Nuclear Chemistry Progress Report

Oregon State University

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I. Summary

In this report, we summarize the highlights of the work done between August 1, 2003, and August 1, 2004, that was supported by USDOE Grant No. DE-FG06-97ER41026. The work reported herein is the result of a collaborative effort between the nuclear chemists at Oregon State University and many other individuals and research groups. Each project discussed was the result of a joint effort of the groups, interchanging roles in data acquisition and analysis. The individuals contributing to each project are listed at the end of each section with the names of the Oregon State scientists being underlined. Some of the work reported here is in its preliminary stages and use of the data contained in the preliminary reports should be made only after consultation with the appropriate authors. Up-to-date versions of many of these reports and new information can be found on our Web page at http://www.orst.edu/dept/nchem.

The work described is part of a project involving the study of low energy (10 MeV/nucleon), and intermediate energy (10-100 MeV/nucleon) heavy ion reactions.

During the past year, most of our effort has gone toward performing four major experiments with one each at LBNL, ANL, MSU and ORNL with collaborative participation in two additional experiments at ORNL. Data analysis is still in progress for three of these experiments (ANL, MSU and ORNL) that should serve as important parts of the Ph.D. theses of students in our group. The analysis of the data for the LBNL experiment has largely been completed and despite an exhaustive search, we are unable to confirm the production of $^{283}_{112}$ (which is now taken to be a 4 s alpha emitter) in the $^{48}_{20}$Ca + $^{238}_{92}$U reaction. (Our two previous unsuccessful searches for a 3-5 min SF activity for $^{283}_{112}$ have been validated, in that, the Dubna laboratory has essentially retracted the reports of the synthesis of this activity). The problem of confirming the formation of $^{283}_{112}$ remains an important one for heavy element studies as it represents one of the few ways to confirm the “hot fusion” studies that have led to the formation of elements 113,114,115,116 and 118.

With LBNL no longer being a national user facility, we have switched our emphasis to study of fusion reactions induced by radioactive beams (at ORNL and MSU) along with continuing our studies of heavy nuclei at ATLAS. As part of this emphasis, we have re-examined the role of radioactive beams in the synthesis of new heavy nuclei, building on previous efforts. We have extended our modeling of these reactions to include the best available representations of the capture cross section, the $P_{\text{CN}}$ factor, and the survival probability. In calculating the latter, we include the effects of collective enhancement and its energy dependence, energy dependent shell corrections and the best global representations of fission barriers and binding energies.

Of special note is the expanded student interest in nuclear chemistry and our research group. We now have four full-time grad students and one post-doc in addition to the usual summer visitors, etc.

II. Low Energy Heavy Ion Research

A. Search for the Production of Element 112 in the $^{48}_{20}$Ca + $^{238}_{92}$U Reaction
In 1999, a Dubna-GSI-RIKEN collaboration [1] reported the successful synthesis of
$^{283}\text{112}$ using the reaction $^{48}\text{Ca} + ^{238}\text{U} \rightarrow ^{286}\text{112} \rightarrow ^{283}\text{112} + 3\text{n}$ with the observation of
two events. The nuclide $^{283}\text{112}$ ($t_{1/2} = 81^{+147}_{-32} \text{s}$) was reported to decay by spontaneous fission
(SF) and was produced with a cross section of $5.0^{+3.3}_{-3.2} \text{ pb}$. The decay mode of $^{283}\text{112}$ is somewhat unexpected as all the other isotopes of element 112 (A=277,284 and 285) decay by alpha emission. The Dubna-GSI-RIKEN collaboration searched for alpha decay in $^{283}\text{112}$ but could not see any events. Subsequently, in the reaction of $^{48}\text{Ca}$ with $^{242}\text{Pu}$, two events were found in which an evaporation residue (EVR) emitted an alpha-particle, producing a daughter nucleus that decayed by SF.[2] These latter SF decays were attributed to the decay of $^{283}\text{112}$ and if taken with the previous work, imply a half-life of $\sim 3 \text{ m}$ for $^{283}\text{112}$. These experiments were done with the VASSILISSA velocity filter separator.

The nucleus $^{283}\text{112}$ and its synthesis play an important role in our understanding of the recent syntheses of elements 114 and 116 by hot fusion reactions [3-5]. $^{283}\text{112}$ is directly populated in the de-excitation of $^{287}\text{114}$ synthesized using the $^{48}\text{Ca} + ^{242}\text{Pu}$ reaction [2]. The long half-life is typical of elements 112 and 114 nuclei produced in the synthesis of elements 114 and 116 [3-5]. The relatively large reported production cross section, 5 pb, is typical of the higher cross sections associated with hot fusion reactions compared to cold fusion reactions for the synthesis of $Z \geq 112$. It is these same cross sections, which challenge our understanding because current theoretical predictions of the survival probabilities in these reactions [6] would not give cross sections of this magnitude. For example, Armbruster [7] using the best available data on the capture cross sections, the probability of evolving from the contact configuration to the completely fused system and the survival probabilities, estimated a evaporation residue production cross section for the reaction of 231 MeV $^{48}\text{Ca} + ^{238}\text{U} \rightarrow ^{286}\text{112} \rightarrow ^{283}\text{112} + 3\text{n}$ of 50 fb.

Previously [8] in 2001, we studied the reaction $^{238}\text{U}(^{48}\text{Ca},3\text{n})$ at the 88-Inch Cyclotron of the Lawrence Berkeley National Laboratory, using the Berkeley Gas-filled Separator (BGS). The details of the experimental setup, its calibration and the tests made to be sure the system was operating properly have been described elsewhere. [8]. Two search strategies were used to look for events corresponding to the implantation and decay of $^{283}\text{112}$ nuclei. The first strategy assumed the decay of $^{283}\text{112}$ would occur in accord with the predictions of Smolanczuk [9]. We searched for EVR-$\alpha$, $\alpha$-$\alpha$ and EVR-fission events occurring within 6 s, restricting the range of $\alpha$-particle energies to be from 8 to 11 MeV and the single fragment fission energies to be $\geq 90$ MeV. (This latter limit was chosen to include 96% of the expected single fragment kinetic energy distribution assuming the SF single fragment kinetic energy distributions have similar shapes for $^{252}\text{No}$ and $^{283}\text{112}$.) No events were observed with a total dose of 1.1x10$^{18}$ ions. This corresponds to a one-event upper limit cross section of 1.8 pb for $^{283}\text{112}$ nuclei decaying by alpha-particle emission and 1.6 pb for spontaneously fissioning $^{283}\text{112}$ nuclei when one takes into account the differing efficiencies of detecting fission fragments and alpha-particle decay chains.

A second strategy involved searching for events similar to those reported by the Dubna-GSI-RIKEN group. [1]. We searched for EVR-$\alpha$, $\alpha$-$\alpha$, and EVR-fission events occurring within 1000 s, using the same energy restrictions as in the first search. No EVR-fission events were found, leading to a one event upper limit cross section of 1.6 pb for the type of event reported by the Dubna-GSI-RIKEN group or any chain terminating in an SF decay.
Because of the importance of this measurement and its implications, during the year 2002, we made another experimental search for the production of $^{283}\text{112}$ in the reaction $^{231}\text{MeV } ^{48}\text{Ca} + ^{238}\text{U} \rightarrow ^{286}\text{112} \rightarrow ^{283}\text{112} + 3\text{n}$. The setup was similar to that used in the previous search. In our calibration of the BGS and our detector system, we measured the cross section for the 215.5 MeV $^{48}\text{Ca} + ^{206}\text{Pb} \rightarrow ^{252}\text{No} + 2\text{n}$ reaction by detecting the SF decay (SF branching ratio of 0.322) of $^{252}\text{No}$. We measured a cross section of $470 \pm 80 \text{nb}$, in agreement with the known value of $515^{+80}_{-47} \text{nb}$, thus demonstrating our ability to detect known SF activities with our apparatus. The total particle dose in the experiment was $1.16 \times 10^{18}$ which leads to an upper limit cross section of $1.9 \text{ pb}$ for this run. Combining these data with the previous measurements [8], leads to overall one event upper limit of $0.8 \text{ pb}$ for production of $^{283}\text{112}$ in the reaction $^{231}\text{MeV } ^{48}\text{Ca} + ^{238}\text{U} \rightarrow ^{286}\text{112} \rightarrow ^{283}\text{112} + 3\text{n}$.

After completion of this experiment, we were informed [10] of an attempt, prompted by our earlier report [8], of the Dubna VASSILISSA group to reproduce their previous measurement. The Dubna group was not successful either in reproducing their earlier measurement, obtaining an upper limit of $2.3 \text{ pb}$ (68% confidence level) for the production of $^{283}\text{112}$ in the reaction $^{231}\text{MeV } ^{48}\text{Ca} + ^{238}\text{U} \rightarrow ^{286}\text{112} \rightarrow ^{283}\text{112} + 3\text{n}$. However, when this group increased the beam energy from 230-231 MeV (the energy given in the original publication [1]) to 234 MeV, they found two SF events which they attributed to the decay of $^{283}\text{112}$. The total dose in this second experiment was $4.68 \times 10^{18}$ ions and corresponds to a cross section of $3.0^{+4.0}_{-2.0} \text{ pb}$.

The situation became more complicated when it was reported that the Dubna Gas-Filled Recoil Separator (DGFRS) group [11] was unable to repeat the observations of the VASSILISSA group but did identify $^{283}\text{112}$ as a $6.1^{+7.2}_{-5.2} \text{s}$ alpha emitter with $E_{\alpha} = 9.54 \pm 0.07 \text{ MeV}$. This nucleus was observed as a daughter or decay product in the decay of $^{287}\text{114}$ and $^{289}\text{116}$. Recently [12] the DGFRS group has reported the direct production of this nucleus in the $^{48}\text{Ca} + ^{238}\text{U}$ reaction with the observed excitation function shown in Figure II-A-1. The best current value of the half-life of $^{283}\text{112}$ (as observed by the DGFRS group) is $4.4^{+1.3}_{-0.7} \text{s}$ with $E_{\alpha} = 9.54 \pm 0.06 \text{ MeV}$. The spontaneous fission branch is assigned a probability of $< 10\%$.

\[ \text{Figure II-A-1} \quad \text{Excitation function for the } ^{238}\text{U}(^{48}\text{Ca},3\text{n})\text{reaction. From [12].} \]

In view of these developments, we made yet another attempt (2004) to observe this activity or any product in the reaction of $^{48}\text{Ca}$ with $^{238}\text{U}$. The experimental setup was similar to
that used in the two previous attempts with some important differences. These differences were:

(a) the size of the focal plane detector was increased from 32 strips to 48 strips, increasing the geometrical efficiency of detecting recoils from 45% to 56%. (b) a beam shutoff system was employed that turned off the beam for eleven half-lives of $^{279}$Ds after a recoil-alpha event had occurred, allowing us to measure EVR-\(\alpha\)-\(\alpha\) correlations and EVR-\(\alpha\)-SF events. (The $^{279}$Ds daughter of $^{283}$112 is an $^{18}_{-0.03}^{+0.05}$ s nucleus that decays 90% by SF and 10% by the emission of a $9.70 \pm 0.06$ MeV \(\alpha\)-particle. (c) the center of target beam energy was 236.5 MeV, near the peak of the $^{238}$U($^{48}$Ca,3n) excitation function (Figure II-A-1) measured by the DGFRS group. (A point worth noting is that the DGFRS group typically reports their results in terms of excitation energies calculated using Thomas-Fermi masses [13] and using energy loss estimates from the tables of Hubert et al. [14] (solids) and Northcliffe and Schilling [15] (gases). (d) a multiwire avalanche counter was used in place of a PPAC to provide time and position signals for the recoils, scattered beam, etc. upstream from the focal plane detector (e) the magnetic rigidity of the BGS was set to 2.21 T-m (rather than the 2.25 Tm used previously) to reflect our best knowledge of the systematics of B$^\rho$ settings of the BGS for similar reactions. UF$_4$ targets deposited on 0.58 mg/cm$^2$ Al foils were used as targets with thicknesses varying from 360 to 440 \(\mu\)g/cm$^2$ of $^{238}$U. The integrated target atom x ion dose was $1.74 \times 10^{36}$. No events corresponding to those reported by the DGFRS group were observed. Correcting for the separator efficiency gives a one event upper limit cross section of 0.96 pb, or an upper limit at the 84% confidence level of 1.8 pb.

The current situation is summarized in Figure II-A-2. All uncertainties shown are 84% confidence limits. Formally, at the 84% confidence level, the individual DGFRS and the LBNL measurements overlap. If one combines the two LBNL measurements, by, in effect, pretending the two beam energies corresponded to a single irradiation with a broad energy distribution, one gets a one event upper limit cross section of 0.5 pb with the 84% upper limit being 0.9 pb. An overall view of Figure II-A-2 demonstrates the difficulties of trying to establish whether a phenomenon exists at this low cross section level. The 2004 LBNL measurements correspond to a $^{48}$Ca dose of $\sim 1.5 \times 10^{18}$ ions. The DGFRS measurements correspond to an integrated dose of $\sim 1.8 \times 10^{19}$ ions, about an order of magnitude greater. To resolve the discrepancies, real or apparent, between these measurements may require significant increases in beam time and delivered ion dose.
Figure II-A-2 Summary of our results and those of the DGFRS group for the \( ^{238}\text{U}(^{48}\text{Ca,3n}) \) reaction. Upper limits (84% confidence level) are denoted by single-sided error bars with the “one-event” limits shown as points.


References

12 J.B. Patin (private communication).
B. Synthesis of Heavy Nuclei Using RNBs

In new radioactive beam facilities, such as the RIA facility, the issue arises as to the potential for synthesizing new heavy nuclei using radioactive nuclear beams (RNBs). While it is tempting to write several potential synthesis reactions leading to very interesting products, we believe that one must make quantitative estimates of the realistic probability of such syntheses. Accordingly, we have been engaged for several years [1,2] in attempts to make these calculations, trying, with each succeeding attempt, to reduce the uncertainties in the calculations and to improve their predictive power. We report here another effort aimed at improving these calculations.

The rate of production of heavy nuclei at a radioactive beam facility is simply
\[ N \sigma \Phi \]
where \( N \) is number of target nuclei, \( \sigma \) the evaporation residue production cross section and \( \Phi \) the beam intensity. Our approach to evaluating production rates is to make brute force calculations in which we calculate the reaction rates (in atoms/s) for reactions involving all possible combinations of projectile and target nuclei that might be available at a facility like RIA. The beam energies are adjusted to give the maximum yield of the desired product. We do a preliminary sort of these results, identifying those reactions with rates exceeding 1 atom/100 years. These candidate cases are more carefully evaluated using a modified version of HIVAP[3].

In calculating the luminosity, \( N\Phi \), we assume the available targets are all stable nuclei (with a target thickness of 1.0 mg/cm\(^2\)) and all longer-lived actinide nuclei (with a thickness of 0.5 mg/cm\(^2\)). For the list of projectiles, we used the RIA list [4], that gives the identity and intensity of all the expected RNBs that have suitable energies (< 15 A MeV). (We have corrected the beam list to take into account the RIA 400 kW driver power and have made decay corrections for the yields of the lighter nuclei [5]).

In setting up the luminosity calculations, one gets a simple physical insight into the feasibility of producing trans-roentgenium nuclei with radioactive beams. Assume for the sake of argument, that the production cross section for such nuclei is \( \approx 5 \text{ pb} \) and that a production rate of 1 event/month is needed for a viable synthesis study. Then, at a modern separator, one needs a beam intensity of \( > 5 \times 10^{10} \text{ particles/s} \). This requirement generally restricts one to using RNBs that are \( < 5 \) neutrons from stability [4].

The cross section for the production of an evaporation residue, \( \sigma_{EVR} \), can be written as
\[ \sigma_{EVR} = \sigma_{CN} W_{\text{sur}} \]
where \( \sigma_{CN} \) is the complete fusion cross section, and \( W_{\text{sur}} \) is the survival probability of the completely fused system. The complete fusion cross section can be written as
\[ \sigma_{CN} = \sum_{J=0}^{J_{\text{max}}} \sigma_{\text{capture}} (E^*,J) P_{CN} (E^*,J) \]
where \( \sigma_{\text{capture}} \) is the capture cross section and \( P_{CN} \) is the probability that the projectile-target system will evolve, inside the fission saddle point, to form a completely fused system. \( P_{CN} \) takes on values from 0 to 1.

The capture cross section can be given in a simplified form [6] as
\[ \sigma_{\text{capture}} = \pi \lambda^2 (2J + 1) T(E_{\text{cm}},J) \]
where $\lambda$ is the reduced wavelength, $T$ the transmission coefficient for a projectile with center of mass energy $E_{cm}$ and forming a system with angular momentum $J$. In a common approximation [6], this expression is simplified to be $\pi \lambda^2 \ell_{lm}^2$ where $\ell_{lm} = 15\hbar$, thus making the capture cross section formally independent of $J$. Alternatively Siwek-Wilczynska, et al. [7] have provided a semi-empirical formulation for the capture cross section that has a specific isospin dependence. We have shifted our calculations to the Siwek-Wilczynska et al. formalism instead of the simple $\pi \lambda^2 \ell_{lm}^2$ formalism used previously [1,2].

We have used the Armbruster formalism for $P_{CN}$ [6]

$$P_{CN}(E_{cm},J) = 0.5 \left [ \exp \left ( x_{eff} - x_{thr} \right ) \right ]$$

where the coefficient $c$ has the value of 106 and the constant $x_{thr}$ is 0.72 for actinide-based reactions and 0.81 for reactions involving Pb or Bi targets. The calculation of the effective fissility, $x_{eff}$, of the fused system is described in [6].

The survival probability $W_{sur}$ can be written as

$$W_{sur} = P_{sn}(E^{*}, J) \prod_{i=1}^{n} \frac{\Gamma_{n}}{\Gamma_{n} + \Gamma_{f}} e^{kE^{*}}$$

where the index $i$ is equal to the number of emitted neutrons and $P_{sn}$ is the probability of emitting exactly $x$ neutrons [8]. For calculating $(\Gamma_{n}/\Gamma_{f})$, we have used the classical expression from Vandenbosch and Huizenga [9]

$$\frac{\Gamma_{n}}{\Gamma_{f}} = \frac{4A^{2/3}(E^{*}_{cn} - B_{n})}{k(2(a(E^{*}_{cn} - B_{n}))^{1/2} - 1)} \exp \left ( 2a^{1/2} \left ( E^{*}_{cn} - B_{n} \right )^{1/2} - \left ( E^{*}_{cn} - B_{n} \right )^{1/2} \right )$$

The constants $k$ and $a$ are taken to be 9.8 and $A/12$, respectively. The fission barriers, $B_{f}$, are written as the sum of liquid drop and shell correction terms as

$$B_{f}(E^{*}_{cn}) = B^{LD}_{f} + U_{shell} \exp \left ( \frac{-E^{*}_{cn}}{E_{D}} \right )$$

where the shell corrections to the liquid drop barriers are taken from [10] and the fade-out of the shell corrections from Ignaytuk et al. [11]. Neutron binding energies are taken from [10]. Collective enhancement effects are only important for spherical product nuclei and they are calculated using the semi-empirical formalism of [6].

The success of the primary sorting code can be evaluated by comparing the results of known synthesis reactions and the predictions. In Figure II-B-1, we show a comparison of the calculated and measured cross sections [12] for the synthesis of elements 102-112 using cold fusion reactions. The model calculations and measurements generally agree within an order of magnitude (and usually better) over several orders of magnitude. Since the overall trend of these data reflects the effect of fusion hindrance, one gains some confidence in our treatment of this factor.

In Figure II-B-2, we show similar plots for hot fusion reactions where (a) there is no expected fusion hindrance ($Z_1/Z_2 < 1200$) and (b) $^{48}$Ca induced reactions where there is fusion hindrance. The agreement between prediction and measurement is nearly not so good although the gross trends are reproduced. The reproduction of the gross trends, i.e., the change in slope of the dependence of the cross sections upon fissility for the $^{48}$Ca based reactions, is not trivial. This is the first time we have been able to reproduce this trend and the “success” is due largely to the use of a particular model [10] for the fission barrier heights and neutron binding energies. Further work is needed to better define the measurements, which themselves show a large
scatter and the calculations.

