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ARGON STABLE ISOTOPE CONCENTRATIONS IN LUNAR REGOLITH

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Abstract: We performed stepwise heating experiments for determination of the two stable isotope ratios of argon fractions and total concentrations of the three stable isotopes ^{36}Ar, ^{38}Ar and ^{40}Ar in lunar regolith acquired from the Apollo 11, Apollo 12 and Apollo16 missions. Also the concentration of *in situ* formed radiogenic ^{40}Ar was estimated on the basis of known ages and potassium concentrations determined by isotope dilution method. The observed excess of ^{40}Ar concentration is interpreted to be due to variable (over geological time) flux of solar energetic particles which were implanted into the material at the Moon surface.

Keywords: Moon, regolith, argon origin, argon isotopes, radiogenic Ar, Sun

1. INTRODUCTION

Argon has three stable isotopes ^{36}Ar, ^{38}Ar and ^{40}Ar, hence two independent isotope ratios can be determined by means of mass spectrometry. However, since the publication of the paper by Suess (1949) on the origin of noble gases in Earth atmosphere, most investigators were focused on ^{38}Ar/^{36}Ar only and this ratio was studied in detail up to present, whilst the second ratio ^{40}Ar/^{36}Ar was usually neglected. Regarding the first ratio it is practically invariable for argon incorporated within the bodies of the Solar System with the exception of small meteoroids and Moon surface material being exposed to cosmic irradiation. In such materials neutrons are generated in the spallation process and this leads to generation of new nuclei. For example, a little fraction of ^{36}Ar is produced by beta decay of chlorine isotope formed in reaction ^{35}Cl(n,γ) ^{36}Cl and thus this process lowers the original ratio ^{38}Ar/^{36}Ar.

The monograph by Ozima and Podosek (2002) and the paper by Ozima *et al.* (1998) contain a detailed review on this argon isotope ratio in solar, Q-gases and terrestrial noble gases. Therefore we focused our efforts on measuring the concentration of argon isotopes extracted from lunar soil samples in a stepwise heating experiment and studying the variation of the ^{40}Ar/^{36}Ar ratio.

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2. MATERIALS AND METHODS

Three lunar regolith samples were acquired from NASA Astronautical Acquisition and Curation Office, Houston, TX, USA. The basic data about the samples are given in **Table 1**.

According to Allen and Todd (2007), the Apollo samples are handled and stored either under vacuum or in clean dry nitrogen gas at the Astromaterials and Curation Office of the NASA's Johnson Space Center.

We have determined potassium content for 3 lunar samples by the isotope dilution method as described by Halas (2001). Aliquots of about 7 mg of each sample were dissolved together with the spike (nearly pure ^{41}KCl solution, the reagent acquired from Oak Ridge National Laboratory, USA) in a platinum crucible and subsequently the mixtures obtained were analyzed on thermal ionization mass spectrometer. The amount of spike was selected in order to obtain mixtures with ^{39}K/^{41}K ratio in the range from 1 to 3. This condition assured the highest accuracy of the method, close to the precision of the isotope ratio determination (0.1%).

Argon for isotope analysis was extracted from about 5 mg aliquots of samples using an ultra high vacuum line equipped with dual vacuum crucible (Halas, 2007). The isotope analysis of individual argon fraction extracted was performed by means of static vacuum mass spectrometry. An example of mass spectrum is shown in **Fig. 1**. The

Table 1. *The list of investigated regolith samples*

Sample No.	Vessel No.	Mission	Location
10084.2000	9-14382	Apollo 11	mare-basalt region
12001.922	9-4079	Apollo 12	mare-basalt region
64501.228	9-19392	Apollo 16	highland

mass spectra, after small corrections for mass discrimination and background, enable us to calculate the two isotope ratios. The absolute concentrations of ^{36}Ar, ^{38}Ar and ^{40}Ar were determined by calibrating the sensitivity of mass spectrometer with known aliquots of ^{38}Ar. Calibration was performed prior to and after the experiment.

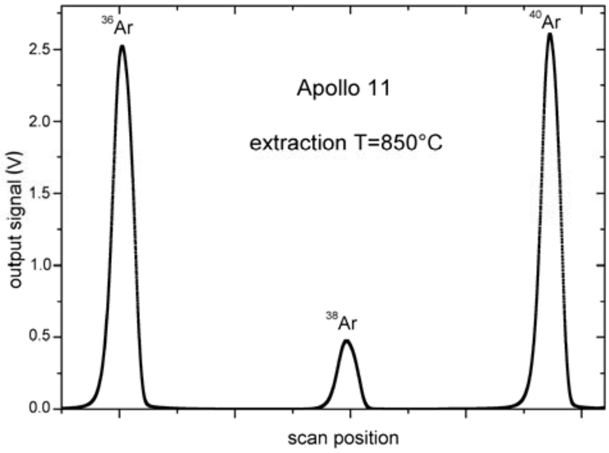

Fig. 1. *An example of mass spectrum recorded for argon extracted from lunar regolith.*

3. RESULTS

The results of argon isotope analysis are summarized in **Table 2**.

Using the average age of regolith for each location compiled by Dalrymple (1994), we estimated the absolute concentrations of radiogenic isotope, ^{40}Ar*, accumulated due to *in situ* decay of ^{40}K. By subtracting of ^{40}Ar* from total ^{40}Ar, we obtain the mass spectra of Ar implanted into regolith.

