Deep Inelastic Transfer Reactions - A New Way to Exotic Nuclei?

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Abstract. We studied deep inelastic multinucleon transfer reactions in collisions of $^{64}\text{Ni}+^{207}\text{Pb}$ and $^{48}\text{Ca}+^{238}\text{U}$ at energies around the Coulomb barrier. The experiments were performed at the velocity filter SHIP at GSI Darmstadt. One of the goals was to investigate if deep inelastic transfer is superior to fragmentation reactions for producing neutron-rich isotopes in the astrophysically interesting region of nuclei along the magic neutron number $N = 126$. With both collision systems, rather neutron-rich transfer products were populated, some of them reaching out to the limits of the present chart of nuclides. New isotopes could not be identified. A comparison of the measured transfer cross-sections and yields with those from fragmentation reactions allowed for interesting conclusions.

1 Introduction

In recent years, intense discussions restarted about the possibility to produce new neutron-rich heavy and superheavy nuclei in deep inelastic transfer (DIT) reactions at Coulomb barrier energies. These considerations were triggered by new theoretical model calculations which result in experimentally feasible cross-sections for DIT products in these regions (see e.g. [1, 2]). One region of interest concerns nuclei along the closed neutron shell $N = 126$ which are of relevance for the astrophysical r-process. These isotopes are presently produced in fragmentation reactions at relativistic energies with heavy projectiles and light target nuclei. However, calculations in macroscopic-microscopic models predict larger cross-sections for these isotopes if they are produced in DIT reactions. Another region of interest is neutron-rich transuranium nuclei which are not reachable in fragmentation reactions or in fusion reactions with stable beams.

Only few experimental data exist on the production of heavy transfer products in the above named regions. In the last decade, collisions of $^{58}\text{Ni}+^{208}\text{Pb}$ and $^{64}\text{Ni}+^{208}\text{Pb}$ at energies around the Coulomb barrier were studied [3, 4]. In these “thick-target” experiments, the transfer products were stopped in the target. The isotopic identification was performed via in-beam and offline gamma decay spectroscopy. Limit cross-sections of about $20 \mu \text{b}$ were reached. Concerning target-like transfer products, a vast region of nuclei around $Z = 82$ was populated. New, previously unknown isotopes were not identified. Also attempts to produce new transactinide nuclei in transfer reactions were made already several decades ago. The most sensitive of those experiments applied chemical separation techniques with subsequent isotope identification via alpha decays. With this method, transfer products up to

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Z = 101 were observed in reactions of $^{48}\text{Ca}$ or $^{238}\text{U}$ beams with $^{238}\text{U}$ and $^{248}\text{Cm}$ targets [5–7]. Limit cross-sections of 50 nb were reached but also in these experiments no new isotopes were observed.

The expected cross-sections for new neutron-rich transfer products are small. Model predictions result in values of $\sigma < 1 \, \mu b$ for isotopes along the N=126 shell and in $\sigma < 1$ nb for new neutron-rich nuclei with $Z \geq 102$. As a consequence, similarly efficient separation and detection techniques have to be applied like have been developed for the identification of single atoms in superheavy element experiments. At GSI Darmstadt, we performed studies of DIT reactions in heavy collision systems using the velocity filter SHIP for separation of the target-like transfer products from background events. The isotopic identification was performed via alpha and/or gamma decay spectroscopy in the focal plane of SHIP. In particular when the isotopes were identified via alpha decays, the method turned out to be significantly more sensitive than the techniques applied in previous experiments. In this case we reached limit cross-sections of 0.1 nb [8]. The identification via gamma spectroscopy resulted in cross-section limits of about 10$\mu$b, similar like in the above named experiments [3, 4].

The sensitivity of our setup allows, in principle, to reach new isotopes in the region of N=126 as well as in the transuranium region if we assume the cross-sections predicted by theory. Here, we want to concentrate on our studies of DIT reactions leading to nuclei in the region of N=126. For this, we investigated collisions of $^{48}\text{Ca} + ^{238}\text{U}$ and $^{64}\text{Ni} + ^{207}\text{Pb}$ and compared the transfer cross-sections and yields with those from fragmentation reactions.

