Achievements using the continuous SISAK α-Liquid Scintillation Technique, and its application in detection of $^{257}$Rf atoms

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Chapter 1

Introduction

1.1 Transactinide Research

The chemical elements of atomic numbers of 104 and higher are denoted transactinides. The Periodic Table of the Elements is shown in Figure 1.1, where the transactinide series, expected to fill the 6d-electronic shell is placed under the 4d and 5d transition elements. The placement of these elements in the Periodic Table is a fundamental objective of transactinide research. D. Mendeleyev arranged the elements after their atomic weight in 1869 [1]. His periodic system had open gaps for elements yet to be discovered and he predicted the chemical and physical properties of these elements. Today’s modern periodic system is organized after the atomic number of the elements, and as new elements are synthesized their characteristics in relation to the chemical periodicities are of great interest. As the charge of the nucleus increases, relativistic theory has to be applied to account for chemical properties of the elements. Relativistic effects scale roughly as $Z^2$, and are often divided in kinematic and spin-orbital effects [2, 3]. The electrons in the core-penetrating $s$ and $p_{1/2}$ orbitals of an atom with a high charge nucleus experience relativistic relaxation, and this will energetically destabilize the $p_{3/2}$, $d$, and $f$ orbitals. Spin-orbital effects, splitting of $l > 0$ states, might increase in size and influence the reaction cross section of atom and molecule. Diverging electronic configurations and unusual oxidation states may be expected for the transactinide elements as a consequence of relativistic effects.

From the perspective of disruptive Coulomb repulsion and attractive nuclear forces, nuclei of charge $\gtrsim 100$ should not exist. This is often referred to as the macroscopic limit of the nucleus [4]. Heavier nuclei exist because of shell effects. The arrangement of nucleons in discrete levels, nuclear shells, was introduced in the late 1940s (the shell model), explaining magical numbers, - certain combinations of protons and neutrons expressing enlarged stability [5]. In the 1950s the shell prescription was extended to models describing the single particle effects on collective motions.
in the nucleus [6] and the deformation of nuclei [7]. Strutinsky merged shell correction with a macroscopic prescription of the nucleus [8], resulting in the macroscopic-microscopic representation of fission. The enhanced stability of nuclei around another shell closure was discussed [9], and early macroscopic-microscopic calculations predicted $Z=114$ and $N=184$ as the next double magic closed shell [10, 11, 12]. Most calculations within the macroscopic-microscopic framework suggest spherical shell closure of this nucleus, but also expected shell closure for a deformed nucleus with $Z=108$ and $N=162$ [13, 14].

Other approaches to describe the nuclei of the transactinide region has been sought during the last three decades employing fully microscopic descriptions of the nucleus. Depending on the method, for instance using relativistic or non-relativistic forces, the next magic nuclide is $^{292}_{172}120$ [15, 16, 17, 18], $^{298}_{184}114$ [19, 15, 16, 20], $^{308}_{184}124$ [17], or $^{310}_{184}126$ [14, 15, 16, 17].

Presently 111 elements are recognized by the International Union of Pure and Applied Chemistry (IUPAC) [21]. As indicated in Figure 1.1, heavier ones are reported, $^{212}_{82}112$ [22], $^{213}_{83}113$ [23, 24], $^{214}_{84}114$ [25, 26], $^{215}_{84}115$ [23], $^{216}_{84}116$ [27, 28] and $^{218}_{84}118$ [29]. Most of the nuclides of the known heavy elements are isotopes of neutron deficient character, far from the proposed closed neutron shell of 184. Disintegration by $\alpha$-particle emission is dominant among these nuclides, but several decay partly or entirely by spontaneous fission. The half-lives are in order of $\mu$s to s. Theoretical macroscopic-microscopic calculations expect partial $\alpha$-decay half-lives ($T_{\alpha}$) to be smaller than the partial spontaneous fission half-lives ($T_{sf}$) in the region around $^{184}_{84}114$. The region in which heavy nuclei is assumed to be deformed spontaneous fission is predicted to be a more pronounced decay mode. The half-life calculations of the nuclides in the area around another closed shell nucleus are
dependent on the variables applied in the model, for example the half-life of $^{184}_{114}$ is predicted to about 12 min \[30\] and around 12 days \[31, 18\].

The transactinide elements are produced in heavy-ion fusion reactions, in which an accelerated beam of particles ($Z_1A_1$) is directed at a suitable target ($Z_2A_2$). The production mechanism is a two step process, first forming a compound nucleus ($Z = Z_1 + Z_2$) which is followed by emission of a few light particles (most often neutrons). This concept was introduced by Bohr \[32\], and according to his classical theory the compound nucleus is in equilibrium with respect to the total mass, charge, energy and angular momentum of the system describing the fusion reaction. In practice the compound nucleus is not completely in equilibrium, since it has a short life time (typically $10^{-14} - 10^{-19}$ s). The synthesis of transactinides via this mechanism is described by the fusion probability and survival probability of the compound nucleus. The fusion probability depends on the projectile energy and the coulomb barrier between $A_1Z_1$ and $A_2Z_2$. Survival of the compound nucleus is related to its excitation energy and angular momentum. (At a high angular momentum the nucleus fissions promptly.) In order to release the excess energy the compound nucleus can evaporate neutrons, but fission is more likely. Lower excitation energy of the compound nucleus implies a higher survival probability. The final state of the nucleus after emission of neutrons is referred to as an evaporation residue (EVR). A complete fusion reaction of an asymmetric combination of $A_1$ and $A_2$, using an actinide target and a suitable projectile is often called hot fusion. The reaction is exothermic and the excitation energy from 35 - 50 MeV result in the emission of 3 to 5 neutrons. More symmetric reactions with targets of lead or bismuth are only slightly exothermic and are referred to as cold fusion reactions. With excitation energies typically from 10 - 20 MeV, the compound nucleus can deexcite by emission of only 1-2 neutrons. Generally asymmetric reactions have higher production cross sections than more symmetric combinations of $A_1$ and $A_2$ since the coulomb repulsion is less, but due to the higher excitation energy the survival probability is lower.

The probability of the $Z_1$ projectile nuclide to completely fuse with the target nuclide $Z_2$ forming a transactinide element is rather small. A large number of other nuclear reactions are likely to take place when a target is bombarded with a projectile beam. Fast reactions in which a few valence nucleons in $Z_1$ and $Z_2$ are involved give rise to the term transfer products. The present reaction cross sections of transactinide nuclides are in the range of $10^{-33} - 10^{-36}$ cm$^2$. In practice these numbers represent a production rate of about 30 atoms/hour for Rf and Db and 1 atom/3 days for element 112. Transactinide research involves studies of single atoms, because each atom produced has disintegrated before the next atom is formed. A particular nuclide is identified by the presence of one or several time-correlated $\alpha$-particles, which are emitted by members of this nuclides decay chain.

Chemical investigations have been conducted on the lighter transactinides, employing fast techniques capable of performing within the short life-times of transactinide nuclides. The methods
developed include chromatography, ion exchange and solvent extraction. The elements Rf to Hs have been studied in gas-phase, while experiments in liquid phase have been performed with the first three transactinide elements. The chemical behavior of the transactinides known so far are covered in [33]. This book also present the instrumental techniques employed in transactinide research, specialized for studies of single atoms with short half-lives. Relatively few atoms (about ten) are needed to establish chemical identity, as shown by Borg and Dienes in their work concerning the validity of single atom chemistry [34]. In a static partition method, like solvent extraction, a single atom can only be present in one phase at the time, and therefore the partition coefficient have to be related to the probability of finding the atom in one phase or the other. Guillaumont et al. have treated this subject and derived an expression equivalent of the law of mass action [35].

1.2 The SISAK-system

The solvent extraction system SISAK allows a continuous separation of nuclides of half-lives down to \(~1\) \(\text{s}\) [36]. SISAK is an acronym for Short-lived Isotopes Studied by the AKUFVE-technique, where AKUFVE [37], a Swedish acronym, refers to arrangement for continuous investigations of distribution ratios in liquid extraction. The system was developed in the 1970s for chemical isolation of short-lived nuclides from complex nuclear reaction production mixtures [38]. Aftertime SISAK has been used for investigating both chemical and nuclear properties of numerous elements and nuclides. Today, the third generation of the system is operating [39], capable of liquid flow rates from 0.5 to 3.3 ml/s. The most significant changes have been reduction of the hold-up volume and residence time of the apparatus. SISAK III and the antecedents were originally constructed of palladium-passivated titanium, but in the mid-1990s the equipment was produced in the plastic material PEK (Poly ether ether ketone) [40]. The motivation for the choice of new material was PEK’s resistance against common acids like hydrochloric and sulfuric acid.

During the early 1990s the system was adapted for transactinide research. A detection system based on liquid scintillation was developed [41], because it appeared to be the most practical method. It was compatible with the large flow-rates of the SISAK-apparatus, and could be designed to operate in a continuous manner. Definite detection of an \(\alpha\)-emitting transactinide nuclide with this method requires the recording of genetically correlated \(\alpha\)-particles.

There have been several attempts of studying various transactinide nuclides with the SISAK-system in the 1990s, all with inconclusive results [42, 43, 44, 45, 46, 47, 48, 49]. In Table 1.1 each of these experiment are listed, giving years and locations, production reactions, and the applied extraction systems. The main problem in all these experiments was to distinguish \(\alpha\)-particles emitted from the transactinide nuclide from the activity of transfer products. The intense projectile also reacts with the target-backing material, aerosol particles and the carrier-gas. Typically, the background of \(\beta\)-particles and electrons was so high that it caused pile-up events disguising the
1.3 Objectives

The main objective of this thesis work has been development of the continuous α-liquid scintillation detection system used with the SISAK-apparatus, in the frame of solvent extraction. SISAK employed as a tool to investigate transactinide elements impose the premises of rapid and reliable detection, due to the short half-lives and low production rates of transactinide nuclides. Unambiguous liquid-scintillation detection of single atoms depend on a veritable understanding of the factors contributing to the radiative basis of this technique. The identification of a transactinide nuclide is based on nuclear properties, and chemical studies with SISAK is conditioned by a com-

Table 1.1: Earlier inconclusive transactinide experiments with the SISAK-system. In 1995 and 1996 extractive scintillator solutions were used, and the contents are given in the table footnote. From 1997 the scintillation solution is added after the extraction procedure.

