Hydrogen, Oxygen and Silicon Isotope Systematics in Lunar Material

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Hydrogen, oxygen and silicon isotope systematics in lunar material

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In this review the following conclusions are supported:

(1) The bulk of the hydrogen in the lunar soils represents protons implanted from the solar wind and is essentially deuterium free.

(2) Rarely samples of relatively detuerium rich hydrogen are found, probably resulting from *in situ* spallation reactions.

(3) The water found in lunar samples is probably entirely terrestrial contamination (which yields a third component of hydrogen in the lunar samples).

(4) Extreme ^{18}O and ^{30}Si enrichments are found in the surfaces of grains in the lunar soil. These probably result from the condensation of material which was vaporized from the lunar surface by bombardment from micrometeorites, etc., and fractionated in the lunar atmosphere, leaving a heavy isotope enriched fraction to condense. The complimentary fraction enriched in light isotopes escapes and about 1 % of the mass of the regolith has been lost in this way.

(5) Oxygen isotope geothermometry on lunar basalts gives temperatures in the approximate range 1000–1150 °C generally close to experimentally determined liquidus temperatures.

(6) Recent oxygen isotope studies by Clayton and co-workers provide exciting new evidence that the Earth–Moon system has a different origin from the higher temperature condensates in the chondritic meteorites.

Introduction

This paper reviews studies of the abundances of hydrogen, oxygen and silicon isotopes in lunar material which have been published between 1970 and 1975. A few unpublished oxygen isotope data for Luna 16 and 20 soils are also briefly discussed. During this relatively brief period of about five years both a large body of analytical data and an extensive literature have been produced. However, interpretation of many of the results has provided scope for considerable controversy, and the points at issue are extremely important to our understanding of the origin of the Moon and the 'geological' processes that occur in the lunar interior and on the lunar surface. In this review attention will be directed mainly to the isotopic evidence relating to the following important, and in some cases controversial, topics:

(*a*) The origin of the water in lunar material. The crux of the problem is whether this water is indigenous to the Moon or whether all or part of it is terrestrial contamination. Indigenous water could be derived from outgassing of the lunar interior or the impacts of hydrous bodies such as comets or carbonaceous chondrites. Terrestrial water contamination could have been added to the samples on the lunar surface from rocket exhaust gases or the astronauts back-packs. Terrestrial water contamination could also have been derived from the atmosphere of either the returning spacecraft or the various laboratories handling the samples, or both.

(*b*) The large variations in oxygen and silicon isotope abundances found associated with the surfaces of grains from the regolith. These variations are thought to reflect isotope fractionation resulting from the bombardment of the lunar surface by the particle flux from the solar wind, solar flares or cosmic rays, and/or micrometeorites and meteorites.

(*c*) Geothermometry: the temperatures recorded in the crystalline rocks by the partition of ^{18}O between coexisting phases.

(*d*) The origin of the Moon. Recent discoveries of new oxygen isotope evidence suggest that the earth and Moon have a similar origin which, at least in terms of the genesis of the abundances of oxygen isotopes, is significantly different from that of the high temperature condensates in chondritic meteorites.

Throughout this review isotope abundance data are reported in standard δ notation in per thousand (mil) (‰) where:

$$\delta = \frac{R_{\text{sample}} - R_{\text{standard}}}{R_{\text{standard}}} \times 10^3$$

$R = D/H$, $^{18}O/^{16}O$, $^{17}O/^{16}O$ or $^{30}Si/^{28}Si$ and the standards adopted are 'standard mean ocean water' (s.m.o.w.) for hydrogen and oxygen (Craig 1961) and Caltech rose quartz for silicon (Taylor & Epstein, 1970*a*; Epstein & Taylor 1971). Thus, for example, a value of $\delta D = 0$‰ (s.m.o.w.) means that the sample has the same deuterium content as s.m.o.w. (about 157 parts/10^6) and a δD value of -1000‰ (s.m.o.w.) means that the sample is deuterium free.

