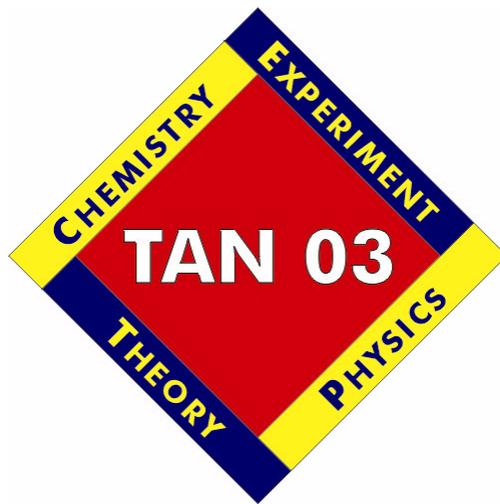


Methods and Future Developments



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Physical and chemical techniques to investigate the heaviest elements – Presence and future

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Early transactinide studies were much influenced by the assumption that isotopes of these elements decay predominantly by spontaneous-fission. As a consequence, rotating drums, wheels or tape systems were used to collect reaction products behind a target that were then transported in front of SF-detectors. Usually mica was applied for fission track counting. Also in the very first chemical investigation a frontal isothermal chromatography separation was coupled to a detection of SF-decaying nuclides by this technique. Clearly, these methods did not fulfil the stringent requirements of an unequivocal assignment of single-atoms to a given nuclide.

Later it turned out that not SF but α -decay is the dominant decay mode. Therefore, second generation experiments used event-by-event mode α detection systems, coupled to on-line physical separators in order to measure correlated decay chains. This approach immediately significantly reduced the random probability for an erroneous assignment of the decay of single atoms to a given nuclide.

Two physical separation techniques have been successfully applied in recent years: velocity filters (e.g. SHIP) and gas-filled magnetic separators (e.g. GNS). SHIP at GSI in Darmstadt was instrumental in the discovery experiments of the elements Bohrium through Darmstadtium, very likely also of the elements 111 and 112. Recently, experiments performed with the gas filled magnetic separator GNS at the FLNR in Dubna yielded evidence for the discovery of elements 113 through 116, possibly also of element 118. Both separation techniques are restricted to products from cold or “warm” fusion processes where evaporation residues have relatively high recoil energies. Current detection limits have reached the sub-pb level.

Chemistry studies of transactinides were much hampered by the technical challenge to couple fast and efficient on-line separations with high-resolution list-mode α -spectroscopy systems. In a first step, evaporation residues are collected in a thermalization chamber that are then continuously transported to a chemistry device. If the products are very volatile a pure gas (e.g. He) is used. For less volatile products solid aerosol particles (e.g. KCl) are added to the gas in order to serve as carrier particles for the adsorbed species (gas-jet technique).

So far, the following chemical techniques have been applied: for liquid phase investigations automated HPLC devices (e.g. ARCA) and continuous extraction systems (e.g. SISAK), and in the gas phase adsorption-chromatographic separators (e.g. OLGA). After chemical isolation, products are usually assayed for α or SF decay with conventional semiconductor detectors. In the SISAK device reaction products in an organic liquid are analysed in flow-through detector systems by liquid scintillation counting applying pulse-shape analysis. This method, however, is known to have a poor energy resolution. SISAK was therefore successful so far only if coupled to a physical separator (BGS at LBL). Promising are recent attempts to perform studies on chemically reactive surfaces (IVO/COLD for element 112, CALLISTO for hassium). As yet, chemistry studies have been applied for transactinides up to hassium and very recently also for element 112. All these investigations are possible for nuclides with half-lives of a few seconds or longer.

Future success of transactinide research will much depend on, first, improvements in beam intensities (including duty cycles), second, novel target technologies, urgently needed to accumulate higher particles

doses in reasonable time, third, devices that are able to assign accurate mass numbers and/or atomic numbers to separated evaporation residues at a single atom level and, fourth, to next generation set-ups, able to perform physical and chemical studies with single atoms (ions) or molecules.

Several projects focus on these goals. Some examples are: at GSI a new high-intensity heavy ion superconducting CW-LINAC is evaluated and new chemical compounds are tested as future targets instead of metallic targets. At FLNR an on-line high-resolution mass separator (MASHA) is being built. At GSI the SHIPTRAP project focuses on the development of ion traps behind the velocity filter for detailed studies on single ions. PSI effort is currently devoted to the development of an ISOL target device that might be coupled to a physical separator (e.g. BGS). Such a system would enable novel investigations (e.g. of organo-metallic compounds).

Ion source developments for heavy element research

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The low cross section experiments for the synthesis of new isotopes and elements in the transactinide and superheavy element region demand intense heavy ion beams (10^{13} particle/sec and higher on target). The duration of these experiments is long (> 200 hours), and frequently accelerated beams of neutron rich isotopes are required. These requirements present a challenge to accelerator injector systems. Therefore, a concentrated ion beam development program is necessary to be able to produce sufficient beam intensities for heavy element experiments.

In addition, high production efficiencies for those high intensity heavy ion beams are desired for various reasons: First, rare isotopes like ^{48}Ca , ^{36}S , or ^{85}Rb are very expensive (for example 250000\$/g ^{48}Ca). Secondly, it is unpractical to reload the oven during long experiments for beams made from solid material. Third, it is desirable to minimize the contamination of the ion source for consecutive runs.

The production of heavy ion beams from solids and gases will be reviewed briefly. Electron cyclotron resonant ion source (ECRIS) development required for heavy element research is described in more detail. The development of an intense ^{48}Ca ion beam at LBNL is described as an example for methods to optimize ion source ionization efficiency.

Recoil separator capabilities for heavy element research and an opportunity for second generation chemistry studies

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Compound nucleus recoil separators have become the tool of choice in the discovery and study of nuclear properties of transactinide isotopes. An overview of recoil separator principles, capabilities and limitations are presented. The recoil separators currently active in the study of heavy element isotopes are presented. An attempt is made to compare the efficiency, selectivity, sensitivity, and resolution of present-day separators for heavy element studies.

The factors limiting efficiency of recoil separators are dominated by energy loss and scattering of compound nucleus evaporation residues in the target material, together with the angular and velocity acceptance of the separators. Selectivity and sensitivity are closely related, with selectivity most dependent on the separator's ability to suppress unwanted nuclear reaction products, and sensitivity strongly dependent on the detection techniques used. Resolution refers to the ability of the separator/detector system to give information on the Z , A , and/or energy of the heavy element isotopes. The small cross sections and resulting low production rates for heavy element isotopes present the most fundamental limitation in transactinide science. Thus, the available integrated luminosity available to an accelerator-separator combination is as important as the capabilities of the separator itself. All of these factors are discussed for the existing separators, and options for future separator programs are presented.

The Berkeley Gas-filled Separator has been coupled with a gas-jet device to provide pre-separated isotopes for transactinide chemical studies. This technique is described, and the new capabilities it provides are discussed. An international effort to design and build a new separator specifically for transactinide chemical studies, "ChemSep," has begun.

Heavy element research with laser spectroscopy and traps

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Recoil separators like the SHIP facility at GSI in Darmstadt open a great variety of new research opportunities in the heavy element research if combined with traps in which the fusion products can be stored.

