Eur. Phys. J. D **45**, 75–80 (2007)
DOI: 10.1140/epjd/e2007-00033-9

THE EUROPEAN PHYSICAL JOURNAL D

Physical preseparation: A powerful new method for transactinide chemists

Ch.E. Düllmann[a]

Gesellschaft für Schwerionenforschung mbH, Planckstrasse 1, 64291 Darmstadt, Germany

Received 22 September 2006

Abstract. In recent years, the concept of physical preseparation of single atoms was introduced into the field of transactinide chemistry. In this approach, the transactinide element of interest is isolated in a physical recoil separator and then extracted from this machine. The beam as well as the unwanted reaction products are strongly suppressed, allowing for the investigation of new chemical systems that were not accessible before. The most important aspects of the technique are discussed and the advantages for chemistry experiments with transactinides are presented, using some of the chemical studies that were performed with preseparated isotopes as examples.

PACS. 25.70.-z Low and intermediate energy heavy-ion reactions – 25.70.Gh Compound nucleus – 25.70.Jj Fusion and fusion-fission reactions – 29.25.Rm Sources of radioactive nuclei

1 Introduction

The chemical study of the transactinide (TAN) elements [1,2] is a challenging field of research as the maximum production rates are of the order of a few atoms per hour, day, or even week and the lifetimes of even the longest-lived isotopes are of the order of a few tens of seconds at most. Nevertheless, experimentalists have reached hassium (Hs, element 108) [3,4] and are currently tackling the chemistry of element 112 [5,6]. Elements up to Hs were shown to generally behave in accordance with their expected position in the periodic table of the elements. Looking at the full body of data available on the transactinide elements [1,2] obtained in both, the gas and the aqueous phase, it becomes clear that a very limited set of chemical systems has been investigated so far, compared to any of the lighter elements, where hundreds or thousands of compounds are known for almost all elements. Aside from the difficulties inherent to such studies, as the short half-lives and the small production rates, also technical problems have prevented more advanced studies. In order to understand this, two common techniques that were used in most of the past transactinide experiments will briefly be described before preseparation, a new method, is introduced.

1.1 Aerosol particle gas-jet

In this technique [7–9], the nuclear reaction products are thermalized in a gas-filled volume located directly behind the target, the recoil-chamber. Not only the transactinide element of interest but also the majority of unwanted products such as transfer products or products of the interaction of the beam with the target assembly or impurities in the target is thermalized in this volume. Furthermore, the intense heavy-ion beam produces a plasma in the recoil chamber. The recoil chamber is flushed with rapidly flowing gas, usually He, which is enriched with 10^6–10^7 aerosol particles/cm^3 (usually KCl, MoO_3, or C). Non-volatile thermalized reaction products attach to the surface of these particles which are transported with the gas flow at high velocities with high yields, thus enabling efficient transport of the elements of interest (but also of unwanted reaction products) to a chemistry setup located outside the irradiation cave.

1.2 Transport in the form of a volatile species without aerosol particles

In this approach [10], which was successfully used, e.g., in the chemical investigation of Hs in the form of HsO_4 [3,4], a highly volatile species is formed in-situ in the recoil chamber and transported to the chemistry setup in this form. This technique is expected to be suitable and highly efficient for the presumably very volatile p elements 112 and 114 [11,12] as tests with Hg and Rn showed [5,6].

Both approaches have inherent disadvantages as far as the chemical investigation of TAN elements is concerned: in the first approach, there is no selectivity between desired and unwanted reaction products and a chemical system with a high separation factor for the element of

---

[a] e-mail: `c.e.duellmann@gsi.de`

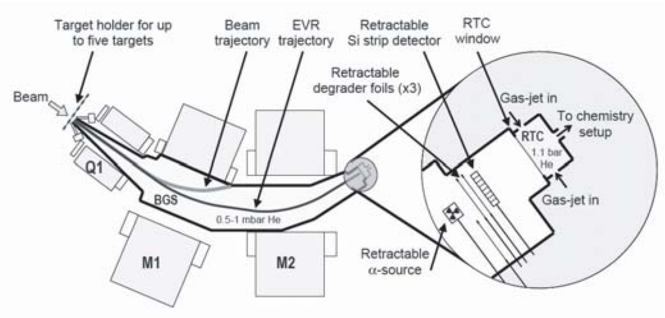

Fig. 1. Schematic of the BGS setup at LBNL in the configuration as a physical preseparator for chemistry experiments [15].

