Multinucleon transfer reactions – a pathway to new heavy and superheavy nuclei?

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Multinucleon transfer reactions – a pathway to new heavy and superheavy nuclei?

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Abstract. Recently, we reported the observation of several new neutron-deficient isotopes with proton numbers Z ≥ 92 in collisions of ^{48}Ca + ^{248}Cm at the Coulomb barrier. The peculiarity is that these nuclei were produced in deep inelastic multinucleon transfer reactions, a method which is presently discussed as a possible new pathway to enter so far unknown regions in the upper part of the Chart of Nuclides. Of particular interest are multinucleon transfer reactions as a possible means to produce neutron-rich superheavy nuclei and nuclei along the magic neutron shell N = 126. Based on present-day physical and technical state-of-the art, we will discuss the question how big are our chances to enter these regions by applying multinucleon transfer reactions.

1. Introduction

New exotic nuclei are usually produced by fragmentation, by fission or in complete fusion reactions. The different regions of nuclei, which can be populated with these reactions are shown in figure 1. These established reactions, however, are exploited or not applicable, respectively, in the region of transuranium and superheavy nuclei, causing the necessity for novel methods to synthesize new heavy isotopes. In the last decade an intense discussion arose on the possibility to apply multinucleon transfer (MNT) reactions for the synthesis of new heavy and superheavy nuclei. One region of interest is neutron-rich transactinide and superheavy nuclei which cannot be produced in complete fusion reactions due to the lack of sufficiently neutron-rich projectile-target combinations. The other goal is to produce new isotopes in the region of astrophysical interest along the closed N = 126 neutron shell. Nuclei in this region are presently produced in fragmentation reactions at relativistic energies but model calculations predict larger cross-sections for new isotopes in this region if they are produced in MNT reactions. Multinucleon transfer reactions might indeed be an appropriate solution. They allow, in principle, to produce neutron-rich as well as neutron-deficient isotopes with proton numbers reaching far beyond uranium. Experimental and theoretical studies to produce heavy nuclei in MNT reactions at Coulomb barrier energies were already performed in the 1970s up to the 1990s [1-4]. The lowest cross-sections reached in some of those experiments were 20 nb. No new isotopes were observed. A revival of the topic was initiated about 10 years ago by new model calculations [5, 6] and measurements (see e.g. [7]) of reaction cross-sections and by the above named necessity to find methods different from the established fusion and fragmentation reactions. Meanwhile, the possible

application of MNT reactions for synthesis of new isotopes has become a topical subject in various laboratories and appropriate separation and detection techniques are in development.

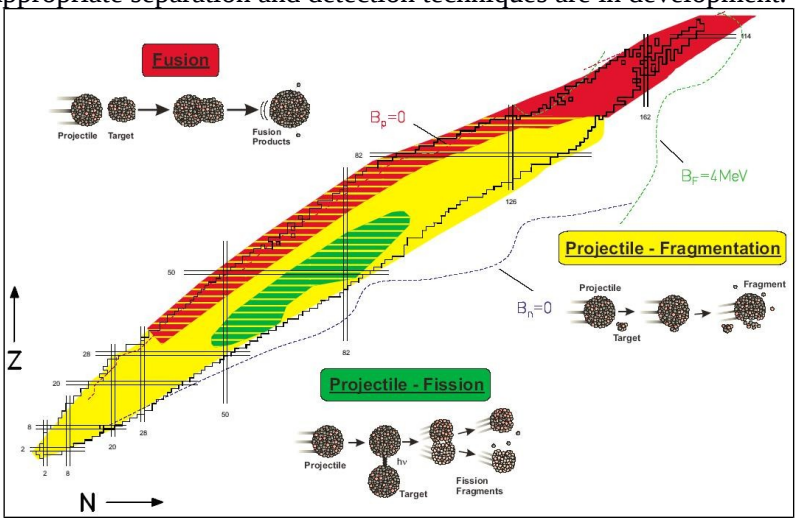

Figure 1. Chart of presently known nuclides and the reactions, which are typically applied to produce them [8].

