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^{40}Ar-^{39}Ar Dating: Investigation of some Technical Problems and its Application to the Deccan Trap Rocks and a Unique Meteorite from Antarctica

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In order to get more reliable ages and extend the application of ^{40}Ar-^{39}Ar dating for terrestrial and exterrestrial samples, some technical problems such as the examination of the efficiency of Cd-shielding to reduce the interference thermal neutron flux and the laser heating method were investigated. It has been reconfirmed that Cd-shielding is effective to reduce the interference ^{40}Ar produced by thermal neutron from potassium.

Further the laser heating method has been successfully developed in our laboratory and give reasonable ages for some age standard and Deccan Trap samples.

Application of ^{40}Ar-^{39}Ar dating method gives ages for Deccan Trap samples which range from less than 60 Ma to about 70 Ma. The clast in a unique meteorite from Antarctica gives a in ^{40}Ar-^{39}Ar age of about 490 Ma which corresponds to a degassing event and concordant with that of the host meteorite.

KEYWORDS: ^{40}Ar-^{39}Ar age, Cd-shielding, Laser heating, Deccan Traps, Meteorite, Antarctica

1. Introduction

Although the ^{40}Ar-^{39}Ar dating method is a modification of the K-Ar dating method, the ^{40}Ar-^{39}Ar method has a lot of merits over the conventional K-Ar dating method. However, in order to get a precise ^{40}Ar-^{39}Ar age, the correction for the interference Ar isotopes is significant. The production of interference Ar isotopes is controlled by the energy spectra of the neutron flux and only the fast neutron flux is required in an ideal case. To approach such a state, Cd-shielding has often been used to reduce the contribution of thermal neutron flux. It is important to check the efficiency experimentally and we have examined it through a series of our studies. Further, we have tried to develop the laser heating ^{40}Ar-^{39}Ar dating method in our laboratory in order to get an ^{40}Ar-^{39}Ar age from a mineral which fulfills the condition as dating materials.

In this paper, we report some recent developments in ^{40}Ar-^{39}Ar dating method in our laboratory and some examples of the application of ^{40}Ar-^{39}Ar dating for terrestrial and exterrestrial samples.

2. Experimental

Through the present studies, samples were wrapped in Al-foil in the case of powdered samples, but rock samples were cut as cylinders (~6 mm diameter). However, samples for laser heating were prepared as granules and 1-5 grains of each sample were kept in the holes (1.5 mm in diameter, 1.5 mm in depth) on the Al dishes. In the case of meteorite from Antarctica, sample chips with grain sizes of 1~5 mm were wrapped in Al foil. These samples were stacked together with age standard samples (either EB-1;91.4+/-0.5 Ma or MMHb-1; 520.4+/-1.7 Ma[1)]) in vacuum sealed quartz ampoules (10 mm in diameter and 70 mm long). CaF_2 and K_2SO_4 were also included to examine the correction factors for interference Ar isotopes produced by thermal neutrons.

Such samples were irradiated by JMTR with a fast neutron flux of (1-5) $\times 10^{17}$ n/cm^2. To reduce interference Ar isotopes, Cd-shielding (0.2 mm thick) was used.

Ar gas in these samples was extracted at the Radioisotope Center, University of Tokyo. Ar isotopes were measured mainly on the on-line noble gas mass spectrometer, VG-3600 (VG-Isotech), but some samples were measured on the noble gas mass spectrometer (VG-5400) at the Institute for the Study of the Earth's Interior, Okayama University. To extract Ar gas either a radio-frequency induction heater or the continuous argon ion laser, BeamLok 2060 (Spectra-Physics) was used. Blanks and interfering Ar isotopes produced by neutron irradiation from Ca and K were corrected to calculate an ^{40}Ar-^{39}Ar age, using the correction factors determined on the basis of the measurements of Ar isotopes for neutron-irradiated CaF_2 and K_2SO_4.

