Presented at "Symposium on Spallation Nuclear Reactions
and Their Applications in Astrophysics and Radiotherapy,"
held at the University of Pennsylvania, May 2, 1975.

MASTER

RADIOACTIVE ISOTOPES ON THE MOON*

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

Raymond Davis Jr.

Department of Chemistry, Brookhaven National
Laboratory, Upton, New York

1. EXPECTATIONS

There has been an active interest in observing and accounting for radioactive products produced by cosmic rays in meteorites since the early suggestion by Bauer (1947) that the helium observed in meteorites was mostly produced by cosmic rays. The first searches for tritium in iron meteorites by Fireman (1958) led to the possibility of measuring the cosmic ray exposure ages of meteorites. By the time of the first lunar landing there were a number of laboratories engaged in measuring radioactive and stable products produced by cosmic rays in meteorites and earth bound satellites. These studies led to new knowledge of the period of time a meteorite fragment was exposed to cosmic rays. It was found that stone meteorites were exposed for periods of 2 to 30 million years, whereas some iron meteorites were exposed for many hundreds of millions of years. By studying selected radioisotope pairs it was found that the galactic cosmic ray intensity has been essentially constant for the last 500,000 years (^{39}Ar , ^{36}Cl), and perhaps there has been a lower cosmic ray intensity a few billion years ago (^{36}Cl - ^{40}K). Radioactive products were also applied to studying the gradient in the galactic cosmic ray intensity within the solar system as a result of modulation by the magnetic fields associated with the solar wind. Many studies were made of the distribution of cosmic ray spallation products in meteorites and these data were interpreted in terms of nuclear cascade processes. Information on these various

* This work supported by the U.S. Energy Research and Development Administration.

topics can be obtained from reviews and numerous original papers [1, 2].

We may ask what new information could be obtained by studying radioactive products in lunar samples? Having a man on the moon made it possible to collect particular samples chosen for the problem of interest. In the case of meteorites the original surface is lost and the location of the sample within the pre-atmospheric-impact body is unknown. On the moon's surface, samples can be collected for studying radioactivity distribution in the first few millimeters of the surface. From these surface samples one could learn about solar flare particle fluxes and derive information about the variations of these particle fluxes in the past. Having the surface material one could search for radioactive products collected from space, for example, supernova debris. By digging or boring holes in the lunar soil, samples could be obtained at depth, so that we could derive information about the nuclear cascade processes produced by cosmic rays in a dense medium. This information could be applied to an understanding of mixing processes in the lunar soil. Exposure age dating of rocks could be useful in determining the history of cratering processes on the moon. It was generally anticipated that a wide range of exposure ages would be obtained. There was hope that from a study of certain radioactive products that might be present in the lunar atmosphere, e.g., tritium and radon, one could learn whether these isotopes were desorbed or vented from the lunar soil or perhaps were injected into the lunar atmosphere by solar wind or solar flare particles. Direct counting of lunar rocks could also measure the primordial radioisotopes ^{40}K , ^{238}U , ^{230}U , and ^{232}Th . Using this technique a survey of these primordial radioactive isotopes can be made without sacrificing material. These are some of the problems that could be studied. There was the further vague hope that by entering this new area of research something new would turn up.

It is interesting to recount how the lunar program was operated, what sort of samples were obtained, and what new information resulted from studying radioactivities in lunar rocks and soil. This report will not be a proper review, but will give only the results of a few studies of special interest from which we can see the broad scope of the program.

2. LUNAR SAMPLES, DISTRIBUTION AND TECHNIQUES

Prior to the first lunar landing the National Aeronautics and Space Administration offered the world scientific community access to lunar samples and funds for studies within the United States. There was a broad response to the offer and appropriate

committees were formed to review the proposals and distribute lunar material. As a result of six missions to the moon a total of 381 kilograms of material was recovered. In the early missions detailed information on sample orientation was not generally available. As the program developed there was direct communication between the astronauts present on the moon, and a panel of scientists directing their choice of samples. A broad range of samples was returned consisting of individual rocks, rock chips, small rocks raked from the surface, surface soil, soil from trenches, and core and drill tubes driven into the lunar surface. The material was transported in sealed rock boxes and the larger rocks were carried in pouches in the spacecraft. There was a preliminary examination of selected material that gave useful data on chemical composition, mineralogical content, and rare gas content. The results of the preliminary examination for each mission were published in Science. A description of each rock and soil sample was given in a rock catalog for each mission. This basic information was available to the scientist, but in general was insufficient to select the best sample for a particular purpose. This was especially true for those studying radioactivities where rock orientation and clean surface samples are needed. Obtaining the most significant samples required a good working arrangement between the individual investigator and a member of the Lunar Sample Planning Team. This arrangement was workable but it was difficult for an investigator to discover what samples were available and obtain them in a short period of time. In addition to the administrative and documentary difficulties there was the overriding requirement of a biological quarantine placed on lunar materials. The biological tests required a period of six weeks before samples could be distributed. The biological quarantine was imposed during the first three lunar missions, Apollo 11, 12, and 14, and after that the restriction was lifted.