interest is needed in order to allow for its unambiguous identification. This selectivity has to be favored over any other property, thus severely limiting the number of available chemical systems. It is also well-known that the yield of aerosol gas-jets drops dramatically as the beam intensity reaches high levels due to the destruction of the aerosol particles in the plasma present in the recoil chamber. Both approaches, additionally, involve very violent conditions with respect to, e.g., the investigation of non-thermally stable compounds such as organometallic ones: in the first approach, high temperatures (∼1000 °C) are required for the destruction of the aerosol particles which then releases the transported atoms and allows their conversion into volatile species. In the second approach, a plasma is present in the recoil chamber as described earlier. Thus, neither of these techniques are particularly well suited to study such chemical systems.

1.3 The method of physical preseparation

Physical preseparation has been pioneered at Lawrence Berkeley National Laboratory (LBNL) [13–15] using the Berkeley Gas-filled Separator (BGS) [16] as a preseparator. The method can be described as follows: the desired nuclear species are produced in a heavy-ion-induced fusion reaction and separated from the beam and the majority of the unwanted nuclear reaction products in a physical recoil separator. At its exit, they are extracted from the separator and thermalized, e.g., in a gas-filled volume which is referred to as the Recoil Transfer Chamber (RTC) [17]. A schematic of the setup used at LBNL is depicted in Figure 1.

At the GSI in Darmstadt, Germany, a new gas-filled separator, the TransActinide Separator and Chemistry Apparatus (TASCA) [18] has recently been constructed and entered the commissioning phase in the spring of 2006. One of the main foreseen applications is its use as a preseparator for chemical studies and all aspects of the system were optimized with respect to this field of research. Other laboratories are currently in the process to upgrade existing physical recoil separators to preseparators, e.g., the group at RIKEN [19] which is operating the GAs-filled Recoil Ion Separator (GARIS) [20].

2 Aspects of preseparation

In order to perform experiments with preseparated isotopes, there are a number of aspects that need to be considered beforehand. Some of the pertinent ones will be discussed in the following, concentrating on the specific features of TASCA.

2.1 Beam/target combination

Most of the current generation setups for the chemical investigation of transactinides require isotopes with lifetimes of at least a few seconds [1,2]. The lifetimes of even the longest lived isotopes are often comparable to the time that the chemical procedure requires. Therefore, the longer the lifetime, the higher is the yield and thus the higher the rate of detected atoms, showing clearly that studies with the longest-lived available isotopes are highly desirable. These are only accessible in relatively asymmetric reactions with light beams with masses in the range of about 20–30 amu and actinide targets. In experiments with preseparation, there is an additional constraint that is absent in studies with more conventional techniques. As the species traveling through the preseparator need to penetrate an interface called the RTC window, which is separating two pressure regimes with a pressure difference of about 1–2 bar, the kinetic energy of these species needs to be relatively high. The exact threshold depends on the design of the interface and the preseparated species. Current setups use thin Mylar foils of $\geqslant 3.3\ \mu$m thickness [15], necessitating highest possible kinetic energies of the reaction products and most likely the development of new interfaces with thinner windows. The lightest target, i.e., the target that can be combined with the highest mass projectile and thus leads to the most energetic recoils, which allows production of the relatively long-lived isotopes of the lighter TANs is ^{244}Pu. In reactions with projectiles such as ^{22}Ne, ^{23}Na, ^{26}Mg, ^{27}Al, and ^{30}Si, isotopes of Rf-Hs with long enough half-lives for chemical studies can be produced. ^{48}Ca beams furthermore would give access to the claimed 3-s ^{289}114 and its daughter, 30-s ^{285}112 [21]. Therefore, ^{244}Pu is currently considered a very good target for studies with preseparated isotopes [22].