The simplest conceivable trap is a buffer gas cell in which the fusion products come to rest either as atoms or singly charged ions. The capture time of the neutral atoms is determined by the diffusion process to the walls. This time of typically 50 ms is long enough to investigate the unknown atomic level schemes or determine the ionization potential of heavy elements by laser spectroscopy employing pulsed lasers with repetition rates of 20 Hz or more. Very powerful methods have been developed like the resonance ionisation spectroscopy with detection of the ionization process either by radioactive decay [1] or directly after mass analysis [2]. The combination of experimental search for atomic levels in the heavy actinide and transactinide region with theoretical level predictions based on Multi-Configuration-Dirac-Fock calculations may provide a critical test of such ab-initio calculations of electron configurations. Once excited atomic states are known, the hyperfine structure of suitable transitions can be studied as well enabling stringent tests of nuclear models applied to the heaviest nuclei.

The fraction of singly charged ions can be used to study gas phase ion chemical reactions with admixtures like O₂, H₂O, CH₄ ... to the buffer gas in the stopping chamber. A measurement of the drift time of the ions or ionic compounds in the buffer gas chamber enables a determination of the ionic mobility and may open up new avenues in studies of relativistic effects on ionic radii and the bond length of simple molecular ions [3]. It is conceivable that such investigations can be extended to transactinides with lifetimes larger than 10 ms produced with cross-sections greater than about 100 pb.

A high-resolution Penning trap mass spectrometer as proposed for the SHIPTRAP facility [4] allows direct mass spectrometry of heavy actinide and transactinide isotopes. Traps with small amounts of reactive gases may also provide access to kinetic studies by investigating the loss rate of stored heavy ions and the formation of ion chemical compounds.

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Status and future developments in the aqueous heavy element chemistry

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Recently, the status of the liquid-phase chemistry of the transactinide elements has been reviewed extensively in a number of reviews and book contributions [1-4]. In Table 1, an attempt is made to summarize the chemical systems studied with elements 104, 105, and 106, so far, and the pertinent references are given. The chemical techniques involved are briefly outlined. These include manual techniques, the multi-column technique (MCT), the automated rapid chromatography apparatus (ARCA), the on-line liquid-liquid extraction system SISAK in conjunction with liquid scintillation counting, and the automated ion-exchange separation apparatus coupled with the detection system for α -spectroscopy (AIDA). Critical comments [1] have been addressed to the results and conclusions presented in Refs. [8-12], and [14].

Technical developments are going on in several directions: The MCT has been coupled to the aerosol collection device ALOHA on which 2-s collection and dissolution cycles are performed so that the dissolved activity can be fed rapidly onto the sequence of chromatography columns in a quasi-continuous mode. The optimized conditions for on-line experiments with seaborgium are discussed in which Kd values on an anion exchange resin in 0.1 M HNO₃/5x10⁻³ M HF are planned to be measured via the long lived descendant of ²⁶⁵Sg, 20-d ²⁵³Es. It is also planned to reduce seaborgium to a lower oxidation state by contacting its aqueous solution with a hot aluminum surface and by distinguishing anionic seaborgium(VI) from cationic seaborgium (III) on a cation exchange column using the MCT.

Continuous efforts have been devoted to improvements of the on-line liquid-liquid extraction system SISAK coupled with liquid scintillation counting (LSC). Analog pulse-shape discrimination (PSD) has been used to reduce the β background in the α spectra by a factor of >1000. Without this technique, the β - and γ -induced background from activities produced in the target will interfere with the α spectrum to such an extent that it becomes useless. However, in the past, when feeding the He/KCl jet directly into the SISAK system, the PSD alone was insufficient to unequivocally identify transactinides with the SISAK - LSC system. Omtvedt et al. [17] have overcome this limitation by producing ²⁵⁷Rf in the ²⁰⁸Pb(⁵⁰Ti,1n) reaction, separating it in the Berkeley Gas-filled Separator (BGS), and transferring it to the gas jet using the Recoil Transfer Chamber (RTC). This was the first time that a transactinide was extracted and unequivocally identified by the SISAK - LSC system and also the first experiment to use pre-separation in a transactinide chemistry investigation. Another improvement comes from the digital recording of the LSC pulses with a fast transient recorder and the pulse-shape analysis by an artificial neural network (PSD - NN) [29]. This has been proven to eliminate pileups to the extent that identification of transactinides even without pre-separation becomes feasible. Its ability to recognize α - γ - and α -conversion-electron-coincidence summing which is known to cause shifted α energies may be used in the future to do nuclear spectroscopy [29].

Table 1.

Element	Isotope	Chemical technique	Chemical system studied	Ref.
104, rutherfordium	^{261m} Rf	manual	Cation exchange/ α -hydroxyisobutyrate	5
	^{261m} Rf	manual	Aliquat 336/12 M HCl, 6 M HCl	6
	^{261m} Rf	manual	Tri-isooctyl amine/12 M HCl	7
	^{261m} Rf	manual	Thenoyltrifluoroacetone/0.05 - 0.24 M HCl	8
	^{261m} Rf	manual	Tributyl phosphate/8 M - 12 M HCl	9
	^{261m} Rf	manual	Tributyl phosphate/9 M - 12 M HBr	10
	^{261m} Rf	manual	Co-ferrocyanide/<6 M HCl	11
	^{261m} Rf	manual	Tri-isooctyl amine/0.4 M HF	12
	^{261m} Rf	MCT	Anion exchange/0.2 M HF	13
	^{261m} Rf	MCT	Anion exchange/0.27 M HF, 0.2 or 0.1 M HNO ₃	14
	^{261m} Rf	ARCA	Tributyl phosphate/6 M HCl	15
	^{261m} Rf	ARCA	Cation exchange/0.1 M HNO ₃ , var. HF Anion exchange/0.1 M HNO ₃ , var. HF	16
	²⁵⁷ Rf	SISAK	Dibutyl phosphoric acid/6 M HNO ₃	17
	^{261m} Rf	AIDA	Anion exchange/8 M HNO ₃ /4 M - 11.5 M HCl	18
	²⁶³ Rf	manual	Cation exchange/ α -hydroxyisobutyrate and Tributylphosphate/9 M HCl	19
105, dubnium	²⁶² Db	manual	Glass surfaces/fuming with HNO ₃	20
	²⁶² Db	ARCA	Tri-isooctyl amine/12 M HCl, 0.02 M HF/10 M HCl, 0.025 M HF/4 M HCl, 0.02 M HF/0.5 M HCl, 0.015 M HF	21, 22
	²⁶² Db	ARCA	Diisobutyl carbinol/conc. HBr/6 M HCl, 0.0002 M HF/0.5 M HCl	23
	²⁶² Db, ²⁶³ Db	ARCA	Cation exchange/ α -hydroxyisobutyrate	24, 25
	²⁶² Db	ARCA	Aliquat 336/10 M HCl/6 M HCl/6 M HNO ₃ , 0.015 M HF Aliquat 336/ 0.5 M HF/4 M HF/6 M HNO ₃ , 0.015 M HF	26
106, seaborgium	²⁶⁵ Sg	ARCA	Cation exchange/ 0.1 M HNO ₃ , 5x10 ⁻⁴ M HF	27
	²⁶⁵ Sg	ARCA	Cation exchange/0.1 M HNO ₃	28

A new development in the field of liquid-liquid extraction is MicroSISAK based on the use of microsystem components [30]. Such a system provides separation times on the order of seconds at considerably reduced flow rates as compared to the conventional SISAK system. For mixing of the aqueous phase with the organic phase, a micro mixer is used where the phases are conducted through 2 mm long and 50 μ m broad channels to a common outlet. The so-produced emulsion is subsequently separated in a filter apparatus on a filter membrane of 0.5 μ m pore size. At a pressure differential of 10 mbar across the membrane and a flow rate of 0.5 ml min⁻¹, the aqueous phase is completely retained by

the hydrophobious membrane while the organic phase penetrates the filter. The goal is to eventually spray the organic phase onto a moving tape where it is evaporated to dryness and assayed for a activity by silicon detectors thus avoiding the drawbacks of LSC. In collaboration with the Institut für Mikrotechnik, Mainz, IMM, extraction yields are being optimized for flow rates below 0.1 ml min^{-1} [31].