There were some observations that were time dependent and required making measurements during the quarantine period. A low-level counting laboratory was built 50 feet underground in the Lunar Receiving Laboratory to make radioactivity measurements. The samples were placed in stainless steel cans, sterilized and passed to the counting laboratory for direct counting of the gamma radiation. The ^{37}Ar (35 day half-life) activity measurements were also performed in the Lunar Receiving Laboratory during the quarantine period. Measurements of the ^{222}Rn (4 day) present in the gas in the rock boxes were made by extracting the gas prior to opening the rock boxes. The searches for magnetic monopoles, and remnant magnetic properties of rocks were also made during the quarantine period.

Radioactivities were measured by a variety of techniques.

First there was the non-destructive technique of observing the gamma radiation directly using a pair of large sodium iodide crystals. Three laboratories were engaged in these measurements, the Lunar Receiving Laboratory already mentioned, a similar prototype system at Oak Ridge National Laboratory, and a laboratory at Battelle-Northwest, Richland, Washington. Although there were other laboratories, these three made the majority of the direct counting measurements. Another approach was to dissolve the sample, make chemical separations of particular elements, and measure the beta and gamma radiation using a counting technique designed to have a high selectivity for a particular radioactive isotope. Another approach was to vacuum melt the sample to extract rare gases, hydrogen and carbon dioxide. These gases were separated using gas separation methods and then placed in proportional counters for measurement.

I would like to give the results of a few selected experiments. These will include some observations made on the first few millimeters of the surface where the effects of solar flare particles are important, some measurements on individual rocks, and some studies of radioactivities produced deep in the lunar soil by galactic cosmic rays.

3. SOME RESULTS

3.1 At the surface

A group of chemists [3] at the University of California, San Diego, under the leadership of J. R. Arnold measured the depth variation of radioactivities in the outer centimeter thick layer of rock 12002. A vertical slice and a top slab of the rock was dissected by carefully grinding off layers, see Fig. 1. Analyses were made by wet chemical methods to study a variety of isotopes, namely ^{10}Be , ^{22}Na , ^{26}Al , ^{36}Cl , ^{53}Mn , ^{54}Mn , ^{55}Fe , ^{56}Co , and ^{57}Co . Their results are given in Table 1. The isotope ^{56}Co (77 day half-life), was produced by (p,n) reaction (threshold 5.4 MeV) on the abundant isotope, ^{56}Fe (91.7%). Cobalt-56 showed the steepest profile, and is the best indicator of solar flare protons. The energy spectrum of solar flare protons is known reasonably well from satellite data, and the (p,n) cross sections on ^{56}Fe are well known. Furthermore, the flares of April 12 and November 3, 1969 contributed 94 percent of the ^{56}Co activity. The contribution from galactic cosmic rays was essentially negligible. These investigators compared their ^{56}Co depth profile from 0.1 to 1.9 g/cm² with the depth profile calculated from solar flare proton intensities and cross-section data. The comparison for this isotope was excellent. Similar calculations for the

Fig. 1. Schematic of rock 12002, showing location of samples (on left) and subdivision of samples (on right).

Table 1. Activities in rock 12002 (all activities in dpm/kg)

Isotope	$T_{1/2}$	OP-1 9.9 g (0-1 mm)	OP-2 9.5 g (1-2 mm)	OP-3 17.7 g (2-4 mm)	OP-4 25.3 g (4-9 mm)	OP-5 19.2 g (9-20 mm)	OP-6 50.4 g (20-60 mm)	OP-7 4.4 g (60 mm)
Be^{10}	2.5×10^6 y					15.4 ± 3.5	11.5 ± 1.4	
Na^{22}	2.6 y	146 ± 18	122 ± 14	91 ± 11	71 ± 8	54 ± 7	39 ± 6	
Al^{26}	7.4×10^5 y	209 ± 26	154 ± 21	131 ± 16	91 ± 12	72 ± 11	64 ± 6	64 ± 11
Cl^{36}	3.1×10^5 y	13.4 ± 3.8		9.7 ± 2.5	14.7 ± 2.4	8.4 ± 2.1	9.4 ± 1.2	
Mn^{54}	3.7×10^6 y	93 ± 6	86 ± 5	73 ± 5	67 ± 4	52 ± 3	47 ± 3	31 ± 3
Mn^{56}	303 d	99 ± 17	77 ± 12	53 ± 11	50 ± 9	39 ± 10	31 ± 5	
Fe^{55}	2.6 y	731 ± 68	454 ± 46	285 ± 30	171 ± 23	101 ± 17	56 ± 12	
Co^{60}	77 d	533 ± 70	203 ± 30	80 ± 15	32 ± 9			
Co^{60}	370 d	16 ± 6	< 6					