Here, one has to keep in mind that the expected production cross-sections for new heavy and superheavy nuclei are small and reach far below the microbarn range. As a consequence, similarly efficient separation and detection techniques have to be applied as used for the identification of single atoms from fusion-evaporation reactions in superheavy element experiments. The techniques have also to be fast due to the short half-lives of exotic nuclei. Up to now no dedicated in-flight separators exist for heavy transfer products but the existing in-flight separators for heavy and superheavy evaporation-residues can also be applied for separation of transfer products. In particular, velocity filters are very sensitive to the reaction kinematics and allow for efficient separation between different isotopes. In recent years, we succeeded to produce for the first time new transuranium nuclei in MNT reactions [8] by using a highly sensitive experimental technique, namely, the velocity filter SHIP at GSI and its detection system.

2. Experimental technique

A sketch of the velocity filter SHIP and its detection system is shown in figure 2. Reaction products, which leave the target at angles of ±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 accepted velocity window at a given setting is $\Delta v/v = 0.1$ (FWHM). All reaction products, which pass the velocity filter are implanted in a position sensitive 16 strip silicon detector (stop detector) where they are identified via α-decay tagging. The identification via α-decay chains is a very sensitive method, which enables in the utmost case the identification of single isotopes. In addition, a germanium clover detector is installed behind the stop detector to measure γ-rays emitted in prompt or delayed coincidence with particle registration in the stop detector. The fast in-flight separation allows, in principle, to detect short-lived nuclei with half-lives down to microseconds. This limit is given by the flight time of the reaction products through the 11 m long SHIP, which is about 1 μs. In our experiment, the shortest accessible half-lives were 20 μs given by the conversion time plus dead tine of the data acquisition system while the longest accessible half-lives are determined by the data recording time and the yield of a specific isotope.

Velocity filters are very sensitive to the reaction kinematics. In the same way as SHIP is typically used to separate heavy and superheavy fusion-evaporation residues, it can equally be applied to separate heavy target-like transfer products. Transfer products, which are scattered to the acceptance

angle of SHIP have velocities between 1.5 times and 2 times the compound nucleus velocity v_{CN}. Usually, a broad variety of transfer products is created in the reactions where each of the nuclei has a characteristic velocity depending on A and Z and the energy dissipation during the reaction. In order to cover the range of different velocities expected in an experiment, we varied stepwise the velocity setting of SHIP and recorded at each setting the α- and γ-decays of the implanted reaction products and their daughter nuclei.

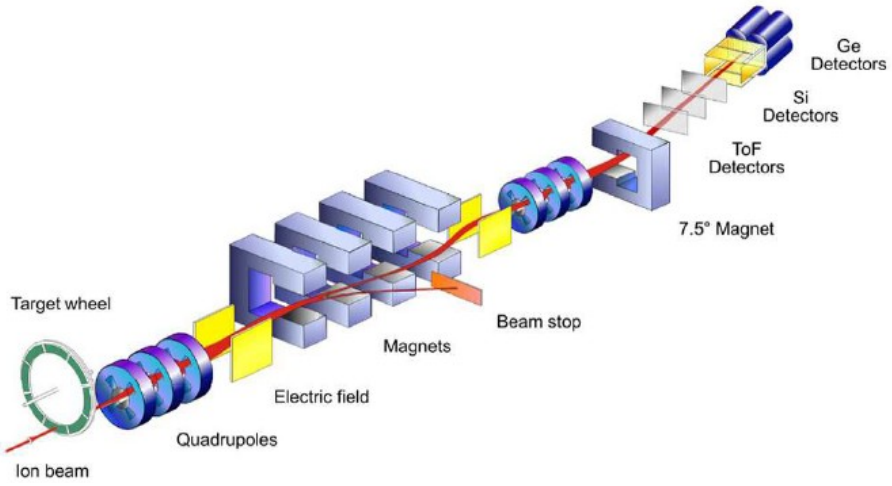

Figure 2. Sketch of the experimental setup: the velocity filter SHIP and the detection system.

