Japan Geoscience Union Meeting 2015
(May 24th - 28th at Makuhari, Chiba, Japan)

SGC51-01 Room:A04 Time:May 24 15:15-15:30

^{40}Ar/^{39}Ar age of Haedean zircon

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Analyses of ages and closure temperatures using SHRIMP and LA-ICPMS U-Pb system of zircon, K-Ar system of hornblende and micas, and fission track method in zircon and apatite in the same rock sample are the standard techniques in thermochronology. Since zircon has no potassium, it has rarely been used in K-Ar geochronology. After intensive microscopic observation and electron microprobe analyses of Haedean zircons from the Labrador and Acasta gneiss, fine grain muscovite and other minerals as well as fluid inclusions are recognized. The presence of such inclusions suggests a possibility of a secondary event, and it might have caused disturbance in U-Pb system in the zircons, implying the zircons may be older. Utilizing susceptive characteristics of K-Ar system in a secondary hydrothermal event, laser step heating ^{40}Ar/^{39}Ar method was applied on the individual zircon grains to investigate the timing of formation of such inclusions in the zircons.

During laser step heating experiment, unirradiated Haedean zircons showed little release of ^{40}Ar below 1000^{o}C. However, without exception, they released large amount of ^{40}Ar (an order of 10^{-7} ccSTP/g) above 1000^{o}C in spite of a fact that zircon has no potassium. This suggested that either the zircons have excess argon trapped or a potassium phase. In ^{40}Ar/^{39}Ar experiment, the released argon isotopes were approximately 10^{-13} ccSTP (^{39}Ar) and 10^{-10} ccSTP (^{40}Ar) from a grain of 400 microgram. The volume of ^{39}Ar was very small, and the relative error was large. However, one of Labrador zircon gave 4.39 ± 0.34 Ga at a fusion step after 1000^{o}C, and the ^{39}Ar fraction was over 70% of the total release. The age could be caused by an excess argon during initial stage of zircon formation or a secondary event. Because of localized nature of excess argon, it tends to produce various ages in the same area rather than a uniform age. The facts that the similar age was observed in another grain, and that the fraction is above 1000^{o}C suggest that the age may have been preserved since the formation of the zircon. The results from other area will be discussed together.

Keywords: Haedean zircon, ^{40}Ar/^{39}Ar age

SGC51-02 Room:A04 Time:May 24 15:30-15:45

Two regional metamorphic belts in assemblages of subduction-related orogens in Wales-England, U.K.

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Cryogenian-Devonian orogenic belt in the British Isles consists of accretionary complex, arc-related volcano-plutonic belt with regional metamorphic belt that have formed in relation to an oceanic-plate subduction in the Iapetus Ocean. This subduction-related orogeny plays an important role in understanding of the geotectonic history of the pre- Ordovician orogenic belt in Wales-England.

Wales-England consists of ca. 677-450 Ma calc-alkaline volcano-plutonic complexes and ca. 608-539 Ma accretionary complexes, suggesting that the subduction-related orogeny continued for ca. 200 My. Four metamorphic units related to this orogeny exist in northwest Wales. However, their metamorphic ages, except for one unit, have not been constrained yet. Therefore, we performed U-Pb dating on detrital zircon in psammitic schists and K-Ar dating on phengite in pelitic/mafic schists from the four metamorphic units in order to constrain their sedimentary and metamorphic ages, respectively.

The K-Ar analysis was carried out at Okayama University of Science, and the U-Pb ratio was determined with LA-ICP-MS at the Kyoto University. Three metamorphic units give K-Ar ages of 578-545 Ma, which overlaps the Ar-Ar age of 560-550 Ma reported by a previous work. During this metamorphic event, accretionary complex and calc-alkaline volcano-plutonic complexes also had been formed. On the other hands, one metamorphic unit, called as New Harbour Group, exhibits a younger K-Ar age of 474±9 Ma. The youngest U-Pb age of detrital zircons from the New Harbour Group is 520±31 Ma. These ages suggest that the New Harbour Group formed in the regional metamorphic event different from that of the above three units. Calc-alkaline igneous rocks of ca. 480-450 Ma exist in Wales-England, which coincide with the second metamorphic event. Therefore, we conclude that at least two regional metamorphic events occurred in the geotectonic history of Wales-England.

