K-Ar Age Detwermination of a Lava Stalagmite in Manjang Cave Jeju Island Korea

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INTRODUCTION

THE K-AR METHOD of age determination is commonly used to date rocks from Pleistocene volcanoes in Japan (e.g. Kaneoka *et al.* 1980, Itaya *et al.* 1984, Shimizu *et al.* 1988, Itaya *et al.* 1989). However, there are still many problems with K-Ar dating of the young volcanic rocks, as reviewed by Itaya and Nagao (1988).

Jeju island is a young volcano of which the geology and petrology have been studied by many authors (e.g. Lee 1966, Kim 1969, Won 1975, 1976, Lee 1982a,b). Recently, K-Ar ages of rocks from the volcano were reported by Won *et al.* (1986), suggesting that the volcano is Middle to Late Pleistocene in age. There are more than 50 lava tunnels on Jeju island, including some which are more than 100 m long such as Kimnyong Sa gul (cave), Jeongbang gul and Manjang gul. The speleology and environmental science of these lava tunnels have been investigated (e.g. Sawa 1987, Hong 1987) but their age of formation is not known.

This paper discusses problems experienced in dating young volcanic rocks and then the age of the formation of the lava tunnel, Manjang gul.

OUTLINE OF GEOLOGY

Jeju island is situated about 90 km south of the southernmost tip of the Korean Peninsula. It has an elliptical plan 80 × 40 km, of which the long axis trends WSW (Fig. 1). Landforms on the island are closely related to the volcanism which built the island; they can be divided topographically into the lava plateau, the shield-shape Halla volcanic edifice and the parasitic cones. The lava plateau is composed of voluminous, gently dipping basaltic lava flows which extend to the coast region. Many lava tunnels have been found within these lava flows. The shield volcano occupies the central part of the island. More than 360 parasitic cones lie along the long axis of the island. The process of growth of the island has been grouped into four stages on the basis of geological and topographical features, as follows (Won 1975, Lee 1982a): (1) the first stage which formed the base of the island about 100 meters below sea level; (2) the second stage which formed the lava plateau of the island; (3) the third stage which formed the Halla volcanic edifice in the central part of the island; (4) the fourth stage which formed the parasitic cones. Won *et al.* (1986) has reported K–Ar ages of 0.75 Ma for the Sanbangsan trachytes of the second stage and 0.025 Ma for the Backlockdam trachytes of the third stage.

Manjang gul is a lava tunnel located in the north-east part of Jeju island, near Kimnyong, which has developed within the Pyeosunri lava flow belonging to the second stage of island growth (Fig. 1). The Pyeosunri flow is a low viscosity basalt flow. The tunnel has been ranked as the world's longest with a total length of 13,269 m; the height of the roof is up to 25 m at the part with several

