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Glenn T. Seaborg and Heavy Ion Nuclear Science

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Transactinium Science — A Symposium
Honoring the Contributions of
Professor Glenn T. Seaborg

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Abstract:

Radiochemistry has played a limited but important role in the study of nucleus-nucleus collisions. Many of the important radiochemical studies have taken place in Seaborg 's laboratory or in the laboratories of others who have spent time in Berkeley working with Glenn T. Seaborg. I will discuss studies of low energy deep inelastic reactions with special emphasis on charge equilibration, studies of the properties of heavy residues in intermediate energy nuclear collisions and studies of target fragmentation in relativistic and ultrarelativistic reactions. The emphasis will be on the unique information afforded by radiochemistry and the physical insight derived from radiochemical studies. Future roles of radiochemistry in heavy ion nuclear science also will be discussed.
I. Introduction

Glenn T. Seaborg has had several different careers, any of which would satisfy a lesser person. In this talk, I deal with Seaborg's contributions to science after his return to Berkeley from his position in the Kennedy, Johnson and Nixon administrations. Seaborg set up a new laboratory at Berkeley for radiochemistry, assembled a distinguished group of collaborators and proceeded to re-establish radiochemistry as an important tool for studying nuclear reactions. For the last two decades, Seaborg's laboratory has served as a training ground for young nuclear chemists and a source of stimulation for visitors. His work has touched the forefronts of modern nuclear science, involving attempts to make superheavy elements and to understand nuclear reactions at energies of tens of MeV to hundreds of GeV.

The Berkeley that Seaborg returned to in 1971 was vastly different from the Berkeley he left in 1961. Among other developments, radiochemistry had ceased to be an important tool for studying nuclear reactions, having been eclipsed by techniques using semiconductor radiation detectors and on-line measurements. Even the rudimentary tools of a radiochemical lab, such as multichannel pulse height analyzers, were not available for Seaborg and his group to use. Undaunted, Seaborg assembled a fine group of younger chemists (Kratz, Norris, Liljenzin, et al.) and proceeded to re-establish radiochemistry as a useful tool for the study of nuclear reaction mechanisms.

A schematic representation of typical radiochemical techniques for studying reactions is shown in Figure 1. The simplest radiochemical method is the thick target-thick catcher method shown in the top portion of Figure 1. A beam of projectile nuclei impinges on a thick (~50 mg/cm²) target and the reaction products are stopped in the target (T) and catcher foils placed in the forward (F) and backward (B) directions. From off-line analysis of the radionuclides found in T, B and F, one can calculate the cross section for the formation of a product of given Z and A. By careful integration of these data, correcting for β-decay and non radioactive product nuclei, one can derive mass yield distributions for the reaction (σ(A)). A model-dependent analysis of the relative amount of radioactivity in F, B and T gives one information about the product energies and the momentum transfer in the reaction.

The middle portion of Figure 1 shows a typical setup for the measurement of a fragment angular distribution. The projectile beam strikes a thin target (~100 μg/cm²) and the recoiling product nuclei are stopped in Mylar or aluminum foils that line the walls of a cylindrical scattering chamber. These catcher foils can be cut in any shape and size to achieve the desired resolution in the 4π angular distribution measurement.

The bottom portion of Figure 1 shows the use of differential range techniques to measure fragment energy spectra. Reaction products recoiling from a thin target impinge on stacks of thin Mylar or aluminum foils placed at a given angle with respect to the incident beam. The measured distribution of product radioactivity in the catcher foil stack can be turned into fragment energy spectra using range-energy relationships.

Radiochemical techniques are, of necessity, off-line techniques for studying nuclear reactions. As such, they suffer because the correlations between many observables in the
RADIOCHEMICAL TECHNIQUES

Isobaric yields
\[ \sigma(Z,A) \implies \sigma(A) \]

Angular distributions
\[ d\sigma(\Theta,Z,A)/d\Omega \]

Energy spectra
\[ d^2\sigma(\Theta,Z,A)/d\Omega dE \]

Figure 1. A schematic representation of various types of radiochemical techniques used in studying nuclear reactions.
reaction are lost in radiochemical measurements. This inability to measure the correlations between product characteristics has led to a sharp decline in the use of these techniques. However, the exquisite sensitivity of these techniques (allowing cross section measurements at the picobarn level), the unit Z and A resolution, the simplicity of the apparatus allowing many survey measurements and the lack of detection thresholds that afflict the use of other techniques has assured a continuing interest in the use of these techniques. Seaborg and his collaborators, students, etc. have played a key role in developing and using these techniques.

