HOW OLD IS THE CRATER COPERNICUS?*

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Abstract. Two KREEP glass concentrates separated from lunar soil 12033 have been dated with the Ar^{39}/Ar^{40} method. Both samples show low-temperature plateaus in accordance with a major outgassing of the KREEP glasses $(800 \pm 40) \times 10^6$ yr ago. This is the age of Copernicus, provided the identification of KREEP glass as ray material ejected during the Copernican event is true (Hubbard *et al.*, 1971). The exposure age of the two KREEP glass concentrates is 200×10^6 yr and thus distinctly smaller than the ejection age. Possible explanations for this are discussed.

1. Introduction

The mass spectrometric Ar^{39}/Ar^{40} dating method, suggested by Merrihue (1965) and applied first by Merrihue and Turner (1966) has been very successful in dating lunar rocks (Turner, 1970). One of the advantages of the technique is its ability to detect and date secondary thermal events which result in a partial outgassing only (Turner, 1969).

The Apollo 12 soil contains abundant material of basaltic composition with unusually high trace element abundances (Hubbard *et al.*, 1971; Meyer *et al.*, 1971). The acronym KREEP is commonly used to designate this soil component. KREEP fragments include different types of breccias as well as ropy and sculptured glasses (Meyer *et al.*, 1971). The ropy KREEP glass is most abundant in the two soil samples 12032 and 12033 (Marvin *et al.*, 1971). The latter was collected from the bottom of a soil mechanics trench at a depth of approximately 15 cm (Shoemaker *et al.*, 1970). The chemical composition of the KREEP material is different from the composition of the local basalts, indicating a non-local origin of the KREEP component in the soil. The results of the Apollo 15 and 16 gamma-ray experiments (Arnold *et al.*, 1972a, b) have verified that KREEP-like material is an abundant soil component in the entire Mare Imbrium and Oceanus Procellarum region. A large number of events, probably spread over a very extended time period, must have been responsible for the distribution of the KREEP component in these mare regions. However, the high abundance of KREEP glass in certain Apollo 12 samples or soil levels, suggest that this particular KREEP glass was formed and deposited by essentially one major event. An absolute dating of this event with the Ar^{39}/Ar^{40} method was considered a possibility.

In this paper we report the dating of this major event with the Ar^{39}/Ar^{40} method. A preliminary report of these results was given at the Third Lunar Science Conference (Eberhardt *et al.*, 1972a). In our laboratory we have also determined exposure ages of a large number of individual KREEP glass and other soil fragments. These investigations are still in progress and the results will be reported elsewhere. Some of these

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results will, however, be used in our discussion of the Ar^{39}/Ar^{40} ages obtained for the KREEP glass.

2. Experimental Technique

Two KREEP glass fractions were prepared from our sample 12033.56 (weight 1.02 g) as follows: First, a coarse fraction ($>150\ \mu m$) was obtained by sieving in acetone. Then, two fractions enriched in the ropy KREEP glass were separated with heavy liquids from this coarse fraction (density ranges 2.88–2.96 g cm^{-3} and 2.96–3.11 g cm^{-3}). The two density separates were further enriched by hand picking impurities. Large KREEP glass grains (larger than approximately 0.5 mm) were removed for subsequent analysis of the noble gases in single grains. The KREEP concentrates were cleaned of adhering dust with acetone in an ultrasonic bath. This treatment did not remove the characteristic welded coating of white, feldspar rich dust (Meyer *et al.*, 1972). From optical inspection we estimate that these two KREEP fractions (fa-38, density 2.88–2.96 g cm^{-3}; and fa-35, density 2.96–3.11 g cm^{-3}) contained more than 90% ropy glass.

The two KREEP glass samples were irradiated, together with four hornblende standards (CC-27) and Ni-wire flux monitors in the FR-2 reactor of the Gesellschaft für Kernforschung mbH in Karlsruhe. A 1 mm Cd shield was used to lower the thermal neutron flux by approximately 7 orders of magnitude. The Ni-wire monitors showed a flux inhomogeneity of less than $\pm 0.3\%$ over the whole volume of the irradiation container.