In Figure II-B-3, we show the predicted production rates for No isotopes using stable and radioactive beams and using hot or cold fusion reactions. Focusing on cold fusion reactions, one sees that stable beams are superior to radioactive beams for the synthesis of most isotopes although the radioactive beams do allow one to reach n-rich isotopes not accessible with stable beams. A similar situation occurs with hot fusion reactions although the n-rich isotopes produced with RNBs have substantial yields.

In Figure II-B-4, we show the same comparisons for the production of Sg isotopes. Now the superiority of the RNBs for making n-rich nuclei is more evident along with the general superiority of hot fusion over cold fusion reactions. Table II-B-1 shows typical examples of the types of hot fusion reactions with RNBs that yield new n-rich isotopes of Sg. The projectiles, as remarked earlier, typically have about five neutrons more than the most n-rich stable isotope. Also shown is a typical cold fusion reaction that reaches into this region and the associated low production rate.

In Figure II-B-5, we show the situation for the synthesis of isotopes of element 112. The RNB induced reactions have too low rates to be useful. Figures II-B-6 and II-B-7 confirm that view for elements 114 and 118.

While we will continue to improve these estimates, we conclude that RNBs do not provide a path to new heavy elements, but are a path to new n-rich heavy nuclei.

(W. Loveland and Z. Evenson

References

2. W. Loveland, Nucl Phys. A (accepted for publication)
5. J. Nolen, private communication
10. P. Moller et al., At Data Nucl, Data Tables 39, 213; 59, 185.
12. The experimental data is from a compilation by K.E. Gregorich and Ch. E. Duellmann.
Figure II-B-1 Comparison of measured and calculated cross sections for the production of elements 102-112 by cold fusion reactions
Hot fusion reactions
(no fusion hindrance)
Figure II-B-2 Comparison of measured and calculated heavy nucleus production rates for hot fusion reactions involving no fusion hindrance and $^{48}\text{Ca}$ induced reactions
Figure II-B-3  Comparison of hot and cold stable beam and RNB induced syntheses of the isotopes of element 102.
Figure II-B-4 Comparison of hot and cold stable beam and RNB induced syntheses of the isotopes of element 106.
Figure II-B-5 Comparison of hot and cold stable beam and RNB induced syntheses of the isotopes of element 112.
Figure II-B-6  Comparison of hot and cold stable beam and RNB induced syntheses of the isotopes of element 114.
Figure II-B-7 Comparison of hot and cold stable beam and RNB induced syntheses of the isotopes of element 118.

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C. Fusion Enhancement in the $^{46}$Ar + $^{124}$Sn Reaction

One of the interesting aspects of the study of nuclear reactions induced by radioactive beams is the possibility of using n-rich radioactive projectiles to synthesize new, n-rich heavy nuclei [1]. It has been shown [1] that new areas in atomic physics and chemistry of the transactinide elements could be developed using intense n-rich radioactive beams.

Various authors [2-4] have suggested that there will be significant enhancements to the fusion cross sections for n-rich projectiles due to a lowering of the fusion barrier and the possible excitation of the soft dipole mode. They have further speculated that the use of these projectiles might lead to the successful synthesis of new or superheavy nuclei, a thought that is echoed in the Long Range Plan for Nuclear Science [5]. We have been engaged in a project to make measurements of the fusion enhancement factors for n-rich radioactive projectiles of possible interest in the synthesis of new heavy nuclei. This experiment represents an extension of that effort to remedy deficiencies in previous experiments and tries to extend the study to more n-rich nuclei, with the further addition of shell-stabilized projectile and target nuclei.

Previously [6] we reported the results of study of fusion enhancement in the reaction of $^{32,38}$S with $^{181}$Ta. (Figure II-C-1) Complete fusion cross sections were deduced from capture-fission cross sections, after large (~50%) corrections were applied for fission events where the interacting nuclei did not go inside the true fission saddle point (quasifission) but reseparated after exchanging mass and energy. These corrections were based on the expected differences [7] between the fragment angular distributions for true fusion-fission events and the quasifission events. The corrections were unexpectedly large given the atomic numbers of the interacting nuclei and have been questioned in subsequent work [8]. The overall study concluded that the fusion enhancement seen with the $^{38}$S projectile was due to a lowering of the fusion barrier of 5.9±0.4 MeV in going from $^{32}$S to $^{38}$S in rough agreement with the systematics of the isospin dependence of the fusion barrier height [9-13]. No evidence was found for any
enhancement apart from this simple shift in barrier height. Subsequently, a study of fusion enhancement in the $^{32,38}$S + $^{208}$Pb reaction showed a very different pattern (Figure II-C-2). A huge shift in barrier height (15±9 MeV) was observed between the two reactions and the reduced excitation functions for the two reactions were quite different (Figure II-C-2).

Apparently the shell stabilized $^{208}$Pb target (as compared to the deformed $^{181}$Ta) either polarized the $^{38}$S or caused some other unusual fusion enhancement.

What we have done as a logical extension of the earlier work is to study fusion enhancement with n-rich projectiles in a system ($^{46}$Ar + $^{124}$Sn) where the principal reaction channel is the formation of evaporation residues (according to PACE simulations [14] (Figure II-C-3)), thus eliminating the ambiguity in the previous work of distinguishing between compound nucleus fission and quasifission. In addition, we increase the relative neutron richness of the projectile (N/Z = 1.56 vs 1.38) and the composite system (N/Z =1.50 vs 1.46).

To simulate the oft-repeated suggestion [5] that “using beams of neutron-rich medium mass radioactive nuclei on Pb or Bi targets” might be useful in heavy element synthesis, we use a reaction where the projectile has medium mass (and is magic) along with a magic target nucleus (which produced an unusual result for $^{38}$S induced reactions). We pick a lighter compound system and restrict our attention to energies below $E_{cm} = 140$ MeV to insure that the total fusion cross section is given by the evaporation residue cross section (Fig. II-C-3) and that reasonable counting rates can be obtained with the expected low intensities of near barrier radioactive beams. (In going from the $^{32,38}$S + $^{181}$Ta system to the $^{36,46}$Ar + $^{124}$Sn system, we also go from a deformed target nucleus to a spherical one and to a system with positive Q values for two neutron transfer. These changes may affect sub-barrier fusion but only limited exploration of these questions is possible with the CCP RNB intensities).

This experiment is similar to the previous experiments in general methodology. A 90 MeV/A $^{48}$Ca beam was fragmented in a Be production target in the A1900 fragment separator at the NSCL CCF at MSU. After passage through an achromatic wedge, degrader and momentum defining slits (to assure high beam purity) in the A1900, the beam was transported to the N3 area. A typical “on-target” beam intensity of 1000-5000 particles/s was obtained. A diagram of the experimental apparatus is shown in Figure II-C-4. The ~25 MeV/A $^{46}$Ar beam is degraded to 140-185 MeV by an Al degrader wheel that was remotely controlled. The straggling of the beam was ~8 \% (FWHM) after degradation to 170 MeV. The degraded beam
passed through a set of microchannel plate detectors and PPACs separated by 96 cm wherein the
time of flight of each beam particle was measured.

The evaporation residues (predicted PACE spectra and angular distributions are shown
in Figure II-C-5) were detected in a two element (25 µ and 1000 µ) silicon telescope placed near
0°. The detector subtended ~2° around the beam axis and was ~50 cm from the target. The
center of mass energy of the beam, E_{cm} was < 140 MeV, insuring that the fusion cross sections
is adequately represented by the evaporation residue cross section (Fig. II-C-3).

Five settings of the Al degrader were used to give primary 46Ar energies from 140 - 185
MeV. Analysis of these data is in progress. The measurement of the 36,40Ar + 124Sn excitation
function is to be made at a stable beam facility in a separate experiment.
(W. Loveland, D. Peterson, D.J. Morrissey, F. Liang, D. Shapira)

References

1. W. Loveland, in Proceedings of the Third International Conference on Radioactive
   Nuclear Beams, edited by D.J. Morrissey (Editions Frontieres, Gif-sur-Yvette, 1993), pp
   526-536.
   C46, R45 (1992).)
14. A. Gavron, in Computational Nuclear Physics 2, K. Langanke, J. Maruhn, and S.E.
Figure II-C-1. Reduced excitation functions for the S + Ta reactions. No evidence is seen for a change in the barrier shape between the two reactions.

Figure II-C-2. Reduced excitation function for the \( ^{32,38}S + ^{208}Pb \) reaction.
Figure II-C-3. PACE simulations of the $^{46}\text{Ar} + ^{124}\text{Sn}$ reaction

Figure II-C-4. Schematic diagram of the experimental apparatus.
D. Fusion Hindrance in the $^{132}$Sn + $^{90,96}$Zr Reaction

There have been several studies [1-11] of the effect of varying the isospin of the projectile and target nuclei upon the fusion cross section. In some of these studies [6], the effect of the changing isospin was a simple shift in the fusion barrier with similar reduced excitation functions for the systems under study. For reactions at sub-barrier energies, however, more complicated nuclear structure effects appear to play a role. These have been described [1,2,5,7-11] in terms of inelastic excitations of the projectile and target nuclei and the influence of transfer reaction channels. Alternate explanations [12,13] use a macroscopic description of neck formation or neutron flow to explain the observed effects of N/Z upon the fusion process.

One of the first reported studies of the effect of using radioactive n-rich medium mass projectiles to produce heavy nuclei was the work of our group [14], that reported fusion excitation functions for the $^{32,38}$S + $^{181}$Ta reaction. That study concluded that the use of the n-rich projectile led to a simple lowering of the fusion barrier ($\Delta V_B \sim 5.9$ MeV) with the reduced excitation functions for the two reactions being identical within experimental uncertainties. An extension of that study by our group to the $^{32,38}$S + $^{208}$Pb reaction has indicated a very large shift in the fusion barrier ($\Delta V_B \sim 12$ MeV) for the n-rich projectile compared to the stable projectile when the target nucleus is the shell stabilized $^{208}$Pb.

This experiment is an extension to this previous work. We extended the measurement of fusion enhancement factors relevant to the synthesis of heavy nuclei to study the fusion of a massive neutron-rich shell-stabilized projectile, $^{132}$Sn, with $^{90,96}$Zr. This extension of the previous work involves, for the first time, a system in which fusion hindrance is expected to be significant. This is important because fusion hindrance factors have played a role in the synthesis of all the transactinide nuclei [15]. Previous studies [16,17] of the isospin dependence of fusion hindrance in the related $^{90,96}$Zr + $^{124}$Sn reaction have shown, contrary to theoretical
expectations [18-23] that the fusion hindrance is more for more n-rich projectile, thus canceling some of the advantage of the n-rich projectile. Alternate explanations have attributed this effect to the shell stabilization of $^{90}$Zr compared to $^{96}$Zr. This study will test that suggestion by making the most n-rich species shell-stabilized.

The reaction chosen for study $^{96}$Zr ($^{132}$Sn,X) has been chosen for a variety of reasons. The radioactive projectile, $^{132}$Sn, is as neutron-rich (N/Z =1.64) as any radioactive beam available today and is significantly more n-rich than any stable beam. The observed beam intensity of ~10-15000 particles/s is high for the current generation of radioactive beam facilities, allowing fusion cross sections of ~10 mb to be measured with ± 30% uncertainty in a period of ~24 hours. (Detailed examination of fusion barrier distributions or sub-barrier fusion or detection of low yield evaporation residues is not possible with this beam intensity). The "speed limit" imposed by the acceleration of the beam in the Tandem is ~625 MeV (assuming foil-foil stripping) which is adequate (see below) to sample the expected fusion excitation function. $^{132}$Sn is doubly magic and can be used to test the role of nuclear structure on fusion hindrance with massive n-rich projectiles. The stable beam analog reactions $^{124}$Sn ($^{90,96}$Zr,X) have already been measured [23,24] allowing comparisons to the radioactive beam measurement.

To simulate the proposed reaction, we have used the code HIVAP [24] that is a semi-empirical representation of what we know about fusion and fusion hindrance in the reactions involving heavy nuclei. In Figure II-D-1, we show the predicted fusion excitation functions (along with the fusion-EVR excitation functions) for the $^{96}$Zr ($^{124,132}$Sn,X) reactions. We see that for the energies of interest (E$_{Sn}$ = 520-625 MeV) the compound nuclei decay mostly by fission with a small (~10 mb, i.e., ~10%) roughly constant EVR component. The HIVAP prediction, based upon previous results [16,17], is that the fusion hindrance associated with the very n-rich projectile will approximately cancel the expected advantages of a lowered fusion threshold. But that is, of course, what we are trying to measure, i.e., to see if the fusion hindrance effects seen previously were the result of having shell stabilized projectiles or more n-rich projectiles.

The experiment was performed at the HRIBF at ORNL during April-May, 2004. A schematic diagram of our experimental apparatus is shown in Figure II-D-2. The radioactive beam passed through a set of microchannel plate detectors (MCPs), separated by about 1 m. The efficiency of this setup has been measured to be 99.9% [25] and it has operated at rates that are 10x those used in this experiment. The time of flight of each beam particle is measured. The fission fragments were detected in an Si strip detectors placed at forward angles. The use of inverse kinematics focuses the reaction products forward, with the expected full momentum transfer fission fragment folding angle being ~80°. Simulations indicate an expected efficiency of ~35% for this geometry. Valid fission events were defined as coincidences between detected fragments with a folding angle corresponding to full momentum transfer. An ion chamber [25] was placed in the beam to monitor the energy and number of beam particles. The efficiency of the detector setup was measured using a stable $^{124}$Sn beam by comparison with existing data [23,24] at several projectile energies.

Measurements were made at ten different energies between 300 and 600 MeV for the $^{124,132}$Sn + $^{90,96}$Zr reactions, which should adequately define the fusion-fission excitation function (Figure II-D-1). Analysis of these data is in progress.

References

The cross section for producing a heavy nucleus in a heavy ion reaction can be expressed as the product of three factors, i.e.

$$\sigma_{ER} = \sigma_c \cdot P_{CN} \cdot W_{sur}$$

where $\sigma_c$ is the capture cross section, $P_{CN}$ the probability of compound nucleus formation after capture and $W_{sur}$ is the survival probability of the excited nucleus. For hot fusion reactions (combinations of lighter projectiles with actinide target nuclei producing compound nuclei excited to 50-60 MeV), the most important term in this expression is $W_{sur}$. For cold fusion reactions (combinations of heavier projectiles with Pb-Bi target nuclei producing compound
nuclei excited to 10-15 MeV), the P\textsubscript{CN} term is the most important term.

**What this experiment deals with is the study of a typical cold fusion reaction, \(^{208}\text{Pb}\ (^{50}\text{Ti}, \text{n})^{257}\text{Rf}, in which we attempt to determine P\textsubscript{CN}.** This reaction has been studied before [1-6]. The evaporation residue cross section, \(\sigma\text{_{ER}}\), is \(10 \pm 1\) nb [6], at the maximum of the excitation function. The capture cross section, \(\sigma\text{_{c}}\), has also been measured for several projectile energies [1]. In a related nuclear structure issue, the survival probability, \(W\text{_{sur}}\), for this reaction has been calculated [7] using two different theoretical prescriptions for the fission barriers involved [8,9]. The resulting survival probabilities for this case (and several similar cases) differ by an order of magnitude (\(W\text{_{sur}} = 10^{-3}\) [8], \(W\text{_{sur}} = 10^{-4}\) [9]). Thus a determination of P\textsubscript{CN} will establish an experimental value of \(W\text{_{sur}}\) that can be compared to divergent theoretical predictions. This question is of interest, to the heavy element community, because of the highly divergent predictions for the survival probabilities involved in using cold fusion reactions to synthesize new heavy nuclei. (Figure II-E-1)

![Figure II-E-1](image.jpg)

**Figure II-E-1. Calculated survival probabilities for cold fusion reactions.**

Questions such as the possibility of success in further heavy element synthesis are strongly tied to understanding these survival probabilities.

To determine P\textsubscript{CN} experimentally, we did an experiment similar to that of Keller, et al., previously done at ANL where this experiment was performed at the ATLAS accelerator facility in June, 2004. In this sort of experiment (see Figure II-E-2 for a sketch of the apparatus) one measures very precisely the fission fragment angular and mass distributions for fission events following full momentum transfer. By analyzing these data, one determines the contributions of deep inelastic, quasifission and complete fusion-fission processes to the capture cross section. From this measurement and the known data for this system, one can deduce P\textsubscript{CN} and consequently, \(W\text{_{sur}}\) for this system. The separation of deep-inelastic events from fission events is based upon the energy-mass correlations of the fragments while the resolution of quasifission and complete fusion-fission is based upon a quantitative analysis of the fragment angular and mass distributions [11].
In the experiment, a $^{50}\text{Ti}$ beam struck a 0.5 mg/cm$^2$ $^{208}\text{Pb}$ target (supported by a 40μg/cm$^2$ C backing). Fission fragments were detected by a series of movable 60-300μ Si detectors mounted at angles between 90° and 170°, positioned 20-30 cm from the target. Coincident fragments were detected in a set of four 5x5 cm Si strip detectors. The time of flight of every fragment was measured.

Data were taken at $^{50}\text{Ti}$ beam energies of 230, 233, 238 and 253 MeV, bracketing the maximum in the EVR excitation function [6]. Analysis of these data is in progress.

(D. Peterson, W. Loveland, P. Sprunger, R. Naik, C. Jiang)

References

III Collaborative Work

During the past year, we have had the pleasure of collaborating with other research groups, aiding them in their research and in turn, enjoying their participation in our experiments. In this section, we summarize some recent experiments at ORNL in which we assisted Liang, Shapira, et al., The measurements involved two separate extensions of the previous work [1] in which a large sub-barrier enhancement was observed in the evaporation residue cross sections for the $^{132}\text{Sn} + ^{64}\text{Ni}$ reaction. In the first of these measurements, an attempt was made to measure the fusion-fission cross section for the $^{132}\text{Sn} + ^{64}\text{Ni}$ reaction at energies above the Coulomb barrier. This measurement should completely characterize the fusion cross section for this system as well as fission-evaporation competition in neutron-rich systems. The second measurement involved an attempt to measure the evaporation residue excitation function for the $^{134}\text{Sn} + ^{64}\text{Ni}$ system. This measurement is designed to determine the role of neutron transfer channels in sub-barrier fusion. The actual measurement involves measuring both the $^{134}\text{Sn}$ and $^{134}\text{Te}$ excitation functions for reactions with $^{64}\text{Ni}$ because the $^{134}\text{Sn}$ beam is mostly $^{134}\text{Te}$ and one must correct for this.


References


IV Technical Developments

A. Changes in the Surface Composition and Morphology of UF$_4$ Targets During Heavy Ion Irradiation

One of the most important technical challenges that faces people who study heavy element production reactions with picobarn cross sections is the changes in and damage of the targets that occurs during the high particle doses ($\sim 10^{17} - 10^{19}$ particles) typical of heavy element
studies. While there has been some work on target damage with lower dose U beams [1], there have been few studies of the changes in target composition and morphology that accompany high dose irradiations.

We have examined such changes in some UF$_4$ targets used in an attempt to synthesize $^{271}$Mt using the $^{238}$U($^{37}$Cl,4n) reaction [2] using an electron microprobe and an atomic force microscope.

Targets of UF$_4$ were prepared by volatilizing UF$_4$ in vacuum onto a 0.54 mg/cm$^2$ Al substrate. The target material (UF$_4$) was evaporated from resistance-heated W boats. For evaporation, the Al substrates were affixed to standard Berkeley Gas Filled Separator (BGS) [3] banana-shaped target frames with conductive carbon glue. The UF$_4$ deposits had an areal density of 400-500 µg/cm$^2$ and covered the entire area of the ~12 cm$^2$ target frames.