For a future application to the spherical superheavy elements 112 and 114, the electrodeposition of these elements on certain electrode materials has been proposed [32]. It is well known that the electrochemical deposition of radionuclides in metallic form from solutions of extremely small concentrations depends strongly on the choice of the electrode material. The associated "underpotential" can deviate considerably from the Nernst potential. In a macroscopic model [32], the interaction between the microcomponent and the electrode material has been described by the partial molar adsorption enthalpy and -entropy. By combination with the thermodynamic description of the electrode process, a potential has been calculated that characterizes the process at 50% deposition. Model calculations for Ni-, Cu-, Pd-, Ag-, Pt-, and Au-electrodes and the microcomponents Hg, Tl, Pb, Bi, and Po have confirmed the decisive influence of the electrode material on the deposition potential. The experimental determination of "underpotentials" for some of these elements [33] is in qualitative agreements with the model predictions [32]. Plans for the application of this electrochemical technique to the spherical superheavy elements are presented.

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The status and future of heavy element research at Dubna

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Relatively long half-lives of isotopes of elements 105-116 produced in reactions between ^{48}Ca and $^{248,245}\text{Cm}$, $^{244,242}\text{Pu}$, ^{243}Am and ^{238}U and chemical properties of the SHE predicted theoretically provide possibilities of new experiments devoted to the chemical identification of SHE, study of their chemical properties, combination of chemical and physical methods for the SHE synthesis and search for SHE in Nature.

The FLNR programme of the heavy element research for 2004-05 includes the following main experiments:

- Chemistry of elements 112 and 114: the $^{244}\text{Pu}(^{48}\text{Ca},xn)$ reaction will be used; the adsorption of E112 and E114 (if E114 will have an enhanced volatility) on the Au surface will be determined in a temperature interval 25°C to -200°C.
- Chemical separation of recoil nuclei produced in the $^{243}\text{Am}(^{48}\text{Ca},xn)$ reaction will be carried out for the ^{268}Db identification.
- Test experiments at the new FLNR separator MASHA (Mass Analyzer of Super Heavy Atoms) will be completed. First experiments on the determination of masses of isotopes of 112 and 114 elements will be carried out.
- Preparation of an experiment on the search for SHE ($Z=108$) in nature (study of Os-samples with a low-background neutron multi-detector).

Development of an on-line hot catcher for volatile species

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New isotopes of elements 112 and 114 have been produced in ⁴⁸Ca beam induced fusion reactions with ²³⁸U and ²⁴², ²⁴⁴Pu targets [1]. Their relatively long half-lives give chemists the possibility to investigate the chemical properties of these super heavy elements. The gas-phase chemical investigation of transactinides has gained remarkable progress during the last decade. Continuously operating gas-phase separations, e.g., some devices based on the On-Line Gas chromatographic Apparatus (OLGA), were instrumental to study the formation of halide, oxohalide and hydroxide compounds of transactinides from Rf through Bh. The In-situ Volatilization and On-line detection apparatus (IVO) was successfully applied to study the volatility of the tetroxide of HsO₄ [2]. For the heaviest element, calculations of the electron configurations predicted increasingly strong relativistic effects that may strongly influence their chemical behavior. Hence, the elements 112 and 114 are predicted to be chemically inert and volatile elements, similar to noble gases [3]. On the other hand, classical extrapolations along the groups of the periodic table predict elements 112 and 114 to behave similar to their homologues in the groups 12(Zn, Cd, Hg) and 14(Ge, Sn, Pb), respectively [4]. The expected *high volatility in elemental states* would allow us the chemical investigation of these elements with gas-phase chemical methods or even better at vacuum conditions. The differentiation between either a metallic or a noble-gas character of the heaviest elements could be achieved by vacuum thermochromatography, that provide very clean surfaces in the adsorption process. It also supplies crucial advantages for the experiment with short-lived isotopes: 1) fast separation under molecular flow condition; 2) no carrier gas; 3) a very stable temperature gradient due to the good heat isolation in vacuum; 4) vacuum chemical apparatus could be friendly coupled with a physical separator.

In order to transfer the fusion-reaction products from the target position to the chemical apparatus in vacuum, an on-line hot catcher system coupled to a vacuum thermochromatographic apparatus is now being developed at Paul Scherrer Institute, Switzerland.

The working principle is as follows: Products from heavy ion induced fusion reactions have a recoil momentum sufficiently high and forward-peaked in order to penetrate the target material ($\approx 1\text{mg}/\text{cm}^2$). Passing a thin carbon window ($\approx 30\ \mu\text{g}/\text{cm}^2$), they are implanted into a solid catcher, which is heated up to 2000K. Due to thermo-diffusion, the implanted atoms drift towards the surface of the catcher and desorb from the catcher surface. Subsequently, they effuse inside of the surrounding crucible until they pass the inlet to the thermochromatographic device.

The yields and velocities of the described processes are closely related to the material constants of the solid catcher and of the crucible material, such as diffusion coefficients and adsorption enthalpies. Hence, in order to find the proper catcher material to release volatile metallic p-elements rapidly with high efficiency and also to investigate their adsorption behavior on different metal surfaces by means of vacuum thermochromatography, off-line experiments based on the long-lived isotopes of Tl, Pb, and Bi were performed. An off-line model set-up for "hot catcher" test experiments (see Fig.1) was developed. Isotopes of the p-elements Bi, Pb, Tl were produced by bombarding a ^{nat}W(9 μm) target foil with a ²⁰Ne beam (110 MeV on the target) at the PSI PHILIPS Cyclotron. The recoiling nuclei of the nuclear reactions with the W target were implanted in a He-atmosphere into various catcher foils just behind the target. Ta,

Ti, Pt, Rh, and C served as catcher materials in these experiments. Isotopes of other elements (Ag, As, Se, Te, Y, Ru, Nb, Mo, Ga) were produced due to the interaction of the beam particles with the catcher material. After the long term irradiation (~ 6 h), the catcher foil was placed into the Ta crucible, which was afterwards closed with a Ta-window. The tantalum crucible with an inner diameter of 1.8 cm and a length of 5 cm (model of a future recoil chamber) has a 2 mm hole, which was attached to the inlet of the chromatographic column by a Ta-connector. For the thermochromatographic separations a pure quartz tube or a quartz tube, which was covered inside by thin Au, Ag, Cu, Ni, Pt, and Pd metal foils (see Fig. 1) were used. The quartz tube was then applied to a copper tube. The whole set-up was evacuated by a turbo molecular pump to a pressure of about 5×10^{-5} mbar. An induction heating device was used to heat up the Ta-crucible to about 1800K. This temperature was measured applying an IR thermometer. Along the chromatography column a temperature gradient between 750°C and -170°C was forced by the gradient oven at the starting point and by the cold finger cooled with liquid nitrogen at the end.