The subsequent Ar analyses were made in an inductively heated extraction system connected on line to a double sector magnetic mass spectrometer. The advantage of the double sector mass spectrometer is its high abundance sensitivity which enables the measurement of extreme ratios without any interference from tails of abundant isotopes. The mass spectrometer is equipped with programmable magnets and a digital ion current measuring system.

The hornblende standard CC-27 was obtained from Dr S. R. Hart (Dept. of Terrestrial Magnetism, Carnegie Institution of Washington). The K/Ar age of $(2650 \pm 25) \times \times 10^6$ yr was determined from (a) K and Ar determinations with the isotopic dilution method made in our laboratory and (b) from comparison with the well dated standards P 207 (Lanphere and Dalrymple, 1967); CAA 12 (Lanphere, 1968); BS 1 (Dalrymple and Lanphere, 1971); Be 4 (Jäger, 1963; Dalrymple and Lanphere, 1971), using the total Ar^{39}/Ar^{40} ratios.

3. Results

The results of the Ar analyses on the irradiated samples are given in Table I. All results are corrected for extraction blank and mass spectrometer background. The Ar^{37} and Ar^{39} abundances are corrected for decay by extrapolating back to the time of irradiation. Corrections for interfering reactions were also applied, using the following relations: $(Ar^{36}/Ar^{37})_{Ca} = (2.7 \pm 0.2) \times 10^{-4}$; $(Ar^{39}/Ar^{37})_{Ca} = (6.85 \pm 0.2) \times 10^{-4}$; $(Ar^{38}/Ar^{37})_{Ca} = (6 \pm 2) \times 10^{-5}$ (values determined from several $CaCO_3$, CaF_2 and

TABLE I

Results of Ar measurements on two KREEP glass concentrates from 12033. All data are corrected for interfering reactions (see text)

Temperature (°C)	Ar^{40} (10^{-8} cm³ STP/g)	$\frac{Ar^{40}}{Ar^{39}}$	$\frac{Ar^{37}}{Ar^{39}}$	$\frac{Ar^{36}}{Ar^{38}}$	$\frac{Ar^{40}}{Ar^{36}}$
KREEP glass concentrate fa-35 (14.5 mg)					
400	63.7	94.2	7.6	2.54	83.0
	±3.6	±6.7	±0.7	±0.20	±5.0
650	78.5	60.6	6.55	3.37	29.6
	±4.3	±4.2	±0.35	±0.15	±1.2
840	204.5	54.5	6.62	2.45	50.8
	±10.5	±1.8	±0.25	±0.06	±1.2
880	98.5	52.2	6.94	2.80	34.8
	±5.2	±2.3	±0.30	±0.10	±1.0
940	234	46.0	7.04	4.25	6.90
	±12	±2.1	±0.30	±0.08	±0.12
1070	1110	52.5	7.52	4.42	5.28
	±57	±3.2	±0.20	±0.07	±0.10
1160	390	80.6	8.51	4.47	5.06
	±20	±4.6	±0.35	±0.09	±0.11
1320	1360	92.5	8.94	4.50	5.80
	±70	±3.8	±0.35	±0.07	±0.12
1600	565	184.7	10.0	3.80	28.6
	±30	±7.4	±0.4	±0.10	±0.7
total	4100	72.7	7.95	4.38	7.00
	±100	±3.5	±0.30	±0.08	±0.14
KREEP glass concentrate fa-38 (10.8 mg)					
400	70.0	118.6	6.0	2.35	158
	±4.2	±4.7	±0.7	±0.35	±14
720	220	53.6	4.21	3.35	37.4
	±15	±1.8	±0.15	±0.10	±1.1
860	420	45.1	5.84	3.59	18.1
	±25	±0.5	±0.25	±0.08	±0.4
940	88.0	52.5	5.45	3.76	17.0
	±6.0	±3.2	±0.35	±0.15	±0.6
1050	568	55.1	7.82	4.46	4.74
	±40	±2.1	±0.30	±0.05	±0.07
1140	495	82.8	8.82	4.26	6.63
	±45	±3.0	±0.25	±0.06	±0.13
1280	805	110.1	9.00	4.52	5.24
	±60	±2.8	±0.30	±0.07	±0.06
1580	920	124.1	9.23	3.50	21.5
	±45	±3.3	±0.30	±0.04	±0.3
total	3585	76.0	7.55	4.24	8.42
	±100	±2.3	±0.30	±0.08	±0.10