<table>
<thead>
<tr>
<th>Year/Location</th>
<th>Reaction</th>
<th>Chemical system</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>1995/LBNL</td>
<td>$^{243}$Am ($^{22}$Ne, 4n)$^{261}$Db</td>
<td>1 M α-HIB/ext. scint.$^a$</td>
<td>[42]</td>
</tr>
<tr>
<td>1995/LBNL</td>
<td>$^{248}$Cm ($^{18}$O, 4n)$^{262}$Rf</td>
<td>2 M HCl/ext. scint.$^b$</td>
<td>[42]</td>
</tr>
<tr>
<td>1996/LBNL</td>
<td>$^{250}$Cf ($^{15}$N, 4n)$^{261}$Db</td>
<td>1 M lactic acid; 1 M α-HIB</td>
<td>[44]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>/1 M Cyanex 471x/ext. scint.$^c$</td>
<td></td>
</tr>
<tr>
<td>1996/PSI</td>
<td>$^{248}$Cm ($^{18}$O, 5n)$^{261}$Rf</td>
<td>2 M HNO$_3$/ext. scint.$^d$</td>
<td>[45]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>/2 M HNO$_3$</td>
<td></td>
</tr>
<tr>
<td>1996/GSI</td>
<td>$^{248}$Cm ($^{22}$Ne, 5n)$^{265}$Sg</td>
<td>1 M α-HIB/ext. scint.$^e$</td>
<td>[43]</td>
</tr>
<tr>
<td></td>
<td>$^{248}$Cm ($^{22}$Ne, 4n)$^{266}$Sg</td>
<td>/1 M α-HIB</td>
<td></td>
</tr>
<tr>
<td>1997/PSI</td>
<td>$^{248}$Cm ($^{18}$O, 5n)$^{261}$Rf</td>
<td>6 M HNO$_3$/0.025 M HDBP</td>
<td>[46]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>/1 M HNO$_3$</td>
<td></td>
</tr>
<tr>
<td>1998/GSI</td>
<td>$^{249}$Cf ($^{12}$C, 4n)$^{257}$Rf</td>
<td>6 M HNO$_3$/0.25 M HDBP</td>
<td>[47]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>/2 M NaNO$_3$</td>
<td></td>
</tr>
<tr>
<td>2000/PSI</td>
<td>$^{248}$Cm ($^{18}$O, 5n)$^{261}$Rf</td>
<td>6 M HNO$_3$/0.25 M HDBP</td>
<td>[48, 49]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>/2 M NaNO$_3$</td>
<td></td>
</tr>
</tbody>
</table>

$^a$2% by vol. TOA, 80 g/l napthalene and 0.5 g/l dimethyl-POPOP in toluene
$^b$0.5 M HDEHP, 80 g/l napthalene and 0.5 g/l dimethyl-POPOP in toluene
$^c$2% by vol. TOA, 2.5% by vol. dodecanol, 80 g/l napthalene, 1 g/L dimethyl-POPOP in toluene
$^d$0.25 M HDBP, 2.5% by vol. dodecanol, 80 g/l napthalene and 0.5 g/l dimethyl-POPOP in toluene
$^e$0.046 M TOA, 80 g/l napthalene and 0.5 g/l dimethyl-POPOP in toluene
plete evaluation and improvement of the method of liquid scintillation detection under on-line conditions.
Chapter 2

Background

2.1 Liquid Scintillation Theory

The method of liquid scintillation is widely applied to detect ionizing radiation. The ingredients of a liquid scintillation solution are usually of aromatic compounds, where one or several components exhibit light emission properties. The excitation of the $\pi$-electron system of the aromatic compounds is related to a series of singlet and triplet states. Only the $\pi$-electron system can deexcite by photon emission. Particularly, it is the transition from the first excited level of the singlet series to the ground state that emits fluorescence light, often called scintillation light. This is illustrated in Figure 2.1, a modified Jablonski diagram [50]. The levels of the singlet and triplet excited series are referred to by $S_1, S_2, \ldots, S_n$ and $T_1, T_2, \ldots, T_n$, the ground state is $S_0$. Each level has a finer structure of vibrational and rotational levels, of about 0.1 and 0.01 eV intervals.

\[
\begin{array}{c}
S_3 \\
\downarrow \text{intersystem crossing} \\
S_2 \\
\downarrow \text{internal conversion} \\
S_1 \\
\downarrow \text{fluorescence} \\
S_0 \\
\end{array}
\]

Figure 2.1: Deexcitation of the $\pi$-electron system of an aromatic molecule. $S_0$ indicates the ground state of the molecule and $S_i$ and $T_i$ represent the singlet and triplet exited states.
The main processes that occur in an aromatic molecule after excitation are indicated in Figure 2.1. Deexcitation from the first level of the triplet state to the ground state, phosphorescens, - is also a radiative transition, but not intense enough to be observed in solutions at room temperature. A radiationless transition between states of the same multiplicity is called internal conversion (e.g. $S_2 \rightarrow S_0$, $T_2 \rightarrow T_1$). The term intersystem crossing is used for radiationless transitions between states of different multiplicity (e.g. $S_2 \rightarrow T_2$, $T_1 \rightarrow S_0$). The mean life time of the higher exited states, $S_n$ are in the in the order of $10^{-12}$ s. The fluorescence is emitted within a few nano seconds (prompt fluorescence), but there is also a delayed scintillation component of a few micro seconds. This delayed fluorescence arise from the interaction of two triplet excited molecules, according to the scheme [51]:

$$T_1 + T_1 \rightarrow S_1 + S_0$$
$$S_1 \rightarrow S_0 + h\nu$$

The relative magnitude of the prompt and the delayed fluorescence is strongly dependent on the ionizing particle. The intensity of the delayed component increases with the LET-value of the ionizing particle. In fact, this relation between the delayed fluorescence and the LET-value, enables the possibility to separate different ionizing particles in liquid scintillation solutions. Analysis of the time dependency of the scintillation light is realized in the technique of Pulse-Shape Discrimination (PSD), that was first applied for n/$\gamma$ separation [52].

The ionization density along the path of an electron is so low that interaction between exited molecules is negligible. For an $\alpha$-particle the situation is quite the opposite, the high ionization density along the path results in ionizing quenching. Thereby the light yield of an $\alpha$-particle is a factor $\sim 10$ lower than for an electron. Mainly, the fast scintillation component is ionically quenched, implying that pulse analysis is the only mean to discriminate between particles.

Although, the delayed scintillation component is unaffected by ionic quenching, it is especially sensitive to chemical quenchers, like oxygen. The ability to separate $\alpha$-particles from electrons in a liquid-scintillation solution is conditioned by the removal of oxygen (the solutions are equilibrated with air). The intensity of the delayed fluorescence is reduced by the interaction of triplet excited molecules of the scintillation solution and oxygen [53]:

$$^3X^* + ^3O_2 \rightarrow ^1X + ^1O_2$$

For an electron of energy $> 125$ keV, the light-yield is a linear function of the particle energy. This is not the situation for highly ionizing particles. The light-yield per MeV for $\alpha$-particles has been documented by Flynn et al. [54], Horrocks [55] and McDowell [56]. These studies showed that the light output is linear for $\alpha$-particles of energy 4 – 7.7 MeV.
2.2 Rutherfordium

2.2.1 Solution chemistry of rutherfordium

Rutherfordium was first identified as the isotope $^{257}$Rf by Ghiorso et al. in 1969 [57]. The isotope was produced in the reaction $^{249}$Cf ($^{12}$C, 4n)$^{257}$Rf, and identified by genetically correlated α-particles from the nuclide and the daughter product $^{253}$No. The first experiments [58] investigating the chemical behavior of rutherfordium using an ion-exchange technique indicated that it was a group-4 element, along with titanium, zirconium and hafnium. The two elements zirconium and hafnium have analog outer electronic configuration and due to the lanthanide contraction near identical atomic and ionic radii, so their chemical properties are similar. They only exist in $+IV$ oxidation state in aqueous solutions and undergo extensive hydrolysis, already at pH $\sim 1$ [59]:

$$M(H_2O)_{8}^{4+} + OH^- \rightarrow MOH(H_2O)_{7}^{3+}$$

where $M=\text{Zr, Hf and Rf}$. The strength of hydrolysis is expected to decrease in the order: Zr > Hf > Rf.

According to the group trend, the electronic configuration of Rf would be [Rn] 5f$^{14}$6d$^{2}$7s$^{2}$. This ground state configuration was the result of an early relativistic Dirac-Fock (DF) calculation for a Rf atom in the gas phase [60]. Later, a possible ”p-like” character of Rf was suggested, as a consequence of relativistic stabilization of the 7$p_{1/2}$ orbital [61]. The result of some concurrent multi-configurational DF calculations was a 6d7p ground state configuration [62, 63], but in these papers it is argued that Rf is a typical d-element. More refined calculations of atomic Rf has concluded that its ground state configuration is 5f$^{14}$6d$^{2}$7s$^{2}$, and the low excitation energy of the 6d7p state can be ascribed to relativistic effects [64]. Kratz’ chapter in [65] is a relevant and interesting presentation and discussion of the chemistry of rutherfordium, both in liquid and gas phase. Most of the liquid phase experiments with rutherfordium have examined the competition between hydrolysis and complex formation reactions. A majority of the data has been obtained using manual extraction and the automated HPLC technique ARCA II [66].

A SISAK liquid-liquid extraction scheme for group-4 elements was proposed by Pense Maskow in his thesis from 1994 [67]. The system was based on selective extraction from HNO$_3$ into HDBP in toluene, in which Hf and Zr were well separated from lanthanides and group 5 and 6 elements (Nb, Ta, Mo, and W). In strong acidic solutions this expression is relevant:

$$M(NO_3)_2^{2+} + 2(\text{HDBP})_2 \rightleftharpoons M(NO_3)_2(\text{DBP} \cdot \text{HDBP})_2 + 2\text{H}^{2+}$$

As indicated in Table 1.1 Rf was sought studied with this extraction procedure at several occasions. In year 2000, SISAK was coupled to the Berkeley Gas-filled Separator (BGS) at Lawrence
Berkeley National Laboratory (LBNL), and this solvent chemistry was employed with success. The result is presented in Paper II, and the system is discussed in Chapter 5.

