BNL 18580

Radioactive Rare Gases and Tritium in the Sample Return Container, and the ^{37}Ar and ^{39}Ar Depth Profile in the Apollo 16 Drill Stem

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The gas was extracted from the sample return container from the Apollo 16 and 17 missions by adsorption on charcoal and activated vanadium metal. The hydrogen, argon, and radon were separated and counted to give the tritium, ^{37}Ar, ^{39}Ar, and ^{222}Rn activities. The tritium and argon activities observed could be explained by diffusive losses of these gases from the fine material in the container. There was no excess tritium present in the Apollo 17 containers that could be attributed to solar tritons remaining from the intense flare of August 4, 1972. The ^{222}Rn observed in the sample return container was interpreted as an emanation product from lunar fines and an emanation yield of 1×10^{-4} was calculated. This yield is consistent with the low radon content observed in the lunar atmosphere.

The tritium, ^{37}Ar, ^{39}Ar, and ^{222}Rn activities and the K, Ca, Ti, Fe, and Mn contents were measured on a set of samples from the Apollo 16 deep drill stem at depths from 83 to 343 g/cm^2. The ^{37}Ar and ^{39}Ar activities combined with similar measurements at more shallow depth by Fireman and associates (SAO) give the complete activity profile in the lunar regolith. Since ^{37}Ar is produced mainly by the ^{40}Ca(n,α)^{37}Ar reaction it is possible to determine the neutron production rate in the regolith as a function of the depth. The ^{222}Rn extracted from the samples by vacuum melting was found to be lower than expected in some samples based upon their uranium contents. The hydrogen and helium contents of the drill stem samples were measured and found to be relatively uniform with depth in contrast to similar measurements on Apollo 15 and 17 drill stems. The H/He atom ratio was higher than the accepted solar wind value by a factor of two, *possibly due to water contamination.*

Introduction

This report will present two separate series of measurements on lunar material from the Apollo 16 mission. The gas in the sample return container (SRC) has been collected in all Apollo missions with the exception of Apollo 11 and analyzed for the radioactive rare gases, ^{37}Ar, ^{39}Ar, and ^{222}Rn, and tritium. We will give the new results for the Apollo 16 and 17 missions and a discussion summarizing all of the results and their relation to the lunar atmosphere.

The second series of measurements to be reported are the ^{37}Ar, ^{39}Ar, tritium, and ^{222}Rn activities in the Apollo 16 drill stem. These gaseous radioactivities were extracted by a vacuum melting procedure from a set of samples extending from 57 cm to 220 cm from the surface. The upper section of this drill stem, from the surface to a depth of 22 cm was measured by Dr. Fireman's group of the Smithsonian Astrophysical Observatory (SAO). In addition to the radioactivity measurements we also determined the hydrogen, helium, K, Ca, Ti, Fe, and Mn concentrations in these samples. These results will be given and the fast neutron production rate and flux will be deduced. A similar set of measurements are in progress in the Apollo 17 drill stem, but these will be reported at a later time. The ^{37}Ar activity may be used to determine the neutron production rate by galactic cosmic rays as a function of depth in lunar soil. A brief analysis of our data will be given here, and a more detailed analysis is given by Kornblum et al. (1973) in these proceedings.

I. Radioactive Rare Gases from the Sample Return Container

The sample return container (SRC) was closed on the surface of the moon by the astronauts and remains sealed until it is opened at the Lunar Receiving Laboratory (LRL). The container is provided with an O-ring and an indium metal seal and therefore should remain vacuum tight during transit. The fine lunar material in the container is essentially in its pristine condition as it existed on the lunar surface. Our interest was in observing the emanation of ^{222}Rn (3.8 day) from ^{226}Ra decay, and the diffusive loss of the radioactive isotopes, ^{37}Ar (35 day), ^{39}Ar (269 year), and tritium (12.3 year) from lunar fines. Our results on the Apollo 12, 14, and 15 missions were reported (Stoenner, et al., 1972), and we will give the results from one box from the Apollo 16 mission (SRC no. 2) and both SRC from the Apollo 17 mission.

Experimental

The gas was removed by adsorption on charcoal at liquid-nitrogen temperatures (77°K) using a stainless steel system located directly under the LRL cabinets used for opening and unloading the sample containers. This system was pumped initially with a cryopump, and finally a high vacuum (10^{-6} Torr) was achieved with a vac-ion pump. The system consisted of bakeable high-vacuum valves (2.5 cm) leading to a charcoal-filled finger (2.5 cm dia.) containing 10 grams of petroleum-base charcoal (low in radium), and a second finger containing 10 grams of activated vanadium metal. A thermocouple gauge was used to measure the pressure in the system and to follow the pressure in the SRC as the gas was removed. The probe was attached through an o-ring seal to the outside surface of the SRC at a point where the wall was thinned to 0.5 mm. Prior to extracting the gas, the system was evacuated and baked to 400°C. The SRC was penetrated with a sharp bevelled rod 6 mm in diameter and the gas was collected over a 60-90 minute period. During this time the pressure in the SRC drops to about 5 percent of its initial value. The charcoal adsorption technique effectively removes the rare gases (Ar to Rn), other gaseous compounds, and partially adsorbs hydrogen. The activated vanadium metal is an effective adsorber for hydrogen gas at room temperature and was used in an effort to determine the chemical form of tritium. Following the extraction, the charcoal and vanadium fingers were closed off with valves and brought to Brookhaven for analysis.

