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December 15, 1970

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AND

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DISCLAIMER

TECHNICAL PROGRESS REPORT ON NUCLEAR CHEMISTRY RESEARCH

CONTRACT AT(11-1)-1167

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This document is a Technical Progress Report for the period December 15, 1969 to December 15, 1970 on nuclear chemical research performed under Contract AT(11-1)-1167 at The University of Chicago. This report contains summaries of work in progress during the contract year, reprints of two papers published during this period, "Radioactivity Induced in Apollo 11 Lunar Surface Material by Solar Flare Protons" by H. R. Heydegger and Anthony Turkevich, Science 168, 575 (1970) and "Production of Manganese 54 and Zinc 65 from Copper in Thick Targets by 0.45-GeV, 1.0-GeV, and 3.0-GeV Protons" by A. Van Ginneken and Anthony Turkevich, Journal of Geophysical Research 75, 5121 (1970), and preliminary drafts of three manuscripts being prepared for publication. Other papers based on work completed during this and earlier periods are also in preparation.

SHORT REPORTS ON WORK PERFORMED SINCE LAST PROGRESS REPORT

1. "Mass Yield and Charge Distribution in 450-MeV Proton Fission of U^{238}" by John A. Panontin and Nathan Sugarman.

This work has been completed and a manuscript will be sent to The Journal of Inorganic and Nuclear Chemistry for publication. [A preliminary draft of the manuscript is appended to this Report.] The Abstract of this paper follows:

"The independent and/or cumulative cross sections of 26 isotopes of silver, cadmium, indium, tin, and antimony from the fission of U^{238} with 450 MeV protons have been measured. The isotopic yield distribution for these elements are presented along with the charge distribution for five mass chains, 115, 117, 125, 127, and 129. The values of the most probable charge, Zp, for fourteen mass chains were calculated and the variation of A/Zp with A defined for the mass region, 100-130. For A values between 100 and 120, A/Zp and the charge distribution width are relatively constant at values of 2.424 ± 0.004 and 2.90 ± 0.10 Z units, respectively. In the mass region, 125-129, a width of 3.21 Z units and a slightly higher value of A/Zp, 2.430 ± 0.004, are required to account for the data. A mass yield curve encompassing this region of A was also calculated."

2. "Isomer Yield Ratios and Average Spin of Primaries in 450-MeV Proton Fission of U^{238}" by John A. Panontin and Nathan Sugarman.

This work has been completed and a manuscript will be sent to the Journal of Inorganic and Nuclear Chemistry for publication. [A preliminary draft of the manuscript is appended to this Report.] The Abstract of this paper follows:

"The isomer ratio of 24 independently formed nuclides from the fission of U^{238} by 450 MeV protons has been determined in the range of charge 46 to 51. There is a marked dependence of the isomer ratio on the Z of the product, and relatively little or no dependence on A or on the average excitation energy of the cascade nuclei leading to the formation of the product. The isomer ratios increase with increasing Z and for a given Z increase by about a factor of 2 at the neutron number 65.

"The average spin of the primaries leading to a given product was calculated from the isomer ratio by means of statistical theory. Near symmetric fission, the primaries are formed with very little (2-4 $\hbar$) or no spin, and with increasing Z of the product, the average spin of the primaries increases, reaching a value of ~15-19 $\hbar$ at Z of 51 (Sb). Anomalously high isomer ratios, and corresponding spin values, were found for the primaries leading to In^{113}, In^{115}, and In^{117}. This result may arise from the reversal of the normal spin ordering in these nuclei in which the low spin

state is the ground state. The total spin of complimentary primaries, without regard to their respective orientation, is estimated to be 4-8 $\hbar$ near symmetric fission and 19-27 $\hbar$ for an assymetric charge ratio of 1.25 (Nb-Sb)."

3. "Thin Target Cross Sections for Some Cr, Mn, Fe, Co, Ni, and Zn Nuclides Produced in Cu by 82- to 416-MeV Protons" by H. R. Heydegger, C. K. Garrett, and A. J. M. Van Ginneken.