Keywords: British Isles, Subduction-related orogeny, Regional metamorphic belt, K-Ar phengite ages

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SGC51-03 Room:A04 Time:May 24 15:45-16:00

Ar/Ar Geochronological analysis of Paka volcano, northern Kenya Rift

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The northern Kenya Rift constitutes one of the most prominent examples of young volcano-tectonic processes in a continental rift system. However, the limited amount of geochronologic information on different generations of lava flows and associated eruptive centers, fissures, and extensional faults has been a major problem in assessing the degree of activity in the inner trough of the rift. Here we report on 30 new ^{40}Ar/^{39}Ar ages of lava flows associated with the evolution of the composite Paka caldera-shield volcano in the northern sector of the Kenya Rift.

Paka is a composite caldera-shield volcano. It rises ˜650 m above the rift floor and covers an area of ˜280 km^2 with lava flows and pyroclastics of trachyte, mugearite or basalt (Dunkley et al, 1993). Dunkley et al. (1993) suggested that the principal volcanic eruptions on Paka occurred at 0.4 Ma, 0.2 Ma and 10 ka based on their four ^{40}Ar/^{39}Ar age determinations. The relationship between the pristine morphology of the summit crater and associated fault systems parallel to the rift orientation suggests that the volcano-tectonic activity at Paka is intimately associated with deformation processes in the inner rift trough, the youngest sector of volcano-tectonic processes in the rift. Thus, knowledge of the timing of volcanic eruptions and the generation of faults is crucial for a rigorous assessment of the youngest tectono-magmatic activity along the volcano-tectonic axis of the northern Kenya Rift.

Thirty-two samples were collected, mostly from lava flows or pyroclastic deposits, for ^{40}Ar/^{39}Ar dating at the geochronology laboratory at the University of Potsdam. Sampling was carried out based on the geologic map of Dunkley et al. (1993) and the morphology of lava flows on and around the flanks of Paka. Concentrated groundmass parts of the rocks after acid treatment were sent to Oregon State TRIGA Reactor for neutron activation of samples for four hours at the CLICIT facility, then ^{40}Ar/^{39}Ar analyses were performed at Potsdam. In most cases, high-resolution ages were successfully obtained that are broadly consistent with Dunkley's ages after careful evaluation of age spectra and isochrons obtained by stepwise-heating with a CO_2 laser.

A total of thirty ^{40}Ar/^{39}Ar ages was obtained, except two negative ages. The age data show protracted volcanic activity during the past 0.6 Ma years, and also three phases of pronounced volcanic activity: (I) between 0.428 and 0.372 Ma, (II) between 0.160 and 0.126 Ma, and (III) between 0.039 and 0.012 Ma (Fig. 1). Based on these three periods, all rocks were grouped in three periods, as (1) 0.6-0.35 Ma, (2) 0.35-0.1 Ma and (3) 0.1-0 Ma. In addition, we obtained geochemical information of the samples and their spatial distribution was investigated. We found that several basaltic lava flows of Dunkley et al (1993) were trachyte or mugearite flows; in addition, we conclude that some stratigraphic positions should be modified. During period (1), only trachyte and mugearite erupted on the western rift floor and the northeastern flank of Paka; instead, the younger trachytes erupted more along a N-S trend on the flank of Paka. Many normal faults affecting flows are only found on the older lava flows in northeastern side of the volcano, which belong to period (1). The trace element ratio of Nb/Zr clearly shows different ratios among the three age groups, which implies a gradual change of magma composition over time. Overall, our new age data are meaningful and consistent with field observations of superposition and cross-cutting relationships and thus help to develop a structural evolution model for the inner trough region of the northern Kenya Rift.

