introduced by choosing a projectile energy about 20 MeV above the interaction barrier<sup>1</sup>. More recent studies show that fusion in similar systems occurs at or even below the barrier.

These arguments led to a further attempt to search for superheavy elements by this reaction, however, at lower excitation energies of the compound nucleus  $^{296}116$  of some 16 to 40 MeV. The whole program consisted of several independent experiments designed to cover

- (i) the half-life region from microseconds to days in thin target experiments with the gas-filled mass separator SASSY at Berkeley and the velocity filter SHIP at GSI<sup>2</sup>
- (ii) the half-life region from seconds to years in thick target experiments with radiochemical techniques carried out off-line to cover the long half-lives and on-line giving access to the short-lived species.

In a first series of experiments which were performed in October 1982 at LBL classical off-line chemical techniques were applied to separate spontaneously-fissioning superheavy elements from other reaction products. These attempts are described in the previous report<sup>3</sup>. In a second series of experiments performed in 1983 on-line chemical techniques were used to get access to much shorter-lived isotopes with half-lives down to the second range. These techniques, combinations of a gas-jet transportation system with several automated chemical separation procedures are described elsewhere<sup>4-6</sup>. The methods are based on the following predictions about chemical properties of superheavy elements around  $Z=114$ :

- (i) Elements 112 through 117 should be volatile in their elemental state at temperatures up to about 1000 °C<sup>4</sup>.
- (ii) Some elements such as 112, 114 and 118 may even be volatile at room temperature<sup>5</sup>.
- (iii) Elements 108 through 116 should form strong anionic bromide complexes in aqueous solutions<sup>6</sup>.

For each of these three separations a  $^{248}\text{Cm}$  oxide target (1,72 mg/cm<sup>2</sup>) was bombarded by 233-248 MeV  $^{48}\text{Ca}$  projectiles (measured energy range within the target) with beam integrals of  $(0.7-1.4)\times 10^{17}$  particles. The samples were counted on-line for alpha decay and spontaneous-fission events. The observed  $\alpha$ -peaks can be attributed to isotopes of astatine, polonium, bismuth radon and lead. In all experiments no indication for any unknown  $\alpha$ -line nor for any fission activity was found. In the Figure the resulting upper cross section limits on a 95 % confidence level are plotted for spontaneously-fissioning superheavy nuclei versus the half-life. In addition to the results from the chemistry experiments, the Figure also depicts the cross section limits obtained from the search experiments applying the SHIP and the SASSY systems<sup>2</sup>.

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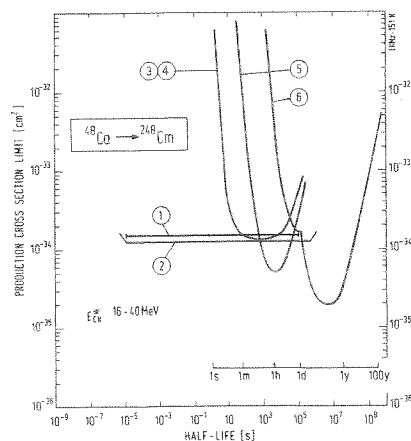
#### References

1. see e.g. J.V. Kratz, *Radiochim. Acta*, **32**, 25 (1983)
2. G. Münzenberg et al., this Scientific Report, p.
3. H. Gäggeler et al., GSI Scientific Report 1982, GSI-83-1, p. 69 (1983)
4. H. Dornhöfer et al., GSI Scientific Report 1981, GSI 82-1, p. 216 (1983)
5. N. Hildebrand et al., GSI Scientific Report 1982, GSI-83-1, p. 234 (1983)
6. W. Brüche et al., this Scientific Report, p.

#### Figure:

Cross section limits for the production of spontaneously-fissioning superheavy elements, obtained with the

- 1: Small angle separating system SASSY (LBL)
- 2: Velocity filter SHIP (GSI)
- 3: High temperature on-line gas phase chemistry (GSI)
- 4: Low temperature on-line gas phase chemistry (GSI)
- 5: Quasi on-line solution chemistry (GSI)
- 6: Off-line chemistry, procedures as 3-5 (LBL, GSI)



A Search For Superheavy Elements Using the  $^{48}\text{Ca} + ^{254}\text{Es}$  Reaction

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We measured upper limits for the production of superheavy elements (SHEs), using the compound-nucleus reaction  $^{48}\text{Ca} + ^{254}\text{Es}$ . This combination permits the closest approach of any practical fusion reaction to the predicted island of stability at the 184-neutron shell. Theoretical calculations by Randrup et al.<sup>1</sup> and Fiset and Nix<sup>2</sup> predict that the maximum stability against spontaneous fission (SF) is at the closed 184-neutron shell for the superheavy elements. The much favored and studied  $^{48}\text{Ca} + ^{248}\text{Cm}$  reaction<sup>3,4</sup> would produce element 116 with 178 neutrons after emission of two neutrons from the compound nucleus. The calculations of Randrup et al.<sup>1</sup> show, however, that the fission-barrier heights decrease very rapidly below 184 neutrons, becoming only 3 to 4 MeV at 178 neutrons, with corresponding SF half-lives of  $10^{-16}$  y as compared with years or more at 184 neutrons. This sensitivity of SF half-life to neutron number for the SHEs dictates a very small choice of nuclear reactions that yield a product as close as possible to the 184-neutron shell. From this standpoint, the  $^{48}\text{Ca} + ^{254}\text{Es}$  reaction is the most favorable compound-nucleus reaction currently available. Randrup et al.<sup>1</sup> predict fission barriers of greater than 6 MeV for the 2n out-product of the einsteinium reaction, with SF half-lives comparable to the alpha half-lives.

We bombarded a  $5\text{-}\mu\text{g}/\text{cm}^2$   $^{254}\text{Es}$  target with  $^{48}\text{Ca}$  at the SuperHILAC accelerator at the Lawrence Berkeley Laboratory (Berkeley, CA). There were four bombardments, each lasting several hours, for a total of  $7.7 \times 10^{15}$   $^{48}\text{Ca}$  particles. Two  $^{48}\text{Ca}$  bombarding energies were used: 238 and 246 MeV. These energies are 2 MeV under and 6 MeV over the calculated Coulomb barrier, respectively. They were chosen to minimize excitation energy in the compound nucleus in order to minimize fission and the number of neutrons emitted during de-excitation.