This work has been completed and a manuscript will be submitted to The Physical Review for publication. [A preliminary draft of the manuscript is appended to this Report.] The Abstract of this paper follows:

"Thin target cross sections for the production of the radionuclides Zn^{65}, Ni^{63}, Ni^{56}, Co^{60}, Co^{58}, Co^{57}, Co^{56}, Fe^{59}, Fe^{55}, Mn^{54}, Mn^{52}, and Cr^{51} by 82, 111, 187, 283, and 416 MeV protons incident on natural copper were determined by radiochemical means. The experimental results are compared with values in the literature and with expected trends."

4. "Thin Target Cross Section for the $Al^{27}(p,3p3n)Na^{22}$ Reaction Relative to that for the $Al^{27}(p,3pn)Na^{24}$ Reaction" by H. R. Heydegger, C. K. Garrett, A. J. M. Van Ginneken, P. H. Walpole, and Anthony Turkevich.

Data reduction is continuing for the purpose of determining the Na^{22}/Na^{24} cross-section ratio in Al. The $Al^{27}(p,3p3n)Na^{22}$ cross section should prove especially useful in studies of the

excitation functions for the production of very long-lived species such as Al^{26} ($T_{\frac{1}{2}} \approx 10^6$ yr). A preliminary investigation of the production of Al^{26} from Al by proton bombardment is also being undertaken.

5. "Cross Sections and Recoil Properties of Pb and Tl Nuclides from 450-MeV and 11.5-GeV Proton Bombardment of U^{238}"

by John A. Panontin and Nathan Sugarman.

The cross sections of a number of lead and thallium nuclides from the bombardment of U^{238} by 450-MeV and 11.5-GeV protons have been measured. The thick-target thick-catcher recoil properties of $Pb^{200,201,203}$ have also been measured at the same bombarding energies.

The cross-section data at 450 MeV were analyzed to give the isotopic yield distribution of isotopes of lead. The results were compared with a Monte Carlo cascade-evaporation calculation, employing the cascade results of the Vegas STEP-NO program for 380-MeV protons incident on U^{238}.

A two-vector model analysis of the recoil data was made in order to obtain average values of $p_{\parallel}$, $p_{\perp}$ and p_{evap}, corresponding to the average momentum imparted to cascade nuclei in the direction of the proton beam, perpendicular to it, and the average momentum resulting from the evaporation stage of the reactions. A comparison of the results with those from a similar analysis of reported data for spallation products from Bi and Ta in the MeV and GeV range of bombarding energies, leads to the observations that (1) p_{evap} is linearly dependent upon ΔA and

independent of bombarding energy and (2) $p_{\parallel}$ is linearly dependent upon ΔA for $\Delta A < 15$, and tends to a constant value of approximately 0.4 to 0.5 the momentum of the incident proton in the hundreds of MeV range of bombarding energy for $\Delta A > 15$. This latter result is inconsistent with the momentum-excitation energy relation ($p_{\parallel}$-E*) derived from Monte Carlo cascade data in which $p_{\parallel}$ increases linearly with E*, which, in turn, is proportional to ΔA. A similar situation is observed in the GeV region in which the value of $p_{\parallel}$ is considerably less than that expected from the Monte Carlo cascade data for the excitation energy required to produce the observed products by a cascade-evaporation mechanism. The implication is that considerable momentum is carried off in the cascade stage of the reaction in the forward hemisphere by a number or group of particles.

The results for a spallation mechanism, when extrapolated to neutron-deficient products in the fission-product region from GeV proton bombardments of heavy elements give values consistent with those observed. The values of $p_{\parallel}$ were essentially equal to those observed for $\Delta A \sim 30$ and inconsistent with the $p_{\parallel}$-E* relation derived from Monte Carlo cascade data, not surprising in view of the similar results found at 450 MeV for $\Delta A > 15$.

6. "Radiochemical Analysis of Freshly Fallen Meteorites"*

by H. R. Heydegger and Anthony Turkevich.

Work in this area has largely been suspended during

the studies of the lunar samples (see Section 7). With the conclusion of those studies, it is expected that the investigation of the production of radionuclides by various projectiles in space will be resumed.