2.2 Configuration of preseparators

In this article, TASCA [18] is used as a model to present the important aspects of preseparation. It is worth mentioning here that preseparators do not necessarily have to be of the gas-filled type. Also, not all preseparators employ the same magnetic configuration. The BGS [16], for example, employs the magnetic configuration QD_hD where D and Q denote dipole and quadrupole magnets, respectively. The second magnet is a gradient-field dipole magnet with a horizontally focusing component as indicated by the index. The TASCA device on the other hand employs the magnetic configuration DQQ [23]. To get maximum possible transmission values for TASCA, new vacuum chambers were constructed. As a special feature, the vacuum chamber in the quadrupole section was designed to have a butterfly like cross section where the walls of the chamber closely follow the contours of the magnet tips. This increases the solid angle and thus the transmission of TASCA significantly.

2.3 TASCA mode: DQ_hQ_v vs. DQ_vQ_h

Another special feature of TASCA is the possibility to operate it in two different modes of polarity for the quadrupole magnets: DQ_hQ_v vs. DQ_vQ_h where the index refers to vertical (v) or horizontal (h) focusing [23]. These two modes lead to different images in the focal plane. The first mode provides maximum available transmission but results in a relatively large image [23,24]. Monte-Carlo simulations [25] suggest an image size of roughly 4×14 cm^2 for the reaction ^{244}Pu(^{22}Ne, 5n)^{261}Rf [24], which has to be verified experimentally. At the cost of a somewhat reduced transmission, the second mode yields a much smaller image. For the reaction ^{208}Pb(^{50}Ti, n)^{257}Rf it is predicted to be roughly 3×4 cm^2 large [24]. Depending on the half-life of the isotope of interest, it might be advantageous to sacrifice some of the transmission in order to be able to use the smallest possible window and thus RTC volume size, as decay losses in the RTC can easily dominate the overall yield over the transmission of the separator [26]. It is therefore expected that experiments with relatively long-lived isotopes will employ the "high transmission mode" DQ_hQ_v while studies of, e.g., ^{257}Rf ($T_{1/2} \sim 4$ s) produced in the relatively symmetric cold fusion reaction Ti+Pb (where the transmission is rather high anyway) will employ the "small image mode" DQ_vQ_h.

2.4 Magnet settings

In order to focus the desired reaction products in the center of the focal plane, it is crucial to properly set the magnetic fields. Deflection in a gas-filled dipole magnet as used in TASCA is governed by the magnetic rigidity $B\rho$ of the ion traveling through the gas and can be described by the following relationship [27]:

$$B\rho = mv/q_{ave}e \qquad (1)$$

where B is the magnetic flux density, ρ is the radius of curvature of the ion's path, m and v are the mass and velocity of the ion, respectively, q_{ave} is the average charge state of the ion, and e is the electron's charge. A good overview of the basic operation principle of gas-filled separators can be found in [27]. As follows from equation (1), the magnetic rigidity of an ion depends on its velocity, mass, and charge. While the first two properties are given by the nuclear reaction parameters, i.e., mass of projectile and target nuclei and the beam energy, evaluation of the average charge state is not as straightforward. Due to collisions with atoms of the fill gas, the ions take on an average charge state independent of their initial charge state. This average charge state follows a general trend $\propto vZ^{1/3}$ [27]. However, deviations that seem to be related to the shell structure of the stripped ion in the gas are observed, as can be clearly seen in Figure 3 in [28]. A recent semi-empirical fit to an extensive set of measured charge states for heavy ions that is based on the shell structure of the stripped ion traveling through the gas allows to predict charge states of reaction products with relatively high accuracy [29] as can be seen from the quality of the fit shown in Figure 2. As q_{ave} is often approximately proportional to v, not only charge focusing but also velocity focusing takes place in a gas-filled separator, leading to high charge and momentum acceptances which are characteristic features of gas-filled recoil separators.

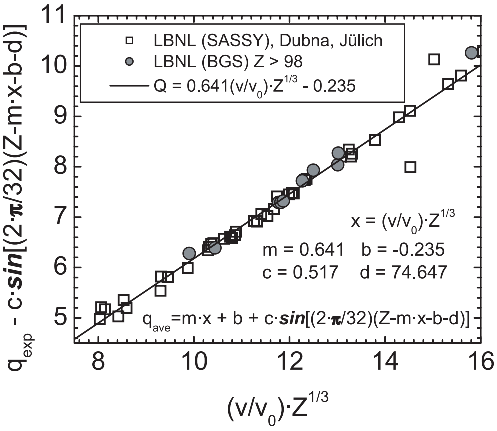

Fig. 2. Average charges of heavy ions passing through dilute He gas. The ordinate shows the experimental average charges, with the sinusoidal correction for the electronic shell structure of the stripped ions removed. The best fit is indicated by the solid line. Adapted from [29].