Nine of the banana-shaped target frames were mounted on the rotating wheel target assembly of the BGS for the irradiations. The target wheel was rotated at ~400 rpm in the 1 torr He environment of the separator. The incident $^{37}$Cl$^{12+}$ beam from the LBNL 88 Inch Cyclotron passed through the 0.45 µg/cm$^2$ C foil that serves as the entrance window of the BGS. In traversing this entrance window, the mean energy of the beam was reduced from 204 MeV to ~203.3 MeV [10] before entering the separator which had a pressure of 1 torr He. Inside the separator, the beam passed through the Al target backing and the UF$_4$ deposit in that order. The beam energy at the center of the target was 195 ± 3 MeV [4]. Typical $^{37}$Cl beam currents were ~0.5–1 µA. A total dose of ~5 x $10^{18}$ ions was delivered to the targets over the 360 hour experiment. (Beam doses were monitored using Rutherford scattering from the targets). At the end of the irradiation, one observed an obvious colored stripe in the area that had been exposed to the ion beam (Figure IV-A-1). The width of this stripe was centered in the target frame and was about 3 mm in width. If most of the beam went through this discolored area, the ion dose was then about (5 x $10^{18}$ ions/(0.3 x 2 x π x 15.7 cm$^2$)) = (5 x $10^{18}$ / 29.6 cm$^2$) = 1.7 x $10^{17}$ ions/cm$^2$.

At OSU we mounted samples of the target foils onto 15mm diameter circular steel stubs using adhesive tape, taking care to stretch the foil to avoid wrinkles. We monitored their chemical compositions using a CAMECA SX-50 electron microprobe and their morphological properties with a Digital Instruments Nanoscope III Multimode Scanning Probe Microscope. The instrument was operated as an atomic force microscope (AFM), operated in contact mode using a standard silicon nitride cantilever tip.

AFM measurements on an unused foil showed an unexceptional topography (Figure IV-A-2) with an averaged roughness of 5-10 nm that depends slightly upon the area included in the measurement. As can be seen in Figure IV-A-3, images of the non-irradiated portions of the used samples showed some larger scale features superimposed on a background that appears to have a very similar topography to the unused foil. Roughness measurements bear out this visual impression. As the area used for estimating the roughness is decreased, the averaged measured roughness dropped from about 10 nm to 4 nm.

However, AFM images (Figure IV-A-4) of the areas of the sample foils that had been exposed to the ion beam revealed remarkable morphological changes. These images reveal dramatic ridges and valleys microns wide by 500 nm high. The orientation of these ridges is predominantly along the long direction of the beam profile. The measured roughness now becomes a sensitive function of the area used. Over a wide area that encompasses these ridges and valleys, the average roughness is of the order of 100 nm, while the roughness over a small (1µm$^2$) area remains relatively close to that of the non-irradiated areas of the sample. The
irradiation seems to have caused a large-scale rumpling of the surface.

A manuscript has been prepared and submitted for publication that provides an account of this work and also includes a description of the chemical changes in the target. (P.R. Watson, W. Loveland, K.E. Gregorich, H. Nitsche, and P.M. Zielinski)

References

Figure IV-A-2. 5x5 μm AFM image of unused foil sample. Data scale 150 nm. Scanning conditions: 256x256 pixels, scan rate 1 Hz/line.
Figure IV-A-3. 5x5 \( \mu \text{m} \) AFM image of unirradiated area of sample. Data scale 250 nm. Scanning conditions: 256x256 pixels, scan rate 1 Hz/line.
Figure IV-A-4. 10x10μm AFM image of irradiated stripe area of sample. Data scale 500 nm. Scanning conditions: 256x256 pixels, scan rate 1 Hz/line.

V. Personnel

W. Loveland .................................................................Professor of Chemistry
Donald Peterson ............................................................. Postdoctoral Research Associate*
A.M. Vinodkumar ........................................................... Postdoctoral Research Associate
Lars Jonas Haller ............................................................. Visiting Scientist
Gustav Pettersson ........................................................ Visiting Scientist

Peter Sprunger ............................................................ Graduate Research Assistant
Radhika Naik ............................................................... Graduate Teaching Assistant
During the past year, we have had the privilege of collaborating with a number of scientists from other institutions. The following list summarizes the names of many of these individuals (and their home institutions) who contributed to work described in this report.

K. Aleklett (Uppsala)
K.E. Gregorich (LBNL)
C.J. Gross (ORNL)
J.J. Kolata (Notre Dame)
J. F. Liang (ORNL)
D.J. Morrissey (MSU)
H. Nitsche (LBNL)
J.P. Omtvedt (LBNL)
C. Rouki (Uppsala)
D. Shapira (ORNL)
R. Varner (ORNL)
P.M. Zielinski (LBNL)

VI. Degrees Granted


VII. Publications

A. Articles in Print


2. “Basic Principles of Radiochemistry”, W. Loveland, in Radioanalytical Methods

B. Articles submitted/accepted for publication

5. “Changes in Surface Composition and Morphology of UF$_4$ Targets During Heavy Ion Irradiation”, P. R. Watson, W. Loveland, P.M. Zielinski, K.E. Gregorich, and H. Nitsche, Nucl Instru Meth. B (accepted for publication).

C. Oral Presentations


6. “New Synthetic Paths to the Heaviest Elements”, W. Loveland, Chemistry Department, Indiana University, Bloomington, IN, April, 2004.

Appendices

To fulfill reporting requirements, we have included copies of reprints and preprints (not previously submitted) corresponding to work performed during this period as part of the Annual Progress Report.
Cross-section limits for the $^{208}\text{Pb}(^{86}\text{Kr},n)^{293}\text{I}$ reaction

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Abstract. In April-May 2001, the previously reported experiment to synthesize element 118 using the $^{208}\text{Pb}(^{86}\text{Kr},n)$ reaction was repeated. No events corresponding to the synthesis of element 118 were observed with a total beam dose of $2.6 \times 10^{17}$ ions. The simple upper-limit cross-sections (1 event) were 0.9 and 0.6 pb for evaporation residue magnetic rigidities of 2.0 Tm and 2.1 Tm, respectively. A more detailed cross-section calculation, accounting for an assumed source distribution function, the energy loss of the beam in traversing the target and the uncertainty in the magnetic rigidity of the $Z=118$ recoils is also presented. Re-analysis of the primary data files from the 1999 experiment showed the reported element 118 events are not in the original data. The current results put constraints on the production cross-section for synthesis of very heavy nuclei in cold-fusion reactions.

PACS. 25.70.Gh Compound nucleus – 25.70.Jj Fusion and fission reactions – 27.90.+b 220 ≤ A

1 Introduction

In 1999, the synthesis of element 118 (and its decay products) using the $^{208}\text{Pb}(^{86}\text{Kr},n)$ reaction was reported [1]. This claim was based on the apparent occurrence of three decay chains, each consisting of an implanted heavy atom and six subsequent alpha decays, correlated in time and position. A fourth event involving a number of "escape" alpha particles (depositing only part of their energy in the detector) was reported too [2,3]. Based upon the above three chains, and a revised estimate of the beam dose in the 1999 experiments, a cross-section of $7 \pm 3$ pb was deduced for the synthesis of element 118 at a projectile energy (center of target, lab system) of 448 MeV.

A new calibration of the magnetic rigidities ($B_{\mu}$) of the Berkeley Gas-Filled Separator (BGS) indicates that the reported element 118 compound-nucleus evaporation residues (ECX Res) recoiling from the target would have had a $B_{\mu}$ of 0.9 Tm in the 130 Pb He gas of the separator. Attempts to re-examine this result by other groups [4, 6] were unsuccessful.

Subsequently, the claim to synthesis of element 118 using the $^{208}\text{Pb}(^{86}\text{Kr},n)$ reaction has been retracted [7]. That retraction was based upon the absence of the reported [1-3] chains in a re-analysis of the binary data on the original 1999 data tapes. An investigation into this matter has concluded that there was no evidence of misconduct and data fabrication by any individual [8]. GSI has also repeated similar systematic data [9]. Despite the rejection of this claim, the current work is important in establishing definitive upper limits on the production of element 118 in the $^{208}\text{Pb}(^{86}\text{Kr},n)$ reaction.

Motivated in part by the erroneous report [1], there have been a number of papers [10–22] predicting the structure and decay properties of element 118 and its daughters. Should element 118 be synthesized, it will be interesting to compare those predictions with the observations. In a similar vein, there have been a number of papers [23–37] dealing with the synthesis of element 118 and the reported production cross-section, which was retracted, respectively large. Special mention should be made of the work of Smolanczuk [25, 30] which prompted the experimental measurement [1]. Smolanczuk originally estimated [25] a production cross-section of 670 pb for the $^{208}\text{Pb}(^{86}\text{Kr},n)$ reaction, an estimate that was later revised [30] to 3.9 pb. Other predictions [32–34] for the evaporation residue production cross-section for the reaction of 448 MeV $^{86}\text{Kr}$.
Table 1. “One-event” upper-limit cross-sections for formation of element 118 in the $^{208}$Pb($^{64}$Kr,n) reaction.

<table>
<thead>
<tr>
<th>$E^*$ (MeV)</th>
<th>Dose</th>
<th>Separator-Bp</th>
<th>One-event upper limit (pb)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.2</td>
<td>1.1x10^{18}</td>
<td>BGS-2.00 Tm</td>
<td>0.9</td>
<td>this work</td>
</tr>
<tr>
<td>13.2</td>
<td>1.5x10^{18}</td>
<td>BGS-2.12 Tm</td>
<td>0.6</td>
<td>this work</td>
</tr>
<tr>
<td>13.2</td>
<td>1.1x10^{18}</td>
<td>GANIL-LISE velocity filter</td>
<td>2.1</td>
<td>[5]</td>
</tr>
<tr>
<td>13.2</td>
<td>2.0x10^{18}</td>
<td>GARS-2.1 Tm</td>
<td>0.6</td>
<td>[6]</td>
</tr>
<tr>
<td>13.2</td>
<td>2.9x10^{19}</td>
<td>GSI-SHIP velocity filter</td>
<td>0.5</td>
<td>[7]</td>
</tr>
<tr>
<td>15.5</td>
<td>0.4x10^{18}</td>
<td>GSI-SHIP velocity filter</td>
<td>3.6</td>
<td>[4]</td>
</tr>
</tbody>
</table>

Figs. 1. The predicted and observed cross-sections for the synthesis of heavy nuclei for cold-fusion reactions involving a $^{208}$Pb target.

with $^{208}$Pb range from 0.005 to 2 pb. These predicted cross-sections are generally larger than expected from a simple log-logistic extrapolation of the trend of cross-sections for reactions of the type $^{208}$Pb(X,n) [4] (fig. 1). Similar predictions of a non-exponential decrease in cold-fusion cross-sections for reactions leading to elements 116 and lighter elements have been made [27]. The physical effect behind this trend was pointed out by Myers and Swiatecki [25, 26] as due to a smearing of the Coulomb barrier below the bombarding energy for symmetric target-projectile combinations, thus “unshielding” the saddle point. As pointed out by Swiatecki and Wilczynski [25], the system in these cases may well evolve from the dimolecular composite system into the compound nucleus. Thus, even the establishment of upper limits for the $^{208}$Pb($^{64}$Kr,n) cross-section may contribute to our understanding of the large scale collective motion in very heavy nuclei.

Besides the work presented herein, three recent attempts to reproduce the observations of [1] have been reported [4]. We summarize these attempts in tabl. 1 in terms of the beam energy used, the particle dose and the observed upper limit cross-section. All upper limit cross-sections are reported as “one-event upper limits”, i.e., the cross-sections that would have been reported had one event been observed with the given particle dose, target thickness, separator efficiency, etc. In the case of the data in [5], a separator efficiency of 50% [38] was used to calculate the cross-section. All excitation energies were calculated from [29].

2 2001 experimental setup

The reaction $^{208}$Pb($^{64}$Kr,n) was studied at the 88-Inch Cyclotron of the Lawrence Berkeley National Laboratory, using the Berkeley Gas-Filled Separator [29]. The experimental apparatus was a modified, improved version of the apparatus used in [1], including improved detectors and data acquisition systems, continuous monitoring of the separator gas purity, and a better monitoring of the $^{64}$Kr beam intensity. A $^{64}$Kr beam was accelerated to 457 MeV with an average current of $\sim 1.3 \times 10^{12}$ ions/s. The beam was gated through the 40 μg/cm² carbon entrance window of the separator before passing through the $^{208}$Pb target placed 0.5 cm downstream from the window. The targets were 470 μg/cm² thick (sandwiched between 20 μg/cm² C on the upstream side and 10 μg/cm² C on the downstream side). Nine of the arc-shaped targets were mounted on a 35 cm wheel that was rotated at 300 rpm. The beam energy in the target was 433-445 MeV [40], encompassing the in-target energies used in [1]. The reproducibility of beam energies from the 88-Inch Cyclotron was determined by measuring the beam energy spectrum in a Si p-i-n diode for three different $^{64}$Kr beam energies between 203 and 213 MeV. Deviations from the expected linear relationship between energy deposit in the p-i-n diode and the square of the cyclotron frequency gave a standard deviation of 0.2% (FWHM = 0.5%) for the beam energy. The beam intensity in the BGS was monitored by two silicon p-i-n diodes (mounted at ±27 degrees with respect to the incident beam) that detected elastically scattered beam particles from the target. Attenuating screens were installed in front of these diodes to reduce the number of particles reaching them (and the subsequent radiation damage to the detector). These detectors, the $^{208}$Pb targets and the separator entrance window were replaced periodically during the run which lasted three weeks.

The DYN ($Z = 104$ MeV) was operated spatially in flight from beam particles and transfer reaction products by their differing magnetic rigidities in the gas-filled separator. The separator was filled with helium gas at a pressure of 130 Pa. The expected magnetic rigidities of
123 MeV $^{209}$Bi EVRs were estimated using the data of Gliorso et al. [41]. These estimates were 2.00 Tm from extrapolation of the data in their fig. 3, and 2.10 Tm from their semi-empirical formula for EVR charge. In the current experiments, two settings of the magnetic fields of the separator were used, 2.00 Tm and 2.12 Tm. These settings differ by 6%, the width in magnetic rigidity of the focal-plane detector. The efficiency of the separator for transport and implantation of $Z = 118$ EVRs was estimated to be $\sim 75\%$ using a Monte Carlo simulation described below.

The detector setup at the focal plane of the separator consisted of a parallel plate avalanche counter (PPAC) [42] placed $\sim 29$ cm upstream of a Si-stripe detector. The 10 cm x 10 cm PPAC registered time, energy loss, and $x$, $y$ position of the particles passing through it. It has a thickness equivalent to $\sim 0.6$ mg/cm$^2$ of carbon. The time of flight of the EVRs between the PPAC and the Si-stripe detector was also recorded. The PPAC was used to distinguish between events arising from beam-related particles being implanted into the Si-stripe detector, and those arising from the decay of previously implanted atoms. During these experiments, the PPAC efficiency for detecting beam-related particles depositing between 8 and 14 MeV in the Si-stripe detector was 97.5-99.5%.

The 300 $\mu$m thick passivated ion-implanted silicon-strip detector had 32 vertical strips and an active area of 110 mm x 90 mm. The strips were positioned relative to the vertical (58 mm) direction. The sources used for position and energy calibrations are summarized in table 2.

The energy resolution of the focal-plane detector was measured during the $^{40}$Ca + $^{144}$Sm experiments using the 7.45 MeV $^{31}$Pb background peak. The energy resolution was 49 keV (FWHM). The differences in measured positions for the $^{209}$Bi - $^{209}$Bi full energy $\alpha$-$\alpha$ correlations in the $^{40}$Ca + $^{144}$Sm study had a Gaussian distribution with a FWHM of 0.52 mm ($\sigma = 0.22$ mm). The measured position resolution for full-energy alpha-particles correlated to “escape” alpha-particles (which deposited only 0.5-3.0 MeV in the detector) was $\sim 1.2$ mm (FWHM).

A second silicon-strip “punch-through” detector was installed behind the detector to reject particles passing through the primary detector. A “top” and a “bottom” detector were installed upstream of the focal-plane detector to detect escaping alpha-particles and fission fragments. The focal-plane detector combined with these “top” and “bottom” detectors had an estimated efficiency of $70\%$ for the detection of full-energy 11 MeV alpha-particles following implantation of a $^{209}$Bi nucleus.

Any event with $E > 0.5$ MeV in the focal-plane Si-stripe detector triggered the data acquisition. Data were recorded in list mode, and included the time of the trigger, the position and energy signals from the PPAC and the Si-stripe detectors, and energy signals from the “top”, “bottom” and “punch-through” detectors. With the use of buffering ADCs and scalers, the minimum time between successive events was 15 $\mu$s.

The energies of the $^{209}$Bi EVRs, after passing through the PPAC, were estimated to be $\sim 75$ MeV by extrapolation of beam energy stopping powers, calculated with SRIM2000 [49]. The $^{209}$Bi implantation pulse height in the Si-stripe detector was estimated to be 24-48 MeV after applying the pulse-height defect determined for Rn EVRs from the 366 MeV $^{86}$Kr + $^{138}$Sn reaction. With a beam current of $1.3 \times 10^{17}$ $^{86}$Kr ions striking the target, the average total counting rates ($E > 0.5$ MeV) in the focal-plane detector were $\sim 30$ s and $\sim 10$ s at BP settings of 2.00 and 2.12 Tm, respectively. The average rate of “alpha-particles” (8-14 MeV with no PPAC signal) was $\sim 0.2$ s.

3 Results

At separation settings corresponding to EVR magnetic rigidities of 2.00 and 2.12 Tm, the projectile doses were 1.1 x $10^{14}$ and 1.5 x $10^{14}$, respectively. Nine events similar to those reported in [1] or, as predicted [10-21], for the decay of $^{209}$Bi, were observed. Two independent searches were performed for superheavy element (SHE) decay sequences. In the first, a search routine in the GOOSY [43] environment was used to search for events similar to those reported in [1], i.e., a search was made for events in which two alpha-particle decays were detected in the focal-plane detector within 30 ns and with positions differing by $< 1.5$ mm. The energies of the alpha-particles had to be greater than 10 MeV and they had to be unrelated to beam events (no PPAC signal). No such $\alpha$-$\alpha$ correlations were found.

A second, less restrictive search was made for EVRs ($> 20$ MeV with a PPAC signal) followed by alpha decays (8-14 MeV in the focal-plane Si-stripe detector, no PPAC signal) correlated in position ($\pm 2$ mm, same strip) and time (within 10 ns). For this search, one detector strip on the low-Bp edge of the detector and three strips on the high-Bp edge of the detector were excluded because they detected relatively high rates of scattered beam particles with trajectories bypassing the PPAC. Two potential EVR-$\alpha$-$\alpha$ chains were identified. Based on the single rates for alpha-like events and EVRs, the expected number of random EVR-$\alpha$-$\alpha$ chains is $\sim 4$. Both of the observed EVR-$\alpha$-$\alpha$ chains had large differences in the vertical positions for the parent and daughter alpha-particles, $|\Delta y| > 0.78$ mm $= 3.5\sigma$. Thus, we conclude that there are no valid EVR-$\alpha$-$\alpha$ correlations in the data set from this experiment.

One must also consider the possibility that SHEs decay by spontaneous fission (SF). Spontaneous fission events in which the fragment was detected in the focal-plane detector ($E > 90$ MeV, no PPAC signal) can be confused with scattered $^{86}$Kr beam particles not vetoed by the PPAC signal. Therefore, we excluded, from the analysis, three strips on the low-Bp edge of the detector and four strips on the high-Bp edge of the detector to discriminate against those scattered beam particles. We observed no single fission fragment signals during the run using these gating conditions, i.e., no EVR-SF events.

There were no correlation chains containing at least EVR-$\alpha$-$\alpha$ or EVR-SF with additional full-energy
alpha-particles (8-14 MeV with no PPAC signal) or escape alpha-particles (with energy deposited in the plastic detector > 0.5 MeV and no PPAC signal).