The extraction of Hf from oxalic and hydrochloric acid into Tri-n-octylamine (TOA) in xylene was investigated by Yakabe et al. in the mid 1970s [68]. Experiments with Hf and Zr were initiated to investigate the applicability of this system for studies of Rf with SISAK. Dyve performed manual and on-line extractions of Hf from a solution of oxalic acid mixed with hydrochloric acid into TOA and dodecanol in toluene for his Masters thesis [69]. Further work was undertaken by Omtvedt et al. to examine the extraction of Zr [70]. Tertiary amines like TOA tend to polymerize in inert solvents. Defining the extracted organic phase specie, which may consist of monomer, dimer, or polymeric amine salt can be difficult. Polymerization of tertiary amines seem to be smaller in aromatic than in aliphatic solvents, and strong Lewis base moderators also reduce aggregation [71]. Yakabes’ work shows that at high acid concentrations monomeric TOA seems to be the extracting specie, so the following overall extraction equation might also be the most relevant under SISAK-conditions:

\[ 4R_3N + 4H^+ + M(C_2O_4)_4^{2-} \rightleftharpoons (R_3NH)_4 \cdot M(C_2O_4)_4, \quad R = C_8H_{17} \]

It should be noted that Yakabes results are obtained with macro amounts of metal ion.

According to Dyve and Omtvedt the transfer of the Hf- and Zr-complexes was rapid enough to use this extraction method with SISAK. At the end of year 2000, Rf was extracted with this system, and the results are presented in Section 4.4. A further evaluation of the procedure is given in Chapter 5.

TOA is well know to extract group-4 elements from H_2SO_4 media. Tertiary amines are for example used large-scale commercially to recover uranium from ores, a procedure in which Zr(IV) is co-extracted. In 2002 extraction studies of Zr and Hf with the SISAK-system was started. It appeared that the SISAK-system could be used to investigate the complexation of group-4 elements, and especially to reveal different features of Zr and Hf. The investigations of this system is still on-going, but an experiment was conducted in the spring of 2003 in which Rf was extracted. Section 4.5 reports on the results of this experiment.

Manual extraction of Hf and Zr from H_2SO_4 into TOA in benzene was investigated by Yagodin et al. in the late 1960s [72]. They found the power dependence of the distribution ratio on the TOA concentration for metal ion extracted at acid concentrations < 1 M to be larger than one. Aggregation of TOA is expected and then the power dependence is the number of [TOA]_n molecules in the extracting compound divided by n. The results of Poláková [73], currently working on this system applying the SISAK-technique, indicate that the power dependence of the D-value on the TOA concentration is about 1.4. Based on her work and Zheng [74], the extraction mechanism of group-4 elements from 0.5 M sulfuric acid into 0.02 M TOA in toluene involve both [M(SO_4)_3]^{2-}
and $M\left(SO_4\right)_2$:

$$
\begin{align*}
R_3\text{N} + H^+ + HSO_4^- & \rightleftharpoons (R_3\text{NH}) \cdot HSO_4, & R = C_8H_{17} \\
2(R_3\text{NH}) \cdot HSO_4 + [M(SO_4)_3]^2- & \rightleftharpoons (R_3\text{NH})_2 \cdot M(SO_4)_{3} + 2 HSO_4^- \\
4(R_3\text{NH}) \cdot HSO_4 + M(SO_4)_2 & \rightleftharpoons (R_3\text{NH})_4 \cdot M(SO_4)_{4} + 2 H_2SO_4
\end{align*}
$$

The metal-complexes are also extracted by the amine sulphate, as suggested in [74], but the listed equations are thought to be the most important at 0.5 M sulfuric acid and 0.02 M TOA in toluene concentrations.

### 2.2.2 Nuclear properties of $^{257}\text{Rf}$ and decay product $^{253}\text{No}$

Studies of rutherfordium utilizing the SISAK-system can be accomplished by using the nuclide $^{257}\text{Rf}$. It has a half-life of 4.7 s and an $\alpha$-decay branch of $79.6 \pm 0.20%$ [75]. The daughter product $^{253}\text{No}$ (1.7 m) decays mainly by $\alpha$-particle emission, as shown in the decay scheme of $^{257}\text{Rf}$ in Figure 2.2. According to [75], based on the work of Ghiorso et al. [57], Bemis et al. [76] and Heßberger et al. [77] the electron capture branch of $^{257}\text{Rf}$ is $18 \pm 2\%$ and $2.4 \pm 0.3\%$ decays by spontaneous fission.

Heßberger et al. [78] have reported of an isomeric state of $^{257}\text{Rf}$ that is populated in the reaction process $^{208}\text{Pb}\left(^{50}\text{Ti,n}\right)^{257}\text{Rf}$. Two $\alpha$-lines of energy 9021 keV (34%) and 8968 keV (26%) were assigned to this isomeric state, that was not observed in the $\alpha$-particle decay of $^{261}\text{Sg}$. The partial half-life of the 9021-keV line is about 8 s, according to Heßberger et al.

The nuclide $^{253}\text{No}$ was first characterized by Mikheev et al. [79]. Ghiorso et al. [80] produced and detected $^{253}\text{No}$ the same year. The branching of $^{253}\text{No}$ is estimated to $\sim 80\%$ $\alpha$-particle emission and $\sim 20\%$ EC-decay. Studies trying to reveal the decay details of this nuclide has been pursued using in-beam spectroscopy and coincidence techniques [81, 82, 83]. The level scheme of $^{249}\text{Fm}$ as proposed by [82] and [83] are shown to the right in Figure 2.2. In [83] conversion electron coefficients of $\alpha_K < 0.35$ and $\alpha_L < 0.1$ are reported for the 279 keV level in $^{249}\text{Fm}$. 

Figure 2.2: Decay scheme of $^{257}$Rf from Table of Isotopes [75]. To the right a more detailed structure of the $\alpha$-decay chain as suggested by [78] and [82, 83].
Chapter 3

Method and Instrumentation

3.1 Radionuclide production and transportation to the SISAK-apparatus

The first SISAK-experiment to unambiguously detect a transactinide nuclide took place in November of year 2000. It was accomplished by coupling SISAK to the Berkeley Gas-filled Separator (BGS) at the Lawrence Berkeley National Laboratory (LBNL) 88-Inch Cyclotron. The nuclide $^{257}$Rf was extracted with HDBP in toluene from nitric acid, at a concentration of which Zr and Hf are known to extract with a high yield. Paper I and II report on the result of this experiment. In Paper I the liquid-scintillation detection of $^{257}$Rf $\alpha$-particles is analyzed in detail, and the transportation of the nuclide from production site to the chemistry apparatus is discussed. The extraction of complexed $^{257}$Rf is presented in Paper II.

In a typical SISAK experiment radionuclides are continuously transported from the production site in an accelerator or a research reactor to the SISAK equipment via a gas-jet transport system, as illustrated in Figure 3.1 - showing a general experimental set-up. The carrier-gas (He or N$_2$) is seeded with aerosols, usually of KCl, to which the recoiling nuclides attach. The gas-jet is connected to a switch yard, and the reaction products are either directed to the SISAK-system or to a collection site, at which the direct yield of the radionuclide production can be measured (not for single atom scale). Entering the SISAK-system, the radionuclides sticking to aerosols are dissolved when mixed with an appropriate aqueous solution, and the carrier gas is removed as the liquid is pumped through a degasser centrifuge. The aqueous phase and an organic extractant of choice are then fed into the centrifuge, in which the extraction takes place and the two immiscible phases are separated. A pre-mixer has recently been added ahead of the centrifuge, to increase the contact time of the two phases. The number of centrifuges depends on the separation system applied, in Figure 3.1 the two centrifuges represent an extraction and a scrubbing step. After
the separation procedure the organic output phase is combined with liquid scintillation solution, mixed with an inert gas and pumped through a degasser centrifuge. The inert gas washes out the equilibrated air of the scintillation solution. Removal of oxygen is crucial for pulse-shape discrimination, as explained in Section 2.1. The degasser centrifuge of the gas-wash step is often referred to as the booster, because its pumping capacity helps overcome the back pressure from the detection cells. The gas-wash step is addressed in Paper IV.

The detection system consists of several detection cells coupled in series. Presently, three detection cells with pulse-shape analyzing electronics are available. A valve is mounted prior to each detection cell, and the liquid will either flow through the cell or bypass it depending on the valve position. The valves can close automatically when an $\alpha$-particle of particular interest has been recorded. The scintillation solution is held in the detection cell to await for the possible decay of a daughter nuclide. This method of detecting genetically correlated $\alpha$-particles are often used to identify transactinide nuclides.

The solution flows through a monitoring cell immediately after passing the detection cells in which the light-yield is measured. Compton scattered electrons from a $^{137}$Cs-source are continuously recorded, and provide a measure of the scintillation-light yield. The energy of the $\alpha$-particles are corrected on-line according to the detection system light-yield. This method is the topic of Paper III.

A LabView [84] control program displays and records the purity of the liquid phases and the gas and liquid flow-rates. This information is stored for approximately 5 minutes as displayed in the program. The purity of the separated liquids are monitored using light diodes and phototransistors. If for some reason the phase separation becomes poor, the light transmission drops and the detection cells are automatically bypassed.

A photograph of the apparatus is shown in Figure B.1 in Appendix B. The SISAK degassers and centrifuges can be seen on the right side and on the left side are the detection cells which are mounted in an array of lead bricks.
3.1 Radionuclide production and transportation to the SISAK-apparatus

The radionuclides are transported via a He-jet to the SISAK apparatus.

3.1.1 The SISAK equipment

There are two static gas-liquid mixer designs, illustrated in Figure 3.2. The mixer in panel a, is a quartz tube with two glass frits (about 1 cm apart) in which the mixing take place. The hold-up volume is about 2.6 ml. Panel b is a drawing of the zigzag-mixer. It is a PEEK rod with tracks carved out, contained in a tube of quartz or PEEK. The liquid and gas mix at the crossing of the tracks. This design is produced in lengths of 4.5 and 8.5 cm. The efficiency of mixing He-jet transported radionuclides (0.5 l/min) into an organic liquid\(^1\) (0.4 ml/s) is in the order of 30 - 40% for the three mixers at room temperature. When the radionuclides are dissolved in an aqueous liquid, like HNO\(_3\) or H\(_2\)SO\(_4\), the mixer efficiencies are typically between 67 to 86%. The numbers depend on the liquid and gas-jet flow rates and

\(^1\)toluene, 0.05 M tri-n-octyl amine in toluene, and scintillation solution with toluene as solvent

Figure 3.1: General arrangement during a SISAK experiment, with α-liquid scintillation detection.

Figure 3.2: Drawings of the SISAK gas-liquid mixers. In Panel (a) the glass-frit mixer is depicted, and Panel (b) shows the zigzag-mixer.
the temperature of the aqueous solution. The highest efficiencies are obtained at around 80°C.