Experimental work has been confined to continuing measurements on the long-lived species of interest, particularly those isolated from Pueblito de Allende.†

A paper (a reprint of which is attached to this report), utilizing some of the results of this effort, was published during the year ["Radioactivity Induced in Apollo 11 Lunar Surface Material by Solar Flare Protons", SCIENCE 168, 575, (1970)]. The emphasis in further calculations will continue to be on siliceous materials.

In addition, several manuscripts concerning the studies of radionuclides in the Hamlet, Harleton, and Aroos meteorites are still under prepraration. Measurements of the long-lived species are being continued in order to verify the half-life values.

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* Program supported jointly by a grant from the National Aeronautics and Space Administration (NAS 9-7883).

† Collaborating on this study were G. W. Reed, Jr. of Argonne National Laboratory and J. E. Keith of MSC-NASA.

7. "The Amounts of the Heaviest Elements in Returned Lunar Material"* by G. W. Reed, Jr.**, H. R. Heydegger, A. Turkevich, and J. Collister.

The uranium content and data on its isotopic composition, and the ^{204}Pb content of Apollo 11 lunar soil have been measured by neutron activation techniques. The $^{238}U(n,2n)^{237}U$ and the $^{235}U(n,f)^{140}Ba$ nuclear reactions were used in the uranium determination, and the $^{204}Pb(n,2n)^{203}Pb$ reaction for the lead determination. Because of the unavailability of the Argonne reactor, the irradiations were carried out at the Brookhaven National Laboratory High Flux Beam reactor. The uranium content has been found to be 0.55 ppm with an isotopic composition consistent with that of terrestrial uranium. The ^{204}Pb content is about 5 ppb with some indication of variability in different samples. The uranium result is in good agreement with the results of other workers; the ^{204}Pb result appears to be the first definite identification of primordial lead in lunar material. A manuscript for presentation at the 1971 Lunar Science Conference at Houston describing these results has been prepared.

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* This program is a collaborative one with the Argonne National Laboratory. The effort at the University of Chicago is mainly sponsored by the National Aeronautics and Space Administration with important equipment, machine- and electronics-shop maintenance and technical support from this AEC grant.

** Chemistry Division, Argonne National Laboratory.

8. "Thermal Release of Hg from Terrestrial, Meteoritic and Lunar Material"* by George W. Reed, Jr.† and S. Jovanovic‡.

The manner in which volatile elements and compounds are incorporated in geological material is of importance for deciphering the physicochemical conditions to which the matter has been subjected. Such measurements may also be the only method of determining in what form and how certain trace elements are present. Stepwise linear and isothermal heating experiments have been carried out on ores and silicate rocksand minerals. The release patterns indicate that Hg is "trapped" in various types of sites, some of which may be distinct chemical compounds, some lattice sites, and others, grain boundaries. In many cases, the release appears to be diffusion-regulated. Thus, activation energies may be calculated and compared with those of the rare gases, the only other elements on which detailed studies have been made. Only for meteorites have activation energies been estimated, so far, and these are lower than those for rare gases. This result raises questions on the mechanism of Hg retention since the time the meteorites became small bodies in space.

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* Some of the meteoritic and all the lunar studies were supported in part by NASA contract T76356 with The Argonne National Laboratory.

† Dr. George W. Reed, Jr. is employed as a member of the scientific staff of The Argonne National Laboratory. He is also a Research

Associate of The Enrico Fermi Institute of The University of Chicago.

‡ Employed by the Argonne National Laboratory.

9. "Production of Rare-Earth Nuclides from Uranium by 12-GeV Proton Bombardment" by P. M. Starzyk and Nathan Sugarman.

A study of recoil properties of rare earths produced by the 12-GeV proton bombardment of uranium is in progress. This study involves the use of solid state detectors for nuclide identification and minimal chemical separation of rare earths from each other. A computer program for the analysis of the relatively complex spectra was adopted from that of Dr. J. Routti, formerly of the Lawrence Radiation Laboratory, for use on the 7094 computer of The University of Chicago. A magnetic tape interface is associated with the analyzer. The data are analyzed by use of energy and shape calibrations for known γ-ray peaks stored as part of the program.

