the rocks and the regolithic material indicate an extended history of rock-forming events for the moon which may be studied with profit by these techniques, particularly when coupled with systematic geologic documentation of sample sources. The question posed by the discrepancy between the apparent ages of the rock fragments and the associated lunar dust and breccia raises the possibility that fundamental and unexpected rock-forming processes exist.

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8. Essential to this work was the dedicated laboratory participation and cheer of Geraldine Baenteli and Dolly Pearson. E. V. Nenow and C. Bauman provided important technical support. E. Brown wrestled untiringly with NASA, which supported the major part of this research under contract NAS 9-7963. California Institute of Technology Contribution No. 1707.

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Rubidium-Strontium, Uranium, and Thorium-Lead Dating of Lunar Material

Abstract. *Rubidium and strontium concentrations and strontium isotopic compositions have been measured on whole rock samples and density fractions of microgabbro. Density fractions on two rocks define isochrons of 3400 and 4500 million years with large uncertainties owing to low enrichment of radiogenic strontium. Lead from fine surface material is highly radiogenic. An age of 4750 million years has been calculated from the ratio of $^{207}Pb/^{206}Pb$. The concentrations of uranium, thorium, and lead isotopes are consistent with the evolution of lead in a 4700-million-year-old closed system characterized by the ratios of uranium to lead and of thorium to lead in this surface material.*

A time scale established by dating significant events in lunar history will permit one to relate lunar history to that of the earth and solar system and will place absolute values on relative chronologies established by photogeology. The first step in this program is to assess the usefulness of available lunar materials for dating purposes and to establish the approximate range of ages with which we will be dealing in the dating of lunar rocks.

Initial Rb-Sr dating has been carried out on samples of crystalline rock, in the belief that the interpretation of results on such rocks should be more straightforward than that on the microbreccia samples. The concentrations of K, Rb, and Sr found by mass spectrometric isotope dilution analysis of five such rocks, as well as in a sample of fine lunar surface material, are given at the top of Table 1. The data are plotted in Fig. 1. These rocks are characterized by low Rb/Sr ratios, resulting in small (~1 percent) enrichments in radiogenic strontium-87.

These values of $^{87}Sr/^{86}Sr$, not greatly enriched over the primordial values found from meteorite studies, indicate that, like the earth, the source of these lunar rocks acquired a Rb/Sr ratio much lower than the chondritic or solar value very early in the history of the solar system.

These small enrichments place constraints on the interpretation of the Rb-Sr data in terms of ages. Consequently, any such interpretations given herein must be regarded as preliminary. Improvements in our techniques for measuring Sr isotope ratios, such as conversion to digital recording, are currently being made, and more definitive results will soon be obtained by remeasurement of these same Sr samples.

Rb-Sr measurements define the time at which a Rb-Sr fractionation took place in the chemical system being studied. In the case of rock systems, this requires measurement of at least two samples with differing Rb-Sr ratios, which had the same Sr isotopic composition at the time of the event being dated. This is the case for cogenetic materials in which the Sr isotopic composition was initially homogeneous. For whole-rock dating, it is therefore preferable that independent evidence exists for the cogenesis of the rocks, in order that the entire burden of the argument for cogenesis does not rest upon the colinearity of the data when plotted on a Sr evolution diagram. This independent evidence is lacking in the case of these lunar rocks. Therefore, our initial effort has been directed to the study of the "internal ages" of single rock samples, obtaining varying Rb-Sr ratios by separation of density fractions of finely ground (< 88 μm) rock. These are not pure mineral separates, but have the advantage that an insignificant quantity of material is rejected in the process of separation.

The separation procedures were described in work on density fractions from enstatite and shocked hypersthene chondrites (*1*). The chemical procedures are identical to those employed in our published work on meteorites (*2*). As in this work, K, Rb, and Sr blanks are ~50, 0.1, and 1 ng, respectively, and they represent a negligible contribution to our results.

The most radiogenic fraction from two samples of microgabbro (Table 1) has a density of ~3.3 g/cm³ and probably represents a pyroxene concentrate. The least radiogenic fraction has a density of less than 2.96 g/cm³ and was found to consist almost entirely of plagioclase. The $^{87}Rb/^{86}Sr$ ratios range over a factor of about three, and with

Table 1. The Rb and Sr analytical results.