The implantation depth in the Sierrp detector for the $^{208}$Pb recoil ($E \sim 73$ MeV after passing through the PPAC) was extrapolated from heavy-ion ranges calculated with SRIM2000 [40], and is estimated to be 7 μm. From this depth, the efficiency for detecting full energy for isotropically emitted $8-14$ MeV alpha-particles is 53%.

The expected decay sequence [22,1] following implantation of a $^{208}$Pb EVR consists of 6 α-particles emitted sequentially within the first 10 seconds after implantation. Using the 50% α-particle efficiency $\varepsilon$, together with a binomial series for the probability $P$ of observing at least $n$ members of an $N$ member chain,

$$P = \sum_{n=n}^{N} \frac{N!}{n!(N-n)!} \varepsilon^n (1-\varepsilon)^{N-n},$$

the probability for detecting at least two full-energy α-particles from a sequence of six is 93%. Such a binomial treatment can be used to calculate the efficiency for detection of other postulated decay chains. Assuming a 100% event chain detection efficiency results in “coincident” upper-limit cross-sections of 0.9 pb and 0.6 pb for $E_\beta$ settings of 2.00 and 2.12 μm, respectively. These cross-section limits assume detection of one event, whereas none were observed, and a constant production cross-section throughout the target thickness.

4 Discussion

4.1 Cross-section calculation

The standard cross-section calculation assumes a constant cross-section, $\sigma_{const}$, for all beam energies within the target. The number of events observed, $N_1$, is given by

$$N_1 = \dot{N} \times N_{elec} \times \varepsilon \times \sigma_{const},$$

where $\dot{N}$ is the integrated beam current, $N_{elec}$ is the area density of target atoms, and $\varepsilon$ is the experimental efficiency. This formalism was used in [4-6] for the cross-section limits quoted in those works and used above in table 1 and related discussion. However, in the case of the $^{208}$Pb($^{82}$Kr,$^{118}$I) reaction, the excitation function is expected to be narrow and the energy loss of the beam in traversing the target material is relatively large ($\Delta E = \sim 8$ MeV) [10], so the assumption of a constant cross-section for all beam energies within the target does not hold. For the purpose of an improved determination of cross-section limits in this experiment, a Gaussian excitation function, $\sigma(E)$, with a full-width at half-maximum (FWHM) of 5 MeV in the lab frame has been assumed:

$$\sigma(E) = \sigma_c \exp \left( \frac{(E - \bar{E})^2}{2\Delta E^2} \right),$$

where $\sigma_c$ is the cross-section at $\bar{E}$, the centroid energy, $E$ is the beam energy at the corresponding target depth, and $\sigma$ is the standard deviation of the Gaussian ($\sigma = 2.12$ MeV for a 5 MeV FWHM). This results in the differential equation

$$\frac{\partial N_1}{\partial E} = \varepsilon(E) \cdot \sigma(E) \cdot \varepsilon \cdot \frac{\partial E}{\partial E},$$

where the separator efficiency, $\varepsilon(E)$, depends on the beam energy (depth in target). Since the area number density of target atoms and the $dE/dx$ of the beam are nearly constant throughout the target, the area number density of target atoms within a differential target thickness element, $\frac{\partial N_1}{\partial E}$, is constant and equal to $N_1/\Delta E$, where $\Delta E$ is 8.0 MeV for the $^{82}$Kr beam in the $^{208}$Pb targets. Integrating over the energy range in the target, and solving for $\sigma_c$ results in

$$\sigma_c = \frac{N_1}{\int \delta \cdot \frac{\partial N_1}{\partial E} \cdot \varepsilon(E) \exp \left[ \frac{-(E-\bar{E})^2}{2\Delta E^2} \right] dE},$$

where $E_1$ and $E_f$ are the beam energies at the upstream and downstream limits of the target. For our experiment, assuming that the centroid of the excitation function corresponds to the center of the target and that the efficiency is constant throughout the target, $\sigma_c/\sigma_{const} = 1.6$, and thus the cross-section limits presented below are larger than those calculated in the traditional way by this factor.

4.2 BGS Efficiency simulation

The efficiency of the BGS is limited by the initial position, energy and angular distributions of recoils exiting the target, and by transmission of these recoils through the BGS under the influence of the magnetic fields, and energy loss, multiple scattering and charge exchange in the He fill gas.
4.3 Improved $^{209}$118 cross-section upper limits

Since neither the centroid of the element 118 excitation function nor the magnetic rigidity of the $Z = 118$ EVR's are known, numerical simulations of the BGS efficiency were run using several different choices for the excitation function centroid energy and average EVR magnetic rigidity. These simulations gave a set of experimental sensitivities as a function of the assumed excitation function centroid, and of the assumed magnetic rigidity. A total of 1.1 x 10$^{18}$ Kr ions at a BGS magnetic rigidity ($R_B$) of 2.0 Tm and the second with 1.5 x 10$^{18}$ Kr ions and $R_B = 2.12$ Tm were combined. The upper cross-section limits (assuming observation of one element 118 chain where zero events were observed) as a function of assumed $R_B$ and $R_B$ were presented in fig. 2. The cross-section limits reached were as low as 20 pb, and a limit of less than 40 pb was reached for compound-nucleus excitation energies from 10.0-16.8 MeV (444.5 < $E_{nuc}$ (MeV) < 454.0), covering magnetic rigidities for the recoiling products from 1.94-2.18 Tm.

5 Summary

Several experiments have led to one-event cross-section upper limits near 0.6 pb for the 440 MeV $^{209}$Pb($^{86}$Kr,n)$^{208}$118 reaction (table 1). In fig. 1, we compare this 0.6 pb limit with various theoretical predictions [27,30,33,34] for the production of $^{208}$118 in the reaction of 440 MeV $^{86}$Kr with $^{209}$Pb. This limit is below some of the predicted values. Combination of sets of upper-limit cross-sections from table 1 results in an upper-limit cross-section of ~ 0.2 pb, placing more stringent limitations on the validity of some of the models. The most pessimistic estimate [34] of the evaporation residue cross-section assumes the probability of forming a true compound nucleus, after capture of the projectile, decreases by approximately four orders of magnitude in going from $^{76}$Zn + $^{208}$Pb to $^{86}$Kr + $^{208}$Pb. This decrease may counter any fusion enhancement due to a lowering of the Coulomb barrier relative to the energy of the fission system in the latter reaction. Observation of the production of element 118 in the $^{208}$Pb($^{86}$Kr,n) reaction will require sensitivity to cross-sections smaller than ~ 0.2 pb.

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References

38. C. Stödel, private communication.
43. See http://www-ansi.cms.infn.it/HCAL/hom.html.
Enhanced Fusion-Evaporation Cross Sections In Neutron-Rich $^{132}$Sn on $^{64}$Ni


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Evaporation residue cross sections have been measured with neutron-rich radioactive $^{122}$Sn beams on $^{64}$Ni in the vicinity of the Coulomb barrier. The average beam intensity was 2 \times 10^{16} particles per second and the smallest cross section measured was less than 3 nb. Large sub-barrier fusion enhancement was observed. Coupled-channel calculations taking into account inelastic excitation significantly underpredict the measured cross sections below the barrier. The presence of several neutron transfer channels with large positive Q values suggests that multinucleon transfer may play an important role in enhancing the fusion of $^{122}$Sn and $^{64}$Ni.

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The interaction of two colliding nuclei consists of an attractive nuclear potential and a repulsive Coulomb potential. This creates a Coulomb barrier which the system has to overcome in order to fuse. At energies below the barrier, fusion occurs by quantum tunneling. Sub-barrier fusion cross sections for heavy ions are often found enhanced over the one dimensional barrier penetration model (BPM) prediction. The enhancement can be explained in most cases by the coupling of the relative motion and the nuclear structure degrees of freedom of the participating nuclei [1]. It has been suggested that the fusion yield would be further enhanced when the reaction is induced by unstable neutron-rich nuclei [2-4]. This is attributed to the large N/Z ratio of these nuclei reducing the barrier height and the presence of a large number of nucleon transfer channels which can serve as doorway states to fusion [5]. Sub-barrier fusion can be used in experiments to produce superheavy elements. Using closed shell neutron-rich projectile and target will lead to compound systems with lower excitation energies and with a smaller facility and, therefore, a higher survival probability [6].

The experimental search for fusion enhancement in heavy ion reactions has been pursued at several laboratories using neutron-rich radioactive beams. The measurements of $^{38}$S + $^{181}$Ta [7] and $^{29,31}$Al + $^{197}$Au [8] found only the enhancement expected from the lowering of the barrier height caused by the larger radii of the neutron-rich nuclei compared to the stable $^{32}$S and $^{27}$Al, respectively. This paper reports the first reaction study using accelerated unstable neutron-rich $^{122}$Sn beams to measure fusion-evaporation cross sections. The doubly magic $^{112}$Sn ($Z = 50$, $N = 82$) has eight extra neutrons compared to the heaviest stable Sn isotope, $^{124}$Sn. The N/Z ratio of $^{122}$Sn (1.64) is larger than that of $^{48}$Ca (1.4) and $^{50}$Pb (1.54) which are closed shell nuclei commonly used to produce heavy elements [9]. The target, $^{64}$Ni, is semimagic ($Z = 28$) and is the most neutron rich stable isotope of nickel. The compound nucleus formed in this experiment, $^{188}$Pt, lies in the valley of β stability. It has initial excitation energies greater than 30 MeV and can decay by particle evaporation or fission.

The experiment was carried out at the Holifield Radioactive Ion Beam Facility (HRIBF) at Oak Ridge National Laboratory. The isotope separator online technique was used to produce radioactive $^{122}$Sn. Isobaric contaminants at $A = 132$ were suppressed by extracting molecular SnS$^2^-$ from the ion source and subsequently breaking it up in the charge exchange cell where the SnS$^-$ was converted to Sn$^+$ [10]. The $^{122}$Sn ions were produced at six energies (453, 475, 489, 504, 536, and 560 MeV) and delivered to the target. The beam intensity was measured by passing it through a 10 μg/cm$^2$ carbon foil and detecting the secondary electrons in a microchannel plate (MCP) detector. Three of these MCP systems were used in this experiment for monitoring the beam and providing timing signals. The average beam intensity was $2 \times 10^{15}$ particles per second (pps) with a maximum near $3 \times 10^{16}$ pps. The purity of the $^{122}$Sn beam was checked by measuring the energy loss in an ionization chamber (IC). A $^{132}$Te beam was used to calibrate the energy loss spectrum. It was determined that the impurity was less than 2% and that all measurable impurities had a higher atomic number (Z) than Sn. This impurity has negligible effect on the measurement because the higher Coulomb barrier suppresses the fusion of the contaminants with the target. A $^{124}$Sn beam was used as a guide beam to set up the accelerator and beam line optics. At the target position, the beam was focused to a spot 1.0 mm horizontally and 2.5 mm vertically. The shape of the
guide beam was recorded by an electronic phosphor [11] located 14 cm in front of the target. This beam was also used for testing the detector system. The $^{132}\text{Sn}$ beam was then tuned by changing the optical elements and comparing the beam shape with that of the guide beam using the electronic phosphor.

The evaporation residues (ERs) were detected along with beam particles by a timing detector and an IC located 16.9 cm from the target at $0^\circ$. They were identified by their time of flight and energy loss in the IC. The acceptance of the timing detector was a 2.54 cm diameter circle and the detection efficiency was approximately 100% for these heavy ions. In the time-of-flight measurement, the coincidence between two timing detectors placed 119 and 315 cm upstream from the target provided the timing references. The data acquisition was triggered by the scaled down beam singles or the ER-beam particle coincidences. With this triggering scheme an overall dead time of less than 5% was achieved. The IC was filled with CF$_4$ gas. The pressure was adjusted between 50 and 60 Torr to optimize the separation of ERs from the beam.

A detailed description of the experimental apparatus will be published elsewhere [12]. Figure 1 shows the histogram of the energy loss in the first two segments of the IC for a beam energy of 536 MeV. Although there is some signal pileup introduced by directly injecting the beam into the detector, it is clear that the ERs are still well separated from the beam. With this setup, measurement of ER cross sections less than 5 mb can be achieved.

The cross section was obtained by integrating the ER yield and summing the beam particles in the IC. Because of the low intensity of radioactive beams, the measurement was performed with a thick target, 1 mg/cm$^2$ self-supporting highly enriched (99.8%) $^{64}$Ni foil. The target thickness was determined by measuring the energy loss of $\alpha$ particles emitted from a $^{244}$Cm source and a 536 MeV $^{132}\text{Sn}$ beam passing through the target, and by measuring the weight and area of the target. The energy loss of $^{132}\text{Sn}$ projectiles in the target was approximately 40 MeV. At energies below the Coulomb barrier, the excitation function falls off exponentially. For this reason, the measured cross section ($\sigma_{\text{meas}}$) is sensitive predominantly to the front portion of the target and is actually a weighted average of the cross section over the range of energy loss in the target, from the energy of the beam entering the target ($E_{\text{in}}$) to the beam energy corrected for the energy loss in the carbon foils) to that exiting the target ($E_{\text{out}}$), namely,

$$\sigma_{\text{meas}} = \frac{\int_{E_{\text{in}}}^{E_{\text{out}}} \sigma(E) dE}{\int_{E_{\text{in}}}^{E_{\text{out}}} dE}.$$  \hspace{1cm} (1)

To determine the effective reaction energy, the cross section was parametrized as an exponential function, $\sigma(E) = N \exp(\alpha E)$, where $N$ is a normalization factor and $\alpha$ is a slope parameter. By solving the integral Eq. (1) for two adjacent data points in the excitation function, $N$ and $\alpha$ were obtained. Subsequently, the effective energy is deduced by inverting the exponential function, namely, $E = \ln(\sigma_{\text{meas}}/N)/\alpha$.

Since this experiment was performed in inverse kinematics (a heavy projectile on a light target) the ERs were very forward focused. However, the shape of the beam spot was not symmetric. Moreover, one of the disadvantages of using a thick target is the multiple scattering which results in broadening the angular distribution. Monte Carlo simulations were used to estimate the efficiency of the apparatus. The angular distribution of ERs was generated by the statistical model code PACE [13] and the width of the distribution of multiple scattering angles was predicted by Ref. [14]. The simulations show that the efficiency of the apparatus changes from 95 ± 1% for the lowest beam energy to 98 ± 1% for the highest energy.

Figure 2 presents the fusion-evaporation excitation function of $^{132}\text{Sn} + ^{64}\text{Ni}$ measured in this work (solid circles) and that of $^{64}\text{Ni}$ on even Sn isotopes measured by Freeman et al. [15]. The open circle is our measurement using the $^{132}\text{Sn}$ guide beam which is consistent with the measurement of Ref. [15] as shown by the open triangles. In Fig. 2 the energy is scaled by the fusion barrier ($V_p$) predicted by the Bass model [16] and the ER cross section is scaled by the size of the entrance channel $R = 1.2(A_p^{1/3} + A_i^{1/3})$ fm, where $A_p$ ($A_i$) is the mass of the projectile (target). It can be seen that at the highest energy the ER cross section for $^{132}\text{Sn} + ^{64}\text{Ni}$ is larger. This can be expected from the higher stability against fission for the neutron-rich compound nucleus.
At energies below the barrier, the ER cross sections for $^{132}\text{Sn} + ^{64}\text{Ni}$ are found much enhanced compared to those of $^{64}\text{Ni} + 112$–124$\text{Sn}$ and a simple shift of the barrier height cannot explain the enhancement.

To compare the measured excitation function with fusion models, it is necessary to estimate fission yields in the reaction. Statistical model calculations were carried out using the code PACE. The input parameters were determined by reproducing the ER and fission cross sections of $^{64}\text{Ni} + ^{124}\text{Sn}$ in Ref. [17]. The following parameters were used: level density parameter $a = A/8$ MeV$^{-1}$ where $A$ is the mass of the compound nucleus, ratio of the Fermi gas level density parameter at the saddle point to that of the ground state $a_f/a_0 = 1$, diffuseness of spin distribution $d = 4\hbar$, and Sierk's fission barrier [18]. The calculations predict that fission is negligible for $^{132}\text{Sn} + ^{64}\text{Ni}$ and $^{64}\text{Ni} + ^{124}\text{Sn}$ at $E_{\text{cm}}$ = 160 MeV. Therefore, the following discussion will be restricted to the data points at $E_{\text{cm}}$ = 160 MeV where the ER cross sections are taken as fusion cross sections.

Large sub-barrier fusion enhancement in $^{132}\text{Sn} + ^{64}\text{Ni}$ can be seen when the excitation function is compared to a one-dimensional DPM shown by the dotted curve in the upper panel of Fig. 3. The nuclear potential was assumed to have a Woods-Saxon shape. The potential parameters were obtained by adjusting them to reproduce the fusion cross section of $^{64}\text{Ni} + ^{132}\text{Sn}$ in Ref. [17] at high energies. They are depth $V_0 = 76.6$ MeV, radius parameter $r_0 = 1.2$ fm, and diffuseness parameter $a = 0.65$ fm.

It is well established that sub-barrier fusion enhancement can be described by channel couplings [1]. The couplings result in splitting the single barrier into a distribution of barriers. The incident flux overcoming the low energy barriers gives rise to the enhanced fusion cross sections [19–21]. Coupled-channel calculations were performed with the code CCFULL [22] which takes into account the effects of nonlinear coupling to all orders. The calculations used the same nuclear potential

**TABLE I.** Parameters used in coupled-channel calculations. $\lambda^+$ is the spin and parity, and $\beta_2$ is the deformation parameter.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>$\lambda^+$</th>
<th>$E^*$ (MeV)</th>
<th>$\beta_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{64}\text{Ni}$</td>
<td>$2^+$</td>
<td>1.346</td>
<td>0.179</td>
</tr>
<tr>
<td>$^{124}\text{Sn}$</td>
<td>$2^+$</td>
<td>1.132</td>
<td>0.095</td>
</tr>
<tr>
<td>$^{132}\text{Sn}$</td>
<td>$3^-$</td>
<td>2.614</td>
<td>0.136</td>
</tr>
<tr>
<td>$^{132}\text{Sn}$</td>
<td>$2^+$</td>
<td>4.041</td>
<td>0.06</td>
</tr>
</tbody>
</table>
excited state and the small reduced transition probability \(\langle B(E2)\rangle\).

In \(^{64}\text{Ni} + \^{124}\text{Sn}\), the \(^{64}\text{Ni} + \^{64}\text{Ni}\) reaction is the only transfer channel which has a positive \(Q\) value. Coupled-channel calculations including this channel with an empirically determined coupling constant of 0.25 MeV and \(J\) are in good agreement with the fusion cross sections near and below the barrier, as can be seen by the solid curve in the lower panel of Fig. 3. It is noted that the code CCFULL is suitable for reactions where multimucleon transfer is less important than \(J\) \cite{22} as is the case in \(^{64}\text{Ni} + \^{124}\text{Sn}\). For the \(^{132}\text{Sn}\)-induced reaction, the \(Q\) values are positive for \(^{64}\text{Ni}\) picking up two to six neutrons which suggests that the observed fusion enhancement may be attributed to multimucleon transfer similar to that observed in \(^{40}\text{Ca} + \^{90}\text{Zr}\) \cite{25}. Although CCFULL is not expected to treat the coupling of multimucleon transfer accurately, exploratory calculations were carried out to provide a preliminary estimate of the effects of coupling to these channels. Results of calculations including \(J\) and these transfer channels using the same coupling constant as in \(^{64}\text{Ni} + \^{124}\text{Sn}\) and assuming clusters of neutrons transferred to the ground state are shown by the solid curve in the upper panel of Fig. 3. The calculation cannot account for the cross sections near and below the barrier, nevertheless, it illustrates qualitatively the enhancement of sub-barrier fusion due to the coupling to multimucleon transfer. More realistic calculations which also consider sequential transfer, as pointed out in Ref. \cite{25}, may account for the discrepancy. It would be interesting to study near-barrier fusion further using even more neutron-rich \(\text{Sn}\) isotopes. However, this will be a very challenging task because the present beam intensity for \(^{132}\text{Sn}\) at HIRIDF is approximately 2000 pps and highly contaminated. On the other hand, HIRIDF can provide other pure neutron-rich radioactive beams such as \(\text{Br}\) and \(\text{I}\) with reasonable intensities for further studies.