Recoil properties of nuclides of lanthanum, cerium, praseodymium, and neodymium resulting from 450-MeV proton bombardment of uranium were determined in the manner just described and found to agree with previously measured results. This agreement provides a check on both the chemistry and the analysis program for the nuclide distribution from 450-MeV proton bombardment. Attempts at fast ion-exchange separation were made, in the event that no distinctive γ rays are present and proportional counting is necessary. Optimal column conditions were established.

Cross sections of nuclides of lanthanum, cerium, and praseodymium formed from 12-GeV proton bombardment of uranium have been determined and are consistent with published values. Presently, data from recoil experiments at this energy are being analyzed.

10. "Recoil Properties of Spallation Products from the Bombardment of Heavy Elements by High-Energy Protons" by Maynard C. Cheney and Nathan Sugarman.

The recoil properties of nuclides considered to be produced by the spallation of heavy elements bombarded by high-energy protons are being investigated. The nuclides being studied are in the region of tantalum and rhenium and target materials used are uranium, bismuth, gold, and tantalum. The short half-lives of several of the expected nuclei made necessary the modification of decontamination procedures in order to provide for fast separation of tantalum and rhenium from solutions. Data from gamma spectroscopy and proportional counting of samples from thick catcher experiments have been subjected to computer analysis for determination of cross sections and recoil properties. Values for F/B of several tantalum isotopes produced from uranium and bismuth at 440 Mev have been found to be in agreement with values extrapolated from literature data for lighter spallation products from other targets. Analysis of the data at 11.5 Gev is as yet incomplete. A similar study of rhenium isotopes produced from the same targets is currently underway, and future experiments will include nuclides of higher mass

in order to clarify the dependence of recoil properties on nuclear composition. It is expected that this information will contribute to the understanding of the interaction of high energy protons with complex nuclei and of the apparently anomalous range results of the neutron-deficient products in the fission mass region.

11. "Angular Distribution and Differential Range Measurements of Isotopes of Iodine and Tantalum from 12-GeV Proton Bombardment of Uranium" by S.-K. Chang and Nathan Sugarman.

The recoil properties of neutron-deficient nuclides formed from high energy proton bombardment of heavy elements in the mass range of the fission products differ markedly from those of the neutron-excess products. In particular, the ratio of the activity projected forward to that projected backward is much closer to unity than expected for products formed from high excitation energy processes and the sum of the forward and backward projected activities (the "effective" range) is much smaller than that of the neutron-excess products. The mechanism for the formation of these neutron-deficient products is not well understood. It is thought that the cascade process involves the injection of a heavy fragment whose momentum affects the recoil properties of these products.

An experiment has been begun to understand this process by a comparison of the angular distribution and differential range distribution of isotopes of iodine and tantalum. Inasmuch as the neutron-excess nuclides of iodine are products of fission,

it is hoped that a comparison of the recoil properties of these with those of tantalum, considered to be formed by spallation, and with the neutron-deficient isotopes of iodine, will lead to an understanding of the formation of the neutron-deficient iodine isotopes.

Two preliminary experiments were performed with the 12-GeV proton beam of the ZGS of The Argonne National Laboratory. The iodine isotopes of mass number 121, 124, 126, 131, and 133 were identified by their characteristic gamma rays and half-lives. The tantalum isotopes in the mass range 173 to 184 were somewhat more difficult to identify since some of their decay schemes are not well established.

The targets for the angular distribution and differential range experiments are prepared by vacuum evaporation of UF_4 on 1/4-mil aluminum foil. The uranium thickness is ~ 0.2 mg/cm^2. The catcher foils are 1-mil aluminum foils for the angular distribution measurements or 1/4-mil Mylar foils for the differential range measurements. The irradiations are to be performed in the internal beam of the ZGS at Argonne National Laboratory.

