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NUCLEAR CHEMISTRY RESEARCH

Progress Report

November 1, 1974 - October 31, 1975

Nathan Sugarman and Anthony Turkevich

The University of Chicago

Chicago, Illinois 60637

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ANNUAL PROGRESS REPORT ON NUCLEAR CHEMISTRY RESEARCH

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October 31, 1975

ABSTRACT

The major effort in the high-energy nuclear chemistry program has been the study of cross sections and kinematic properties of products formed from the interaction of 1-300 GeV protons with heavy- (Bi and U) and medium-mass (Cu, Ag) nuclei. The products studied ranged in mass from ~20 to that of the target nucleus. Except in the case of near-target product nuclei ($\Delta A \sim 10$-20), no substantial change in cross section or recoil properties were observed at 300 GeV from those measured at 11.5 GeV. New experiments are in progress to relate the production mechanisms of selected products to fundamental particle interactions and the production of exotic particles.

Extensive preparations have been made for another experiment to be performed soon at higher intensity on the search for polyneutron aggregates at the LAMPF Accelerator.

Work is continuing (in collaboration with Los Alamos and NOAA) on the use of stable isotope tracers (methane-20 and 21) for the study of long-ragne transport and diffusion phenomena in the atmosphere.

A collaborative effort (with Dr. George W. Reed, Jr., ANL) on the study of extraterrestrial objects (meteorites and lunar samples) is continuing. The work has involved correlations among trace and minor elements, the evolution of the lunar regolith, differentiation of lunar magmas, and a study of lunar agglutinates.

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Senior Investigators: Nathan Sugarman and Anthony Turkevich

Assisted by: H. R. Heydegger, S.-K. Chang, Karoline Wielgoz, Jerome LaRosa, James Cadieux, Dale Boyce, T. Economou, Robert A. Johns.

This document is a Technical Progress Report for the period November 1, 1974, to October 31, 1975, on nuclear chemical research performed under Contract E(11-1)-1167 at The University of Chicago. This report contains:

1) Reprints of four papers published during this period.

a) "Recoil Properties of Ta Isotopes Produced in the Interaction of High-Energy Protons with Bi and U" by S. K. Chang, M. C. Cheney, and Nathan Sugarman, Physical Review C10, 2467 (1974).

ABSTRACT

Thick-target thick-catcher recoil properties were determined for neutron-deficient Ta isotopes produced in the interaction of 0.45- and 11.5-GeV protons with Bi and U. The production mechanism was shown to be a binary mass division process at 0.45 GeV. At 11.5 GeV, either a high-excitation-energy ("deep") spallation process or a fission process from light nuclei is a possible mechanism. The half-life of ^{179}Ta, measured on two samples over a period of two years, was determined to be 588 ± 10 days.

b) "High Energy Nuclear Reactions in Our Planetary System" by Anthony L. Turkevich, Proceedings of the Sixth International Conference on High Energy Physics and Nuclear Structure, Santa Fe, and Los Alamos, New Mexico, 1975 June 8-13, American Institute of Physics, pp. 351-363.

ABSTRACT

Nuclear reactions of high energy particles with complex nuclei are occurring at significant rates in many parts of the solar system. The particles involved are galactic cosmic rays, solar flare particles and radiation belt particles. The experimentally observable quantities are stable and radioactive products, gamma rays and neutrons. Most of these have been used in investigations of the state and history of planetary system bodies, and may play an even larger role in future space missions.

c) "Labile and Non-labile Element Relationships Among Apollo 17 Samples" by Stanka Jovanovic and George W. Reed, Jr., Proceedings of the Fifth Lunar Conference, Geochim. Cosmochim. Acta, Suppl. 5, 2, 1685 (1974).

ABSTRACT

The trace element--F, Cl, Br, I, Li, U, Ru, Os, Hg, and Te--contents of a suite of Apollo 17 soils and rocks are reported. Unusually large concentrations of the halogens are found in the orange and associated gray soils and unusually low concentrations are found in dunite and troctolitic granulite. There is a high degree of coherence among the halogens; this suggests that they

evolved under closed system conditions on the moon.