The first papers to emerge from Seaborg's newly created laboratory for heavy ion radiochemistry dealt with the fragment mass distribution from the interaction of near-barrier $^{40}\text{Ar}$, $^{84}\text{Kr}$ and $^{136}\text{Xe}$ with $^{238}\text{U}$ (Figure 2). In this work, Seaborg and his collaborators showed the utility of radiochemical techniques in revealing the gross characteristics of nuclear reactions. In the fragment mass distribution for the $^{40}\text{Ar} + ^{238}\text{U}$ reaction, one can identify the relative importance of quasielastic scattering (E and F) deep inelastic scattering (C), fusion-fission (A) and the fission de-excitation of transfer and deep inelastic products (B). In the Kr and Xe-induced reactions, one saw a feature labelled G (and called the "goldfinger" in honor of a popular novel by Ian Fleming and the later movie). This class of reaction products was unexpected and puzzling until people recognized them as the non-fissioning remnants of the target nucleus. The fission of these nuclei had been inhibited by the high fission barriers in the Au region. Their yield increased as the distributions of the target-like fragments became broader, i.e., with increasing projectile Z and A.

However interesting these studies were, the clear cut primary goal of Seaborg and his collaborators at this time was the synthesis of superheavy nuclei. Figure 3 shows our current understanding of the expected half-lives of the heavy and superheavy nuclei. The main feature of our current understanding is that we think there should be a peninsula of long-lived nuclei extending from the region of known nuclei to the superheavy region (rather than an island of superheavy nuclei separated from the known nuclei by a "sea of instability"). Superheavy nuclei are defined as those nuclei whose half-lives increase with increasing Z (rather than decrease as expected from the occurrence of spontaneous fission). The half-lives of the longest lived superheavy nuclei (Z = 112) are expected to be $\sim 10^5$ sec. No matter how this situation changes in the future, we will always be blessed (cursed) with the allegorical language (islands, peninsulas, seas, mountains, etc.) introduced by Seaborg in his many popular accounts of superheavy nuclei.

The most definitive experiments to synthesize superheavy nuclei involved the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction and were carried out at Berkeley involving Seaborg's group. In Figure 4, I show the upper limit of the superheavy production cross sections that were measured in these reactions. Here the superior sensitivity of radiochemical techniques was used to establish (for the longer-lived nuclei) the lowest upper limit cross sections $\sim 10^{-35}$ cm$^2$. Unfortunately, hindsight has shown use that this failure to synthesize the superheavy nuclei was expected. Using Armbruster's systematics of s-wave fusion cross sections, we would estimate a fusion cross section for the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction of $\sim 3 \times 10^{-32}$ cm$^2$. The excitation energy of the composite species can be calculated to be $\sim 28$ MeV, allowing 2-3 chances to fission. A simple-minded, estimate of the energy independent average value of $\Gamma_r/\Gamma_f$ would be $\sim 10^{-2}$, resulting in an estimated production cross section for superheavy nuclei of $10^{-38} - 10^{-36}$ cm$^2$, well below the experimental upper limit cross sections.
Low Energy Nuclear Reactions
The first studies

\[ {}^{40}\text{Ar} + {}^{238}\text{U} \]

\[ {}^{84}\text{Kr} + {}^{238}\text{U} \]

\[ {}^{136}\text{Xe} + {}^{238}\text{U} \]

Figure 2. Fragment isobaric yield distributions for the reaction of \(^{40}\text{Ar}, {}^{84}\text{Kr}\) and \(^{136}\text{Xe}\) with \(^{238}\text{U}\) (ref. 3-5)
Figure 3. A representation of the predicted half-lives of heavy and superheavy nuclei vs. their atomic number Z and their mass number A. The height of each bar is proportional to the logarithm of the half-life in seconds.
Attempts to Synthesize Superheavy Nuclei

Figure 4. The upper limits for the production of superheavy nuclei in the $^{48}$Ca + $^{248}$Cm reaction.\textsuperscript{7}
Undaunted by these failures, Seaborg and collaborators, particularly Darleane Hoffman, showed how multinucleon transfer reactions could be used as effective tools to synthesize new heavy nuclei (Figure 5). The upper left panel of Figure 5 shows a typical set of experimental data showing the production of Bk, Cf, Es and Fm nuclei from the Ne + 248Cm reaction. Once again, the exquisite sensitivity of radiochemical techniques is shown in these carefully done measurements of minuscule cross sections. Because of the low production cross sections, few, if any, detailed studies of the mechanism(s) operating in these reactions have been done. However, the work of Hoffman and Hoffman\(^{10}\) and Magda et al\(^{11}\) has demonstrated a key, unexpected feature of these reactions, i.e., the production of "cold" reaction products (E* < 10 MeV). The excellent agreement between the experimental data and the calculations appears to only occur if one assumes E\(_{\text{product}}^*\) < 10 MeV. These reactions promise to be important tools for production of new transuranium nuclei.