When the ions are traveling through He with a velocity below a certain threshold which is approximately $(v/v_0)Z^{1/3} \lesssim 6$ where v_0 denotes the Bohr velocity ($\approx 2.2 \times 10^6$ m/s), the fit shown in Figure 2 is not as good as for fast ions. This is thought to originate from the relatively high ionization potential of He. Systematic studies of slow evaporation residues are under way at LBNL [30]. These data together with data from [31,32] provide a basis to add a correction term for slow ions to extend the validity of the fit.

Therefore, all properties that govern the deflection of ions in a gas-filled dipole seem to be understood well enough that remaining uncertainties are small enough to not cause a substantial fraction of evaporation residues of interest to miss the RTC window.

The other type of magnetic elements commonly used in gas-filled separators is multiplets of quadrupole magnets which are used to focus the species into the focal plane. For TASCA, ion-optical calculations [23] with the code TRANSPORT [33,34] as well as Monte-Carlo simulations [25] have been performed [24] and provide a basis for the proper magnet settings. The experimental verification of these predictions is under way.

2.5 Fill gas

The above discussion concentrated fully on He as fill gas. This is connected with the fact that a large body of data is available for He and that there are no safety concerns connected with its use, in contrast to, e.g., hydrogen. During the commissioning phase, TASCA will be used exclusively with He as a fill gas. However, the option to later use other gases such as H_2, Ne, N_2, or any kind of gas mixture is already being implemented into the separator control and safety system from the very beginning on.

2.6 Design of the RTC window unit

The technical realization of the interface between the separator and the RTC regimes is quite demanding [15,17]. The following requirements have to be fulfilled: (i) the interface has to withstand a pressure difference of 1–2 bar; (ii) the leak rate of the gas in the RTC into the separator has to be as small as possible (the maximum tolerable leak rate depends on the type of separator with vacuum systems putting much more stringent limits. In case of one and the same gas being used in the RTC as well as the fill gas in the separator, the acceptable leak rate from the RTC into the separator is substantially higher than in the other cases. In the extreme case, the full amount of gas supplied to the separator is coming from the RTC through the RTC window); (iii) the smallest possible layer thickness in terms of stopping power for high-Z low-velocity ions should be used, and (iv) in the case of chemicals being fed through the RTC, the window has to be made from a material that is able to withstand these chemicals.

In the first generation RTC built in Berkeley [17], 6-μm thick Mylar held by a grid made from metal wires was used. The geometrical transparency of the supporting structure was very high, about 90%. However, the thick layer of material allowed the use of relatively symmetric reactions only. This structure was used for experiments with, e.g., the following reactions: ^{118,120}Sn(^{56}Fe, xn)$^{169-173}$Os [17]; ^{116,120,124} Sn(^{50}Ti, xn)^{162,165,169}Hf [15]; ^{208}Pb(^{50}Ti, n)^{257}Rf [14,26]; ^{209}Bi(^{50}Ti, n)^{258}Db [35]. Further studies of group 4 elements including the lighter homolog, Zr, necessitated fabrication of a thinner window as the recoil range of Zr recoils produced in the reaction $^{\mathrm{nat}}$Ge(^{18}O, xn)^{85}Zr in Mylar was less than 6 μm. Therefore, a new support structure with a honeycomb pattern of 6.25-mm diameter holes was built that allows use of 3.3-μm thick Mylar foils [15].

For asymmetric reactions as they will be used at TASCA, even this is still too thick. Therefore, work is ongoing within the TASCA collaboration to construct a new interface based on yet thinner materials. Such windows need support structures with smaller open areas in order to withstand the pressure difference; however, the transmission should still be as high as possible.

3 What can chemists do with preseparated isotopes?

Three examples will be briefly introduced to show the power of the method of physical preseparation.