Ion microprobe studies of orange glass spherules have yielded F and Cl concentration ratios consistent with those from the bulk sample.

For most samples, F and Cl are associated with P_2O_5 as apatites. In a few samples, controls other than apatite stoichiometry determined the correlation with P_2O_5. These controls were probably either very late crystallization or very early partial melting and they also appear to have determined the correlation of Li with Cl and possibly that found among other lithophile trace elements. The lack of specificity in terms of sample type suggests that these controls are independent of parental rock or magma.

Ru and Os exhibit several correlation trends indicating chemical processing on the moon. The samples which have meteoritic ratios are interpreted as being primitive and not extensively recycled.

Local mixing of soils from different geologic units is evident in the U data.

Evidence for lunar atmospheric Hg is provided by the Hg found in the sealed sample return containers.

d) "A Study of ^{204}Pb Partition in Lunar Samples Using Terrestrial and Meteoritic Analogues" by R. O. Allen, Jr., S. Jovanovic and G. W. Reed, Jr., Proceedings of the Fifth Lunar Conference, Geochim. Cosmochim. Acta, Suppl. 5, 2, 1617 (1974).

ABSTRACT

^{204}Pb, Bi, Tl, and Zn have been measured in Apollo 16

samples and Apollo 17 orange soil. These data in addition to already reported Apollo 14 and 15 ^{204}Pb results and literature data are used to support the ^{204}Pb-metallic phase coherence. The partition of ^{204}Pb into very fine metallic grains appears to be the result of an impact process rather than metal segregation in a magma.

2) Summaries of work in progress during this period. Papers based on other work completed during this and earlier periods are also in preparation.

SHORT REPORTS ON WORK PERFORMED SINCE LAST PROGRESS REPORT

1. "Studies on New Atmospheric Tracers" by Anthony L. Turkevich (in collaboration with George A. Cowan of Los Alamos Scientific Laboratory, Lester A. Machta of the National Oceanic and Atmospheric Agency and Norman Daly of the Atomic Weapons Research Establishment, England).

A paper on the successful meteorological experiment with methane-21 ($^{13}CD_4$) has been accepted for publication by SCIENCE.

ABSTRACT

Methane-21 ($^{13}CD_4$) is potentially a useful nonradioactive tracer for testing atmospheric transport and diffusion models on a continental scale. In an experiment to demonstrate this long-range utility, the release of 84 grams of methane-21 was detected at distances of 1500 to about 2500 km at concentrations of ~5 x 10^{-17} by volume in the air, by separating methane and measuring the methane-21 content mass spectrometrically.

* * *

Preparations are continuing on another experiment to be performed at Savannah River to check on specific meteorological models using two tracers, $^{12}CD_4$ and $^{13}CD_4$, released at different locations.

2. "Search for Polyneutron Systems at LAMPF" by Anthony Turkevich, H. R. Heydegger*, J. B. Cadieux, J. LaRosa and T. Economou.

No experiments were performed to date during this contract

period because of the long shutdown of the LAMPF accelerator, and the lack of readiness of the hot cell facilities at Los Alamos and of the appropriate facilities and procedures being developed at Chicago. Four alpha detecting assemblies were constructed and tested at Chicago and experiments were performed at Los Alamos during last summer to chase down sources of contamination in local air and water. These could limit the ultimate sensitivity of the experiment. Experiments with the full beam at Los Alamos are tentatively scheduled for mid-December, 1975, and January, 1976.

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*Chemistry Department, Purdue University, Calumet Campus, Hammond, Indiana 46323.

3. "Interaction of High-Energy Protons with Complex Nuclei" by S.-K. Chang, Karoline Wielgoz, R. A. Johns and Nathan Sugarman.