Ca-metal monitors); $(Ar^{40}/Ar^{37})_{Ca} \leqslant 5 \times 10^{-3}$ (Dalrymple and Lanphere, 1971), $(Ar^{40}/Ar^{39})_K = (6 \pm 2) \times 10^{-3}$ (Dalrymple and Lanphere, 1971; Mitchell, 1968), $(Ar^{38}/Ar^{39})_K = (2 \pm 1) \times 10^{-2}$ (estimated from our terrestrial monitors with high K/Ca). Corrections for these interfering reactions were always smaller than 4%.

For the calculation of the Ar^{39}/Ar^{40} ages corrections for spallation and trapped Ar^{40} have to be applied. For the fractions extracted at higher temperatures the corrections for Ar^{40}_{tr} become significant (cf. Table II). The following ratios were used to make these corrections: $(Ar^{36}/Ar^{38})_{sp} = 0.65 \pm 0.1$; $(Ar^{40}/Ar^{38})_{sp} = 0.2 \pm 0.1$; $(Ar^{36}/Ar^{38})_{tr} = 5.32 \pm 0.15$; $(Ar^{40}/Ar^{36})_{tr} = 0.7 \pm 0.3$. The first three ratios are essen-

TABLE II

Ar^{39}/Ar^{40} ages of KREEP glass concentrates from 12033

Temperature (°C)	Ar^{40}_{rad} (10^{-8} cm³ STP/g)	$\frac{Ar^{40}_{rad}}{Ar^{40}}$	$\frac{Ar^{40}_{rad}}{Ar^{39}}$	Age (10^6 yr)	$\frac{Ar^{38}_{sp}}{Ar^{37}}$
KREEP glass concentrate fa-35					
400	< 60	0.99	< 93.6	< 1450	0.035
650	77	0.98	59.3	1035	0.039
	± 8		± 4.1	± 35	± 0.002
840	202	0.99	53.9	960	0.041
	± 12		± 1.8	± 17	± 0.002
880	97	0.98	51.3	925	0.042
	± 8		± 2.3	± 25	± 0.003
940	212	0.90	41.5	780	0.052
	± 20		± 2.6	± 30	± 0.004
1070	965	0.87	45.7	845	0.059
	± 85		± 3.8	± 40	± 0.003
1160	340	0.87	70.2	1175	0.078
	± 35		± 5.8	± 65	± 0.005
1320	1200	0.88	81.7	1315	0.072
	± 90		± 5.4	± 60	± 0.004
1600	555	0.98	180.5	2210	0.056
	± 40		± 7.5	± 50	± 0.002
total	3710	0.90	65.2	1115	
	± 140		± 4.3	± 60	
KREEP glass concentrate fa-38					
400	< 70	0.99	< 120	< 1700	0.034
720	217	0.98	52.7	945	0.043
	± 15		± 1.8	± 20	± 0.002
860	405	0.96	43.5	810	0.045
	± 25		± 0.8	± 10	± 0.001
940	85	0.96	50.5	915	0.051
	± 6		± 3.2	± 30	± 0.003
1050	485	0.86	47.2	865	0.063
	± 45		± 3.5	± 30	± 0.002
1140	445	0.90	74.4	1230	0.077
	± 45		± 4.3	± 35	± 0.002
1280	700	0.87	95.8	1470	0.091
	± 65		± 6.0	± 40	± 0.002
1580	890	0.97	120.4	1720	0.069
	± 45		± 3.6	± 35	± 0.004
total	3230	0.94	69.9	1175	
	± 110		± 3.2	± 35	