longer lived isotopes, ^{54}Mn (303 day), ^{22}Na (2.6 year), and ^{55}Fe (2.6 year) were not nearly so satisfactory, the measured activities were about a factor of two higher than the activity derived from proton fluxes from satellites and cross-section data. The lack of agreement could be attributed to errors in the solar flare particle fluxes, since a large fraction of the total flux was due to large flares from the previous solar cycle and accurate satellite measurements were not yet available at that time.

Of particular interest are the profiles for the longer lived ^{26}Al (7.4×10^5 year) and ^{53}Mn (3.7×10^6 year). These isotopes can be used to test whether or not the solar flare intensity has varied over the last million years. The usual stellar structure

arguments favor no change in the solar luminosity over the last 10⁸ years, although it is entirely possible that major climatic changes can be attributed to variations in the solar luminosity [4]. It would be interesting to know whether there was a change in the solar flare intensity, and if so, the depth variation of ²⁶Al and ⁵³Mn may be the only way of testing for this variation. Unfortunately, the answer is not clear. The measurements on these isotopes agree reasonably well with flare particle fluxes that are similar in energy distribution and spectral shape to those of the last two solar cycles. However, there may be errors in the cross-sections (mostly unpublished). Additionally, there is the question of the long period orientation of rock 12002, and the degree of space erosion is a serious problem. I feel the conclusion of essentially constant solar proton intensity-energy reached is perhaps only true within a factor of two or so. This is not of adequate accuracy to determine whether or not there is a solar derived cause for glaciation. This important question warrants further study, and it would be advisable to use the surface of a very large boulder less likely to have been disturbed.

Another study that could give information about past solar intensities is the study of the ¹⁴C profile. Begeman et al. [5] measured ¹⁴C at three depths (0-0.5; 0.5-2; 2-6.5 cm) in rock 12053 and found the exterior layer was 2.3 times higher than the two inner layers. This high surface value cannot be explained by (p,3p) reaction on oxygen with the same average solar proton intensity and energy spectrum that fits the ²²Na, ⁵⁵Fe, and ⁵⁴Mn data, that is, a 4 π integral flux above 10 MeV of 100 protons cm⁻² sec⁻¹ and an exponential rigidity of 100 MV. To explain the high ¹⁴C value the authors say a flux 3 to 5 times higher, averaged over the past 10,000 years, is needed. There is also the mundane possibility that the cross-sections used are incorrect. A further possibility is that the ¹⁴C was produced directly in the photosphere of the sun and implanted in the surface. A ¹⁴C depth profile was measured by Boeckl [5] on rock 12002, the same rock studied by the Arnold group (Fig. 1). His measurements were made with a finer depth structure (4 points 0-9 mm) than those of Begeman et al. Boeckl obtained a steep profile that was consistent with a flare proton rigidity that fitted the ²²Na, ⁵⁵Fe, and ⁵⁴Mn data, but required a flux a factor of two higher. It is clear that further studies of the ¹⁴C activity in the surface of lunar rocks is needed to resolve these important questions.

It is known from the historical record that sunspots were essentially absent over a 70 year period from about 1645 to 1715. It would be interesting to observe this depression in the solar activity with radionuclides in lunar surface material. However, there are no radionuclides with a half-life of a few hundred

years that are suitable for this purpose. Argon-39 (269 y) has the required half-life but is produced primarily by neutron reactions on potassium and calcium. Silicon-32 (~280 y) is another isotope with the correct half-life, but it is not produced in good yield by solar protons on lunar materials. So it is unfortunate that this solar quiet period cannot be verified quantitatively by radioisotopes, or conversely, that the radioisotope technique be tested!

The surface of the moon is exposed and collects infalling material for billions of years. The temporal record is disturbed in the lunar soil by mixing processes mainly as a result of meteorite impacts. Mixing by impacts and land slides will be serious in some locations but at other locations the soil is relatively undisturbed for long periods of time. There is quantitative information on these processes based upon measurements of the changes in the gadolinium and samarium isotope abundances induced by neutron capture. Studies on an Apollo 15 core allowed Russ et al. [6] to conclude that the upper 10 cm of soil is turned over every 0.1 b.y. and the turnover time at a few meters depth is more than 0.5 b.y.