In reactions at Coulomb barrier energies, the created MNT products have wide angular distributions, but due to the narrow acceptance angle of SHIP, only nuclei which are emitted in forward direction are accepted which results in typical efficiencies for MNT products between 0.1% and 1%. The selection of nuclei in forward direction means at the same time selection of nuclei from central collisions where projectile and target nucleus reach maximum possible overlap and sticking time at the given beam energy. This is the most favourable configuration for the exchange of large numbers of nucleons leading to MNT products far from the original projectile and target nuclei.

3. Experimental results

In reactions of ^{48}Ca + ^{248}Cm at 5.3 MeV/u we observed about 100 different nuclides, which were populated in MNT reactions (see grey squares in figure 3). The isotope identification was performed via the α-decay properties of the nuclei. Usually, a contiguous region of isotopes around the target and projectile nucleus is populated in transfer reactions. However, the nuclei, which are represented by blank squares in figure 3 could not be detected in our experiment due to too short or too long half-lives and / or unfavourable decay channels like fission or β-decay. Apart from a large number of already known nuclides, our data revealed also some new isotopes (marked by dots), which are located in the region $Z \geq 92$ on the neutron-deficient side of the Chart of Nuclides. All of them lead to decay chains of already known nuclei which enabled their assignment. For a detailed description of the experiment and results we refer to [10].

The cross-sections of the observed isotopes for elements Z = 84 – 95 are shown in figure 4 as a function of their mass number A, in the following called isotopic distributions. The cross-sections are related to the cumulative number of α-decays detected at (0 ± 2) degree. This means, they comprise for each isotope nuclei which were directly produced in the reaction as well as nuclei populated by precursor decays. The lowest measured cross-sections were on the level of 10 pb/sr, which corresponds to an average event rate of one nucleus per day in the stop detector at the given beam intensity of $2 \cdot 10^{12}$ projectiles / s and target thickness of 460 μg/cm^2.

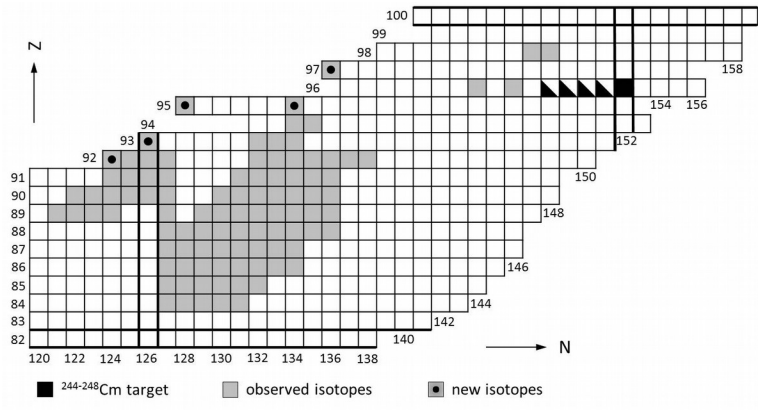

Figure 3. Survey of all multinucleon transfer products which we identified in reactions of ^{48}Ca + ^{248}Cm at the beam energy 5.3 MeV/u (grey squares). Beside ^{248}Cm with a contribution of 96.85% (black square) the target contained also traces of the isotopes $^{244-247}$Cm (black triangles). The five new isotopes, which we published in [9] are marked by dots.