3. Results and discussions

3.1. Efficiency of Cd-shielding to reduce the production of interference Ar isotopes due to thermal neutron irradiation

In the ^{40}Ar-^{39}Ar method, the production of ^{39}Ar from ^{39}K by the reaction $^{39}K(n,p)^{39}Ar$ is required, but the other Ar isotopes produced by the neutron irradiation interfere the calculation of an ^{40}Ar-^{39}Ar age and desirable to be reduced as much as possible It has been known that Cd-shielding is effective to reduce the thermal neutron flux. Since the required reaction is most effective by fast neutron irradiation, application of Cd-shielding for ampoules is expected to reduce the production of interference Ar isotopes for ^{40}Ar-^{39}Ar dating. Its efficiency has been estimated by comparing the correction factors for monitored samples between those of Cd-shielding and non Cd-shielding samples by summarizing the data reported for JMTR.

The result indicates that there are no significant differences for correction factors for Ca-derived Ar isotopes such as $(^{36}Ar/^{37}Ar)_{Ca}$ and $(^{39}Ar/^{37}Ar)_{Ca}$. However, a clear difference is observed for $(^{40}Ar/^{39}Ar)_K$: those without Cd-shielding range from 0.03-0.02(average: about 0.12), while those with Cd-shielding range from 0.0025 to 0.03(average:0.015). Thus, it is clearly shown that the Cd-shielding is quite effective to reduce the interference ^{40}Ar isotope produced from potassium by thermal neutron irradiation.

3.2. Development of laser heating ^{40}Ar-^{39}Ar dating method

Laser heating ^{40}Ar-^{39}Ar dating method makes it possible to determine a very small sample including grain analysis and *in-situ* analysis. Hence, since the application of this method for volcanic rocks in early '80[2], this method has been widely used nowadays.

In 1993, we obtained an argon ion continuous laser (BeamLok 2060). By combining with a high sensitivity mass spectrometer (VG3600), we have tried to establish a system for laser heating ^{40}Ar-^{39}Ar dating method, and succeeded in getting reasonable ^{40}Ar-^{39}Ar ages.

Dating standard samples, EB-1 (two runs), LP-6 and MMHb-1 samples were measured by laser heating total fusion method. Stepwise heating method was tried for two SORI samples. Total fusion analysis for five grains of MMhb-1 yields the age of 508+/-24 Ma. Relatively large error for this measurement is originated from the estimation of J-value by the radio frequency induction heating method. This result agrees with those of previous analyses[4] within the 1 sigma error range.

Based on the J-value that is estimated from conventional ^{40}Ar-^{39}Ar dating method, 132.3+/-1.8 Ma was obtained for single grain LP-6 sample. Recommended age of LP-6 (127.7+/-1.4 Ma) doesn't include our result. However, Ingamell and Engells (1976)[3] reported this standard behavior as a mixture of two biotites of ages 121 and 136 Ma. Baksi et al. (1996)[4] indicated that the analyses of 1-2 flakes (~10-30 μg) of LP-6 with a laser system yield ages ranging from 105-130 Ma and flakes showing high (> 0.05) Ca/K ratios yield ages <115 Ma with >10% atmospheric contamination, whereas flakes with low Ca/K ratios gave ages >120 Ma and showed ~5% atmospheric contamination. In our results, LP-6 reveals the low Ca/K ratio of 0.011 (K/Ca =91.4). There is no confliction between the relatively old age and low Ca/K ratio for the single grain experiment of the LP-6.

Two EB-1 analyses yield relative old age of 102.3+/-2.6 and 100.7+/-4.4 Ma, respectively. As a reference age for this standard, 91.4+/-0.5 Ma was reported. Possible reasons of these old age might reflect the heterogeneity in the grains and/or recoil loss of ^{39}Ar during neutron irradiation. Such effect may be significant due to the small amount (<1mg) of the sample in the case of laser heating. However, further studies are needed to settle the problem.