It is of great interest to search in this material for debris from supernova explosions and for radioisotopes accumulated from the sun. Fields and his associates [7] at Argonne National Laboratory have made a search for ^{244}Pu ($t_{1/2} = 82$ m.y.) in 12 lunar samples. The lowest limit set was 10^{-17} g ^{244}Pu /g on a few gram sample of an Apollo 15 soil. It is difficult to estimate the expected ^{244}Pu collection rate from supernovas, or cosmic rays. If we take the present upper limit to the cosmic ray intensity for particles of charge 92-96 of 10^{-11} cm $^{-2}$ sec $^{-1}$, assume 10 per cent of the incoming ^{244}Pu atoms survive stopping in 1 g/cm 2 , and collect for a period of 10^8 years, then there may be 10^{-18} g Pu/g of lunar soil. However, if there were low energy ^{244}Pu atoms arriving as supernova debris a higher concentration could be present in lunar soil. It seems clear that further searches for ^{244}Pu should be made with carefully selected samples. There has been a great reluctance to using lunar samples for exploratory work of this nature in spite of the high interest in this problem.

The group at Argonne [7] also made measurements of the $^{236}\text{U}/^{238}\text{U}$ ratio (^{236}U $t_{1/2} = 23.4$ m.y.) and the ^{237}Np ($t_{1/2} = 2.14$ m.y.) concentration in a number of lunar soils and rock samples. One expects a $^{236}\text{U}/^{238}\text{U}$ ratio $\sim 5 \times 10^{-9}$ from slow neutron capture on ^{235}U (total U 0.1 to 2 ppm), and indeed five samples are in this range. However, one rock (12073) had a ratio of $40-50 \times 10^{-9}$ and two samples of soil (12070) had ratios of $30-50 \times 10^{-9}$ (coarse fraction) and 235×10^{-9} (total sample). These very high ratios cannot be explained by nuclear reactions on ^{238}U such as the (n,3n) or (p,t) reactions. They require a

higher production rate sometime in the past, or some other source for the ^{236}U . If it were from an increased solar or galactic cosmic ray intensity over long periods of time, all the exposure ages derived from present cosmic ray fluxes would be shortened considerably. However, if there were short periods of very high particle fluxes exposure ages would not be affected. The fact that ^{53}Mn activities are within a factor of two of the concentrations expected from present day solar and galactic cosmic ray fluxes requires that fluxes as high as indicated by ^{236}U concentrations did not occur during the last 10^7 years. This interesting result needs a clear explanation and it would be very useful to have additional measurements on both rock and soil samples.

3.2 At depth

During the last three missions to the moon the astronauts recovered long columns of the lunar soil with a special coring drill [8]. The length of the three cores were 2.4, 2.4, and 2.9 meters. Obtaining these long cores was a major achievement in sampling the lunar surface. These and the shorter drive tubes are our only samples that give a view of regolith stratigraphy. Studies on these cores using a variety of techniques, gadolinium and samarium isotope mass spectrometry, rare gas isotope analyses, fossil track distributions, chemical analysis, and mineralogical studies have lead to new knowledge of the history of the lunar regolith. I would like to confine my discussion to an in situ measurement of the neutron flux distribution and some radio-isotope measurements that were made on these cores.

A basic factor in understanding the gadolinium and samarium isotope distribution changes as a result of neutron capture is to know the present-day neutron fluxes produced in the lunar soil by cosmic ray interactions. The flux distribution was calculated theoretically by Lingenfelter, Canfield, and Hampel and by Armstrong and Alsmiller [9], and it was of great interest to measure the flux experimentally. The measurement was made by a special probe designed by Don Burnett (CIT) that was introduced into the hole in the lunar regolith after the Apollo 17 coring drill was removed. After insertion a mica fission track detector was rotated over a ^{235}U foil, and a plastic track detector was rotated over a ^{10}B foil to register slow neutron captures. The probe was in place registering the flux for 49 hours. The results [10] for both target elements showed a sharp rise from the surface to a broad peak at a depth of 100-160 g/cm² and slow decrease with increasing depth. The shape of the curve fitted the theoretical curve very well, see Fig. 2. The total neutron capture rate in ^{235}U was 11 ± 17 percent lower, and in ^{10}B it was 19 ± 13 lower than the theory of Lingenfelter et al. predicted.

Fig. 2. Neutron depth profile obtained by Woolum and Burnett [10] compared to the theory of Lingenfelter et al. [9].

The results for ^{10}B normalized to the theory are shown in Fig. 2.

