Gas-Retention and Cosmic-Ray Exposure Ages of Lunar Rock 15555

Abstract. *The last lava flow in the Hadley Rille area of Mare Imbrium, as inferred from an argon-40–argon-39 experiment on a plagioclase separate from the lunar basalt 15555, occurred $3.31 \pm 0.03 \times 10^9$ years ago. An argon-40–argon-39 experiment on a whole rock sample shows significant loss of radiogenic argon-40 and yields a well-defined, high-temperature plateau indicating a lower age of $3.22 \pm 0.03 \times 10^9$ years. A cosmic-ray exposure age of $90 \pm 10 \times 10^6$ years is determined from the ratio of spallogenic argon-38 to calcium.*

Lunar rock 15555 is a large rock from near the rim of Hadley Rille. It is presumed to be a fragment of the local bedrock representing the final lava flow that flooded the Hadley Rille area of Mare Imbrium. We present here our conclusions regarding the age of formation and exposure to cosmic rays of rock 15555, based on isotopic dilution K-Ar and ^{40}Ar-^{39}Ar methods.

An age for 15555 was obtained by measurements of Ar and K in plagioclase separate of high purity. Plagioclase generally shows less Ar loss than whole rock samples of lunar materials and provides stronger lower limits on the time of formation (*1, 2*). The concentrations determined mass spectrometrically by isotope dilution (*1*) are $1.17 \pm 0.05 \times 10^{-5}$ cm^3/g (STP) for ^{40}Ar and 323 ± 2 ppm for K; these give a gas-retention age of $3.32 \pm 0.05 \times 10^9$ years. The errors include uncertainties in the system blanks, mass spectrometry, and amounts of ^{38}Ar tracer.

Samples of whole rock and plagioclase and pyroxene separates of high purity were examined by the ^{40}Ar-^{39}Ar method. Except for minor differences (*3*) the procedures for neutron irradiation, gas extraction, mass spectrometry, and data correction were the same as those detailed by Turner *et al.* [see (*1*)]. The results are given in Table 1.

The ^{40}Ar-^{39}Ar age of an unknown sample is computed by

$$\frac{e^{\lambda t}-1}{e^{\lambda t_o}-1} = \frac{f_o\,(^{40}Ar^*/^{39}Ar^*)}{f\,(^{40}Ar^*/^{39}Ar^*)_o} \qquad (1)$$

where t is the age, λ is the decay constant, f is the neutron fluence, and an asterisk designates data corrected for all contributions other than radiogenic ^{40}Ar or $^{39}K(n,p)^{39}Ar$. Nonsubscript variables denote the sample, and the subscript o refers to a monitor of independently measured $^{40}Ar^*/K$. The monitor is used only to establish the ratio of $^{39}Ar^*$ to ^{39}K; its actual age is irrelevant, and its appearance in Eq. 1 is only through the definition

$$\left(\frac{R}{R+1}\right)\left(e^{\lambda t_o}-1\right) \equiv \left(\frac{^{40}Ar^*}{^{40}K}\right)_o = \left(\frac{^{40}Ar^*}{^{39}Ar^*}\right)_o\left(\frac{^{39}Ar^*}{^{39}K}\right)\left(\frac{^{39}K}{K}\right)\left(\frac{K}{^{40}K}\right) \qquad (2)$$

where R is the ratio of electron capture to $\beta-$ decay in ^{40}K. The accuracy of the absolute age determination of the sample is dependent on the accuracy to which $^{40}Ar^*/K$ in the monitor is measured. This case must be distinguished from experiments in which the use of Eq. 2 is inverted and the $^{40}Ar^*/K$ ratio of the monitor is inferred from an age determined by other means that must be presumed identical to an argon-retention age. For this study the monitor is a hornblende (hb3gr) from the Llano uplift (*4*) for which $^{40}Ar/K$ is $5.69 \pm 0.09 \times 10^{-3}$ cubic centimeters (at STP) of ^{40}Ar per gram of K (*1*) ($t_o = 1.062 \times 10^9$ years).