We collected recoils from the einsteinium target on metal foils, which were then chemically processed to yield SHE fractions for SF counting. Two types of chemistry were applied based on predicted properties of SHEs<sup>5</sup>. First, a solution chemistry procedure using sulfide precipitation and cation exchange chromatography and, second, a gas-phase chemistry was performed<sup>5</sup>. The time needed to run the chemical

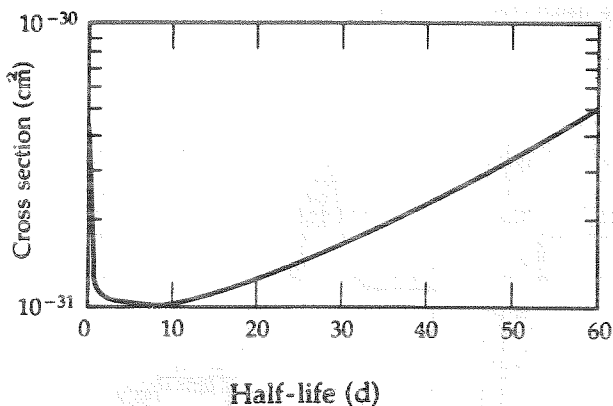
separations were about 0.5 - 2 hours and typical chemical yields for homologues of SHEs were 25 through 60%. Both types of chemistry provided excellent decontamination from the actinide elements, whose isotopes interfere by also decaying by SF. In over two months of counting, we observed no SF events in samples from either of the chemistries. By assuming the detection of two SF events in coincidence and a 50% chemical yield, we determine that the cross section for SHE production is  $\leq 5 \times 10^{-32}$  cm<sup>2</sup> for half-lives of a few days for all bombardments. The Figure shows the half-life vs cross-section limits we obtained for the two solution-chemistry experiments only.

We believe that the  $^{48}\text{Ca} + ^{254}\text{Es}$  reaction deserves further attention as a pathway to SHEs. The proposed production of 40  $\mu\text{g}$  of  $^{254}\text{Es}$  at Oak Ridge National Laboratory could produce a target 100 times larger than that used in our experiments. With a larger target and increased bombardment times, cross sections of about  $10^{-25}$  cm<sup>2</sup> are achievable.

#### References

1. J. Randrup et al., *Physica Scripta* **10A**, 60 (1974)
2. E. O. Fiset and J. R. Nix, *Nucl. Phys.* **A193**, 647 (1972)
3. H. Gäggeler et al., this Scientific Report, p.
4. G. Münzenberg et al., this Scientific Report, p.
5. see J. V. Kratz, *Radiochim. Acta*, **32**, 25 (1983)

Figure: Cross-section limits for the production of SHEs as a function of half-life, from solution-chemistry experiments.





Search for Volatile Reaction Products in the Reaction  $^{48}\text{Ca} + ^{248}\text{Cm}$

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For the search of superheavy elements in the reaction  $^{48}\text{Ca} + ^{248}\text{Cm}$  on-line chemical methods, based on the predicted volatility of the elements 112-118 and on the formation of strong anionic bromide complexes of the elements 108-116 have been applied<sup>1</sup>. The elements 112, 114 and 118 may even be volatile at room temperature and therefore a cryogenic chamber equipped with an annular surface barrier detector and a solar cell<sup>2</sup> has been used for the detection of very volatile reaction products. The reaction products from the bombardment of  $1.72 \text{ mg/cm}^2$   $^{248}\text{Cm}$ -oxide with 233-248 MeV  $^{48}\text{Ca}$  projectiles were transported with an argon gas-jet using pure argon to the cryo-system. In this way the transport efficiency for non-volatile elements was decreased. The volatile products were passed through a quartz tube filled with quartz powder (64-144 mesh/cm<sup>2</sup>) and heated to 1250 K to trap the clusters with attached non-volatile products formed during the irradiation. In a second quartz tube filled with tantalum foil (T=1000 K) traces of water were removed. After passing these traps the transport gas contains only noble-gases and very volatile products which were condensed together with the heavy noble gases in the cryo-system on a cooled solar cell (T=50 K) operating as a fission fragment detector. With an annular surface barrier detector installed in front of the solar cell  $E_1/E_2$  fission fragment measurements in coincidence with the photovoltaic cell and  $\alpha$ -spectroscopy were carried out. Furthermore single fission events could be measured with the ring detector. During the irradiation time  $\alpha$ -spectra were collected and afterwards multiscale measurements were performed to follow the decay of the deposited activities. Fig. 1 shows an  $\alpha$ -spectrum

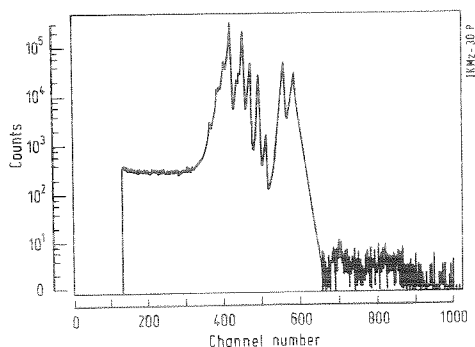


Fig. 1: On-line  $\alpha$ -spectrum of the gaseous products in the reaction  $^{48}\text{Ca} + ^{248}\text{Cm}$  measured for 23 h

measured on-line for 23 h during the deposition of the gaseous products. The main activities in the  $\alpha$ -spectra can be assigned to the radon isotopes 210-222 and their decay products. From the evaluated activities after the end of irradiation the production cross sections for the radon isotopes  $^{210}\text{Rn}$ ,  $^{211}\text{Rn}$ ,  $^{212}\text{Rn}$ ,  $^{219}\text{Rn}$ ,  $^{220}\text{Rn}$ ,  $^{221}\text{Rn}$ ,  $^{222}\text{Rn}$  were obtained. The results are shown in fig. 2.

During the whole experimental period no spontaneous fission event was observed yielding a cross-section limit for the production of spontaneously-fissioning superheavy elements as reported in another contribution<sup>1</sup> to this report.