The charcoal finger was connected to a glass vacuum system containing a 700 cc Toepler pump that removes the gas from the finger, and places it in a tube containing in series a small charcoal trap (0.1 g C) and a quartz tube containing activated vanadium metal. The system was that used for vacuum melting and extracting gases from lunar samples and will be described later. The collected gas volume was measured, an aliquot was taken for mass spectrometer analysis, and then the gas was heated to 1000°C over vanadium metal to getter all reactive species. The vanadium metal was cooled to room temperature to retain hydrogen, and the rare gases were removed in sequence by adjusting the temperature of the charcoal. With the charcoal at 77°K helium was removed. The volume of helium in all cases was too small to measure accurately, and was less than 10^{-3} cm^3 STP. The charcoal temperature was raised to -76°C. Argon carrier with 10 percent CH_4 was introduced and removed into a small proportional counter. Finally, the charcoal temperature was raised to 250°C. Argon carrier with 10 percent CH_4 was introduced once again and removed together with radon into a second proportional counter. After the rare gases were removed the vanadium was heated to 1000°C to release hydrogen. The hydrogen was passed through a palladium thimble, collected with a Toepler pump, its volume measured, and placed in a small proportional counter for tritium measurements.

The stainless steel finger containing the vanadium absorber was treated separately. The vanadium metal was in a quartz tube with a taper joint so that it can be quickly removed from the finger and plugged into the processing system. It was evacuated, and then heated to 1000°C to release hydrogen. The hydrogen was passed through a palladium thimble, collected with a Toepler pump, its volume measured, and finally introduced together with argon-CH_4 into a small proportional counter for tritium measurement.

Results and Discussion

One Apollo 16 box and both Apollo 17 sample return containers were analyzed. The first SRC from the Apollo 16 mission was not sealed properly on the lunar surface and consequently had a pressure of one atmosphere and thus could not be analyzed. The volumes collected by the charcoal adsorber plus the vanadium metal are given in Table 1. The volume of gas collected can be used to estimate the pressure in the SRC prior to puncturing. The total volume of the box is 23 liters, the rocks occupy approximately 5 liters of this space leaving a void volume of 18 liters. The corresponding pressure in the box is given in the last column of Table 1, and it can be seen that the calculated pressure agrees reasonably well with the thermocouple gauge reading. An aliquot of the gas recovered from the charcoal finger was analyzed with a mass spectrometer and found to contain predominantly N_2 and a small amount of water. Thus most of the pressure in the sample return container is air, and probably resulted from a small leak in its cover seal.

The volumes of hydrogen ultimately recovered from the charcoal and vanadium adsorbers are listed in Table 1. The volume of hydrogen obtained from the charcoal absorber would include water, hydrogen, and any other hydrogen containing compound that existed as gas in the SRC. It can be seen that the volumes were quite low for containers 16-2 and 17-2, comparable to the system blank, but much higher for 17-1. The vanadium absorber will not react with water or hydrocarbons at room temperature but does adsorb hydrogen efficiently. The volumes of hydrogen from the vanadium absorber are a fair measure of the actual volume of hydrogen in the SRC gas. However, there is a system blank in the range of 0.05 to 0.10 cm^3 H_2 at S.T.P., which is small compared to the volume of hydrogen from 17-1, but comparable to the volume of hydrogen from 17-2.

It will be shown later that lunar fines contain about 1.0-1.4 cm^3 of H_2 at S.T.P. per gram. It is interesting to estimate the fraction of hydrogen present in fine material that diffuses out into the gas phase during the 8 to 21 days the material was present in the SRC. The weights of fine material in these three SRC's are given in Table 2. From these weights one estimates that the fraction of hydrogen released was 8×10^{-5}, 7×10^{-4}, and 2×10^{-5}, respectively from the individual containers 16-2, 17-1, and 17-2. These values will be compared to the corresponding ones for tritium release.

The radioactive ^{37}Ar, ^{39}Ar, ^{222}Rn, and tritium observed in the Apollo 16 and 17 SRC are listed in Table 2. We have also included in the table the reported values obtained from earlier missions. The argon activities and tritium observed in the SRC gas can be attributed to the diffusion of these radioactive products out of the lunar fines (Stoenner, et al. 1971 and 1972).

The tritium activities in the Apollo 16 and 17 SRC are very low; the uncertainties listed are counting statistics (1 σ). The lowest activity observed (17-1), is approximately twice counter background. It may be noticed that the tritium activity was approximately equally divided between the charcoal and vanadium adsorbers. The average tritium activity we observed in Apollo 16 samples was approximately 200 dpm/kg soil, see section II. It would follow that the fraction of the tritium that diffused out of the soil was about 10^{-4}. This fraction compares well with the fraction of hydrogen gas that diffused out of the soil, and about a factor of ten lower than was observed in the Apollo 15 mission. These diffusion processes were discussed in more detail in our earlier report (Stoenner, et al., 1972).

Tritium was observed in the solar flare particles from the flare of November 12, 1960 by Fireman, DeFelice and Tilles (1961). This flare had an intensity of 10^9 protons/cm^2 with energy above 30 MeV. In our analysis of lunar samples we have been interested in obtaining further evidence for tritium from the sun. Very high amounts (20-200 dpm) of tritium were observed lightly adsorbed on two Apollo 12 samples (12063 and 12065), and searches for sources of tritium contamination within LRL and the spacecraft were made (Stoenner, et al., 1971, 1972). Searches for adsorbed tritium on rock 12065 made six months later by D'Amico, et al., 1971, revealed that less than 0.01 dpm T was present. There still remains a question whether the high tritium level observed on the two Apollo 12 rocks was from the sun or from some unknown source of contamination. If there is a high concentration of tritium present in solar flare particles or solar wind one might expect to observe it in the gas in the SRC, particularly after a solar flare event. Tritium measurements were not carried out on the Apollo 12 because of the difficulties with biological quarantine procedures, but measurements were made on succeeding missions. Very low tritium activities were observed, and these could be explained by diffusive losses of hydrogen from the fine material in the container. There were no flares prior to the Apollo 15 mission, but there were flares prior to the other missions. The flare dates, days before mission, and proton fluxes (>50 MeV protons/cm^2) were as follows (see Fireman, 1972): (12) Nov. 2, 1969, 21 days, $(3.4 \pm 0.9) \times 10^7$; (14) Jan. 24, 1971, 12 days, $(3.7 \pm 0.6) \times 10^7$; (16) April 19, 1972, 2 days, $(0.7 \pm 0.3) \times 10^7$. The intensity of these flares prior to the Apollo 12, 14 and 16 missions were about two orders of magnitude less than that of Nov. 12, 1960 that Fireman and his associates found to contain tritons. However, the flare of Aug. 4-9, 1972, 124 days prior to the Apollo 17 mission was very intense, and the total number of protons/cm^2 with energy above 60 MeV was 2.3×10^{10} (Solar Geophysical Data, 1973). However, we found very low tritium levels in both SRC's from this mission. We have concluded that the tritium observed in the sample return containers from the Apollo 14, 16, and 17 missions could be accounted for by diffusion of cosmic ray produced tritium from the fine material present. There is no evidence for a triton component in the solar flare particles from these experiments. The fact that lightly adsorbed tritium was not observed from the Apollo 17 SRC's can perhaps be explained by noting that the lunar surface at the collection site was exposed to two months of direct solar radiation, and all easily evolved hydrogen was lost.