For each release fraction of the 15555 samples, the apparent age computed from Eq. 1 is listed in Table 1 and plotted as a function of fractional $^{39}Ar^*$ release in Fig. 1. The assigned errors are compounded of the statistical errors in the spectrometry and data corrections and are appropriate for a comparison of one release fraction with another of the same sample. A comparison of different samples must include an uncertainty of 0.010×10^9 years due to possible uncertainty in the relative neutron fluence. The absolute ages are uncertain by an additional 0.02×10^9 years due to uncertainty in the $^{40}Ar^*/K$ ratio of the monitor. A comparison of results from different laboratories must allow for the

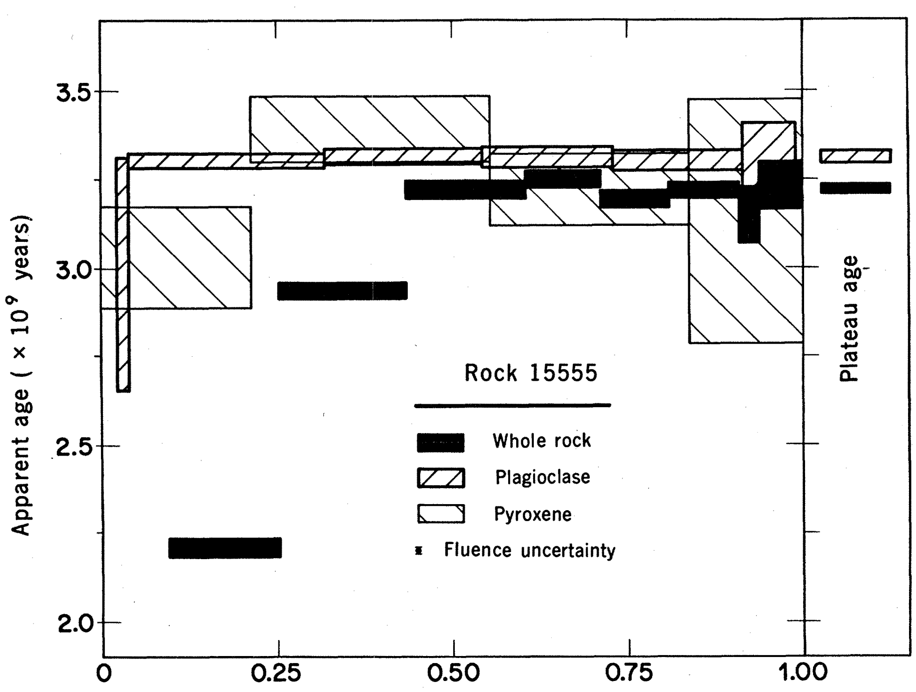

Fig. 1. Apparent K-Ar ages (data from Table 1) for individual thermal release fractions of three samples of lunar rock 15555, plotted as a function of fractional $^{39}Ar^*$ release. For each release datum, the calculated age is at the center of the box, with the horizontal length equal to the fraction of $^{39}Ar^*$ released at that temperature. The vertical extent of the box is the error limit compounded of the statistical error and data corrections. A comparison of the ages of different samples must also include the uncertainty in the relative fluence, shown separately for scale. Whole rock and plagioclase gas-retention ages obtained from the weighted mean of the high-temperature plateau points are shown at the right; these errors include fluence uncertainty. The monitor uncertainty is not included in this comparison.

possible use of different decay constants.

Within error limits the apparent ages vary in the orthodox fashion, increasing monotonically as the more retentive sites are degassed. This is a common pattern and is most reasonably attributed to prior partial diffusive loss of radiogenic $^{40}Ar^{*}$ from the least retentive sites. The 15555 data do not show the high-temperature decrease in apparent age observed but not understood in some Apollo 14 samples (*1*).

A plateau is a sequence of apparent ages that are, within error limits, the same for all the fractions spanning a significant portion of the total release pattern. If the apparent ages increase monotonically with temperature but do not define a plateau, one can only set a lower limit on the true age of the sample. The existence of a high-temperature plateau including all the points above a given temperature is taken to mean that the corresponding gas-retention age is well defined, insofar as this is the only pattern that is as yet understood in any straightforward way.