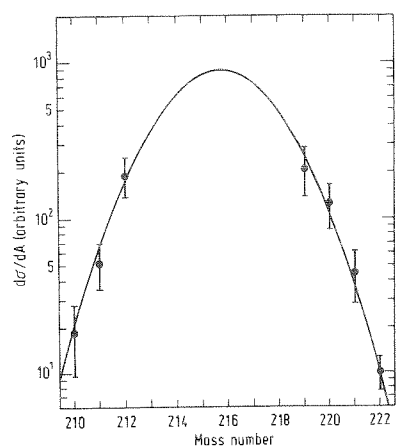


Fig. 2: Cross-sections for the production of radon isotopes in the reaction  $^{48}\text{Ca} + ^{248}\text{Cm}$

<sup>1</sup>H. Gäggeler et al., this Scientific Report 1983, p.

<sup>2</sup>N. Hildebrand et al., GSI Scientific Report 1982, GSI 83-1, p. 234 (1983)

Excitation Functions for Heavy Actinides from the Reaction of  $^{48}\text{Ca}$  with  $^{248}\text{Cm}$ 

B

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In 1982, a collaboration was started with the goal of trying once again to produce superheavy elements from the reaction of  $^{248}\text{Cm}$  with  $^{48}\text{Ca}$  projectiles at energies near the Coulomb barrier<sup>1,2</sup>. In addition we measured the excitation functions for the production of the actinides formed during such bombardments.

The target of curium oxide was prepared by stepwise electroplating and contained 1.7 mg/cm<sup>2</sup> of Cm (96.5%  $^{248}\text{Cm}$ , 3.5%  $^{246}\text{Cm}$ ). The target was mounted in a gas cooled target holder<sup>2</sup>. A gold or copper catcher foil of about 6 mg/cm<sup>2</sup> was placed behind the target to collect recoiling products. The  $^{248}\text{Cm}$  target was irradiated with  $E_{\text{lab}} = 239, 263$  and 288 MeV  $^{48}\text{Ca}$  projectiles. The energy loss within the target was measured to be about 16 MeV. After each bombardment, the catcher foils were dissolved and the actinides Pu, Bk, Cf, Es and Fm were chemically isolated and final samples were prepared on discs as described elsewhere<sup>3,4</sup>. These samples were analysed for alpha, spontaneous fission and gamma activities. The chemical yields for the actinide separations ranged from 50 to 75%.

The evaluated cross sections for the production of isotopes of Pu, Bk, Cf, Es and Fm are listed in Table I. The peak of the excitation functions for Cf, Es and Fm appear to occur at the middle energy of 263 - 247 MeV (within the target) which is about 20 MeV above the calculated barrier of 236 MeV. This observation is in agreement with estimates<sup>5</sup> based on ground state Q-values and the Coulomb barriers of the initial and final systems, which show that the excitation energy,  $E^*$ , for these products at the Coulomb barrier are negative by more than 10 MeV.

The yields for the Bk isotopes which are produced most likely in quasi-elastic reactions and for the below-target transfer isotopes of Pu are highest at the highest energy.

Independent of the bombarding energy, the mass distributions for Pu, Bk, Cf, Es, and Fm peak at the most probable mass numbers  $A_p$  of 243, 249, 250, 252 and 254, respectively. These maxima correspond within

one mass unit to the predicted primary most probable masses  $A_p$  calculated on the basis of potential energy considerations<sup>6</sup>. This tends to support the general idea of a binary transfer mechanism which results in a nucleus with low  $E^*$ , i.e. a "cold" nucleus which is not immediately destroyed by prompt fission or particle emission.

## References

1. G. Münzenberg et al., this annual report
2. H. Gäggeler et al., this annual report
3. D. Lee et al., Phys. Rev. C 25, 286 (1982)
4. M. Schädel et al., Phys. Rev. Lett. 41, 469 (1978)
5. D. C. Hoffman and M. M. Hoffman, Report LA-UR-82-824, March, 1982
6. H. Freiesleben and J. V. Kratz, Physics Reports, in press

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TABLE I. Cross sections for the production of heavy actinides in the bombardment of  $^{248}\text{Cm}$  with  $^{48}\text{Ca}$

	223-239(MeV) <sup>a</sup>		247-263(MeV) <sup>b</sup>		272-286(MeV) <sup>b</sup>	
	Cross section ( $\mu\text{b}$ )	s <sup>b</sup> (%)	Cross section ( $\mu\text{b}$ )	s <sup>b</sup> (%)	Cross section ( $\mu\text{b}$ )	s <sup>b</sup> (%)
Pu	243	295	17	515	17	1965
	245	113	19	261	17	1050
	246	22	50	115	18	413
Bk	245	11.7	22	39	15	66.5
	246	73	16	272	14	482
	248m	730	17	1690	17	2660
	250	690	11	2010	11	2920
Cf	246	0.25	5	1.4	2	1.0
	248	101	9	238	9	209
	250	413	22	2540	8	1935
	252	50.9	21	231	11	224
	253	0.81	17	8.2	21	4.0
	254	0.32	28	1.5	10	1.0
Es	252	9.46	7	28.4	4	24.3
	253	4.08	17	10.5	9	7.8
	254m	0.74	30	1.8	6	1.4
Fm	252	0.02	38	0.09	30	0.06
	254	0.28	6	0.94	6	0.71
	255	0.21	7	0.90	2	0.62
	256	0.06	30	0.25	10	0.14

- a) Range of projectile energy within the target (laboratory system)
- b) Statistical standard deviation based on the analysis of the decay data.

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The connection of a gas-jet recoil-transport system with a thermochromatographic column offers a fast method for the separation of complex reaction product mixtures<sup>1</sup>. In most applications of such technique reported so far, KCl-clusters loaded with reaction products were collected and destroyed in a quartz-wool catcher, kept at 1000 °C and the released products were volatilized by adding a reactive gas with subsequent separation in a thermochromatographic column. Another possibility consists in direct chemical reactions with the jet gas itself by mixing a reactive component to the carrier-gas and operating the gas-jet without any cluster material. Here volatile compounds of fission products are formed by recoil reactions within the target-area and can be transported to a separation or detection system<sup>2</sup>. Following these approaches we have investigated the application of fluorinating agents for efficient volatilization of fission products as model elements for a broad region of the periodic table.