The ^{39}Ar activities are all very low and are at the limit of the sensitivity of our counters. The ^{37}Ar activities are considerably higher, well above counter backgrounds, and in all cases a clearly resolved ^{37}Ar peak at 2.8 keV was observed (see Stoenner, et al., 1970, for counting technique). The ^{37}Ar is produced in lunar materials principally by the ^{40}Ca$(n,\alpha)^{37}$Ar reaction from galactic and solar cosmic rays. The amount of ^{37}Ar present in lunar fines, therefore, depends upon the calcium content and the cosmic ray intensity. Since ^{37}Ar has a 35 day half-life it is particularly sensitive to periodic solar flares. However, flare particles only increase the ^{37}Ar levels at shallow depths (<10 g/cm^2) in the lunar soil (Fireman, et al., 1973). The high amount of ^{37}Ar observed in the SRC from the Apollo 16 mission is the result of a large amount of fine material in the container (4.3 kg), with high calcium content, and there was a low intensity solar flare two days before the sample was recovered. The ^{37}Ar activity in both of the Apollo 17 containers is extremely low, whereas one might expect a high level resulting from the large solar flare of Aug. 4, 1972 124 days prior to the Apollo 17 mission. Our ^{37}Ar measurements on surface samples collected during this mission are as high as 120 dpm/kg (~900 dpm/g on Aug. 4), but samples at depths greater than 50 g/cm^2 were approximately a factor of two lower than samples from the corresponding depth from the Apollo 16 mission, possibly because of lower calcium content. The low ^{37}Ar values obtained from the Apollo 17 containers cannot be attributed to incomplete collection of the gas from the container, because the drop in pressure was carefully observed during the collection of the gas, and the quantity of gas recovered corresponds closely to the initial pressure in the container. Perhaps the diffusion of ^{37}Ar from the material in these containers was unusually low. We have not studied the rate of ^{37}Ar diffusion from Apollo 17 samples to test this possibility.

The loss of radon from the lunar soil and into the lunar atmosphere has been the subject of many investigations. Of special interest are the spacecraft observations that have revealed that a region containing the Crater Aristarchus is unusually high (Gorenstein and Bjorkholm, 1973). However, in general the amount of radon in the lunar atmosphere is low, and this may be attributed to the low emanating power of lunar soil. We have been particularly interested in observing ^{222}Rn in the sample return containers because they contain a large mass of lunar fines, and the physical and chemical characteristics of the soil in the container were essentially the same as they were when present in the high-vacuum, dry environment of the lunar surface. We have improved our system for recovering radon by using charcoal low in radium, and have eliminated grease-lubricated stopcocks from our system. The sample recovered is counted in a small proportional counter with a well measured counting efficiency for radon and its daughters. Thus by using a large mass of lunar sample and a counter with a very low background we can measure very low emanation rates. The measured radon activities at the time of puncturing the box, and corrected for the radioactive growth factor for the period of time the container was closed are given in Table 2. From these saturation ^{222}Rn production rates, the weight, and average uranium content of the fine material in each container, we have calculated the fraction of ^{222}Rn emanated. The values are listed in Table 3. The emanation rates obtained are considerably lower than the values reported by Adams, et al. (1972), and in the same range as the values reported by Yaniv and Heyman (1972). The relation between these emanation measurements and the lunar atmosphere was discussed in our earlier report (Stoenner, et al., 1972). Low emanation rates observed here are consistent with the observed low alpha particle emission rates from the surface of the moon.

II. Radioactive ^{37}Ar, ^{39}Ar, Tritium, and ^{222}Rn, and the Hydrogen and Helium Concentrations in the Apollo 16 Drill Stem

The primary interest in these investigations was to measure the ^{37}Ar (35.1 day) production rate in the lunar soil by cosmic rays. This isotope is of particular interest because it is produced primarily by the fast neutron reaction ^{40}Ca(n,α)^{37}Ar, and can therefore be used to measure the fast neutron production rate as a function of the depth. The slow neutron intensity was measured to a depth of 2 meters by the Apollo 17 neutron probe experiment (Woolum, et al. 1973). These slow and fast neutron fluxes are of special interest in understanding the long term development of the lunar regolith as revealed by isotopic variations in gadolunium and

and samarium (Russ III, et al. 1972). The Apollo 16 drill stem samples were measured in two laboratories. Fireman and his associates (1973) at the Smithsonian Astrophysical Observatory measured samples from the surface to a depth of 27 g/cm^2, and we measured samples from 83 g/cm^2 to 342 g/cm^2 depth. In addition to ^{37}Ar, we measured three other radioactive products, ^{39}Ar (269 year), tritium and ^{222}Rn. Argon-39 is produced as a spallation product from titanium and iron, and by (n,p) reaction on potassium. Radon-222 arises from ^{226}Ra decay and it is of interest to test whether the equilibrium amount is present in the lunar regolith that would be expected from the decay of ^{238}U. Our vacuum melting and gas separation techniques also permitted measuring the hydrogen and helium concentrations in the lunar soil as a function of the depth. These measurements are of interest in understanding the solar wind fluxes and H/He ratios in the past. Elemental analyses of these same samples are very important in the interpretation of the results, and for this reason 25 mg aliquots of the samples were analyzed for K, Ca, Ti, Fe, and Mn. A similar set of measurements on a more comprehensive set of samples are in progress on the Apollo 17 drill stem, but these will not be reported here but will be referred to in the discussion. Again we would mention that we will not give a detailed analysis of the results, but will do so after the Apollo 17 results are completed.