tially independent of the type of lunar material involved. The assumed uncertainties, including any possible variations, introduce negligible errors in the Ar^{40}_{tr} corrections. The $(Ar^{40}/Ar^{36})_{tr}$ ratio is more critical and, furthermore, large variations have been observed in lunar material. The value assumed was obtained from a Ar^{40} vs Ar^{36} correlation diagram (Heymann and Yaniv, 1970) based on the Apollo 12 samples 12033, 12024, 12028, 12042, and 12060 (cf. also Pepin *et al.*, 1972; data from Funkhouser *et al.*, 1972). The error obtained from the least square fit of the correlation line is only ± 0.02. We assumed a much larger uncertainty of ± 0.3 to include in addition the different $(Ar^{40}/Ar^{36})_{tr}$ ratios observed in other Apollo 12 and Apollo 11 fines and possible variations of the ratio with release temperature. The corrected ratios and concentrations relevant for the following discussion are given in Table II as well as the Ar^{39}/Ar^{40} ages.

4. Ar^{39}/Ar^{40} Ages

The release pattern obtained for the two KREEP glass fractions (Figures 1 and 2) are quite similar. The apparent ages for the lowest temperature fractions, up to a total Ar^{39} release of approximately 10%, are between 925×10^6 yr and 1035×10^6 yr. The next temperature fractions, comprising 45% of the total Ar^{39} release of both samples, show a lower apparent age with a quite well defined plateau. For higher temperatures the apparent age then increases monotonically to values above 1500×10^6 yr. The

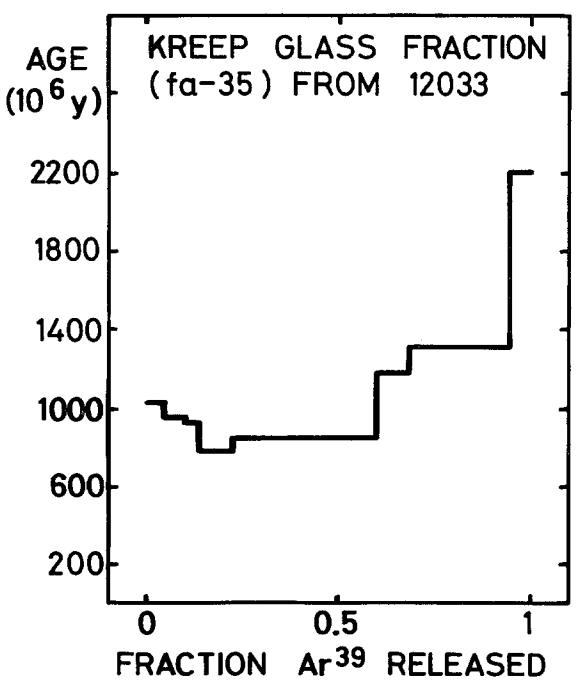

Fig. 1. Ar^{40}/Ar^{39}-release pattern of the KREEP glass fraction fa-35 (14.5 mg) which was separated from lunar fines 12033.