In the future, it is necessary to measure fission for \(^{132}\text{Sn} + \^{64}\text{Ni}\) in order to obtain the fusion cross sections and study the survival probability of the compound nucleus. In addition, it was found that the extra-push energy \cite{26} is needed for compound nucleus formation in \(^{64}\text{Ni}\) on stable even \(\text{Sn}\) isotopes at high energies \cite{17} and the extra-push energy diminishes as the number of neutrons in \(\text{Sn}\) increases. The threshold for requiring the extra-push energy given in Ref. \cite{17} is near the \(^{132}\text{Sn} + \^{64}\text{Ni}\) system. This can be investigated by measuring ER and fission cross sections at higher energies.

In summary, fusion-evaporation cross sections using neutron-rich \(^{132}\text{Sn}\) beams on a \(^{64}\text{Ni}\) target were measured at energies near the Coulomb barrier. Large sub-barrier fusion enhancement using neutron-rich radioactive heavy ion beams was observed in this experiment. The enhancement cannot be explained by a simple shift of the barrier height, or by the coupling to inelastic excitation channels. There are five neutron transfer channels which have large positive \(Q\) values. These reaction channels may serve as doorway states to fusion. Further experiments using neutron-rich radioactive beams would advance our understanding of the mechanism for the fusion enhancement and provide valuable information for using such beams to produce superheavy elements at future radioactive beam facilities.

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\begin{thebibliography}{10}
\bibitem{6} S. Hofmann, Prog. Part. Nucl. Phys. 46, 293 (2000).
\bibitem{9} S. Hofmann and G. Münzenberg, Rev. Mod. Phys. 72, 733 (2000).
\bibitem{12} D. Shapiro et al. (to be published).
\bibitem{14} J. B. Marion and B. A. Zimmerman, Nucl. Instrum. Methods 51, 93 (1967).
\bibitem{17} K. T. Lesko et al., Phys. Rev. C 34, 2155 (1986).
\bibitem{23} S. Raman et al., At. Data Nucl. Data Tables 61, 1 (1997).
\bibitem{26} W. J. Swiatecki, Phys. Scr. 24, 113 (1981).
\end{thebibliography}
Synthesis of Heavy Nuclei Using Radioactive Nuclear Beams

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A calculation of the expected production rates of the nuclei with $100 \leq Z \leq 120$ using radioactive nuclear beams has been performed. Starting with the expected beam intensities for the proposed RIA accelerator, all possible projectile-target combinations were considered. A semi-empirical formalism was used to predict the best cases for further study, which was done with a detailed model, IIIIVAP. The formalism was tested with known reactions, where production cross sections were predicted to within an order of magnitude (over a range of $10^3$). The calculations show several exciting new opportunities for atomic physics, chemistry and nuclear spectroscopy of the most n-rich heavy nuclei. Stable beams are still the preferred path for new element synthesis.

1. INTRODUCTION

In proposing new radioactive beam facilities, such as the RIA facility, the issue arises as to the potential for synthesizing heavy nuclei using radioactive nuclear beams (RNDs). While it is tempting to write several interesting possibilities, we believe that one must make quantitative estimates of the realistic probability of such studies. Accordingly, we have quantitatively evaluated the possibilities of synthesizing heavy nuclei using the proposed RIA facility.

We begin by noting the potential value of n-rich beams in synthesizing heavy nuclei. A plot (Figure 1) of the calculated half-lives [1] of the heavy nuclei sorted by $Z$ and $A$ shows that in addition to the expected increases in half-life at the magic numbers $N=162$ and $N=184$, one sees a general overall increase in half-life with increasing neutron number. This increase amounts to an increase of orders of magnitude in half-life, that could qualitatively change the character of the studies of the atomic physics and chemistry of these elements. We would also expect that the lowered fusion barriers for the n-rich projectiles might lead to lower excitation energies and greater survival probabilities.

2. METHODOLOGY

What we have done to examine the possibilities of synthesis of new nuclei using the RIA facility is to do a brute force calculation. We have taken the RIA beam list [2] that gives the identity and intensity of all the expected RNDs that have suitable energies (<15 A MeV) and considered all possible combinations of these projectile nuclei with all "stable"
Figure 1. Variation of calculated half-lives of heavy nuclei with Z and A.

target nuclei (including radioactive actinide nuclei) with appropriate target thicknesses (¬1 - 1.0 mg/cm²). The optimum projectile energy was determined by scanning a range of projectile energies near the Bass barrier [3] for each projectile-target combination. The fusion probability was calculated using a semi-empirical formalism developed by Armbruster [4] that considers fusion hindrance (which is evaluated as a function of the effective fissionability of the completely fused system) and assumes fusion to be primarily a s-wave process. \( \Gamma_a/\Gamma_f \) values used to evaluate the survival probabilities are calculated using values of the neutron binding energies and fission barriers from Smolanczuk [5] and Möller et al. [6]. For the most promising cases, the fusion probability was re-calculated using the HIVAP computer code [7]. The yield of each projectile-target combination was evaluated in atoms/day.

2.1. Details of the Calculational Model

The cross section for the production of an evaporation residue, \( \sigma_{EVR} \), can be written as

\[
\sigma_{EVR} = \sigma_{CN} W_{evr}
\] (1)
where $\sigma_{CN}$ is the complete fusion cross section and $W_{\text{sur}}$ is the survival probability of the completely fused system. The complete fusion cross section can be written as

$$\sigma_{CN} = \sum_{J} \sigma_{\text{capture}}(E_{\text{cm}}, J) P_{CN}(E_{\text{cm}}, J)$$

(2)

where $\sigma_{\text{capture}}$ is the capture cross section and $P_{CN}$ is the probability that the projectile-target system will evolve, inside the fission saddle point to form a completely fused system rather than reseparating (quasifission). The capture cross section can be given in a simplified form [4] as

$$\sigma_{\text{capture}} = \pi \lambda^2 (2J + 1) T(E_{\text{cm}}, J)$$

(3)

where $\lambda$ is the reduced wavelength, $T$ the transmission coefficient for a projectile with cm energy $E_{\text{cm}}$ and forming a system with angular momentum $J$. For heavy element synthesis reactions, this can be simplified [4] to be $\pi \lambda^2 f_{\text{lim}}^2$ where $f_{\text{lim}} = 15\hbar$, and thus the capture cross section becomes formally independent of $J$. The key approximation, due to Armentrout [4], is that $P_{CN}$ can be represented by a simple function

$$P_{CN}(E_{\text{cm}}, J) = 0.5[\exp(c(x_{\text{eff}} - x_{\text{thr}}))]$$

(4)

where the coefficient $c$ has the value of 106 and the constant $x_{\text{thr}}$ is 0.72 for actinide-based reactions and 0.81 for reactions involving Pb or Bi targets. The calculation of the effective fissility of the fused system is described in [4]. The survival probability is evaluated with the classical expression [8] for an excitation energy dependent value of $\Gamma_{n}/\Gamma_{f}$

$$\frac{\Gamma_{n}}{\Gamma_{f}} = \frac{4\lambda^{2}/(E_{\text{cm}}^* - B_{n})/(k(2a(E_{\text{cm}}^* - B_{f})^{1/2} - 1))}{\exp(2a^{1/2}(E_{\text{cm}}^* - B_{n})^{1/2} - (E_{\text{cm}}^* - B_{n})^{1/2})}$$

(5)

where one assumes $a_{n} = a_{f} = a = \lambda/12$ MeV$^{-1}$ and $k = 9.8$ MeV.

2.2. Tests of Model

This formalism was checked by calculating the cross sections of the formation reactions used to synthesize elements 102-112 and comparing them to the observed cross sections. The results (Figure 2) show an agreement between predicted and observed cross sections within an order of magnitude for cross sections ranging over seven orders of magnitude.

3. RESULTS

In Figure 3, we show the overall results of the calculation. Neutron-rich radioactive beams offer significant advantages in the production of neutron-rich heavy nuclei, but stable beams are the tool of choice for the synthesis of nuclei near the valley of $\beta$-stability or for $Z$ near 120.

Focusing on the neutron-rich heavy nuclei, we show (Figure 4) the predicted production rates of the most neutron-rich isotopes of the elements from Fm to Hs. Most of the nuclei shown are "unknown" and represent new opportunities for systematic studies of the nuclear structure of the trans-ferrmium nuclei. Many of these nuclei have predicted halflives of minutes to days which can qualitatively change studies of their chemistry and
atomic structure. The reactions used to produce these nuclei involve projectile nuclei that are not very far from stability because the most n-rich projectile nuclei have low production rates [2].

The situation regarding the synthesis of new heavy elements is mixed. While it is possible to make new elements using radioactive beams, the use of stable beams gives higher production rates. (In the stable beam calculations, we have assumed 1.0 nA beam currents.)

One might inquire why the conventional cold fusion reactions do not play a more important role in these simulations. In Table 1, we summarize the situation for some typical reactions. The product of the cross section and available flux is not sufficient to give suitable production rates.

It should be noted that these estimates of production rates do not include any fusion enhancement effects, other than the lowering of the fusion barriers (and excitation energy) with the n-rich projectiles. Also the formalism used in these calculations, which does reproduce the production cross sections for elements 102-112, does not describe correctly the recent reports from Dubna involving the synthesis of elements 112, 114, 115, 116 and 118 using reactions of $^{40}$Ca with actinide targets.

REFERENCES

Figure 3. Comparison of production of heavy nuclei with RIA beams and stable beams.

5. R. Smolanczuk et al., Phys. Rev. C 52, 1871; C59, 2634
6. P. Möller et al., ADNDT 39.213: 59, 185

Table 1
Cold fusion reactions with RNBs

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Cross Section (cm^2)</th>
<th>RIA flux (ions/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>^{196}Pt(^{88}Kr,n)^{283}114</td>
<td>0.7 x 10^{-38}</td>
<td>10^{11}</td>
</tr>
<tr>
<td>^{204}Hg(^{94}Kr,n)^{297}116</td>
<td>1.1 x 10^{-38}</td>
<td>10^{10}</td>
</tr>
<tr>
<td>^{209}Bi(^{88}Cu,n)^{274}112</td>
<td>0.3 x 10^{-36}</td>
<td>10^{10}</td>
</tr>
<tr>
<td>^{208}Pb(^{93}Fe,n)^{290}Hs</td>
<td>4.6 x 10^{-38}</td>
<td>10^{9}</td>
</tr>
<tr>
<td>^{208}Pb(^{59}Cr,n)^{296}Sg</td>
<td>7.5 x 10^{-34}</td>
<td>10^{7}</td>
</tr>
</tbody>
</table>
Figure 4. Production rates of n-rich isotopes of elements 100-108 using the RIA facility.
Changes in Surface Composition and Morphology of UF$_4$ Targets During Heavy Ion Irradiation

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**ABSTRACT**  
The changes in surface composition and morphology have been measured for UF$_4$ targets subjected to high dose irradiation ($5 \times 10^{18}$ ions) with ~195 MeV $^{37}$Cl (~5.3 AMeV). Using atomic force microscopy and an electron microprobe, we observed significant morphological changes in the targets along with changes in chemical composition.

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**Keywords:** Ion-induced changes; nuclear reaction targets; atomic force microscopy; uranium tetrafluoride

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1. Introduction

The effect of heavy-ion bombardment on solid surfaces has been extensively studied particularly with respect to sputtering [1], surface modification [2] and ion track formation [3]. In the context of nuclear reactions involving heavy ion collisions with actinide targets, knowledge of beam-induced changes in target thickness or composition is very important. For example, any loss of material during the irradiation, will affect production rates in the reaction under study. To date, little data on this topic has appeared in the literature. Stiebing et al.[4], using proton backscattering of un-irradiated and irradiated actinide targets showed that radiation damage from heavy ions ($^{238}$U) with energies of ~ 6 MeV/u can cause massive microstructural changes in targets. They postulated that radiation-induced granularization of the material produced a redistribution of material that lead to significant changes in thickness, although this did not lead to major changes in the scattering yield. Bethge [5] found a loss of material without a change in chemical composition in the irradiation of ThF$_4$ targets by 5.9 A MeV $^{238}$U in a 2.2 hour irradiation with a beam current of 45 enA. In a similar vein, Munzenberg et al.[6] and Kindler et al. [7] have reported on the changes in lead and bismuth targets upon irradiation with heavy ions.

As part of an effort to synthesize $^{271}$Mt, using the $^{238}$U($^{37}$Cl,4n) reaction [8], an intense beam of $^{37}$Cl$^{12+}$ ions was used to irradiate thin targets of UF$_4$ that had been deposited on Al backings using vacuum volatilization. The ion dose was large (~5 x 10$^{18}$), approximately four orders of magnitude larger than in the work cited above, and we observed a clear discoloration in the region where the ion beam struck the targets. To better understand the changes in these targets, we have examined the chemical and morphological changes induced in these targets.
using an electron microprobe and a scanning probe microscope. In this paper, we summarize these observations.

2. Experimental

Targets of UF\(_4\) were prepared by volatilizing UF\(_4\) in vacuum onto a 0.54 mg/cm\(^2\) Al substrate. The target material (UF\(_4\)) was evaporated from resistance-heated W boats. For evaporation, the Al substrates were affixed to standard Berkeley Gas Filled Separator (BGS) [9] banana-shaped target frames with conductive carbon glue. The UF\(_4\) deposits had an areal density of 400-500 \(\mu\)g/cm\(^2\) and covered the entire area of the \(\sim 12\) cm\(^2\) target frames.

Nine of the banana-shaped target frames were mounted on the rotating wheel target assembly of the BGS for the irradiations. The target wheel was rotated at \(\sim 400\) rpm in the 1 torr He environment of the separator. The incident \(^{37}\)Cl\(^{12+}\) beam from the LBNL 88 Inch Cyclotron passed through the 0.45 \(\mu\)g/cm\(^2\) C foil that serves as the entrance window of the BGS. In traversing this entrance window, the mean energy of the beam was reduced from 204 MeV to \(\sim 203.3\) MeV [10] before entering the separator which had a pressure of 1 torr He. Inside the separator, the beam passed through the Al target backing and the UF\(_4\) deposit in that order. The beam energy at the center of the target was 195 \(\pm 3\) MeV [10]. Typical \(^{37}\)Cl beam currents were \(\sim 0.5-1\) p\(\mu\)A. A total dose of \(\sim 5 \times 10^{18}\) ions was delivered to the targets over the 360 hour experiment. (Beam doses were monitored using Rutherford scattering from the targets). At the end of the irradiation, one observed an obvious colored stripe in the area that had been exposed to the ion beam (Figure 1). The width of this stripe was centered in the target frame and was about 3 mm in width. If most of the beam went through this discolored area, the ion dose was then about \((5 \times 10^{18} \text{ ions}/(0.3 \times 2 \times \pi \times 15.7 \text{ cm}^2)) = (5 \times 10^{18} / 29.6 \text{ cm}^2) = 1.7 \times 10^{17} \text{ ions/cm}^2\).

At OSU we mounted samples of the target foils onto 15mm diameter circular steel stubs.
using adhesive tape, taking care to stretch the foil to avoid wrinkles. We monitored their chemical compositions using a CAMECA SX-50 electron microprobe and their morphological properties with a Digital Instruments Nanoscope III Multimode Scanning Probe Microscope. The instrument was operated as an atomic force microscope (AFM), operated in contact mode using a standard silicon nitride cantilever tip.

3. Results and Discussion

The chemical composition of an irradiated foil deposit showed significant changes from that of an un-irradiated foil deposit. Interestingly, the un-irradiated foil deposit did not show the expected stoichiometric ratio of UF$_4$, but rather one of UOF$_3$. (Please note the formula UOF$_3$ should be understood as representing the stoichiometric atom ratio but not necessarily the actual compound present). The source of the oxygen is not likely to be from the thin oxide layer on the Al backing foil, as no Al signal was detected by the microprobe. Hence, the O detected must either be from adsorbed water or from oxidation of the UF$_4$ film. As the sample was under vacuum during the microprobe measurements, if the O signal were from adsorbed water, that water must be quite strongly bound to the surface. If the O signal were coming from adsorbed water we would not expect much variation from place to place on the foil and we might also expect that signal to decrease due to electron-stimulated desorption of water during the microprobe measurement. As we shall see shortly, the amount of oxygen present on different areas of the foils varied substantially. In addition, no decrease in the O signal with time was observed so it seems the more likely that the oxygen was present as O atoms in the UF$_4$ film rather than as adsorbed water.

It is possible that some oxygen incorporation into the film occurred during its preparation. The source of this oxygen would have to be in the starting material or in the
background gases in the evaporation system. It seems unlikely that the bulk UF$_4$ source material would contain this large an amount of oxygen. The measured stoichiometry in the unused film would be consistent with equimolar amounts of UF$_4$ and UO$_2$F$_2$. The oxidation of UF$_4$ to UO$_2$F$_2$ is a well-known reaction at high temperatures [11,12] and it is possible that the hot film was oxidized during condensation. Alternatively, it is also possible that a slow surface oxidation of the film occurred during storage. There appear to be no direct measurements of such surface oxidation processes at low temperatures in the literature.

In Figure 2 we show the variation of the composition in the non-irradiated areas and the visible irradiated stripe area of the samples. There is a significant loss of fluorine and concomitant increase in the oxygen content in the stripe that extends for sample A from about -15500 to -21000 position units. At one end of the scan (-13000 position units) the stoichiometry is approximately UOF$_3$, the same as was found for the unused foil. The O concentration is higher, and the F lower, at the other end of the scan (-22000 positions units), but this is probably due to the scan not being continued far enough to completely escape the stripe area.

It is well-known that UF$_4$ has a surprisingly high sputter yield. Meins et al. [13] measured U yields as high as 200 for sputtering with high-energy $^{35}$Cl ions, compared with a value of 0.06 for UO$_2$ under similar conditions. Unfortunately, the composition of the sputtered species has not been measured. The measurements in [13] counted only the loss of U from the substrate. One interpretation [14] has speculated that electronic relaxation produces uranium halide dimers on the surface that are efficiently sputtered. The model could accommodate sputtering of species varying in composition from U$_2$ to U$_2$F$_6$. The stoichiometry in the stripe area is about UO$_{2.5}$F$_2$. The change in stoichiometry from the un-irradiated foil could be accommodated by a model where there is sputtering of a U$_2$F$_x$ species with x = 7-8:
AFM measurements on an unused foil showed an unexceptional topography (Figure 3) with an averaged roughness of 5-10 nm (Table 1) that depends slightly upon the area included in the measurement. As can be seen in Figure 4, images of the non-irradiated portions of the used samples showed some larger scale features superimposed on a background that appears to have a very similar topography to the unused foil. Roughness measurements bear out this visual impression. As the area used for estimating the roughness is decreased, the averaged measured roughness (Table 1) dropped from about 10 nm to 4 nm.

However, AFM images (Figure 4) of the areas of the sample foils that had been exposed to the ion beam revealed remarkable morphological changes. These images reveal dramatic ridges and valleys microns wide by 500 nm high. The orientation of these ridges is predominantly along the long direction of the beam profile. The measured roughness now becomes a sensitive function of the area used. As Table 1 indicates, over a wide area that encompasses these ridges and valleys, the average roughness is of the order of 100 nm, while the roughness over a small (1 \( \mu m^2 \)) area remains relatively close to that of the non-irradiated areas of the sample. The irradiation seems to have caused a large-scale rumpling of the surface.