ABUNDANCE OF HYDROGEN ISOTOPES IN LUNAR MATERIAL

The interpretation of the hydrogen isotope abundance data for lunar material is not straightforward. The difficulty stems from the fact that both elemental hydrogen and water are present in most, if not all, samples. Two extraction techniques have been used to obtain hydrogen for isotopic analysis: (1) Combustion, in which (after initial outgassing) the sample is heated in oxygen converting all the hydrogen present to water. (2) Pyrolysis, in which the sample is heated *in vacuo* and the evolved water and hydrogen may be collected separately, and in successive temperatures fractions, for isotopic analysis. Clearly using the combustion technique the relative contributions to the total extracted hydrogen from water and from elemental hydrogen are indeterminate. With the pyrolysis technique it is usually difficult or impossible to preclude the possiblity that isotopic exchange between elemental hydrogen and water occurs during heating; modifying the isotopic composition of both the hydrogen and the water.

The ranges of concentration of water and hydrogen in lunar samples are, 4–40 and 0.1–120 μmol g^{-1} respectively. The relation between the total hydrogen content and isotopic composition is summarized in figure 1 which is adapted from Friedman, Hardcastle & Gleason (1974*a*). It is evident that regularities in the data exist and we will divide the results into three groups. First, the extremely deuterium enriched datum is the single analysis of the interior of basalt sample 12051 by Friedman, O'Neil, Gleason & Hardcastle (1971) with a δD value of $+307$‰. The description of the experiments surrounding this analysis (Friedman *et al.* 1971) appears to be convincing evidence that, although the result seems unusual for lunar material, it is not an artefact. Epstein & Taylor (1972, 1973) have also recorded unusually deuterium rich hydrogen fractions extracted from samples of certain lunar fines (14240, 14422 and 61221) during the highest temperature stage of pyrolysis (700 °C – melting). The results obtained for these three fractions of gas were -250, -255, and -88‰ respectively. Although these values are very much less enriched in deuterium than the value of Friedman *et al.* (1971) for basalt sample 12051, they are nevertheless relatively 'heavy' for lunar hydrogen (see below). Epstein & Taylor (1972, 1973) also suggest that it is not possible to explain their δD values for 14240, 14422 and 61221 as effects of cross-contamination with water during pyrolysis, because the water fractions released at lower temperatures do not contain sufficient deuterium.

Two possible explanations have been offered for the relative deuterium enrichment in this first group of hydrogen data:

(1) Deuterium may be produced in spallation reactions, in which deuterons are 'spalled' off heavier nuclides as a result of bombardment by high energy particles in the flux of cosmic rays.

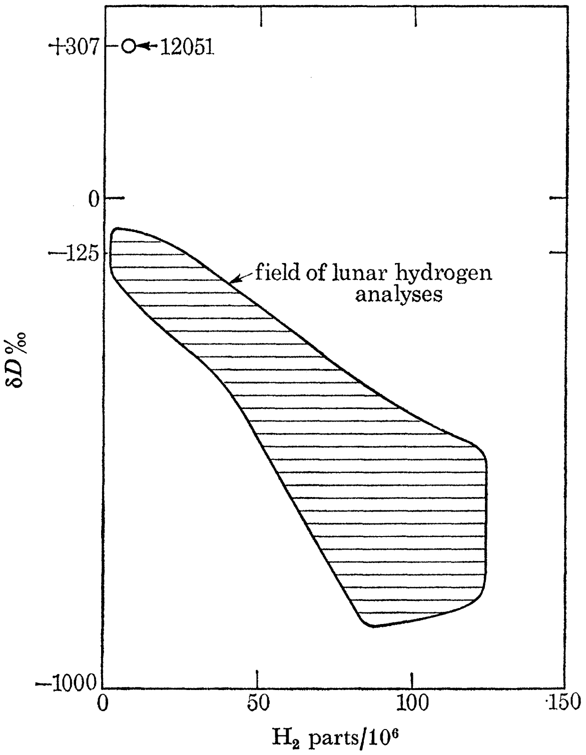

Figure 1. The relation between total content and isotopic composition of hydrogen in lunar samples (adapted from Friedman, Hardcastle & Gleason 1974).