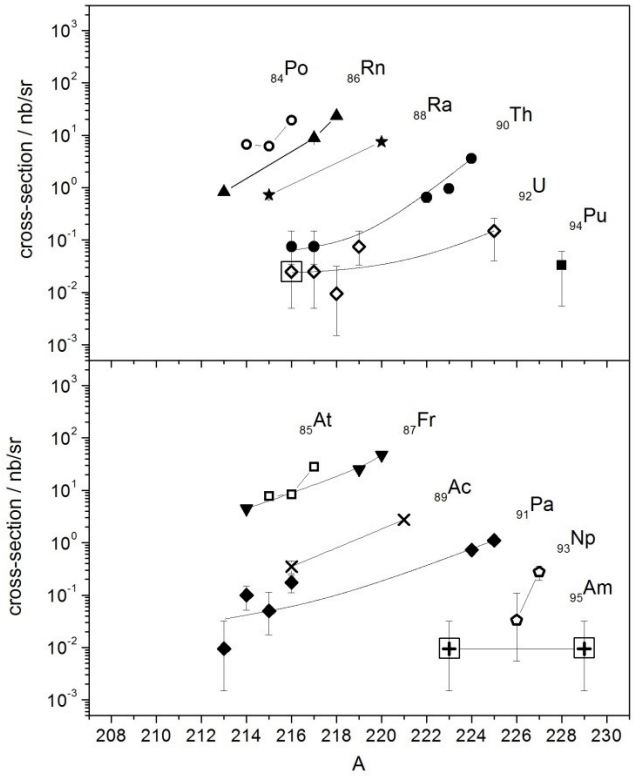

Figure 4. Isotopic distributions of multi-nucleon transfer products with Z = 84 – 95 measured in collisions of ^{48}Ca + ^{248}Cm at 5.3 MeV/u. The cross-sections are related to the cumulative number of α-decays which comprise decays of directly populated nuclei and of nuclei populated by precursor decays. Upper figure: distributions for even-Z nuclei; lower figure: distributions for odd-Z nuclei. The new isotopes are framed. The lines are drawn to guide the eye.

4. Discussion

The results of our experiment reveal that the application of a velocity filter in combination with α-decay tagging for the identification of the reaction products allows to reach in few days of beamtime cross-sections on the level of 10 pb/sr. According to model predictions, this is, in principle, already sufficient to reach new isotopes in the envisaged regions named above. In the following, we are going to shed more light on the possible application of MNT reactions to populate these regions.

4.1 Neutron-deficient transuranium isotopes

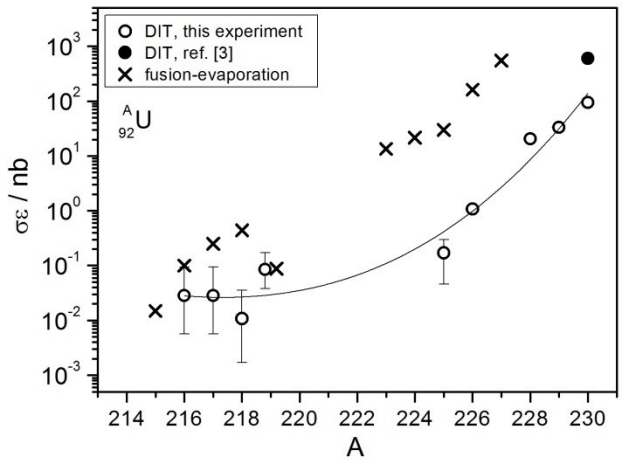

Figure 5. Product of cross-section and experimental efficiency, σε, for uranium isotopes measured in fusion-evaporation reactions (crosses) and in multinucleon transfer reactions of ^{48}Ca + ^{248}Cm in our experiment (open circles) and in the experiment in ref. [3] (full circle). For the transfer products we show cumulative cross-sections. For ^{219}U we put for better visibility a small offset on the A value to avoid the overlap of the data points from transfer and fusion.

The established method to produce neutron-deficient transuranium isotopes is fusion-evaporation reactions. Therefore, it is interesting to compare them with MNT reactions concerning the capacities of the two methods for isotope production. As a starting point we show in figure 5 measured fusion-evaporation cross-sections for various uranium isotopes [11 - 17] and MNT cross-sections measured in our experiment and in [3]. Figure 5 can be regarded as a representative example, because the behaviour of the cross-sections is similar for other elements in the uranium region. The fusion cross-sections for the same residual nucleus can vary by orders of magnitude depending on the applied projectile-target combination and beam energy. If cross-sections from more than one experiment were available for the same isotope, we put the largest known value in figure 5. Concerning MNT reactions, it is very likely that the given cross-sections have not the maximum achievable values, because systematic studies to find the optimum projectile-target combinations and measurements of excitation functions for the given MNT products were not performed yet.

