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FINAL REPORT ON NASA CONTRACT

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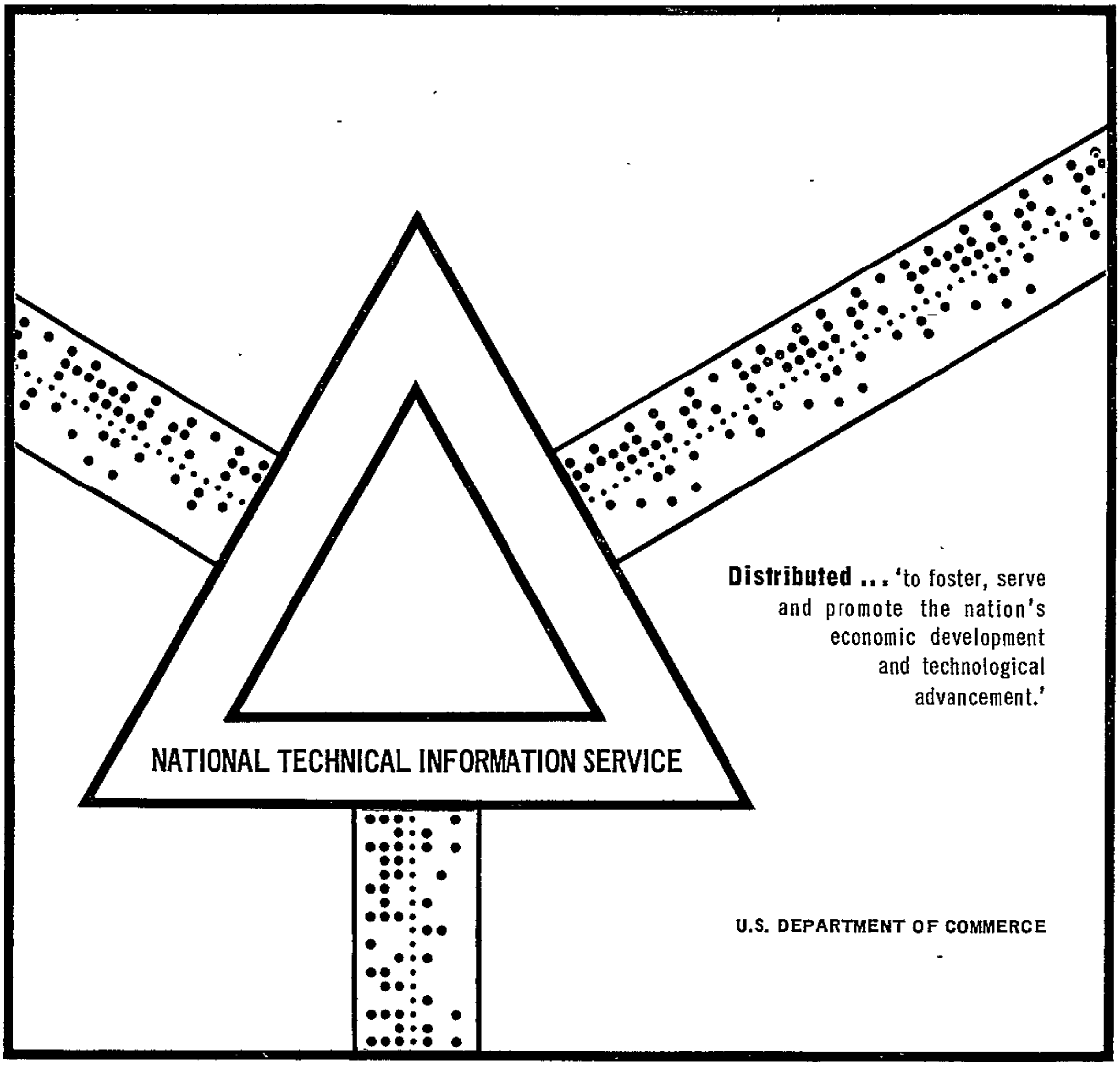

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Contents

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COMPARISON OF THE ANALYTICAL RESULTS FROM THE SURVEYOR, APOLLO, AND LUNA MISSIONS*

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ABSTRACT

The principal chemical element composition and inferred mineralogy of the powdered lunar surface material at five mare and one terra sites on the moon are compared. The mare compositions are all similar to one another and comparable to that of terrestrial ocean ridge basalts except in having higher titanium and much lower sodium contents than the latter. These analyses suggest that most, if not all, lunar maria have this chemical composition and are derived from rocks with an average density of 3.19 g cm^{-3}. Mare Tranquillitatis differs from the other three maria in having twice the titanium content of the others.

The chemical composition of the single highland site studied (Surveyor 7) is distinctly different from that of any of the maria in having much lower amounts of titanium and iron and larger amounts of aluminum and calcium. The inferred mineralogy is 45 mole percent high anorthite plagioclase and the parent rocks have an estimated density of 2.94 g cm^{-3}. The Surveyor 7 chemical composition is the principal contributor to present estimates of the overall chemical composition of the lunar surface.

* Presented at the Apollo 12 Lunar Science Conference, Houston, Texas, January 11-14, 1971; submitted for publication in the Proceedings of this Conference.

Analytical information for the principal chemical elements is available at present from six sites on the moon. The location of these sites, their characteristics, and the nature of the analytical technique used, are summarized in Table 1. The phrase "principal chemical elements" is used here to denote the elements usually present in rocks in amounts greater than about 0.3 atom percent. They comprise the elements O, Na, Mg, Al, Si, Ca, Ti and Fe. These elements constitute about 99% of the atoms, and, therefore, determine the gross chemical nature of the lunar surface. The analyses compared in this report are those of the powdered material at a given site. It is felt that, in the case of the principal chemical elements, the chemical composition of this material represents the average of a given area of the moon better than the composition of individual rocks.

Although the Surveyor analyses, obtained by the remote control alpha scattering technique, are usually appreciably less precise than those on returned lunar samples, they represent half of the sites on the moon from which information is available. Their accuracy is adequate to establish the gross rock type and even some of its special characteristics. In addition, the Surveyor 7 chemical analysis is at present the only one from a highland site on the moon.

Finally, at the present stage of lunar and planetary investigations, a comparison between remote control analytical results and those obtained on returned samples can help make proper decisions about future exploration of the moon, asteroids and planets. In this connection it should be borne in mind that the capabilities of remote control analyses by various techniques are continually improving. The Surveyor missions represent the state of technology of one particular technique almost ten years ago. Not only could the accuracy for the principal chemical elements be improved today, but there are possibilities, also, of measuring some of the minor constituents.

Table 2 presents the results of chemical analyses of surface lunar fines for the principal chemical elements at the five lunar mare sites that have been studied so far. Estimates of the accuracies of the Surveyor analyses are indicated; the accuracies of the analyses of the returned samples are probably all less than 3% at the same confidence level. Although the Surveyor analyses for Ca, Ti, and Fe are, strictly speaking, for groups of elements with about the same atomic weight, assignment of the values primarily to the elements indicated appears justified on geochemical grounds.

The comparison between the Surveyor 5 and Apollo 11 analyses, columns 2 and 3 of Table 2, is particularly to be noted since both apply to Mare Tranquillitatis, at sites less than 30 km apart. It is seen that the agreement is just about within the Surveyor analytical errors except in the case of Fe, where the Surveyor answer is some 25% lower than the Apoll result. The analytical results for all the maria lead to a similar geochemical picture - that of a silicate rock with Al and Ca each comparable to or greater than Mg, low Na (0.3 to 0.6 atom percent) and relatively high Ti (0.9 to 2.2 atom percent). The Fe content is also relatively high at 3.8 to 5.4 atom percent. Although there may be some indication of systematic biases in the alpha scattering method leading to high values of Si and low values of Mg and Fe, the variations in mare compositions illustrated in Table 2 are relatively small.

Since the first chemical analysis of the lunar surface on the Surveyor 5 mission (Turkevich et al., 1967), the compositions of the lunar maria have been compared with those of terrestrial basaltic rocks. The more extensive data available now make possible a more detailed comparison. This is done in Fig. 1, where the amounts of Mg, Al, Ca, Fe, Ti and Na in the lunar maria are compared with the amounts of these elements in terrestrial ocean ridge basaltic rocks (Kay et al., 1970). The comparison is not made for O or Si since these are not too sensitive indicators of rock type. There is, however, an indication that the amount of Si in the lunar maria may be one to two atom percent lower than in terrestrial basaltic rocks.

Fig. 1 shows that the Al, Mg and Ca contents of the lunar maria are well within the ranges observed by Kay et al. In the case of Fe, also, there is overlap, although the overlap is provided primarily by the alpha scattering results, both of which are lower than those on returned lunar samples. Thus, for the six chemical elements present in amounts greater than three atom percent, the chemical composition of the lunar maria is quite well represented by that of terrestrial ocean ridge basaltic rocks.

The striking differences between the maria compositions and those of terrestrial basaltic rocks are in the high Ti and low Na contents of the former. Both these aspects are well illustrated in Fig. 1. Even larger differences in the amounts of minor constituents have, of course, been noted (see e.g. LSPET, 1969, 1970).

The principal variation in composition among the different maria is in the Ti content, which is twice as high in Mare Tranquillitatis than in the other three lunar maria studied. The apparently even larger variation in Mg content appears to be at least partially due to the larger errors in the Surveyor analyses for this element. On the other hand, the Al content at all five sites is remarkably constant at 6.6 ± 0.3 atom percent.