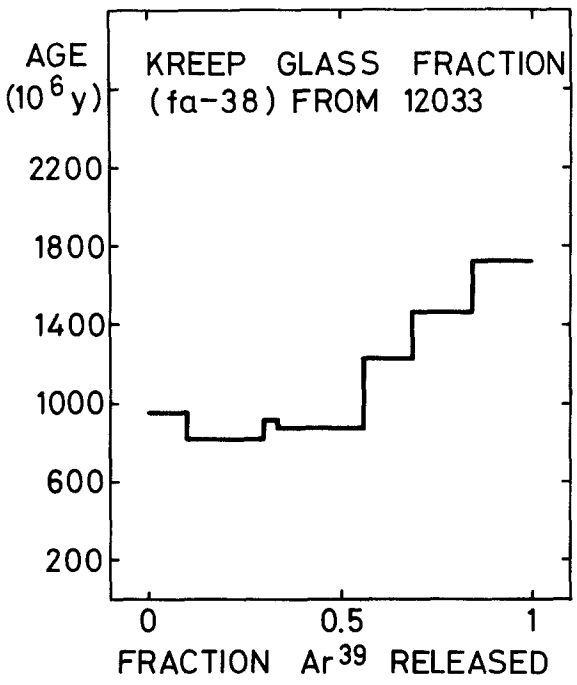

Fig. 2. Ar^{40}/Ar^{39} release pattern of the KREEP glass fraction fa-38 (10.8 mg) which was separated from lunar fines 12033.

average ages for the low temperature plateau are $(830 \pm 40) \times 10^6$ yr for the fa-35 concentrate (average of 940° and 1070° fractions, weighted on the basis of released Ar^{39}) and $(845 \pm 25) \times 10^6$ yr for the fa-38 concentrate (weighted average of 860°, 940°, and 1050° fractions). Virtually the same average age is obtained for fa-38 if the 940° fraction is not used.

Figure 3 shows a comparison of the average Ar^{40}/Ar^{39} release pattern observed for the two KREEP fractions with theoretically calculated release curves. The average for the two KREEP fractions was obtained by unweighted averaging the two observed release curves. The theoretical release curves were adapted from Figure 2 of Turner (1969), assuming a primary cooling age of 4000×10^6 yr, partial outgassing 800×10^6 yr ago and a lognormal distribution of spherical grains. The observed release pattern, except for the initially higher Ar^{40}/Ar^{39} ratio, is in good overall agreement with the theoretically calculated release patterns for a lognormal distribution of old spherical grains subjected to a recent episodic partial outgassing. The outgassing, which occurred 800×10^6 yr ago, must have been nearly complete. On the basis of the theoretical model employed for the calculation of the curves in Figure 3 we estimate that less than 5% of the radiogenic Ar^{40} was retained.

The theoretical curves show that even for a very severe outgassing event, the low temperature plateau will not be exactly horizontal. The release pattern for both KREEP glass concentrates (Figures 1 and 2) as well as the calculated average pattern

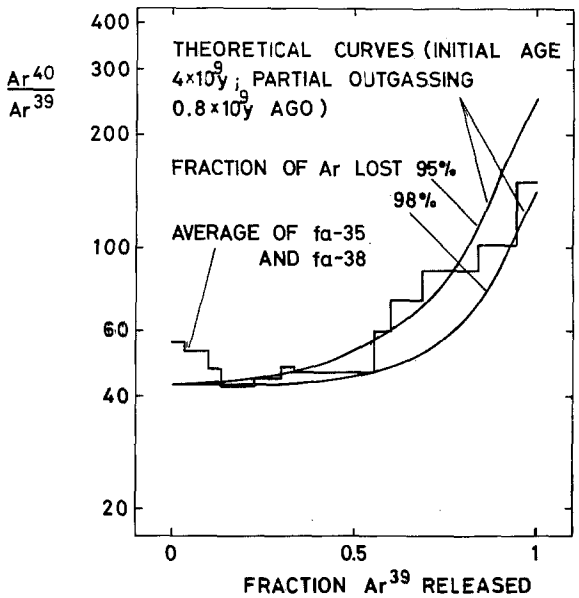

Fig. 3. Ar^{40}/Ar^{39}-release pattern obtained by averaging the two observed release curves. The theoretical curves were adapted from Turner's (1969) work, assuming a primary cooling age of 4000×10^6 yr, partial outgassing 800×10^6 yr ago and a lognormal distribution of spherical grains. It is seen that an Ar loss between 95 and 98% in an episodic event 800×10^6 yr ago fits the data.