Figure 3.3 is a principle drawing of the HG-0.1 degassing unit [39]. The gas and liquid is separated by centrifugal force in the 0.1-ml separation volume, comprising three small chambers. The pumping capacity of the unit depends on the rotational speed, typically the degasser is operated at 20,000 - 23,000 rpm. The maximum liquid flow rate of the degasser is 3.3 ml/s. The efficiency of the degasser centrifuge is reported to be \( \geq 99.5\% \) [39].

![Figure 3.3: Cut-away drawing of the HG-0.1 degasser centrifuge, the darker parts are rotating. The gas and liquid mixture enters the separation chamber from the center. The gas moves up and exits through an opening in the outer wall (not shown in the drawing). The liquid is forced downwards, and is pumped out of the centrifuge due to a pump wheel, which is indicated in the drawing.](image)

A schematic drawing of the H-0.3 centrifuge is shown in Figure 3.4 [39]. The separation volume consists of four chambers, from where the phases are forced into a light and heavy phase collection chamber and from there on to the outlets. There are two manually operated valves regulating the pressure balance in the centrifuge. Under normal conditions the centrifuge runs at 16,000 - 25,000 rpm. A pre-mixer is mounted on the centrifuge input to increase the contact time of the liquids (not shown in Figure 3.4). It consists of a 4.3-cm PEEK cylinder, filled with PEEK-spon shavings.

### 3.1.2 Experimental facilities

**The 88-Inch Cyclotron**

The 88-Inch Cyclotron at LBNL provided beams of \( ^{50}\text{Ti}^{12+} \) and \( ^{48}\text{Ca}^{10+} \) for production of \( ^{257}\text{Rf} \) (Paper I and II, and Section 4.4 and 4.5) and \( ^{253}\text{No} \) (Section 4.6), respectively. The radionu-
3.1 Radionuclide production and transportation to the SISAK-apparatus

Figure 3.4: Cut-away drawing of the H-0.3 centrifuge, the darker parts are rotating. The liquids enter the separation chamber from the center of the centrifuge. The phases are separated due to different centrifugal forces and each phase is discharged via a stationary pump wheel.

eclides $^{155}$Hf and $^{206}$Rn were also produced to provide activity for transport time measurements and test of the detection system. Table 3.1 list the production reactions and some decay data for these nuclides. The cyclotron, fed by two Electron Resonance Sources (ECR) is capable of delivering a range of ion beams from hydrogen up to uranium. Typical beam intensities from the cyclotron were 200-300 pnA $^{50}$Ti$^{12+}$ and 750 pnA $^{48}$Ca$^{10+}$. 
Table 3.1: Production reactions and decay data for the nuclides produced at LBNL 88-Inch Cyclotron. The beam energies were 237 MeV $^{50}$Ti$^{12+}$ and 218.5 MeV $^{48}$Ca$^{10+}$.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Target thickness ($\mu g/cm^2$)</th>
<th>Cross section</th>
<th>$T_{1/2}$</th>
<th>Branching $\alpha/EC$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{208}$Pb ($^{50}$Ti, n)$^{257}$Rf</td>
<td>427</td>
<td>10.4 nb$^a$/ 15 nb$^b$/ 20 nb$^c$</td>
<td>4.7 s</td>
<td>0.796/0.18</td>
</tr>
<tr>
<td>$^{207}$Pb ($^{48}$Ca, 2n)$^{253}$No</td>
<td>470</td>
<td>$\sim$ 0.5µb$^d$/ $\sim$ 1.3µb$^e$</td>
<td>1.7 m</td>
<td>0.80/0.20</td>
</tr>
<tr>
<td>$^{120}$Sn ($^{50}$Ti, xn)$^{170}$−xHf</td>
<td>310</td>
<td>$\sim$ mb range</td>
<td>2.86 s</td>
<td>0.44/0.6</td>
</tr>
<tr>
<td>$^{164}$Dy (48Ca, 6n)$^{206}$Rn</td>
<td>380</td>
<td>$\sim$ mb range</td>
<td>5.67 m</td>
<td>0.62/0.38</td>
</tr>
</tbody>
</table>

$^a$ Heßberger et al. [78]
$^b$ GSI [85]
$^c$ Berkeley 2005 [86]
$^d$ T. L. Khoo et al. [87]
$^e$ Dubna 2004 [88]

A multiple target system is located inside the BGS, consisting of a 35-cm wheel of nine arc shaped target areas. The target wheel is located 0.5 cm downstream of the carbon (40 $\mu g/cm^2$) entrance window of the separator, and rotates with 300 rpm. Two silicon p-i-n detectors mounted ± 27 degrees incident to the beam, measure elastically scattered beam particles. The separator is filled with helium gas of 70-130 Pa, and the beam particles, and transfer products are spatially separated in-flight from the EVRs due to different magnetic rigidities in the gas. The transmission efficiency of the BGS is about 60% for the production of $^{257}$Rf [86].

The EVR are collected in the Recoil-product Transfer Chamber (RTC) [89], after passing through a 6-$\mu$m thick mylar-foil at the end of the separator. The pressure inside the chamber is 1.6-2.0 bar and it is connected to a He-jet transfer system. The He gas is seeded with aerosols, usually of KCl, for an efficient transport of the reaction products to the chemistry apparatus. Figure 3.5 is a schematic representation of this set-up. In Appendix B, Figure B.2 a picture of the RTC is shown.

A Focal Plane detector can be mounted at the end of the separator to measure the number of EVR produced and transmitted through the BGS. The position-sensitive detector consists of 32 silicon strips. The strip number provides the horizontal position and the vertical one is given by the difference in the charge collected at the top and the bottom of the strip.

**Oslo Cyclotron Laboratory**

The MC35 Scanditronix Cyclotron of the Oslo Cyclotron Laboratory (OCL) can deliver beams of hydrogen and helium ions. For the on-line experiments (Section 4.1) the cyclotron provided a 45-MeV $^3$He$^{2+}$ of 100-120 pmA intensity for the production of Yb, Hf, Zr and Cu isotopes. The
production reaction and $\gamma$-decay data for the nuclides of interest are listed in Table 3.2. In Figure 3.6 the target-chamber is shown. The carbon collimator diameter of 7 mm, is equivalent to the target size. The vacuum windows are 0.025-mm thick HAVAR-foils, and a 10-mm long Al cylinder is placed between the entrance window and the target. A 13-cm$^2$ carbon disk, 28 cm downstream of the target serves as a charge collector and provides a measure of the beam dose on target. The reaction products recoil out of the target foil and are retarded and stopped in the gas flowing through the target chamber. Here, the products are attached to aerosols, and are transported through an 18-m long steel capillary (i.d.=1.59 mm) to the SISAK apparatus.

Figure 3.5: General plan of the target chamber and the BGS coupled to the RTC at the 88-Inch Cyclotron at LBNL. A He-jet system transports the recoils to the chemistry area.
Table 3.2: Production reactions and γ-decay data for the nuclides produced at OCL.

<table>
<thead>
<tr>
<th>Target</th>
<th>Reactiona</th>
<th>Foil thickness (mm)</th>
<th>$T_{1/2}$ (s)</th>
<th>$E_\gamma$ (keV)</th>
<th>$I_\gamma$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Er(nat)</td>
<td>$^{164}$Er ($^3$He, 4n)$^{165}$Yb</td>
<td>0.025</td>
<td>9.9</td>
<td>80.1</td>
<td>49</td>
</tr>
<tr>
<td>Er(nat)</td>
<td>$^{166}$Er ($^3$He, 4n)$^{167}$Yb</td>
<td>0.025</td>
<td>17.5</td>
<td>113.34</td>
<td>55.4</td>
</tr>
<tr>
<td>Ni(nat)</td>
<td>$^{58}$Ni ($^3$He, n)$^{59}$Cu</td>
<td>0.01</td>
<td>81.5</td>
<td>1301.46</td>
<td>14.78</td>
</tr>
<tr>
<td>Ni(nat)</td>
<td>$^{60}$Ni ($^3$He, n)$^{60}$Cu</td>
<td>0.01</td>
<td>23.7</td>
<td>1332.50</td>
<td>88</td>
</tr>
<tr>
<td>Yb,Sr</td>
<td>$^{170}$Yb ($^3$He, 4n)$^{169}$Hf</td>
<td>0.025</td>
<td>3.24</td>
<td>492.86</td>
<td>84</td>
</tr>
<tr>
<td>Yb,Sr</td>
<td>$^{86}$Sr ($^3$He, 4n)$^{85}$Zr</td>
<td>na\textsuperscript{b}</td>
<td>7.86</td>
<td>454.20</td>
<td>45</td>
</tr>
<tr>
<td>Yb,Sr</td>
<td>$^{88}$Sr ($^3$He, 2n)$^{89m}$Zr</td>
<td>na\textsuperscript{b}</td>
<td>4.16</td>
<td>587.83</td>
<td>90</td>
</tr>
</tbody>
</table>

\textsuperscript{a} the cross sections for these reactions are in the mb range

\textsuperscript{b} na: not applicable, the weight of electrodeposited layer of Sr was not determined. The main interest was to produce enough activity for practical measures.

Figure 3.6: A simplified drawing of the target chamber at the OCL.

The combined target for production of Zr and Hf was prepared by electrodepositing Sr-oxide on a Yb-foil. The electroplating cell, similar to those presented in [90, 91] confines an area of 0.785 cm$^2$. A small volume of strontium in chloride solution (pH $<$ 3), was added to a solvent of molecular-grade 2-propanol and deposited [91].
3.2 Liquid scintillation detection of $\alpha$-particles

Three component scintillation mixtures are normally applied in $\alpha$-liquid scintillation spectroscopy. Several mixtures were investigated during the development work of the SISAK liquid-scintillation detection system [92]. It was decided to use a toluene-based extractive scintillator, with naphthalene (primary scintillator) and dimethyl-POPOP$^2$ (secondary scintillator). The use of an extractive scintillator was motivated by the advancements in commercial $\alpha$-liquid scintillation spectroscopy developed by McDowell [93].

Several compounds present in the extraction procedures, for example water and nitric acid, are quenchers. In the SISAK detection system, the scintillation-light yield was enhanced by adding the scintillation solution to the organic output phase after the extraction [45], see Table 1.1. It was further improved when the gas-wash (booster) step was implemented, and naphthalene was substituted with 1-methyl naphthalene, a derivate more soluble in toluene. The gas-wash step is discussed in Paper IV.