Reference: Dunkley P. M., M. Smith, D. J. Allen and W. G. Darling (1993): International Series, Research Report SC/93/1, 185pp, British Geological Survey

Keywords: Kenya Rift, Ar/Ar dating, Quaternary volcano, tectonics, whole rock chemistry

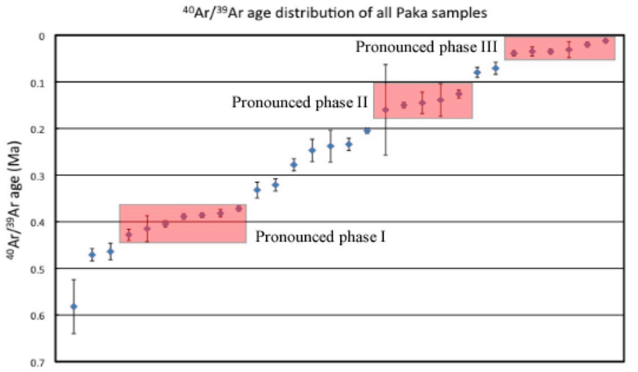

Fig 1. Distribution of ^{40}Ar/^{39}Ar ages of Paka samples and three inferred periods of pronounced volcanic avtivity.

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SGC51-04 Room:A04 Time:May 24 16:15-16:30

Quantifying seawater assimilation across a single MORB pillow

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Halogens and noble gases are critical tracers of volatile interaction between geological reservoirs. The incompatibility of the heavy halogens (Cl, Br, I) coupled with their unique elemental composition and concentrations within surface reservoirs make them suitable tracers of volatiles recycled back into the mantle [1]. However, as the halogens are concentrated within seawater and other marine reservoirs it is possible that they are also incorporated within submarine basalts during eruption, causing the original mantle volatile signature of the basalt to be overprinted [2].

This study combines halogen (Cl, Br and I), noble gas, K and H_2O concentrations within a single pillow basalt to quantify the amount of seawater assimilation during eruption, and to further elucidate the mechanisms of assimilation. In order to determine the scale and heterogeneity of seawater contamination within the pillow, multiple sections taken at 2.5mm intervals were analysed along a transect from the glass rind to the crystalline interior.

The two outer sections of the glass rim show enrichment in the halogens compared to the deeper glass and crystalline section of the pillow, which have distinctly MORB-like I/Cl and Br/Cl ratios. Lower K/Cl and Br/Cl ratios measured within the outermost sections (#1 and #2) indicate the glass has incorporated a Cl rich seawater component. The H_2O/Cl ratios within sections #1 and #2 show that the assimilant is a high salinity brine produced by hydrothermal boiling and phase separation of seawater caused by the eruption of the basalt.

The extent of brine assimilation within the glass ranges between 8% in section #1 to $>$50% in section #2 indicating brine assimilation within basaltic glasses is considerably heterogeneous. The high level of brine contamination within the glass as calculated from the halogens is in contrast with the noble gases within the glass, which still retain their magmatic signatures. This suggests that the halogen and noble gases within submarine basalts may be decoupled, with loss of atmospheric noble gases from the assimilant during hydrothermal boiling.

The crystalline interior shows an increase in the Xe and I concentration relative to the glass rim. The $^{132}Xe/^{36}Ar$ ratio and elevated iodine concentrations when compared to the rest of the pillow indicate the pillow contains a sedimentary component within its interior, most likely due to incorporation of fine sediments during eruption.

Combined halogen, noble gas, K and H_2O data presented here for a single pillow basalt demonstrates that contamination from the marine environment can be variable and extensive. This combined analytical approach demonstrates the need for careful characterization of samples to ensure only samples free from contamination are chosen for analysis of mantle volatiles.

[1] Sumino et al (2010) EPSL, 294, 163 - 172 [2] Kumagai and Kaneoka (1998) Geophys. Res. Lett. 25(20), 3891 - 3894

Keywords: Halogen, Noble gas, Seawater contamination, MORB

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Halogen analysis of mantle xenoliths by extending Ar-Ar and I-Xe dating methods

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An extension of the Ar-Ar and I-Xe dating methods enables us to determine trace amount of halogens (Cl, Br, and I), K, Ca, Ba, and U. These elements are converted to corresponding isotopes of Ar, Kr, and Xe by neutron irradiation in a nuclear reactor. Noble gases in the irradiated samples are analyzed by noble gas mass spectrometry and the conversion factors from parent elements to noble gas isotopes are determined by analyses of standard samples [1]. The detection limits of this technique for Cl, Br, and I are 10^{-10} to 10^{-11}, 10^{-13} to 10^{-14}, and 10^{-14} to 10^{-15} mol, respectively, which are several orders of magnitude lower than those of conventional methods. Using this method, we are investigating halogen geochemistry in the mantle.