(Figure 3) show a small slope in the low temperature plateau but the uncertainty in the Ar^{40}_{tr} correction (cf. Table II) is too large to allow a definite conclusion. Because of this small slope the average ages calculated for the low temperature plateau fractions are slightly higher than the age of the thermal event responsible for the outgassing. From Figure 3 we conclude that the true age for this major event is $(800 \pm 40) \times 10^6$ yr.

The slightly higher apparent age of the lowest temperature fractions might be explained by a contamination of our KREEP glass samples with older material of low Ar retentivity. Obviously, this material could not have been subjected to the same thermal history as the KREEP glass or it would have lost its loosely bound Ar^{40}. We might tentatively identify this contamination as the white, feldspar rich dust which coats most of the KREEP glass particles.

5. K, Ca Concentrations, Exposure Ages

From the total Ar^{37} and Ar^{39} concentrations and the known K and Ca content of our hornblende monitor we can calculate the K and Ca concentrations for the two KREEP glass concentrates (cf. Table III). The Ar^{37}/Ar^{39} ratio of the different temperature fractions is constant within $\pm 20\%$ (except 720 °C fraction of fa-38), indicating a relatively close association of Ca and K (the Ca/K ratio can be obtained from the Ar^{37}/Ar^{39} ratio by multiplying the latter by 1.87). The average Ca content

TABLE III

K and Ca concentrations and exposure ages of the two KREEP glass concentrates

Sample	K (%)	Ca (%)	Exposure age (10^6 yr)
fa-35	0.61	9.2	190
fa-38	0.51	7.2	210

of 8.2% for the two KREEP concentrates is in good agreement with the average Ca content of KREEP glass reported by Hubbard *et al.* (1971) and Meyer *et al.* (1971). Our K content is, however, a factor of two lower than the values reported by these authors. In a large (6.4 mg) KREEP glass fragment Hubbard *et al.* (1971) also measured a low K content of 7030 ppm. Funkhouser (1971) found in several different KREEP glass fragments K contents of approximately 4000 ppm. We have no satisfactory explanation for this apparent variability of the K content.

The Ar^{38}_{sp}/Ar^{37} ratio in the different temperature fractions is variable (cf. Table II), probably because of the different outgassing behaviour of the Ca and Fe target-atom sites. It is thus not feasible to deduce an exposure age from the Ar^{38}_{sp}/Ar^{37} ratio. Exposure ages were calculated from the total Ar^{38}_{sp} concentrations (cf. Table III). The production rates compiled in the caption of Table VII of Eberhardt *et al.* (1972b), the K and Ca abundances given in Table III and the average Fe and Ti abundances for KREEP glass reported by Meyer *et al.* (1971) were used.

6. Discussion

Each of the two KREEP glass samples investigated consisted of more than one hundred individual particles. To prove that the low temperature Ar^{39}/Ar^{40} ages obtained for these two samples correspond to a single major outgassing event for the 12033 KREEP glass we must show that the history of a large fraction of these KREEP glass fragments is identical. The arguments which favor the interpretation of the low-temperature Ar^{39}/Ar^{40} ages as a true event are:

(1) No low temperature plateau would be expected if the ages and histories of the individual grains were significantly different.

(2) The two investigated KREEP glass fractions, comprising different density ranges, have the same low-temperature ages.

(3) The 12033 soil sample is heavily enriched in KREEP glass (Marvin *et al.*, 1971).

(4) The exposure ages of individual KREEP glass particles in soil 12033 show a strong clustering. This is evident from our own measurements (Eberhardt *et al.*, 1972, and unpublished results) as well as from the results of Funkhouser (1971). The exposure ages given by Funkhouser (1971) and Eberhardt *et al.* (1972) cluster around slightly different values. This is essentially an artifact of the different production rates used by the two groups. A trivial explanation for the homogeneity of the 12033

KREEP glass exposure ages could be given if the KREEP glass-rich soil region encountered in the soil trench originated from a large KREEP glass fragment broken up in situ. However, on the basis of the observed morphology of the ropy KREEP glass (Meyer *et al.*, 1971; Marvin *et al.*, 1971) this possibility is easily excluded. The exposure age distribution of the 12033 KREEP glass fragments thus confirms that these particles had a similar history.