Figure 3.7 is a detailed drawing of the detection cell (University of Mainz), and its photo is shown in Figure B.3, Appendix B. It is a flow cell of Teflon with a meander-shaped track. The diameter is 65 mm and the volume is 5.5 ml. An 80-mm long light-guide is mounted on the Teflon block, with a thin disk of Kel-F$^3$ in between to protect the plexi-glass from the organic solvent. The detection system is equipped with photo-multiplier (PM) tubes from Hamamatsu, model R6091, with a diameter matching the detection cell. The light-guide conducts and spread the scintillation light uniformly over the photo cathode. The spectral response peak of the bi-alkali cathode of this tube is 420 nm, which corresponds to the fluorescence emission spectrum of dimethyl-POPOP expressing a maximum at 427 nm [94]. The performance of the scintillation mixture is reported in Paper IV, where the function of the light guide is also discussed.

The pulse from the PM-tube is amplified using a specially designed charge-sensitive preamplifier (University of Mainz) and a delay-line amplifier (Ortec, model 460). The unipolar output signal contains the pulse-height, and the bipolar output signal is analyzed with Ortec's 552 Pulse-Shape Analyzer and Timing Single Channel Analyzer. The falltime of the signal is determined by constant fraction triggering [95]. The pulse is assigned a pulse-shape parameter, reflecting the type of particle and aligned with the pulse-height signal. Figure 3.8 is a schematic representation of the electronic set-up for this pulse-shape analysis.

---

$^2$1,4-Di-2-(4-methyl-5-phenyloxazolyl)-benzene

$^3$Poly-trifluoro-chloroethylene
Figure 3.7: An illustration of the meander-cell, used for continuous liquid-scintillation detection of $\alpha$-particles.
3.2 Liquid scintillation detection of α-particles

Figure 3.8: Schematic representation of the pulse-shape analyzing electronics. The abbreviations are; PMT: Photo-Multiplier Tube, CSP A: Charge-Sensitive Pre-Amplifier, DLA: Delay-Line Amplifier, DA: Delay Amplifier, PSD/T -SCA: Pulse-Shape Discriminator and Timing Single Channel Analyzer, TAC: Time-to-Pulse Height Converter, ADC: Analog-to-Digital Converter, MUX: Multiplexer, ACQ PC: Acquisition PC.
For digital sampling of pulses, the PXD 222 DIGITIZER from LeCroy was used [96]. Its bandwidth is 200 MHz and the maximum sampling rate is 2.5 GSamples/s.

During operation the scintillation-light yield of the detection system is continuously monitored. The Compton spectrum of a $^{137}$Cs-source is recorded at fixed intervals, and the Compton-inflation point gives a measure of the scintillation-light yield. The detectors are calibrated with $\alpha$-particles from the decay of 3.96-s $^{219}$Rn and daughter product 1.78-ms $^{215}$Po. The nuclide $^{219}$Rn, a noble gas, is introduced into the scintillation solution via the flushing gas in the booster step. For the calibration a $^{227}$Ac-source (1.7 kBq on beryllium foil) is available, of which decay chain $^{219}$Rn is a member:

$$
^{227}\text{Ac} \xrightarrow{\beta^-} 21.8\text{y} \quad ^{227}\text{Th} \xrightarrow{\alpha} 18.7\text{d} \quad ^{223}\text{Ra} \xrightarrow{\alpha} 11.4\text{d} \quad ^{219}\text{Rn} \xrightarrow[6.819\text{MeV}]{3.96\text{s}}
$$

The radon emanating out of the $^{227}$Ac-source flows with the flushing gas streaming over the source. The $\alpha$-particles and the Compton spectrum are concurrently recorded when purposely varying the scintillation-light yield by adding a quencher. A numerical relationship is derived from the energy of an $\alpha$-particle as a function of the Compton-inflation point. A detailed description of this procedure is given in Paper III. The method enables on-line $\alpha$-particle energy correction for light-yield variations during an experiment.

Figures 3.9 and 3.10 show matrix plots of liquid scintillation detection of $\alpha$-particles emitted from $^{243}$Am and $^{241}$Am, respectively. The energy of the particles is given on the abscissa and the ordinate represents the pulse-shape of the particles. These two spectra illustrate an effect of relevance in $\alpha$-liquid scintillation spectroscopy, namely summing of a coincident electron with the $\alpha$-particle. The scintillation light produced when a conversion electron ejected immediately after $\alpha$-particle decay of a nuclide, may add to the light emitted due to the stopping of the $\alpha$-particle. In the decay of $^{243}$Am, a 74.7-keV level in $^{239}$Np is populated. The transition to the ground state has a total conversion coefficient, $e/\gamma$, of 0.280 [97], and the life-time of the level is 1.40 ns. This is not enough time to resolve the electron from the $\alpha$-particle in an liquid-scintillation solution. (Implicit that the electron has moved in the direction of the $\alpha$-particle track.) The $\alpha$-peak is asymmetrical and has a high-energy tail of sum-pulses. Figure 3.11 shows a simplified decay scheme of $^{243}$Am, and so of $^{241}$Am, in which the situation is different. A 59.5-keV level in $^{237}$Np with life-time 67 ns is populated in the $\alpha$-particle decay of $^{241}$Am, which is long enough to resolve the conversion electron from the $\alpha$-particle. The total conversion coefficient from this state is 1.16 [98].
3.2 Liquid scintillation detection of $\alpha$-particles

Figure 3.9: Decay of $^{243}$Am recorded with $\alpha$-liquid scintillation detection. $^{243}$Am decays with a 5.28-MeV $\alpha$-particle branch of 87.4%.

Figure 3.10: Liquid scintillation spectrum of $\alpha$-particles from the decay of $^{241}$Am, 85.2% of the $\alpha$-particles are emitted with an energy of 5.49 MeV.
3.3 Analysis

3.3.1 Acquisition

The pulse-height and -shape of detected events are digitized with two Analog-to-Digital Converters (ADC) as shown in the electronic set-up in Figure 3.8. To separate the ADCs from each other, they are assigned consecutive numbers (1,2,3,...), where 1 and 2 are pulse-height and -shape for detector cell 1, and so on. The data from all the ADCs are added into one stream of data by a specially designed multiplexer [99]. The stream of data is collected by a PC with a fast AT-DIO32F digital input/output-card from National Instruments [100]. The data are organized in 16 bits words, where 13 bits are data and 3 bits are used as an index code to identify the ADC which generated the data.

The acquisition program Loke [101], reads data from the I/O-card, and since the stream of data is random in time the acquisition mode is driven by hardware triggered interrupts. Loke is written in Delphi [102], - Borland’s implementation of the computer language Pascal. The low level interface to the acquisition card is written in C++ and developed with the driver development tool WinDriver from Jungo [103].

The groups of data that carry pulse-height and -shape information need to be identified for each single detector event. Using a "false" ADC, a word with index code '0' is the designated separator signal. The acquisition program generates five words which are inserted and replace the separator signal. The first three words have index code '0' and the last two have '7'. Word one...
carries the Compton-edge inflection point and is subindexed as '0'. Whether the valve is open or closed is stored in the second word, which has subindex '1'. The third word, with subindex '2', was not used in the experiments presented in this thesis, but was reserved for the corrected energy of the event. The two words indexed as '7' contain the time information for each event into the stream of data. A counter on the I/O-card drives the clock with a precision of 780 Hz. The clock time is stored as a 26 bit integers of the 2 words, the 3 remaining bits of each word are used to index the time as '7'. Figure 3.12 is a schematic representation of the data stream, with one highlighted event consisting of the five words inserted by the software followed by the number of the triggering ADC with pulse-height and -shape information.

![Figure 3.12: Illustration of the data stream generated by the multiplexer. The abbreviations are; MSB: Most Significant Byte and LSB: Least Significant Byte](image)

It may also be necessary to bypass the detection cells for other reasons than waiting for a time correlated \( \alpha \)-particle. A gate signal, related to the purity of the liquid phases after the extraction step, can close the valves to all the cells if the scintillation solution is contaminated with aqueous phase. It is recorded within the second word indexed as '0' in the stream of data.

### 3.3.2 Digital pulse analysis

It is intended to develop tools for digital pulse-shape analysis to improve the evaluation of data recorded with the SISAK liquid scintillation detection system. The analysis of digitally sampled PMT-pulses might unveil information that is lost in the analog processing of the pulse, for example sum-pulses of an electron coincident with an \( \alpha \)-particle. The basis for digital analysis of pulses is established with a program called *LokeDigitalAnalysis* [104]. This program is work in progress.

Langrock’s thesis work [105] was concerned with digital pulse-shape analysis in liquid scintilla-
tion detection, sampling pre-amplified scintillation pulses. A method based on neural networks was applied to discriminate between $\alpha$-particles and electron/$\beta$-particles. He also suggested how digital analysis in liquid scintillation detection can, in certain instances, be used for nuclear spectroscopy, and showed its application on the decay of $^{241}\text{Am}$.

Typical digitally sampled scintillation pulses from the PM-tube are shown in Figure 3.13. These are used in the analysis presented in Section 4.6.

![Typical α- and electron/β-scintillation pulses from the PM-tube recorded with the PXD 222 DIGITIZER. The pulses are inverted.](image-url)

Figure 3.13: Typical $\alpha$- and electron/$\beta$-scintillation pulses from the PM-tube recorded with the PXD 222 DIGITIZER. The pulses are inverted.
Chapter 4

Results

4.1 Helium-jet transport of radionuclides at the OCL

Experiments were carried out at the OCL to investigate the mixing efficiency of He-jet transported radionuclides into an organic liquid in a SISAK degasser centrifuge. The experimental set-up is shown in Figure 4.1. Toluene was used instead of the combined organic extractant and scintillation solution in the experiments presented here for simplicity and economy. The two liquid phases were compared and no significant difference in activity yield could be found. The toluene carrying the activity was collected in 250-ml bottles and counted on an 80% HPGe detector. In a direct catch run, the collected activity (on filter) was counted after dissolution in 6 M HNO₃.

The nuclides given in Table 3.2 were produced to measure the influence of the ionic charge on the mixing efficiency in toluene. The yields in toluene related to direct catch measurements for the four radionuclides of different valences are given in Table 4.1. No significant difference in the yield of the nuclides was found.

Traditionally, KCl has been the applied and preferred aerosol material in transactinide research. Transporting nuclides produced in heavy-ion fusion reactions (like transactinides) imposes
Table 4.1: The yield of different He-jet transported nuclides in toluene using KCl aerosols.