(2) Deuterium-enriched hydrogen may represent part of the debris from carbonaceous chondrites colliding with the Moon. Boato (1954) has shown that certain examples of both type I and type II carbonaceous chondrites, which contain about 10% and 1% combined water respectively (i.e. water remaining after baking the samples at 180 °C *in vacuo*), can yield hydrogen with δD values in the approximate range +200 to +300‰. Although the currently available evidence in favour of this process is far from conclusive the possibility that a certain amount of water could have been incorporated in the Moon from impacting carbonaceous chondrites should not be overlooked.

Turning next to consider the main body of hydrogen isotope abundance data (figure 1) it is apparent that there is a correlation between the hydrogen content and δD value of lunar material. Samples with more than about 50 μmol g^{-1} of hydrogen yield depleted δD values, less than about −400‰. Samples with less than about 50 μmol g^{-1} hydrogen yield δD values up to about −125‰. At this point it is useful to refer to figures 2 and 3 which have been adapted from Epstein & Talyor (1973). These diagrams show respectively, the relation between the δD value of extracted water and the molar ratio H_2O/H_2 in lunar soils and the relation between the δ*D* value of extracted hydrogen gas and the molar fraction of H_2O in the total H_2O+H_2 mixture in soils and breccias. Epstein & Taylor (1970, 1971, 1972, 1973) suggest that the δ*D* value of about -125 ‰ which is approached asymptotically in figure 2 as the molar ratio H_2O/H_2 increases is probably the best present estimate of the deuterium content of the water in lunar material. The increasingly depleted δ*D* values observed as the molar ratio H_2O/H_2 falls are interpreted as artefacts reflecting either isotopic exchange between water and deuterium-depleted hydrogen during pyrolysis (the technique used by Epstein & Taylor), or mixing of hydrogen from these two components during combustion (Friedman's group have used both the pyrolysis and combustion techniques). The second group of hydrogen isotope abundance data (at one end of the band of results in figure 1) therefore represents a water component in lunar material with a δ*D* value of about -125 ‰. The origin of this 'lunar water' will be discussed in the next section.

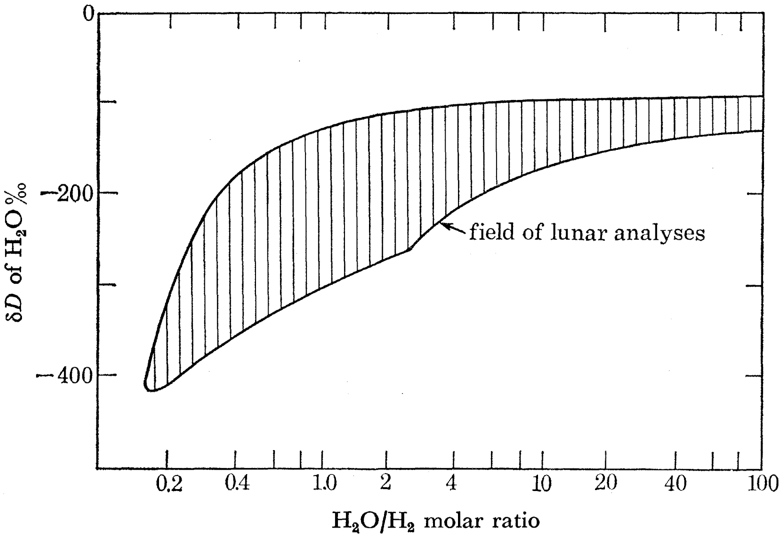

FIGURE 2. The relation between the δD value of extracted water and the molar ratio H_2O/H_2 in lunar soils (adapted from Epstein & Taylor 1973).