Experimental

The vacuum melting procedure and counting techniques were described in our earlier report (Stoenner, et al., 1971). However, there were some important improvements that will be described. Radon can readily dissolve in grease, and our glass system used prior to the Apollo 16 utilized grease-lubricated stopcocks. For the drill stem measurements reported here our glass system was completely rebuilt and high vacuum Teflon plunger type valves with viton O-ring seals were used. The system is similar to that diagrammed in our earlier paper, but without quarantine restrictions it could be combined into a single system (Stoenner, et al., 1970).

The sample (1.5 to 5 g) was weighed into a platinum crucible that fitted into a molybdenum crucible with a close-fitting alumina protective liner. The crucible was heated by induction inside a quartz tube that was attached to an automatic Toepler pump to collect the evolved gases. The purification, separation, and subsequent treatment of He, Ar, Rn, and H_2 utilizes the same equipment and procedures as described in the previous section.

The system was evacuated, the crucible baked out, and the line flamed prior to each sample to eliminate water contamination. The sample was ground to 80 mesh, a 25 mg aliquot taken for chemical analysis, and the remainder placed in the platinum crucible. The system was evacuated for four minutes reaching a vacuum of 10^{-4} Torr. With the sample present in the system for 16 to 72 hours, the line was flamed and the accumulated gas collected and carried through the procedure outlined above. This procedure tests the line blank for hydrogen, and tests for the presence of lightly adsorbed tritium. The line hydrogen blank was in the range 0.05 to 0.15 cm^3 S.T.P., and less than 0.020 dpm T/g was observed. Argon and hydrogen (0.15 cm^3 S.T.P. each) were added to the system for carriers, and the sample was melted. The sample was maintained at 1300°C for one hour, and finally the gases were collected with the Toepler pump while the sample was held at the maximum temperature.

The counting procedures used were identical to those described earlier, except for the ^{37}Ar measurements on samples 69941,16 and 69961,17.

These samples were received very late and it was necessary to use a rise-time analysis of the pulses to resolve ^{37}Ar decay events from ^{39}Ar events. A counting system was used that measures both the rise-time of the pulse, and the pulse height for each event (Davis, et al., 1972). The events are plotted on a two dimensional plot, the rise-time (called amplitude of the differentiated pulse) on the ordinate, and the pulse height (energy) on the abscissa. The 2.8-keV Auger electron from an ^{37}Ar decay event has a very short range in the counter gas ($\sim$ 0.05 mm), and therefore the ion pairs created are localized in the counter gas. These ions are collected at the center-wire as a bunch and give a fast-rising pulse. On the other hand the energetic betas from ^{39}Ar decays, in passing through the counter gas, produce ion-pairs along the track of the beta. If in a given event the same number of ion-pairs are produced, they will be more widely separated in the gas, consequently collected over a longer period of time, and result in a slower rising pulse. The energy scale and region of fast events was calibrated with an ^{55}Fe x-ray source. Figure 1 shows a plot of counting events recorded for sample 69941,16 during a 36.7 day period. The region of fast events is defined by the heavy lines, and the energy region for ^{37}Ar decays centered about 2.8 keV is indicated by the vertical lines (fwhm). The cluster of ^{37}Ar events (146) can be clearly observed in the region defined by the correct rise-time and energy for ^{37}Ar Auger electrons. The multitude of ^{39}Ar events (971) occur well outside of the region of fast events.

To test our procedures for measuring radon a sample of a standard rock (W-1) containing 0.58 μg U per gram was melted following the identical procedures used with lunar material. The ^{222}Rn observed for two aliquots of this rock was 0.41 and 0.43 dpm/g. These values can be compared to the equilibrium rate of 0.43 dpm ^{222}Rn/g corresponding to the uranium content of the rock. The ^{222}Rn counting efficiency of the small proportional counters used was determined by counting the ^{222}Rn from a National Bureau of Standards standard ^{226}Ra solution.

Results and Discussion

The radioactive argon and tritium activities observed in the Apollo 16 drill stem are given in Table 4 and the elemental compositions are listed in Table 5. The errors noted are statistical (1σ). The depth of each sample is given, based on the assumption that although section 60005 was returned with only 76 grams of material compared to approximately 200 grams contained in the other sections, it was in fact full at the time of sampling. If there was truly a void at the level of 60005, the depth of the four lowest data points would be decreased by about 40 g/cm^2.

Argon-37, Argon-39, and Tritium. In figures 2 and 3 are plotted the ^{37}Ar and ^{39}Ar as a function of the depth. Included in this plot are four points from the upper section of the deep drill stem measured by Fireman and his collaborators at the Smithsonian Astrophysical Observatory (SAO). The calcium composition of all deep samples was very uniform. Since ^{37}Ar was produced primarily by (n,α) reaction on ^{40}Ca one expects the ^{37}Ar to be a smooth function of the depth. The results of the measurements at SAO and BNL fit together reasonably well. The flare of April 19, 1972, two days before sample collection, could increase the ^{37}Ar levels in the surface samples and have little effect on the deeper samples (Fireman, et al., 1973). The ^{37}Ar increases by a factor of 1.9 from the surface to a maximum in the range 50-100 g/cm^2 and then falls with an exponential mean free path of 205 ± 30 g/cm^2. This general shape and magnitude when scaled for calcium content agrees well with the calculations of Reedy and