By combining the neutron flux distribution in the lunar soil with measurements of the total neutron fluences (flux $\times$ time) measured by ^{157}Gd captures and the regolith depth (4-5 m) inferred from observations, it has been concluded that the total fluence is a factor of 2-3 times lower than expected for a uniformly mixed soil exposed for 3.8 billion years. Workers in this field have made a great issue of this problem [10, 11]. The various suggested solutions include (1) the regolith is not well mixed, (2) the regolith is not as deep as is thought, (3) the cosmic ray intensity averaged over the last 3-4 billion years is lower than present intensities, (4) large quantities of lunar soil are ejected by meteoroid impacts [12] (40-100 cm/ 10^9 y), or (5) some combination of these or other discrepancies give an apparent reduction in neutron fluences. These observations give an interesting insight into the development of the lunar regolith, a topic of broad significance in the evolution of the solar system.

It was of interest to have a measure of the fast neutron flux generated in the soil by cosmic rays. The 35-day isotope ^{37}Ar is

Fig. 3. ^{37}Ar depth profile measured in the Apollo 16 and 17 drill cores [14, 15].

an ideal one for this purpose. It is produced by the reaction $^{40}\text{Ca}(n,\alpha)^{37}\text{Ar}$ on the abundant element calcium (7-12 percent). Argon-37 is a radioisotope that is easily extracted and counted making it possible to perform measurements on modest amounts of sample (1-2 g). The production cross section has been measured. The excitation function shows an effective threshold at about 3 MeV, rising to broad maximum at 6 MeV (205 mb) and then it drops slowly with increasing energy (138 mb at 14 MeV) [13]. The ^{37}Ar activity as a function of the depth was measured in the Apollo 16 core as a joint project between Smithsonian Astrophysical Observatory [14] and Brookhaven National Laboratory [15]. Only the deeper portions of the Apollo 17 core were analyzed because the upper portions of this core were not opened in time for the measurements. All samples were analyzed for calcium so that the measurements for the two missions could be compared. A plot of dpm $^{37}\text{Ar}/\text{g Ca}$ is given in Fig. 3. The ^{37}Ar production exhibits a steep rise with depth, corresponding to the development of the nuclear cascade, reaching a maximum at a depth of about 50 g/cm², and then it dropped exponentially with an attenuation length of 240 ± 20 g/cm². This general behavior is expected from the nuclear cascade process in the lunar soil [16]. There were no solar flares immediately before the Apollo 16 mission, so these measurements give a valid ^{37}Ar production rate from galactic

cosmic rays. Kornblum and his associates [17] have made calculations of the ^{37}Ar depth profile with a defined neutron source function, the neutron moderating factors derived from the lunar composition (ANISN neutron transport code), and the measured $^{40}\text{Ca}(n,\alpha)^{37}\text{Ar}$ excitation function. Their calculated profile fits the measurements, and they obtain a total neutron production rate in lunar soil of $26 \pm 4 \text{ cm}^{-2} \text{ sec}^{-1}$ by normalizing the calculations to the experimental measurements. This value is higher than that of Lingenfelter et al. and Armstrong and Alsmiller, 16 ± 5 and 17.5, respectively.

The Apollo 17 mission followed only 124 days after the intense solar flare of Aug. 4-9, 1972. This flare was the most intense flare ever observed, similar in magnitude to the great flare of 1956. This flare produced approximately 2×10^{10} protons/cm² with energy above 60 MeV. The effects of this flare were clearly observed in many samples returned from the Apollo 17 mission. The ^{37}Ar activities were found to be very high in the samples from the surface (depth 0-5 g/cm²), but samples at greater depths (>50 g/cm²) were not affected. The flare protons interact at the surface to produce ^{37}Ar by the reaction $^{40}\text{Ca}(p,\alpha)^{37}\text{K}(g^+ \text{ decay})^{37}\text{Ar}$. During the Aug. 1972 flare the production rate at the surface was over 400 times the normal cosmic ray production rate.

The long lived argon isotope, ^{39}Ar ($t_{1/2} = 265 \text{ y}$), was also measured on the Apollo 16 and 17 cores [14, 15]. This isotope is produced by several processes, $^{39}\text{K}(n,p)^{39}\text{Ar}$, $^{40}\text{Ca}(n,2p)^{39}\text{Ar}$, and by spallation of iron. The excitation functions for the neutron reactions have not been measured so the relative cosmic ray production from these various processes cannot be sorted out. The depth distribution curve exhibits the characteristic increase from surface values (factor of 1.6) reaching a maximum at a depth of 50 g/cm² and then dropping exponentially with an attenuation length of 166 g/cm². The general behavior is in agreement with expectation.