Considering the fact that the four maria sampled are widely separated on the moon - although all close to the equator - these analyses suggest that most, if not all lunar maria have this gross chemical composition. An average lunar mare chemical composition calculated on the assumption that each analysis is representative of its mare (the Surveyor 5 and Apollo 11 data for Mare Tranquillitatis were averaged) is given in column 2 of Table 3.

Column 3 of Table 3 gives the average lunar soil composition determined by the alpha scattering technique on the Surveyor 7 mission to a highland site outside the crater Tycho (Patterson et al., 1970). It is seen that the composition here is distinctly different from that of the maria. There is about a factor of three fewer atoms of elements heavier than Ca (i.e. much less Fe and very little, if any, Ti), and about a 40% increase in Al and Ca contents. The amount of Si is, within the Surveyor errors, the same as in the maria.

The applicability of this analysis at one highland site on the moon to the other highland areas is at present only speculative, even though fragments of similar composition have been found in the Apollo 11 soil (e.g. Smith et al., 1970; Wood, 1970) and in some of the Apollo 12 breccia rocks (e.g. Anderson et al., 1971) and these have been attributed to highland material. If it is assumed that the Surveyor 7 analyses are representative of all the lunar highlands, it has been pointed out (Turkevich et al., 1968b; Phinney et al., 1969; Patterson et al., 1969) that the differences in chemical composition between the lunar terrae and maria may be the explanation for the albedo differences of the major topographical features of the moon. They also imply lower density for the highlands as compared to the maria, with a suggestion that isostatic adjustment has occurred in many regions of the moon.

The chemical composition of the highlands as represented by the Surveyor 7 analyses, also suggests the existence of a crust on the moon, chemically as well as physically different from the main body of the moon (Patterson et al., 1970).

The average chemical composition of the lunar surface, on the assumption that the mare composition of column 2 of Table 3 is applicable to 20% of the surface and the Surveyor 7 composition to the rest, is given in column 4 of Table 3.

Although an elemental chemical composition cannot provide the detailed information about rock type and mineralogy that can be obtained from examination of returned samples, some data of this type can be reasonably inferred from chemical analyses. This was first done using the early Surveyor analyses by Turkevich et al., (1968a) and Phinney et al., (1969). A normative mineral composition is derived following an order that minimizes the uncertainties due to the errors of the Surveyor analyses. The amount of ilmenite is determined by the Ti content. The amount of Al, with its relatively small error, determines the amount of plagioclase feldspar ($CaAl_2Si_2O_8$ and $NaAlSi_3O_8$ with the Na content determining the relative amounts of the two). The remaining atoms of Ca, Mg, Fe, and Si are assigned to pyroxenes and olivine or quartz. It is in the detailed assignment of the different types of pyroxenes and in the amounts of olivine or of silica minerals that the relatively large errors of the alpha scattering method for Mg and Si play the largest role. The results of the above procedure are shown in Table 4 for the six lunar sites for which analyses are available.

The table illustrates, from a mineralogical standpoint, the similarities and differences among the different maria. The higher ilmenite content at Mare Tranquillitatis than at the other maria follows directly from the higher Ti content there. The constancy of the plagioclase contribution at 24.5 ± 2.4 mole percent is a consequence of the constant Al contents of the maria. The anorthite fraction of the plagioclase is constant at 88%, although here the inability of the alpha scattering method to distinguish between Ca and K must be remembered. Likewise, the detailed composition of the pyroxenes and the apparent considerable amount of silica minerals in the results from Surveyors 5 and 6 are less certain because of the rather large errors (and anticorrelation) attached to the Si and Mg results. The gross mineralogical characteristics inferred from the treatment of Table 4 have actually been observed in the case of the powdered lunar material returned to earth by the Apollo 11 and 12 and Luna 16 missions (LSPET, 1969; LSPET, 1970; Vinogradov, 1971).

The last column of Table 4 indicates that the material outside the crater Tycho is representable by negligible ilmenite, and a 50% increase in plagioclase content (again highly anorthitic) over lunar mare material. There appears to be no silica excess; in fact, some olivine content is suggested.

The bottom row of Table 4 gives the particle densities for the material making up the lunar fines at the different sites. They were calculated according to the procedure of Phinney et al., (1969), and represent estimates of the densities of the average rock from which these lunar fines were derived. These average densities at all five mare sites are relatively constant at 3.19 ± 0.06 gm cm^{-3}. The density of the corresponding terra material, as represented by the Surveyor 7 analyses, is significantly lower, 2.94 gm cm^{-3}. These numbers are in good agreement with the estimates made by Phinney et al., (1969) from the preliminary results of the Surveyor missions, namely 3.20 ± 0.05 and 3.00 ± 0.05 gm cm^{-3}.

A result of special interest from the Surveyor 7 chemical analysis is the apparent presence of 0.3 atom percent of F. Since the amount of P that would be required to have this F in the form of apatite was definitely excluded by the analyses, this F may be in the form of CaF_2 or as a partial replacement for O in the silicates. It will be of interest to see if any F is detected in those particles of mare material that are considered as coming from highlands.

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Table 1

Locations on the Moon from which Chemical Composition Data are Available

Mission	Date	Selenographic Coordinates			Geographical Area	Type of Area	Type of Analysis
		Long.	Lat.	Ref.			
Surveyor 5	1967, Sept. 11	23.20°E	1.42°N	a	Mare Tranquillitatis	Mare	in-situ α scattering
Surveyor 6	1967, Nov. 10	1.37°W	0.46°N	a	Sinus Medii	Mare	in-situ α scattering
Surveyor 7	1968, Jan. 10	11.44°W	40.97°S	a	outside Crater Tycho	Terra	in-situ α scattering
Apollo 11	1969, July 20	23.43°E	0.69°N	b	Mare Tranquillitatis	Mare	returned samples
Apollo 12	1969, Nov. 19	23.34°W	2.45°S	c	Oceanus Procellarum	Mare	returned samples
Luna 16	1970, Sept. 19	56.18°E	0.41°S	d	Mare Foecunditatis	Mare	returned samples

(a) Jaffe (1969).

(b) LSPET (1969).

(c) LSPET (1970).

(d) Vinogradov (1971).

Table 2

Concentrations of Principal Chemical Elements in Lunar Mare Material (Fines)

Percent by Atom

Mission	Surveyor* 5	Apollo 11	Surveyor* 6	Apollo 12	Luna 16
Reference	a	b	a	c	d
Element					
O	61.1 ± 1.0	59.87	59.3 ± 1.6	59.9	(60.15)
Na	0.47± 0.15	0.33	0.6 ± 0.24	0.30	0.37
Mg	2.8 ± 1.5	4.57	3.7 ± 1.6	6.8	4.99
Al	6.4 ± 0.4	6.30	6.5 ± 0.4	6.3	6.95
Si	17.1 ± 1.2	16.31	18.5 ± 1.4	16.0	15.97
Ca*	5.5 ± 0.7	4.92	5.2 ± 0.9	4.1	4.99
Ti*	2.0 ± 0.5	2.19	1.0 ± 0.8	0.9	0.98
Fe*	3.8 ± 0.4	5.12	3.9 ± 0.6	5.4	5.39

* For the Surveyor analyses, the elemental symbols Ca, Ti and Fe represent a range of elements (see text and Economou, 1970). The principal contributors are expected to be the elements listed. The quoted errors of the Surveyor analyses are estimates at the 90% confidence leve In the case of Surveyor 7, an amount of fluorine equal to 0.29 ± 0.12 atom percent was also found.

(a) Franzgrote (1970).

(b) Averages of the more accurate of many lunar soil analyses reported in Levinson (1970).

(c) LSPET (1970).

(d) Vinogradov (1971).

Table 3

Comparison of the Chemical Composition of Lunar Maria and Terrae

(Percent by atom)

Element	Average Mare	Terra (Surveyor 7)*	Average Lunar Surface**
O	60.0	61.8 ± 1.0	61.4
Na	0.4	0.5 ± 0.2	0.5
Mg	4.8	3.6 ± 1.6	3.8
Al	6.5	9.2 ± 0.4	8.7
Si	16.8	16.3 ± 1.2	16.4
Ca	4.9	6.9 ± 0.6	6.5
Ti	1.3	0 ± 0.4	0.3
Fe	4.8	1.6 ± 0.4	2.2

* The errors are estimates at the 90% confidence level (see Patterson et al., 1970)

** See text.

Table 4

Normative Mineral Composition Inferred from Chemical Analyses

(mole percent)

		Surveyor 5	Apollo 11	Surveyor 6	Apollo 12	Luna 16	Surveyor 7
Ilmenite		13.1	14.7	6.8	6.8	7.6	-
$FeTiO_3$	il						
Plagioclase Feldspars		22.8	22.2	24.5	24.9	28.4	45.0
$NaAlSi_3O_8$	ab	3.1	2.2	4.1	2.3	2.9	4.6
$CaAl_2Si_2O_8$	an	19.7	20.0	20.4	22.6	25.5	40.4
Pyroxenes		46.5	63.1	59.9	43.6	42.3	39.4
$MgSiO_3$	en	16.4	12.9	15.0	8.3	13.2	22.9
$CaSiO_3$	wo	18.3	30.6	25.2	20.3	15.5	11.0
$FeSiO_3$	fs	11.8	19.6	19.7	15.0	13.6	5.5
Olivine		-	-	-	24.8	21.8	15.6
Mg_2SiO_4	fo	-	-	-	15.4	11.6	11.0
Fe_2SiO_4	fa	-	-	-	9.4	10.2	4.6
Quartz		17.7	-	8.8	-	-	-
SiO_2	qtz	-	-	-	-	-	-
estimated density ($gm\ cm^{-3}$)		3.17	3.27	3.13	3.17	3.22	2.94

Average mare rock density: $\bar{\rho} = 3.19\ gm\ cm^{-3}$

Figure Caption

Fig. 1. Comparison of the amounts of some of the principal chemical elements in lunar mare material with the amounts in terrestrial ocean ridge basaltic rocks. The abscissae are in atom percent. The histograms are the distributions in the amounts of the element as determined by Kay, Hubbard, and Gast (1970). The shaded areas about the Surveyor analyses represent the 90% confidence range of these analyses.