Arnold (1972). However, their calculated mean free path, 300 g/cm^2, is greater than we observed. A comparison of these results with a more detailed calculation will be made by Kornblum, et al. (1973) in these proceedings. One may integrate under the depth curve to obtain the total ^{37}Ar production rate of 0.35 ± 0.04 dis sec^{-1} cm^{-2} in the soil. It is presumed that the ^{37}Ar is produced mainly from (n,α) reaction with calcium, though there is some ^{37}Ar produced by spallation of titanium and iron. It may be possible to determine the relative spallation yield of ^{37}Ar by comparing the Apollo 16 and 17 drill stems at equivalent depths if their calcium contents are different. Argon-39 is produced primarily by spallation on iron and titanium and the activity levels are about a factor of 7.5 lower than the ^{37}Ar activities. The spallation cross sections for producing ^{37}Ar and ^{39}Ar at 600 MeV are about comparable for both iron (5.6 mb and 6.3 mb, respectively) and titanium (11.6 and 15.7, respectively, Stoenner, et al., 1970). It would follow that approximately 12 percent of the ^{37}Ar is produced by spallation on iron and titanium in the Apollo 16 drill stem.

The ^{39}Ar activity as a function of the depth is shown in figure 3, and it may be observed that the production rate increases from surface values by a factor of approximately 1.6 and thereafter falls off with depth with an exponential mean-free-path of 166 g/cm^2. Comparing this behavior with the Reedy and Arnold (1972) calculations, we find the measured ^{39}Ar activities are about 1.7 times higher than calculated (they used 16.8 percent Fe and 1.5 percent Ti) and the calculated exponential mean-free-path is greater (280 g/cm^2). The tritium values appear to vary considerably. There is an apparent decrease with depth but a reliable value of L cannot be derived. Tritium is a high energy evaporation product, and one would expect it to be produced primarily from oxygen and aluminum. It is difficult to understand why the tritium results are so erratic, particularly since the fines appear to retain hydrogen reasonably well.

Two soil samples were measured that were collected from station 9. One sample (69941) was scooped from the surface and the ^{37}Ar and ^{39}Ar activities were comparable to values Fireman and his associates (1973) obtained for surface samples. The second sample (69961) was collected under a 0.5 meter boulder, corresponding to a depth of roughly 100 g/cm^2. The ^{37}Ar activity is higher than would be expected from the drill-stem measurements, but the ^{39}Ar activity compares well with the drill-stem measurements. These station 9 samples have Ca, Ti, and Fe contents similar to those of the drill stem at station 10 (170 meters from LM).

Radon. The ^{222}Rn values are a direct measurement of the ^{226}Ra present in the samples. One would expect the ^{226}Ra in these samples to be in equilibrium with the ^{238}U present. As mentioned earlier our extraction system was tested by melting samples of the standard rock W-1 and the quantity of ^{222}Rn was obtained that corresponded to the uranium content. Uranium was not measured on these particular samples, but one can compare our ^{222}Rn measurements with the uranium concentrations of the drill stem measured by Silver (1973). The radon extracted from the drill stem samples and the two station 9 samples are listed in Table 6, and compared with the equilibrium ^{222}Rn disintegration rate calculated from Silver's uranium contents. Our ^{222}Rn activities are about a factor of two low in the top two samples of the drill stem series, and in the remaining samples the agreement is better, though far from satisfactory. The lack of agreement possibly is due to the fact that Silver's uranium measurements may not be applicable to our specific samples, and to resolve this question we are measuring the uranium content on an aliquot of the particular sample melted in the Apollo 17 drill stem. These data will be reported later, but do show in general that the ^{222}Rn activities are about 20 percent lower than the calculated equilibrium production rates. This behavior for lunar soil samples is hard to understand particularly in view of the excellent agreement with the equilibrium rates obtained with W-1 rock standards.

Hydrogen and Helium. Since rather large samples were required for the radioactivity measurements, it was important to measure the hydrogen and helium recovered, and compare our volumes with those measured by other investigators. The hydrogen and helium contents of the drill stem samples and the two station 9 samples are given in Table 5, estimated errors are 10 percent for hydrogen and 5 percent for helium. The hydrogen reported would include water hydrogen in the sample. It may be observed that the hydrogen content of the lunar soil from the drill stem is relatively constant, 1.2 ± 0.2 cm^3 STP per gram. These results compare well with total hydrogen reported for soil samples by other investigators (for example, Epstein and Taylor, 1972; and Friedman, et al., 1972), and our earlier results (Stoenner et al., 1970). The helium content of the drill stem is also relatively constant except for the deepest sample. It is especially interesting to compare these hydrogen and helium contents of the Apollo 16 drill stem with those measured by Kaplan and Petrowski (1973) in the Apollo 15 drill stem at similar depths. They found the helium content was relatively constant, 0.050 to 0.067 cm^3 STP, except for the deepest sample (0.039 cm^3 STP). They obtained hydrogen contents at three depths and found that the hydrogen content decreased with depth: 0-40 cm, 0.37 cm^3; 120-160 cm, 0.25 cm^3; 200-240 cm, 0.05 cm^3 STP). We found in the Apollo 17 drill stem that both hydrogen and helium increased with depth (He, 0.069 to 0.14 cm^3 STP;

H_2, 0.80 to 1.63 cm^3 STP). It is clear from these results that the depth variation of the hydrogen and helium contents of the lunar regolith varies with the location. The hydrogen and helium are derived from the solar wind and one might hope to observe long period variations in the solar wind flux by studying the depth variation of these gases in the lunar soil.

The H/He ratio in the sun is a very important quantity, and plays an important role in astrophysics. This ratio has been measured spectroscopically in solar prominences, and the chromosphere, in solar cosmic rays, and the solar wind (see review of Hirshberg, 1973). The spectroscopic values are considered by Hirshberg to be the most accurate and give a H/He atom ratio of 12 to 20. Solar wind H/He ratios give a value of 7 to 10, and solar cosmic rays indicate a H/He ratio of about 17 at the flare site. The H/He atom ratio we observed in the Apollo drill stem and the station 9 samples is relatively constant (with the exception of the bottom of the drill stem), and give an average H/He ratio of 49. This ratio is higher than the solar wind value, and can be explained by contamination with terrestrial water. Epstein and Taylor (1972) finds about 50 percent of the hydrogen in lunar fines is present as water. Our samples were also probably contaminated with terrestrial water, and if so this would explain the high H/He ratios. Studies of drill stem hydrogen and helium contents coupled with exposure age measurements should be a valuable method for studying the history of the solar wind flux, and H/He ratios. It is possible that the sun's luminosity has varied in the past few million years (see recent review by Cameron, 1973), and it would be interesting to search for possible changes in solar wind and solar cosmic ray intensities in lunar material.