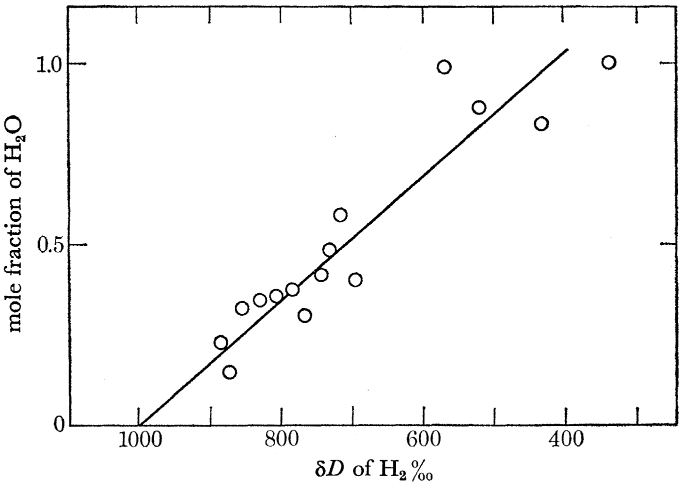

FIGURE 3. The relation between the δ*D* value of extracted hydrogen gas and molar fraction of H_2O in the H_2O+H_2 mixture in soils and breccias (adapted from Epstein & Taylor 1973).

Finally a third group of hydrogen isotope data can be identified in figure 3 where it can be seen that samples with very low water contents yield very low δD values. Extrapolation of the data to a water content of zero suggests that lunar samples contain elemental hydrogen which is almost deuterium free. This component of the total hydrogen is interpreted as representing protons from the solar wind implanted in the samples on the lunar surface. Once again the relatively deuterium rich δD values defining the trend of increasing δD with increasing H_2O content in figure 3 are interpreted as experimental artefacts. In more detail, extrapolation by Epstein & Taylor (1972) of the data summarized in figure 3 gives a δD intercept of about -995‰, corresponding to a deuterium content of about 1 part/10^6, for a water-free sample. Epstein & Taylor (1971) have shown that at least for sample 10061 this level of deuterium could have been generated *in situ* by spallation reactions. It is concluded that the bulk of the hydrogen in typical lunar material is contained in surface (i.e. soil) samples, it is derived from the solar wind and is essentially deuterium free.

THE ORIGIN OF THE WATER IN LUNAR SAMPLES

As noted above the concentration of water in lunar samples ranges (approximately) from 4 to 40 μmol g^{-1}, and the interpretation of figure 2 suggests that the best estimate of the δD value of this water is about -125 ‰. Oxygen isotope ratios for the water in lunar material have been determined in only three samples. Epstein & Taylor (1972) analysed water extracted from rock breccia sample 14321 and fines sample 15301 during pyrolysis up to about 550 °C, obtaining $\delta^{18}O$ values of about -18 ‰ and -6‰ respectively. They suggested that these must be maximum values of ^{18}O enrichment because of the likelihood of isotope exchange between the water and relatively ^{18}O-enriched silicates and oxides (see below) in the samples at the high temperatures used for pyrolysis. Both Friedman, Hardcastle & Gleason (1974*b*) and Epstein & Taylor (1974) have analysed the water in the 'rusty rock' 66095. However although the δD values determined by both groups are reasonably consistent (about -100 ‰), the $\delta^{18}O$ values are completely different. Friedman *et al.* (1974*b*) found a $\delta^{18}O$ value of about $+5$‰ (the water was extracted by pyrolysis, converted to oxygen by either carbon reduction or fluorination using bromine pentafluoride and then to carbon dioxide for analysis). Epstein & Taylor (1974), on the other hand found $\delta^{18}O$ values averaging about -17 ‰, not too different from their earlier analysis cited above (the water was extracted by pyrolysis, converted to oxygen by reaction with fluorine and then to carbon dioxide for analysis). The magnitude of the difference in $\delta^{18}O$ values determined by these two groups is between about ten and twenty times the analytical error. Moreover the two $\delta^{18}O$ values allow two fundamentally different interpretations of the origin of the water in lunar samples. If the $\delta^{18}O$ value of the water in 66095 is close to -17 ‰ it plots exactly on the locus of δD against $\delta^{18}O$ values defined by terrestrial meteoric precipitation. This is illustrated in figure 4 and strongly suggests that the 'lunar water' is in fact terrestrial contamination. If the water in 66095 has a $\delta^{18}O$ value close to $+5$‰ it is unlike any normal terrestrial contamination and may indeed be indigenous to the Moon.