3.1 Automated liquid-liquid extraction studies of Rf with SISAK

The automated on-line liquid-liquid extraction system SISAK [36] was developed for the investigation of very short-lived species. In this technique, the distribution ratio of the element of interest between two phases is measured. The SISAK version used for investigating the chemical properties of transactinides is coupled to liquid scintillation (LS) detectors [37] that are very sensitive for the detection of α-particles and fission fragments. A drawback of this detection scheme is the fact that the light yield per energy unit for β-particles is roughly a factor of ten higher than for α-particles, leading to similar pulse heights in the spectra for the two kinds of events. Additionally, the energy resolution of the LS detectors is significantly lower than for solid state detectors. In experiments performed in the 1990s where SISAK was used to investigate non-preseparated Rf isotopes [38], the background from transfer products and reaction products of the beam with impurities in the target, the target backing, and the target assembly, was relatively high and prevented the unambiguous identification of single Rf atoms. Only after SISAK was installed at LBNL and used ^{257}Rf preseparated in the BGS, it was possible to identify single Rf atoms with this technique [14,26,39]. These studies signify the first successful investigation of a TAN element using preseparated isotopes and as of 2005, Rf was successfully investigated in three different chemical systems [26,35].

3.2 Extraction of Zr and Hf with crown ethers: studying trends in group 4

An important advantage of studies with preseparated isotopes is the almost complete absence of unwanted reaction products. Therefore, no isotopes with decay properties similar to those within the decay chains of the transactinide element of interest are present in the RTC. In contrast to conventional experiments, where the burden of isolating the element of interest was on the chemical separation procedure, there are no such restrictions present anymore and chemical systems with only low separation factors for the studied element from other problematic elements can now be investigated. As an example of such studies, extraction experiments with Zr and Hf [40] performed in Berkeley shall briefly be mentioned. Relatively short-lived Zr and Hf isotopes were produced and preseparated in the BGS. They were transported to the chemistry setup with a KCl-gas-jet and transferred to an aqueous phase. Their extraction from concentrated hydrochloric acid into two different crown ethers (Fig. 3), dicyclohexano-18-crown-6 (DC18C6) and

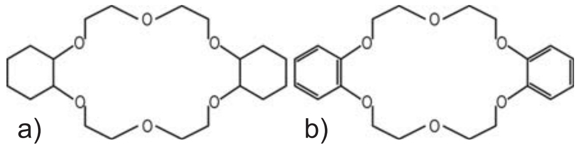

Fig. 3. The structure of dicyclohexano-18-crown-6 (DC18C6) (a) and dibenzo-18-crown-6 (DB18C6) (b).

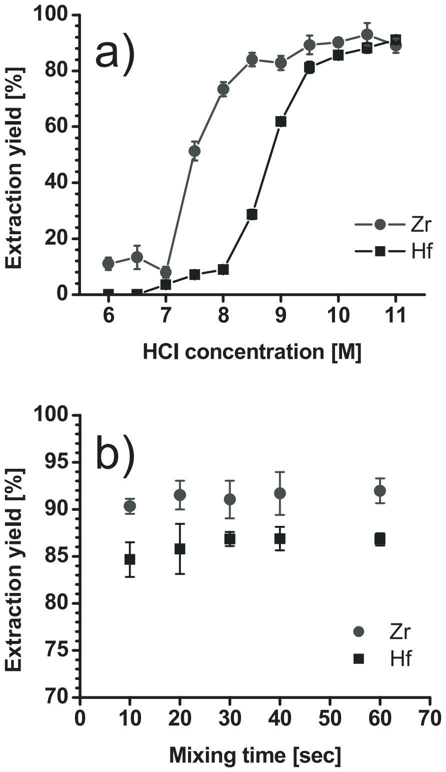

Fig. 4. (a) Extraction of ^{85}Zr and ^{169}Hf into 0.025 M dicyclohexano-18-crown-6 in dichloromethane as a function of hydrochloric acid concentration. (b) Extraction yields for Zr and Hf extraction from 10.0 M HCl into 0.025 M dicyclohexano-18-crown-6 as a function of mixing time [40].

dibenzo-18-crown-6 (DB18C6) in dichloromethane was studied as a function of acid and crown ether concentration.

Similar extractions are used industrially to separate these two homologs of rutherfordium, which are known to notoriously exhibit extremely similar chemical properties, rendering their separation problematic in many chemical separation systems. The crown ether extraction system therefore represents one of the few chemical systems where they behave differently. As many mono- and divalent species are also extracted with the crown ethers, such experiments depend on the availability of preseparated isotopes.

Some of the results obtained in [40] are shown in Figure 4.