Instead of the pure reaction cross-sections we show in figure 5 the product of cross-section and experimental efficiency, σε, because this is the experimentally relevant parameter which reflects the event count rate. In our case, ε is mainly determined by the angular acceptance of the applied separators for fusion and transfer products, respectively. The following trend is noticed in figure 5: for the more neutron-rich isotopes well above A = 220, σε is significantly larger in fusion-evaporation reactions. This is on one hand caused by the significantly larger angular acceptance of the separators for fusion-evaporation residues. Another reason is that these isotopes can be produced with very asymmetric projectile-target combinations like ^{22}Ne + ^{208}Pb → ^{230}U* where the low entrance channel Coulomb barriers and favourable Q-values lead to relatively large fusion residue cross-sections. In this region, fusion-evaporation reactions appear superior to MNT reactions. For very neutron-deficient isotopes the situation changes and σε for fusion and MNT products tend to approximate each other despite the small efficiencies for MNT products. To reach these nuclei in fusion reactions, more symmetric projectile-target combinations have to be applied like e.g. ^{40}Ar + ^{182}W → ^{222}U*. The larger entrance channel Coulomb barriers and less favourable Q-values lead to relatively small fusion-evaporation residue cross-sections. In this region, a clear advantage of MNT reactions comes into play, namely, their broad excitation functions. Resulting from this, a vast region of isotopes can be populated and detected simultaneously at a given beam energy while fusion-evaporation reactions are only selective on few isotopes at a given experimental setting. Therefore, MNT reactions appear indeed as an attractive way for the production of new neutron-deficient transuranium isotopes.

4.2 Neutron-rich superheavy nuclei

There is presently no experimental method to produce neutron-rich nuclei well beyond uranium. Fragmentation reactions are not applicable because the heaviest available projectile or target nucleus, respectively, is uranium and with fusion reactions only neutron-deficient heavy nuclei can be produced due to the bending of the stability line toward the neutron axis with increasing Z. Therefore, MNT reactions are presently the only potential method to access these nuclei. The sensitivity reached in our experiment on ^{48}Ca + ^{248}Cm allows already, in principle, to enter the region of neutron-rich nuclei with Z > 100 where cross-sections in the nanobarn region are expected from model calculations for new neutron-rich isotopes around Z = 102. The main bottleneck, which presently prevents the observation of MNT products in this region is the lack of appropriate identification techniques. Alpha decay tagging is not applicable because the expected dominant decay mode of neutron-rich superheavy nuclei is spontaneous fission. Moreover, also the half-lives can become too long for decay tagging in this region. Also the determination of A and Z by measurement of energy, energy loss and time-of-flight is not possible because at energies around the Coulomb barrier plasma effects and the resulting pulse height deficit do not allow to determine A and Z of very heavy nuclei with an accuracy of one unit.

A satisfactory solution for this problematic is not yet existing. There are different approaches for new identification techniques which are, however, not satisfactory, mainly from terms of their present efficiency. One approach is to determine the nuclear charge number by selective laser ionization while the mass number of the singly ionized nuclei can be obtained with a magnetic dipole field. Another approach is the application of precision mass measurements with a Penning trap or multiple reflection time-of-flight mass spectrometer where a resolving power of 10^5 – 10^6 is already sufficient for an isobaric separation of most of the heavy nuclei. A third approach, especially for A identification, is the application of E-TOF telescopes where calorimeters are used for energy measurement instead of silicon detectors. Calorimetric low temperature detectors were already studied for slow heavy ions and allow for a resolution of A close to one unit even for nuclei in the uranium region [18]. However, all these methods lead presently to tiny detection efficiencies, which are far from allowing the detection of single nuclei.