In a first series of experiments the thermochromatographic system was coupled to a cluster jet and hydrogen fluoride (37 vol.-% in the carrier-gas) was used as the reactive gas. Although the fluorides are normally the most volatile compounds among the halides, the measured deposition temperatures of tellurium, molybdenum and technetium were found to be about 400 °C higher than with other hydrogen halides. Therefore, another reagent, bromine trifluoride, was used for a more efficient fluorination. Bromine trifluoride dissociates at 800 °C into bromine monofluoride and atomic fluorine. It was used in a 1 % mixture with argon (0.1 l/min). The experiments were carried out with an empty column and a charcoal trap placed at the cold end of the column for the adsorption of very volatile fluorides. The temperature of the quartz-wool plug was varied between room temperature and 995 °C. Under these conditions, fluorination and volatilization occurred at about 800 °C, the temperature needed for destroying the KCl-clusters. The fluorides of antimony, technetium, iodine, tellurium and molybdenum were found in the charcoal trap.

Separation of these elements was achieved in a column filled with quartz powder (64 mesh/cm<sup>2</sup>). As fig. 1 shows, tellurium, molybdenum and part of the technetium are now distributed along the column. The rest of technetium, antimony and iodine are found in the charcoal trap. The less volatile fluorides of the alkaline earths, yttrium and the lanthanides are deposited close to the starting position. Zirconium is deposited separately from these elements at 800 °C.

In another series of experiments, hydrogen fluoride was fed directly into a target-chamber made of teflon. Under these conditions short-lived arsenic isotopes could be separated continuously from the other fission-products, which were either retained in the target-chamber or trapped by means of selective adsorbing materials: Volatile compounds of Te, Sb and Mo were caught in a quartz-wool plug at room temperature. A second trap kept at 350 °C was filled with silver-coated quartz powder to retain the halides and the decay products of the noble gases. Finally arsenic was absorbed on di-(2-ethylhexyl)orthophosphoric acid (HDEHP). A  $\gamma$ -ray spectrum of the HDEHP-trap, measured on-line for 1 min shows the isotopes <sup>80-84</sup>As (fig. 2). <sup>85</sup>As ( $t_{1/2}=2$  s) was also identified by summing up 50 spectra measured for 2 s each. Contaminations occurred only by noble gases passing the HDEHP-trap and by <sup>20</sup>F (an activation product of HF), which could not be absorbed entirely in the Ag/SiO<sub>2</sub>-trap.

<sup>1</sup>U. Hickmann et al., Nucl. Instr. Meth. **174**, 507 (1980)

<sup>2</sup>M. Zendel et al., Radiochim. Acta **29**, 17 (1981)

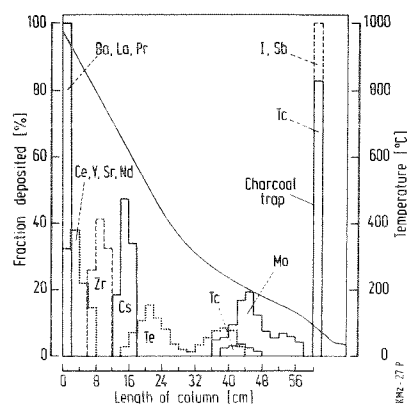


Fig. 1: Distribution of the fluorides in the thermochromatographic column with 1 vol.-% of bromine trifluoride

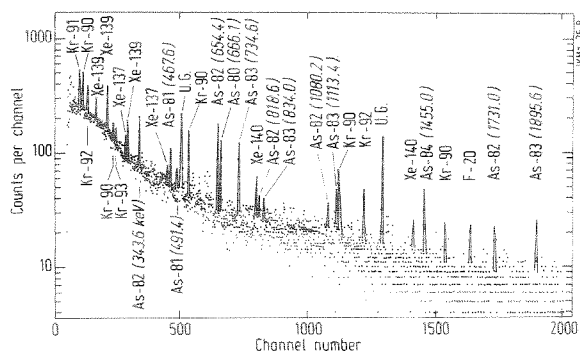


Fig. 2:  $\gamma$ -ray spectrum of the As-fraction measured on-line for 1 min

## Preparation and Gaschromatographic Separation of Actinide Hexafluoroacetylacetonates

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Gaschromatography is a potential, fast method for unravelling actinide mixtures, if suitable actinide compounds with high volatility and sufficient thermal stability can be synthesized. Homologous lanthanide elements have already been gaschromatographically separated as mixed ligand complexes of hexafluoroacetylacetone (HHFA) and tri-*n*-butylphosphate (TBP)<sup>1</sup>. In this report, first experiments are described to extend this method to the separation of <sup>232</sup>Th, <sup>233</sup>Pa, <sup>238</sup>U, <sup>239</sup>Np, <sup>239</sup>Pu and <sup>241</sup>Am in their most stable oxidation states, as well as to trivalent <sup>239</sup>Pu.

For preparing the mixed ligand complexes, the actinides were extracted within 30 s from 1.0 ml of 0.1 N nitric acid (Pu<sup>3+</sup> was stabilized with hydroxylamine) with 1.0 ml of 0.05 M HHFA and 0.035 M TBP, DPSO (dipropylsulfone) or TOPO (trioctylphosphineoxide) as synergistic reagents in cyclohexane<sup>2</sup>. The distribution coefficients (see table 1) were calculated either from  $\gamma$ -measurements for <sup>232</sup>Th, <sup>233</sup>Pa, <sup>238</sup>U and <sup>239</sup>Np or from integral  $\alpha$ -measurements for <sup>239</sup>Pu and <sup>241</sup>Am after evaporation of the solvents.

Table 1

Distribution coefficients for the extraction of some actinide ions from 0.1 N HNO<sub>3</sub> with a mixture of 0.05 M HHFA and synergistic reagents (0.035 M) in cyclohexane

Actinide ion	Synergistic reagent		
	TOPO	DPSO	TBP
Th <sup>4+</sup>	58±16	-	1.3±0.2
PaO <sub>2</sub> <sup>+</sup>	4.0±0.8	1.5±0.4	0.25±0.04
UO <sub>2</sub> <sup>2+</sup>	31±8	-	14±3
NpO <sub>2</sub> <sup>+</sup>	0.22±0.03	0.18±0.03	0.083±0.01
Pu <sup>3+</sup>	22±6	12±3	14±1.3
Pu <sup>4+</sup>	34±1.3	26±4	32±3
Am <sup>3+</sup>	75±16	68±9	56±5

The variation of the distribution coefficients within one column of table 1 can be explained by differences in the ionic radii and the chemical properties of the central ions. The distribution coefficients increase with increasing nucleophilic power of the applied synergistic reagent.