12. Activities of Anthony Turkevich during period July 1, 1970 to January 1, 1971.

Anthony Turkevich spent six months as a National Science Foundation Senior Postdoctoral Fellow with partial AEC contract support at Professor Marc Lefort's laboratory at Orsay, France. An appreciable amount of time was spent working with Dr. R. Bimbot

of Lefort's nuclear chemistry group in trying to understand the mechanism of (α,xn), (α,pxn) and $(\alpha,\alpha xn)$ reactions in Pb and Bi at alpha energies of 80 to 160 MeV. It appears as if alpha particles of this energy may be thought of as having an appreciable probability of preserving their identity in nuclear matter. The cross sections for collisions of alpha particles with free nucleons are large for collisions involving little energy transfer. Such collisions should be suppressed by Pauli principle considerations in nuclear matter. These considerations help explain why non-compound nucleus reactions constitute a very large fraction of (20-50%) the reactions induced by 80 - 160 MeV alpha particles in heavy nuclei.

13. "Report on Radiation Detection Equipment" by Adam Zafrans, Nathan Sugarman, Karoline Wielgoz, Patricia M. Starzyk, Maynard C. Cheney, George W. Reed, Jr., H. R. Heydegger, and John A. Panontin.

a. Ge-Li Gamma Spectroscopy System

After extensive field testing with the aid of a computer "dumping" program, the lithium-drifted Ge-Li solid state detector system for gamma spectroscopy was made available for computer analysis of spectral data. A computer program was developed to take full advantage of the speed and accuracy of the computer at The University of Chicago. In addition, the system has the capability of storing analyzed spectra and those of "standards" on magnetic tape for instant use.

The recent acquisition of a second Ge-Li detector for gamma spectroscopy, with the electronics on hand, will greatly enhance the versatility and capability of the present detection system. With some modification, the present 4096 channel system can operate effectively as two 2048 channel single parameter systems enabling the simultaneous analysis of two samples. In addition, with the simple changing of a program plug, there are two other possible modes of operation, a two-parameter mode, with several memory configurations, and a single-parameter mode, where either detector can use the analyzer with a 2048 or full 4096 channel memory. Two clock-timers permit each detector system to operate with pre-set analysis time.

The two Ge-Li detectors can also be used in coincidence arrangements.

b. Low Energy Photon Spectroscopy System

An addition to the radiation detection capability of the laboratory is the Low Energy Photon Spectroscopy System. The LEPS detector is a lithium-drifted germanium detector with excellent resolution characteristics for the X-ray region, and the solid state analyzer has a 4096-channel ADC and a 1024-channel memory which can be expanded in the future. The electronics of this analyzer is compatible with that of the gamma detection system, an advantage when dealing with complex "state-of-the-art" equipment. An IBM Selectric typewriter serves as the readout service.

c. Low-Level Laboratory

All of the equipment in the Low-Level Laboratory has been in continuous use during the past year. Measurements of the Pb^{203} and U^{237} radioactivities isolated from neutron-irradiated lunar samples (see Section 7) have been made by X-γ and β^--γ coincidences, respectively. The former system includes a 42 cm^3 Ge-Li detector for γ measurements. A second beta detection system for use in the Low-Level Laboratory is under construction.

d. Proportional Counting Systems

The radiation detection laboratories (counting rooms) housing 12 methane-flow proportional counters used for alpha and beta particle detection were rehabilitated. The new "silent" grounding system isolated from the methane gas line and line voltage regulation have resulted in steadier and more reliable operation of the counters.

e. Sharp-Beckman Low Beta Counting System

The Low Beta Counting System has been in almost continuous operation during the past year and has operated satisfactorily.

Effort of Principal Investigators Devoted to Project

Principal Investigator Nathan Sugarman estimates that he has devoted about 70% of his time to this project from April 1, 1970 to December 15, 1970. He estimates that he will devote about 50% of his time from December 15, 1970 to April 1, 1971.

Principal Investigator Anthony Turkevich estimates that he has devoted about 10% of his time to this project for the period April 1, 1970 to July 1, 1970 and 50% of his time for the period July 1, 1970 to December 15, 1970. He estimates that he will devote about 25% of his time from December 15, 1970 to April 1, 1971.