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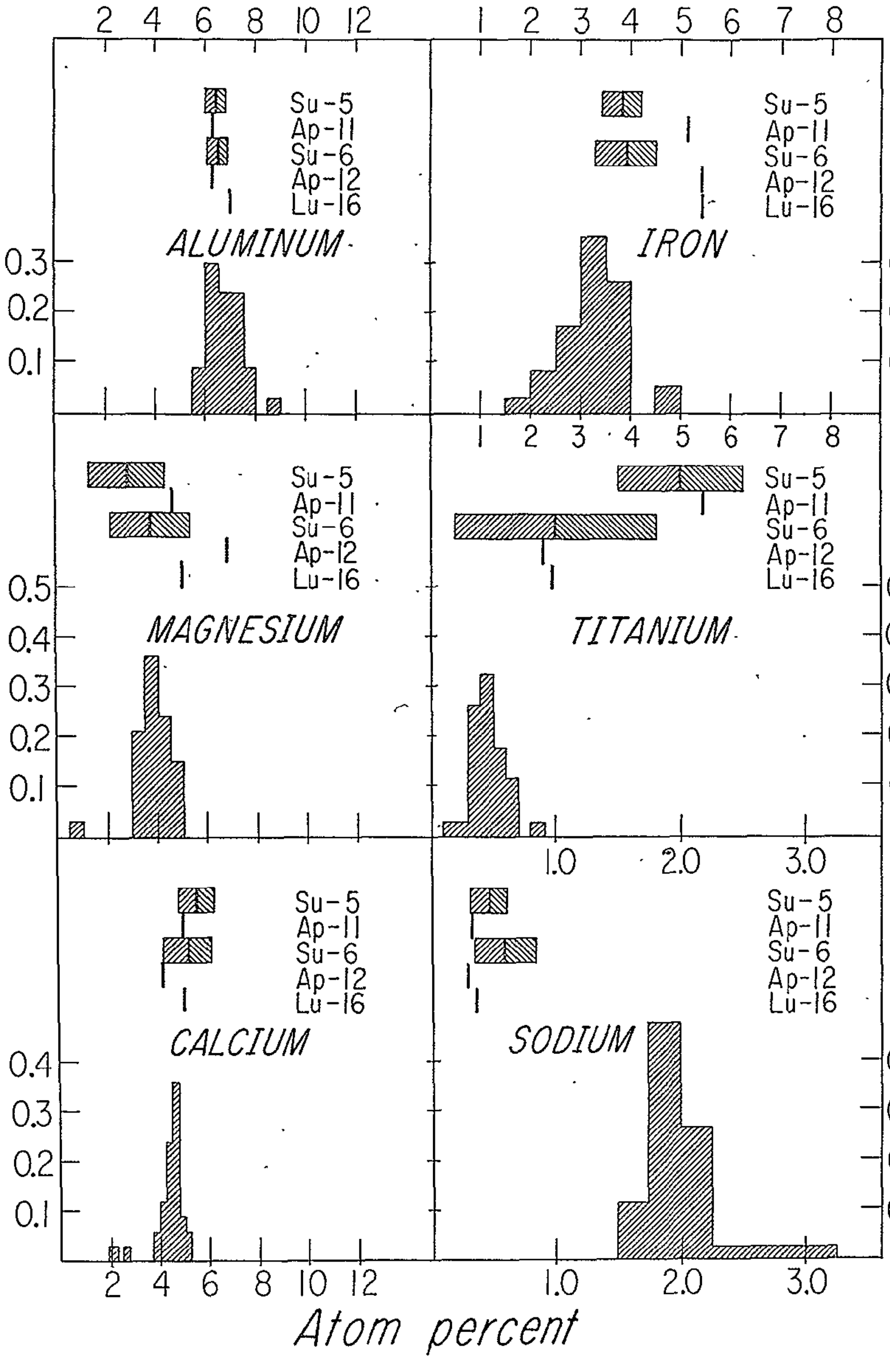

<u>Comparison of Chemical Composition of Six Principal Elements in Lunar Mare Material</u>

Histograms are distributions of the elements in terrestrial ocean ridge basalts (Kay, Hubbard and Gast, 1970).

U and Pb
Analyses

19 February 1971

ACTIVATION ANALYSIS DETERMINATION OF URANIUM AND ^{204}Pb IN APOLLO 11 LUNAR FINES*

N71 24560

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ABSTRACT

The ^{238}U abundance in the Apollo 11 Type D material (10084,58) has been determined via the $^{238}U(n,2n)^{237}U$ reaction with a resultant mean value from four replicates of 0.54 ± 0.02 ppm.

Neutron activation analysis has also been applied to the determination of the ^{204}Pb in the same sample via the $^{204}Pb(n,2n)^{203}Pb$ reaction. Three samples gave results varying from 2.2 to 7.1 ppb. In addition, some data were obtained on the bismuth and thallium contents of this sample.

The ^{204}Pb and ^{208}Pb contents of two chondritic meteorites have also been determined.

I. Introduction

This report summarizes the results obtained on the abundances of some of the heaviest elements in returned lunar material. These elements, thallium, lead, bismuth, thorium and uranium, although present in only trace amounts, are important in understanding the geochemical history of lunar material, as well as the heat production in the moon. There have been several reports on the abundances of these elements in Apollo 11 material. The data for uranium and thorium appear to be well established (e.g. Fields et al., 1970; Gopalan et al., 1970; Silver, 1970; Tatsumoto, 1970). Likewise, the lead has been shown to be very radiogenic (e.g. Gopalan et al., 1970; Silver, 1970; Tatsumoto, 1970), and the absolute amounts found by various workers appear to be in reasonable agreement.

The situation in the case of the other two elements of this group is less satisfactory. Kohman (1970) and Ganapathy et al. (1970) find an overall thallium abundance in Apollo 11 fines of up to 3 ppb, but the latter, as well as Anders et al. (1971) get results as low as 0.3 ppb with an indicated inverse correlation with particle size. The values for bismuth in lunar fines are likewise from Ganapathy et al. (1970) and Anders et al. (1971) who find values up to 1.5 ppb with, again, evidence for a dependence of the results on particle size. The low amounts of these elements make their determination difficult. There is, however, great interest in their abundances since, in meteorites, they have been found to be sensitive indicators of the geochemical history of these objects.

The determination of the amount of ^{204}Pb in lunar material is in somewhat less satisfactory condition. Lunar lead is so radiogenic that the corrections for laboratory contamination in isotope dilution measurements make the published values for the amount of this isotope (again in the range of ppb) particularly uncertain. There has even been some question raised as to whether ^{204}Pb indigenous to lunar material has actually been detected in some of the samples. At the same time, there is considerable interest in the amount of primordial lead in such material since, as in the cases of thallium and bismuth, it is likely to be an indicator of the possible role of the volatility of an element in the geochemical history of the material.

Because of this interest in the heavy elements, it was considered worthwhile to pursue their determination by neutron activation techniques. Although these are almost always less accurate than isotope dilution methods, results obtained thereby have the advantage of being less likely to be invalidated by contamination of the sample. In addition, in the case of ^{204}Pb, the technique used determines specifically this isotope even in the presence of hundreds of times the abundance of the heavier, radiogenic, isotopes. A major effort was, therefore, devoted to the lead measurement, but concurrent bismuth and thallium determinations were made when possible, not only to establish their abundances, but also as internal checks on the lead results. It has been shown (Reed, Kigoshi and Turkevich, 1960) that, in meteorites at least, these three elements exhibit parallel abundance trends.

The nuclear reactions that have been used in the present work and the radiations of the products that were measured are summarized in Table 1. It is seen that both in the case of ^{238}U and ^{204}Pb use has been made of the (n,2n) reaction to supplement the more usual slow neutron induced reactions. Attempts to measure the ^{208}Pb via the (n,γ) production of the 3.3 hr ^{209}Pb isotope were unsuccessful. Other work, cited above, has adequately established the amount of this radiogenic isotope.

II. Experimental*

Except for one irradiation of a breccia, the lunar material investigated was the powdered "fines" from the Apollo 11 mission (sample 10084)**. The samples for our experiments were taken from the shipment containers and transferred inside a N_2 dry box to Super-Sil fused silica tubes, and were sealed immediately after removal from the box. Monitor samples, between 1 μg and 1 mg of the element, were evaporated in similar fused silica tubes. Inert MgO was often added to help retain the recoiling fission products of uranium. Samples and monitors were placed side by side in irradiation cans. The irradiations were carried out in the core of the High Flux Beam Reactor at the Brookhaven National Laboratory. The high energy (>1 MeV) neutron flux in this reactor is of the order of 10^{13} $cm^{-2}sec^{-1}$, while the thermal flux is $\sim 10^{14}$ $cm^{-2}sec^{-1}$. In order to try to isolate the 3.3 hr ^{209}Pb, the samples were worked up within a few hours of removal from the reactor in the Hot Laboratory facilities at the Brookhaven National Laboratory.