In this situation it is clearly important to look at additional lines of evidence for the origin of the water in lunar samples. Three sets of observations are particularly convincing: First a considerable percentage (20 % or more) of the water in 66095 is released *in vacuo* at 25 °C and most of the water is released at 200 °C (Friedman *et al.* 1974*b*; Epstein & Taylor 1974).

High temperatures are attained during the two week lunar day and it is clearly impossible that the water in 66095 could have survived the lunar vacuum unless the Moon is actively outgassing water at this locality. Secondly the concentration profiles of hydrogen, carbon and fluorine versus depth in the surfaces of lunar samples determined by Padawer, Kamykowsi, Stauber & D'Agostino (1974) and Leich, Goldberg, Burnett & Tombrello (1974) indicate that prominent surface enrichments occur for each of these elements.

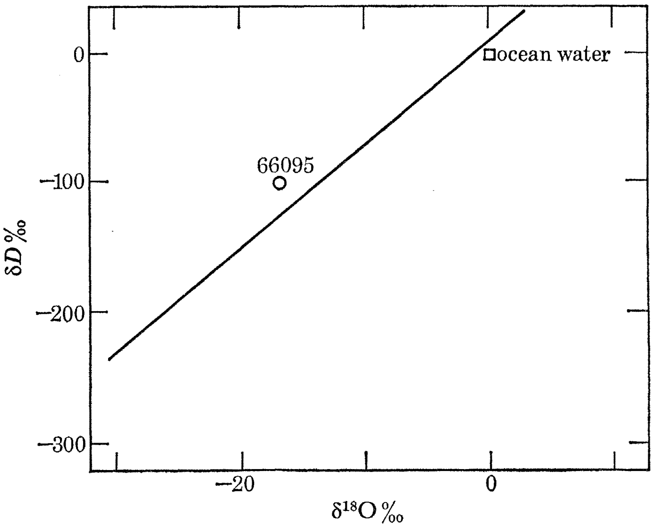

FIGURE 4. Isotopic composition of water extracted from the 'rusty rock' 66095 in relation to the locus of δD and $\delta^{18}O$ values of all terrestrial meteoric precipitation (snow, rain, etc.) which is depicted by the solid line (adapted from Epstein & Taylor 1974).

Both Padawer *et al.* (1974) and Leich *et al.* (1974) conclude that terrestrial contamination is at least partially responsible for these surface enrichments. Various possible sources of terrestrial contamination were indicated in the introduction. All the above authors attribute the observed fluorine contamination to bagging the samples in Teflon. Thirdly, Taylor, Mao & Bell (1973, 1974) have shown that the 'rust' in 66095 contains the metastable mineral akaganeite (βFeOOH) and significant quantities of chlorine. Akaganeite is believed to have formed from lawrencite ($FeCl_2$) originally present in the samples which is known to hydrate and oxidize very rapidly in a terrestrial environment.

In conclusion of this section the writer accepts the possibilities suggested by Friedman *et al.* (1974*b*) that some true lunar water may not be too different in isotopic composition from terrestrial water and that 'perhaps we should be more open-minded in trying to define lunar water'. Nevertheless the evidence cited above that the water in the lunar samples is probably entirely terrestrial contamination is too strong to be dismissed lightly.

^{18}O AND ^{30}Si ENRICHMENTS IN LUNAR SOIL

Studies by Epstein & Taylor (1970, 1971, 1972, 1973), Taylor & Epstein (1970*a*, *b*, 1973*a*, *b*), Clayton, Onuma & Mayeda (1971), Clayton, Hurd & Mayeda (1972, 1973), Clayton (1972, 1973), Javoy, Marette & Pineau (1973), Javoy & Fourcade (1974), Onuma, Clayton & Mayeda (1970), and O'Neil & Adami (1970) have shown that the ranges in $\delta^{18}O$ and $\delta^{30}Si$ values for whole-rock samples from the localities sampled on the lunar surface are amazingly small. Values of $\delta^{18}O$ range from about $+5.4$ to $+6.8$ ‰ and $\delta^{30}Si$ values range from about -0.5 to $+0.3$ ‰. Note that the $\delta^{18}O$ values quoted are based on the Pasadena definition of s.m.o.w. on which the results are systematically about 0.3 ‰ higher than those determined in Chicago. However all the whole-rock surface soil samples show slight enrichments in the heavy isotopes ^{18}O and ^{30}Si. This is due to very large heavy isotope enrichments in the surface layers of the agglutinates and grains that comprise the lunar soil.