2 Theoretical Predictions

Extensive calculations on the production of heavy transfer products in the region of N=126 are available from two different macroscopic-microscopic theories. Both models follow different philosophies concerning the optimum projectile/target combinations for this kind of reactions. The dinuclear system model which uses diabatic potentials suggests the application of neutron-rich intermediate heavy projectiles like $^{48}\text{Ca}$ and heavy target nuclei. As a possible target nucleus, $^{238}\text{U}$ is suggested [1] because due to its large neutron number the population of very neutron-rich transfer products is expected. However, with uranium targets a very large flow of nucleons from the target to the projectile nucleus is necessary to populate transfer products around N=126. The other model, which uses adiabatic potentials and transport (Langevin type) equations of motion, suggests the application of rather heavy neutron-rich projectiles like $^{136}\text{Xe}$ and $^{208}\text{Pb}$ as target material [2]. In this case, the total neutron number of the system is large and one might expect relatively large cross-sections for neutron-rich transfer products. The application of the adiabatic model results in long nuclear interaction times also in very heavy collision systems. This is a necessary condition for a large flow of nucleons between the colliding nuclei. In the dinuclear system model, in contrast, sufficiently long interaction times are only obtained with intermediate heavy projectiles while in very heavy systems the Coulomb repulsion becomes dominant and prevents the sticking of the nuclei. As a consequence, this model predicts only direct transfer reactions in very heavy systems which do not lead to the population of isotopes far from the projectile and target nucleus, respectively.

Calculated cross-sections for the production of nuclei around N=126 in the suggested collision systems $^{48}\text{Ca} + ^{238}\text{U}$ [1] and $^{136}\text{Xe} + ^{208}\text{Pb}$ [2] are shown in fig. 1. In both models, the optimum beam energies for the production of neutron-rich nuclei are just at the Coulomb barrier. Already at such low energies a large flow of nucleons takes place but the excitation energy of the primary transfer products is still moderate (typically a few 10 MeV). The excitation energy has to be kept as low as possible in order to reduce the probability for fission of the excited primary transfer products and to keep the number of evaporated neutrons small (this was also confirmed by our experimental results from the systems $^{58,64}\text{Ni} + ^{207}\text{Pb}$ [8]). In fig. 1 (a), (b) one can see that in both calculations the cross-sections
Figure 1. Calculated cross-sections of deep inelastic transfer products in the region of \( N = 126 \) from two different theoretical models. The cross-sections are drawn as a function of the neutron number \( N \) of the transfer products. (a) Cross-sections of transfer products with \( Z = 74 − 82 \) from collisions of \(^{136}\text{Xe} + ^{208}\text{Pb} \) at 5.5 MeV/u, calculated in the macroscopic-microscopic model with adiabatic potentials [2]. Dashed curves represent primary transfer products; full lines represent secondary transfer products. (b) Cross-sections of Pt isotopes (\( Z = 78 \)) from deep inelastic transfer reactions in \(^{48}\text{Ca} + ^{238}\text{U} \) at 4.7 MeV/u, calculated in the dinuclear system model [1]. Full symbols represent primary transfer products and open symbols represent secondary products. The horizontal lines in both figures mark the cross-section of 1 \( \mu \text{b} \).

of the surviving (i.e. secondary) transfer products with neutron numbers around \( N = 126 \) are mostly below microbarns and decrease rather steeply with increasing neutron number. Comparing the two model predictions, the reactions of Xe + Pb result in about (100 – 1000) times larger cross-sections for the same isotopes than reactions of Ca + U and seem therefore more favourable.