After irradiation, the samples were quantitatively transferred to a zirconium crucible containing appropriate carriers and tracers and were fused with Na_2O_2, Na_2CO_3 and NaOH. The HNO_3 solution of the melt was first passed through a silica-gel column and then the Pb and Bi were separated by electrolytic deposition. Additional recovery of the Pb was accomplished by precipitation of the nitrates of Sr, Ba and Pb. The Pb samples were purified by repetition of cycles

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*See Appendix A for some details of the chemical processing of the samples.

**See Appendix B for results on two meteorites.

including sulfide, sulfate and nitrate precipitations and chloroform extractions with dithizone.

The uranium was separated from most of the solution of the fusion melt by an anion exchange column and purified by ether extractions and removal of contaminating radioactivities by scavenging precipitations and solvent extractions. It was finally electroplated onto platinum, with the recovery being measured by the recovery of the ^{234}U tracer added at the start. In the case of lead, barium and thallium, the final samples were in the form of chromates. The final bismuth sample was in the form of the oxychloride.

The samples from the monitors were usually subjected to less extensive chemical purification, but were finally prepared for radioactivity measurements in the same fashion as were the samples isolated from the irradiated lunar material.

For each nuclide of interest, the measurements of the radioactivity were carried out in such a manner as to enhance the sensitivity of the technique, while at the same time making the measurements as specific as possible for that nuclide. In the case of ^{237}U, this involved measuring the 60 KeV γ rays in coincidence with the β^- decay of the nuclide. In the case of the electron capturing ^{203}Pb, the Tl X-rays were measured with a NaI(Tl) crystal operated in coincidence with the 0.28 MeV γ rays as detected by a 42 cm^3 Ge(Li) crystal. The counting rates of the samples were very low (of the order of events pe 1000 min). However, the low backgrounds, the specificity, and the possibility of detecting and identifying any contaminating species made these detection systems very suitable for this program.

IV. Results

The results of our measurements on the uranium and ^{204}Pb contents of Apollo 11 samples (principally soil) are shown in Table 2*. The errors quoted are the statistical errors (1 σ) from the radioactivity measurements. It is evident that the uranium contents of the soil as determined by the ^{238}U(n,2n) reaction in different irradiations agree within the errors. The standard deviation of the mean for sample 10084,58, i.e. 0.54 ppm, is ±0.02 ppm.

There are only two measurements of uranium via the ^{235}U(n,f) reaction to compare directly with those via the ^{238}U(n,2n) reaction. Although in both cases they give slightly lower answers, these are well within even the statistical errors. In view of the larger number of ^{238}U(n,2n) determinations, the average of these is taken to be the best estimate of the uranium content of sample 10084,58.

There are three determinations of the ^{204}Pb in lunar soil presented in Table 2. In view of the possible uncertainties in the determinations of this isotope by other techniques, it is worth presenting some details of the present results. Figures 1 and 2 give the evidence that the lead radioactivity isolated in irradiation L-8 was actually ^{203}Pb ($t_{\frac{1}{2}}$=52 hr). Figure 1 indicates that, in spite of the low counting rates for the X-ray-/γ-coincidence system, an acceptable 52 hr decay was followed. Figure 2 shows that the X-ray spectrum in coincidence with the γ-ray was that expected in

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*See Appendix B for results on two meteorites.

the decay of ^{203}Pb. Although this was the best example, the other two samples behaved adequately in regard to these two tests of radiochemical purity.

On the other hand, the three values for ^{204}Pb in Table 2 scatter appreciably more than can be accounted for by the statistical uncertainties. This may be a reflection of other, less recognizable, errors in the experiment; it may, however, indicate a variability in the ^{204}Pb content even within a given sample.

In addition to the results presented in Table 1, a value of 7.7 ± 1.4 ppb for the thallium content and an upper limit of 5.4 ppb for the bismuth content were obtained on sample 10084,58 (run L-8). A value of about 1.7 ppb was found for the bismuth content of the same sample in run L-7.

IV. Discussion

The average values obtained in this work for the uranium content and the ^{204}Pb content of lunar soil are compared with the more precise of the published experimental values for these abundances in Table 3. The weighted average value found in the present work, 0.54 ± 0.02 ppm, for the uranium content of sample 10084 is seen to be in agreement with the other values, whose unweighted average is 0.56 ppm. The meagre data obtained in this study on the isotopic composition of the uranium are consistent with the terrestrial value as established by other workers (Silver, 1970; Tatsumoto, 1970).

The ^{204}Pb values obtained in this work are also compared in Table 3 with those deduced by other workers, usually after making considerable blank corrections. It is seen that the present investigation tends to confirm the existence in lunar soil of ^{204}Pb at the levels previously reported. Those values lie within the range of the present more direct determinations. Likewise, the bismuth and thallium results presented here agree with the other published work (Ganapathy et al., 1970; Kohman et al., 1970; Anders et al., 1971).

This confirmation of ^{204}Pb in Apollo 11 lunar soil must still be taken with some caution. There is always the possibility that the extremely small amounts detected are the result of contamination of the samples with terrestrial lead before analysis by the various workers. The observed variation of the ^{204}Pb content within the same sample tends to support this possibility.

Appendix A: Some Details of the Chemical Processing of the Irradiated Samples

Because of the rather low expected abundance of ^{204}Pb, with the consequent low production of ^{203}Pb, and the three hour half-life of ^{209}Pb, it was desirable to separate the lead fraction from the irradiated sample as promptly as possible. It was, therefore, necessary to perform the dissolution of the irradiated solid sample and the initial lead separation in a full Hot Cell at the Hot Laboratory of the Department of Applied Sciences at Brookhaven National Laboratory. The sample was usually delivered to the Hot Cell about one hour after removal from the reactor. The sample had an unshielded radiation level of about 30 r hr^{-1} at two feet. The following sections describe the general chemical procedures employed.

1. Dissolution and General Separation of Elements of Interest.

The irradiated powder was quantitatively transferred to a zirconium crucible containing the appropriate carriers (about 10 mg each of Pb, Bi, Tl, and Ba) and tracers (about 200 dpm each of ^{234}U and ^{231}Pa; in one early experiment ^{232}U was used in place of ^{234}U). The sample was then completely fused with 5 g Na_2O_2, 1 g Na_2CO_3, and 1 g NaOH. The cake was transferred to a Teflon beaker and dissolved with distilled water plus nitric acid, as necessary. The solution was adjusted to 20% (by volume) HNO_3, heated to boiling to decompose the excess peroxide, and poured onto a silica gel column (about

200 ml gel volume) in order to collect the Pa fraction. The effluent solution was then electrolyzed, depositing lead and bismuth onto the Pt gauze electrodes. Electrolysis was carried out in three or four separate 10 to 15 minute intervals at 4 to 5 volts and 2 amps. After each electrolysis period, the Pb and Bi deposit was stripped from the electrodes with a 1 + 1 solution of 30% H_2O_2 and dil. HNO_3. The radiochemical purification of the lead and bismuth is described below in Section 2.

Since this electrodeposition of Pb was rarely complete, the residual solution was immediately subjected to the following treatment in order to collect the remaining Pb. About 50 mg Sr carrier was added, the solution was evaporated almost to dryness, and about 100 ml 90% HNO_3 was added to precipitate the Ba, Sr, and Pb nitrates; these were collected by suction filtration onto a fritted disc. The mixed nitrates were dissolved; PbS was precipitated; the lead was combined with that recovered in the electrodeposition and purified as per section 2a. Ba was recovered from the supernate by precipitation as the sulfate and purified if a determination of ^{235}U was to be made via the (n,f) reaction.

The filtrate from this step was then evaporated to small volume and transferred to a small polyethylene bottle for shipment to the University of Chicago Hot Laboratory, where the following chemical treatment was performed:

the solution was adjusted to 1 + 1 (by volume) HNO_3, and placed on a DOWEX-1 x 10 (100-200 mesh; 200 ml resin volume) anion-exchange column. The column was washed with about 200 ml 1 + 1 HNO_3; uranium was eluted with distilled water and was purified as described in Section 2b.

2. Purification of Individual Elements.

a. Lead. The lead fraction was purified by repetition of a cycle which consisted of the following steps:

(i) precipitation from moderately acid solution as PbS;

(ii) dissolution with H_2SO_4 + heat, and precipitation as $PbSO_4$ by evaporation and dilution with distilled water (the supernate from the $PbSO_4$ precipitation contained the Bi fraction, which was frequently purified for the determination of ^{210}Bi from the $^{209}Bi(n,\gamma)$ reaction);

(iii) dissolution with ammonium acetate and heat, and extraction of lead as the dithizonate from a basic buffer solution with cyanide and citrate present into a chloroform solution of dithizone, and back extraction of the lead with 1% (by volume) nitric acid;

(iv) precipitation as the hydroxide with $NH_3(g)$;

(v) dissolution with nitric acid and precipitation as the mixed nitrate with Sr carrier by addition of 90% HNO_3;

(vi) dissolution by distilled water and precipitation of the sulfate (as above under (ii)), and dissolution as above under (iii), and, finally;

(vii) precipitation as the chromate by the addition of $K_2Cr_2O_7$ solution.

After each cycle, the lead chromate was generally dried and weighed to determine chemical yield. If the yield was low after a given cycle, an additional, measured amount of Pb carrier was added in order to enhance yield in the next cycle.

The second cycle generally also included a fusion of the nitrate precipitate with scandium carrier and Na_2O_2, Na_2CO_3 and NaOH (as above) in a Zr crucible, and passage through an SiO_2 column (as above), as well as an iron hydroxide scavenging step from strong NaOH solution. Often, a CuS or a ScF_3 scavenging step were also performed. Ba, Sr, Sc, and Fe holdback carriers were added at appropriate points in each cycle.