All three samples of 15555 define high-temperature plateaus. The plagioclase yields a particularly excellent plateau, and the data for the whole rock, by usual standards, define a good plateau. The pyroxene points have larger errors, but the three high-temperature points agree with each other and thus also define a plateau. For each sample an argon retention age computed from a weighted mean of the plateau points is given in Table 2.

The whole rock and the plagioclase define different ages. Errors contributed by the monitor play no role in this intercomparison, and the difference cannot be attributed to experimental artifact, such as ambiguous definition of a plateau, uncertainty in relative neutron fluence, or any of the known interferences in the ^{40}Ar-^{39}Ar technique (*5*). The pyroxene age is somewhat closer to that of the plagioclase than that of the whole rock, but it is too imprecise to permit discrimination between them.

A gas-retention age of an igneous rock is ambiguous by the length of time required for a specimen to cool from a temperature at which the gas is quantitatively lost to one at which the gas is subsequently retained quantitatively. Since the appropriate temperature may be different for different phases of a given rock, there is no a priori reason why the different phases cannot have gas-retention ages that are both well defined and significantly different. The difference between phases may also be due to later reheating sufficient to degas one phase but not another more retentive phase. For such a complex but reasonable history, a gas-retention age, even if well defined, is not necessarily the same as a formation age in the usual sense of solidification from a melt, either of the total rock or of a particular mineral phase.

The K concentration of the plagioclase is essentially the same as that of the whole rock, and that of the pyroxene is more than ten times lower (Table 2). While plagioclase and pyroxene account for 95 percent of the mass of the rock (*6*), they thus contribute less than 40 percent of the K, which indicates that the difference between the whole rock and the plagioclase Ar data is due to a minor phase or phases containing some 60 percent of the K and having an effective Ar retention time some 150×10^{6} years after that of the plagioclase. Microprobe investigations of 15555 (*6*) indicate that about ½ percent quintessence (*7*) containing 2 to 5 percent K accounts for most of the K not contained in the plagioclase and pyroxene.

The quintessence is expected to be one of the last phases to solidify from a melt, and from a correspondence between K/Ca and apparent age, Turner *et al.* (*1*) conclude that most low-temperature gas loss occurs from potassium-rich interstitial phases, presumably quintessence. The quintessence separates in two Apollo 14 rocks studied by the ^{40}Ar-^{39}Ar method (*8*) yield good plateaus at somewhat lower apparent ages than do the corresponding whole rock or the more retentive minerals such as plagioclase and ilmenite, although the effect is smaller than that seen in 15555.

Attribution of the difference between 15555 whole rock and plagioclase to a lower gas-retention age of a minor phase is a plausible hypothesis, but quantitative evaluation is difficult. Little can be said about the time and temperature regimes that would differ-

Table 1. Argon in neutron-irradiated lunar rock 15555. Temperatures above 800°C were calibrated with an optical pyrometer and an assumed spectral emissivity of 0.7. Below 800°C, temperatures were inferred from values of the oven current. Relative amounts of $^{40}Ar^{*}$ are ± 5 percent as indicated by the reproducibility of pipette calibrations; the absolute amounts are uncertain up to 10 percent. The ages were computed from Eq. 1 (see text) for $1/\lambda_{tot} = 1.885 \times 10^{9}$ years by comparison with a hornblende monitor for which $t_{o} = 1.062 \times 10^{9}$ years and $(^{40}Ar^{*} : {}^{39}Ar^{*})_{o} = 13.91$.