The efforts of Seaborg's group were not restricted to the study of low energy nuclear collisions. Instead Seaborg and collaborators brought radiochemical techniques to bear on a wide variety of nuclear reactions. In the late 1970's, Seaborg and collaborators measured the target fragment mass yield distributions for many relativistic nuclear collisions. Figure 6 shows a sample of this type of data. These data\(^{12}\) proved not to be very definitive concerning the mechanisms of these reactions as shown in Figure 7. In Figure 7, we compare the data of Figure 6 with predictions of two disparate models for the collisions, the intranuclear cascade model\(^{13}\) and the nuclear firestreak model.\(^{14}\) Despite the significant differences between these two models that emphasize the nucleon-nucleon interaction and collective aspects, respectively, both models seem to do an equally acceptable job of representing the data. This insensitivity of the theoretical models to the results might indicate the property being measured, the fragment mass yield distribution, is largely determined by the collision geometry and the rough distribution of excitation energy in the target residues, i.e., "gross" features of the reactions.

One result from these series of measurements has not been fully appreciated. This is the finding that the fragment N/Z distributions were very narrow, much narrower than one would expect by simple stochastic considerations (Figure 8). Morrissey and co-workers\(^{15}\) pointed out that the narrowness of these charge distributions reflected an intrinsic correlation between neutrons and protons in excited nuclear matter. These workers made a crude model for this effect in terms of the zero point oscillations of the giant dipole resonance, which fit a large amount of experimental data. Bondorf and collaborators\(^{16}\) cast these same ideas more general terms involving isosprin correlations in the nuclear ground state. Nifenecker\(^{17}\) utilized these ideas to explain the shape of the charge distribution in fission. I suspect that these concepts might find further fertile ground in understanding the N/Z distributions in energetic nuclear matter as seen in other heavy ion reactions.

The ability of simple phenomenological models to describe the results of radiochemical studies of intermediate energy and relativistic nuclear collisions has largely been restricted to estimates of the cross sections. As had been established previously for p-nucleus collisions, the momentum transfer to the target nucleus in relativistic nuclear collisions appears to have been grossly overestimated (Figure 9). The work of Seaborg and collaborators\(^{18}\) has documented this problem thoroughly.

-8-
Figure 5. A composite of the measured cross sections for the production of heavy nuclei using multinucleon transfer reactions. (1) 115 MeV $^{20}$Ne, (open symbols), 116 MeV $^{22}$Ne (solid symbols) + $^{248}$Cm (2) 245 MeV $^{40}$Ar + $^{248}$Cm (3) (a) 97 MeV $^{16}$O + $^{254}$Es (b) 97 MeV $^{18}$O + $^{249}$Cf. In panels 2 and 3, the dashed lines indicate the predictions of ref. 10 and 11, respectively.
Figure 6. Measured 12isobaric yield distributions for relativistic heavy ion-\(^{238}\)U reactions.
Figure 7. The data of Figure 6 compared to the predictions of the intranuclear cascade model\textsuperscript{13} (solid histogram) and the nuclear firestreak model\textsuperscript{14} (dashed histogram).
A comparison of the measured Au fragment charge distributions from the 25 GeV/nucleon $^{12}$C + $^{208}$Pb reaction with the predictions of a "hypergeometric (stochastic)" model and the nuclear giant dipole resonance model (solid line-without evaporation, dashed line, with evaporation).

Figure 8.
Radiochemistry has also played a role in the latest studies of ultrarelativistic nuclear collisions. Radiochemical techniques\textsuperscript{19} have extended the general range of validity of \"limiting fragmentation\" to the production of intermediate mass fragments (IMFs) in ultrarelativistic nuclear collisions (Figure 10a). Radiochemists\textsuperscript{20} discovered one of the few surprises of our studies of ultrarelativistic reactions, the finding of very unusual angular distribution for the IMFs produced in these reactions. These workers found that a crude range-weighted measure of the fragment angular distribution, F/B, had a value of 0.85 in the interaction of 14 GeV/nucleon \textsuperscript{16}O with \textsuperscript{197}Au (Figure 10b). This measurement is the first example of an F/B value less than unity and must imply an unusual fragment angular distribution in the cm system. Grabez\textsuperscript{21} has corroborated this finding.

The high intensity accelerator beams available for the study of intermediate energy nuclear collisions have allowed the fullest use of radiochemical techniques for studying nuclear reactions. Once again, Seaborg and his associates have played a central role in these efforts. In a series of measurements involving various heavy targets (\textsuperscript{154}Sm, \textsuperscript{165}Ho, \textsuperscript{197}Au and \textsuperscript{238}U) Seaborg et al. found that as the projectile energy increases, for a given projectile-target combination, the fraction of primary target-like fragments that decay by particle emission relative to fission increases, with heavy residue formation (particle emission) becoming the dominant mode of de-excitation\textsuperscript{22} (Figure 11). Thus the study of these heavy residues becomes an important aspect of the study of intermediate energy nuclear collisions.