Work has continued during the past year on the cross sections and kinematic properties of products formed in the interaction of 1-300 GeV protons with complex nuclei. For heavy nuclei (Bi and U), the products studied were in the three mass regions, ΔA = 8-20, and ΔA ~ 40, and A ~ 20-50. In the case of the medium-mass target nuclei studied (Cu and Ag), products spanning the mass region ~20 to that of the target nucleus were studied. These experiments on the energy dependence of cross section and recoil properties for thick targets will soon be completed. New experiments designed to study angular distribution and differential

momentum distribution of products from thin targets will be initiated in the near future. Following are short reports on results achieved in the thick-target studies.

A. Heavy Target Nuclei (Bi and U)

1) Study of near-target spallation products from uranium (ΔA = 8-20).

Peripheral reactions involving low deposition energy in the primary reaction are being studied by examination of the cross-section and thick-target recoil-property dependence on bombarding energy and ΔA of the products formed. The nuclides in this mass range (Po, At, Rn, Fr, Ra, Ac, Th, Pa, U) are α-active and are members of radioactive decay chains. Using an α-spectrometer it is possible to distinguish individual α emitters and from growth-decay measurements one can assign charge and mass numbers to the product whose cross section and recoil properties are measured. In this manner the properties of the species whose particles are being observed are determined as well as those of a long-lived progenitor "feeding" this species. The measurements are made by direct counting of α particles in the Al catchers and in an aliquot of the uranium target without chemical separation. That the method is not subject to serious error was demonstrated by the agreement of the results of ^{149}Tb from these experiments with others where radiochemical separations were performed.

Some general results to date are:

a) The cross section for formation of a given product

stays relatively constant as the bombarding energy increases from 1.8 to 300 GeV. For example, the cross section for ^{225}Ac is 1.4 mb at 1.8 GeV, 1.3 mb at 11.5 GeV and 1.6 mb at 300 GeV. Only ^{230}U shows a markedly higher cross section, ~2 mb, at 11.5 and 300 GeV than at 1.8 and 5 GeV, ~0.2 mb (literature values). The constancy of cross section implies that the probability of low-energy deposition processes (peripheral) does not change with bombarding energy.

b) The recoil properties of the near-target nuclides have been compared with the formalism for "deep spallation" products of Crespo, Cumming and Alexander. The kinetic energy results at 300 GeV, ~0.5 MeV, agree well with those calculated, ~0.6 MeV. This is not the case for 11.5 GeV protons (see next section).

c) The most striking result from these experiments in the very large change in the "range" (2W[F+B]) and in the forward-to-backward ratio (F/B) of the products in going from 11.5 GeV to 300 GeV. The "range" values for these near-target species decrease by a factor of ~2, from ~0.12 mg/cm^2 U to ~0.05 mg/cm^2 U, considerably more than that observed for other species, ~10%. Whereas the "range" values at 300 GeV agree with those calculated for "deep spallation," those at 11.5 GeV are in serious disagreement. The F/B values <u>increase</u> by a factor of ~2, from ~1.7 to ~3.7, whereas for all other products studied to date, F/B <u>decreases</u> by ~10-30%. Much of the large increase observed in F/B at 300 GeV is a consequence of the large decrease in the "range".

The large effect of bombarding energy on the recoil properties of these near-target species represents the first big difference in mechanism for production of a product in the multi-GeV energy range. More effort will be expended to assure the correctness of the observations and to explain the basic change in mechanism with bombarding energy.

2) Study of spallation products from uranium ($\Delta A \sim 40$).

The cross-section and recoil properties of Tl, Bi, and Pb products are being examined. These products represent interactions of higher deposition energy than the near-target nuclei being examined by direct α-counting. A major purpose of this effort besides that of examining the systematics of spallation processes is the study of the fissionability of highly excited nuclei, the fission-evaporation competition as affected by excitation energy. Radiochemical separations coupled with X- and γ-ray spectroscopy are used to identify the nuclides. From the comparison of results with 3-, 11.5- and 300-GeV protons it is seen that the reaction mechanism responsible for the production of these nuclides does not change appreciably with proton energy in this energy range. A change in mechanism (from binary division to spallation) had earlier been observed between 0.5 and 11.5 GeV for the production of Ta isotopes from U and Bi.