Acknowledgments

We would like to acknowledge the assistance of Dr. Donald Bogard and Mr. Lewis Sims in setting up the apparatus and collecting the gas from the sample return container. We are indebted to LSAPT, the Curator's office and the staff at the Lunar Receiving Laboratory for their help in making early allocations of samples from the drill stems. Dr. Don Burnett stimulated our interest in making the drill stem measurements, and assisted us on several technical problems. We would like to acknowledge many helpful discussions with Dr. John C. Evans during the progress of this work.

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Table 1. Gases collected from the sample return containers.

Mission-SRC	SRC Pressure (Torr)	Volume Gas Collected (cm^3 STP)	Volume H_2 on Vanadium (cm^3 STP)	Volume H_2 on charcoal (cm^3 STP)	SRC Pressure Calculated from Recovered gas (Torr)
16-2	--	3.93	0.35	0.05	0.17
17-1	0.170	4.79	0.87	0.90	0.20
17-2	0.025	0.277	0.12	0.07	0.012

Table 2. Radioactivities from the sample return container

		SRC Pressure (Torr)	Wt. fines in SRC (kg)	Observed activity (dpm)				
Mission	Date			^{222}Rn	^{37}Ar	^{39}Ar	T from Charcoal	T from Vanadium
12	Nov 19-20, 1969	0.030	2.7	4.9 ± 0.3	0.040 ± 0.002	0.0023 ± 0.0010	---	---
14	Feb 5-6, 1971	0.070	0.6	1.74 ± 0.02	0.040 ± 0.002	0.0008 ± 0.0005	0.280 ± 0.020	---
15	July 30-Aug 2, 1971	0.032	2.3	0.65 ± 0.04	0.106 ± 0.003	0.0030 ± 0.0012	0.279 ± 0.005	0.275 ± 0.007
16	April 21-24, 1972	--	4.3	0.116 ± 0.007	0.155 ± 0.005	<0.002	0.045 ± 0.002	0.045 ± 0.002
17-1	Dec 11-14, 1972	0.170	2.0	0.181 ± 0.012	0.0052 ± 0.0014	<0.002	0.034 ± 0.002	0.051 ± 0.002
17-2	Dec 11-14, 1972	0.025	7.7	0.131 ± 0.007	0.0087 ± 0.0014	<0.002	0.005 ± 0.001	0.006 ± 0.001

Table 3. Radon emanation rates in sample return containers

Mission, SRC	Average U Content (ppm)	Total ^{238}U Disintegration Rate in Fines (dpm)*	Fraction Emanated (X 10^3)
12	0.9	1800	2.7
14	3.4	1500	1.2
15	1.4	2400	0.3
16-2	0.64	2000	0.06
17-1	0.25	370	0.5
17-2	0.25	1400	0.09

* 0.74 dpm $^{226}Ra/\mu g$ U.

Table 4. Radioactive argon and tritium in the Apollo 16 drill stem

Sample No.	Depth (g/cm^2)	Activity (dpm/Kg) ^{37}Ar	^{39}Ar	T
60006,12	83	62.8 ± 2.4	11.5 ± 0.6	130 ± 4
60004,18	143	59.5 ± 2.0	8.0 ± 0.5	442 ± 10
60004,10	205	39.4 ± 1.9	6.6 ± 0.5	166 ± 2
60003,6	272	31.7 ± 1.7	3.5 ± 0.5	151 ± 5
60001,13	342	20.8 ± 1.2	2.7 ± 0.4	227 ± 6
69941,16	Scooped	56 ± 4	8.2 ± 0.4	246 ± 5
69961,17	~100	97 ± 6	10.7 ± 0.5	283 ± 4

Table 5. Chemical composition of samples from the Apollo 16 drill stem

Sample	Percent Composition					H_2	He
No.	K	Ca	Ti	Fe	Mn	(cm^3 STP/g)	(cm^3 STP/g)
60006,12	0.25	11.7	0.33	3.9	0.063	1.0	0.055
60004,18	0.27	11.5	0.41	4.8	0.069	1.3	0.054
60004,10	0.23	11.4	0.39	4.8	0.065	1.1	0.047
60003,6	0.35	11.7	0.39	5.0	0.068	1.4	0.056
60001,13	0.34	11.7	0.37	4.7	0.067	1.1	0.025
69941,16	0.094	11.3	0.36	4.4	---	0.8	0.041
69961,17	0.096	10.4	0.37	4.7	---	0.6	0.033

Table 6. Comparison of ^{222}Rn activity and uranium concentrations in the Apollo 16 drill stem