These AFM observations differ from those reported for a number of studies of single crystal alkali metal and other fluorides by Neumann and coworkers [3,15,16]. In these cases tracks of the heavy ions could be imaged as very small (20-50 nm in width) hillocks. In addition, out-of-plane swelling was apparent and was particularly pronounced for LiF. Unlike the similar swelling seen in oxides, such as SiO\(_2\) where there is a straightforward interpretation in terms of a density change due to amorphization [17], the origin of the swelling in LiF, where amorphization is thought to be less likely, is said to be more likely to arise from the formation of specific defects or defect clusters [18].
Our observations do echo those of Stiebing et al. [4], who found massive microstructural changes in UF₄ targets sandwiched between C foils that had been exposed to a variety of heavy-ion beams. In some cases UF₄ appeared to have migrated from some areas and piled up on others. Stiebling et al. [4] recommended that targets be exposed to no more than 10¹⁵ ions/cm² before changing them. As stated earlier, the dose in this experiment was ~1.7 x 10¹⁷ ions/cm². Although their estimates might be taken to strictly apply to the reaction they studied which involved ~6 A MeV ²³⁸U projectiles whose energy loss is about 10 times greater than the 5.5 A MeV ³⁷Cl used in this work, scaling of their rule about target changes would have suggested the targets were to be changed about every 4 hours during the measurement, which is not a practical strategy for heavy element research.

4. Conclusions

This study represents one of the first studies of the damage to actinide targets in the high dose irradiations typical of studies to synthesize new heavy nuclei. We have observed a change in the physical structure of the UF₄/Al targets upon irradiation. Contrary to the lower dose studies [4], we have apparently observed material flow during the irradiation in addition to radiation-induced processes. We have observed a change of chemical composition of the target during irradiation in addition to what appears to be a pre-irradiation oxidation of the UF₄. All of these observations would indicate care must be taken to carefully monitor the condition of the targets in such experiments, using techniques like Rutherford scattering that directly measure the amount of material on the target. However, one must be aware that substantial variations in target properties can occur during irradiation leading to changes in the efficiency at which recoils are transferred out of the target or changing the beam energy distribution in the target.
Acknowledgements

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References


Table captions

Table 1. Averaged root-mean-square ($R_q$) roughness measurements over various areas (A) for irradiated samples and an unused foil.

Figure captions

Figure 1. Typical target after irradiation. A colored stripe is clearly visible.

Figure 2. Variation in elemental composition across a sample. The visible irradiated stripe covered a region from roughly -15500 to -21000 units.

Figure 3. 5x5 $\mu$m AFM image of unused foil sample. Data scale 150 nm. Scanning conditions: 256x256 pixels, scan rate 1 Hz/line.

Figure 4. 5x5 $\mu$m AFM image of unirradiated area of sample. Data scale 250 nm. Scanning conditions: 256x256 pixels, scan rate 1 Hz/line.

Figure 5. 10x10$\mu$m AFM image of irradiated stripe area of sample. Data scale 500 nm. Scanning conditions: 256x256 pixels, scan rate 1 Hz/line.
<table>
<thead>
<tr>
<th></th>
<th>A (μm²)</th>
<th>R_q (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unused foil</td>
<td>1</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>8</td>
</tr>
<tr>
<td>Samples - non-irradiated region</td>
<td>1</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>9</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>Samples - irradiated region</td>
<td>1</td>
<td>12</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>90</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>100</td>
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</table>

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Figure 5. 10x10μm AFM image of irradiated stripe area of sample. Data scale 500 nm.
Chapter 3

Basic Principles of Radiochemistry

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What is radiochemistry?

Radiochemistry is defined as "the chemical study of radioactive elements, both natural and artificial, and their use in the study of chemical processes" (1). Operationally radiochemistry is defined by the activities of radiochemists, i.e., (a) nuclear analytical methods (b) the application of radionuclides in areas outside of chemistry, such as medicine (c) the physics and chemistry of the radioelements (d) the physics and chemistry of high activity level matter and (e) radiotracer techniques. Radiochemistry is closely allied with nuclear chemistry, a three-pronged endeavor made up of: (a) studies of nuclear properties such as structure, reactions, and radioactive decay by people trained as chemists (b) studies of macroscopic phenomena (such as geochronology or astrophysics) where nuclear processes are intimately involved and (c) the application of measurement techniques based upon nuclear phenomena (such as activation analysis or radiotracers) to study scientific problems in a variety of fields. All radiochemists are, by definition, nuclear chemists, but not all nuclear chemists are radiochemists. Many nuclear chemists use purely non-chemical techniques to study nuclear phenomena, and thus, their work is not radiochemistry. Neither nuclear chemistry nor radiochemistry should be confused with radiation chemistry, the study of the chemical effects of ionizing radiation.
Nuclear chemists and radiochemists must possess knowledge of radioactivity, radioactive decay and nuclear reactions, the interaction of radiation with matter and nuclear radiation detectors. These subjects are treated in a number of fine textbooks (2-5). Radiochemistry is treated in these textbooks as well as a number of excellent textbooks that focus primarily on radiochemistry (6-8).

II. What makes radiochemistry unique?

Radiochemistry involves the application of the basic ideas of inorganic, organic, physical and analytical chemistry. However, the need to manipulate radioactive materials imposes some special constraints (and features) upon these endeavors. The first of these involves the number of atoms involved and the concentration of solutions. The range of activity levels in radiochemical procedures ranges from pCi to MCi, but for the sake of argument, let us assume an activity level, A, typical of radiotracer experiments, 1 μCi (= 3.7 x 10⁶ dis/s = 3.7 x 10⁶ Bq), of a nucleus with mass number ~ 100. If we assume a half-life for this radionuclide of 3 days, the number of nuclei involved can be calculated from the equation

\[ N = \frac{A}{\lambda} = \frac{(1\mu Ci)(3.7 \times 10^4 \text{ dps/μCi})(3 \text{ days})(24 \text{ hr/day})(3600 \text{ s/hr})}{\ln 2} \]

where \( \lambda \) is the decay constant of the nuclide (\(- \ln 2 / t_{1/2} \)). Then

\[ N \sim 1.4 \times 10^{10} \text{ atoms} \]

Wt. of sample \(- 2.3 \times 10^{-12} \text{ g} \)

This quantity of material, if prepared as an aqueous solution of volume 1 L would have a concentration of \(10^{14}\) mol/L. This simple calculation demonstrates a number of the important features of radiochemistry, i.e., (a) the manipulation of samples involving infinitesimal quantities of material (b) the ability to conduct tracer studies since the quantity of radioactive material added to a system is so small as to not affect the system and (c) the power of nuclear analytical techniques. In the previous example, the quantity of radioactivity, 1 μCi is a significant, easily detectable quantity of radioactivity. Thus it is possible to speak of detection sensitivities in terms of pg or less. In fact, since the decay of a single atom (such as a heavy element) can lead to the emission of an alpha-particle that can be detected with 100% efficiency, it is possible to speak of detecting single atoms—or doing chemistry on an atom by atom basis.
The small numbers (< 100) of atoms involved in some radiochemical procedures can cause problems. The extreme dilution in some solutions can mean that equilibrium is not reached due to kinetic limitations. One must be sure that all phase space is appropriately sampled in every chemical procedure.

Conventional analytical techniques generally operate at the ppm or higher levels. Some techniques such as laser photoacoustic spectroscopy are capable of measuring phenomena at the $10^{-8} - 10^{-6}$ mol/L level. The most sensitive conventional analytical techniques, time-resolved laser-induced fluorescence and ICP-MS are capable of measuring concentrations at the part per trillion level, i.e., 1 part in $10^{12}$, but rarely does one see detection sensitivities at the single atom level as routinely found in some radioanalytical techniques. While techniques such as ICP-MS are replacing the use of neutron activation analysis in the routine measurement of ppb concentrations, there can be no doubt about the unique sensitivity associated with radioanalytical methods.

Along with the unique sensitivity and small quantities of material associated with radiochemistry, there are certain problems associated with the use of radioactivity. The most important of these is the need to comply with the regulations governing the safe use and handling of radioactive material. This task is a primary focus in the design and execution of radiochemical experiments and is often a significant factor in the cost of the experiment. Because so many of these rules are site specific, they are not treated in this short review.

There are some chemical effects that accompany high specific activities that are unique to radiochemistry and worth noting. Foremost among these are the chemical changes accompanying radioactive decay. The interaction of ionizing radiation from a radioactive source with air can result in the generation of ozone and the nitrous oxides, which can lead to corrosion problems. Sources containing radium or radium (Ra) produced from the decay of heavier elements, such as uranium, will emanate radon (Rn) gas as the decay product of Ra. The decay products of Rn are particulates that deposit on nearby surfaces, such as the interior of the lungs, leading to contamination problems. In high activity aqueous solutions, one can make various species such as the solvated electron, $e_{aq}^-$, hydroxyl radicals, OH·, as well as the solvated proton, $H_3O^+$. The hydroxyl radical, OH·, is a strong oxidizing agent with

$$\text{OH}^- + e^- \rightarrow \text{OH}^- \quad E_0 = 2.8 \text{ V}$$

while the solvated electron, $e_{aq}^-$, is a strong reducing agent

$$e_{aq}^- + H^+ \rightarrow \frac{1}{2} \text{H}_2 \quad E_0 = 2.77 \text{ V}$$
Solutions involving high activity levels will change their redox properties as a function of time. For example, a solution of 100 Bq/mL (10\(^{-7}\) mol/L) of \(^{239}\)Pu will have all the atoms undergo a redox change in a period of one year. In general, it is hard to keep high specific activity solutions stable. Reagents, column materials, etc. can suffer radiation damage also. In radiotracer studies, the self-decomposition (radiolysis) of \(^1\)H or \(^14\)C-labeled compounds can lead to a variable concentration and number of products. In addition to the usual concerns about the purity of chemicals, one must be concerned with radiochemical purity, the presence of other radioactive species in low concentration. Some tracers (usually cations) in solution behave as colloids rather than true solutions. Such species are termed radiocolloids and are aggregates of \(10^3 - 10^7\) atoms, with a size of the aggregate of 0.1 – 500 nm. They are quite often formed during hydrolysis, especially of the actinides in high oxidation states. One can differentiate between real colloids and pseudocolloids, in which a radionuclide is sorbed on an existing colloid, such as humic acid or Fe(OH)\(_3\). The chemical behavior of these radiocolloids is difficult to predict, as the systems are not at equilibrium.

### III. Radiochemical Separation Techniques

Radiochemical separations involve the conventional separation techniques of analytical chemistry adapted to the special needs of radiochemistry. Radiochemical purity is generally more important than chemical purity. When dealing with short-lived nuclides, speed may be more important than yield or purity. The high cost of radioactive waste disposal may require unusual waste minimization steps. A recent review summarizes some newer developments of relevance to radiochemistry (5) along with the chapters by Dietz and Nash et al. in this book.

**Precipitation**

The oldest, most well established chemical separation technique is *precipitation*. Because the amount of the radionuclide present may be very small, carriers are frequently used. A carrier is usually a stable or long-lived radionuclide in question or a chemically homologous element. The carrier is added in macroscopic quantities and insures the radioactive species will be part of a kinetic and thermodynamic equilibrium system. It is important that there is an isotopic exchange between the carrier and the radionuclide. There is the related phenomenon of co-precipitation wherein the radionuclide is incorporated into or adsorbed on the surface of a precipitate.
that does not involve an isotope of the radionuclide. Examples of this behavior are the sorption of radionuclides by Fe(OH)₃ or the co-precipitation of the actinides on LaF₃. Co-precipitation can be minimized by the use of holdback carriers, inactive materials that are isotopic with the radionuclide and replace it in co-precipitates. Separation by precipitation frequently involves the chemical process of scavenging, which is the removal of unwanted activities from solution by precipitation. In a typical application, the desired element is put in a soluble oxidation state, a precipitation of some scavenger compound such as Fe(OH)₃ is performed, removing unwanted activities, the element of interest is oxidized or reduced to a different oxidation state and removed. Separation by precipitation is largely restricted to laboratory procedures and apart from the bismuth phosphate process used in WWII to purify Pu, has little commercial application.

Solvent Extraction

Separation by liquid-liquid extraction (solvent extraction) has played an important role in radiochemical separations. Ether extraction of uranium was used in early weapons development, and the use of tri-butyl phosphate (TBP) as an extractant for U and Pu was recognized in 1946, resulting in the commercial PUREX process for reprocessing spent reactor fuel. In recent years, there has been a good deal of activity in the development of solvent extraction processes for the removal of transuranic elements, ⁵²Sr and ¹⁵⁷Cs from acidic high level waste. Laboratory demonstrations of the TRUEX process that uses the neutral extractant CMPO (octyl(phenyl)-N,N-diisobutylicarbamoyl-methylphosphine oxide) to separate the transuranium elements from acidic high-level waste have been successful. More recently crown ethers have been used as specific extractants for strontium and cesium.

In solvent extraction, a metal ion is distributed between two immiscible phases, usually an aqueous phase and an organic phase. To achieve sufficient solubility in the organic phase, the metal ions must be in the form of a neutral, non-hydrated species. The distribution ratio D is defined as

\[ D = \frac{[M]_{\text{org}}}{[M]_{\text{aq}}} \]

where \([M]_i\) is the metal ion concentration in the \(i\)th phase. The relevant equilibria to describe the extraction in systems of lipophilic acidic chelating agents are

\[ \text{HL}_{\text{org}} \rightleftharpoons \text{HL}_{\text{aq}} \]

\[ \text{HL}_{\text{aq}} \rightleftharpoons H^+ + \text{I}^-_{\text{aq}} \]

\[ M_{\text{aq}}^{3+} + 3\text{HL}_{\text{org}} \rightleftharpoons (\text{ML}_3)_{\text{org}} + 3H^+_{\text{aq}} \]
where $K_c$ is the equilibrium constant for the last reaction. The distribution coefficient $D$ is given as

$$D = [\text{ML}]_{\text{org}}/[\text{M}^{2+}]_{\text{aq}} = K_c [\text{HL}]_{\text{org}}^3/[\text{H}^+]_{\text{aq}}^3$$

If one introduces a water-soluble complexing agent into the system, the $[\text{M}^{2+}]_{\text{aq}}$ becomes $[\text{M}^{2+}]+[\text{MX}^{2+}]+[\text{MX}_2^{+}] + \ldots$ and the measured distribution ratio will include these species as well. The separation factor between two ions, $S$, is given by the ratio of their distribution coefficients

$$S = D_a/D_b$$

Thus, the most effective separations will involve cases where the target ion interacts strongly with the extractant but is less strongly complexed by the aqueous ligand $X$.

Besides chelating agents, extractant molecules can be of other types. Cation transfer from the aqueous to organic phase can be accomplished by solvation of a neutral metal complex (TBP, for example) or formation of an ion-association complex (quaternary amines) or by a surfactant that forms extractable aggregates. Cation selectivity is best for the lipophilic chelating agents. Recent developments have suggested the utility of using supercritical fluids, such as $\text{CO}_2$, as solvents, mainly for lesser costs in waste disposal.

**Ion Exchange**

Ion exchange is one of the most popular radiochemical separation techniques due to its high selectivity and the ability to perform separations rapidly. In ion exchange, a solution containing the ions to be separated is brought into contact with a synthetic organic resin containing specific functional groups that selectively bind the ions in question. In a later step the ions of interest can be removed from the resin by elution with another suitable solution that differs from the initial solution. Typically the solution containing the ions is run through a column of resin. The resins are typically cross-linked polystyrenes with attached functional groups. Most cation exchangers (such as Dowex 50) contain free sulfonic acid groups, $\text{SO}_3\text{H}$, where the cation displaces the hydrogen ion. Anion exchangers (such as Dowex 1) contain quaternary amine groups, such as $\text{CH}_2\text{N}((\text{CH}_3)_2\text{Cl}$ where the anion replaces the chloride ion. The resin particles have diameters of 0.08 – 0.16 mm and exchange capacities of 3-5 meq per gram of dry resin.

It is common to adsorb a group of ions on the column material and then selectively elute them. Complexing agents, which form complexes of varying
solubility with the absorbed ions, are used as eluants. There exists a competition between the complexing agent and the resin for each ion and each ion will be exchanged between the resin and the complexing agent several times as it moves down the column. This is akin to a distillation process. The rates at which the different ions move down the column vary causing a spatial separation between “bands” of different ions. The ions can be collected separately in successive eluant fractions (see Figure 1).

![Graph](image)

**Figure 1. Elution of tripositive lanthanide and actinide ions on Dowex-50.**

The most widely cited application of ion exchange techniques is to the separation of the rare earths or actinides from one another. This is done with cation exchange using a complexing agent of α-hydroxyisobutyric acid (“α-but”). The order of elution of the ions from a cation exchange column is generally in order of the radii of the hydrated ions with the largest hydrated ions leaving first; thus lawrencium elutes first and americium last among the tripositive actinide ions (see Figure 1). In the case of the data of Figure 1, the separation between adjacent cations and the order of elution is derived from the comparative stability of the aqueous actinide or lanthanide complexes with α-
hydroxyisobutyrate. As shown in Figure 1, there is a strikingly analogous behavior in the elution of the actinides and lanthanides that allowed chemists to prove the identity of new elements in the discovery of elements 97-102 (Bk-No). For cation exchange, the strength of absorption goes as $M^{4+} > M^{3+} > MO_2^{2+} > M^{2+} > MO_2^{+}$.

The anion exchange behavior of various elements has been extensively studied. For example, consider the system of Dowex 1 resin and an HCl eluant. Typical distribution ratios for various elements as a function of [Cl] are shown in Figure 2. One usually sees a rise in the distribution coefficient D until a maximum is reached and then D decreases gradually with further increases in [Cl]. The maximum occurs when the number of ligands bonding to the metal atom equals the initial charge on the ion. Figure 2 or similar data can be used to plan separations. For example, to separate Ni(II) and Co(II), one needs simply to pass a 12 M HCl solution of the elements through a Dowex 1 column. The Co(II) sticks to the column while the Ni(II) is not absorbed. A mixture of Mn(II), Co(II), Cu(II), Fe(III), and Zn(II) can be separated by being placed on a Dowex 1 column using 12 M HCl, followed by elutions with 6M HCl (Mn), 4 M HCl (Co), 2.5 M HCl (Cu), 0.5 M HCl (Fe) and 0.005 M HCl (Zn). Du et al., for instance, prove in their chapter the viable application of the ion exchange technique to separate and purify plutonium.

In addition to the organic ion exchange resins, some inorganic ion exchanges, such as the zeolites, have been used. Inorganic ion exchangers are used in situations where heat and radiation might preclude the use of organic resins although the establishment of equilibria may be slow.

Newer developments have emphasized the preparation of more selective resins. Among these are the chelating resins (such as Chelex-100) that contain functional groups that chelate metal ions. Typical functional groups include iminodiacetic acids, 8-hydroxyquinoline or macrocyclic units such as the crown ethers, calixarenes or cryptands. The bifunctional chelating ion exchange material, Dipholin® resin—a substituted diphosphonic acid resin, shows promise in treating radioactive waste. Important newer resins include those with immobilized phosphorus ligands. (9)

**Exchange Chromatography**

Exchange chromatography is an analytical separation technique that is closely related to solvent extraction. Extraction chromatography is a form of solvent extraction where one of the liquid phases is made stationary by adsorption on a solid support. The other liquid phase is mobile. Either the aqueous or the organic phase can be made stationary. Extraction chromatography has the selectivity of solvent extraction and the multistage character of a chromatographic process. It is generally used for laboratory scale experiments although some attempts have been made to use it in larger scale
operations. The common applications involve the adsorption of an organic extractant onto a variety of inorganic substrates such as silica or alumina or organic substrates such as cellulose or styrene-divinyl benzene copolymers.

The same extracting agents as used in solvent extraction can be used in extraction chromatography. Early applications of extraction chromatography have employed various traditional extractants such as the acidic organophosphorus compounds (di-2-ethylhexyl) phosphoric acid, HDEHP) or TBP as extractants for the actinide elements. Recent advances have led to a variety of new solvent exchange extractants such as the crown ethers, cryptands or bifunctional organophosphorus compounds. A particularly successful application is the selective sorption of actinides on TRU resins, involving solutions of CMPO in TBP sorbed on Amberlite XAD-7. This resin has found a number of applications in the determination of the actinides in complex matrices.

Rapid Radiochemical Separations

Many of the separation techniques we have described take hours to perform. Many interesting nuclei, such as the heavier actinides, the transactinides, or the light nuclei used in PEI have much shorter half-lives. Thus, we review the principles of rapid radiochemical separations (procedures that take seconds to minutes) and refer the reader to (11, 12) for details.