Two SORI samples were dated by the stepwise heating method. Results of these analysis are shown in Figure 1. SORI-1 yields a total age of 91.8+/-1.4 Ma and a plateau age of 91.1+/-0.7 Ma. SORI-2 yields a total age of 92.5+/-1.5 Ma and a plateau age of 91.2+/-0.7 Ma. These results are consistent with the K-Ar and ^{40}Ar-^{39}Ar dating results of SORI samples analysed by some researchers of Geological Survey of Japan[5].

Thus, results of dating standard samples indicate that the precision and accuracy of laser heating ^{40}Ar-^{39}Ar dating method satisfy the required measurement level.

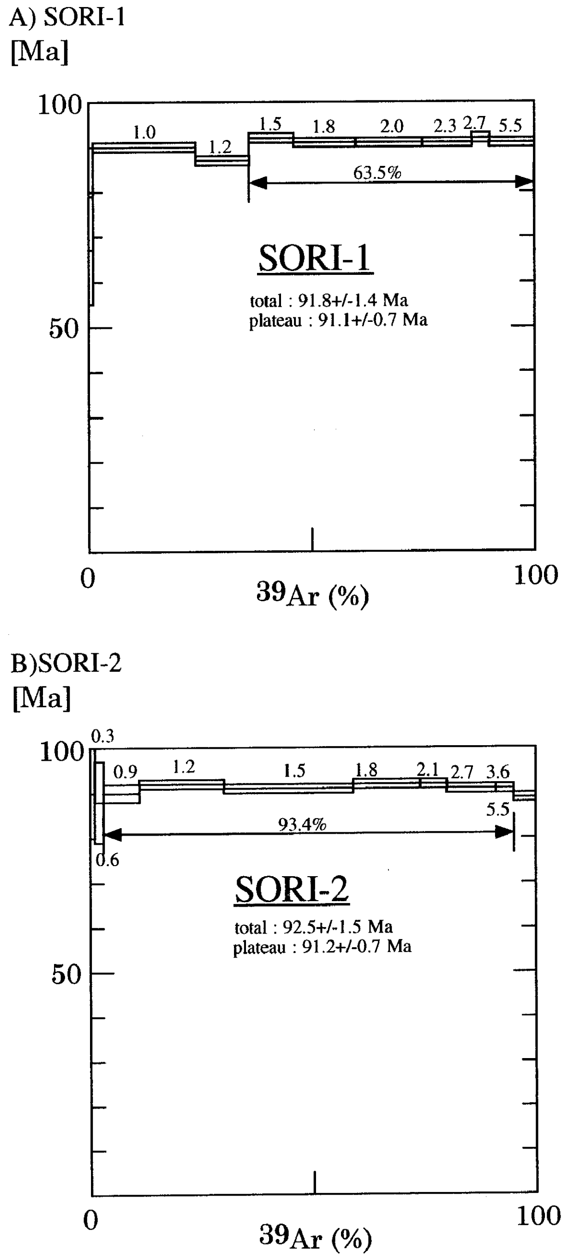


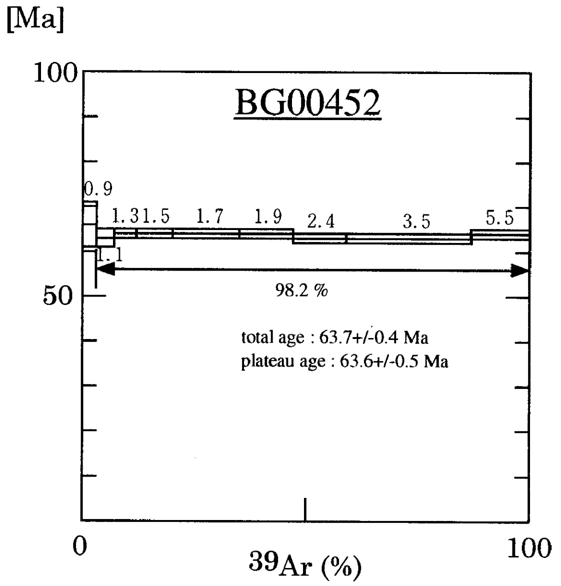
Figure 1 ^{40}Ar-^{39}Ar age spectrum of A) SORI-1 and B) SORI-2. Numbers on each steps indicate output power of continuous laser(in Watt).