Lead was finally collected as the chromate, weighed, and mounted on a Cu disc for radioactivity measurement. Overall chemical yields were $\sim$10%.

The lead monitor was generally collected for radioactivity measurement after the equivalent of one cycle of the chemical procedure described above. When no other monitor was irradiated in the same vial, only the equivalent of one half of the cycle was employed. In any case, the final collection and mounting were performed in the same fashion as employed for the sample.

b. Uranium. Uranium was collected from the column effluent as the hydroxide (with Ce^{+3} carrier) by the addition of $NH_3(g)$. This precipitate was dissolved with HCl and distilled water, about 10 mg $NH_2OH \cdot HCl$ was added and the solution was heated for about one hour, about 5 ml conc. HF was added and the resultant rare earth fluoride precipitate was discarded. The supernate was then evaporated to dryness several times with conc. HNO_3 to drive off the excess HF. Uranyl hydroxide was again precipitated as above (with La^{+3} carrier). This precipitate was dissolved with HCl, $NH_2OH \cdot HCl$ and cupferron were added and the solution was contacted with three 4 ml portions of $CHCl_3$, the organic phase being discarded. Uranium was again precipitated as the hydroxide from the aqueous phase (with La^{+3} as carrier), and this precipitate was taken up with HNO_3. The solution was evaporated to dryness several times with HNO_3. The cake was finally taken up with a saturated solution of NH_4NO_3. Uranium was extracted from the aqueous phase with three portions of diethyl ether. The organic phase was washed with two portions of saturated NH_4NO_3 solution, and the uranium was back-extracted into the aqueous phase with three portions of distilled water.

This final aqueous phase was evaporated and adjusted to a volume of about 10 ml, containing 5 ml 4% (by weight) $(NH_4)_2 C_2O_4$. This solution was transferred to a Teflon cell, and uranium was electrodeposited onto a Pt disc (1 mil thick, 2.4 cm diameter) using the technique of Warren (1967)*.

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*W. G. Warren, LA-1721, 3rd Ed., p. 148 (1967).

Overall chemical yields varied from 1 to 50%.

The chemistry employed for the monitor was generally the same as that for the samples.

Appendix B: Results on the ^{208}Pb and ^{204}Pb Contents of Two Meteorites

In the course of preparation for the lunar sample analyses, results were obtained for the ^{208}Pb and the ^{204}Pb contents of the meteorites Indarch and Bruderheim. The irradiations and chemical procedures were essentially the same as used in the lunar sample analyses. The ^{208}Pb content was established by measuring the ^{209}Pb ($t_{\frac{1}{2}} = 3.3$ hr) β^- activity induced by the (n,γ) reaction during the irradiations.

The results are presented in Table 4. In the case of Indarch, the values found for the lead content agree adequately both in magnitude and in isotopic composition with the results on the same meteorite by Reed et al. (1959). The lead content of Bruderheim found in the present work is appreciably higher than the average in other ordinary chondrites ($\sim$ 90 ppb of ^{208}Pb); however, it is less than twice the amount found in one of them (Holbrook).

Acknowledgement: The authors gratefully acknowledge the cooperation of the Brookhaven National Laboratory in this work in providing the irradiation facilities of the High Flux Beam Reactor, the Hot Laboratory facilities of the Department of Applied Sciences and the laboratory and radioactivity measurement equipment of the Department of Chemistry. This work was supported mostly by NASA contract NAS-9-7883; most of the radioactivity detection equipment employed in this work was purchased under AEC contract AEC AT(11-1) 1167.

Table 1

Nuclear Processes Utilized in Neutron Activation Determination of Heavy Elements

Element	Nuclear Reaction	Half-life	Radiations
uranium	$^{238}\mathrm{U}(n,2n)^{237}\mathrm{U}$	6.75 d	β^-,γ
	$^{235}\mathrm{U}(n,f)^{140}\mathrm{Ba}$	12.8 d	β^-,γ
	$\downarrow$ $^{140}\mathrm{La}$	38.0 hr	β^-,γ
bismuth	$^{209}\mathrm{Bi}(n,\gamma)^{210}\mathrm{Bi}$	5.0 d	β^-
	$\downarrow$ $^{210}\mathrm{Po}$	138 d	α
lead	$^{204}\mathrm{Pb}(n,2n)^{203}\mathrm{Pb}$	52 hr	X, γ
thallium	$^{203}\mathrm{Tl}(n,\gamma)^{204}\mathrm{Tl}$	3.8 y	β^-

Table 2.

Uranium and ^{204}Pb Contents of Apollo 11 Samples

Irradiation	Sample		Uranium Content (in ppm) via		^{204}Pb Content (in ppb)
	Mass (mg)	Number	^{238}U(n, 2n)^{237}U	^{235}U(n, f)^{140}Ba	
L-2	206	10062, 24	-	0.25 ± 0.02	-
	192	10084, 58	-	0.43 ± 0.01	-
L-4	103	10084, 2	0.49 ± 0.06	0.48 ± 0.05	-
	70	10084, 58	0.50 ± 0.07	0.47 ± 0.05	-
L-6	817	10084, 58	0.51 ± 0.04	-	2.2 ± 0.8
L-7	797	10084, 58	0.62 ± 0.07	-	7.1 ± 1.5
L-8	730	10084, 58	0.55 ± 0.02	-	4.7 ± 0.4
Weighted Average		10084, 58	0.54 ± 0.02		

Table 3

Uranium and ^{204}Pb Contents of Apollo 11 Soil

Comparison of different determinations

Sample	Uranium (ppm)	^{204}Pb (ppb)	Reference
10084,58	0.54 ± 0.02	2.2, 4.7, 7.1	This work
10084,75	0.591 ± 0.018		Fields et al. (1970)
10084,25	0.549 ± 0.005	2.16*	Gopalan et al. (1970), Wetherill (1971)
10084	0.544 ± 0.005	1.95*	Tatsumoto (1970)
10084,35	0.562 ± 0.005	3.10*	Silver (1970)
10084,45		5.6,* 5.8,* 6.8*	Huey et al. (1971)

*These values are after correction for laboratory blanks.

Table 4.

The ^{204}Pb and ^{208}Pb Contents of Some Chondritic Meteorites

Meteorite		^{208}Pb (ppb)	^{204}Pb (ppb)	$\frac{208}{204}$	Reference
Type	Sample				
Enstatite	Indarch	850 ± 90	24 ± 1	35 ± 4	This work
		930 ± 30	34.3 ± 0.5	26.8 ± 1.2	Reed et al. (1959)
Ordinary	Bruderheim	378 ± 19	9.3 ± 0.5	41 ± 3	This work
	Forest City Modoc Holbrook Beardsley	20 - 230	0.8 - 5.5	24 - 42	Reed et al. (1959)