Halogens, except for F, are incompatible elements that are strongly partitioned into aqueous fluids. They are concentrated at the Earth's surface in distinct compositions and scarce in the mantle. These features enable us to trace water subducted into the mantle using halogens. Halogens are also thought to play an important role in transporting LIL (large-ion lithophile) elements. Enrichment of LIL elements in arc magma is explained by addition of slab-derived aqueous fluids. These elements are up to 10 times more strongly partitioned into aqueous fluids when the fluids contain halogens [2]. Because of these geochemical characteristics, halogens are among the most important elements in slab-derived fluids. The fluid inclusions of mantle xenoliths provide the most primary information on slab-derived fluids supplied to the mantle wedge. However, the halogen concentrations in such materials are too low to analyze by conventional methods and thus little research has been conducted on them.

We determined the halogen concentrations in mantle xenoliths from various geological settings: Western-Pacific subduction zones (Kamchatka, the Philippines, and N. Japan), Europe (Eifel), and North America (San Carlos and Kilbourne Hole). The halogen compositions of fluid inclusions were obtained by crushing extraction of noble gases. Melting extraction was also conducted to obtain those of whole samples.

The H_2O-rich fluid inclusions of the mantle xenoliths from volcanic fronts of subduction zones show high I/Cl ratios with relatively constant Br/Cl ratios, which are similar to those of marine sedimentary pore fluids (seawater trapped in pores of deep-sea sediments) [3]. Halogen compositions obtained from melting of the same samples show a small addition of Cl; however, they are still similar to sedimentary pore fluids. Similar signatures were found in mantle peridotites from the Sanbagawa metamorphic belt, suggesting subduction of sedimentary pore-fluid-derived halogens thorough incorporation into hydrous minerals such as serpentine in the subducting oceanic lithosphere [4]. Although the halogen concentrations are lower, some mantle xenoliths from rear-arc regions of subduction zones also show sedimentary pore fluid-like signatures. The I/Cl and Br/Cl ratios of mantle xenoliths from intraplate settings (Europe and North America) show common correlation, suggesting halogen fractionation from MORB source-like composition.

The halogen signatures of mantle xenoliths from subduction zones suggest that subduction of sedimentary pore-fluid-derived halogens [4] significantly extend into the mantle beneath volcanic front. These subducted halogens also extend to rear-arc regions, although their influence is small. The correlated I/Cl-Br/Cl ratios of mantle xenoliths from intraplate settings may have resulted from a metasomatic event, which could generally take place in subcontinental lithospheric mantle.

[1] Turner (1965) JGR 70, 5433. [2] Kawamoto et al. (2014) EPS 66, 1. [3] Kobayashi et al. (2014) JpGU Meeting 2014, SCG62-04. [4] Sumino et al. (2010) EPSL 294, 163.

Keywords: halogen, noble gas, mantle xenolith

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Across-arc variation in noble gas and halogen compositions of volcanic rocks from the Izu-Ogasawara subduction zone

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Recently, subducted halogens and noble gases with seawater and sedimentary pore-fluid signatures were discovered in exhumed mantle wedge peridotites and eclogites from the Sanbagawa-metamorphic belt, southwest Japan [1,2]. These findings along with seawater-derived heavy noble gases (Ar, Kr, and Xe) in the convecting mantle [3] provide observations that allow us to investigate the processes that control the return of volatile and highly incompatible elements into the mantle. Serpentinized lithosphere of subducting oceanic plates can transport noble gases and halogens acquired from pore water in the overlying sediment [1,2,4]. To verify whether and how such subduction fluids modify the composition of the mantle beneath subduction zones, we determined the noble gas and halogen compositions of olivines in arc lavas of the northern Izu-Ogasawara subduction zone and IODP sediments and basalts recovered from the northwestern margin of the Pacific plate. Trace amounts of halogens (Cl, Br, and I) in the olivine samples were measured by a combination of neutron irradiation and noble gas mass spectrometry [5].