3 Experimental Setup and Method

In our experiments we used the velocity filter SHIP for separation of the target-like transfer products from background events. A sketch of the setup is shown in fig. 2 (a detailed description of our method applied for DIT reaction studies can be found in [8]). Reaction products which leave the target at forward angles of (0 ± 2) degree with respect to the beam direction are accepted by the entrance aperture of SHIP. All nuclei which enter SHIP are separated according to their velocities. The ratio of the electric and magnetic field values, \( E/B \), determines at which velocity an ion can pass SHIP. The accepted velocity window at a given setting is \( \Delta v/v \approx 0.1 \) (FWHM). For the here described experiments, \( E \) and \( B \) were chosen such that the target-like transfer products could pass SHIP while the much faster projectiles and projectile-like reaction products were deflected to the beam stop. All reaction products which pass the velocity filter are implanted in a position sensitive 16 strip silicon detector (stop detector) where their time of implantation, position, kinetic energy and radioactive decays (alpha, beta and fission decays) are registered. In addition, a germanium clover detector is mounted behind the stop detector. It consists of four crystals which register gamma rays accompanying the decays of implanted nuclei. With this method, nuclei with half-lives down to about 5 \( \mu \text{s} \) can be detected and studied. In order to reduce background in the alpha and gamma spectra, we utilize the structure of the UNILAC beam which consists of 5 ms long beam pulses followed by 15 ms long beam-off periods. With this, background events can strongly be suppressed if the radioactive decays are recorded during beam-off periods.
Figure 2. Scheme of the velocity SHIP and its detection system which was used for the study of deep inelastic transfer products. Typical beam intensities at the target and total count rates at the implantation detector during our experiments are also given. Further, an example of a measured velocity spectrum is shown for the case of $^{222}$Ra nuclei produced in deep inelastic transfer reactions in $^{48}$Ca+$^{238}$U collisions at 4.90 MeV/u. The velocities in the spectrum are relative to the compound nucleus velocity $v_{CN}$ (i.e. centre-of-mass velocity).

Usually, a broad variety of transfer products is created in the reactions where each of the nuclei has a characteristic velocity, depending on its A and Z. In order to cover this range of different velocities, we varied stepwise the E and B values of the velocity filter. In this way, one obtains the velocity spectrum of each individual isotope. An example of such a spectrum is shown in fig. 2 for the transfer product $^{222}$Ra from collisions of $^{48}$Ca+$^{238}$U at 4.90 MeV/u. In this spectrum, the transfer product velocities $v$ are given relative to the compound nucleus velocity $v_{CN}$ (a peak at $v/v_{CN} = 1.0$ would be expected for nuclei resulting from complete fusion reactions). Typically, the spectra of target-like transfer products from reactions with medium heavy projectiles and heavy targets, like discussed here, reveal peak structures like in fig. 2 where the maxima are located at velocities between 1.5 and 2.0 times the compound nucleus velocity $v_{CN}$. The velocity of the transfer products reflects also the energy dissipation during the reaction. Therefore we used the locations of the peak maxima to calculate the total kinetic energy (TKE) of target-like and projectile-like transfer products in the exit channel. For this we calculated the kinetic energy of the target-like transfer product from the velocity which corresponds to the peak maximum. Then we reconstructed the kinetic energy of the corresponding projectile-like transfer product by considering the reaction as a two-body process and taking into account the energy and momentum conservation laws.

4 Experimental Results and Discussion

4.1 Transfer Products from Collisions of $^{64}$Ni+$^{207}$Pb

We investigated the population of transfer products in reactions of $^{64}$Ni+$^{207}$Pb at six different beam energies between 4.80 and 5.92 MeV/u (the Coulomb barrier is 5.0 MeV/u). Here, we will show the results measured at 5.2 MeV/u because at this energy we observed the largest cross-sections for neutron-rich transfer products. The beam intensity was typically several times $10^{12}$ particles/s. The targets consisted of 380 $\mu$g/cm$^2$ thick layers of the chemical compound $^{207}$PbS. We measured the
Figure 3. Gamma spectra from collisions of $^{64}\text{Ni} + ^{207}\text{Pb}$ measured in the focal plane of SHIP during beam-off periods. The energy regions (0 - 500) keV and (500 - 1000) keV are displayed separately. The figures contain the integral data from all investigated beam energies. The isotopes which we identified via these gamma lines are shown in the chart of fig. 4.

Figure 4. Transfer products identified in collisions of $^{64}\text{Ni} + ^{207}\text{Pb}$ at 5.2 MeV/u via the gamma lines shown in fig. 3 (hatched squares). The black squares mark stable isotopes. For orientation, the location of the target nucleus $^{207}\text{Pb}$ is marked by the white asterisk. The limits on the neutron-rich side correspond to the limits of the present Karlsruhe chart of nuclides of 2012.

velocity spectra of the transfer products in the range $(1.6 - 2.0) v_{CN}$ in steps of $0.1 v_{CN}$. The total measurement time at 5.2 MeV/u was about four hours. In this experiment, we identified transfer products in the region of $Z = 76 - 89$ by alpha and gamma spectroscopy. For isotopes which could be identified via alpha decays we reached very low limit cross-sections of about 0.1 nb. This concerns most of the transfer products above Pb ($Z > 82$). In this region we performed extensive studies of the characteristics of DIT reactions like excitation functions, excitation energies, influence of shell effects and the beam energy dependence of isotopic yields. The respective results are discussed in [8].