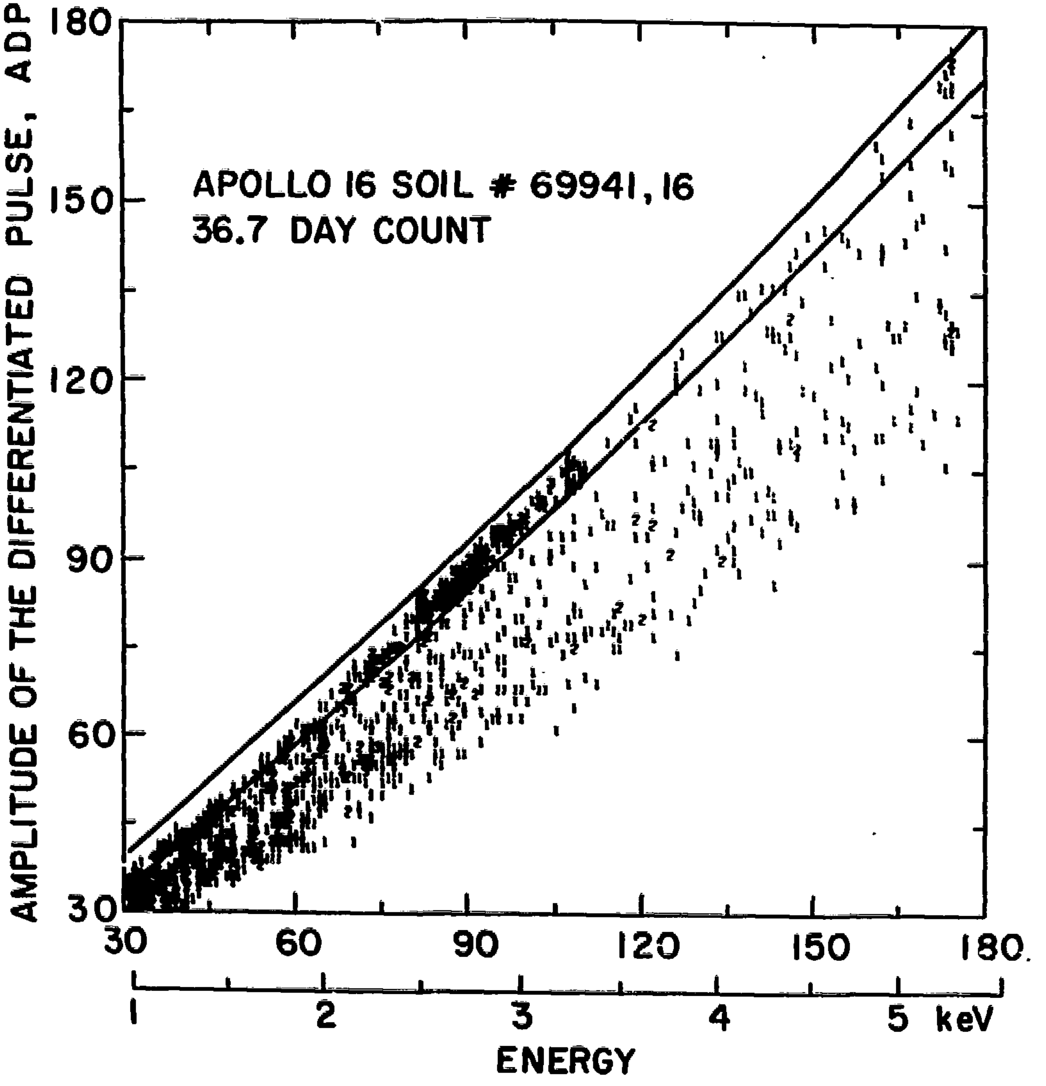
Sample No.	^{222}Rn (dpm/g)	^{222}Rn calc. from U content* (dpm/g)
60006,12	0.26 ± 0.01	0.53 (6)
60004,18	0.29 ± 0.01	0.47 (21)
60004,10	0.38 ± 0.01	0.50 (12)
60003,6	0.54 ± 0.01	0.48 (8)
60001,13	0.61 ± 0.01	0.61 (6)
69941,16	0.58 ± 0.02	0.56 (22)
69961,17	0.43 ± 0.01	0.53 (25)

*U from Silver (1973), number in parenthesis is the fraction number of the same parent sample. ^{222}Rn in dpm/g = 0.74 x (ppm U).

Fig. 1. Pulse rise-time (ADP) versus pulse height (Energy) plot for Apollo 16 soil sample. Fast-rising ^{37}Ar pulses should fall in fast region defined by heavy lines, and half-width (fwhm) defined by channels 80 to 107 on energy scale.

Fig. 2. Argon-37 as a function of the depth in the Apollo 16 deep drill stem.

Fig. 3. Argon-39 as a function of the depth in the Apollo 16 deep drill stem.



"Radioactive Rare Gases ..."