4.3 Nuclei along the N=126 neutron shell

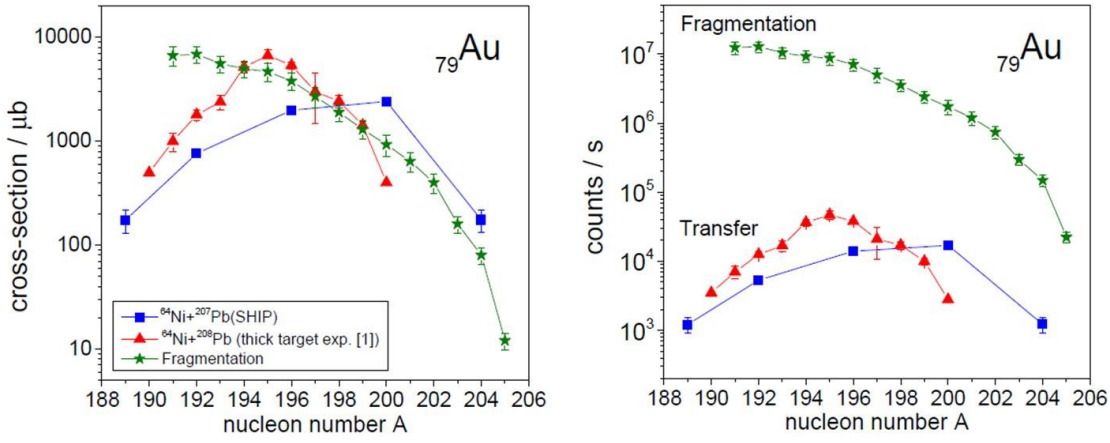

Figure 6. Production cross-sections and expected yields (at the target) for Au isotopes produced in multinucleon transfer reactions of ^{64}Ni + ^{207,208}Pb and in fragmentation reactions.

In collisions of ^{64}Ni + ^{207}Pb at 5.0 MeV/u we studied the population of isotopes below Pb in MNT reactions [19]. The nuclides below Pb are β^- emitters (on the neutron-rich side of the stability line) or

undergo electron capture decay (neutron-deficient side). Therefore, we used delayed γ-rays emitted by the reaction products, which are implanted in the stop detector, for isotope identification. This method is less sensitive than α-decay tagging and lead to lower cross-section limits of 10 μb//sr. We identified several isotopes of elements Z = (76 – 81). As a representative example, we show in figure 6 the results for Au (Z = 79) isotopes (full squares). Similar results were obtained 10 years earlier by another group in a thick-target experiment performed with collisions of ^{64}Ni + ^{208}Pb [20]. The results of this experiment are shown by triangles in figure 6.

The left part of figure 6 shows the production cross-section of Au isotopes as a function of their nucleon number A. Our cross-sections and the ones measured in [20] differ in most cases within one order of magnitude. The data from MNT reactions of both experiments indicate a rather steep downward slope toward the neutron-rich side after the maximum is reached around nuclei located close to the stability line. In both experiments no new isotopes were observed and as well the N = 126 shell was not reached for nuclei with Z < 82. Since the presently applied technique to produce neutron-rich isotopes below Pb is projectile fragmentation, it is interesting to compare the isotopic distributions and cross-sections measured in MNT reactions with those from fragmentation reactions [21, 22]. The fragmentation data are represented by asterisks. Also the fragmentation cross-sections show a steep decrease towards neutron-rich nuclei but the experiments are more sensitive and allowed to reach cross-sections below 10 nb. The fragmentation and MNT cross-sections are mostly within the sane order of magnitude for the isotopes discussed here. We could notice the trend that the MNT cross-sections are going to exceed the ones from fragmentation if one goes further below Pb, which is well in agreement with theoretical predictions.

Table 1. Typical parameters for beam intensities N_{beam}, target thicknesses d_{target} and angular efficiencies ε in multinucleon transfer and in fragmentation reactions.