<table>
<thead>
<tr>
<th>ion</th>
<th>yield (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu(II)</td>
<td>34.8 ± 1.2</td>
</tr>
<tr>
<td>Yb(III)</td>
<td>34 ± 3</td>
</tr>
<tr>
<td>Hf(IV)/Zr(IV)</td>
<td>41 ± 6</td>
</tr>
</tbody>
</table>

restrictions on the choice of aerosol material. The reason for this is when low-Z aerosol materials are used, byproducts from beam interactions with the aerosol interfere with detection of the wanted reaction product. When preseparation is applied as in the experiment described in Paper I and II other aerosol materials can be used, since the beam is deflected by the mass separator. The salts PbCl$_2$ and PbI$_2$ were tested, along with some organic compounds. In Table 4.2 the yields are given as the number of counts per second normalized to $10^5$ beam integral pulse. PbI$_2$ appeared to be the most efficient aerosol material, but it was decided to pursue PbCl$_2$ in the oncoming experiments. The organic compounds did not transport the produced nuclides through the gas-jet.

Table 4.2: Activity of $^{165,167}$Yb transported through a He-jet and mixed with toluene for three different aerosols.

<table>
<thead>
<tr>
<th>material</th>
<th>relative yield (c/s$\cdot10^5$ beam integral unit)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KCl (670)</td>
<td>105 ± 6</td>
</tr>
<tr>
<td>PbI$_2$ (400)</td>
<td>260 ± 5</td>
</tr>
<tr>
<td>PbCl$_2$ (430)</td>
<td>246 ± 9</td>
</tr>
</tbody>
</table>

Using PbCl$_2$ as aerosol material, the gas-jet flow was varied for toluene flowing at rates of 0.4 and 0.6 ml/s. Figure 4.2 shows the yield as number of counts per second normalized to $10^5$ beam integral unit. The bars indicate the counting uncertainty. The maximum appears at a He flow rate of 0.25 l/min and toluene flowing at a rate of 0.4 ml/s. During the 0.4 ml/s series the pressure in the target chamber increased from 1.12 atm to 1.15 atm, and from 1.91 atm to 2.00 atm in the 0.6 ml/s series. This figure clearly shows the span in one set of condition, here actualized at 0.5 l/min gas and 0.4 ml/s toluene flow. The large variation in the yield at this gas and liquid flow-rate is shown in Figure 4.3. Here, the ratio of the beam count to the number of counts in sample is plotted against the sample number, for two different experimental periods. PbCl$_2$ and KCl were used as aerosol material and repeatedly measured at this condition throughout the periods of beam time,
as presented in the upper and lower panel of Figure 4.3, respectively. The pressure in the target chamber was 1.27-1.29 atm at these flow-rates.

Figure 4.2: The activity of $^{165,167}$Yb in toluene for different He-flow rates and PbCl$_2$ aerosol particles, measured at 0.4 and 0.6 ml/s toluene flow.

During a period of beam time, extending over several separate days, the yield is decreasing. The lower panel of Figure 4.3, presenting a beam time period with the application of KCl aerosols shows a steady yield reduction. The samples were measured over three days, but the majority, samples seven to thirty-two, on day two. The yield for the PbCl$_2$ aerosol samples, presented in the upper panel, seem to vary more randomly. Under this beam time period, lasting one week, the capillary was cleaned with isopropanol and dried after samples 42 and 92, indicated with dashed lines in the figure. In both instances the yield increases for the succeeding samples, indicating that material deposited in the capillary affects the yield.
Figure 4.3: The ratio between the beam integral count and the number of $^{165,167}$Yb counts in the sample varying with the sample number. The experimental period in the upper panel investigated the transport yield using PbCl$_2$ aerosols. KCl aerosols was used in the experiment depicted in the lower panel.
4.2 Liquid scintillation detection of $^{257}\text{Rf}$.

The liquid scintillation detection of $^{257}\text{Rf}$ α-particles is reported in Paper I. The nuclide $^{257}\text{Rf}$, transported via a He-jet seeded with KCl aerosols was directly mixed with liquid scintillation solution, 1-methyl naphthalene and dimethyl-POPOP in toluene. This type of measurement was carried out in order to evaluate the detection system and to determine a maximum $^{257}\text{Rf}$ detection rate for normalization of the extraction yield discussed in Paper II. The recorded events are discussed in relation with the measured transport- and hold-up times and decay properties of the nuclide.

Alpha-particle spectra of the data presented in Paper I are shown in Figure 4.4 and 4.5. The α-particles closing the detection cell (selected by user-defined energy gates in Loke) are plotted in the upper panel. The cross hatched areas represent the closing events where another (or several) α-particle was recorded during the time interval the detection cell was closed. In the lower panel the events registered during cell closure are depicted with the time-correlated closed-cell events. According to the known decay properties of $^{257}\text{Rf}$ (Section 2.2.2), the probability of detecting α-particle decay of $^{257}\text{Rf}$ and daughter $^{253}\text{No}$ is 0.64.

![Figure 4.4: Energy spectra of closing (upper panel) and closed-cell events (lower panel) in detection cell 1. The cross hatched bins show the closing events, during which cell-close time one or more α-particles were recorded.](image)

Totally, 46 decay series of two or three time-correlated α-particles were observed in this exper-
Figure 4.5: Energy spectra of closing (upper panel) and closed-cell events (lower panel) in detection cell 2. The cross hatched bins show the closing events, during which cell-close time one or more $\alpha$-particles were recorded.

iment. The valve position of detection cell 1 was closed for 54% of the closing events in detection cell 2.

4.3 Extraction of $^{257}$Rf from nitric acid with HDBP

Rutherfordium was extracted from 6 M nitric acid into 0.25 M HDBP in toluene in the experiment presented in Paper II. The decay losses were approximated by using the program SimSISAK [106]. The program comprises two pieces. The first generates the probability function of an atom’s location after a certain time period. The latter, calculates the probability of an atom to survive transport through the system, and decay during its residence time in the detection cells using a Monte Carlo technique. The hold-up time in the gas-jet/target chamber is simulated by the volume divided by the gas-flux. This is a crude approximation, especially in these experiments using the RTC, which has a large volume. So the numbers given in Paper II are for the relative comparison in the analysis given within and not to be treated as absolute yields.

The transport time was examined equivalently to the measurement described in Paper I, with activity from $^{164-166}$Hf. Figure 4.6 shows the residence time distribution, which has a break-
4.4 Extraction of $^{257}\text{Rf}$ from oxalic acid with TOA

through time of 10 s, and peak at 17 s.

Figure 4.6: The transport time to detection cell 1, for set-up in Paper II. $^{164-166}\text{Hf}$ were produced with a pulsed beam.

The energy spectra of the recorded events are shown in Figure 4.7. In the upper panel of the figure the closing events are depicted, ten $\alpha$-particles are assigned to $^{257}\text{Rf}$, and a few closing events are ascribed to $^{253}\text{No}$. The lower panel shows the closed-cell events, with the five correlated $^{253}\text{No}$ $\alpha$-particles that are emphasized in the correlation plot in Figure 3 in Paper II.

4.4 Extraction of $^{257}\text{Rf}$ from oxalic acid with TOA

The extraction of rutherfordium from oxalic acid into TOA in toluene was examined using pre-separated $^{257}\text{Rf}$ produced at the LBNL 88-Inch Cyclotron. Production details for $^{257}\text{Rf}$ are given in Section 3.1.2. The SISAK schematics for this experiment is shown in Figure 4.8. The aqueous phase was 0.1 M oxalic acid and 0.3 M hydrochloric acid, and the organic phase, 0.1 M TOA in toluene, contained some 1-dodecanol as a moderator to improve the phase-separation in the centrifuge. This experiment was performed concurrently with the experiments presented in Paper
Figure 4.7: Energy spectra of the closing events (upper panel) and closed-cell events (lower panel). Each event is put in one of five groups according to its decay assignment.

I and II. The transport of $^{257}$Rf from production site to the chemistry set-up is discussed in Paper I, and is assumed to apply for this experiment. The effective beam-time was about 20 hours. Two detection cells were used and the detection system was calibrated according to the method presented in Paper III.

Figure 4.9 shows the energy spectra of the closing events (upper panel) and the events recorded during cell closure (lower panel), for detection cell 1 and 2. The cross hatched areas indicate closing events with energy $\geq 8.0$ MeV, correlated with a closed-cell event. The time-correlated closing and the closed-cell events (the shaded events of Figure 4.9) are displayed in Figure 4.10.

As seen of Figure 4.9 and 4.10 definitive assignments of decay sequences are not straightforward for this set of data. The energy calibration must have been off-set by several MeV during the experiment, since the closing event spectrum contains counts in the whole region of 6 to 9 MeV. In practice this means that the detection cells were also closed by $\alpha$-particles from the decay of $^{253}$No, $^{249}$Fm, and $^{211}$Bi (present due to the calibration procedure, see Section 3.2 and Paper I). Regardless of all the cell closing events, the analysis does show qualitatively that rutherfordium extracts in this system.
4.4 Extraction of $^{257}$Rf from oxalic acid with TOA

Figure 4.8: Diagram of the SISAK-system for studying the extraction of $^{257}$Rf from a mixture of 0.1 M oxalic acid and 0.3 M HCl into 0.1 M TOA and 2.5% by volume 1-dodecanol in toluene.

Figure 4.9: Energy spectra of the closing and closed-cell events, in the upper and lower panel respectively. The cross hatched bins of the upper panel indicates the closing events succeeded by one or more closed-cell event of energy $> 7.5$ MeV. The lower panel shows the events recorded during cell closure caused by a closing event of energy $> 8.0$ MeV.
Figure 4.10: Graphic representation of the time-correlated closing events and the closed-cell events. $^{257}$Rf emits $\alpha$-particles with energies from 8.5 MeV to 9.0 MeV, and the $\alpha$-particles from $^{253}$No have energy around 8.0 MeV. The events are grouped according to time interval between them as a fraction of the half-life of $^{253}$No, 102 s.
4.5 Extraction of $^{257}\text{Rf}$ from sulfuric acid with TOA

$^{257}\text{Rf}$ was produced at the 88-Inch Cyclotron at LBNL for studying its extraction from 0.5 M H$_2$SO$_4$ into 0.02 M TOA in toluene. The general plan for the experiment is shown in Figure 4.11.

![Figure 4.11: Experimental set-up to study the extraction of $^{257}\text{Rf}$ from 0.5 M H$_2$SO$_4$ into 0.02 M TOA in toluene with SISAK.](image)

4.5.1 Transport time measurements

The transport time from target to the detection cells was measured with activity from $^{164-166}\text{Hf}$. A pulsed beam of $^{50}\text{Ti}$, 3 s on and 87 s off, was used to produce hafnium in the reaction $^{120}\text{Sn} (^{50}\text{Ti}, xn)^{170-8}\text{Hf}$. The gas-jet carried radionuclides and liquid-scintillation solution were mixed in the first stage depicted in Figure 4.11 and pumped through the remaining apparatus and detection cells as shown in the set-up. The scintillation solution was pumped in a closed loop at 0.8 ml/s and changed when needed.