These remarkable heavy isotope enrichments have been demonstrated by fluorine stripping experiments in which bulk samples are fluorinated at progressively higher temperatures releasing the oxygen for analysis in time and temperature fractions. A typical set of oxygen isotope results is illustrated in figure 5 (curve A). The $\delta^{18}O$ and $\delta^{30}Si$ normally correlate closely and enrichments up to $\delta^{18}O$ and $\delta^{30}Si$ values of about $+55$ and $+30$ ‰ respectively have been observed in the first fractions.

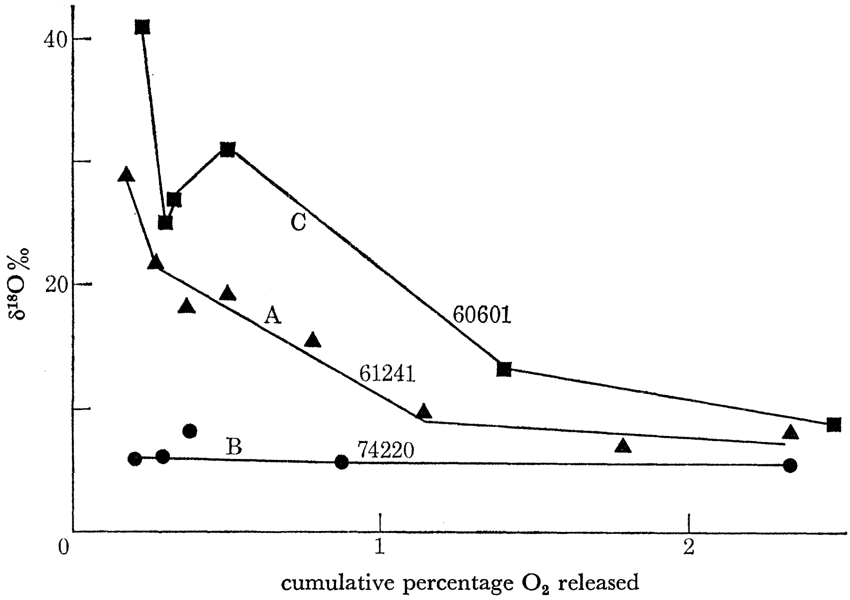

FIGURE 5. Typical oxygen isotope results of fluorine stripping experiments on lunar material.

All authors agree that these isotope enrichments must be related to the production of the amorphous coatings observed on the grain surfaces which are believed to result from vaporization of material from the grain surfaces and/or condensation of material on the grain surfaces. The vaporization of material on the lunar surface is thought to be due to bombardment by meteorites, micrometeorites, particles from the solar wind or solar flares and cosmic rays. Clayton, Hurd & Mayeda (1974) have suggested it is likely that these extreme isotope enrichments are in vapour condensates derived from lunar material volatilized by bombardment as described above and fractionated in the lunar atmosphere. Clayton *et al.* (1974) also calculate that a fraction enriched in the light isotopes ^{16}O and ^{28}Si amounting to about 1 % of the mass of the lunar regolith has been lost from the Moon during the course of its history. It is clear that these surficial heavy isotope enrichments are a crude indication of the duration of surface exposure or maturity of the soil.