In most chemical separation procedures, the goal is to selectively transfer the species of interest from one phase to another, leaving behind any unwanted species. The phase-to-phase transfer is rapid, but the procedures to place the species in the proper form for transfer are slow. The goal of rapid radiochemical separations is to speed up existing chemical procedures or to use new, very fast chemical transformations.

Two procedures are commonly used for rapid radiochemical separations, the batch approach and the continuous approach. In the batch approach, the desired activities are produced in a short irradiation, separated and counted with the procedure being repeated many times to reduce the statistical uncertainty in the data. In the continuous approach, the production of the active species is carried out continuously and the species is isolated and counted as produced.

One of the most widely used techniques for rapid chemical separation is that of gas chromatography, which has been developed for use with the transuranium elements by Zvara and co-workers (13). In gas chromatography, volatile elements or compounds are separated from one another by their differences in distribution between a mobile gas phase and a stationary solid phase. Thermochromatography involves passing a gas through a column whose temperature decreases continuously with distance from the entrance. Thus the less volatile species condense on the column walls first with the more volatile species depositing last. This technique was used to show the chemical properties
(14-16) of the transactinides Rf-Hs and their behavior relative to their chemical congeners.

IV. Applications of Radiochemistry

The discipline of radiochemistry has been actively pursued since the early 1900s. Because of this, there are a very large number of successful applications of radiochemistry. In this short review, we can only describe a few of the most important of these applications and refer the reader to standard textbooks that catalog many of these applications (2-8).

Radioanalytical Techniques

Activation Analysis

As indicated earlier, the exquisite sensitivity of radiochemistry lends itself to the development of Radioanalytical techniques. One of the most important of these is activation analysis. Activation analysis is an elemental analysis technique that allows one to determine the amount of an element X contained in some material Y. The basic steps in activation analysis are: (a) Irradiate Y with ionizing radiation, so that X will change into a radioactive isotope of X, X*. (b) Using chemical or instrumental techniques, “isolate” X and X* from all other elements in Y (not necessarily quantitatively) and measure the activity of X*. “Isolation” can involve a chemical separation, followed by counting X* or use of a technique like γ-ray spectroscopy that allows the detection of γ-rays associated with the decay of X* without any intervening chemistry. (c) Calculate the amount of X present. This last step (c) is usually done by irradiating and counting a known amount of X under the same conditions used for Y. Then the mass of X in Y = (known mass of X)(activity of X* in Y)/activity of X* in known.

The most common choice for the irradiation of samples is to irradiate them with reactor thermal neutrons. In this case, the activation reaction is $^{A}X(n,\gamma)^{A+1}X$. This choice is motivated by the high capture cross sections for thermal neutrons, the copious fluxes of neutrons available in reactors ($>10^{12}$ n/cm$^2$-sec), the ease with which neutrons penetrate matter allowing analysis of rocks and other solids and the small activation of C, N, and O, major elements in biological samples, by these neutrons. Activation analysis using neutrons is called NAA. If the “isolation” method involves a radiochemical separation, it is termed RNAA, while instrumental “isolation” of the activity of interest is INAA. Other activating particles include photons, fast (~ 14 MeV) neutrons and charged particles, which are used primarily for surface analysis.
Typical detection sensitivities for NAA are ~ ppb with some elements being detected at parts-per-trillion levels. NAA is a multi-elemental analysis technique, allowing the simultaneous determination of 30-40 elements in most samples. It is a non-destructive technique, a fact of some importance for analysis of very valuable specimens such as lunar samples, forensic samples, art artifacts, etc. The drawbacks of NAA include the need to handle radioactivity, the availability of nuclear reactors for irradiations, the long times and high labor costs frequently associated with assaying longer-lived (t_{1/2} > 1 day) radionuclides, and the lack of chemical speciation information. While activation analysis continues to be an important tool for the analysis of geological samples, some biological materials and environmental samples, and in art and archeology, it is being replaced in many applications by ICP-MS. This latter technique has similar or better sensitivities than NAA, and can be used to get chemical speciation information and is generally more cost effective.

The chapter by Landsberger in this book contains more information about this subject.

**PIXE**

**PIXE** (particle induced x-ray emission) is an elemental analysis technique based upon the bombardment of thin samples with charged particles, usually 1-3 MeV protons. The charged particles interact with the inner shell electrons of the atoms causing ionization and subsequent x-rays that follow the rearrangements of the atomic electrons. The x-ray energies are characteristic of the atomic number Z of the ionized atom and the number of emitted x-rays can be used to determine the number of atoms present. Either absolute or comparative measurements can be used for quantitative determinations. Because of the limited penetrating power of both the exciting protons and the emitted x-rays, PIXE samples should be thin. Sensitivities for PIXE analyses are typically at the ppm level and are good for elements with Z > 10-30. PIXE is a non-destructive, multi-elemental, rapid analysis technique that has found widespread use in measuring elemental abundances in environmental samples such as filters loaded with particulate matter. Because the detected radiation is the result of atomic rather than nuclear transitions, PIXE is really a non-nuclear technique, but because of the use of nuclear particle accelerators and x-ray detectors, it is usually classified as a radioanalytical method. The chapters by Robertson in this book describe applications of PIXE.
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**Radioimmunoassay (RIA)**

There are a large number of nuclear analytical techniques based upon the use of radiotracers. One technique that is most widely used and utilizes
immensely radiotracer is radioimmunoassay (RIA). RIA is a sensitive quantitative method for determining trace amounts of biological molecules based upon their ability to displace a radioactively labeled form of the molecule from combination with its antibody. A known amount of the biomolecule, such as a hormone, is labeled with a radiotracer and mixed with a sample containing an unknown, unlabeled amount of that hormone and a limited amount of its antibody. The radiolabeled hormone and the unlabeled hormone compete for binding sites on the antibody. If the antigen(hormone)-antibody complex is isolated and its activity determined, the amount of unlabeled hormone can be determined using standard calibration curves. This technique can be used to detect concentrations of hormones, drugs, vitamins, enzymes, viruses, serum proteins, etc. at levels as low as $10^{-10}$ mol/L.

Nuclear Medicine

Perhaps the most important and most rapidly developing application of radiochemistry today is in nuclear medicine. The use of radionuclides in medicine can be for either diagnosis or therapy. An estimated 10 to 12 million nuclear medicine imaging and therapeutic procedures are performed each year in the U.S. and over 100 million laboratory tests using radionuclides are performed. Therapeutic procedures may involve direct irradiation of target organs using x-ray machines, particle accelerators, etc. and in-situ irradiation by physically implanted radioactive material or chemically administered radionuclides like $^{131}$I. Of the diagnostic procedures, over 90% use either $^{99}$Tc$^m$ or one of the iodine isotopes ($^{123,125,131}$I). Common nuclear medicine diagnostic applications include diagnosis of hyperthyroidism (Grave’s Disease), cardiac stress tests to analyze heart function, bone scans for detecting tumors, etc. The radionuclides used in these procedures are generated using reactors ($^{131}$I), radionuclide generators ($^{99}$Tc$^m$), medical (in hospital) cyclotrons (the PET nuclides $^{11}$C and $^{18}$F), or ordinary cyclotrons ($^{123,125}$I, $^{201}$Tl).

$^{99}$Tc$^m$ is a favored radionuclide for diagnosis because it only emits 141 keV $\gamma$-rays, which are very convenient for imaging. $^{99}$Tc$^m$ ($t_{1/2}=6.0$ h) is produced using radionuclide generators or “cows”. The nuclide $^{98}$Mo is produced in reactors using the $^{98}$Mo($n,\gamma$) reaction or fission. The $^{98}$Mo is isolated, oxidized to MoO$_4^{2-}$, loaded on an alumina column to which it binds. The $^{98}$Mo decays to $^{99}$Tc$^m$, producing pertechnetate ions, TcO$_4^-$. The TcO$_4^-$ is eluted off the column with 0.9% NaCl. The column is a “generator” or “cow” in that the $^{98}$Mo is continually decaying to $^{99}$Tc$^m$, replacing the removed species and the column can be “milked” repeatedly to produce the $^{99}$Tc$^m$.

Another important application of radiochemistry in nuclear medicine is in PET (positron emission tomography). Positron emitters emit $\beta^+$ particles which annihilate in matter producing two 0.511 MeV photons that travel in opposite directions from the point of annihilation. If both of these $\gamma$-rays are detected in
coincidence, one can locate the point of emission. The two most important PET nuclides are $^{11}$C ($t_{1/2} = 20.3$ m) and $^{18}$F ($t_{1/2} = 1.83$ h). These nuclides can be attached to glucose molecules and used for real time imaging of organs like the brain that take up glucose.

Physics and Chemistry of the Heaviest Elements

The actinides and transactinides are all radioactive and thus studies of their physics and chemistry will necessarily involve radiochemistry. Among the active areas of research are the study of the fate, transport and speciation of the actinides in the environment, the physics and chemistry associated with nuclear waste management and site remediation, stockpile stewardship, the fundamental chemistry and physics of the actinide elements (thermodynamics, crystal structure, etc.) and the characterization of the basic chemical and physical properties of the transactinides.

This latter area dealing with the properties of the transactinides is among the most challenging (and high profile) aspects of radiochemistry. These elements have short half-lives and the typical production rates are about one atom/experiment. The experiments must be carried out hundreds of times and the results summed to produce statistically meaningful results.

The elements Lr - 112 are expected (non-relativistically) to be d-block elements because they are expected to involve the filling of the 6d orbital. However relativistic calculations have shown that rutherfordium prefers a 6d7p electron configuration rather than the 6d2 configuration expected non-relativistically and predicted by a simple extrapolation of periodic table trends. This prediction also implies that RFCl₄ should be more covalently bonded than its homologue HfCl₄ and ZrCl₄. In particular, the calculations show RFCl₄ to be more volatile than HfCl₄ which is more volatile than ZrCl₄ with bond dissociation energies in the order RFCl₄ > ZrCl₄ > HfCl₄. (The periodic table extrapolations would predict the volatility sequence ZrCl₄ > HfCl₄ > RFCl₄).

The first aqueous chemistry of rutherfordium showed that it eluted from liquid chromatography columns as a 4+ ion consistent with its position in the periodic table as a d-block element rather than a trivalent actinide. Gas chromatography of the rutherfordium halides has shown the volatility sequence ZrCl₄ > RFCl₄ > HfCl₄ with a similar sequence for the tetra bromides. Thus rutherfordium does not follow the expected periodic table trend nor is its behavior in accord with relativistic calculations.

The aqueous chemistry of hahnium has also shown unexpected trends. Hahnium does not behave like its homologue tantalum in aqueous solutions but similar to niobium or the pseudo-group 5 element, protactinium, under certain conditions. Gas phase thermochromatography of NbBr₅, TaBr₅, and HaBr₅,
shows NbBr₅ and TaBr₅ to behave similarly while HaBr₅ is less volatile. Just the opposite trend was predicted by relativistic calculations.

Thus the chemistry of hahnium and rutherfordium deviates significantly from periodic table trends, a fact that is partly explained by relativistic calculations.

The study of the chemistry of scaborgium (14) is remarkable for its technical difficulty as well as the insight offered. In an experiment carried out over a two-year period, 15 atoms of scaborgium were identified in a thermal chromatography experiment. From this experiment, one concluded that the volatility sequence MoO₂Cl₂ > WO₂Cl₂ > SgO₂Cl₂ was followed. This observation agreed with both the extrapolations of periodic table trends and relativistic calculations. In an aqueous chemistry experiment, three atoms of scaborgium were detected, showing scaborgium to have a hexavalent character expected of a Group 6 element. The most stable oxidation state of scaborgium is +6 and like its homologues molybdenum and tungsten, scaborgium forms neutral or anionic oxo- or oxohalide compounds.

More recently, hohrium (element 107) has been characterized (15), in a thermochromatography experiment involving the oxychloride. BhO₂Cl was found to be less volatile than the oxychlorides of the lighter members of group VII, in agreement with relativistic calculations. Hassium (element 108) has also been characterized (16) in the form of its tetraoxide, HsO₄, in a thermochromatography experiment. HsO₄ appears to be less volatile than its group VIII congener OsO₄.

**Radiotracers**

The basic idea behind the use of radiotracers is that all the isotopes of a given element will behave the same chemically. Thus the atoms of radioactive $^{24}_1$Na will behave in the same way as those of stable $^{23}_1$Na in a chemical system. So what? The point is that it is easier to follow the radioactive $^{24}_1$Na atoms than it is to detect the non-radioactive $^{23}_1$Na atoms, due to the exquisite sensitivity associated with the detection of radioactivity. The other unique advantages of radiotracer experiments include their simplicity and small expense (compared to competing technologies such as mass spectrometry). In a well-designed experiment, the presence of radiotracers does not affect the system under study and any analysis is non-destructive. Interference from other species that may be present is not important (as compared to conventional methods of analysis where interferences may thwart the analysis.) Perhaps the most outstanding advantage of the use of radioisotopes is the opportunity offered to trace dynamic mechanisms. Such biological phenomena as ion transport across cell membranes, turnover, intermediary metabolism, or translocation in plants could, before the advent of radiotracer methods, be approached only indirectly. Medical imaging with radiotracers is another example of the ability to study dynamic processes.
Atomic Nucleus

The atomic nucleus is a tiny massive entity at the center of an atom. Occupying a volume whose radius is 1/100,000 the size of the atom, the nucleus contains most (99.9%) of the mass of the atom. In describing the nucleus, we shall describe its composition, size, density, and the forces that hold it together. After describing the structure of the nucleus, we shall go on to describe some of the limits of nuclear stability.
The nucleus is composed of protons (charge = +1; mass = 1.007 atomic mass units (amu)) and neutrons. The number of protons in the nucleus is called the atomic number Z and defines which chemical element the nucleus represents. The number of neutrons in the nucleus is called the neutron number N, whereas the total number of neutrons and protons in the nucleus is referred to as the mass number A, where \( A = N + Z \). The neutrons and protons are referred to collectively as nucleons. A nucleus with a given N and Z is referred to as a nuclide. Nuclides with the same atomic number are isotopes, such as \(^{12}\text{C}\) and \(^{13}\text{C}\), whereas nuclides with the same N, such as \(^{16}\text{O}\) and \(^{17}\text{O}\), are called isotones. Nuclides such as \(^{14}\text{N}\) and \(^{14}\text{C}\), which have the same mass number, are isomers. Nuclides are designated by a shorthand notation in which one writes \(\text{Chemical Symbol}\), that is, for a nucleus with 6 protons and 8 neutrons, one writes \(^{14}\text{C}\), or \(^{14}\text{O}\), or just \(^{14}\text{C}\).

The size of a nucleus is approximately 1 to 10 \(10^{-15}\) m, with the nuclear radius being represented more precisely as 1.2 \(A^{1/3}\times10^{-13}\) m. We can roughly approximate the nucleus as a sphere and thus we can calculate its density.

\[
\text{density} = \frac{\text{mass}}{\text{volume}} = \frac{A \times 1.66 \times 10^{-27} \text{kg}}{\frac{4}{3} \pi r^3} = \frac{A \times 1.66 \times 10^{-27} \text{kg}}{\frac{4}{3} \pi (1.2A^{1/3} \times 10^{-13} \text{m})^3}
\]

where \(1.66 \times 10^{-27}\) kg is the mass of the nucleon. Thus the nuclear density is about 200,000 tonnes/m\(^3\) and is independent of A. Imagine a cube that is 1 mm on a side. If filled with nuclear matter, it would have a mass of about 200,000 tonnes. This calculation demonstrates the enormous matter/energy density of nuclei and gives some idea as to why nuclear phenomena lead to large energy releases.

Of the 6,000 species of nuclei that can exist in the universe, about 2,700 are known, but only 270 of these are stable. The rest are radioactive, that is, they spontaneously decay. The driving force behind all radioactive decay is the ability to produce products of greater stability than one had initially. In other words, radioactive decay releases energy and because of the high energy density of nuclei, that energy release is substantial. Qualitatively we describe radioactive decay as occurring in three general ways: \(\alpha\), \(\beta\), and \(\gamma\)-decay. Alpha-decay occurs in the heavy elements, and consists of the emission of a \(^{4}\text{He}\) nucleus. Beta-decay occurs in nuclei whose Z/N ratio is different from that of a stable nucleus and consists of a transformation of neutrons into protons or vice versa to make the nucleus more stable. Gamma-decay occurs when excited nuclei get rid of some or all of their excitation energy via the emission of electromagnetic radiation, or via the radiationless transfer of energy to orbital electrons.

The force responsible for holding the neutrons and protons together within the very small nuclear volume must be unusually strong. The nuclear force, or strong interaction, is one of the four fundamental forces of nature (namely, the gravitational, electromagnetic, strong, and weak forces). The nuclear force is charge-independent, meaning that the nuclear force between two protons, or two neutrons, or a neutron and a proton, is the same. The nuclear force is short-ranged, meaning it acts over a distance of \(10^{-15}\) to \(10^{-14}\) m, that is, the size of nuclei. Of course the nuclear force is attractive, as it binds the nucleons in a nucleus. But some experiments have shown the nuclear force has a "repulsive core," meaning that at very short distances,
the force switches from attractive to repulsive, preventing the nucleus from collapsing in on itself. The nuclear force is an "exchange" force, resulting from the virtual exchange of pions (short-lived particles with integral spin, produced1 mutually in nuclear reactions) between interacting nucleons. More recently we have come to understand the nuclear force using a theory called quantum chromodynamics (QCD), which describes protons and neutrons as being made up of quarks. In particular, the proton is thought of as a combination of two up quarks (uu) and a down quark (d), whereas the neutron is thought to consist of one up quark (u) and two down quarks (dd). The up and down quarks are small particles with charges of +2/3 and −1/3, respectively. The quarks account for approximately 2 percent of the mass of the proton. The rest of the mass consists of gluons, which are the particles exchanged between the quarks to bind them together. The force acting between the quarks has the unusual property of being small when they are close together, and increasing as the distance between them grows. Because of this, no isolated quarks have been observed in nature.

In close analogy to atomic structure, we speak of the structure of various nuclei. Many nuclear properties can be described using a nuclear shell model in which the nucleons are placed in orbitals like electrons in atoms. These orbitals and their properties are predicted by applying quantum mechanics to the problem of defining the states of the nucleons, which move under the influence of the average force in the nucleus. Like atoms, there are certain configurations of nucleons that have special stability, for example, the so-called magic numbers akin to the inert gas structures in chemistry. In addition to those nuclear properties that are best described in terms of a shell model, there are other properties that seem to be best explained by the large-scale collective motion of a number of nucleons. These motions lead to nuclear rotations and vibrations, which are described by a nuclear collective model.

Current research on nuclei, their properties, and the forces that hold them together focuses on studying nuclei at the limits of stability. The basic idea is that when one studies nuclei under extreme conditions, one then has a unique ability to test theories and models that were designed to describe the "normal" properties of nuclei. One limit of nuclear stability is that of high Z, that is, as the atomic number of the nucleus increases, the repulsion between the nuclear protons becomes so large as to cause the nucleus to spontaneously fission. The competition between this repulsive Coulomb force and the cohesive nuclear force is what defines the size of the Periodic Table and the number of chemical elements. At present there are 112 known chemical elements, and evidence for the successful synthesis of elements having the atomic numbers 114 and 116 has been presented.

Another limit of nuclear stability is the extreme of the neutron to proton ratio, N/Z. For certain very neutron-rich nuclei, such as \(^{11}\)Li, an unusual halo structure has been observed. In halo nuclei, a "core" of nucleons is surrounded by a "misty cloud, a halo" of valence nucleons that are weakly bound and extend out to great distances, analogous to electrons surrounding the nucleus in an atom. Halo nuclei are fragile objects, are relatively large, and interact easily with other nuclei (have enhanced reaction cross sections). The halo nucleus \(^{11}\)Li, which has a Li core surrounded by two-neutron halo is shown in Figure 1. \(^{11}\)Li is as large as \(^{208}\)Pb. \(^{11}\)Li and other
two-neutron halo nuclei are three-body systems (2 neutrons and a $^9$Li core), which pose a special challenge to nuclear theorists. They are also examples of Borromean systems, in which the nucleus is no longer bound if any one of the three components is removed. (The name derives from the heraldic emblem of medieval princes of Borromeo, which has three rings interlocked in such a way that removal of any one ring will make the others fall apart.) See also Atomic Structure; Rutherford, Ernest.