Sample	K (μg/g)	Rb (μg/g)	Sr (μg/g)	$^{87}Sr/^{86}Sr$ (atomic)
84,25 (fines)	1100	2.83	164.7	0.7017
72,38	2539	5.72	168.8	0.7043
24,24	2814	6.20	183.9	0.7044
17,41	2500	5.80	174.2	0.7044
50,30	665	0.788	188.9	0.7002
22,45		5.66	165.5	0.7047
17,41 fractions:				
$\rho<2.96$		9.52	548.3	0.7022
$2.96 <\rho<3.32$ (coarse)	4090	9.19	257.3	0.7047
$3.32 <\rho$ (coarse)	1545	4.01	77.9	0.7070
$3.15 <\rho<3.25$ (fine)		7.79	134.0	0.7080
$3.25 <\rho$ (fine)	843	2.30	45.8	0.7065
24,24 fractions:				
$\rho<2.96$		14.27	598.2	0.7026
$2.96 <\rho<3.15$		19.42	520.0	0.7050
$3.15 <\rho<3.32$		15.43	286.2	0.7081
$3.32 <\rho$		2.82	47.7	0.7083

improvements in measurement of ^{87}Sr/^{86}Sr ratios should permit age measurements of reasonable quality.

The data from these density fractions seem to define linear arrays with slopes corresponding to ages of 3400 ± 600 and 4050 ± 700 million years, respectively ($\lambda = 1.39 \times 10^{-11}$ year^{-1}) (Fig. 2). The errors represent the extreme isochrons which can be drawn through the error brackets of the individual points. As stated previously, identification of these results in terms of ages is tentative, pending improvements in the measurement of the ^{87}Sr/^{86}Sr ratios. The error brackets represent mean deviations of sets of at least ten ^{87}Sr/^{86}Sr ratios, normalized to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. A limited amount of work done with isotope ratio standards indicates that we can resolve differences in ^{87}Sr/^{86}Sr ratios of 0.0005. Absolute values could have somewhat larger systematic errors, pending more complete study of the linearity of our recording system in this range of 1 per mil (*3*).

Uranium, Th-Pb measurements have been carried out on fine lunar surface material (sample 84,25). Isotope dilution measurements give a concentration of uranium of 0.549 μg/g and a concentration of thorium of 2.08 μg/g. Blank corrections are less than 1 percent. Two isotope dilution lead measurements have been made, the first with ^{208}Pb tracer, and the second with ^{206}Pb tracer. This permits calculation of the abundance and concentration of all the Pb isotopes, after subtraction of the contribution from the tracers. In this way it was found that the lead in the lunar surface was highly radiogenic. Uncorrected for the blank, the ratio ^{206}Pb/^{204}Pb was 108 and the ratio ^{207}Pb/^{204}Pb was 76.5. After correction for a total lead blank of about 0.2 μg, the ratios become 231 and 161, respectively. These ratios may well be still higher, since the residual ^{204}Pb may represent terrestrial contamination beyond that introduced by the blank. Assuming the small amount of nonradiogenic lead remaining after subtraction of the blank to be of primordial composition (*4*), a ^{207}Pb/^{235}U age of 4720 ± 100 million years and a ^{206}Pb/^{238}U age of 4655 ± 100 million years can be calculated. The age calculated from the ^{207}Pb/^{206}Pb ratio is 4750 ± 50 million years. The data from the analysis with ^{206}Pb tracer were of poor quality. The ^{208}Pb/^{232}Th age calculated from these data is 4720 million years, but this value may be in error as much as 10 percent. Of course, these numbers cannot be interpreted as the age of any particular rock. The fine surface material is probably a sample of many different rocks, possibly from a large area of the lunar surface. However the ^{207}Pb/^{206}Pb age can be interpreted as the time of the major U-Pb fractionation which took place at least in the surficial regions of the moon, and also as an "age of the moon," analogous to Houterman's (*5*) age of the earth and subject to similar assumptions. The U-Pb ages indicate that Houterman's closed system model is characterized by a ^{238}U/^{204}Pb ratio essentially that of the present surface material.