Other investigators have used different methods to estimate the age of the secondary thermal episode related to the Apollo 12 KREEP. From the study of the U–Th–Pb system, Silver (1971) concluded that the 'exotic' radioactive debris in the Apollo 12 soil was subjected to an important thermal episode less than 10^9 yr ago. His estimate of $(850 \pm 100) \times 10^6$ yr for its age is in excellent agreement with our low-temperature Ar^{39}/Ar^{40} age. Applying a two component systematics to K–Ar dates of Apollo 12 bulk soils, Pepin *et al.* (1972) deduced an K–Ar age of $(950 \pm 50) \times 10^6$ yr for the KREEP component in these soils. This total K-Ar age is intermediate between our low-temperature and total Ar^{39}/Ar^{40} ages. Our KREEP glass fractions are relatively coarse grained compared with bulk soil samples and the lower total K–Ar age deduced for the bulk KREEP material is consistent with the expected higher outgassing of the finer grain sizes during the 800×10^6 yr event.

A much higher age of at least 2000×10^6 yr for the original deposition of the KREEP stratum at the Apollo 12 site was deduced by Funkhouser (1971). His estimate is based on K–Ar ages of relatively large (>1 mg) KREEP glass fragments. His age is based on the assumption that the Ar outgassing during the thermal event related to the deposition was complete and that, furthermore, subsequent Ar loss occurred in most KREEP glass grains. From our results it is evident that neither assumption is valid, i.e. the outgassing during the thermal event was incomplete, especially in larger grains and there is no apparent evidence for subsequent Ar loss.

The possible origin of the KREEP glass at the Apollo 12 landing site has been discussed by several authors. Hubbard *et al.* (1971) came to the conclusion that KREEP glass is Copernican ray material (cf. also Meyer *et al.*, 1972; Marvin *et al.*, 1972). Quaide *et al.* (1972) argue that a swarm of Copernican secondary craters, 45 km to the north of the Apollo 12 landing site, might be the source. For both models the major thermal event, dated by the low-temperature Ar^{39}/Ar^{40} plateau, is most likely the melting and ejection of the KREEP material in one of these cratering events. Within the present-day precision of the Ar^{39}/Ar^{40} method these cratering events occurred simultaneously and, provided the association of the KREEP glass with the Copernican event is not refuted in the future, we can conclude that the age of the crater Copernicus is $(800 \pm 40) \times 10^6$ yr.

The average exposure age of 200×10^6 yr of our two KREEP glass concentrates is essentially in agreement with the exposure ages measured for individual KREEP glass particles (Funkhouser 1971; Eberhardt *et al.*, 1972a, and unpublished results). The exposure age of the KREEP glass is thus a factor of four lower than the Ar^{39}/Ar^{40} age. Two simple explanations can be suggested:

(1) After the initial deposition during the Copernican event the KREEP glass was

rapidly covered by a sufficiently thick regolith layer to shield the glass from cosmic radiation. Relatively recently this shielding layer was removed. Local events could be responsible for both the covering and uncovering of the KREEP glass layer. Such a model was discussed by Funkhouser (1971) and Eberhardt *et al.* (1972a).

(2) Variations of the galactic cosmic ray intensity during the last 800×10^6 yr could also explain the low exposure age. A four times lower average flux would be required to bring exposure age and Ar^{39}/Ar^{40} age in agreement. However, the results of K^{40}/K^{41} exposure age measurements in iron meteorites do not support such a large variation in the cosmic ray intensity during this time period (Voshage, 1967).

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During the 1950's two of the authors had the privilege of working in Dr Urey's laboratory in Chicago and La Jolla. The close association with Dr Urey and the constant exposure to his thoughts and ideas on meteorites, the Moon and the early history of the solar system has greatly influenced and stimulated the subsequent development of meteorite and lunar research at the Physikalisches Institut in Bern.

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