3) Study of "fragmentation" products from heavy elements, ($A \sim 20-50$).

The products in the mass range 20-50 are considered to be

"fragments" produced by a binary division process from highly excited nuclei. Excitation functions, where available, indicate deposition energies from the interaction of the proton with the target nucleus of the order of 1 GeV. The recoil properties of these fragments should represent those of truly high-energy interaction.

Cross-section and recoil properties of the "fragmentation" products, Na, Mg, P, and Sc from high energy proton bombardment of U and Bi are being determined. The cross-section and recoil properties are essentially independent of proton energy for E_p>10 GeV. Some knowledge of the gross angular distribution (anisotropy) will be obtained from a comparison of "forward-backward" experiments with "perpendicular" experiments. Preliminary results of scattering-chamber experiments performed at Brookhaven National Laboratory show that the angular distribution is anisotropic, with preferential emission of fragments perpendicular to the beam direction.

B. Medium Mass Nuclei (Cu and Ag)

The recoil properties and production cross sections for ~20 nuclides from copper and ~30 nuclides from silver were measured over the proton energy range of 3 GeV to 300 GeV. The radioactivities were measured by Ge(Li) counters on chemically separated samples and on untreated foil samples. The two techniques yielded results in good agreement. The cross sections agreed well with those reported in the literature (English et al., silver; Rudstam

et al., copper). The cross sections and recoil properties do not change appreciably from 3 to 300 GeV. For a copper target, the 2W(F+B) values range from 0.17 mg/cm^2 Cu for ^{61}Cu to 2.50 mg/cm^2 Cu for ^{24}Na. For silver, the 2W(F+B) values range from 0.072 mg/cm^2 Ag for ^{105}Ag to 3.90 mg/cm^2 Ag for ^{28}Mg. The "range" values, as calculated from 2W(F+B), yield the kinetic energies of the product nuclides. It is found that the "range" increases linearly with the mass difference between the target and the product for masses down to A~35. The kinetic energy corresponding to the "range" is in agreement with a spallation mechanism in the same mass range. For products of mass less than 35, the range is higher by 15% than the linear extrapolation from the ranges of the products close to target. It is thought that binary fission contributes to the production of these A<35 nuclides. Further work in this area is planned.

4. "Momentum-Deposition Energy Relation for Highly Relativistic Reactions" by Nathan Sugarman.

The interaction of a highly relativistic proton with a complex nucleus may be written in its simplest form as follows:

$$p + {}^{A}Z \rightarrow ({}^{A'}Z')^{EXCITED} + X$$

where X is a conglomerate of relativistic secondary particles (mostly mesons and at least one baryon) and $^{A'}Z'$ is the residual excited nucleus which later evaporates particles and fragments and/or undergoes fission. If all particles in X are also highly relativistic

and if X has a baryon number of 1, then the forward component of the momentum of $^{A'}Z'$, $p_{||}$, is related to the excitation energy of $^{A'}Z'$, E*, by the simple relation:

$$p_{||} \approx E^*$$

where $p_{||}$ is in the unit GeV/c and E* in GeV. If X is not totally relativistic or if its baryon number differs from 1, then appropriate corrections must be made. This formalism was applied to recoil results of ^{24}Na from U bombarded with 28-GeV and 300-GeV protons and the E* value of the excited nucleus was found to be ~1 GeV, about the same as that deduced from the excitation function of ^{24}Na from U. (The derivation is given in some detail in the accompanying Outline of Talk at Frostavallen Conference, "Measurement of Excitation Energy for Binary Division in High Energy Reactions").

5. "Radiochemical Studies of High Energy Reactions Involving Mesons and other Exotic Particles" by J. LaRosa and Anthony L. Turkevich.

In the interaction of high energy (>1 GeV) particles with complex nuclei, some radioactive products can be formed only if mesons or more exotic particles are involved. A classical case is the formation of ^{65}Ni in the proton bombardment of copper. This nuclide can be formed directly only via reactions of the type (p,X^{++}) where X^{++} must have a baryon number of +1 and involve at least one meson. Literature data on this reaction are available up to 28 GeV. Studies on this reaction and others that may uniquely

involve mesons or other exotic particles have been initiated using the ANL and the FNAL accelerators.