These Pb results indicate that the lunar surface material has a very high ratio of ^{238}U/^{204}Pb (>210) in contrast with the terrestrial value of 9 for this ratio. Furthermore, the ^{207}Pb/^{206}Pb ratio indicates that this enrichment of uranium with respect to lead took place very early in the history of the solar system. If this very old and highly radiogenic lead proves to be generally characteristic of lunar material, this result argues against a terrestrial origin for the moon at any time significantly later than the time of

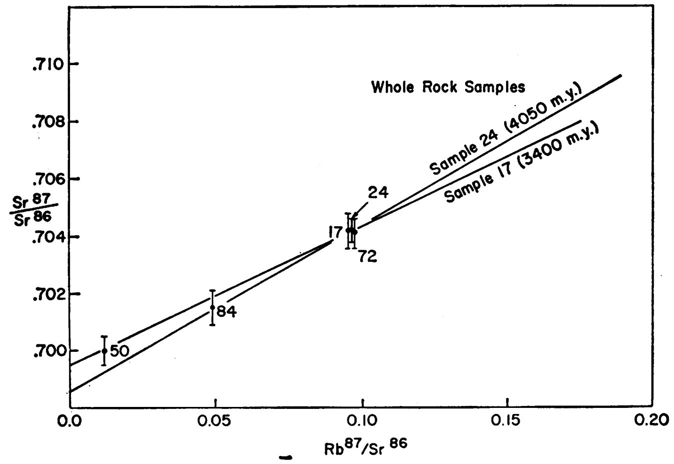
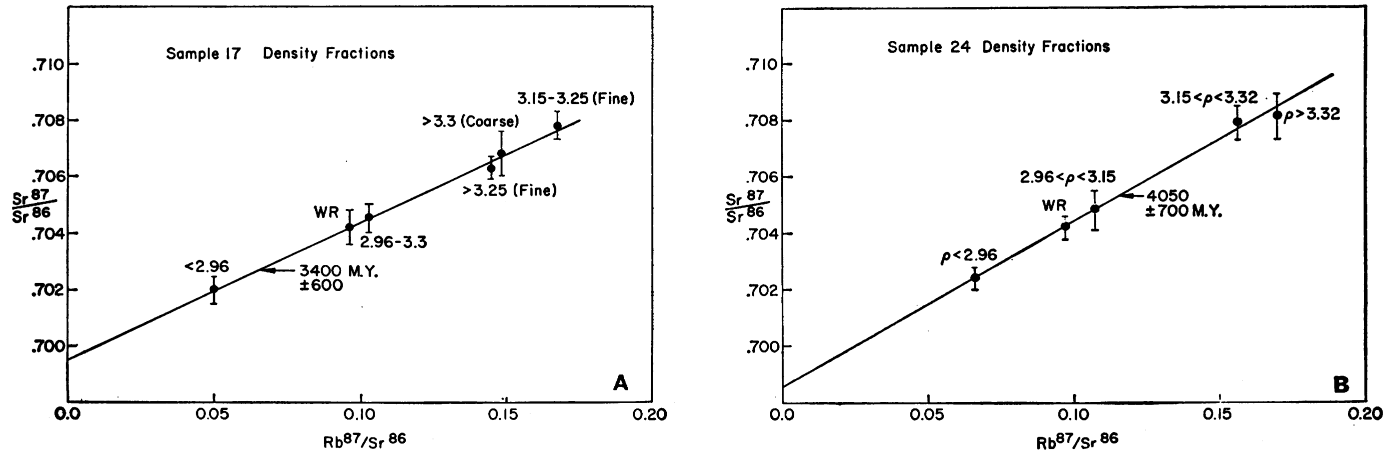


Fig. 1 (top left). Strontium evolution in whole rock samples. the lines drawn are the 3400- and 4050-million-year isochrons fitted to the data obtained from the density fractions. Fig. 2 (below). (A) Strontium evolution for density fractions from sample 17,41. (B) Strontium evolution for density fractions from sample 24,24.



formation of the earth. Also, since Tilton has shown that tektites contain lead resembling closely the isotopic composition of modern terrestrial lead (*6*), a lunar origin of these objects can be ruled out unless some lead much more like that found on earth is found elsewhere on the moon.

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* Deceased 2 December 1969.

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Rubidium-Strontium Relations in Tranquillity Base Samples

Abstract. *Preliminary total rock analyses disclosed a greatly different rubidium depletion between two groups of these igneous rocks, and ratios of strontium-87 to strontium-86 indicate that the rubidium depletion in these materials must have occurred during or shortly after the accretion of the terrestrial planets.*

Five rock samples weighing 0.82 g (No. 22.43), 1.27 g (No. 20.28), 1.14 g (No. 47.25), 2.36 g (No. 72.25), and 1.2 g (No. 58.36) and a 5.16-g sample of fines from the lunar soil (No. 84.39) were available for analysis. This progress report gives analyses on total rock samples only; work on separated minerals and the fined-grained soil material has not been completed. Rubidium, strontium, and the isotopic composition of Sr were determined by mass spectrometric techniques in separate whole pieces of rock ranging upward in size from a few tens of milligrams. These samples were not necessarily assumed to be representative of the rock chips selected by the Lunar Receiving Laboratory, but, by relating the isotopic data from each sample separately and using different whole fragments in repeated analyses, we hoped to obtain additional information on the homogeneity of the rocks, test the assumption of a lack of short range migration of components, and derive meaningful age evaluations and average compositions.