	N_{beam}	d_{target}	ε
Transfer	$5 \cdot 10^{12}$ / s	500 μg / cm^2	< 5% (SHIP)
Fragmentation	$5 \cdot 10^{9}$ / s	5 g / cm^2	< 50% (FRS)

Next, we are going to compare the production *yields* for the observed isotopes from MNT and fragmentation because this is finally the decisive criterion for the successful application of a reaction type for isotope production. To deduce yields from the crtoss-sections, one has to take into account the different conditions concerning beam intensities, target thickness and detection efficiencies for MNT and fragmentation products. Typical parameters are shown in table 1. The yields (at the target) for Au isotopes are shown on the right side of figure 6. One can see that the estimated yields for the same isotopes are one or more orders of magnitude larger in fragmentation reactions. If we regard in addition the expected yields at the detector, the difference between the two production methods is still increasing in favor of fragmentation reactions caused by the very different angular distributions of MNT and fragmentation products. In projectile fragmentation reactions the nuclei are emitted in a very narrow forward cone due to the relativistic energies and inverse kinematics. Resulting from this, very large angular efficiencies of nearly 100% can be reached despite of the small acceptance angles of the separators. MNT products, in contrast, are emitted in a broad angular window of typically 50 degree or more which makes an effective application of in-flight separators difficult. Finally, another point has to be mentioned. In fragmentation reactions, the identification of the isotopes is performed by measuring energy, energy loss, time-of-flight and magnetic rigidity of the nuclei. Due to the relativistic energies, A and Z of the nuclei can be resolved with one unit. In MNT reactions at

Coulomb barrier energies, plasma effects and the resulting pulse height deficit in the silicon detector limit the A and Z resolution, which is usually not better than three units in the region around Pb.

These results and considerations show that fragmentation reactions appear much more favorable for the production of neutron-rich nuclei along the N = 126 shell. And based on present day knowledge, this will be also valid for future experiments, in particular after the upgrades of beam intensities which are ongoing in present radioactive ion beam facilities.

5. Summary and Outlook

As outlined in the previous sections, MNT reactions in heavy collision systems are presently discussed as a possible new pathway to produce new isotopes in the following regions on the upper part of the Chart of Nuclides: (i) neutron-rich transuranium and superheavy nuclei, (ii) neutron-deficient transuraniun nuclei and (iii) neutron-rich nuclei with Z < 82 along the N=126 neutron shell. What can we conclude for the success of this enterprise? Based on the presently available, still rather scarce experimental data one can say that there is no overall valid answer concerning the three regions of interest. Rather, it seems that we have to treat each region individually and arrive at quite different conclusions. The answer is rather optimistic concerning the production of very neutron-deficient transuranium isotopes. Here it even seems that MNT reactions might be more profitable compared to the presently applied fusion reactions. The expected yields from MNT are comparable to the ones from fusion reactions, but a big advantage of MNT reactions is that a wide region of isotopes can be populated in the same experiment while fusion-evaporation reactions are selective on only few isotopes at given beam energy and experimental setting. This is due to the broader excitation functions of MNT products.

For the production of neutron-rich transuranium nuclei, the cross-sections obtained in MNT reactions seem at least sufficient to produce isotopes of elements up to Z ≈ 105. However, here appears the bottleneck of missing identification techniques for single heavy nuclei. The only existing powerful method of alpha decay tagging cannot be applied because nuclei in this region are very long-living and mostly do also not decay via alpha emission. Nevertheless, MNT reactions seem presently the only possibility to enter the region of neutron-rich transuranium and superheavy nuclei. Therefore, the first and decisive step is the development of new and powerful detection methods, which allow the identification of these nuclei independent of their decay properties.

For the third region of nuclei along N=126, Z < 82 the answer seems quite sure that projectile fragmentation reactions will also in the future remain the most powerful method for their production. Even if the production cross-sections in fragmentation and MNT turned out to be similar, the yields in fragmentation reactions are several orders of magnitude higher because of the more favourable conditions concerning target thickness and experimental efficiencies. Further, in fragmentation reactions the problem of isotope identification does not appear like in MNT reactions. Due to their relativistic energies, A and Z of the fragmentation products can be determined by measuring energy, energy loss, time-of-flight and magnetic rigidity.

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