The measurement of heavy residue properties is difficult because of their low energies\textsuperscript{23} (Figure 12). In reactions induced by 85 MeV/nucleon ions, the residue energies are \textasciitilde 100 keV/nucleon. The threshold-free differential range technique is ideal for measuring these low fragment energies. Coupled with unit Z and A resolution, one has been able to make significant measurements of some features of intermediate energy nuclear collisions. For example, Seaborg et al.\textsuperscript{24} were able to show the disappearance of the deep inelastic reaction mechanism in Xe-Au collisions as the projectile energy increased from 21 to 45 MeV/nucleon (Figure 13). In deep inelastic reactions, the heavy residue energies result from the Coulomb repulsion of the touching fragments (dashed line). This behavior was observed in the 21 MeV/nucleon \textsuperscript{129}Xe + \textsuperscript{197}Au reaction but not the 45 MeV/nucleon \textsuperscript{129}Xe + \textsuperscript{197}Au reaction.

Without denigrating the scientific output of Seaborg and his collaborators, perhaps it is fair to say that the most important contribution of Seaborg's group to heavy ion science is Glenn's influence upon the people who worked with him (Figure 14). In Figure 14, I show a list of the co-authors of the papers cited in this review with the names of students being underlined. The list is a subset of a Who's Who of nuclear chemistry. Regrettably I have only had the time to discuss a small subset of all the scientific work done in Glenn Seaborg's research group during this time. To partially rectify this oversight, I include, as on Appendix of this document, a list of all the papers from this group during this period, with the names of participating students being underlined.

As one who has had the pleasure of working with Glenn for over 15 years, I am deeply grateful for his guidance, his enthusiasm, his scientific wisdom and his unique ability to create the opportunity for all of us who worked with him of participating in research at the forefront of nuclear science.
The measured values of the longitudinal component of the neutron-deficient target fragment velocities from the reaction of 4.8 GeV/nucleon $^{12}\text{C}$ with $^{238}\text{U}$ compared to predictions of the intranuclear cascade model and the nuclear fire streak model (ref. 18).
Radiochemistry and Ultrarelativistic Nuclear Collisions

Figure 10. a) Excitation functions for the production of IMFs in p-nucleus and nucleus-nucleus collisions.
b) Energy dependence of the F/B ratio for $^{24}$Na produced in the reaction of protons and heavy ions with $^{197}$Au.
Figure 11. Excitation functions for fission and heavy residue production in C-Au and Ar-Au collisions.
Figure 12. Heavy residue energy spectra from the 85 MeV/nucleon $^{12}$C + $^{197}$Au reaction.
Figure 13. Mean heavy residue kinetic energies from the reaction of 45 MeV/nucleon $^{129}$Xe with $^{197}$Au. Also shown are the expectations for deep inelastic scattering (dashed line) and the 21 MeV/nucleon $^{129}$Xe + $^{197}$Au reaction.
The People Who Contributed to the Work Described.

Y. Agarwal
K. Aleklett
P. Armbruster
T. Blaich
M. Bronikowski
W. Brüchle
M. Brügger
C. Casey
Y.Y. Chu
J.B. Cumming
W.R. Daniels
H. Dornhöfer
J.P. Dufour
M. Fowler
C. Frink
H. Gäggeler
K. Gregorich
N. Greulich
H. Groening
H. von Gunten
P. Haustein
G. Herrmann
F.P. Hessberger
V. Hickmann
N. Hildebrand
D.C. Hoffman
S. Hofmann
K. Hulet
B. Jacak
S. Katcoff
J. Kratz
J. Landrum
D. Lee
M. Leino
P. Lemmertz
M. Lerch
J.O. Liljenzin
Y.F. Liu
R. Lougheed
W. Loveland
C. Luo
W. Marsh
P. McGaughey
K. Moody
D.J. Morrissey
G. Münzenburg
A.E. Norris
M. Nurmia
R.J. Otto
K. Poppensieker
N.T. Porile
W. Reisdorf
M. de Saint-Simon
M. Schädel
K.-H. Schmidt
J.H.R. Schneider
W.F.B. Schneider
L. Sihver
K. Sümmerer
N. Trautmann
D. Vermeulen
R. Welch
J. Wild
P. Wilmarth
G. Wirth
Z. Xu
S. Yashita

Figure 14. Names of Seaborg's coworkers in the papers discussed in this review. Names of students are underlined.
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Appendix


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