The sample fragments were gently broken in a boron carbide mortar, weighed, and digested in a Parr Industries Teflon-lined bomb at 120° to 140°C. Before acid digestion the samples were spiked with ^{87}Rb. Minimum amounts of reagents were used: 10 ml of HCl carrying the spike, 3 ml HF and 1 ml $HClO_4$ per 0.15 g of sample were the approximate amounts. The solution was washed into a Pt dish, covered with a Teflon lid, and heated with a hot plate below and a heat lamp above. After achieving total solution (no visible mineral or chemical residues) the solution was evaporated to get rid of excess HF and SiF_4 and diluted with 0.5*N* HCl to 50 ml in a volumetric flask, and aliquots were taken for Sr spiking and for Rb and Sr isotope dilution and Sr isotope ratio analysis. Element separations were achieved on cation exchange columns using Dowex 50, 8 percent cross-linked, 350 mesh.

Table 1. Preliminary contamination and spike analyses.

Reagent	Rubidium (10^{-9} g/ml)	Strontium (10^{-9} g/ml)
H_2O	0.003	
HCl	0.005	0.05
HNO_3	0.009	
HF	0.05	
$HClO_4$	0.15	
Total blank	1.5	
Comparison of normal solutions		
CIT Sr	14.61 × 10^{-9} mole ^{88}Sr/g	
MIT Sr	14.70 × 10^{-9} mole ^{88}Sr/g	
CIT Rb	0.440 × 10^{-9} mole ^{87}Rb/g	
MIT Rb	0.449 × 10^{-9} mole ^{87}Rb/g	
Fused silica	Rb, 10^{-9} g/g	Sr, 10^{-9} g/g
C-17-A	<9	<190
F-19-A	<3	<160

The mass spectrometer used for this work consisted of a 12 inch radius, 60° sector analyzer section (Consolidated Electrodynamics Corporation) with detachable source, in-line valve, and cold finger in source housing using single Ta filament surface ionization, Faraday cup collector, Cary vibrating reed electrometer, and W. Shields design expanded scale system of recording residual voltages (after 70 to 90 percent precise nullification) on a modified Leeds-Northrop Speedomax G recorder. All scale settings on vibrating reed electrometer and expanded scale system were kept the same for all runs, including the calibration and standardization analyses, so that instrumental fixed bias was removed.

In analyses of Sr isotopic ratios the internal consistency of repeated ratio measurements in a single analysis gave a standard deviation of the mean of less than 0.01 percent. Allowing for possible contamination, we believe that the overall error will not exceed a standard deviation of 0.00015.

Mass spectrometer errors in analyses of isotope dilution are generally not more than 1 percent (standard deviation) for Sr and 2 percent for Rb, but in these cases of low Rb content the error due to contamination may outweigh the measurement error. Repeated analyses on separate small rock fragments show also the inhomogeneity of the rock, so that overall errors determined on this basis would reflect this added factor. Because the Rb, Sr, and Sr isotope analyses are performed on each piece separately, the inhomogeneity does not affect the age relationship. The uncertainty in the error at this stage of the investigation has resulted in an assignment of a larger error than is probably necessary.

Analyses of the California Institute of Technology (CIT) Rb and Sr normal solutions are given in Table 1, relative

Table 2. Tranquillity Base samples : total rock fragments.

Sample	Rubidium (10^{-6} g/g)	Strontium (10^{-6} g/g)	$^{87}Rb/^{86}Sr$ Atomic ratio	$^{87}Sr/^{86}Sr$ Normalized*
22.43	6.0†	174†	0.099	0.7051‡
72.25	5.4	168	0.093	0.7051
20.28	0.77	152	0.0146	0.7000
58.36	0.62	173	0.0103	0.6998
47.24	1.18	189	0.0181	0.6996
Sargasso Sea water				0.7090

* Normalized to $^{86}Sr/^{88}Sr = 0.1194$. † Preliminary values only; errors not yet established by replicate analyses to ascertain inhomogeneity between fragments. ‡ Error in replicate ratio measurements in single analysis $\sigma_m = 0.0001$.