In order to estimate the number of $^{257}\text{Rf}$ atoms to be detected, a transport-time distribution was estimated by a Monte Carlo simulation based on the measured residence time distribution of $^{164-166}\text{Hf}$ and the decay curve of $^{257}\text{Rf}$. This distribution is shown in Figure 4.12. The expected number of $^{257}\text{Rf}$-atoms to be recorded in the first detection cell is 7 out of 1000 produced atoms, using 50% gas-jet, 67% mixer, and 80% extraction yields. It should be noticed that this rough simulation does not include the probability of the nuclide to survive past the detection system.

Alpha-particle correlations from the decay of $^{257}\text{Rf}$ and daughter $^{253}\text{No}$ were measured with a focal plane detector (Section 3.1.2) to determine the count rate of scattered beam per $^{257}\text{Rf}$-atom.
produced. The numbers and efficiencies are given in Table 4.3, and the expected production rate was estimated to 1 $^{257}$Rf atom per 393 counts in the Rutherford scattering detectors.

Table 4.3: Number of $^{257}$Rf atoms per number of counts in the Rutherford scattering detectors.

<table>
<thead>
<tr>
<th>Number of $^{257}$Rf</th>
<th>EVR-α correlation: 5.129 cps</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scattered beam count:</td>
<td>1.370 · 10$^{-3}$ cps</td>
</tr>
</tbody>
</table>

Efficiencies:

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>BGS</td>
<td>21%</td>
</tr>
<tr>
<td>$\epsilon_{FP}$</td>
<td>50%</td>
</tr>
</tbody>
</table>

Expect 1 $^{257}$Rf per 393 counts of scattered beam

4.5.2 Recorded events

Energy spectra of the closing events and the closed-cell events are shown in Figure 4.13. The events are given in different colors to indicate the nuclide of which the event originated. The closing events presented in the upper panel contain ten events assigned to the $\alpha$-particle decay of $^{257}$Rf. The lower panel representing the closed-cell events, of which six are assigned to $^{253}$No. The unassigned event are correlated to the closing event which have not been ascribed to $^{257}$Rf. The six cell closures and subsequent $\alpha$-particle decay assigned to $^{257}$Rf and daughter $^{253}$No, are listed in Table 4.4. For each chain the energy and the time elapsed between the particles are given. Figure 4.14 shows the time-correlated closing events plotted against the closed-cell events, and the $\alpha$-particles from $^{257}$Rf and $^{253}$No (Table 4.4) are enclosed in the dashed-line box.
Figure 4.12: An estimated detection probability of $^{257}\text{Rf}$ in detection cell 1, based on a Monte Carlo simulation using the decay curve of $^{257}\text{Rf}$ and the measured residence time distribution of $^{164-166}\text{Hf}$. 
Figure 4.13: Energy spectra of the closing events (upper panel) and closed-cell events (lower panel). The events are categorized according to their decay assignments.

Table 4.4: The energy and time difference for time-correlated α-particles in the detectors. The parenthesis of the first column indicates in which detection cell the α-particles were recorded.

<table>
<thead>
<tr>
<th>Event (DC#)</th>
<th>Energy (keV)</th>
<th>∆t (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 (1)</td>
<td>8398</td>
<td>57</td>
</tr>
<tr>
<td></td>
<td>8108</td>
<td></td>
</tr>
<tr>
<td>2 (2)</td>
<td>8823</td>
<td>270</td>
</tr>
<tr>
<td></td>
<td>7817</td>
<td></td>
</tr>
<tr>
<td>3 (1)</td>
<td>8946</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>8127</td>
<td></td>
</tr>
<tr>
<td>4 (1)</td>
<td>8954</td>
<td>143</td>
</tr>
<tr>
<td></td>
<td>8085</td>
<td></td>
</tr>
<tr>
<td>5 (1)</td>
<td>8736</td>
<td>246</td>
</tr>
<tr>
<td></td>
<td>8103</td>
<td></td>
</tr>
<tr>
<td>6 (1)</td>
<td>8630</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>7787</td>
<td></td>
</tr>
</tbody>
</table>
Figure 4.14: Time correlated $\alpha$-particles from the decay of $^{257}$Rf and daughter $^{253}$No. The $\alpha$-particles from $^{257}$Rf have energies from 8.5 MeV to 9.0 MeV, and $\alpha$-particle decay of $^{253}$No has about 8.0-MeV energy. The correlated closing and closed-cell events are plotted with time-difference dependence as a fraction of the half-life of $^{253}$No, 102 s.
4.6 Digital pulse-shape analysis of \( \alpha \)-decaying nuclides

Scintillation-pulses from the PMT were sampled digitally during the detection of \( ^{253}\text{No} \), \( ^{206}\text{Rn} \), \( ^{219}\text{Rn} \), \( ^{215}\text{Po} \), \( ^{243}\text{Am} \) and \( ^{241}\text{Am} \). \( ^{253}\text{No} \) and \( ^{206}\text{Rn} \) were produced at the LBNL 88-Inch Cyclotron and transported to a SISAK mixer and degasser centrifuge in a He-jet with aerosols of PbCl\(_2\). In Section 3.1.2 the details of the production of \( ^{253}\text{No} \) and \( ^{206}\text{Rn} \) are given. The radionuclides were mixed with scintillation solution and pumped through a detection cell as shown in the experimental set-up in Figure 4.15. The calibration nuclides \( ^{219}\text{Rn} \) and \( ^{215}\text{Po} \) were measured as explained in Section 3.2. Similarly, \( \alpha \)-particles from \( ^{241}\text{Am} \) were recorded. A small amount of \( ^{241}\text{Am} \) extracted with HDEHP\(^1\) in toluene from hydrochloric acid was added to \( \sim 100 \) ml of scintillation solution.

The sampled PMT pulses were smoothed\(^2\) before its cumulative distribution was fitted with the following expression:

\[
A_1 \cdot \ln (A_2 (x - x_0) + 1) + A_3 \frac{x - x_0}{1000} + A_0
\]

where \( x \) is the channel number. When the value of \( x = x_0 \), the second derivative of 4.1 equals \( A_1 \). In Figure 4.16 the cumulative distribution of a digitally sampled \( \alpha \)-pulse is shown with the fitted curve. The parameter \( A_1 \) represents the curvature of the cumulative distribution and take a different value for \( \beta \)-particles/electrons and \( \alpha \)-particles.

Pulse-shape analysis based on a digital representation of the PMT scintillation-pulses are not straightforward. Figure 4.17 shows that only for high-energy scintillation pulses the separation between \( \beta \)-particles/electrons and \( \alpha \)-particles is good. The ADC in the DIGITIZER might be a limiting factor for this procedure. The dynamic range of the PXD 222 DIGITIZER-ADC is 256 channels, and the precision for small pulses are not satisfactory.

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\(^1\)bis(2-ethylhexyl)phosphoric acid
\(^2\)10 point Fast Fourier Transform
4.6 Digital pulse-shape analysis of α-decaying nuclides

Figure 4.16: Cumulative distribution of a typical α-particle PMT-pulse recorded digitally. The distribution is fitted with the expression in 4.1, which yielded the parameters shown in the box.
Figure 4.17: The pulse-shape of digitally sampled \( \alpha \)- and electron/\( \beta \)-particles represented by the curvature \( A_1 \) of the cumulative PMT-pulse distribution.
Chapter 5

Discussion and future perspectives

5.1 Alpha LS-detection of single atom decay

The method of liquid scintillation has over a few decades proven itself as a useful and powerful tool for detection of α-decaying nuclides. The work presented in Paper I and II of this thesis shows that α-liquid scintillation is also feasible for identification of single atoms, and count rates of a few atoms per hour. The foremost advantage of liquid scintillation detection is sensitivity, - an almost 4π-detection efficiency can be achieved. Since all charged particles can be registered in a liquid scintillation solution, a careful assessment of the background is required. The first SISAK-experiments to extract and subsequently, detect a transactinide nuclide owes its success to the use of the Berkeley Gas-filled Separator to reduce the background of beam particles and other reaction products. The BGS as a preseparator seems to be a necessity for liquid scintillation detection of α-decaying nuclides produced at low cross sections, such as transactinide elements. The immediate success of coupling the SISAK-system to the BGS suggests new possibilities in transactinide research. A physical preseparator combined with a chemical procedure offers more freedom in choosing chemical systems. Without the intense background of projectile beam particles, it is for example, possible to improve the aerosol/gas-jet transport by employing other aerosol materials.

A recoil separator, especially designed for (super)heavy element chemistry is currently under construction at the GSI UNILAC-facility. It is named TASCA, TransActinide Separator and Chemistry Apparatus. TASCA will be optimized for separation of (super)heavy elements produced in asymmetric reactions (hot and warm\textsuperscript{1} fusion). The BGS is most efficient for nuclides produced in fusion reactions of Z~20 bombardment on Pb/Bi. In more asymmetric reactions the probability increases for breakthrough of transfer products. A too symmetric combination will make the separation of beam particles and product nuclide more difficult.

\textsuperscript{1}here warm fusion is used to denote fusion reactions of \textsuperscript{48}Ca (beam) with an actinide element (target)
Running the SISAK-apparatus in combination with the BGS at the LBNL 88-Inch Cyclotron is limited by the transport time from production site to the chemistry area. It is clear from the experiments on $^{257}$Rf, - in which the decay losses are severe, that this experimental set-up might not be able to identify nuclides with shorter half-lives and similar or lower production rate. The transport through the SISAK-apparatus is quite long as illustrated in Figure 4.6 in Section 4.3.

Single atom chemistry and detection with the current set-up of SISAK coupled to the BGS is optimum for nuclides of half-life of $\sim 4$ to 30 seconds. Detection of nuclides with longer half-lives can be achieved with additional detection cells, but a detection cell and the analog electronic modules are an expensive investment. Further development of the detection cell should result in a larger hold-up volume. Another feature interesting for a new detection cell design, is the possibility of $\alpha$-particle energy calibration during experiment without contaminating the cell. A detection cell of two identical (symmetrical) separate compartments or tracks, in which only one compartment is used for the calibration activity might be possible. It is important that a new design is as practical as the meander cell and preform as well.