The volatilization of the synthesized mixed ligand complexes was investigated with a gaschromatographic system described elsewhere<sup>1</sup>. The outlet of the separation column was connected to an apparatus for gas-chemical preparation of samples for  $\alpha$ - and  $\beta$ -measurements<sup>3</sup>. The investigated actinides with a valence state higher than 3+ could not be volatilized and remained entirely in the injector of the gaschromatograph. Signi-

ficant volatilization was observed with the trivalent complexes of Am and Pu. Fig. 1 shows the yields of Pu<sup>3+</sup> and Am<sup>3+</sup>-HHFA/TBP-complexes collected on a nickel-foil as a function of the column temperature. The experimental conditions were: Carrier-gas flow 56 ml/min N<sub>2</sub> loaded with 27 mol-% HHFA, empty glass column 2 m long by 2.4 mm inner diameter, injection temperature 30 °C above column temperature, retention time < 5 min. The resolution of the  $\alpha$ -peaks was 43 keV FWHM, if TBP was used as synergistic reagent, with TOPO- and DPSO-adducts a much worse resolution was obtained. The described method should enable the separation of trivalent actinides from higher valence states within a few minutes.

1. N. Greulich et al., GSI Scientific Report 82-1, 219 (1981) and GSI Scientific Report 83-1, 221 (1982)
2. W.C. Butts and C.V. Banks, Anal. Chem. 42, 133 (1970)
3. U. Hickmann et al., GSI Scientific Report 82-1, 217 (1981)

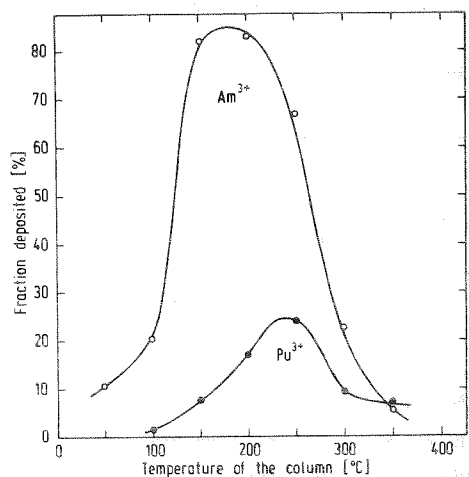


Fig. 1: Yield of americium- and plutonium-hexafluoroacetylacetonate (TBP as synergistic reagent) collected at the outlet of a gaschromatographic system and plotted as a function of the column temperature. For further explanations see text.

Determination of Adsorption Enthalpies for Polonium on Metal Surfaces by On-Line  
Isothermal Gaschromatography

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In the course of recent attempts to produce superheavy elements by heavy ion reactions, the element polonium received a lot of attention because its chemical behaviour should be quite similar to that of some of the expected superheavy elements around  $Z=114^1$ . In addition, since no stable isotope of polonium exists, its chemical properties can be studied with a few atoms only, a prerequisite for extrapolations into the region of superheavy elements, because expectations of production rates for such elements in heavy ion reactions are on a one-atom-per-day scale only! Therefore, in recent years investigations of the gaschromatographic behaviour of polonium have been performed<sup>2-5</sup> with the same technique frequently used in chemical attempts to separate superheavy elements<sup>6</sup>. The most recent attempts to search for superheavy elements applying chemical techniques have been performed using on-line separations such as isothermal gaschromatography<sup>7</sup>. In this contribution we study the adsorption behaviour of polonium, in the form of its short-lived isotope  $^{211m}\text{Po}$  ( $T_{1/2}=25.5$  s), with different surface materials such as quartz or several metals. This nuclide was produced at the Göttingen cyclotron and was continuously transported into a chromatographic column using a gas-jet transportation system. We then measured the retention time within the chromatographic column as a function of the temperature using the radioactive decay of  $^{211m}\text{Po}$  as a clock. This method was first outlined by Rudolph et al.<sup>8</sup>. All experimental details are given elsewhere<sup>9</sup>. In Table I the measured retention data are listed for Po on  $\text{SiO}_2$ , Pt, Au, Ag, Cu, and Pd. In order to deduce adsorption enthalpies from these on-line measurements, we derived a thermodynamical equation which is based on the same model assumptions used already in gas-adsorption studies with gas-thermochromatography<sup>10</sup>, finally resulting in

$$(1) \quad \Delta H_a^0 = -RT \ln \left[ \left( \frac{t_r v_o T}{z T_o \phi} - 1 \right) v / (a \exp\{\Delta S_a^0/R\}) \right]$$

with  $t_r$ =retention time,  $v_o$ =gas flow rate,  $z$ =column length,  $v$ =free column volume,  $a$ =free column surface per unit of length,  $\phi$ =cross section of the tube, and  $\Delta S_a^0$ =adsorption entropy, calculated according to

Ref.10. The  $\Delta H_a^0(\text{exp.})$  values from Table I were thus calculated from the experimental retention data. Also given are  $\Delta H_a^0$ -values from the literature which were obtained from thermochromatographic or desorption studies<sup>11</sup>. The last column of Table I summarizes calculated  $\Delta H_a^0$ -values as they result from a semiempirical model of metal-metal interactions<sup>12</sup>. Clearly there is good agreement between our data and the literature values and also with the calculated enthalpies. This result gives us confidence to apply the same technique in investigations of the thermodynamical properties of exotic species such as very heavy elements which can be produced on a scale of only a few atoms.

#### References

1. B. Eichler, *Kernenergie*, **19**, 302 (1976)
2. B. Eichler et al., Preprint JINR P12, Dubna (1976)
3. B. Eichler et al. *Radiochim. Acta* **26**, 193 (1979)
4. H. Gäggeler et al., Report GSI-79-11, p. 155 (1979)
5. S. Hübener et al., *Radiochim. Acta*, **27**, 157 (1980)
6. see e.g. J.V. Kratz, *Radiochim. Acta*, **32**, 25 (1983)
7. H. Gäggeler et al., this Scientific Report, p.
8. J. Rudolph and K. Bächmann, *Radiochim. Acta*, **27**, 105 (1980)
9. N. Greulich et al., GSI Scientific Report 1981, GSI-82-1, p. 218 (1982)
10. B. Eichler and I. Zvara, *Radiochim. Acta*, **30**, 233 (1982)
11. B. Eichler et al., to be published
12. B. Eichler Report ZfK-396, Rossendorf (1979)

Table I:

#### Adsorption behaviour of polonium on different surfaces

Surface	Column Length (cm)	Temperature <sup>a)</sup> (K)	$-H_a^0(\text{exp.})^b$ (kJ/Mol)	$-H_a^0(\text{exp.})^c$ (kJ/Mol)	$-H_a^0(\text{calc.})^d$ (kJ/Mol)
$\text{SiO}_2$	15	633	123.4	---	---
Ag	10	833	165.4	---	232
Au	10	1133	223.7	196/200	200
Cu	10	893	180.8	157/193	178
Pd	10	>1373	>277.9	302	301
Pd	0.8	1298	289.7		
Pt	7	923	186.9	196/211/ 279/280	238

a) for a retention time  $t_r=T_{1/2}({}^{211m}\text{Po})=25.5$  s

b) calculated with eq.(1)

c) values from Ref. 11

d) calculated according to Ref. 12

Chemical Separation of Astatine from Bi/Pb-Targets<sup>B</sup>

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For the radiochemical detection of the presumably very rare reaction channel  $^{207}_{82}\text{Pb} (^4_2\text{He}, \pi^-) ^{211}_{85}\text{At}$  (pionic fusion<sup>1</sup>) a selective chemical separation of astatine from polonium, bismuth, lead and from spallation and fission products with high decontamination factors is necessary. At the same time it is desirable to prepare the astatine sample as a weightless source for  $\alpha$ -particle spectroscopy. To this end, the volatilization of elementary astatine and its re-condensation on a cooled metal surface<sup>2</sup> seemed to be most attractive.

We have produced  $^{210,211}\text{At}$  tracer activities at the Karlsruhe cyclotron by bombarding  $^{209}\text{Bi}$ -targets with 30 MeV  $\alpha$ -particles. These, together with 20 mg of lead, were heated to 1000° C in a gas flow of 100 ml/min Ar and 10 ml/min H<sub>2</sub> for 15 min in a quartz tube, see Fig. 1. Under these conditions both astatine and polonium are volatilized. Inside oven 2 (600° C) the quartz tube was lined with Pd foil of 4 cm length which absorbs polonium while astatine passes through. Fig. 2 shows the yield of astatine as a function of the temperature of oven 2. It is evident that temperatures  $\geq 500^\circ\text{C}$  are necessary in order to avoid losses of astatine to the Pd surface. Astatine is then condensed on a cooled Ni-foil (1 mg/cm<sup>2</sup>) covered with  $\sim 40\ \mu\text{g}/\text{cm}^2$  of vapour-deposited Pd. Decontamination factors from polonium were of the order of 10<sup>5</sup> in this one-step chemistry and the chemical yield of astatine was 91 $\pm$ 7%. The Pd-Ni-substrate foils for the astatine sources are sufficiently thin to allow for  $\alpha$ -particle spectroscopy on both sides of the source by large area silicon detectors with a total efficiency of 65%.

First bombardments of 20 mg/cm<sup>2</sup> nat. Pb-targets with 2.5  $\mu\text{A}$   $\alpha$ -particles of 130 MeV at the Jülich cyclotron showed that the decontamination from polonium of this one-step chemistry was not sufficient to detect subnanobarn cross sections of astatine in the presence of tens of millibarns of polonium. Also, an extremely high level of beta radiation caused by fission products, in particular by iodine, jeopardized the function of the silicon detectors for  $\alpha$ -particle spectroscopy. This called for additional chemical purification: The bombarded lead-targets are now dissolved in 500  $\mu\text{l}$  of 3.5M HNO<sub>3</sub>. Astatine is then deposited in the presence of hydroxyl-ammonium chloride and ammonium acetate buffer on a 2 cm<sup>2</sup> Au-foil. The latter is rinsed with acetone, dried, and heated in the quartz tube (Fig. 1)

for 15 min to 900° C (oven 2 at 600° C) under otherwise identical conditions as given above for the one-step gas-phase chemistry. Chemical yields for the combined procedure are close to 80%. The separation now produces sources which are well-suited for a highly sensitive search for  $^{211}\text{At}$  from the  $^{207}_{82}\text{Pb} (^4_2\text{He}, \pi^-)$  reaction by  $\alpha$ -particle spectroscopy.

## References

1. K. Klingenberg et al., Phys. Rev. Lett. **47**, 1654 (1981)
2. U. Hickmann et al., GSI Scientific Report 1981, p. 217

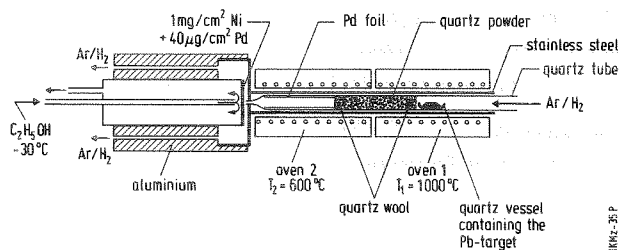


Fig. 1: Apparatus for the volatilization and re-condensation of astatine

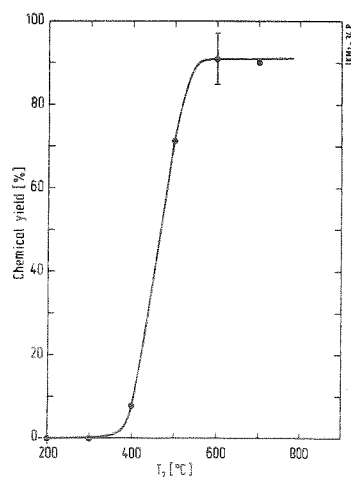


Fig. 2: Chemical yield of astatine measured on the Pd-Ni foil as a function of the temperature of oven 2. The temperature of oven 1 is 1000° C. The error bar on the data point at 600° C corresponds to the standard deviation of ten experiments.