In the following, we focus on the population of isotopes below Pb ($Z < 82$). Radioactive nuclei in this region are $\beta^-$-emitters or undergo electron capture decay. If the decay of an implanted mother nucleus populates an excited state in the daughter, the daughter nucleus will decay to the ground state by emission of gamma rays and/or conversion electrons. In our case, we used the emitted gamma rays for isotopic identification of the reaction products. The method is applicable for identification of isotopes which populate with sufficient branching an excited state in the daughter nucleus and which are not too long-lived. In the experiment discussed here, the upper limit for accessible half-lives was less than one day, given by the measurement time and the production yield of the respective nucleus. The gamma spectrum recorded during beam-off periods in the focal plane of SHIP is shown in fig. 3 for the energy ranges (0 - 500) keV and (500 - 1000) keV. The identified transfer products with $Z \leq 82$ are shown in the chart in fig. 4 (hatched squares). The velocity spectra of these isotopes
revealed a large energy dissipation during the reaction. The TKE values, which we deduced from the velocities, are independent of the beam energy and are located below the Viola energy [9] (the Viola energy usually denotes the TKE of fission fragments from an equilibrated compound nucleus). These are typical signatures of deep inelastic reactions where all kinetic energy is dissipated and the exit channel nuclei reseparate with their Coulomb barrier energy.

From the intensities of the measured gamma lines we deduced total production cross-sections for the respective isotopes taking into account the detection efficiency and the angular efficiency of SHIP for DIT products. Concerning the angular efficiency, experimental data are not available. Therefore, we used the efficiencies from simulations in the theoretical model [2] for $^{64}$Ni+$^{208}$Pb [10]. Here, one has to stress that the transfer products discussed here result from deep inelastic reactions with large amount of energy dissipation and very broad angular distributions - in strong contrast to direct transfer reactions which peak at the grazing angle. As a consequence, the target-like transfer products are in our case emitted in a cone with an opening angle of about 50 degree in the laboratory system. The opening angle depends on A and Z of the transfer products and becomes smaller if A and Z increase. The fraction of the created nuclei which is accepted by SHIP for the transfer products shown in fig. 4 is in the range (0.1 - 0.5) % according to the simulations of [10].

The deduced total cross-sections for the examples of Ir (Z=77) and Pt (Z=78) isotopes are shown in fig. 5 (black squares). In the same figure, also the cross-sections for Ir and Pt transfer products from the other experiment with the very similar system $^{64}$Ni+$^{208}$Pb [3] are shown (open squares). The cross-sections from both experiments agree surprisingly well, within less than a factor of ten, despite the uncertainties concerning angular efficiency in the case of SHIP and concerning beam energy in the thick target experiment [3]. The data from both experiments indicate a rather steep decrease of the cross-sections towards the neutron-rich side after the maximum is reached around nuclei located close to the stability line. The same holds for the data measured for other elements beside Ir and Pt which we are not going to show here. In both experiments no new isotopes were observed or identified, respectively, and the N = 126 shell was not reached for nuclei with Z < 80. Very interesting is the comparison of the isotopes and related cross-sections reached in DIT reactions with the ones from fragmentation reactions. In experiments on fragmentation reactions with $^{208}$Pb projectiles on beryllium targets, rather neutron-rich isotopes were detected [11]. The measured fragmentation cross-sections are represented by the asterisks in fig. 5. One can notice that fragmentation and transfer

![Figure 5](image-url)
Table 1. Typical values for beam intensities \( N_{\text{beam}} \), target thicknesses \( d_{\text{target}} \) and angular efficiencies in deep inelastic transfer reactions and in fragmentation reactions. The efficiency of \(< 5\%\) for heavy deep inelastic transfer products is specifically given for SHIP.

|                  | \( N_{\text{beam}} \) | \( d_{\text{target}} \) | Angular efficiency |
|------------------|-------------------------|--------------------------|---------------------|
| Deep inelastic transfer | \( 5 \times 10^{12} / \text{s} \) | \( 500 \ \mu \text{g/cm}^2 \) | \(< 5\%\)            |
| Fragmentation     | \( 10^8 / \text{s} \)   | \( 5 \ \text{g/cm}^2 \)  | (50 - 100)\%        |