The calibration of the SISAK detection system is improved with the real-time gain shift correction method presented in Paper III. The continuous recording of the Compton inflection point of $^{137}$Cs electron spectra throughout an experiment provides the means to track any changes in the scintillation-light output.

The SISAK detection system is developed for $\alpha$-particle spectroscopy, but the frequently unexplored decay of the transactinide nuclides is a challenge for an indubious analysis. Studies of the decay properties of heavy-element nuclides are in progress, and No and Lr isotopes have been subject to $\gamma$- and conversion electron spectroscopy. As discussed in Paper I conversion electrons in coincidence with $\alpha$-particle emission is a plausible explanation of broadening of $\alpha$-particle energy spectra. Summing effects have to be expected in the decay of other nuclides which are candidates for future SISAK experiments. In addition to summing events, Figures 4.5, 4.7, 4.9, and 4.14 contains some unexplained events of higher energy. This might be events originating from the natural background, or some kind of pile-up effect. It should certainly be subject for investigation, as also the liquid scintillation detection of spontaneous fission ($sf$) events in this system needs to be examined. Wierczinski et al. [92] reported that decay by spontaneous fission produced a signal around 15 MeV, in an earlier generation SISAK detection cell. If the signals from spontaneous fission decay are well understood, it should be possible to use $\alpha$-sf correlations to identify a nuclide.
5.2 Preseparation of transactinides before chemical studies with SISAK

Earlier SISAK extraction schemes developed for different transactinide elements, had to be concerned with the activity present from the decay of other reaction products. The selectivity of the extraction system for a certain transactinide was modeled by the system’s ability to separate the members of the periodic group. Physical preseparation of transactinide atoms/ions permits another approach. SISAK extraction schemes can be developed to focus on the chemical properties within one periodic group. For example, does Rf behave more like Zr or Hf.

It would be ideal to study a transactinide element when its homologs are produced and investigated at the same time. As the measurements in Section 4.1 indicate the uncertainty inherent in the aerosol/gas-jet technique might contribute to different experimental conditions even though the set-up is intact. When physical preseparation is employed, a simultaneous production of homologs is not attainable. Düllmann et al. [107] propose using a cocktail of heavy ions for production of the transactinide and its homologs. With the targets mounted on a ladder inside the target chamber, no parts of the experimental equipment have to be opened or changed when switching from production of one element to another.

Studies of higher Z transactinides with SISAK seem achievable in the near future when the BGS is employed as a preseaparator. Some nuclides are suggested in Table 5.1, produced in hot-fusion reactions using Pu-targets. Düllmann has calculated the energy of the recoil and its range in mylar [108], these numbers are also given in Table 5.1. The SISAK experiments performed so far have used a 6-µm thick mylar foil between the BGS and the RTC, to withstand the large pressure difference. As the numbers in Table 5.1 show, the suggested reactions produce recoils which will be stopped in a 6-µm foil. A modified degasser centrifuge to comply with a thinner foil between the RTC and the BGS would be necessary. The degasser centrifuge operates at atmospheric pressure, and the gas-jet pressure will vary typically from 1-3 atm, depending on the mixer and liquid flow rate. The pressure in the gas-jet could be lowered if the degasser centrifuge would, for example, draw the gas with suction.

Other isotopes of the transactinides in Table 5.1 are possible. Db can be produced with a cold-fusion mechanism, for example in the reaction $^{208}\text{Pb} (^{51}\text{V}, n)^{258}\text{Db}$. This reaction has a cross section of a few nb, and the 4.4-s half-life of $^{258}\text{Db}$ should be sufficiently long for SISAK conditions. Unfortunately, the decay of $^{258}\text{Db}$ is complicated, involving both α-particle emission and electron capture. Another pertinent reaction for $^{269}\text{Hs}$-production is $^{248}\text{Cm} (^{26}\text{Mg}, 5\text{n})^{269}\text{Hs}$ with 7 pb cross section. A SISAK experiment on Hs is not dependent on physical preseparation, because a volatile compound of Hs will be the basis of a chemical extraction scheme.
Table 5.1: Some transactinide nuclides that might be relevant for studies with the SISAK-system. Listed with a possible production reaction, the recoil energy of the EVR and its range in mylar. The recoil energy and range in mylar is calculated by Düllmann [108].

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>( T_{1/2} ) (s)</th>
<th>Reaction</th>
<th>( \sigma ) (nb)</th>
<th>( E_{\text{recoil}} ) (MeV)</th>
<th>Range (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{261}\text{m} \text{Rf})</td>
<td>4.2</td>
<td>(^{244}\text{Pu} \left( {^{22}\text{Ne}, 5n} \right) ^{261}\text{m} \text{Rf})</td>
<td>( \sim 4 ) nb(^a)</td>
<td>9.5</td>
<td>1.7</td>
</tr>
<tr>
<td>(^{262}\text{Db})</td>
<td>34</td>
<td>(^{244}\text{Pu} \left( {^{23}\text{Na}, 5n} \right) ^{262}\text{Db})</td>
<td>0.2-0.5 nb(^b)</td>
<td>10.8</td>
<td>1.9</td>
</tr>
<tr>
<td>(^{265}\text{Sg})</td>
<td>7.9</td>
<td>(^{244}\text{Pu} \left( {^{26}\text{Mg}, 5n} \right) ^{265}\text{Sg})</td>
<td>60-200 pb(^b)</td>
<td>13.5</td>
<td>2.4</td>
</tr>
<tr>
<td>(^{267}\text{Bh})</td>
<td>17</td>
<td>(^{244}\text{Pu} \left( {^{27}\text{Al}, 4n} \right) ^{267}\text{Bh})</td>
<td>2-10 pb(^b)</td>
<td>14.4</td>
<td>2.6</td>
</tr>
<tr>
<td>(^{269}\text{Hs})</td>
<td>14</td>
<td>(^{244}\text{Pu} \left( {^{30}\text{Si}, 5n} \right) ^{269}\text{Hs})</td>
<td>0.5-4 pb(^b)</td>
<td>18.1</td>
<td>2.9</td>
</tr>
</tbody>
</table>

\(^a\) Lazarev et al. [109]
\(^b\) Estimates by Düllmann [108]

5.3 Chemical separation of rutherfordium with SISAK

Three different extraction systems have been applied to study rutherfordium. Further investigations of the HNO\(_3\)/HDBP extraction system, showed sorption of group-4 elements on the PEEK surface for high nitric acid concentrations. The focus was directed towards identifying a more suitable system to study Rf, as undertaken by other members of the SISAK-group. In the oxalic acid/TOA scheme already used to extract Rf (Section 4.4), the homologs Zr and Hf exhibit near identical extraction behavior. Thus this system was left in favor of the sulfuric acid/TOA extraction system, in which the extraction of Zr and Hf clearly differs from each other.

The distribution coefficient proposed in Paper II is estimated from the total yield, represented by the activity measured separately in liquid scintillation solution. For a more precise measurement of the D-value, the activity in both outgoing phases after extraction should be detected. A SISAK experiment on \(^{257}\text{Rf}\) with aqueous phase detection implemented has recently (spring 2005) been conducted. The outgoing aqueous phase was fed into an additional mixer and centrifuge with an organic solution which extracted the activity contained in the aqueous phase. In this experiment \(^{257}\text{Rf}\) was extracted from sulfuric acid into TOA in toluene, similar to the experiment presented in Section 4.5. At low TOA concentrations Rf is expected to extract with a low yield from 0.5 M sulfuric acid, and during such a measurement the \(\alpha\)-particle decay of \(^{257}\text{Rf}\) was recorded in both liquid phases. A diagram is shown in Figure 5.1.
5.4 Outlook

Nuclides produced in cold-fusion reactions with enough recoil energy to pass a several $\mu$m-thick Mylar foil are relevant candidates for future experiments with the current SISAK equipment. Produced in hot-fusion reactions, nuclides are accessible if they can easily be separated from other reaction products and projectile particles by chemical means (for example Hs, as mentioned in Section 5.2). Production rates of most known transactinide nuclides of $Z > 105$ are today of an order that would correspond to a count rate in the SISAK detection system < 1 atom/3-4 days. Such low count rates demand a revision of the SISAK system.

A modified degasser would in fact imply considerable changes to the whole system, and might be the first step to a SISAK4 version. It would be desirable for a new version to run with smaller volumes, such as the MicroSISAK [110], which is under development and has not defined a detection system yet.

A possible direction to develop a new SISAK system is one in which the gas-jet is omitted. The recoils could be transported to the apparatus, perhaps on a rolling tape construction and then dissolved in liquid for chemistry experiments. This set-up would be a significant undertaking.

It needs to be discussed whether MicroSISAK and modifications of the current equipment - a possible SISAK4, are two separate ways of utilizing the original SISAK-system or if efforts should be joined.
Appendix A

List of acronyms and abbreviations

AKUFVE Arrangement for continuous investigations of distribution ratios
ADC Analog-to-Digital Converter
BGS Berkeley Gas-filled Separator
dimethyl-POPOP 1,4-Di-2-(4-methyl-5-phenyloxazolyl)-benzene, C$_{26}$H$_{20}$N$_{2}$O$_{2}$
EVR Evaporation Residues
GSI Gesellschaft für Schwerionenforschung
HDBP Di-butyl phosphoric acid, (C$_{4}$H$_{9}$)$_{2}$PO$_{4}$H
LBNL Lawrence Berkeley National Laboratory
LET Linear Energy Transfer
OCL Oslo Cyclotron Laboratory
PSI Paul Scherrer Institut
PMT Photo-Multiplier Tube
PSD Pulse-Shape Discrimination
RTC Recoil-product Transfer Chamber
SISAK Short-lived Isotopes Studied by the AKUFVE-technique
TOA Tri-n-octylamine, (C$_{8}$H$_{17}$)$_{3}$N
Appendix B

Photos of equipment

Figure B.1: The chemistry area at LBNLs 88-Inch Cyclotron during a SISAK experiment with the transactinide nuclide $^{257}$Rf. The SISAK degassers and centrifuges mounted in the white rack in the middle of the hood. The detection cells, to the far right in the picture, are set-up in an array of lead bricks. Photo of Jon Petter Omtvedt
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[84] National Instruments Corporation, 11500 N Mopac Expwy, Austin, TX 78759-3504, USA, LabVIEW.


REFERENCES


[96] LeCroy Corporation, 700 Chestnut Ridge Road, Chestnut Ridge, NY 10977-6499, USA, PXD 222 DIGITIZER.


[102] Borland Software Corporation, 100 Enterprise Way, Scotts Valley, CA 95066-3249, USA, Borland Delphi programming software.


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