6. "Lunar Sample Studies" by G. W. Reed, Jr.,* S. Jovanovic,** and R. O. Allen, Jr.†

The results summarized herein are based on work carried out with support from the U. S. Energy Research Development Administration and NASA. The facilities available at The Enrico Fermi Institute of The University of Chicago have been utilized in the research reported here.

1) In last year's progress report we alluded to some work as a part of a consortium studying a Boulder at the Apollo 17 landing site. The data acquired have permitted a more detailed consideration of the processes involved in the formation of the boulder. A paper on this work has been submitted for publication in "The Moon," "History of Boulder 1 at Station 2, Apollo 17 Based on Trace Element Interrelationships," by S. Jovanovic and George W. Reed, Jr.

ABSTRACT

Correlations among the trace and minor elements, Cl and Br, Cl and P_2O_5, and Ru and Os present in parent igneous rocks generally survived the processes of boulder breccia formation.

Fractions of the Cl, Br and Hg, that are mobilized by water leaching and/or volatilization at moderate temperatures of $\leq 450^{\circ}C$ place constraints on the thermal history of Boulder 1 and

its component breccias. Since, and possible during, consolidation the boulder has probably not been subjected to temperatures of $\geq 450^{\circ}C$.

The parent rocks of the Apollo 17 boulder and breccia samples studied could have been derived from two initial magmas. Boulder 1, Station 2 gray competent breccias 72255 and 72275 clast #2 appear to be genetically unrelated to gray competent breccia and anorthositic 72215 material or to light friable breccia 72275; they do appear to be related to 72394 (Boulder 2) and 76315 (Station 6 boulder) samples.

Vapor clouds from apparently external sources permeated the source regions of the boulders.

2) New data were obtained in an effort to understand better the evolution of the lunar regolith. Samples of soils, breccias and glass coating breccias were studied in an attempt to observe the effects of local cratering events and rock weathering on the distribution of trace elements, both labile and non-labile. The results of this study are given in a paper submitted for publication in "Proceedings of the Sixth Lunar Science Conference," "Soil-breccia Relationships and Vapor Deposits on the Moon," by S. Jovanovic and George W. Reed, Jr.

ABSTRACT

Sets of Apollo 15 and 17 samples consisting of soil, breccia, and breccia glass coating were selected for a study of regolith evolution. The data are inconclusive with respect to local cratering effects or rock weathering.

A large number of samples from all missions fall into 1 of 5 groups based on their water leachable Cl and Br fractions. A limited number of vapor clouds with distinct labile Cl-Br ratios is suggested. One such vapor cloud may have particular significance since it has a primordial Cl-Br ratio.

3) On the basis of data previously reported, a paper, "Cl and P_2O_5 Systematics: Clues to Early Lunar Magmas," by S. Jovanovic and G. W. Reed, Jr., was prepared and accepted for publication in "Proceedings of the Sixth Lunar Science Conference." We propose that the early several hundred kilometer thick magma ocean at the surface of the moon was not homogeneous. It consisted of convection cells having somewhat different chemistries and the evidence for such cells persists in trace element patterns of lunar rocks and soils. The inhomogeneities in the chemistry may have been due to accretional or to geophysical processes.

ABSTRACT

On the basis of the relationships of trace elements associated with late residual liquids we identify groups of samples which may be representative of complementary end members in the differentiation of lunar magmas. A model for the lunar crustal evolution involving convection cells is proposed to account for the different magmas required.

4) A consequence of the interaction of micrometeorites with lunar surface material is the formation of glass-bonded aggregates called agglutinates. We propose that this process led

to chemical and isotopic fractionations and have demonstrated this with our data on volatile heavy metals and halogens and have used this interpretation to explain literature data on isotopic enrichments.