Figure 6. Count rates of Ir isotopes from deep inelastic transfer (DIT) and fragmentation which can be expected at the detection system. The count rates are related to the cross-sections in fig. 5, left. They were calculated with the beam intensities and target thicknesses given in table 1. Angular efficiencies of 50% for fragmentation products and 5% for transfer products were assumed.

cross-sections are quite similar (i.e. within the same order of magnitude) in the region where data from both, DIT and fragmentation, are available. In some cases the transfer cross-sections are even larger. This confirms theoretical predictions. Also the fragmentation cross-sections reveal a steep drop towards the neutron-rich side but the experiment was more sensitive and allowed to reach limit cross-sections below 10 nb. One has still to remark that the cross-sections which we measured in Ni+Pb reactions in our experiment and the ones given in [3] are on the same order of magnitude than the theoretical cross-sections predicted in [2] for reactions of Xe+Pb (theoretical calculations for the system Ni+Pb are presently not yet available for the nuclei discussed here).

Even more interesting is a comparison of the count rates which can be expected at the detector in DIT and fragmentation reactions because this is the decisive criterion for a successful application of a reaction for isotope production. To deduce the count rates from the cross-sections, one has to take into account the different conditions concerning beam intensity, target thickness and efficiencies for DIT and fragmentation reactions. Typical parameters are shown in table 1. The given angular efficiency range for DIT products is specifically for SHIP and for reactions with intermediate heavy projectiles and heavy target nuclei. Figure 6 shows count rates for Ir isotopes which can be expected at the detector from DIT and fragmentation reactions. The count rates have been calculated from the measured cross-sections given in fig. 5. For this, the beam intensities and target thicknesses given in table 1 were used. The angular efficiencies were assumed as 50% for fragmentation products and 0.1%
Figure 7. Deep inelastic transfer products identified in collisions of $^{48}\text{Ca} + ^{238}\text{U}$ at 4.90 MeV/u (hatched squares). The question marks indicate that the correct identification of the two respective isotopes is not certain. The border of the chart on the neutron-rich side corresponds to the border of the present Karlsruhe chart of nuclides from 2012. The black squares mark stable isotopes. The target nucleus $^{238}\text{U}$ is marked by the asterisk.

for transfer products. The angular efficiency for transfer products was taken from the calculations in [10] for Ir nuclei. Detector efficiencies were assumed as 100% for this estimate. The count rates for the same Ir isotopes are typically at least 1000 times higher in fragmentation reactions. The same result we obtained also for isotopes of the other observed elements. This means, in order to become more profitable than fragmentation reactions, the count rates of transfer products would have to be increased by clearly more than three orders of magnitude. Therefore, also experimental setups with 100% angular efficiency for DIT would not allow to reach count rates larger than in fragmentation reactions.

Finally, another point has still to be mentioned: In fragmentation reactions, the isotopic identifications of the reaction products can be performed by measuring energy, energy loss, time-of-flight and magnetic rigidity of the nuclei. Due to the relativistic energies of the ions, $A$ and $Z$ can be resolved with one unit. This method cannot be applied for the identification of transfer products from reactions at Coulomb barrier energies because in the case of heavy and low-energetic reaction products, plasma effects and the resulting pulse height defect limits the $A$ and $Z$ resolution which is usually not better than three units in the region of Pb and above. Because of this, the isotope identification is presently performed via radioactive decays which limits the method to nuclei with appropriate decay properties while the method applied in fragmentation reactions gives access to all nuclei with halflives larger than several 100 ns. This time limit is given by the time of flight of the nuclei through the fragment separator.