It is of great interest to observe the depth profile of a long lived radioactive isotope to search for long period variations in solar activity or galactic cosmic ray intensities. As mentioned earlier ^{53}Mn is an ideal isotope for this purpose. It has a long half-life, 3.7×10^6 years, and it can be measured by a neutron activation technique that requires only a 0.2 g sample. This isotope is made at the surface by low energy protons mainly by the $^{56}\text{Fe}(p,\alpha)^{53}\text{Mn}$ and $^{56}\text{Fe}(n,p3n)^{53}\text{Mn}$. Imamura et al. [18] measured the ^{53}Mn depth profiles in the Apollo 15 and 16 cores. The solar proton produced ^{53}Mn is very high at the surface, dropping rapidly with depth to a depth of 2 g/cm², see Fig. 4. Below a depth of 10 g/cm² it decreases exponentially with an attenuation length of $220 \pm 50 \text{ g/cm}^2$. The results agree well

Fig. 4. ^{53}Mn depth profile of Imamura et al. obtained on the Apollo 15 drill core, soil and rock samples [3, 18].

with the model calculations of Reedy and Arnold [16]. It was concluded that the soil column has remained undisturbed for the last 5 million years, consistent with the Gd isotope conclusions mentioned earlier that extend over a much longer period of time.

It would be interesting to compare the depth profile of various radioisotopes with actual measurements with accelerated protons and alpha particles. In the past there has been a number of these thick target bombardments, the first being B. S. P. Shen's bombardment of an iron target 30 x 30 x 90 cm with 3 GeV protons. The results of these early bombardments are summarized in a review by Kohman and Bender [2]. Two studies have been made recently with thick targets of simulated lunar soil. Bar and Herr [19] studied thermoluminescence of minerals as a function of the depth in a target 30 x 30 x 140 cm bombarded with 600 MeV protons. Lyman [20] measured ^{37}Ar , ^{39}Ar , ^{42}Ar , and tritium activity distribution in a simulated lunar target 20 x 20 x 43 cm bombarded by 600 MeV protons. Experiments of this type require counting numerous samples and it is further necessary to perform the measurements at a series of energies. In view of the enormous effort involved, most experimenters are discouraged. Also the basic nuclear cascade process is understood, and it does not seem worth the effort at the present time to establish the detailed

parameters involved.

3.3 Individual rocks

The largest amount of data on radioactivities in lunar materials was obtained by a few laboratories that measured gamma radiation by NaI scintillation counters. Large pure activated NaI(Tl) crystal scintillation detectors were developed industrially and a number of laboratories devised sophisticated low-level counting arrangements for measuring lunar and environmental samples. This approach has the decided advantage of being able to measure lunar samples without affecting them in any way. Once the elaborate counting facility was built a large number of samples can be surveyed. In fact the accumulation of data was limited by the ability of the curator's office to process the sample, especially during the quarantine period. One extremely useful observation that was made immediately was that lunar rocks are relatively low in the volatile element potassium. If a plot of the ratio K/U versus K concentration is made, it can be shown that igneous earth rocks, lunar rocks, and various meteorite classes, eucrites, carbonaceous chondrites and ordinary chondrites fall into distinct groups [21]. This useful plot not only sorts out solar system materials but also allows a classification of lunar basalts, soils, highland rocks, and breccias. These measurements of primordial radioactive elements also demonstrated that the Th/U ratio for lunar rocks is 3.8, almost identical to the ratio generally observed for earth rocks [21, 22].

The usual cosmogenic radioactivities measured were: ^{22}Na (2.6 y), ^{26}Al (3.7×10^6 y), ^{46}Sc (84 d), ^{48}V (16 d), ^{51}Cr (28 d), ^{54}Mn (303 d), ^{57}Co (270 d), ^{58}Co (71 d) and ^{60}Co (5.3 y). The activity levels are dependent upon the chemical composition of rock, and where relevant cross-sections and chemical analyses are known, the agreement between measured and calculated activities is satisfactory. There is interest in deriving information about cosmic ray intensities in the past and about lunar surface events from the pair of isotopes ^{22}Na and ^{27}Al . These isotopes are produced by various reactions on the elements Si, Al, Mg, and Na. Yokoyama et al. [23] and Keith and Clark [24] have attempted to normalize the observed activities by adopting a production rate for each element. Their techniques are useful for selecting rocks with exposure ages comparable to the half-life of ^{26}Al , the so-called undersaturated rocks.