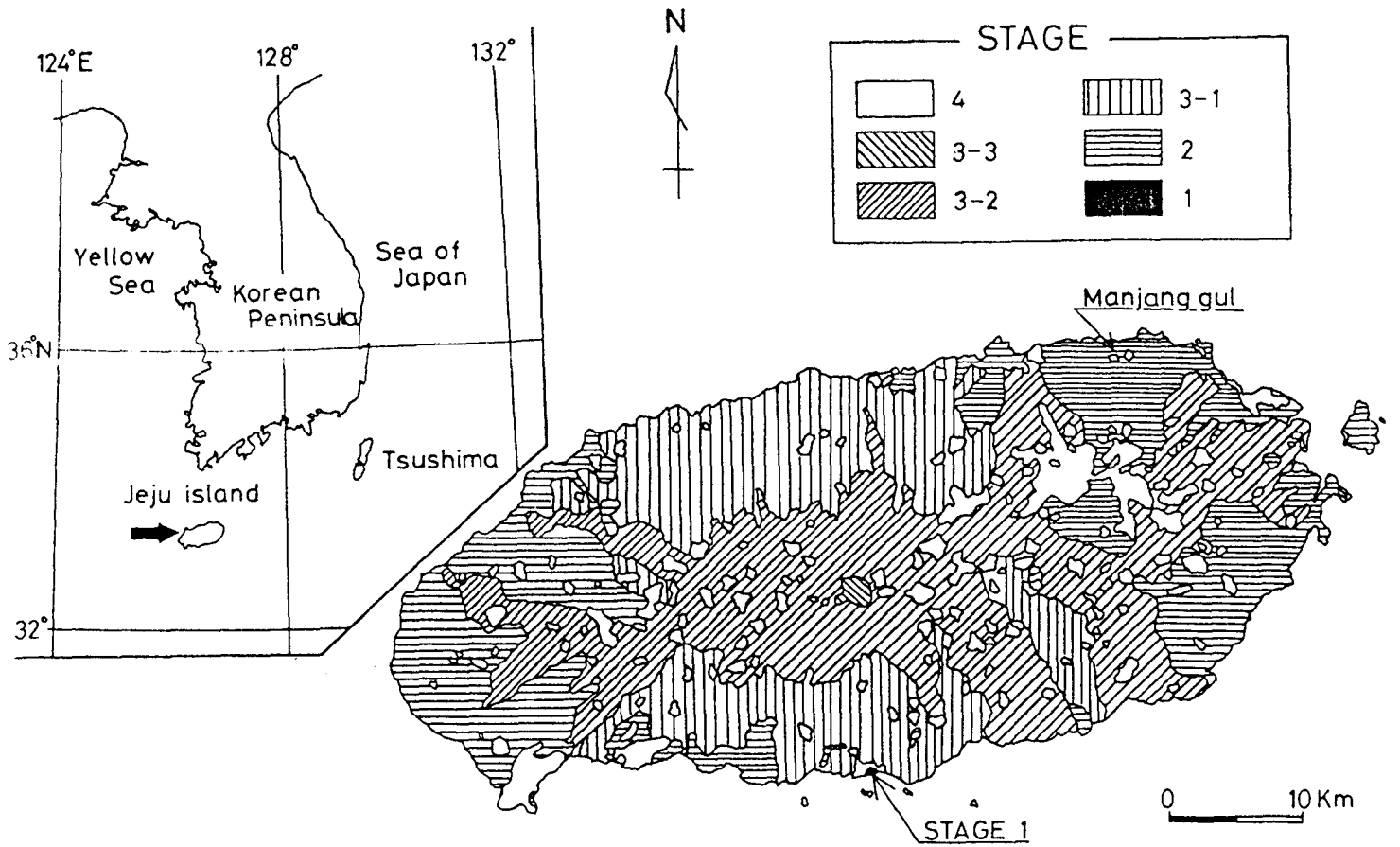

Fig. 1. Simplified geological map of Jeju island showing major stratigraphic divisions (after Lee 1982a). The volcanism of Jeju island has been grouped into four stages: 1, 2, 3 and 4. The third stage is also divided into three sub-stages (see text).

floors. The tunnel contains a twin lava stalagmite taller than 8.9 m (Sawa, 1988).

Two rock samples for K–Ar dating were collected from Manjang gul; Sawa61–102 from the twin stalagmite and Sawa62 from the floor near the stalagmite. These basalts have a phenocryst assemblage of olivine + augite + plagioclase. Sawa61–102 has well crystallized groundmass and Sawa62 has glassy groundmass which has been oxidized, as seen in the clinker part of the lava flow.

ANALYTICAL PROCEDURE

The rock samples were crushed and sieved to take 32–60 mesh size fraction for the argon analysis. Magnetic materials such as oxide minerals and non-magnetic phases such as plagioclase phenocrysts were removed from the fraction. A portion of the 32–60 mesh size fraction was ground using an agate mortar, and the resulting powder was analysed for potassium.

Potassium analysis was carried out by flame photometry using a 2000 ppm Cs buffer. Powder samples were decomposed for analysis by flame photometry with HF and HNO_3 treatment in a teflon beaker. Multiple runs of two chemical standards (JG-1 and JB-1) indicate that the accuracy and reproducibility of this method were within 2%. An average value of duplicate runs was used in the age calculation.

Argon was analysed on a 15 cm radius sector type mass spectrometer with a single collector system at Okayama University of Science, designed and constructed by Nagao and Itaya (1986), using an isotopic dilution method and a ^{38}Ar spike. Calibration of the ^{38}Ar

spike is accurate to within about 1% (Nagao *et al.* 1984, Itaya and Nagao 1989). The sample was wrapped in aluminum foil, preheated for about 24 hr at 180–200°C in a vacuum, and the argon was extracted at 1500°C in an ultra-high vacuum line (atmospheric ^{40}Ar blank was less than 2×10^{-9} ccSTP). Clean-up of reactive gases was done by a Ti–Zr scrubber.