Results of calculated absolute concentrations and the corrected ^{40}Ar/^{36}Ar ratios are shown in **Table 3**. These ratios vary from 0.33 to 1.68 and they are representative for argon implanted into regolith by the solar wind and by ejecta during huge explosions or impacts on the Sun. In this way, we could reproduce the Ar isotope composition of the solar matter ejected to space. The ^{38}Ar/^{36}Ar ratio represents the isotope ratio of primordial argon which existed already in the presolar nebula, whilst the ratio ^{40}Ar/^{36}Ar of the solar argon must be highly affected by the radioactive decay of ^{40}K. Below we discuss the possible origin of the ^{40}Ar excess.

4. SPECULATIONS ON THE ORIGIN OF ^{40}Ar-RICH COMPONENT IN SOLAR GASES

In most experimental studies no significant variations of ^{38}Ar/^{36}Ar ratio was recorded so far. Basically, the same ratio is observed in the Earth atmosphere, meteorites and lunar regolith samples of various ages, from zero up to 4.5 Ga (Levine *et al.,* 2007; Berra *et al*., 2006). This is the evidence of the homogeneity of the presolar nebula from which all the bodies in the Solar System were formed. In contrast, we have observed highly variable ^{40}Ar content in the lunar regolith samples. The older regolith acquired by Apollo 11 and Apollo 16 missions contain larger amount of ^{40}Ar than the younger one obtained by Apollo 12, see **Table 3**. This result leads us to favor the idea that the older regolith acquired argon from a deep zone of the Sun located below the convection zone which is homogeneous with respect to both chemical and isotope composition due to continuous mixing. The thickness of the convection zone remains to be about 0.15 of the solar radius and it contains only 2% of the total solar mass.

The remaining mass of the Sun must contain identical ^{38}Ar/^{36}Ar ratio, but significantly higher ^{40}Ar/^{36}Ar ratio. Such deeply located solar matter could be ejected most likely during the bombardment of the Solar System bodies by unknown origin large bodies which produced

Table 2. *Yield of stable argon isotopes from regolith samples released in stepwise heating experiment.*

Sample and mass	T (°C)	t (min)	^{36}Ar (%)	^{38}Ar (%)	^{40}Ar (%)	^{38}Ar/^{36}Ar	^{40}Ar/^{36}Ar
Apollo 11	850	30	57.54	56.00	57.19	0.193	1.070
m = 4.78 mg	1200	30	23.23	23.36	23.41	0.200	1.085
	1250	35	5.16	5.45	5.22	0.210	1.088
	1250	45	3.47	3.71	3.50	0.213	1.088
	1300	40	6.53	6.98	6.59	0.213	1.086
	1300	180	4.06	4.58	4.09	0.225	1.086
Total yield (nmol/g)	-	-	19.396	3.855	20.883	-	-
Apollo 12	850	30	70.12	68.12	67.79	0.194	0.571
m = 4.97 mg	1250	40	25.80	27.17	27.81	0.210	0.636
	1250	130	3.18	3.80	3.42	0.239	0.636
	1250	130	0.90	0.91	0.98	0.200	0.640
Total yield (nmol/g)	-	-	11.214	2.233	6.619	-	-
Apollo 16	850	30	71.53	70.16	69.08	0.191	1.847
m = 4.80 mg	1250	40	23.71	24.69	25.73	0.202	2.076
	1250	60	1.38	2.49	2.58	0.203	2.072
	1250	150	2.41	2.66	2.61	0.214	2.071
Total yield (nmol/g)	-	-	12.141	2.357	23.215	-	-

Table 3. *Yield of stable argon isotopes from regolith samples released in stepwise heating experiment.*

Sample	Approx. age (Ga)	%K measured	$^{40}Ar^{*}$ (nmol/g) estimated	^{40}Ar (nmol/g) corrected	$^{40}Ar/^{36}Ar$ corrected
Apollo 11	3.6	0.107±0.005	2.13	18.75	0.967
Apollo 12	3.15	0.198±0.004	2.93	3.69	0.329
Apollo 16	3.9	0.116±0.005	2.78	20.43	1.68

maria on the visible surface of the Moon. Norman *et al.* (2006) have identified these enormous impact events to have taken place between 3.75 to 3.96 Ga ago. Also Sun could suffer of such cataclysmic impacts which could violate locally the whole convection zone and eject some matter from deeper zones of the Sun.

The following two mechanisms may explain higher $^{40}Ar/^{36}Ar$ isotope ratio in the deeper zones of the Sun:

(1) Elemental differentiation which may lead to higher concentration of heavier elements in the Sun centre. For this reason the nuclei of potassium can produce proportionally larger concentration of the radiogenic argon in the central zones than in the outer convection zone.

(2) Radioactive isotope ^{40}K decays faster by electron capture in plasma comprising fully ionized atoms and electrons, due to higher availability of electrons in dense plasma in comparison to unionized atoms in solid or melted minerals.

The first mechanism requires a significantly higher K concentration, at least one order of magnitude than that in the convection zone, which seems to be opposite to the concept of initially uniform Sun. However, no star remains chemically uniform with its age, because the central parts contain decreasing amount of H which is burned to He.

The second reason may be proven in future in some laboratory experiments and by theoretical calculation (cf. Takahashi *et al.,* 1987). The following condition of the plasma, which follows from the astrophysical models, must be taken into account: central density = 134 g/cm^3 and T = $14.6 \cdot 10^6$ K. Also a redistribution of the electron density around K^{19+} ions, due to the screening effect, is in favor to higher availability of electrons to be captured by ^{40}K nuclei.

5. CONCLUSION

In conclusion we may state that the surface material of the Moon reflects not only sunshine, but it also records the Ar isotope composition present in the Sun. Our study reveals huge variations of ^{40}Ar abundance. The implanted argon into lunar regolith may be derived from two diverse sites: the convection zone with $^{40}Ar/^{36}Ar \sim 0.33$ and from deeper zones of the Sun with that ratio being at least 3 times higher.

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