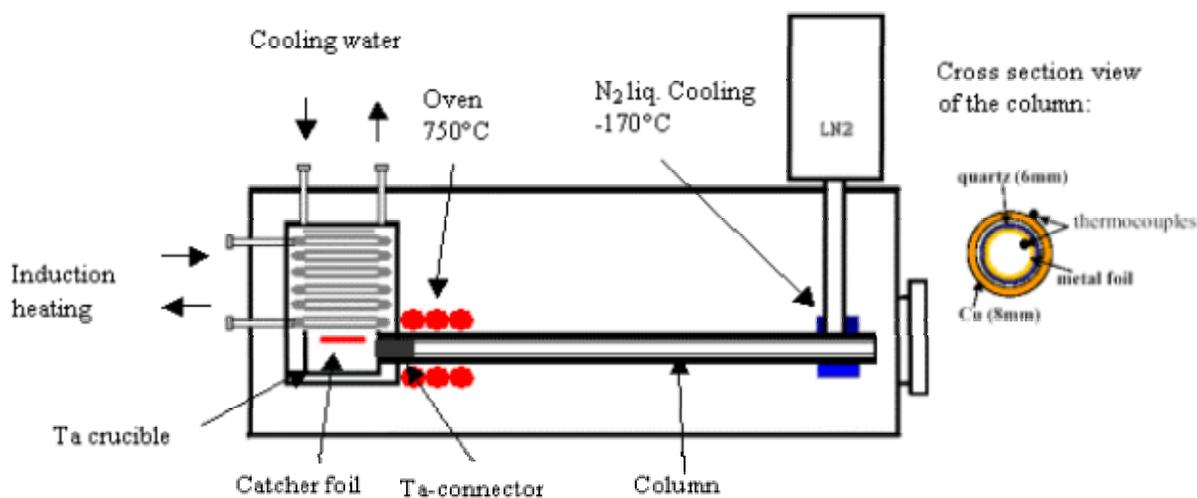


Figure 1. Schematic view of the set-up used for off-line release studies and vacuum thermochromatographic experiments.

Each experiment lasted about 30 min. Release efficiencies for volatile elements from the particular catcher material were determined by γ -activity measurement of the given catcher sample before and after heating. The distribution of the elements in the catcher system and their distribution along the chromatography column were determined with a HPGe- γ - detector with a 1 cm lateral resolution applying a lead collimator.

The determined release efficiencies of some volatile species from different catcher materials are compiled in Table 1. Generally, a higher release efficiency may be expected if the working temperature is close to the melting point of the catcher material. We observed a nearly complete release of Pb, Tl, and Bi from the Pt and Rh catcher at a working temperature of 1800K, whereas these elements partly remained in the catcher made of Ta, Ti, and graphite. Despite a He flushing during the irradiation the Ta and Ti catcher were always oxidized in the beam spot. For future applications, Rh was determined to be the best-suited catcher material, since in nuclear reactions of the heavy ion beams with the Pt catcher large amounts of interfering activities (e.g. $^{211,212}\text{Po}$) are produced, which disturb the identification of transactinides. All of the released volatile species could be observed deposited in the quartz or metal chromatographic tube. Se was found in the crucible and in the connector, most probably because of the formation of very stable and not volatile Ta-Se compounds. The measured release efficiencies are well

correlated with the release enthalpies ΔH_r^M calculated from the sum and the formation enthalpy of solid solution based on the Miedema model [5] and of the desublimation enthalpy (see Tab. 1). These calculations seem to be very useful for a prediction of release efficiencies for unknown catcher-product combinations. The low release efficiencies for the Ta catcher originated probably from the very low diffusion velocities in a metal with an exceptionally high melting point or from the formation of a Ta₂O₅ layer on the surface of the catcher foil during the irradiation, which may remarkably disturb an efficient release.

Table 1. Release efficiency for some volatile species from different catcher materials.

% { ΔH_r^M , kJ/mol}	Catcher/ thickness (μm)	Pt/ 5	Rh/25	Ta/5	Ti/6	C/200	Fe/25
Element	Melting point (K)	2045	2239	3269	1941		1808
Pb		100{281}	88{228}	65{188}	81{298}	70{332}	100{100}
Tl		100{267}	100{211}	58{125}	86{224}	100{324}	{75}
Bi		100{319}	{266}	{237}	{352}	{368}	{138}
Ag		94{288}	30{247}	{225}	62{291}	{348}	{162}
Y		38{763}	67{675}	75{296}	76{350}	67{973}	{429}
Ru		56{661}	{652}	{808}	100{825}	{718}	{677}
Nb		10{986}	9{910}	12{722}	51{714}	41{1168}	{793}
Mo		0{764}	0{717}	0{676}	49{671}	16{900}	{665}
Te			39				
Se							96
As		{493}	{470}	{557}	{622}	{527}	90{433}
Ga		{465}	{421}	{361}	{418}	{531}	87{327}

Adsorption properties of Tl, Pb, Bi, and other volatile species on the metals Au, Ag, Cu, Pt, Pd, and Ni as well as on quartz were determined from the results of the vacuum thermochromatography (see Tab. 2). In order to obtain clean metal surfaces, each metal column was pre-treated with the mixture of He and H₂ gas flow at 850°C and then with a pure He gas at the same temperature just before the experiment. The deposition temperatures were determined from the peak maximum of element distribution along the tube. From the experimental parameters the adsorption enthalpies ($-\Delta H_{\text{ads}}^{\text{TD}}$) have been calculated using the thermodynamic model of vacuum thermochromatography from [6,7]. A Monte Carlo simulation based on kinetic model of mobile adsorption [8] was also used to evaluate the adsorption enthalpies ($-\Delta H_{\text{ads}}^{\text{kin}}$) from the experimental results.

Using different catcher materials several determinations of the adsorption enthalpies of Pb and Tl on quartz surfaces were performed. Reasonable reproducibility was found. The determined adsorption data are consistent with data from earlier vacuum thermochromatography experiments ($-\Delta H_{\text{ads}}^{\text{Lit}}$) [7]. In the Pd and Ag column, Pb and Tl were deposited at the starting point. Therefore, only an upper limit of the adsorption enthalpy is given. For the other metal columns, the adsorption enthalpies determined from the present experiment are well comparable with data obtained by gas thermochromatography ($-\Delta H_{\text{ads}}^{\text{Lit}}$) [9].

The developed coupling principle between a catcher system and vacuum thermochromatography was shown to work properly. The chemical inertness, the high release efficiencies at moderate temperatures and the absence of beam induced interfering α -decaying by-products make Rh the most attractive catcher material for future experiments with transactinides. Moreover, extremely stable compounds of Rh with actinide elements are known. Hence, the metal chemistry inside the catcher

material will be valuable for a chemical pre-separation already in the recoil chamber. Such separation effects can be predicted using calculated data of the release enthalpies ΔH^M . However, only on-line studies can provide information about the very important kinetics of the release. Therefore, on-line hot-catcher experiments with short-lived isotopes of Hg, Pb, Tl, and Bi using a similar experimental set-up are planned.