Checking of mass discrimination of the mass spectrometer should be done very carefully in dating young volcanic rocks. The mass discrimination factor depends on argon amount introduced into the mass spectrometer (see Appendix). Therefore, the mass discrimination factor was determined using the calibration curves given by analyses of several different amounts of atmospheric argon. Sample argon was analysed in the amount of $1.0 \pm 0.5 \times 10^{-7}$ ccSTP for ^{40}Ar and its isotopic ratio was calibrated using the mass fractionation factor determined by the calibration curve (see Appendix).

K–Ar AGES OF ROCKS

Table 1 shows K–Ar age data of the two rock samples collected from Manjang gul; ^{36}Ar and ^{40}Ar contents of each specimen are also tabulated to compare their non-radiogenic argon content. Calculations of age and error followed the method described by Nagao *et al.* (1984) and Nagao and Itaya (1988). We first dated these rocks without careful checking of the mass discrimination. The results showed very poor reproducibility. The age data in Table 1 which were dated with careful checking of the mass discrimination show good reproducibility. However, these two rock samples have large differences in age with a large error. The older (Sawa62) has extremely high non-radiogenic argon content

Table 1. K-Ar age data of basalts from Manjang gul in Jeju island. $^{40}Ar^{R}$ shows radiogenic ^{40}Ar. Physical constants used in age calculation are after Steiger and Jäger (1977)

Number of specimen	Potassium (wt. %)	^{36}Ar	^{40}Ar ($\times 10^{-8}$ ccSTP/g)	$^{40}Ar^{R}$	Age (Ma)	$^{40}Ar^{R}$ (%)
Sawa61-102	0.81 ± 2	0.0288 ± 3	8.65 ± 8	0.13 ± 13	0.042 ± 41	0.7
		0.0315 ± 4	9.45 ± 9	0.16 ± 14	0.049 ± 46	1.5
Sawa62	0.91 ± 3	0.3339 ± 40	100.15 ± 94	1.5 ± 1.5	0.42 ± 42	1.6
		0.3341 ± 39	99.84 ± 92	1.1 ± 1.5	0.32 ± 42	1.1

DISCUSSION

Itaya *et al.* (1984) and Itaya and Nagao (1988) have shown that Quaternary volcanic rocks in Japan had 5×10^{-8} to 1×10^{-5} ccSTP/g of ^{40}Ar, although the estimated radiogenic argon was less than 10^{-7} ccSTP/g. This shows that the primary argon content of the rocks has marked variation. The variation is mainly due to the variability of argon content in the magma. Itaya and Nagao (1988) and Itaya *et al.* (1989) reported that glass-bearing lava samples have three to ten times as much non-radiogenic argon as glass-free samples from the same lava flow. They proposed that the present argon, except for the radiogenic one, in the rocks represents the argon trapped during various stages of a series of argon-release processes during the cooling of the flowing lava, not in equilibrium with atmospheric argon. Sample Sawa62 with extremely high non-radiogenic argon is from the clinker part of the lava flow. As the clinker part often has significantly higher non-radiogenic argon content than the massive part of the lava flow (Shimizu 1987), sample Sawa62 which has high non-radiogenic argon content could have experienced comparatively minor argon release during the cooling of the lava.

The primary isotopic ratio of argon in volcanic rocks is not always that of atmospheric argon, 295.5 for 40/36 ratio and 0.188 for 38/36 ratio, as reviewed by Itaya and

Nagao (1988). This would impose a constraint on the validity of the K–Ar age determination method for young volcanic rocks, and means that it is essential to check all samples with primary isotopic ratios different from the atmospheric values.

Itaya and Nagao (1988) recognised three situations whereby excess or depleted argon may be derived: (1) depleted ^{40}Ar fractionated from atmospheric argon isotopes observed commonly in island arc volcanic fields; (2) excess ^{40}Ar in quenched rim of submarine pillow basalt lava from hot spot and ridge fields; (3) excess ^{40}Ar in coarse grained quartz and plagioclase phenocrysts in andesite and rhyolite lavas. They also proposed a correction method for the age data for the mass fractionated type of rocks which involved analysing the primary argon 38/36 ratio by using a mass spectrometer for the rare gas analysis, in which ^{38}Ar spike has not been introduced.