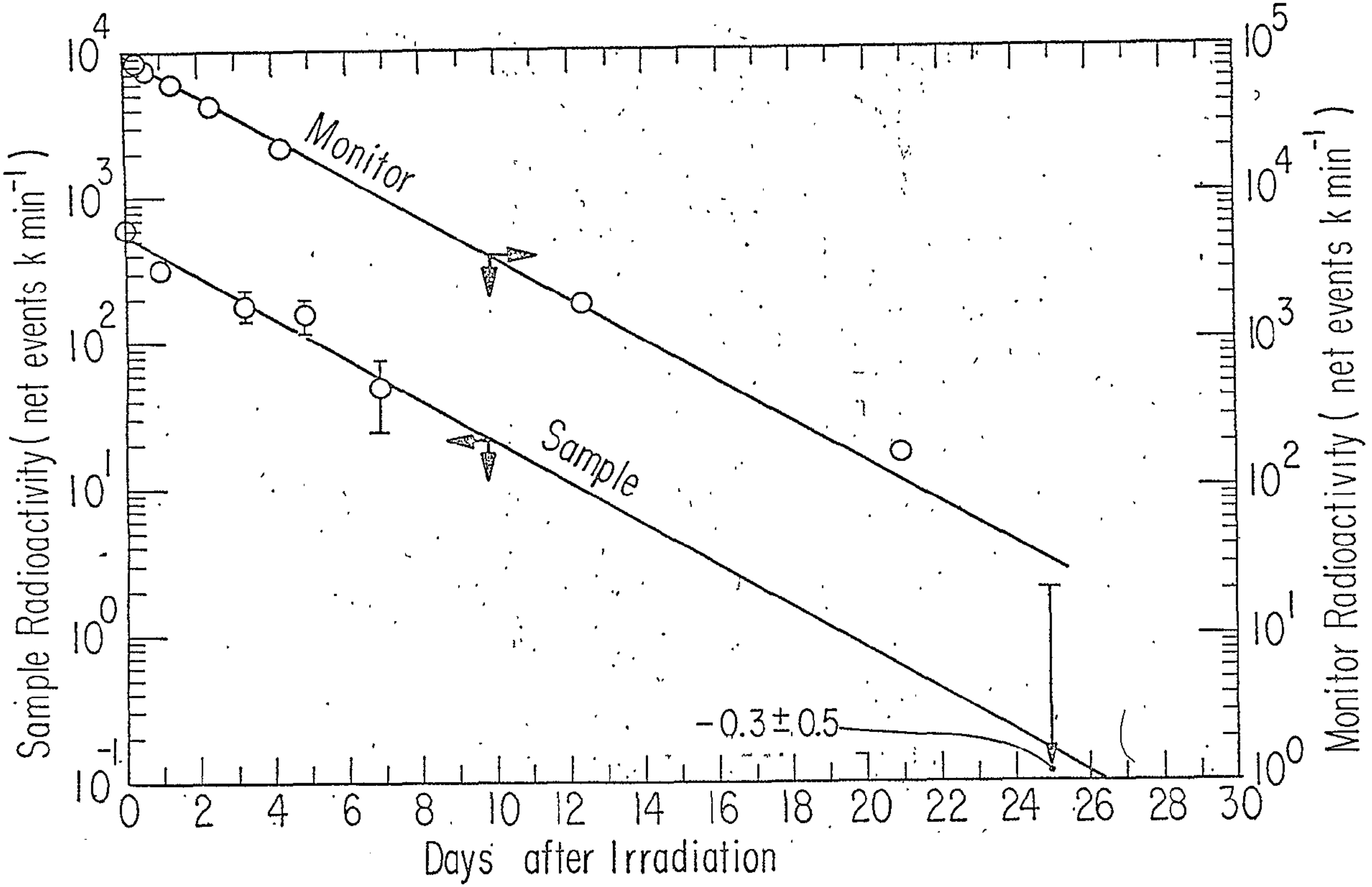
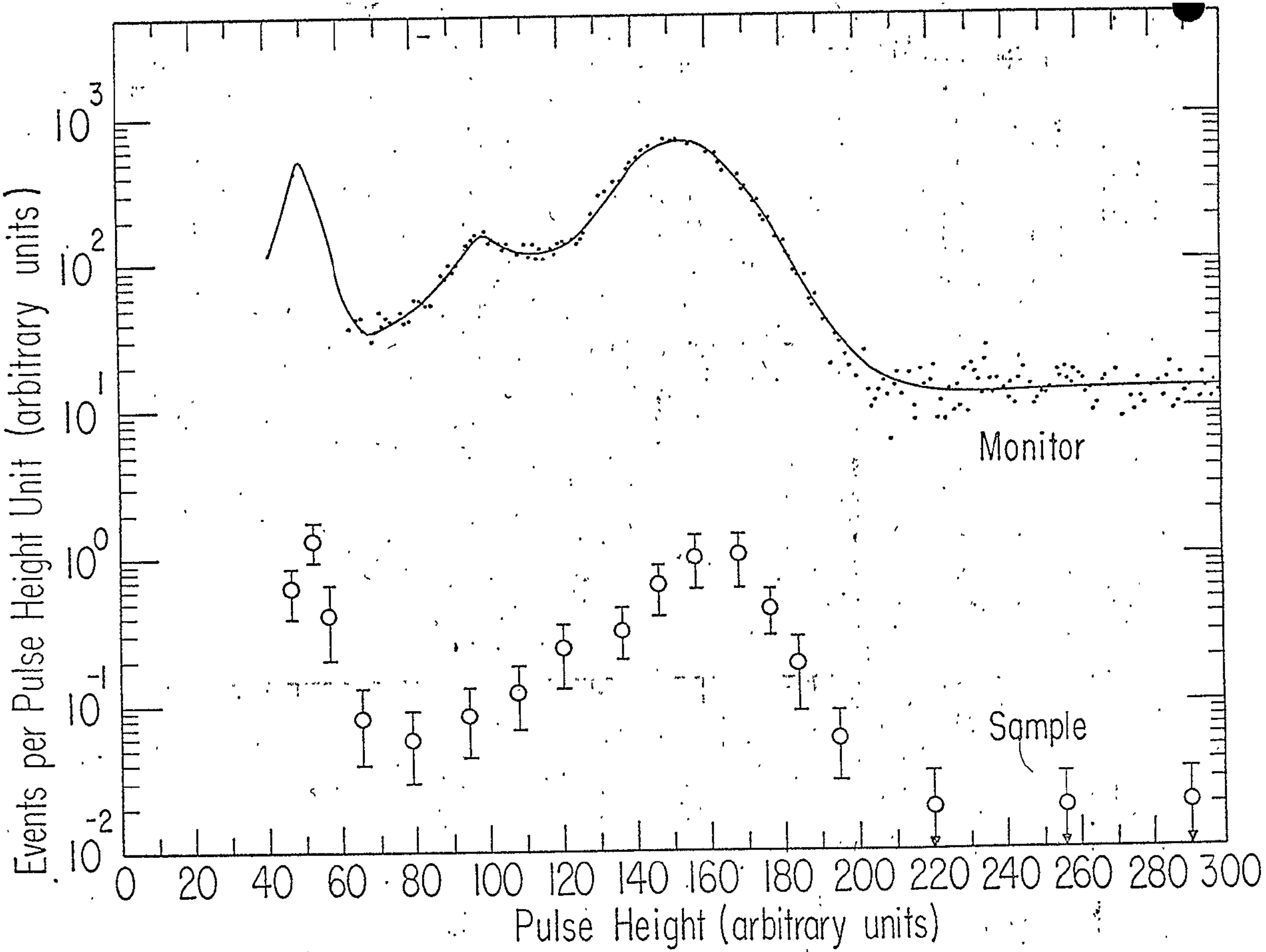

Monitor
Sample
-0.3±0.5
Sample Radioactivity (net events k min^{-1})
Monitor Radioactivity (net events k min^{-1})
10^{4}
10^{3}
10^{2}
10^{1}
10^{0}
10^{-1}
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
10^{0}
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30
Days after Irradiation

Fig. 1



Events per Pulse Height Unit (arbitrary units)
10^3
10^2
10^1
10^0
10^{-1}
10^{-2}
Monitor
Sample
0
20
40
60
80
100
120
140
160
180
200
220
240
260
280
300
Pulse Height (arbitrary units)

Fig. 2 -

FIGURE CAPTIONS

Figure 1. Decay of ^{203}Pb ($t_{\frac{1}{2}}$ = 52 hr) radioactivity in lead separated from neutron irradiated lunar fines (Run L-8). The radiation measured is the (73 ± 15)-keV X-ray in coincidence with (278 ± 20)-keV γ ray. A background rate of (2.3 ± 0.3) events kmin^{-1} has been subtracted. The lunar sample (lower curve, left hand ordinate) is seen to decay with the same half-life as the monitor lead (upper curve, right hand ordinate).

Figure 2. ^{203}Pb in lead separated from neutron irradiated lunar fines (Run L-8). The spectrum is that of X-rays in coincidence with (278 ± 20)-keV γ rays. The abscissae are proportional to the energy of the X-rays. The top spectrum is that of the lead monitor; the lower spectrum is that of the sample.

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α Activity
SC/3 Camera

Examination of returned Surveyor 3 camera visor for alpha radioactivity

N71 24561

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Abstract - The Surveyor 3 camera visor brought back by the Apollo 12 astronauts has been examined for an alpha radioactive deposit formed by the decay of radon isotopes diffusing out of the lunar surface. An upper limit for the equilibrium amount of Po^{210} at Oceanus Procellarum has been set at less than 0.005 dis cm^{-2} sec^{-1}. This number is appreciably lower than the amount of Po^{210} observed by Surveyor 5 at Mare Tranquillitatis.

INTRODUCTION

On April 19, 1967, Surveyor 3 landed in the eastern part of Oceanus Procellarum at 23.34° W. longitude and 2.99° S. latitude (ACIC coordinate system). On November 20, 1970, the same site was revisited by Apollo 12 astronauts Alan Beam and Charles Conrad who brought back the Surveyor 3 television camera together with 32 kilograms of moon rocks. Part of this camera, the visor, was made available to the University of Chicago to examine for the presence of a deposit of alpha radioactivity.

The possibility of such a radioactive deposit on the surface of the moon was suggested by several authors (e.g. KRANER et al., 1966). Radon isotopes formed by the decay of uranium and thorium, diffuse out of lunar material into space where they undergo further decay and some of their daughters end up on the lunar surface. In the thorium decay series the daughters have relatively short half-lifes, and all had decayed before the visor could be examined. On the other hand, the alpha emitting Po^{210} in the uranium decay series is held up by the 22-year half-life of its grandparent Pb^{210}. A measurement of the amount of Po^{210} (5.31 MeV) alpha activity on the visor, together with knowledge of the time spent on the moon, and on the earth before the measurement, provides a measure of the rate of radon decay product deposition on the lunar surface at Oceanus Procellarum.

The existence of such deposit would be interesting in providing information on the emanating power of lunar material and on the amount of radon "atmosphere" on the moon. In addition, it may have an effect on the isotopic composition of the lead in lunar fines.

EXPERIMENTAL METHOD AND RESULTS

Measurements on the Surveyor 3 camera visor (Fig. 1) were started at the University of Chicago 0.64 years after it was taken off the Surveyor 3 spacecraft. The visor was placed in a vacuum chamber and examined for alpha radioactivity with the Alpha Scattering Instrument (TURKEVICH et al., 1966). In order to increase the sensitivity, the proton system of the instrument was used. The active area of the proton detectors of this instrument is about ten times that of the alpha detectors and, in addition, the examined visor could be placed closer to the proton than to the alpha detectors. The gold foils in front of proton detectors, which normally screen out the alpha particles in the Alpha Scattering Instrument were removed for these measurements and replaced by the VYNS films. The visor was measured for a period of 9714 minutes using all 4 detectors and an additional period of 4475 minutes with less than the full complement of detectors in order to check on possible asymmetries in the deposit. The backgrounds in the instrument were negligible.

Fig. 2a shows the experimental data obtained from the visor of the Surveyor 3 television camera. There are several unexpected surprises that characterize these data:

(1) The continuous flat spectrum indicates that the source of alpha activity is not on the surface.