3.3. ^{40}Ar-^{39}Ar ages of the Deccan Trap samples

The Deccan Traps cover the area of about 10^6 km^2 in the central part of India and the total volume of lavas is estimated to be more than 10^6 km^3, which is one of the largest volcanism on the Earth. The main volcanic activity of the Deccan Traps has been regarded to have occurred around 67-65Ma [6], but there are still controversies on the duration of the volcanic activity.

We have tried to examine it by measuring samples from different localities in the Deccan Traps by induction heating method, from where limited number of ^{40}Ar-^{39}Ar age data have been reported so far. Furthermore, three samples were dated by laser-heating method.

1) Bakatgarth



2) Girnar hill

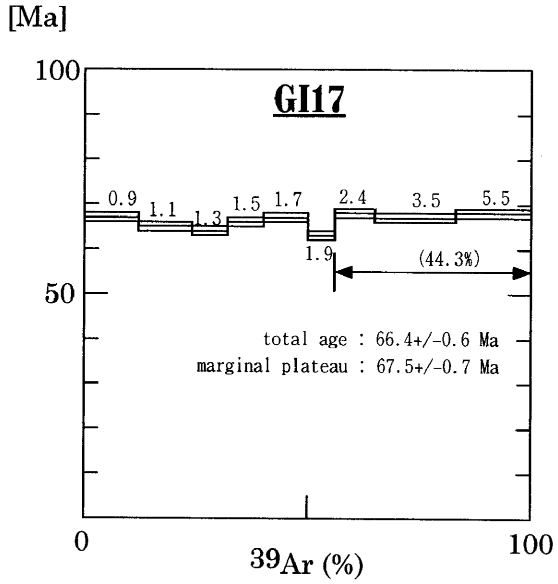


Figure 2 ^{40}Ar-^{39}Ar age spectrum of the Deccan Trap samples.

The results indicate that although many volcanic rocks show ^{40}Ar-^{39}Ar ages around 70-65 Ma, some samples show ^{40}Ar-^{39}Ar ages of around 63-62 Ma. Even ^{40}Ar-^{39}Ar ages of less than 60 Ma have been observed for a few flows from different localities. Such young ages cannot always been attributed to the Ar loss due to the alteration of samples and may correspond to some later volcanic activity in the Deccan Traps. Further, some samples from the eastern part of the Deccan Traps show ^{40}Ar-^{39}Ar ages of around 70 Ma, which are slightly older than the concentrated values of 67-65 Ma. On the basis of age spectra, they cannot always be attributed to excess ^{40}Ar and suggest a possibility that the volcanic activity of the Deccan Traps already started around 70 Ma.

For three samples from the Deccan Traps, biotites were separated and their ^{40}Ar-^{39}Ar ages were measured by the laser-heating method. One sample from the lamprophyre dike of the Bakatgarth along the Narmada river shows a good plateau ^{40}Ar-^{39}Ar age of 63.6+/-0.5 Ma (Figure 2), which agrees well with the plateau ^{40}Ar-^{39}Ar age of 63.2+/-0.1 Ma, obtained by the induction heating method. Biotite sample separated from granodiorite of the Girnar Hill, located at the northwestern part of the Deccan Traps, indicates a total ^{40}Ar-^{39}Ar age of 66.4+/-0.6 Ma, which agrees well with the K-Ar age of 67.0+/-1.3 Ma reported before [7]. The sample shows a pseudo-plateau ^{40}Ar-^{39}Ar age of 67.5+/-0.7 Ma(Figure 2). The other sample from Bombay indicates a pseudo-plateau ^{40}Ar-^{39}Ar age of 63.9+/-0.5 Ma with a total ^{40}Ar-^{39}Ar age of 66.0+/-1.2 Ma. For this sample, total ^{40}Ar-^{39}Ar age of 88 Ma and an apparent isochron age of 72.0+/-6.9 Ma have been obtained [8]. The reason about the large difference from the present result is not clear, but the sample's alteration might have caused such difference reflecting different experimental conditions.