There is a possibility that the rocks from Manjang gul have experienced the mass fractionation process from atmospheric isotopes if they have the isotopes different from the atmospheric values as they have no coarse grained phenocrysts and no quenched part in the lava flow.

Itaya and Nagao (1988) and Itaya *et al.* (1989) discussed the mass fractionation stage in rocks and proposed that the fractionation could take place by magma–sea water interaction in the shallow reservoir within the Earth's crust, and not during the argon release process in the eruption-cooling stage of the magma. This shows that the rocks have the same primary argon isotopes even though they have different non-radiogenic argon contents trapped in the argon release process. It follows that if the two samples, Sawa61–102 and

Sawa62, have undergone the same mass fractionation but different argon release processes, both samples may still have the same content of primary argon isotopes as observed in the study of two types of rhyolite/andesite lava samples from Aso volcano in southwest Japan (Itaya and Nagao 1988) and from Takahara volcano in northeast Japan (Itaya *et al.* 1989). To check this possibility for these two samples, the following calculations were done.

For young K–Ar age dating, the following approximate equation to calculate the age can be used (Nagao *et al.* 1984):

$$t(\mathrm{Ma}) = 0.258 \times {}^{40}\mathrm{Ar}^{\mathrm{R}}(\mathrm{ccSTP/g})/\mathrm{K}(\mathrm{wt.\%}) \qquad (1)$$

where $^{40}Ar^{R}$ and K are radiogenic ^{40}Ar and potassium contents in the sample, respectively. $^{40}Ar^{R}$ is given by the following equation:

$$^{40}\mathrm{Ar}^{\mathrm{R}} = {}^{40}\mathrm{Ar}^{\mathrm{S}} - {}^{(40/36)}\mathrm{p} \times {}^{36}\mathrm{Ar}^{\mathrm{S}} \qquad (2)$$

where $^{40}Ar^{S}$ and $^{36}Ar^{S}$ are ^{40}Ar and ^{36}Ar contents in the analysed sample, respectively, and $^{(40/36)}p$ is the primary $^{40}Ar/^{36}Ar$ ratio of the sample, of which the value was assumed to be 295.5 in conventional age calculation.

If the two samples, Sawa61–102 and Sawa62 have the same primary $^{40}Ar/^{36}Ar$ ratio that differ from 295.5, we can calculate the value in the following steps. We can take two simple equations for the age against the primary $^{40}Ar/^{36}Ar$ ratio using the ^{40}Ar, ^{36}Ar and potassium contents of Sawa61–102 and Sawa62 in Table 1. As each sample has two analytical data for the argon, four sets of simple equations are available. By solving independently each set of equations, the simple and the simultaneous equations, we take four sets of values for the age and primary $^{40}Ar/^{36}Ar$ ratio. The results showed

that the variations of the age and the ratio were 0.001–0.018 Ma and 300.0–298.7, respectively. The calculated age is too young because these rocks are from the second stage of volcanism in Jeju island which is older than 0.03 Ma (Won *et al.* 1986). It follows that the working hypothesis mentioned above is incorrect for these two samples, suggesting that the primary $^{40}Ar/^{36}Ar$ ratio was different between these two samples.

Generally, the sample with low non-radiogenic argon content, in which the argon has been well released on cooling, has a higher reliability for K–Ar age dating (Itaya *et al.* 1989). From this point of view, Sawa61–102 with lower non-radiogenic argon content is better for the K–Ar age dating, and its age of 0.04–0.05 Ma can be used to estimate the formation age of Manjang gul.

To discuss further the discordant ages of the two rock samples requires independent analysis of $^{38}Ar/^{36}Ar$ ratio for each sample by using a mass spectrometer for the rare gas analysis, as proposed by Itaya and Nagao (1988) and carried out by Takaoka *et al.* (1989). Further work is planned on samples of basalts collected systematically from Manjang gul to clarify the argon release processes in cooling lava during the formation of the tunnel.