Fig 1 Stoenner and Davis

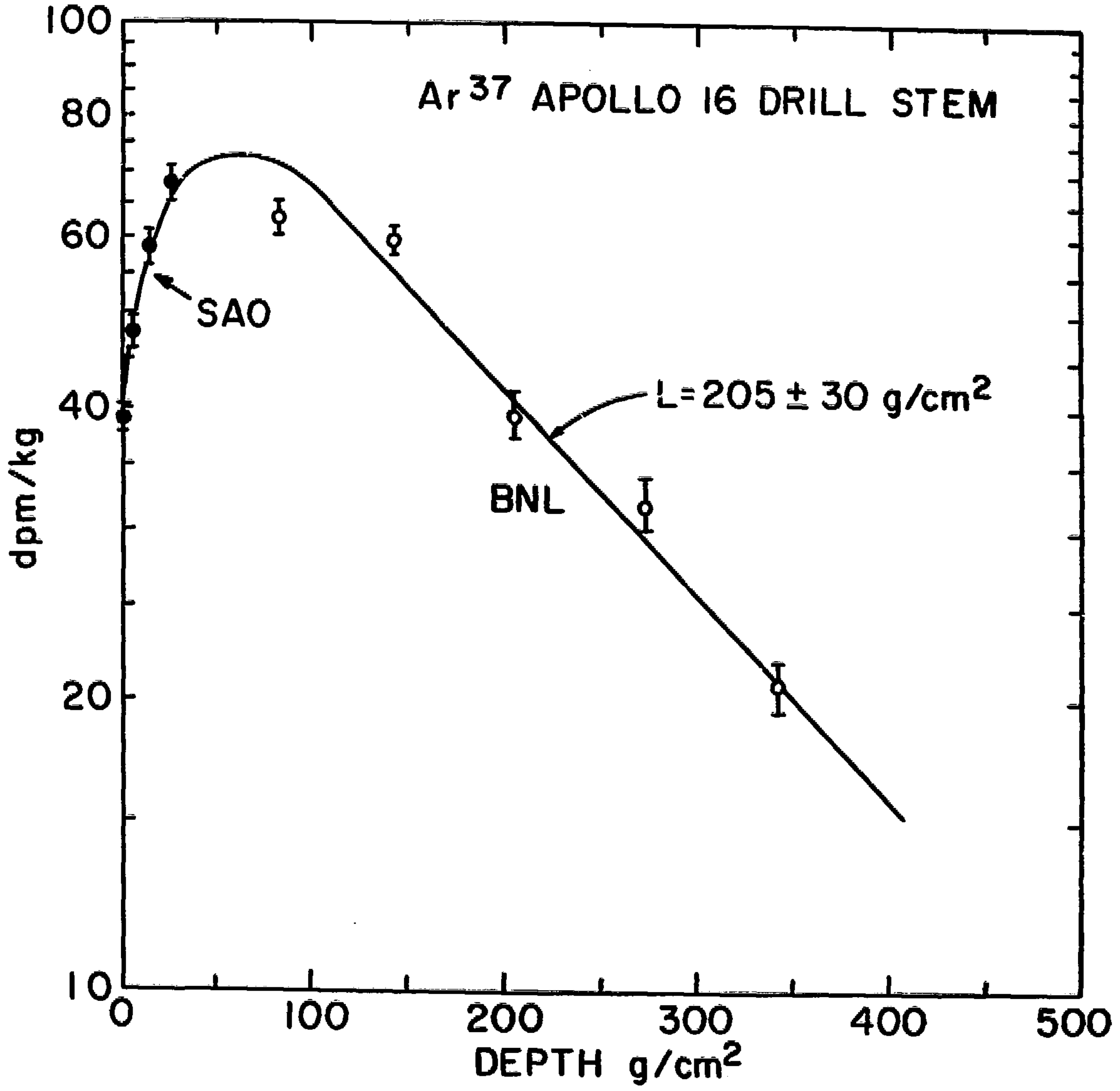


Fig 2 Stoenner and Davis

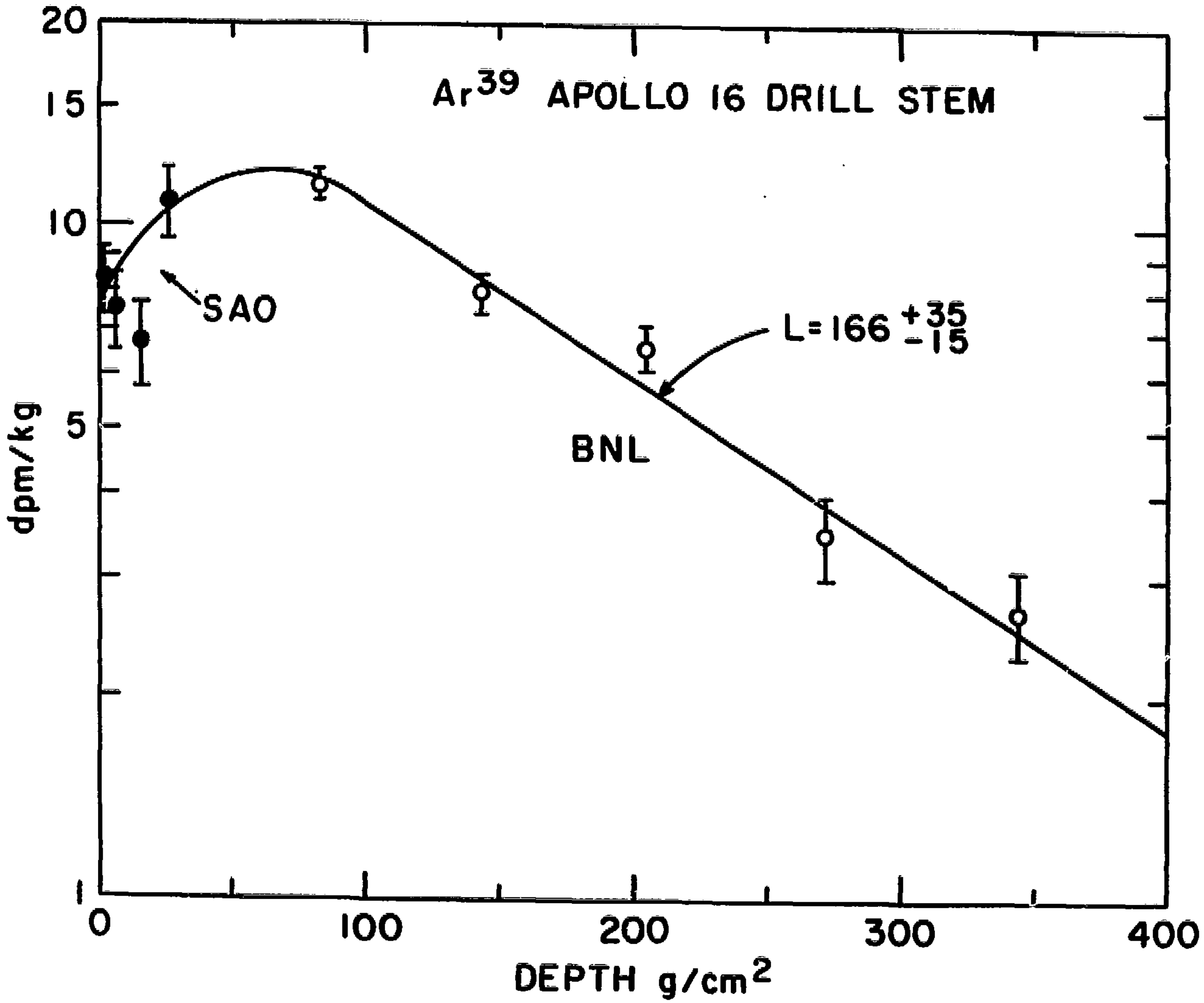


Fig. 3 Stoenner and Davis