Table 2. Deposition temperatures and the evaluated adsorption enthalpies together with values given in literature [7,9]

Catcher / surface	Element	Deposition temperature (K)	$-\Delta H_{\text{ads}}^{\text{TD}}$ (kJ/mol)	$-\Delta H_{\text{ads}}^{\text{kin}}$ (kJ/mol)	$-\Delta S_{\text{a, mob}}$ (J/mol*K)	$-\Delta H_{\text{ads}}^{\text{Lit}}$ (kJ/mol)
Rh-02/quartz	Pb	550±25	146±7	154	166	142
Ta-06/quartz	Pb	541±25	142±7		166	
Ta-04/quartz	Pb	589±25	156±7		166	
Ti-02/quartz	Pb	589±25	155±7		166	
Rh_02/quartz	Tl	370±25	98±6	106	167	113
Ta-06/quartz	Tl	465±25	122±6		167	
Ta-04/quartz	Tl	413±25	109±6		167	
Ti-02quartz	Tl	475±25	125±6		167	
Rh_02/quartz	Bi	360±25	96±6	102	168	109
Rh_02/quartz	Ag	648±25	172±8	187	163	
Rh_02/quartz	Se	>1023±25	<271±10	<295	159	
Rh_02/quartz	Te	389±25	103±6	112	165	151
Pt/Au	Pb	931±25	247±12	260	163	229
Pt/Au	Tl	871±25	231±12	240	163	205
Pt/Ag	Pb	>961±25	<254±12	<250	164	
Pt/Ag	Tl	>961±25	<254±12	<250	164	
Pt/Pt	Pb	>1023±25	<271±12	<266	165	
Pt/Pt	Tl	871±25	231±12	226	165	
Pt/Pd	Pb	>1023±25	<271±12	<300	165	326
Pt/Pd	Tl	>1023±25	<271±12	<300	165	326
Pt/Cu	Pb	783±25	207±12	225	168	222
Pt/Cu	Tl	812±25	215±12	235	167	119
Pt/Ni	Pb	931±25	246±12	262	168	227;231; 243;272
Pt/Ni	Tl	389±25	103±6	115	172	117;149
Pt/Ni	Bi	>1023±25	<271±12	<300	168	252; 322; 328; 273
Pt/Ti	Pb	976±25	258±12	264	164	
Pt/Ti	Tl	948±25	250±12	251	164	
Pt/Ti	Zn	842±25	224±12	215	160	

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Application of artificial neural networks in LSC for transactinide research

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Introduction

In Liquid Scintillation Counting (LSC), α - or fission events can be distinguished from β - and γ - events by characteristic differences in their light curves. The reason is a predominant population of triplet states of the scintillator by strong ionizing particles. These triplet states are metastable which leads to a tailing in the light curve (Fig. 1a).

In a common experimental setup for α -LSC, analog pulse shape discrimination (PSD) is used to separate the β/γ - part of the spectrum. However, β/γ - pile up events in a time window of about 150 ns - 250 ns (Fig.1b) can simulate a heavier particle event in the PSD circuit. This leads to a pseudo α -background in the spectrum. Pile ups are occurring randomly at high count rates but can also be produced by decay cascades of a nuclide.

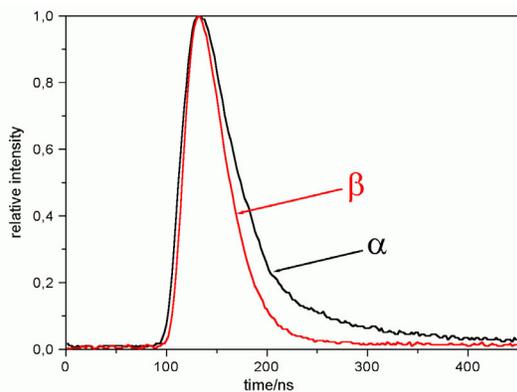


Figure 1a.

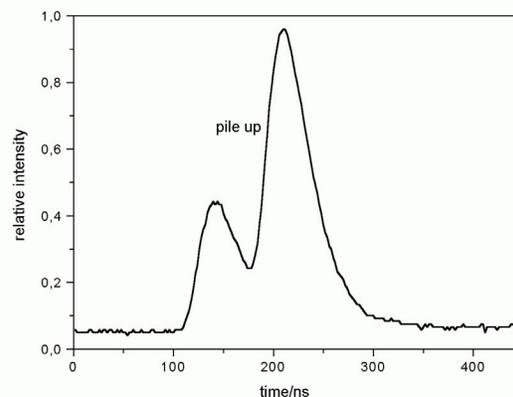


Figure 1b.

In transactinide research with the fast liquid-liquid extraction system SISAK-3 [1], the transactinide element under investigation is extracted into an organic phase containing a liquid scintillator. In these experiments, very few α - decays have to be identified at a high level of β/γ - events. Therefore, any pile up background makes the unambiguous identification of the transactinide nuclides difficult or impossible. In order to apply LSC also in those experiments, the analog PSD has been coupled with a digital pulse recorder. The digitally recorded pulses are off line analysed by an artificial neural network.

Electronics

The setup used is shown schematically in Fig. 2. The photomultiplier (PM), the charge sensitive preamplifier (CSPA), the PSD module, and the time to amplitude converter (TAC), are the standard modules for common LSC. From the built in single channel analyser of the TAC, a trigger signal for the digital pulse recorder is derived. The CSPA integrates the signal. Therefore, it has to be differentiated by the timing filter amplifier (TFA) to get the original shape of the signal. As digital pulse recorder, the PC module acqiris DP110 is used. This module allows a maximum sampling rate of 1 GS/s at a bandwidth of 500 MHz. Besides the pulse also the time of the corresponding trigger event is recorded which allows the search for correlated events.

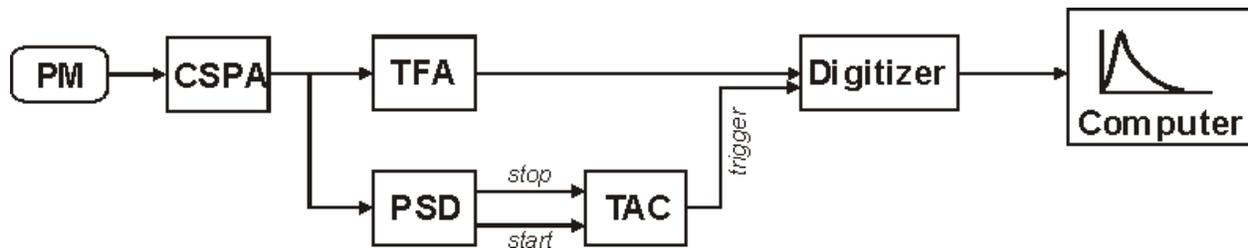


Figure 2.