Results shown in Figure 4a clearly show the behavior of Zr and Hf to be different, and an experiment with Rf at 8 or 8.5 M HCl will show whether this element is more closely resembling Zr, or Hf, or following the trend established by its two lighter homologs in group 4. Results of kinetic studies shown in Figure 4b where the mixing time was reduced down to 10 s show that extraction is in equilibrium after already 10 s (or even faster), rendering this system fast enough for an experiment with 78-s ^{261}Rf.

3.3 Studies of volatile Zr and Hf complexes with hexafluoroacetylacetone

Gas chemical methods have proven powerful for investigations of the heaviest elements and were used for the first chemical investigations of the heaviest elements currently chemically characterized [1,2]. However, many chemical systems were not accessible in conventional experiments due to the presence of a plasma in the recoil chamber. Preseparation is a promising technique to overcome this limitation. As a first system to test the hypothesis that the direct introduction of organic ligands into the RTC should allow the in-situ formation of volatile metal complexes was tested with β-diketonates of Zr and Hf [41–43] the lighter homologs of Rf. Hexafluoroacetylacetone (hfa) complexes of these elements are well-known to be very stable and highly volatile [44].

Zr and Hf isotopes with half-lives of a few tens of seconds to a few minutes were produced at the Lawrence Berkeley National Laboratory 88-Inch Cyclotron. The nuclear reaction products were preseparated in the BGS [16] and thermalized in the RTC, which was flushed with hfa-vapor-containing helium. The reaction products were transported with the gas flow to an oven installed directly at the exit of the RTC. There, formation of volatile complexes of the type M(hfa)$_4$ (M = Zr, Hf) occurred. Single molecules of these complexes are volatile at room temperature and were transported with the gas flow to activated charcoal catchers through a Teflon capillary. The absolute overall yield of this process was measured for ^{162,165,169}Hf. From these data, the overall "reaction plus transport" time was determined. The minimum time achievable with the current setup is of the order of a few tens of seconds [42], indicating that the system should be suitable for an experiment with 78-s ^{261}Rf.

These experiments, which nicely illustrate the possibilities that the technique of preseparation opens up, are expected to be extended to Rf in the near future.

4 Summary

The coupling of chemistry setups for experiments with transactinide elements to a physical recoil separator promises to open up new possibilities and allow expanding the investigations of these elements to chemical systems that were not previously accessible. The conditions in the recoil transfer chamber are much more favorable for chemical studies than those in a recoil chamber directly attached to the target: (i) the intense primary heavy-ion beam is absent and hence there is no plasma present, and (ii) the unwanted byproducts of the nuclear reaction are strongly suppressed. At GSI, the new gas-filled separator TASCA has entered the commissioning phase. One of the research programs to be conducted at TASCA will be its use as a preseparator and it was designed to be especially suitable for this task. The important parameters for performing such experiments are understood. First experiments with preseparated isotopes were conducted at LBNL using the BGS and have proven the power of the method.

It is my pleasure to thank many colleagues working in this interesting field of nuclear science for interesting discussions, especially the ones from the heavy element and nuclear chemistry groups at LBNL and GSI as well as from the TASCA community whose work is cited in the references. Ken Gregorich, Hiromitsu Haba, Jon Petter Omtvedt, and Alexander Yakushev communicated advances and recent results prior to publication, which is gratefully acknowledged.