ROMA - A Rotating Wheel Multidetector Apparatus used in Experiments with  $^{254}\text{Es}$  as a Target

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Rotating wheel systems connected to gas jet transport capillaries have proven to be useful devices for studying short-lived activities with half-lives in the order of seconds. We have set up such a system at GSI to detect  $\alpha$ -particles and correlated fission fragments from the decay of neutron-rich actinide and trans-actinide isotopes formed in the reactions  $^{16,18}\text{O}$ ,  $^{22}\text{Ne}$  +  $^{254}\text{Es}$ <sup>1</sup>. An earlier version of this device, in conjunction with on-line gas phase chemistry (OLGA), has been used in a search for superheavy elements with the  $^{48}\text{Ca}$  +  $^{248}\text{Cm}$  reactions<sup>2</sup>. We utilize a He/KCl-cluster jet to transport recoiling reaction products into the ROMA system, where they are deposited on thin polypropylene foils (about  $70 \mu\text{g}/\text{cm}^2$ ) mounted on a 64-position wheel. After a preset collection time (typically 2-30 seconds), the activity is moved between pairs of surface barrier detectors to measure  $\alpha$ - and spontaneous fission decays. The wheel is rotated by a stepping motor controlled by a GSI 8085 microprocessor which is in turn coupled via CAMAC to the PDP11 computer of the GSI data acquisition system.

The inner part of the ROMA system is shown in the photograph (fig.1). Detector pairs are mounted at 15 out of 64 available positions. Three sets of 16 polypropylene foils are located in 48 out of the 64 positions around the perimeter of the rotating catcher wheel. To reduce the build-up of longer lived activities, a fresh foil set is brought into position every two hours. After the three sets available have been used, the wheel chamber is opened and the entire collector wheel exchanged.

The remaining 16 positions of the wheel are left blank to allow for observation of daughter decays: when, during the on-line analysis, an  $\alpha$ -event is found to fall within a preset energy window (mother decay), the wheel is moved in such a way that the blank positions are located in front of the detectors. In this way the observation of subsequent decays of the daughter activity implanted by nuclear recoil into thin carbon foils covering the detectors is enhanced. After a preset time during which possible daughter decays are recorded, normal wheel indexing is resumed.

The data acquisition includes standard  $\alpha$ - and sf-spectroscopy recorded event-by-event to measure energies, half-lives, and time correlations. To facilitate data taking with 30 surface barrier detectors, the analog signals of sets of eight detectors each were multi-

plexed by an ORTEC 476-8 multiplexer. Each detector was identified by routing bits stored in a bit pattern unit. A test pulse distributor<sup>3</sup> was specifically designed for this set-up to facilitate the time-consuming calibration of the detectors.

1. M. Schädel et al., contribution to this report, p.
2. H. Gäggeler et al., contribution to this report, p.
3. W.Bröchle, contribution to this report, p.

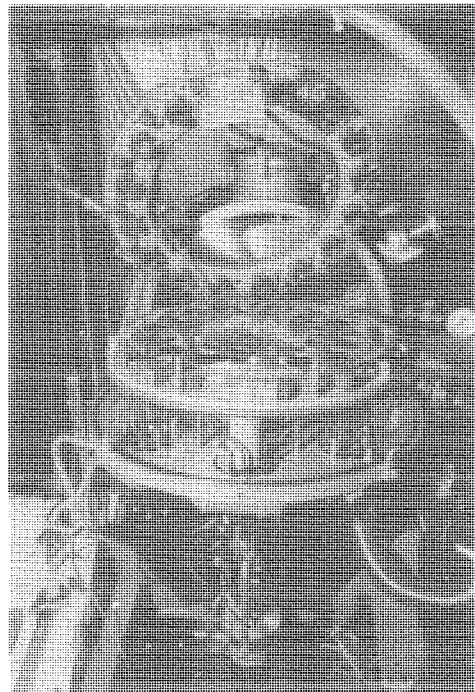


Fig.1: The rotating wheel multidetector apparatus, ROMA, as used in experiments with  $^{254}\text{Es}$  as a target. The photograph shows the lid of the vacuum chamber slid outwards and turned by  $90^\circ$ . The catcher wheel and one set of 15 surface barrier detectors are in a horizontal position now. The second set of detectors is moved upwards to replace the wheel and ease access to the detectors.

The Status of ARCA - Automated Rapid Chemistry Apparatus

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We adapted our microprocessor-controlled automated rapid chemistry apparatus, ARCA<sup>1</sup>, to the needs of two different types of experiments.

The first one, a search for superheavy elements (SHE) in the  $^{48}\text{Ca} + ^{248}\text{Cm}$  reaction<sup>2</sup>, required a fast and effective separation of elements Os through Po and their homologues Z=108 through 116 from actinide elements so that the expected spontaneous fission (sf) decay of the SHE's would not be obscured by interfering spontaneously fissioning actinides. We used a very simple cation exchange column separation with a separation factor of about  $10^4$ . This method is based on the predicted tendency that elements 108 through 116 should form strong anionic complexes in aqueous solution thus being not bound to a cation exchange resin while the cationic species of actinides are strongly retarded. Reaction products recoiling out of the  $^{248}\text{Cm}$  target are stopped and transported in a KCl loaded cluster jet with He as a carrier gas from the irradiation position at Y3 over ca. 80 m into our chemistry laboratory. No significant losses were observed, and the transport time of approximately 10 s is negligible compared to the 30 min time interval while the activity from the jet was collected on a quartz frit (D4) in ARCA. Every 30 min the activity was dissolved in 0.1 m HBr/Br<sub>2</sub> and pumped through the (100x1.75) mm cation exchange column (AG 50 W x 8, - 400 mesh) at a flow rate of 3. ml/min for 10 s. Eluting a SHE fraction was completed after another 60 s of pumping 0.8m HBr/Br<sub>2</sub>. The solution at the outlet of the column was continuously evaporated. This made it possible to use just the last few drops to rinse the glass and to have a sample ready for counting on a Au-coated 33  $\mu\text{g}/\text{cm}^2$  C-foil within 2.3 min after end of bombardment (EOB). Counting each sample between two Si(Au) surface barrier detectors for sf and  $\alpha$ -activity started 3 min after EOB. ARCA went successfully through 88 irradiations and automated chemistry cycles accumulating  $1.3 \times 10^{17}$   $^{48}\text{Ca}$  particles on the  $^{248}\text{Cm}$  target<sup>2</sup>.