Certain soils do not, however, exhibit enrichment patterns characterized by a simple monotonic decrease in $\delta^{18}O$ during fluorine stripping as described above. The Apollo 17 orange soil

74220 (Taylor & Epstein 1973) gives a unique fluorine stripping pattern (curve B in figure 5) with no surface enrichment in either ^{18}O or ^{30}Si. This is interpreted as indicating that the orange soil must have been exposed very recently at the lunar surface. A number of soils from the Apollo 16 and 17 landing sites exhibit somewhat irregular δ^{18}O patterns during fluorine stripping characterized by reversals in the normal simple monotonic decrease in δ^{18}O (e.g. curve C in figure 5). It is not certain at present whether these irregularities in the δ^{18}O curves are correlated with similar effects in δ^{30}Si enrichments because insufficient silicon analyses have been made over the critical regions. Taylor & Epstein (1973) attribute the irregularities in the δ^{18}O curves to the presence of adsorbed terrestrial water contamination which is depleted in ^{18}O (see above).

TABLE 1. δ^{18}O VALUES FOR SIEVED SIZE FRACTIONS OF LUNA 16 AND 20 SOIL

	size fraction			
sample	25 μm	25–40 μm	40–48 μm	weighted average
Luna 16	+6.73 ‰	+8.40 ‰	+5.10 ‰	+6.33 ‰
Luna 20	+6.17 ‰	+5.04 ‰	+7.19 ‰	+6.65 ‰

Some unpublished δ^{18}O values determined on very small samples of sieved size fractions of Luna 16 and 20 soils are given in table 1 (Beckinsale, Bowie & Durham 1977). The weighted average δ^{18}O values for the bulk samples are within the range of normal soils although there is appreciable variation in the different size fractions. One possible interpretation of these data is that each grain size fraction could have a different exposure history and that the soil at each locality is made up of grain size fractions each reflecting the increment which arrived from a specific impact vent.

TABLE 2. TEMPERATURES OF CRYSTALLIZATION OF LUNAR BASALTS DERIVED BY BOTTINGA & JAVOY (1975) USING OXYGEN ISOTOPE GEOTHERMOMETRY

sample	T/°C	sample	T/°C
10020	1116	12021	1061
10022	1147	12038	1009
10071	1159	12040	1062
10017	1178	12052	1065
10044	1088	12063	1130
10044	1078	12064	989
10047	1055	14053	972
10057	996	14072	937
10058	1114	14321	1115
12018	1110	15016	1110

GEOTHERMOMETRY

The principles of geothermometry based on the temperature dependence of the partition of ^{18}O between different crystalline phases in equilibrium are well established (Urey 1947), although experimental calibration of mineral geothermometers has proved very difficult. Recently Bottinga & Javoy (1975), have used a semi-empirical approach to the calibration problem and derived recalculated crystallization temperatures from most of the presently available oxygen isotope data for minerals from lunar basalts. Their results are summarized in table 2.

It can be seen that the temperatures derived by Bottinga & Javoy (1975) form a consistent group ranging from about 1000 to 1150 °C. These values appear to agree in general with experimentally determined liquidus temperatures. There is no evidence for sub-solidus exchange of oxygen isotopes leading to significantly lower apparent temperatures which supports the view that the lunar basalts cooled rapidly from high temperatures.

The origin of the Moon

Finally brief mention will be made of recent oxygen isotope studies by Clayton, Grossman & Mayeda (1973), Clayton, Hurd & Mayeda (1973) and Grossman, Clayton & Mayeda (1974) which are one of the most exciting contributions to lunar science. Clayton and co-workers have determined $\delta^{18}O$ and $\delta^{17}O$ values for terrestrial and lunar materials and the high temperature condensate aggregates in Allende and other chondrites. They find that when they plot $\delta^{18}O$ values against $\delta^{17}O$ values for lunar and terrestrial material the points fall close to a line with a slope of +0.5. Such isotope systematics would be expected from samples derived from the same primordial composition by ordinary chemical processes. The meteoritic high temperature condensate aggregates however are grossly enriched in ^{16}O and plot along a line with a slope of +1 in the $\delta^{18}O$ versus $\delta^{17}O$ diagram. Clayton, Grossman & Mayeda (1973) attribute the slope of the chondritic inclusion line to mixing of pure, or nearly pure, ^{16}O with normal solar system oxygen.

Whatever the origin of this contrast in oxygen isotope composition it is clear that at least in this respect the Earth–Moon system has a completely different origin from the high temperature condensates in the chondrites. This raises fundamental questions about the validity of the chondritic analogy in cosmology.

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