Extraction temperature (°C)	$^{40}Ar^{*}$ [10^{-6} cm^{3}/g (STP)]	$^{40}Ar^{*}$ / $^{39}Ar^{*}$	Apparent K-Ar age (10^{9} years)
		Whole rock (0.054 g)	
540	0.20	14.6 ± 1.6	1.095 ± 0.091
630	0.91	41.2 ± 0.8	2.209 ± 0.026
740	1.78	69.3 ± 1.1	2.936 ± 0.023
850	2.03	83.5 ± 1.4	3.220 ± 0.025
930	1.28	85.2 ± 1.4	3.251 ± 0.025
1040	1.12	82.1 ± 1.3	3.194 ± 0.024
1130	1.17	83.5 ± 1.2	3.220 ± 0.021
1200	0.33	79.7 ± 4.0	3.148 ± 0.078
1450	0.70	84.5 ± 3.3	3.237 ± 0.061
Total	9.52		
		Plagioclase (0.022 g)	
540	0.68	(220) †	(4.8) †
630	0.17	71.5 ± 15.6	2.98 ± 0.33
850	3.53	88.1 ± 1.1	3.303 ± 0.019
930	2.82	88.8 ± 1.3	3.314 ± 0.023
1040	2.38	88.6 ± 1.6	3.311 ± 0.028
1200	2.32	88.0 ± 1.6	3.302 ± 0.028
1450	0.96	88.7 ± 5.5	3.313 ± 0.096
Total	12.87		
		Pyroxene (0.064 g)	
630	0.18	73.7 ± 7.0	3.03 ± 0.14
850	0.36	93.3 ± 5.6	3.39 ± 0.09
1130	0.27	83.6 ± 5.4	3.22 ± 0.10
1450	0.15	79 ± 18	3.13 ± 0.35
Total	0.96		

† Data unreliable because of memory scrubbing by large amounts of contaminant gas. This is common in samples treated in heavy liquids before irradiation.

entiate between plagioclase and quintessence, since the effective grain sizes, diffusion coefficients, and activation energies are not known. As observed in the release patterns, neither plagioclase nor pyroxene has experienced significant loss of $^{40}Ar^{*}$. Since the whole rock has lost some 20 percent of its $^{40}Ar^{*}$, a loss of 30 to 40 percent from the quintessence is indicated. With a loss of this magnitude, it is plausible that the difference between the whole rock and the plagioclase is due to reheating (*9*) strongly affecting the quintessence but not the plagioclase or pyroxene. Alternatively, if the age difference is to be attributed to slow cooling, a cooling time of the order of 10^8 years requires formation at a great depth, which is inconsistent with the presumed formation in a lava flow near the surface.

When the Ar pattern of the whole rock is so strongly influenced by a minor nonretentive phase, whether because of slow cooling or later reheating, the true crystallization age cannot be determined by measurements on a whole rock sample, even when they define a satisfactory plateau. In this circumstance, the evaluation of the formation time of the rock as a whole must be based on more retentive primary major mineral phases. We thus conclude that the formation age of rock 15555 is that defined by the plagioclase: $3.31 \pm 0.03 \times 10^9$ years. The error represents all the experimental uncertainties, including that of the monitor (but not that of the decay constant).

The inferred formation age is in agreement with that found by the Rb-Sr method (*10*). Alexander *et al.* (*11*) report for the whole rock an ^{40}Ar-^{39}Ar plateau of 3.33×10^9 years with a different decay constant, or 3.29×10^9 years with the decay constant used here. A recalculation of our data with the decay constant they use gives a plateau for the whole rock of 3.33×10^9 years (3.42 for the plagioclase). The different results are thus largely attributable to different decay constants, although the uncertainty in the assumed age of their monitor may also be significant.

The age of 3.31×10^9 years is nearly the same as that found for Oceanus Procellarum basalts (*12*), although this does not necessarily imply a genetic relationship. Comparison with the age of 3.7×10^9 years for Mare Tranquillitatis basalts (*13*) does indicate that (at least the final) lava flows in three widely separated areas of the moon occurred within a relatively short period of time.

Table 2. Summary of results for rock 15555. The tabulated errors are compounded from all the relevant uncertainties. In a comparison of the relative ^{40}Ar-^{39}Ar ages the uncertainty due to the monitor can be ignored, and the errors in the ages of the whole rock and plagioclase samples are $\pm$ 0.016 $\times$ 10^9 years each.

Sample	K (ppm)	Argon retention age ($\times$ 10^9 years)	Cosmic ray exposure age ($\times$ 10^6 years)
Whole rock†	330	3.219 ± 0.025	
Plagioclase†	341	3.308 ± 0.025	90 ± 10
Plagioclase‡	323	3.32 ± 0.05	
Pyroxene†	26	3.28 ± 0.09	

† Argon-40–argon-39 method. ‡ Isotope dilution method.