References

1. M. Schädel, Angew. Chem. Int. Ed. **45**, 368 (2006)
2. *The Chemistry of Superheavy Elements*, edited by M. Schädel (Kluwer Academic Publishers, Dordrecht, 2003)
3. Ch.E. Düllmann et al., Nature **418**, 859 (2002)
4. A.V. Zweidorf et al., Radiochim. Acta **91**, 855 (2003)
5. A. Yakushev et al., Radiochim. Acta **91**, 433 (2003)
6. R. Eichler et al., Radiochim. Acta **94**, 181 (2006)
7. H. Wollnik et al., Nucl. Instrum. Meth. **127**, 539 (1975)
8. H. Wollnik, Nucl. Instrum. Meth. **139**, 311 (1976)
9. H.W. Gäggeler et al., Nucl. Instrum. Meth. A **309**, 201 (1992)
10. Ch.E. Düllmann et al., Nucl. Instrum. Meth. A **479**, 631 (2002)
11. B. Eichler, Kernenergie **19**, 307 (1976) (*in German*)
12. K.S. Pitzer, J. Chem. Phys. **63**, 1032 (1975)
13. *Proc. Workshop on the Physics Using Compound Nucleus Separators*, edited by K.E. Gregorich, E.O. Lawrence Berkeley Laboratory, Berkeley, CA, USA, April 10-12, 1997, LBNL-40483 (1997)
14. J.P. Omtvedt et al., J. Nucl. Radiochem. Sci. **3**, 121 (2002)
15. Ch.E. Düllmann et al., Nucl. Instrum. Meth. A **551**, 528 (2005)
16. V. Ninov et al., *Proc. 2nd Int. Conf. on Exotic Nuclei and Atomic Masses*, edited by B.M. Sherril, D.J. Morrissey, C.N. Davids, ENAM98, Bellaire, MI, USA, 1998, AIP Conf. Proc. **455**, 704 (1998)
17. U.W. Kirbach et al., Nucl. Instrum. Meth. A **484**, 587 (2002)
18. M. Schädel et al., "The TASCA Project", GSI Scientific Report 2005, Gesellschaft für Schwerionenforschung mbH, Darmstadt, Germany, Report 2006-1, 2006, p. 262; see also `http://www.gsi.de/TASCA`
19. H. Haba, private communication
20. K. Morita et al., Nucl. Instrum. B **70**, 220 (1992)
21. Yu.Ts. Oganessian et al., Phys. Rev. C **69**, 054607 (2004)
22. Ch.E. Düllmann, Czech. J. Phys. **56** (Suppl. D), D333 (2006)
23. A. Semchenkov et al., "Envisaged TASCA Configuration", GSI Scientific Report 2004, Gesellschaft für Schwerionenforschung mbH, Darmstadt, Germany, Report 2005-1, 2005, p. 332
24. A. Semchenkov, private communication
25. K.E. Gregorich et al., Eur. Phys. J. A **18**, 633 (2003)
26. J.P. Omtvedt et al., Eur. Phys. J. D **45**, 91 (2007)
27. M. Leino, Nucl. Instrum. Meth. B **204**, 129 (2003)
28. A. Ghiorso et al., Nucl. Instrum. Meth. A **269**, 192 (1988)
29. K.E. Gregorich et al., Phys. Rev. C **72**, 014605 (2005)
30. K.E. Gregorich, private communication
31. H.-D. Betz, Rev. Mod. Phys. **44**, 465 (1972)
32. A.B. Wittkower et al., Phys. Rev. A **7**, 159 (1973)
33. K.L. Brown et al., "TRANSPORT/360 - a computer program for designing charged particle beam transport systems", Stanford Linear Accelerator Center, Stanford University, Stanford, CA 94305, USA, SLAC-91, 1970
34. U. Rohrer, "Computer programs for designing, investigating and commissioning charged particle beam lines", PSI Scientific and Technical Report 2000, Volume VI (Large Research Facilities), Paul Scherrer Institut, Villigen, Switzerland, 2001, p. 24
35. J.P. Omtvedt, private communication
36. H. Persson et al., Radiochim. Acta **48**, 177 (1989)
37. L. Stavsetra et al., Nucl. Instrum. Meth. A **551**, 323 (2005)
38. J.P. Omtvedt et al., J. Alloys Comp. **271–273**, 303 (1998)
39. L. Stavsetra et al., Nucl. Instrum. Meth. A **543**, 509 (2005)
40. R. Sudowe et al., Radiochim. Acta **94**, 123 (2006)
41. Ch.E. Düllmann et al., in *Advances in Nuclear and Radiochemistry*, edited by S.M. Qaim, H.H. Coenen (Forschungszentrum Jülich GmbH: Jülich, Germany, 2004), Vol. 3, p. 147
42. Ch.E. Düllmann et al., "Gas chemical investigation of hexafluoroacetylacetonates of Zr and Hf with preseparated isotopes", GSI Scientific Report 2005, Gesellschaft für Schwerionenforschung, Darmstadt, Germany, GSI Report 2006-1, 2006, p. 203
43. Ch.E. Düllmann et al., Radiochim. Acta (to be published)
44. S.C. Chattoraj et al., Inorg. Chem. **7**, 2501 (1968)