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Figure 1. Schematic views of the nuclear halo nuclei $^{11}$Li and $^{19}$O that compare them to $^{206}$Pb and the Borromean rings of medieval times. (Source: http://www.phy.anl.gov/rla/index)
Protactinium

Melting Point: 1,506°C
Boiling Point: Unknown
Density: 15.37 g/cm³
Most common ion: \(\text{Pa}^{4+}\), \(\text{PaO}(\text{OH})_{2}^{2-}\)

An isotope of protactinium (having mass number 234 and a half-life of 1.1 minutes) was first identified by Kasimir Fajans and O. Gohring in 1913 as a short-lived member of the naturally occurring \(^{238}\text{U}\) decay series and was given the name \(\text{kevlenium}\), meaning brief. The existence of protactinium was confirmed in 1918 when another isotope of protactinium (of mass 231 and a half-life of \(3.3 \times 10^4\) years) was studied independently by Otto Hahn and Lise Meitner in Germany, and by Frederick Soddy and John Cranston in Great Britain. The current name of the element is a shortened version of the original \(\text{protaacinium}\), derived in part from the Greek \(\text{pret}\), meaning parent, protactinium thus meant parent of actinium (its decay product). There are twenty-four known isotopes of \(\text{Pa}\), having mass numbers ranging from 214 to 238, the most stable isotope being \(\text{\text{Pa}}^{231}\). Protactinium \text{metal}

is silvery and relatively nonreactive. It occurs at ppm levels in uranium ores and is extracted from these ores. There are about 125 grams (4.4 ounces) of protactinium in the world today. Its ground state electronic configuration is \([\text{Rn}]\text{SF}^76\text{d}^{7}\text{f}\), placing it in Group IIIB. Its principal oxidation state

is +5, but there is no stable \(\text{Pa}^{3+}\) ion because it is hydrolyzed so quickly to species such as \(\text{PaO}_2(\text{OH})_{2}^{2+}\), or forms complexes with anions such as fluoride. Protactinium in its +4 state may exist in \text{aqueous solution} or in compounds. The most important solid compound of protactinium is \(\text{Pa}_{2}\text{O}_{5}\), see also Actinium; Berkelium; Einsteinium; Fermium; Lawrencium; Mendeleevium; Neptunium; Nobelium; Plutonium; Rutherfordium; Thorium; Uranium.

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Bibliography

Plutonium

MELTING POINT: 640°C
BOILING POINT: 3,228°C
DENSITY: 19.84g/cm³

MOST COMMON IONS: Pu⁺⁺, Pu⁺⁺⁺, PuO₄²⁻, Pu₂O₇⁴⁻, PuO₂⁻⁻

Plutonium was discovered by Glenn Seaborg, Edwin McMillan, Joseph Kennedy, and Arthur Wahl in 1940. They prepared a new isotope of neptunium, ²³⁷Np, which decayed by β-emission to ²³⁶Pu.

\[ {}^{238}\text{U}(d,2n){}^{239}\text{Np} \rightarrow {}^{239}\text{Pu}(t_{1/2} = 87.7\text{y}) \]

Their work as part of the Manhattan Project was kept secret and was finally reported in 1946, after World War II, although the existence of plutonium had been revealed to the world earlier, when the atomic bomb was dropped over Nagasaki, Japan. There are sixteen isotopes of plutonium, having mass numbers ranging from 232 to 247. The principal isotopes of Pu are those having mass numbers 238, 239, 240, 241, 242, and 244. Ton quantities of ²³⁹Pu (having a half-life of 2.4 × ₀⁶ y) are available. The isotope ²³⁹Pu is the source material for nuclear weapons and is produced via neutron capture reactions on ²³⁸U in nuclear reactors.

\[ {}^{238}\text{U}(n,γ){}^{239}\text{U} \rightarrow {}^{239}\text{Np} \rightarrow {}^{239}\text{Pu} \]

About 110 tons of ²³⁹Pu are generated in nuclear power plants each year, with approximately 40 percent of the energy produced in the nuclear fuel cycle coming from ²³⁹Pu. About three times as much electricity is generated from ²³⁹Pu in the United States as from oil-fired electrical generating plants.

The ground state (outer orbital) electronic configuration of Pu is [Rn]⁵⁷f⁷σ². The most stable oxidation state for plutonium ions in solution is +4, although appreciable amounts of plutonium in its +3, +5, and +6 oxidation states can exist. The aqueous chemistry of plutonium is further complicated by the successive, stepwise hydrolysis of Pu(IV) compounds to form polymers of colloidal dimensions. Plutonium is the transuranium element that is most abundant in the environment, due to the atmospheric testing of nuclear weapons during the 1950s and 1960s that deposited approximately 4.2 tons of plutonium in the environment. Most of this plutonium is in the soil, in which it has no discernable effects. See also Actinium; Berkelium; Einsteinium; Fermium; Lawrencium; Mendelevium; Neptunium; Nobelium; Protactinium; Rutherfordium; Seaborg; Glenn T. Seaborg; Thorium; Uranium.

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Bibliography

Rutherfordium

MEETING POINT: Unknown
BOILING POINT: Unknown
DENSITY: Unknown
MOST COMMON IONS: Unknown

Rutherfordium is the first transactinide element. It was discovered in 1969 by Albert Ghiorso and his coworkers, who carried out the reactions $^{252}$Cf

$(^{126}$C $4n) \rightarrow 257$Rf (half-life of approximately 3.8 seconds) and $^{259}$Cf $(^{126}$C, 3n) $(\rightarrow 259$Rf (half-life of approximately 3.4 seconds). There are ten known isotopes of rutherfordium, having mass numbers that range from 253 to 262, the isotope with the longest measured half-life being $^{260}$Rf (half-life of approximately 1.1 minutes). After its discovery, it was postulated that rutherfordium, as the first transactinide element, should be a member of a new, fourth transition series, extending from Z=104 to Z=112, and an atom in which the 6d electronic shell is filled. The outer orbital electronic configuration of Rf should be [Rn]7f$^{14}$5d$^{10}$6s$^2$. As such, it should behave similarly to its analogs titanium and hafnium and should form volatile tetrachlorides that are less volatile than HCl. The first gas chromatographic studies showed that rutherfordium forms more volatile chlorides than hafnium—a deviation from Periodic Table trends that was predicted from relativistic calculations. The first aqueous chemistry studies showed the neutral and anionic complexes of Rf to behave like the corresponding complexes of the Group IVB elements rather than of the trivalent actinides, demonstrating that Rf is indeed a transactinide. Detailed studies of the extraction chromatography of Rf complexes have shown chemical behavior similar to that observed of Rf complexes in gas chromatographic experiments—that is, Rf does not behave as expected according to a simple extrapolation of Periodic Table trends, but in a more complicated manner. For example, in the extraction of substances with tributylphosphate, the extraction sequence Zr > Rf > Hf is observed. These and other experiments show that the aqueous chemistry of rutherfordium is a challenge to current relativistic molecular theories. See also Actinium; Berkelium; Einstenium; Fermium; Lawrencium; Mendelevium; Neptunium; Nobelium; Plutonium; Protactinium; Thorium; Uranium.

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Bibliography

Comparison of Measured and Calculated Mass Distributions of Fission Fragments in Proton Induced Fission of $^{232}$Th, $^{235}$U, $^{238}$U and $^{237}$Np at Intermediate Energies.

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Abstract. The mass distributions of fragments produced by proton-induced fission of $^{232}$Th, $^{235}$U, $^{238}$U and $^{237}$Np at proton energies of 50 and 96 MeV have been measured. The measurements were carried out at The Svedberg Laboratory, Uppsala, Sweden using a high precision time-of-flight spectrometer for fission fragments and fission neutrons. The measured mass distributions are compared to ones calculated using the TALYS code. Using proton-induced reactions to simulate the outcomes of more difficult measurements with neutron beams is discussed.

INTRODUCTION

The properties of the fission fragment mass distributions and, in particular, the presence of symmetric and asymmetric mass splits of the fissioning nucleus, remains one of the major questions in fission physics. Study of these properties over a wide excitation energy range may contribute to a better understanding of fission process. Fission fragment mass distributions are relatively well known only at low excitation energies (below 20 MeV). New technological developments, such as ADS, stimulate the study of fission fragment mass distributions at higher energies. This paper presents experimental measurements of the fission fragments. The experimental setup involves a thin-walled stainless steel vacuum chamber with an array of fission fragment detectors inside. Fission fragment velocities were measured using a time-of-flight technique. The fission fragments passed through a pair of microchannel plate detectors (MCP) placed on either side of the target at 90° with respect to the beam. Electrons, knocked out by fission fragments in passing through aluminum oxide foils (first foil: 60 μg/cm², 15 mm in diameter, located at distance of 10 mm from the target, second foil: 60 μg/cm², 30 mm in diameter, located at distance of 80 mm from the target), were focused on MCPs (start and stop detector). After passing through the time-of-flight arm, fission fragments could also strike silicon detectors (30 mm diameter, 80 mm distance from the target), which were used to measure the fragment energies and to act as stop detectors for the time-of-flight measurement. The time resolution of the MCP detectors was 80 ps. This did not worsen the intrinsic fission fragment mass resolution (due to the experiment was done with the use of a high precision time-of-flight spectrometer for fragments and neutrons [1]. Experimental details and results of neutron emission study (neutron multiplicity and energy distributions) are given in [2]. This paper presents experimental measurements of the properties of the fission fragments. The experimental setup involves a thin-walled stainless steel vacuum chamber with an array of fission fragment detectors inside. Fission fragment velocities were measured using a time-of-flight technique. The fission fragments passed through a pair of microchannel plate detectors (MCP) placed on either side of the target at 90° with respect to the beam. Electrons, knocked out by fission fragments in passing through aluminum oxide foils (first foil: 60 μg/cm², 15 mm in diameter, located at distance of 10 mm from the target, second foil: 60 μg/cm², 30 mm in diameter, located at distance of 80 mm from the target), were focused on MCPs (start and stop detector). After passing through the time-of-flight arm, fission fragments could also strike silicon detectors (30 mm diameter, 80 mm distance from the target), which were used to measure the fragment energies and to act as stop detectors for the time-of-flight measurement. The time resolution of the MCP detectors was 80 ps. This did not worsen the intrinsic fission fragment mass resolution (due to the
recoil of emitting neutrons) significantly. This resolution was found to be about 2 amu. As fission neutrons do not change the average velocity of fragments, fission fragment masses defined according to this time-of-flight technique are the primary, “pre-neutron emission” values.

FIGURE 1. Mass yield curves for the proton-induced fission of $^{232}\text{Th}$, $^{235}\text{U}$, $^{238}\text{U}$, $^{237}\text{Np}$ at $E_p=50$ MeV and 96 MeV.

The measured mass distributions (normalized to 200%) are shown in Fig. 1. The results for the fission of $^{232}\text{Th}$ at 50 MeV are compared to the results obtained in [4,5] for the same nucleus at $E_p=53$ MeV by measuring energies of coincident pairs of fission fragments using surface barrier detectors. The results for $^{238}\text{U}$ at 50 MeV are compared to results [6] at the same energy, obtained by time-of-flight technique with two position sensitive avalanche counters, placed at 90° to the beam and having angular acceptance of 56 degrees. The data for $^{232}\text{Th}$ at the higher excitation energy as well as the data for $^{238}\text{U}$ and $^{237}\text{Np}$ are new measurements. Our data agrees with the previous measurements for the widths of the fission mass distributions. However, our data show more pronounced structure for near symmetric mass splits. At 96 MeV for $^{232}\text{Th}$ and $^{238}\text{U}$, a symmetric fission peak in the mass distribution is especially pronounced, particularly for $^{232}\text{Th}$. In comparing data from the various papers [4-6] at the same projectile energy one should take into account that the difference in mass distribution behavior may be due, in part, to an angular anisotropy of “cold” (asymmetric) and more warm (symmetric) fission. The higher symmetric fission yields for $^{232}\text{Th}$ may represent a nuclear structure effect in fission.

MASS DISTRIBUTION CALCULATIONS AND COMPARISON TO EXPERIMENTAL DATA.

Calculations of the fission mass distributions were carried out with the nuclear reaction code TALYS [10]. Fission cross sections were calculated assuming a Hill-Wheeler penetrability through a double-humped fission barrier, including an explicit treatment of the collective effects on the level densities. The competition of all other reaction channels is taken into account over the whole evaporation chain. Subsequently, the fission fragment mass yields are computed per fissioning system in two steps [3]: (i) the
relative contributions of the different fission modes are evaluated using Hill-Wheeler penetrabilities with ground-state level densities and temperature-dependent barrier parameters. (ii) the mass yield curve corresponding to each fission mode is calculated in the framework of the multi-modal random-neck rupture model (MM-RNRM) by Brosa [7]. For each of the reactions studied in the present paper two results are presented: (a) Default TALYS calculations using the fission barriers for the fission modes SL, STI and STII as they are predicted by the Brosa model (calc def) and (b) TALYS calculations with all SL-barriers reduced by 5% (calc 95).

As shown in Figure 1, the calculations are sensitive to the height of the symmetric fission barrier. By means of a mass distribution decomposition into symmetric and asymmetric components, it was determined, that the best fit to the experimental results was obtained with a 5% reduction of the SL barrier. A comparison of the calculated and measured results reveals a systematic underestimation of the mass distribution width. Furthermore, the calculation is not able to reproduce the distinct symmetric hump in the experimental mass yield curve of $^{232}$Th and $^{235}$U. Apparently, the MM-RNRM model overestimates the width of the SL mode and underestimates the width of the ST mode. In general, the fission-product mass yields are predicted within 30% for the mass range 90-140. The pre-fission (pre-equilibrium, $v_{\text{pre}}$, and equilibrium, $v_{\text{eq}}$) neutron multiplicities are a very important input for the theory, because they allow one to estimate the nuclear temperature at the scission point. Measured and calculated numbers of pre-fission neutrons are compiled in Table 1 for the reaction studied in this work. There are some disagreements between theory and experiment as to the relative numbers of pre-equilibrium and equilibrium neutrons. The total number of neutrons, however, agrees on average within the limits of the experimental uncertainties.

<table>
<thead>
<tr>
<th>Target</th>
<th>$^{232}$Th</th>
<th>$^{238}$U</th>
<th>$^{235}$U</th>
<th>$^{237}$Np</th>
</tr>
</thead>
<tbody>
<tr>
<td>Exp.</td>
<td>0.5±0.3</td>
<td>0.5±0.3</td>
<td>0.5±0.3</td>
<td>0.5±0.3</td>
</tr>
<tr>
<td>Calc.</td>
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<td>2.6±0.3</td>
<td>1.5±0.3</td>
<td>1.9±0.3</td>
</tr>
<tr>
<td>$v_{\text{pre}}$</td>
<td>0.3</td>
<td>0.2</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>$v_{\text{eq}}$</td>
<td>3.2</td>
<td>1.3</td>
<td>1.6</td>
<td>1.6</td>
</tr>
<tr>
<td>Exp.</td>
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<td>1.1±0.3</td>
<td>1.1±0.3</td>
<td>1.1±0.3</td>
</tr>
<tr>
<td>Calc.</td>
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<td>2.9±0.3</td>
<td>2.7±0.3</td>
<td>3.0±0.3</td>
</tr>
<tr>
<td>$v_{\text{pre}}$</td>
<td>0.4</td>
<td>0.3</td>
<td>0.4</td>
<td>0.4</td>
</tr>
<tr>
<td>$v_{\text{eq}}$</td>
<td>5.0</td>
<td>3.0</td>
<td>3.7</td>
<td>3.2</td>
</tr>
</tbody>
</table>

TABLE 1. Number of $v_{\text{pre}}$ and $v_{\text{eq}}$ neutrons emitted prior to fission.
Comparison of fission fragment mass distributions in proton and neutron induced fission.

Fission fragment yields at intermediate neutron energies (above 20 MeV) are of interest in accelerator transmutation of waste. Theoretical studies of the mass distributions in proton- and neutron-induced fission at intermediate energies have been carried out[3]. If the same compound nucleus is produced in neutron and proton-induced reactions, then one expects the characteristics of fission will be identical. In reactions of neutrons and protons with heavy nuclei at energies less than 10-15 MeV, one expects a small contribution of direct reactions and most reactions will involve compound nucleus formation. In a recent paper [8], where variation of the ratio of yields of symmetric and asymmetric fission versus number of neutrons in the compound nucleus at the same excitation energy (14 MeV above fission threshold) was studied, no significant differences between neutron and proton-induced reactions was found. In all cases, the fraction of symmetric fission grows as the number of neutrons in the compound nucleus decreases. Generally speaking, at higher energies, as a result of the difference in direct processes for protons and neutrons, a difference in the properties of the fissioning nuclei (Z,N,E*) is possible, and hence, of integral yields observed in the experiment. Mass distributions obtained in this work and in [6] for the proton-induced fission of $^{238}$U at proton energies 20-60 MeV were compared to results of [9] for neutron-induced fission of $^{235}$U at neutron energies up to 200 MeV. In all cases the experimental mass distributions were represented by sum of three Gaussians (two of them corresponding to asymmetric fission and one to symmetric fission). It was assumed that positions of maximum of the Gaussians were defined by the mass of composite nucleus (corrected for pre–fission neutron emission), and that the widths and areas of the Gaussians corresponding to asymmetric component of fission, are equal. The symmetric fission yields so determined are presented in Fig 2 as a function of the energy of the incident particles. It is seen that in each reaction the fraction of symmetric fission increases as the incident nucleon energy increases, but the rate of increase decreases with increasing energy. The probability of symmetric fission increases as the number of neutrons in the compound nucleus decreases: from N=147 for $^{238}$U+n to N=146 for $^{238}$U+p and N=144, 143, 142 for $^{237}$Np, $^{235}$U and $^{232}$Th+p, respectively. The observed trends are consistent with calculations[10].

The role of the nuclear structure of the residual nucleus in intermediate energy reactions, similar to the role of compound nucleus in low energy reactions, was noted in [11]. In the latter work, the proton- and neutron-induced fission cross-sections for residual nuclei with the same Z/A values were equal. This was explained by the similarity of the relevant nuclear characteristics (Z,N,E*) prior to fission. Thus, it seems that a determination of the dependence of fission mass distributions on the nuclear structure of the residual nuclei at various incident proton energies may replace measurements with neutrons. The latter are much more labour-intensive and uncertain since intermediate energy neutron fluxes as $10^4$-$10^6$ times lower than those of protons. Moreover the necessity for experimental setups with high detection efficiency (ionization chambers) do not allow direct determination of the masses because they do not measure velocities of fission fragments but their kinetic energies (which are distorted by neutrons emitted by the fragments.) Low intensity neutron beams do not allow experiments with coincidences with neutrons or charged particles following the fission process, and thus information about the energy of the fissioning system at different stages of the fission process is lost.

FIGURE 2. A comparison of the experimental and calculated [10] (calc95) values of the percentage of symmetric fission for proton and neutron induced reactions.
Conclusion

Fission fragment mass distributions were analyzed for a number of newly studied nuclei in a wider energy range and in coincidence with fission neutrons. One was able to study the “sensitivity” of calculations of mass distributions to the symmetric fission barrier height, to compare mass distributions for neutron- and proton-induced fission in the energy range up to 100 MeV, and to make conclusions about the possibility of using measurements with proton beams as surrogates for the more difficult measurements with neutron beams.

References
2. O. Batenkov et. al., Nucl.Phys. A (submitted)
6. V.A. Rubchenya et. al., NIM A 463(2001)653,
11. A. Smirnov et. al., this conference