If rock 15555 is representative of extensive lava flows in Mare Imbrium, the generation of these overlying lavas must be dissociated from the impact that excavated the Imbrium basin and due to a different, presumably internal, heat source. If the age 3.9×10^9 years for the Apollo 14 samples from the Fra Mauro formation (*1*) represents the Imbrium excavation, the interval between the excavation of the basin and the last lava flow filling it is about 600×10^6 years. Even if the Apollo 14 samples do not date the impact, an interval of at least 400×10^6 years is indicated by the flooding of Imbrium ejecta by Mare Tranquillitatis lava.

In a sample irradiated by neutrons the cosmic-ray exposure age may be determined from the ratio of spallogenic ^{38}Ar to the ^{37}Ar derived from calcium, the principal spallation target (*1*). The most favorable sample for this measurement is the plagioclase, from which we calculate an exposure age of $90 \pm 10 \times 10^6$ years. This is an upper limit to the time of surface residence and corresponds to prior burial below 1000 g/cm^2. The opposite extreme is recent excavation following a residence of 3.3×10^9 years at a depth of about 600 g/cm^2. If 15555 is to be regarded as an in situ bedrock fragment, the sampling site must be at least 600 g/cm^2 below the original local topography.

The surface in the vicinity of 15555 is a relatively mature surface with no fresh craters, and most rocks (including 15555) have rounded surfaces and well-developed fillets (*14*). The maximum surface exposure of 15555 indicates that the time scale for the generation for these features is not greater than 90×10^6 years. Because of the thinning of the regolith at the edge of the rille, the erosional processes in this vicinity are probably dominated by relatively small impacts, and most rocks along the rille edge are probably excavated locally in several small impacts rather than in one large impact. If it is assumed that 15555 was deeply buried along the edge of the rille and gradually exposed to cosmic rays by continuous erosion of the overlying material, the exposure age of 90×10^6 years determines an average erosion rate of 1.5×10^{-6} g/cm^2 year for this area.

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References and Notes

1. G. Turner, J. C. Huneke, F. A. Podosek, G. J. Wasserburg, *Earth Planet. Sci. Lett.* **12**, 19 (1971).
2. A. L. Albee, D. S. Burnett, A. A. Chodos, O. J. Eugster, J. C. Huneke, D. A. Papanastassiou, F. A. Podosek, G. Price Russ II, H. G. Sanz, F. Tera, G. J. Wasserburg, *Science* **167**, 463 (1970); P. Eberhardt, J. Geiss, H. Graf, N. Grögler, U. Krähenbühl, H. Schwaller, J. Schwarzmüller, A. Stettler, *ibid.*, p. 558.
3. J. C. Huneke, F. A. Podosek, G. J. Wasserburg, in preparation.
4. R. E. Zartman, *J. Petrology* **5**, 359 (1964).
5. G. Turner, *Earth Planet. Sci. Lett.* **10**, 227 (1971).
6. A. Albee, personal communication.
7. D. A. Papanastassiou and G. J. Wasserburg, *Earth Planet. Sci. Lett.* **11**, 37 (1971).
8. G. Turner, J. C. Huneke, F. A. Podosek, G. J. Wasserburg, in preparation.
9. G. Turner, in *Origin and Distribution of the Elements*, L. H. Ahrens, Ed. (Pergamon, New York, 1968), p. 387.
10. D. A. Papanastassiou and G. J. Wasserburg, *Earth Planet. Sci. Lett.*, in press.
11. E. C. Alexander, Jr., P. K. Davis, R. S. Lewis, *Science* **175**, 417 (1972).
12. G. Turner, *Earth Planet Sci. Lett.* **11**, 169 (1971).
13. ———, *Geochim. Cosmochim. Acta* **2** (Suppl. 1), 1665 (1970).
14. G. A. Swann, M. H. Hait, G. G. Schaber, V. L. Freeman, G. E. Uhlrich, E. W. Wolfe, V. S. Reed, R. L. Sutton, *U.S. Geol. Surv. Interagency Rep. 36* (1971).
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