Artificial neural network

Artificial neural networks are successfully used for pattern recognition especially if an analytical description of the characteristics of the patterns is difficult or impossible. The artificial model of a neuron is rather simple. Each input to a neuron is multiplied by a weighting factor. The weights are corresponding to the synaptic strength in a natural neural network. The output is some function, the so called transfer function, of the weighted sum of all inputs. The weights are initialized by some random values. Learning of the network means adjusting the weighting factors.

For the digital pulse shape discrimination with neural networks (PSD-NN), a 3 layer feed forward network was modeled with the program SMART [2]. The input layer has 175 neurons corresponding to 175 samples at every 2 ns of a pulse. The second (hidden) layer has 5 neurons, and the output layer has two neurons corresponding to an alpha event and any other type of event, respectively. Each output of a layer is connected to each neuron of the next higher layer. The sigmoid function is used as transfer function which gives output values between 0 and 1. The weights are adjusted during the training phase with the backpropagation algorithm [3]. In principle, the weights are corrected in proportion to the error of the network, i.e., the difference between the current and correct output for a given training pattern. This error is propagated backwards from the output layer to the inner layers in order to determine correction values for all weights of all layers. Training patterns for α -events were recorded from a ^{219}Rn / ^{215}Po source. Training data for β -, γ -, and pile up events were recorded mostly from neutron irradiated dibutylphosphate (DBP) solved in toluene. All pulses are normalized to a fixed amplitude thus avoiding that the network learns an energy calibration. After each training cycle the network was tested with test data recorded from the same sources. Fig. 3 shows the error of the network with proceeding training. The error decreases very rapidly and is almost constant after 600 iterations.

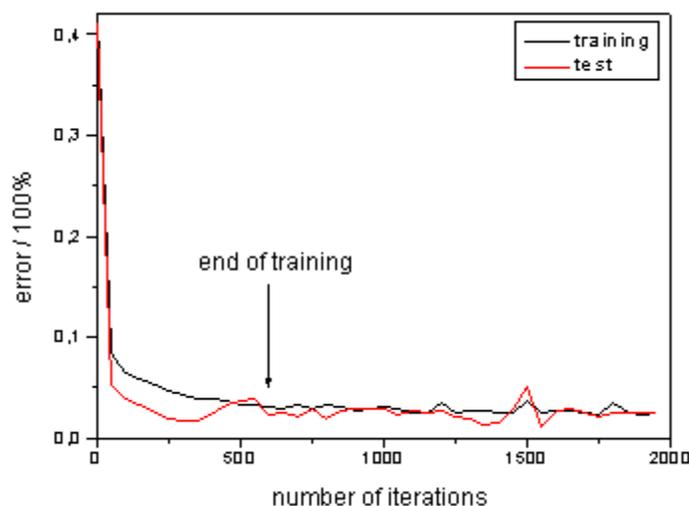


Figure 3.

Test experiment and results

The PSD-NN system was first tested in a SISAK experiment at the Paul Scherrer Institute in February 2000. In this experiment, rutherfordium was produced in the reaction $^{248}\text{Cm}(^{18}\text{O},5n)^{261}\text{Rf}$ ($E_{\alpha}=8.8$ MeV, $T_{1/2}=78$ s). Fig. 4 shows the setup schematically.

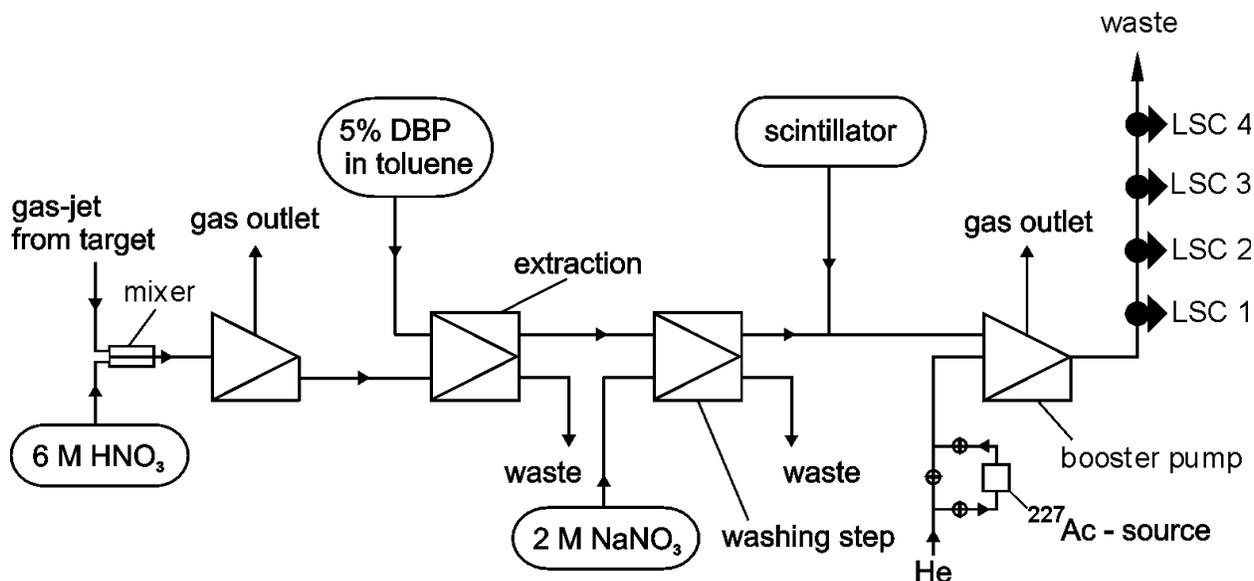


Figure 4.

The reaction products transported with a gas - jet are dissolved in 6M HNO_3 . Group IV elements are then extracted with 5% DBP in toluene. In a washing step with 2 M NaNO_3 the remaining nitric acid is removed. Then, the scintillator consisting of dimethylPOPOP (3g/l) and methylnaphtalene (30vol%) dissolved in toluene is added. In this experiment, 4 detector cells of 5.5 ml each connected in series are used with the analog PSD electronics. The first detector was also connected to the digital pulse recorder.

The power of the artificial neural network is demonstrated in Fig. 5. Fig 5a shows a typical “ α -spectrum” in the energy range of interest from the analog PSD. In Fig. 5b the same spectrum is shown after treatment with the neural network. In the total spectrum 95.8% of the events could be identified as pile up events. These results show that low level α -LSC can be significantly improved by the application of digital pulse recording in combination with artificial neural networks.

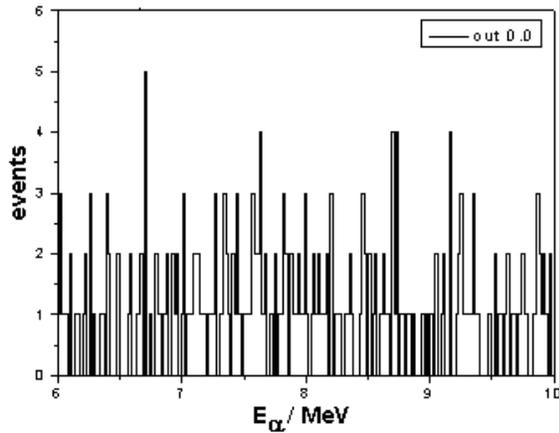


Figure 5a.