Thus, present results suggest a possibility that the total length of the volcanic Deccan Traps might have been longer than the one which is commonly regarded.

3.4. ^{40}Ar-^{39}Ar age of a H-type clast included in a shocked L-chondrite from Antarctica

Among a lot of chondrites of meteorite collections of NIPR (National Institute of Polar Research) from Antarctica, there are a few unique meteorites which include different type of igneous inclusions from that of the host meteorite [9].

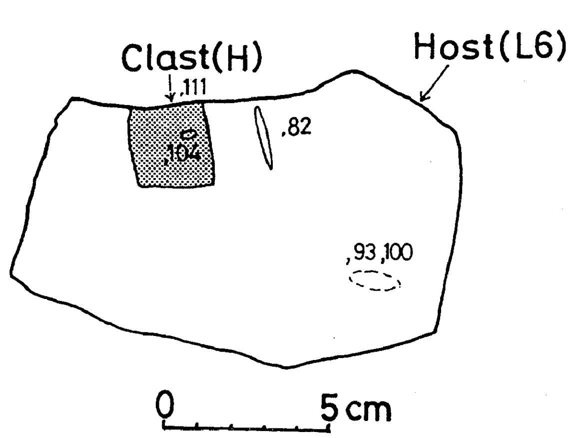


Figure 3 Schematic sketch of the original specimen of Y-75097. In the present study, the results for portions 104 (clast) and 82 (host) are reported.