(2) The intensity is too high - several orders of magnitude higher than was expected.

(3) The presence of high energy alpha particles (higher than 6 MeV) indicates that the source is probably daughter products of Th^{228} or U^{234}.

The surface of the visor, as most of the Surveyor parts, was covered with white paint for thermal control purposes. Because of the unavailability of a model television camera we were not able to measure directly the natural background from the visor. Fig. 2b shows the results of measurements made on plates covered with the same paint and made at the same time as the visor itself. In these measurements, the plates were placed very close to each detector in a position where the absolute efficiency of detecting their activity could be calculated.

This spectrum is very similar to that obtained from the visor. After comparing the absolute intensities, the conclusion was reached that the gross activity on the visor returned from the moon was entirely due to the activity of the paint.

Although the presence of this alpha activity of the paint reduced the sensitivity of identifying an alpha radioactive deposit, the data can still be used to set upper limits for the Po^{210} radioactivity on the moon. Fig. 3 shows a comparison of the alpha activity in the region of Po^{210} (5.3 MeV) for the paint and the visor. Using the gross alpha radioactivity as a measure of the relative efficiencies of detecting radiation from the visor and paint, the paint background could be subtracted from the visor data in the region of interest to get a net activity on the visor of $(0.7 \pm 3.5) \times 10^{-3}$ d min^{-1} cm^{-2}.

In order to estimate the amount of Po^{210} activity to be expected on a square centimeter of the lunar surface after a very long period, this net activity has to be corrected for the shadowing of the visor by assorted spacecraft parts (the "view factor" to space was 0.65), for the decay since removal from the moon (0.64 years) and for the fact that the visor was on the moon for only 2.58 years (keeping in mind the genetic relationship of the Rn^{222} decay chain). Application of these corrections leads to the measurement implying an activity, after infinite time, of

$$D = (0.88 \pm 4.43) \times 10^{-3} \text{ d sec}^{-1} \text{ cm}^{-2}$$

on the lunar surface at Oceanus Procellarum.

DISCUSSION

The limit on the Po^{210} alpha radioactivities obtained in this work is compared with the predictions of KRANER et al. (1966) and of subsequent attempts to measure this quantity in Table 1. The original prediction was an average for the whole moon, as was the limit set by YEH and VAN ALLEN (1969) from Explorer 35 observations. The only reported observation of the presence of the radioactivities is the work of TURKEVICH et al. (1970) from the data obtained by the Alpha Scattering Instrument on the Surveyor 5 mission to Mare Tranquillitatis.

Recently, LINDSTROM et al.(1970) determining the excess of Pb^{210} (over that in equilibrium with uranium) on the surface of the rock brought back by Apollo 11 astronauts, also from Mare Tranquillitatis, gave a limit which is 70 times lower than the value reported by TURKEVICH et al.(1970).

The limit set by the present work on such radioactivity in Oceanus Procellarum, a different site, but one at which the uranium content of the soil is actually appreciably higher than in Mare Tranquillitatis, is also lower than the reported value of TURKEVICH et al.

Although the results of LINDSTROM et al.and the present work appear to contradict the results reported by TURKEVICH et al., it must be remembered that the radioactive deposit should be confined to the topmost fraction of micron of the lunar surface. Any disturbance of the surface, such as the shaking off of a dust layer, or abrasion of the surface, would carry away the deposit also, Thus, although these two most recent attempts to detect the alpha radioactivities have failed,and therefore contradict the observation of TURKEVICH et al., there is some probability that they are not valid checks on the existence of the deposit. In both cases there is no assurance that the topmost layer was not removed. It may be that the Surveyor 5 mission, making an in situ measurement, was better able to detect this fragile deposit than were the examinations of samples brought back from the moon.

Stoenner et al. (1971), in examining the gas in the sealed sample return container (SRC) from the Apollo 12 mission, found evidence for about 5 d min^{-1} of Rn^{222} in the container atmosphere, although the confidence in their number is not large because of a high radon blank from the charcoal in their system. If it is assumed that 5.d min^{-1} of Rn^{222} represents roughly the equilibrium amount that would diffuse out of the 2.8 kg of lunar fines in the particular container, it would imply that 10^{-3} of the Rn^{222} diffuses out of lunar soil particles. Assuming that the same efficiency is applicable on the moon, the limit set for the Po^{210} deposit by the present work at Oceanus Procellarum means that radon diffuses out of less than about one meter of lunar soil on the moon. If the true equilibrium amount of Po^{210} is higher than found in the present work, the parent radon would be derived from greater depths.

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Table 1

Equilibrium Po^{210} alpha radioactivity of lunar surface

	$dis\ cm^{-2} sec^{-1}$	
1966 Kraner et al.	2.0*	whole moon
1969 Yeh and Van Allen	<0.16	whole moon
1970 Turkevich et al.	0.03 ± 0.01	Mare Tranquillitatis
1970 Lindstrom et al.	<0.0004	Mare Tranquillitatis
1971 Present work	<0.005	Oceanus Procellarum

*Predictions

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Figure Captions

Fig. 1. The visor of Surveyor 3 television camera brought back by Apollo 12 astronauts.

Fig. 2a. The alpha spectrum obtained from the Surveyor 3 television camera visor as measured by the Alpha Scattering Instrument.

Fig. 2b. Background obtained from the paint used on visor.

Fig. 3. Comparison of α-radioactivity from the visor and the paint in the vicinity of Po^{210} (5.3 MeV).

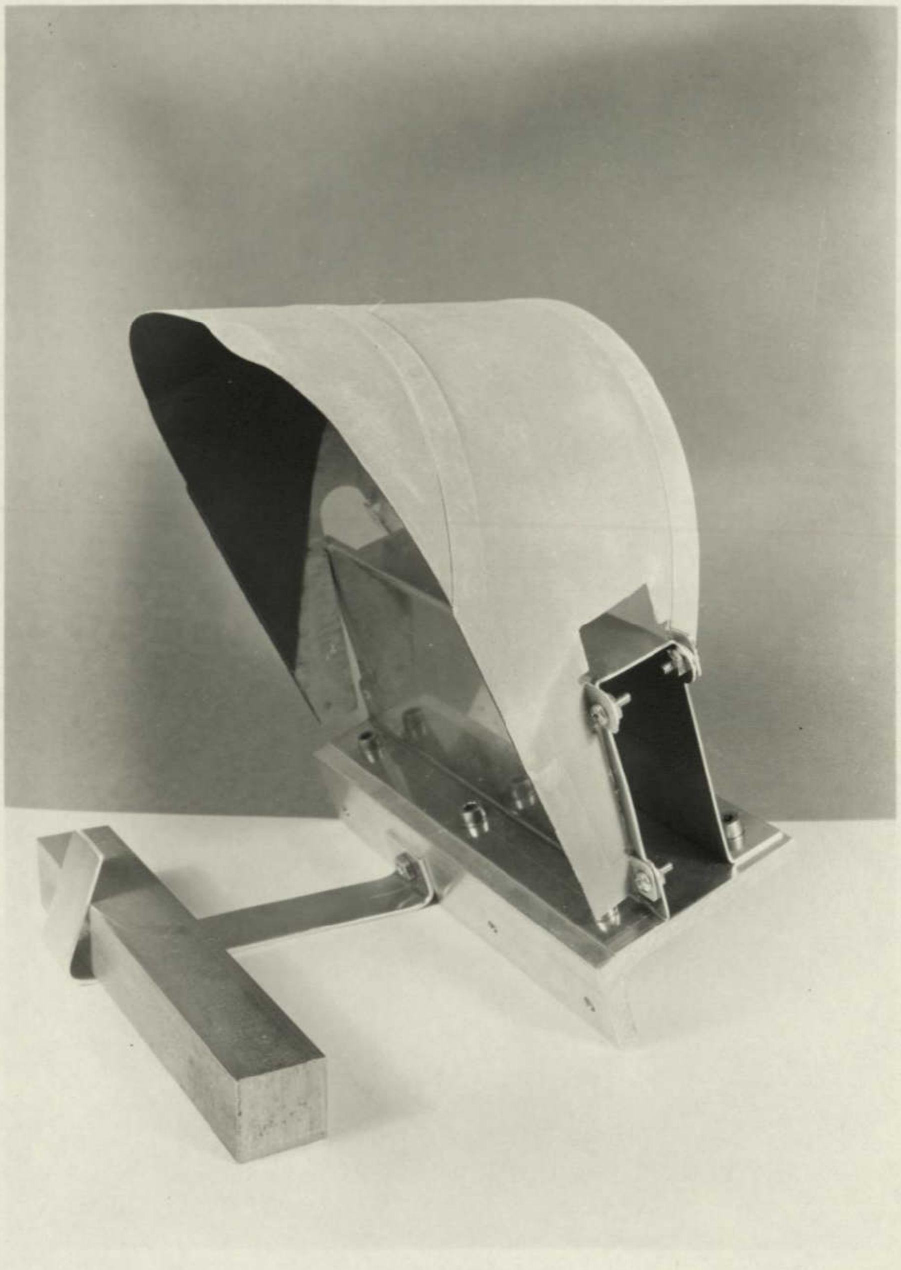
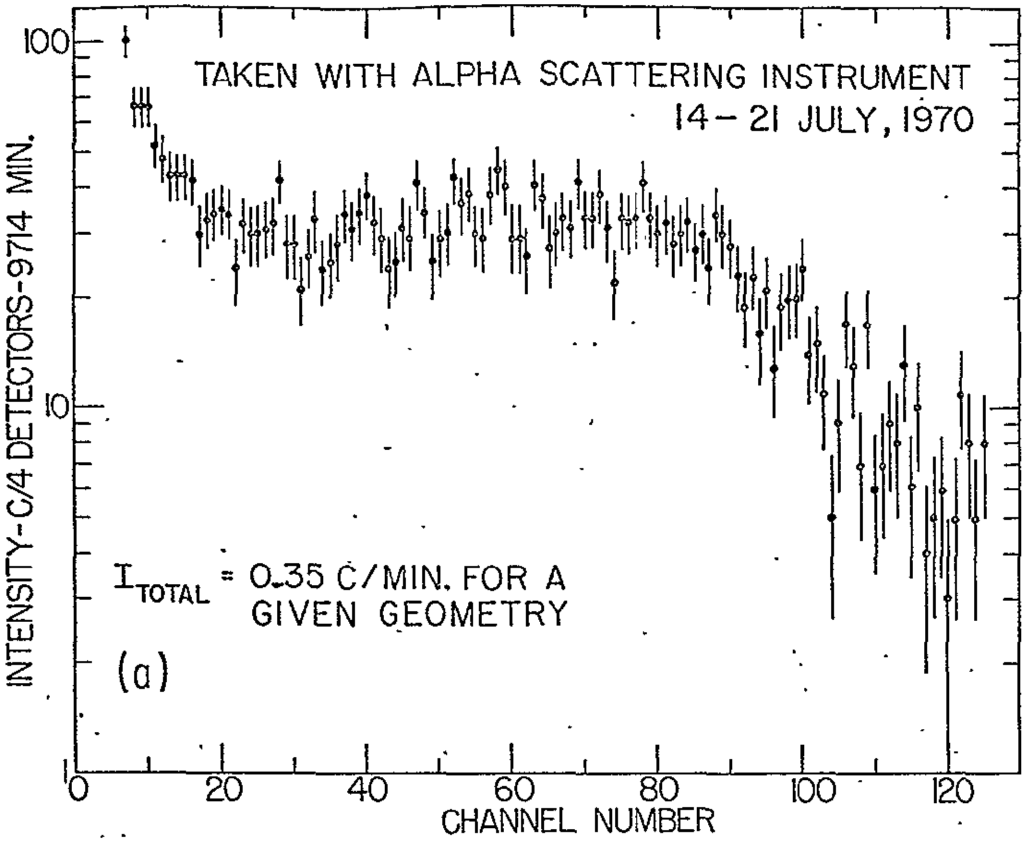