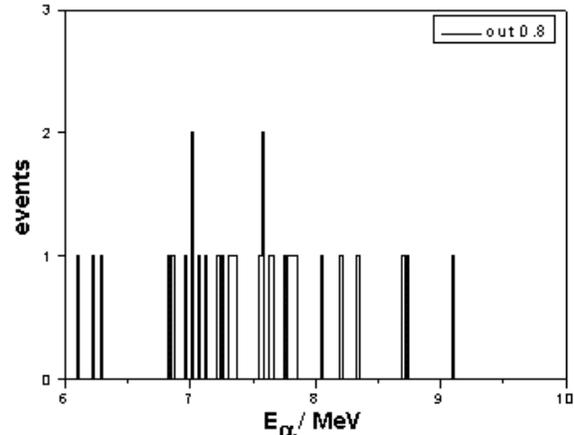


Figure 5b.

A new application of LSC with the digital recording of pulses is the determination of spectroscopic data. Pile ups are often associated with coincident nuclear transitions. If these transitions occur in the nanosecond regime, a pile up can be resolved such that the two radiations can be separated. This enables one to determine lifetimes of excited states. An example is given in Fig. 6 for a measurement of ^{241}Am . The α -decay of ^{241}Am populates the $5/2^-$ level in ^{237}Np at 59.5 keV having a lifetime of 67 ns. This correlated α - γ decay produces pile ups in the scintillator. Fig. 6a gives an example. From the distance between the two peaks, a time difference Δt can be determined. The number of pile ups of a given Δt plotted vs. Δt is shown in Fig. 6b. From the fit of an exponential decay law to these data, the lifetime of the excited state can be determined to 67 ns in good agreement with published data. Thus, this application is promising as it allows the determination of hitherto unknown decay properties of short-lived nuclei.

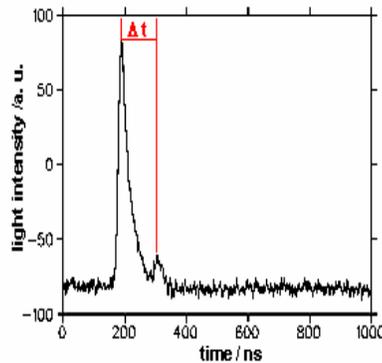


Figure 6a.

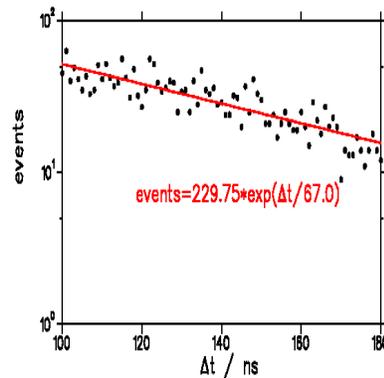


Figure 6b.

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Perspectives of the some computer methods using for the transactinide experimental and theory data treatment

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Spontaneous radioactive decay of nuclear belongs to that category of the natural phenomena which reasons, by virtue of complexity of objects, have not received a strict scientific explanation yet. The advanced interest to this phenomena, as having major significance in human life, results, on the one hand, in accumulation of the comprehensive experimental material and, with another hand, in the plenty of theories about its physical nature. On the basis of theories the different mathematical models are created for experimental data processing and the nuclear behavior predicting. Computer modelling is modern and quickly developing method for the complicated physical and chemical processes studying, both for the treatment of the experimental data bulk, and for the theoretical researches. Therefore it seems to be useful to apply some computer methods to the radioactive decay processes investigating.

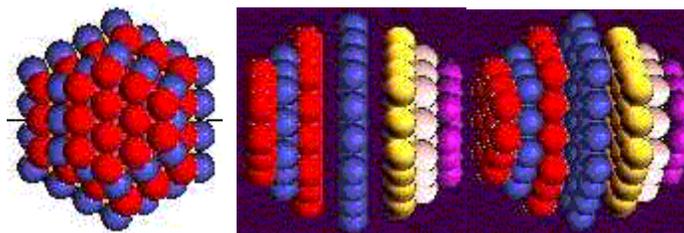
Spontaneous radioactive decay such as α -decay and spontaneous fission are the properties intrinsic in main to heavy nucleus: actinides and transactinides. One of the problem in modern nuclear physics and radiochemistry is to establish dependence between nuclear composition and half-life of radioactive isotopes and to predict stability of still undiscovered nucleus. Theoretical calculations of nucleus stability are based on the two main physical models: 'liquid -drop' and shell model [1]. The essence of these models is reduced to consideration of nuclear binding energy and calculations for α -particles or fragment particles after spontaneous fission of the potential barrier penetration probability that radioactive decay constant is depended on.

Properly process of nuclear disintegration is described by "liquid-drop" model. On analogy to a drop of a liquid any small distortion of the nuclear sphere will be associated with the surface tension and Coulomb energies increasing so the total nuclear binding decreasing. The main conclusion of this model was that fissionability of nucleus may be expected to correlate with parameter Z^2/A . By several semiempirical energy equations this parameter was calculated as 45-49. Thus all nuclei with $Z^2/A > 49$ are expected to undergo fission instantaneously and limit is set to the synthesis of transuranium elements at $Z \approx 110-120$. Though the liquid-drop model explains many features of the nuclear disintegration process some of the experimentally facts could not be interpreted in this framework, especially the "magic numbers" of protons and neutrons at which nucleus are most stable and existence of heavy and superheavy elements. Dependence between number of the nucleons in nucleus and its stability is well described by the shell model, where shell structure in nuclei is proposed as analogous to the electron structure in atoms. The quantum calculations of energy levels showed that the filled shells have numbers of neutrons or protons equal to "magic numbers" and that nuclei are more stable which compositions are closer to the filled shells. Influence of the "shell effects" is important for all nuclei, however for the heaviest nuclei the most essentially. The introduction of the shell term in the formula for energy of a nucleus [2] has essentially improved the convergence between calculations and experimental data of the nuclear stability. Theoretical calculations using the unified "drop-shell" model has allowed to predict an opportunity of existence of unknown stable superheavy nuclei - "islands of stability", presumably in the area of $Z=114$. The experiments which have been carried out by scientists from Lawrence Berkeley National Laboratory (USA), Flerov Laboratory in Dubna (Russia) and Research Center Rossendorf (Germany) have confirmed theoretical prediction [3]. Though the "island of stability" is not achieved yet, experimenters have precisely shown, that influence of neutron environment with $N=162$ (the next filled

shell predicted) on half-life of the synthesized nucleus already is observed [4] . Here interests of theorists closely adjoin to interests of experimenters as on the basis of theoretical predictions of "islands of stability" the new techniques of synthesis were development and new superheavy elements have been produced.

Nevertheless it seems to be noticed that functional dependence between nuclear composition (Z and A) and stability of isotopes is so complicated that in correspondence mathematical models semiempirical functions and parameters are usually used and sometimes experimental and calculation data are diverging. In this connection it seems to be useful to apply computer modelling methods to the treatment of the heavy elements experimental data and theoretical models complement and evolving.