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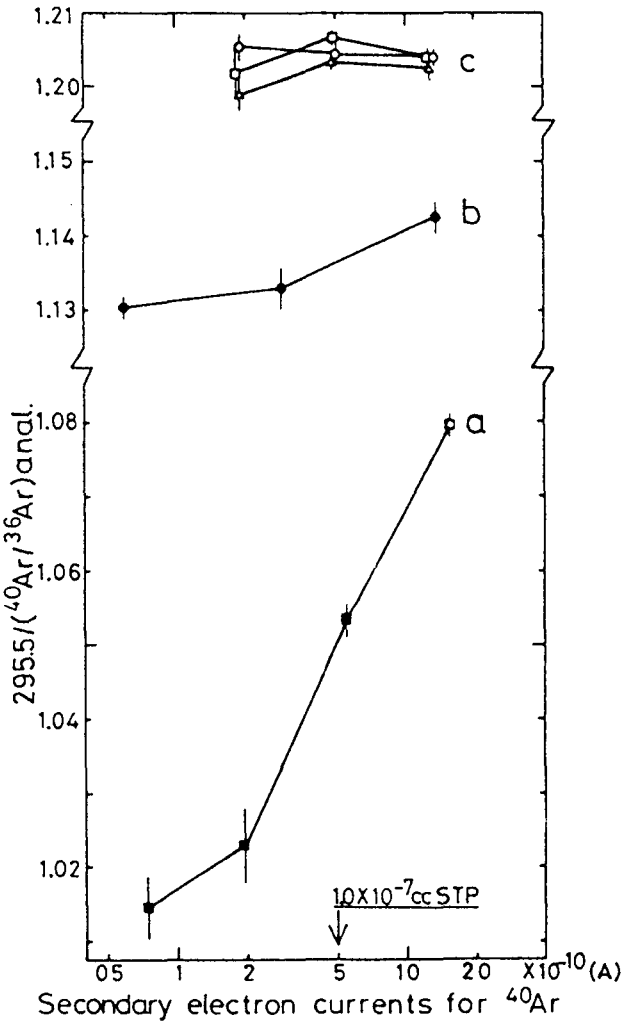

Fig. A1. Diagram showing the relationships between the mass discrimination factor ($295.5/^{40}Ar/^{36}Ar$ ratio analysed) and the amount of atmospheric argon introduced into the mass spectrometer in three differently aligned/adjusted conditions of mass spectrometer (see text).

APPENDIX

Since young geological materials have a lower $^{40}Ar/^{36}Ar$ ratio close to the atmospheric one, 295.5, the analytical precision of the argon isotopic ratios affect directly the reliability of dating. The mass spectrometer always has its own mass discrimination. Therefore, the analysed isotopic ratios of sample argon are calibrated by using the mass discrimination factor obtained in analysis of atmospheric argon. The mass discrimination changes easily with the condition of the mass spectrometer, and varies with different amounts of argon introduced into the mass spectrometer. Fig. A1 shows the relationships between the mass discrimination factor ($295.5/^{40}Ar/^{36}Ar$ ratio analysed) and the amount of atmospheric argon introduced into the mass spectrometer in the three differently aligned/adjusted conditions, a, b and c. These relationships also depend on the proficiency of the analytical technique, such as the taking of a series of mass spectrum (Itaya and Nagao 1989). Thus, K–Ar dating of young volcanic rocks requires proficiency in the analytical technique and the careful checking of the mass discrimination of mass spectrometer before analysis of unknown samples.

The best calibration method of analysing isotopic ratios of sample argon is to use the mass discrimination factor obtained from the same amount of atmospheric argon as the sample argon. Unfortunately, the present system has yet to be equipped so that the argon amount can be continuously controlled. Therefore, we have applied the following conventional calibration method for dating: (1) taking the relationships shown in Fig. A1 using several different amounts of atmospheric argon, and (2) analysing the argon sample and calibrating the isotopic ratio by the mass discrimination factor estimated from the calibration curve in Fig. A1. This procedure has been applied to K–Ar dating of the Middle Pleistocene Takahara volcano in central Japan, where the K–Ar ages of individual flows of the well defined succession were successfully determined (Itaya *et al.* 1989). We are now dating young materials in the conditions such as b or c in Fig. A1. The error is less than 0.5% in each isotope analysis using atmospheric argon. However, the error for calibration of the isotopic ratio of sample argon should be bigger than 0.5% because of the uncertainty of the calibration curves as seen in duplicate analyses on the condition c in Fig. A1. So we took 1.0% of the error value in the calculation of this paper. The numerical approach of a series of calculations for argon analysis has been done in detail by Nagao *et al.* (1984), Nagao and Itaya (1988) and Itaya and Nagao (1989).