Figure 1

ALPHA SPECTRUM FROM THE SURVEYOR 3 T.V. CAMERA VISOR



ALPHA BACKGROUND FROM THE PAINT

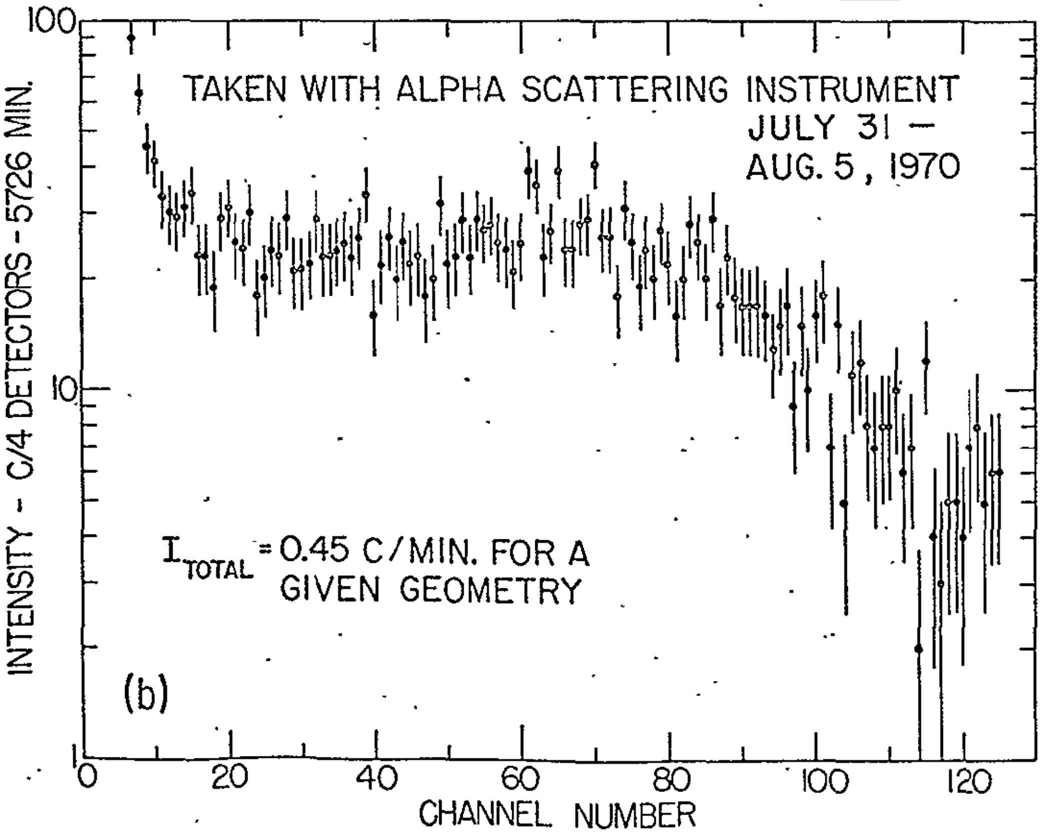


Fig. 2

COMPARISON OF α-RADIOACTIVITY FROM THE VISOR AND THE PAINT IN THE VICINITY OF Po^{210} (5.3 MeV)

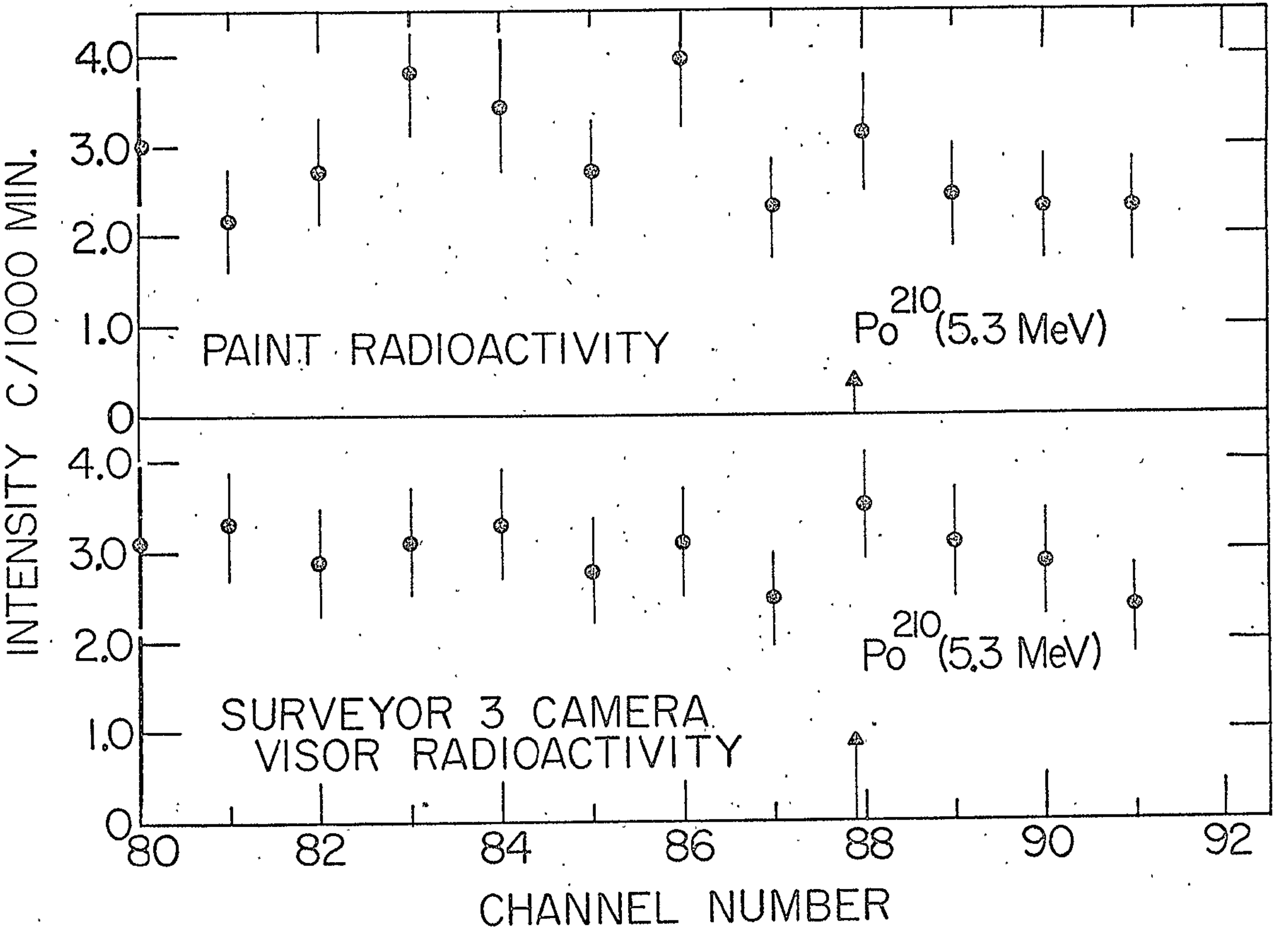


Fig. 3