4.2 Transfer Products from Collisions of $^{48}\text{Ca} + ^{238}\text{U}$

An option which was suggested by the dinuclear system model is the application of uranium targets and $^{48}\text{Ca}$ beams to populate transfer products along $N = 126$, below Pb [11]. In the following, we will briefly present our results on DIT reactions in this system at 4.90 MeV/u which we collected during a relatively short beam time of about one day. The isotope identification was performed via alpha decay spectroscopy or, for beta emitters, via gamma spectroscopy in the focal plane of SHIP. The limit cross-sections reached in this experiment were $\sim 0.1$ nb for alpha emitters and about $5 \mu$b for beta emitters. We identified transfer products in the region of $Z = 84 - 93$ which are shown in the chart in fig. 7.
Nuclei with Z < 84 were not observed, most probably because their production cross-sections were already below the limit cross-section of 5 µb for beta emitters. This is consistent with the theoretical cross-sections given in [1] for these nuclei. It is also consistent with the cross-sections which we measured for isotopes with Z ≥ 84. For these, we found that the cross-sections decrease continuously with decreasing Z of the DIT product (i.e. with increasing distance from the target nucleus). For the isotopes with the lowest observed Z (Rn and Po) we measured cross-sections of several 10 µb which is already close to the limit cross-section.

In total, our experimental results show that reactions with Pb targets are more favourable for the production of neutron-rich transfer products in the region of N = 126, Z < 80. However, the transfer products which we observed in reactions of 48Ca + 238U reach out far to the neutron-rich side in - and close to - the actinide region. For Z = 87 − 93 we detected nuclei which are only one or two neutrons away from the limits of the present chart of nuclides. The largest cross-sections were measured for nuclei which are located around the valley of beta stability. But also the most neutron-rich observed transfer products have still cross-sections up to several 100 µb. For example, the isotope ^238Pa is populated with a cross-section of 180 µb and the isotope ^232Fr with 15 µb. This indicates that also nuclei with still larger neutron numbers, and eventually also new isotopes, must have been populated in this experiment but could not be identified with our detection method due to their decay properties.

4.3 Summary, Conclusions and Outlook

Our studies of ^64Ni + ^207Pb and ^48Ca + ^238U collisions at Coulomb barrier energies showed that a vast region of nuclei is populated in deep inelastic transfer reactions with proton numbers around the target nucleus. In both systems, we identified relatively neutron-rich but not new isotopes. Concerning the population of nuclei in the region of N = 126, Z < 82, reactions with Pb targets turned out to be more favourable than reactions with U targets because they lead, for the same transfer products, to several orders of magnitude larger cross-sections. Our data and the ones from a previous experiment revealed that in transfer reactions cross-sections on the same order, or even larger, can be reached than in fragmentation reactions. This confirms the predictions of theoretical model calculations. However, a significant difference appears if one regards the expected count rates at the detector from both reactions which are about 1000 times larger in fragmentation reactions, resulting from the larger angular efficiency and effective target thickness. Even if an efficiency of 100% is assumed for a separation and detection setup for transfer products, fragmentation would still lead to comparable or higher count rates, respectively. Moreover, the isotope A and Z identification in fragmentation reactions can be performed by measuring energy, energy loss, time-of-flight and magnetic rigidity. A method which is not restricted to nuclei with appropriate decay properties like the identification of transfer products via alpha or beta decays. Concluding from the present situation, fragmentation reactions seem therefore superior to transfer reactions for producing as well as identifying neutron-rich nuclei along N = 126, Z < 82.

However, deep inelastic transfer reactions, in particular with the heaviest and most neutron-rich available targets like ^238U or ^248Cm, seem promising for the synthesis of new neutron-rich nuclei in the region Z > 92 - a region which cannot be accessed in fragmentation reactions and where transfer reactions would presently even be the only possibility. Already in our short beamtime of one day with the system ^48Ca + ^238U, we observed nuclei in the region Z > 84 with neutron numbers up to the limits of the present chart of nuclides. The data indicate that still more neutron-rich nuclei and also nuclei with Z > 93 must have been populated but could not be identified in our experiment due to their decay properties. Therefore, the most urgent point and necessary requirement for a successful application of transfer reactions for the synthesis of new isotopes is the development of detection techniques which allow the A and Z identification of very heavy and low-energetic nuclei, independent of their
decay properties. A possible method is precision mass measurements where a resolving power of $10^5$ is already sufficient for an isobaric identification of most of the heavy isotopes. A promising existing setup is the multiple reflection time-of-flight mass spectrometer MR-TOF-MS [12] which was developed for the Super-FRS project. It allows for a broadband detection of different isotopes and the complete setup, consisting of an ion catcher and the MR-TOF-MS, has an overall efficiency of about 20%, comparable to the efficiencies of present detection systems used in heavy element research.

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