The solar flare of Aug. 4-9, 1972 produced dramatic increases in activity levels of all measured isotopes with half-lives shorter than a few years. By careful measurements of various isotopes with well measured excitation functions it was possible to determine the average rigidity parameters and fluxes that

characterize the energy spectrum of the solar flare protons from this event [21, 25]. Also there is evidence from the measurements on rock surfaces at various angles of inclination that the flare particles were anisotropic [25].

I might conclude by some remarks about cosmic ray exposure ages. Since lunar rocks are exposed on the surface, one can apply mass-spectroscopic measurements of the abundant rare gas spallation products like ^3He or ^{38}Ar to derive exposure ages, assuming a constant production rate for all samples. Helium-3 exposure ages are questionable because of the loss of both tritium and helium. A much more reliable method is to determine the temperature release pattern of the argon isotopes from neutron activated samples following the technique now common in ^{40}Ar - ^{39}Ar dating. In this method the release of spallation ^{38}Ar from calcium can be related to the ^{37}Ar produced by (n,α) reaction on ^{40}Ca . By adopting an assumed ^{38}Ar cosmic ray spallation production rate on calcium ($1.4 \times 10^{-8} \text{ cm}^3 \text{ STP/g Ca} \cdot 10^6 \text{ y}$), the exposure age can be obtained. The usual assumption of constant spallation production for all samples is of course not correct, as there is a change in production rate with depth. Another method of determining exposure ages depends on mass spectroscopic measurements of ^{81}Kr ($t_{1/2} = 2.1 \times 10^5 \text{ y}$), a spallation product of Sr, Y, and Zr. The method depends upon the rock being exposed for a period long compared to the half-life of ^{81}Kr permitting the concentration of ^{81}Kr to be used for the production rate in the sample. The concentration of ^{81}Kr relative to the stable isotope ^{83}Kr is used to determine the exposure age. It is assumed that the production cross section of ^{81}Kr is equal to the average of the ^{80}Kr and ^{82}Kr production cross-sections. One would expect the relative production cross-sections of ^{81}Kr and ^{83}Kr to depend upon target element, and thereby depend upon the Sr, Y, and Zr composition of the rock. However, in spite of this obvious difficulty, this method is held in high esteem.

Exposure ages of lunar rocks vary considerably, as one might expect from random impacts on the lunar surface so precise exposure age measurements are not particularly required. There are numerous exposure age measurements of variable quality throughout the lunar literature. The range of exposure ages are from 600 million years to rocks that have not been exposed long enough for ^{26}Al activity to reach saturation, about 1 million years.

4. CONCLUSIONS

It is interesting to look back over the accomplishments of the last five years of studying radioactivities in lunar samples, and to inquire whether we have learned anything new. There is no

question that an enormous amount of new information has resulted from studying lunar samples and we can address our inquiry at three levels: (1) were the new observations consistent with or do they support earlier views, or (2) were there some surprises that now need confirmation, or (3) did we discover something totally new. I believe the studies of radioactivities in lunar material are essentially characterized as being consistent with earlier knowledge, but there are a few interesting results that need further work.

First, concerning the production of radioactivities by galactic cosmic rays. The measurements in individual lunar rocks and at depth in the lunar soil are consistent with what was already known. The measurements of the depth profile of radioactive products with half-lives in the order of months to millions of years agree reasonably well with theoretical calculations of the nuclear cascade processes and present galactic cosmic ray intensities and energy spectra. The lunar results are consistent with a constant cosmic ray flux. However, the best test of the constancy of the galactic cosmic ray flux are the older measurements of ^{39}Ar , ^{36}Cl , and ^{40}K in iron meteorites. It seems unlikely that further measurements on lunar material will lead to any new knowledge on this question. However, one can see that there will be further interest in applying our knowledge of cosmic ray interactions in lunar material to understanding mixing processes in the lunar soil, and possibly measuring accurate exposure ages of lunar rocks.

Studies of the activities on the surface of lunar rocks and surface soil samples have produced some interesting results. The question of the solar flare intensity in the last 10^6 or 10^8 years is an important question that needs further study. New measurements of the depth profile are needed of ^{22}Na , ^{14}C and ^{53}Mn on the surface of large rocks likely to be fixed in their position for periods longer than 10^7 years. Additional studies of ^{236}U and ^{237}Np are certainly needed. It is very important to understand how some samples can have 10 to 100 times higher concentrations of ^{236}U than others. Perhaps ^{236}U is the only isotope with a half-life long enough to retain the effects of events that occurred 10^8 years ago.

Finally a thought on the use of lunar samples for exploratory work. Experiments like the ones searching for ^{244}Pu , and super heavy elements have not been especially encouraged. It may be a few decades before new samples are obtained from the moon, but when lunar exploration is revived our present closely guarded collection will be only of secondary interest. I feel the present collection should be more available to the scientific community and more emphasis should be placed on significant exploratory studies.