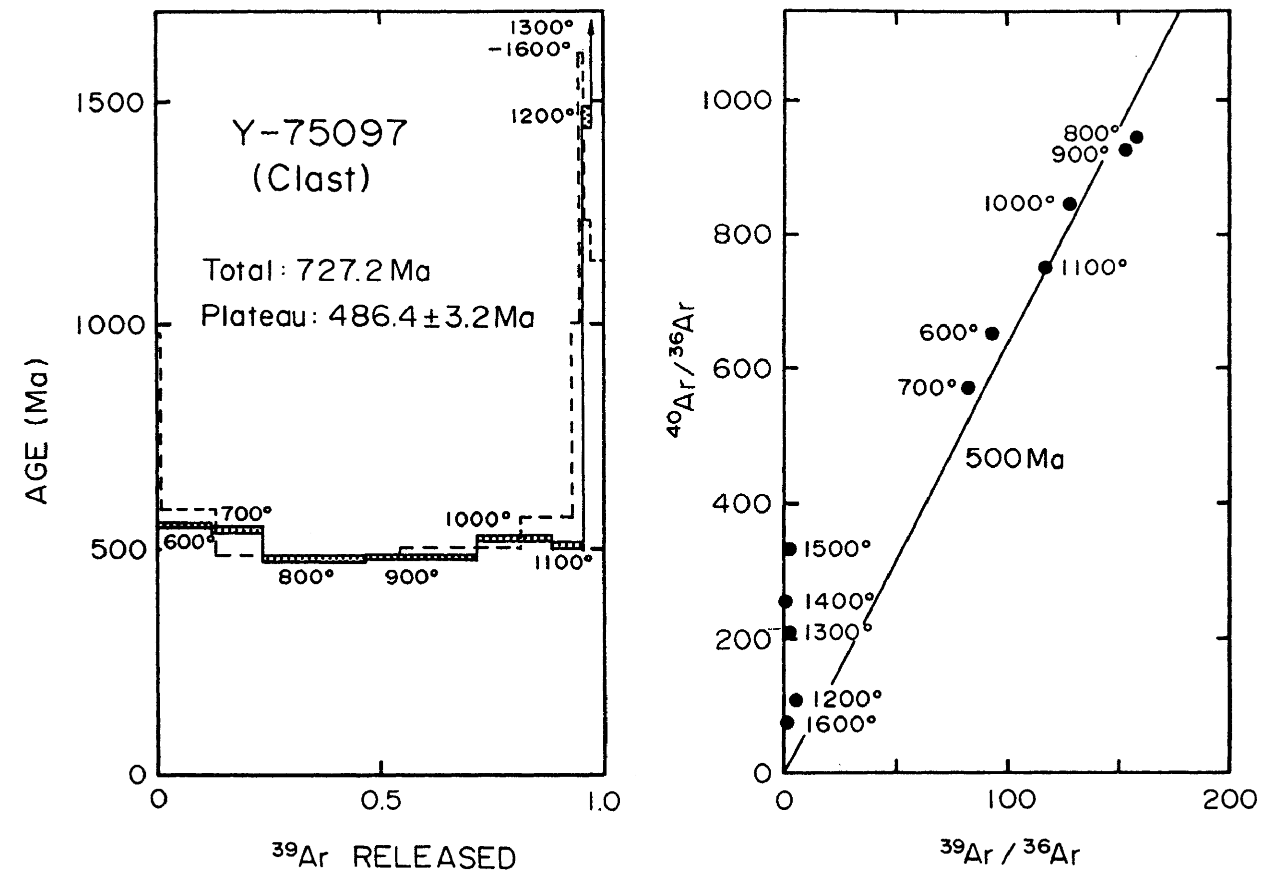


Figure 4 The ^{40}Ar-^{39}Ar age spectrum and the ^{40}Ar/^{36}Ar-^{39}Ar/^{36}Ar isochron plot for the sample Y-75097, 104 (clast). The dotted line in the left figure indicates a spectrum obtained for the sample Y-75097, 82 (host).

Yamato 75097 (Y-75097) is a L6 chondrite which contains a H-like clast and has signs of shock event (Figure 3). For this sample, ^{40}Ar-^{39}Ar ages were determined for both the clast and the host chondrite.

The result is shown in Figure 4, where the ^{40}Ar-^{39}Ar age spectrum and the ^{40}Ar/^{36}Ar-^{39}Ar/^{36}Ar isochron plot for the clast are drawn. As shown in this figure, the clast indicates a U-shaped pattern with a plateau age of about 490 Ma at 800-900℃. Similar plateau ^{40}Ar-^{39}Ar age is observed for the host meteorite, whose ^{40}Ar-^{39}Ar age spectrum is drawn by the dotted line in the same figure. In the lower and higher temperature fractions, ^{40}Ar-^{39}Ar ages are higher than the plateau age for both the clast and the host, which indicate the presence of inherited ^{40}Ar in these portions. These results suggest that both the host and the clast lost most of radiogenic ^{40}Ar about 490 Ma. Such ages have often been observed in shocked L-chondrites [10]. Although both the host and the clast recorded the age of an event of probably shock, the inherited ^{40}Ar in the higher temperature fractions remains in the clast more abundantly than in the host meteorite. This indicates the difference in the degree of degassing due to a shock between the host and the clast, probably reflecting the difference of their textures. Such point is also supported by an evidence that the clast retains the radiogenic ^{129}Xe due to the decay of ^{129}I (half-life: 17 m.y.), which should have been established before 4.0 Ga, but not found in the host meteorite [11].

Thus, based on ^{40}Ar-^{39}Ar analyses, the characteristics of Ar degassing during a shock for a meteorite or its present body and inclusion can be clarified to some extent.

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