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* Senior Chemist, Argonne National Laboratory and Senior Research Associate, The Enrico Fermi Institute, The University of Chicago.

**Employed by Argonne National Laboratory.

† Permanent address: Department of Chemistry, University of Virginia, Charlottesville, Virginia.

"Report on Radiation Detection Equipment" by Adam Zafrans, Nathan Sugarman, S.-K. Chang, H. R. Heydegger, and Karoline Wielgoz.

a. Proportional Counting Systems.

Due to the increased demand for methane-flow, beta-proportional counting equipment, six systems were completely overhauled, new detector tubes were made and the assembled units were placed in operation. At the same time, most of the other proportional units were checked thoroughly and malfunctioning components were replaced. Since then, only periodic attention is required, including the replacement of some mechanical timers and printers.

Under consideration is a plan to have the Electronics Shop of The Enrico Fermi Institute design and construct completely solid-state counting units using integrated circuitry only. Eventually it is hoped that the aging existing equipment will be replaced and the mechanical timers will be eliminated.

b. Intrinsic Germanium Low Energy Spectroscopy Systems.

Early in the year we added two important tools for detection of low energy photons (X rays and low-energy gamma rays). One unit was purchased from Princeton Gamma Tech which also included a preamplifier of their design with optical feedback and a matching amplifier. This unit is much less susceptible to catastrophic destruction in case of loss of liquid nitrogen and sudden warm-up. The spectrometer exhibits excellent resolution (400 eV at 6 keV and

650 eV at 122 keV) and is being used with a Nuclear Data 2200 Analyzer. The resolution is lower than optimum because of mismatch of the amplifier and the analyzer.

The second intrinsic detector was acquired from Ortec and coupled to a Nuclear Data 2400 Analyzer System. This unit, too, has excellent resolution (440 eV at 6 keV and 585 eV at 122 keV). A new high frequency High Voltage bias supply was added to this system thus enhancing its excellent overall performance.

Both units have been used intensively throughout the year.

c. Ge-Li Gamma Spectroscopy Systems.

During this past period our Ge-Li detector systems performed well and we had little trouble with the associated electronics.

Concentrating our efforts on the "fail-safe" problem of Ge-Li detectors, we had designed and built in our Electronics Shop simple but accurate and effective liquid nitrogen Level Detectors. We are incorporating one of these into each detector dewar. The "state-of-the-art" solid state circuits used are extremely sensitive and provide us with a double-level two-color visual LED (light emitting diode) nitrogen level check.

Additionally, we equipped our last Ge-Li detector with a high frequency high voltage bias supply in order to eliminate possible over-voltage of the detector.

d. Alpha Counting Systems.

Three new silicon Alpha Detectors were added to our detection equipment. These required a considerable amount of additional electronics (pre-amplifiers, amplifiers) and the construction by the Electronics Shop of 2 special low-voltage power supplies as well as additional cabling to an adjacent counting room in order to share time on our Nuclear Data Analyzers.

e. Wide-beta Beckman System.

We invested considerable time and effort during the latter part of the year and succeeded in upgrading our low-level Beckman beta counting system and its associated peripheral units. The system, which had been inoperative for a time, is now being thoroughly tested for consistent low background.

f. Plotter System

A skillful modification to an older piece of equipment has proved very gratifying. We converted our Houston Omnigraphic plotter from a primarily point plotter to a more desirable pen type, giving us virtually the usefulness of a new instrument.

Effort of Principal Investigators Devoted to Project

Principal Investigator Nathan Sugarman estimates that he devoted about 70% of his time to this project from February 1, 1975 to October 31, 1975 (including 100% for the months of July and September). He estimates that he will devote about 60% of his time from November 1, 1975 to January 31, 1976.

Principal Investigator Anthony Turkevich estimates that he devoted about 30% of his time to this project for the period February 1, 1975 to June 15, 1975, and 50% of his time from June 15, 1975 to October 31, 1975 (the summer under Los Alamos auspices). He estimates that he will devote 25% of his time from November 1, 1975 to January 31, 1976.