REFERENCES

The abbreviation PLSC-2 stands for Proc. 2d Lunar Sci. Conf.,
Geochim. Cosmochim. Acta, Suppl. 2.

1. O. A. Schaeffer, Ann. Rev. Phys. Chem. 13, 151 (1962); M. Honda and J. R. Arnold, Handbuch der Physik XLVI/2, p. 613 (1967), Springer-Verlag, Berlin.
2. T. P. Kohman and M. L. Bender, High-Energy Nuclear Reactions in Astrophysics, ed. by B. S. P. Shen, Benjamin Press, 1967.
3. R. C. Finkel, J. R. Arnold, M. Imamura, R. C. Reedy, J. S. Fruchter, H. H. Loosli, J. C. Evans, and A. C. Delany, PLSC-2, Vol. 2, p. 1773 (1971).
4. A. G. W. Cameron, Rev. Geophys. and Space Physics 11, 505 (1973).
5. F. Begeman, W. Born, H. Palme, E. Vilcsek, and H. Wanke, PLSC-3, Vol. 2, p. 1693 (1972); R. S. Boeckl, Earth and Planetary Sci. Letters 16, 269 (1972).
6. G. P. Russ III, D. S. Burnett, and G. J. Wasserburg, Earth and Planetary Sci. Letters 15, 172 (1972).
7. P. R. Fields, H. Diamond, D. N. Metta, and D. J. Rokop, PLSC-4, Vol. 2, p. 2123 (1973), contains earlier references.
8. D. S. Woolum, D. S. Burnett, and C. A. Bauman, Apollo 17 Preliminary Science Report NASA SP-330. These reports are a very useful source of general information on each mission.
9. R. E. Lingenfelter, E. H. Canfield, and V. E. Hampel, Earth and Planetary Sci. Letters 16, 355 (1972); T. W. Armstrong and R. G. Alsmiller, PLSC-2, Vol. 2, p. 1729 (1971).
10. D. S. Woolum and D. S. Burnett, Earth and Planetary Sci. Letters 21, 153 (1974); D. S. Burnett and D. S. Woolum, PLSC-5, Vol. 2, p. 2061 (1974).
11. O. Eugster, F. Tera, D. S. Burnett, and G. J. Wasserburg, Earth and Planetary Sci. Letters 8, 20 (1970); G. Price Russ III, ibid, 19, 275 (1973) and references therein.
12. E. L. Fireman, PLSC-5, Vol. 2, p. 2075 (1974).
13. J. W. Barnes, B. P. Bayhurst, B. H. Erkkila, J. S. Gilmore, N. Jaimie, and R. J. Prestwood, J. Inorg. Nucl. Chem. 37, 399 (1975).
14. E. L. Fireman, J. D'Amico, and J. De Felice, PLSC-4, Vol. 2, p. 2131 (1973).
15. R. W. Stoenner, R. Davis Jr., E. Norton, and M. Bauer, PLSC-5, Vol. 2, p. 2211 (1974).
16. R. C. Reedy and J. R. Arnold, J. Geophys. Res. 77, 537 (1972).
17. J. J. Kornblum, E. L. Fireman, M. Levine, and A. A. Aronson, PLSC-4, Vol. 2, p. 2171 (1973); J. J. Kornblum, Thesis, State University of New York at Stony Brook, 1975.
18. M. Imamura, R. C. Finkel, and M. Wahlen, Earth and Planetary Sci. Letters 20, 107 (1973); M. Imamura, K. Nishiizumi, M. Honda, R. C. Finkel, J. R. Arnold, and C. P. Kohl, PLSC-5, Vol. 2, p. 2093 (1974).

19. K. Bar and W. Herr, Earth and Planetary Sci. Letters 22, 188 (1974).
20. W. J. Lyman, Preliminary Report, Chemistry Department, Brookhaven National Laboratory, Upton, N.Y. (1971).
21. J. S. Eldridge, G. D. O'Kelley, and K. J. Northcutt, PLSC-5, Vol. 2, 1025 (1974) and earlier references therein.
22. J. E. Keith, R. S. Clark, and L. J. Bennett, PLSC-5, Vol. 2, 2121 (1974) and earlier references therein.
23. Y. Yokoyama, J. L. Reyss, and F. Guichard, PLSC-5, Vol. 2, p. 2231 (1974).
24. J. E. Keith and R. S. Clark, PLSC-5, Vol. 2, p. 2105 (1974).
25. L. A. Rancitelli, R. W. Perkins, W. D. Felix, and N. A. Wogman, PLSC-5, Vol. 2, p. 2185 (1974).