For that purposes we selected database and interactive graphics computer methods and created the programs RADCHEM 1.0 and RADCHEMGRAF 1.0 presented here. RADCHEM 1.0 is the database containing as experimental data about isotopes (half-life, a kind of radioactive decay, decay energy characteristics, products of decay etc) as theoretical models explaining received experimental data. Submitting the data in such form is convenient for statistical and correlation analysis. So the investigations for actinides isotopes showed, that at the specified sampling of the unstable nucleus configurations by the probability theory and combinatorial numerical method the correlation between half-life and number of matching of protons and neutrons laid out above the last filled shell could be observed. RADCHEMGRAF 1.0 is the 3D-graphic program for the visual representation and analysis of several computed configurations of nuclei. As shown on the figure nucleus are imaged using mathematical model of "spheres density packing" (as more corresponded to nuclear "liquid drop –shells" model).



Figures 1-3. Surface energy differences for number of configurations for actinides isotopes calculated by the RADCHEMGRAF 1.0 also correlated with its stability.

We believe that the development of presented computer method lead to carry out more accurate calculations and to obtain further information, which could be supplement to the theoretical models. Then it will be perspective to apply RADCHEM 1.0 and RADCHEMGRAF 1.0 for education of specialists in radiochemistry as information resource and visual aids . For this purposes it seems to be very useful to replenish RADCHEM 1.0 with data for transactinides and we hope that TAN 03 Conference open up opportunities to make it.

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Optical spectroscopy of the heaviest elements

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The investigation of atomic, chemical and nuclear properties of heavy elements with charge numbers of $Z > 100$ is a real challenge. Heavy elements are produced in nuclear fusion reactions with rates of sometimes only a few atoms per week [1]. Their life-times are short, occasionally only in the order of milliseconds. At present, the most advanced method for the investigation of the properties of heavy elements is chemistry on single atoms in aqueous solutions [2] and in the gas phase [3]. This technique has already yielded detailed chemical information up to $Z=108$ [4]. Such experiments aid in the investigation of relativistic effects. For the heaviest elements these may result in alterations from expectations within a period of homologue elements. Relativistic effects, roughly speaking, originate from a shrinkage of the wave functions of inner shell electrons which, in turn, influence the binding energy of the valence electrons and thus the chemical properties. In the actinide region these are the 5f-, 6d-, 7p- and 7s- orbitals. Therefore, a more direct approach to investigate relativistic effects may be to study first ionization potentials (IP) or, even better, the atomic level schemes, both experimentally and by relativistic ab-initio multiconfiguration Dirac-Fock (MCDF) calculations or other methods [5-7]. However, any atomic spectroscopy, even with the most sensitive laser methods, is hampered by the fact that a broad band search for levels is limited due to the small number of atoms available for the studies. To obtain reliable information under these circumstances, it is necessary to incorporate theoretical level predictions in performing the experiments.

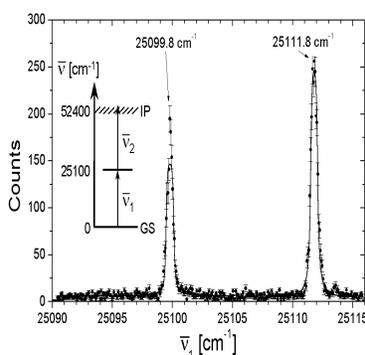


Figure 1. Observed levels in Fermium. The full line shows the best fit of a power broadened line profile to the data.

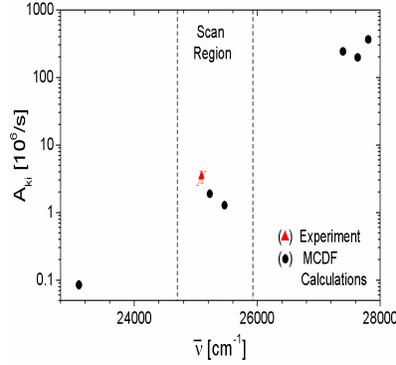


Figure 2. Partial transition rates to the $5f^{12}7s^2\ ^3H_{e_6}$ ground state as function of the level energy $\bar{\nu}$.

In this contribution we report our successful search for predicted atomic levels for elemental fermium for which experimental information was not available previously. The experiments [8] were performed with only a sample of 2.7×10^{10} atoms of the isotope ^{255}Fm which was bred in the high flux isotope reactor at ORNL, USA. The atoms with a half-life of 20.1 h were evaporated at a temperature of about 1,000 °C from a filament and stored in the argon buffer gas of an optical cell [9]. Atomic levels were sought by the method of resonance ionization spectroscopy using an excimer-dye-laser combination. Two atomic levels were found at wave numbers $(25,099.8 \pm 0.2)\text{ cm}^{-1}$ and $(25,111.8 \pm 0.2)\text{ cm}^{-1}$, see Fig. 1. Partial transition rates to the $5f^{12}7s^2\ ^3H_{e_6}$ ground state have been determined from their saturation characteristics. By comparison of the absolute level energies, as well as the partial transition rates with Multiconfiguration-Dirac-Fock (MCDF) calculations, term assignments of the observed levels are proposed. The calculations suggest that the leading orders of these levels could be the $5f^{12}7s7p\ ^5I_6$ and $5f^{12}7s7p\ ^5G_5$ term.

In a follow up experiment the three optical transitions around 27,500 cm^{-1} with large Einstein-coefficients of about $3 \cdot 10^8/\text{s}$ were sought for. Five additional lines were found in the wave number region of 27,100 to 28,400 cm^{-1} , see Fig. 3. A possible term assignment will be discussed.

Due to the small samples which were available, the ionization potential of fermium could not be determined, so far. However, an upper limit of 52,140 cm^{-1} was deduced from the experiment. This is in agreement with extrapolations of spectral properties, which yield $\text{IP}=52,400(600)$ [10] and MCDF calculations which yield $\text{IP}=50,800(2400)\text{ cm}^{-1}$ [11].

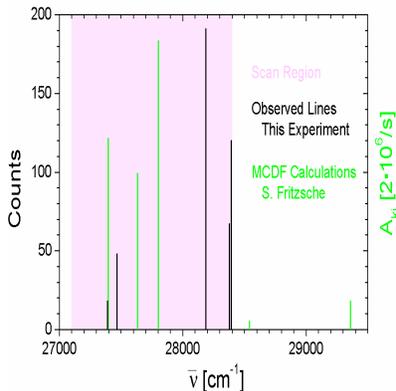


Figure 3. Wavenumber scan from 27,100 cm^{-1} to 28,400 cm^{-1} and MCDF calculations.

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Development of a plutonium ceramic target for the MASHA separator

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We are currently working on the development of a solid Pu ceramic target for use in the MASHA (Mass Analyzer of Super Heavy Atoms) on-line mass separator in Dubna. Along with recent upgrades of the U400 Cyclotron, MASHA will provide for at least a ten-fold increase in the production and detection rate for element 114 atoms, and will allow us to measure their atomic weights precisely. The MASHA separator will employ a thick Pu ceramic target capable of tolerating temperatures in the range of 2000 °C without vaporizing the actinide target. Promising candidates for the Pu target include Pu carbides and nitrides, although more research into the thermodynamic properties of these compounds will be required. Reaction products will diffuse out of the target and will drift to an ECR ion source after which they will be transported through the separator and will impinge on a position-sensitive focal-plane detector array. Furthermore, operation of the MASHA hot target/ion source combination will provide chemical volatility information that will support our assignment of an atomic number of 114 to these nuclei. Taken together, we expect that experiments on MASHA will allow us to make measurements that will cement our identification of element 114 and provide for future experiments in which the chemical properties of the heaviest elements are studied.