In a second experimental series we wanted to measure cross sections for transfer products from an  $^{254}\text{Es}$  target and searched for new isotopes of elements 101 (Md) through 103 (Lr) with minute half-lives<sup>3</sup>. This required a very delicate and precise separation between individual transcurium elements which was difficult to achieve in an automated chemistry apparatus. The flow scheme is shown in Fig.1. The activity was washed from the quartz frit in 0.03 m HCl and loaded onto a HDEHP column, a Sr<sup>2+</sup> fraction was discarded and a

No<sup>2+</sup> fraction eluted with 0.03 m HNO<sub>3</sub> and loaded onto a cation exchange column. With two pumps in operation we simultaneously eluted a No<sup>2+</sup> fraction with 4m HCl from the cation exchange column and, after washing with 1.0m HNO<sub>3</sub>, a Md, Lr fraction with 4m HCl from the HDEHP column. Using a T connector, a stream of 2.0 m NH<sub>4</sub>OH was added to the 4m HCl from the HDEHP column to bring down the H<sup>+</sup> concentration to about 1.0 m. The Md, Lr from this solution was loaded on a second cation exchange column. After washing with H<sub>2</sub>O and NH<sub>4</sub>Cl the elements Lr and Md were eluted with  $\alpha$ -hydroxyisobutyric acid as individual fractions. The cycle time (collection time) was 30 min, and the final separation of the No, Lr and Md fractions was completed after 4.7, 12.5 and 14.2 min, respectively. We ran a total number of 29 automated separation cycles while the  $^{254}\text{Es}$  target was bombarded with  $^{16}\text{O}$  and  $^{22}\text{Ne}^3$ .

As a major change from the early version of ARCA we decided to control the system now with a GSI 8085 microprocessor system<sup>4</sup> with two floppy disc drives. BASIC is now the command language, which makes programming simple and flexible. To operate more than one valve at a time all four 16 channel driver units for the 24V magnetic valves were modified.

1. W. Schorstein et al., GSI Scientific Report 1981, GSI-82-1, p. 220 (1982)
2. H. Gäggeler et al., this Scientific Report, p.
3. M. Schädel et al., this Scientific Report, p.
4. G. Englert et al., GSI Scientific Report 79, GSI-80-3, p. 196 (1980)

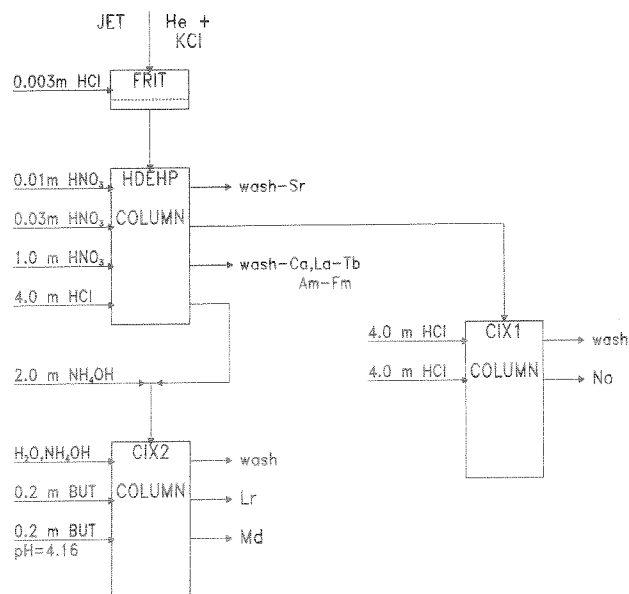


Fig.1: Flow scheme for a three column Md, No and Lr separation used in experiments with  $^{254}\text{Es}$  as a target<sup>3</sup>.



## A PROGRAMMABLE PULSE DISTRIBUTOR FOR 32 DETECTORS

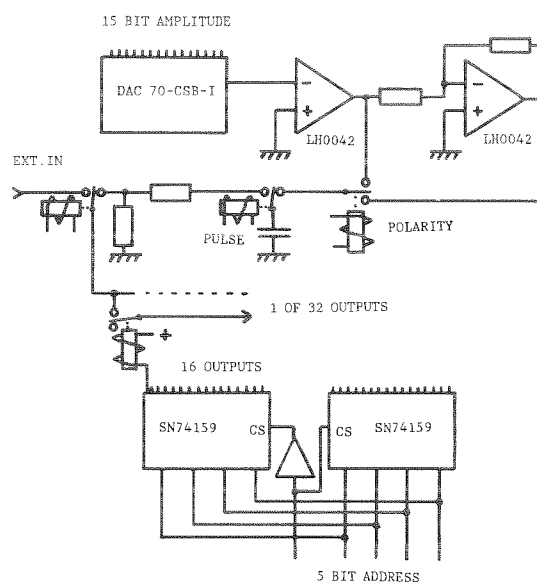
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To check the stability of several surface-barrier detectors we formerly used a GSI- designed pulse distributor (FX086) which could direct calibration pulses from an Ortec pulser to 32 preamplifiers. A disadvantage was that all detectors were supplied with the same pulse amplitude and polarity.

With the increasing use of multidetector counting systems like "ROMA" <sup>1</sup> a new and more flexible design of a test pulse distributor suggested itself. This new pulse distributor contains a computer controlled precision pulser. Amplitude, polarity, and frequency can be defined by a low-cost homecomputer VC20. 32 outputs can be active in any order. For these purposes the 8 bit userport of the VC20 was extended to 24 bit with two 8 bit D-registers (74LS374). By use of drivers (SN49701) and optical coupling (ILD74) a trouble-free operation can be achieved at more than 100m distance between the control system and the pulser.

The reference voltage (max.  $\pm 10V$ ) is produced by a 16 bit DAC (DAC70-CSB-1) and two operational amplifiers (LH0042). The maximum nonlinearity is  $\pm 0.003\%$  of full scale range with  $\pm 14\text{ppm}/^\circ\text{C}$  maximum gain drift. With this reference voltage (polarity switched by a computer controlled relays) a capacitor is charged. This capacitor is discharged when a special mercury relay is energized. The discharge path is determined by the setting of one out of 32 relays. Which output is active can be defined by computer via two decoders (SN74159). There is an option to use only external pulses and distribute them. In this case the computer can count the external pulses by an external trigger input.

The use of the machine is simplified by a Basic- controlled menu which is displayed on screen. After calibration with a standard source the computer calculates calibration coefficients (comparable with normalize- and attenuation- values in a normal pulser) , which also can be stored on tape. Now the desired energies can be typed in, instead of choosing relative amplitudes. In a continuous mode used for calibration control a high-energy pulse alternates with a low-energy pulse, every two seconds the next output is switched on. Maximum frequency is 20Hz.



Schematic drawing of the Pulse Distributor

1) K.Sümmerner et al., this report