The ^{3}He/^{4}He ratios of samples from the volcanic front (Izu-Oshima, Miyakejima, Mikurajima, Hachijojima, and Aogashima) and rear-arc (Niijima, Higashi-Izu monogenetic volcanoes, Nanzaki, and en-echelon seamount chains) regions are in the range of the mid-ocean ridge basalt (MORB) value, without systematic differences among the regions. This indicates a considerably low contribution to the mantle wedge beneath the arc of radiogenic ^{4}He in the subduction fluids observed in the Sanbagawa samples exhumed from a depth ranging from 30 to 100 km [1,2].

In contrast, the ^{40}Ar/^{36}Ar ratios for each volcano (299-620) are significantly lower than the MORB source (up to 32,000 [3]), indicating significant involvement of atmospheric Ar (^{40}Ar/^{36}Ar = 296) in the magmas. Systematically higher ^{40}Ar/^{36}Ar ratios in the rear arc than in the volcanic front, and a comparison with those of subducting materials, suggest that subduction of seawater-derived Ar significantly affects the noble gas composition of the magma-generation region. A simple mass balance calculation of subducted and mantle-derived Ar isotopes reveals that higher subduction flux than that of seawater-derived Ar in the pore fluids in the subducting sediment/crust is required. The serpentinized lithosphere in the subducting slab is the best possible carrier of seawater-derived Ar with high subduction flux.

Although the halogen compositions of most of the olivines are close to that of MORB-source mantle, some samples from the en-echelon seamount chains show a significant contribution from pore-fluid-derived halogens. Combined with the noble gas results, halogen-poor fluid associated with atmospheric noble gases may be dominantly released from the subducting slab beneath the Izu-Ogasawara arc, while halogen-rich fluid significantly contributes to the magma generation region beneath some volcanoes in the rear arc.

It remains unclear whether noble gases and halogens in the subducting materials are completely released beneath the rear arc. However, the presence of seawater-like noble gases in the convecting mantle [3] implies that a small portion of seawater-dissolved atmospheric noble gases, carried in the serpentine, might be transported to greater depths in the deeper mantle.

[1] Sumino et al. (2010) Earth Planet. Sci. Lett. 294, 163-172. [2] Sumino et al. (2011) Mineral. Mag. 75, 1963. [3] Holland & Ballentine (2006) Nature 441, 186-191. [4] Kendrick et al. (2011) Nature Geosci. 4, 807-812. [5] Turner (1965) J. Geophys. Res. 70, 5433-5445.

Keywords: Noble gas, Halogen, Subduction zone, Izu-Ogasawara arc, Pore fluid

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SGC51-07 Room:A04 Time:May 24 17:00-17:15

Noble gas isotopes; Solar evolution parameter

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Noble gas isotopic parameters are essential reference parameters for planetary evolution.

Keywords: noble gas isotopes, planetary evolution, meteorite

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New analytical methods and a system for noble-gas measurements of extraterrestrial materials

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Noble gas isotopes in extraterrestrial materials, meteorites, cosmic dust (micrometeorites), and spacecraft-return samples, are one of the most useful tools of cosmochemistry. Radiogenic (e.g., ^{40}Ar, ^{129}Xe, and ^{136}Xe) and cosmogenic (^{21}Ne, ^{81}Kr, ^{83}Kr, and ^{128}Xe) isotopes are used for dating when gases have begun to be accumulated in the relevant minerals or phases. Primordial or trapped (neither radiogenic nor cosmogenic) isotopes are informative to infer the origin of the materials. Conditions of physical processes, such as evaporation and condensation, will be discussed based on elemental/isotopic fractionation effects.

The mass spectrometry system at Kyushu Univ. consists of a conventional mass spectrometer (MS) and a resonance ionization MS, called RING (Resonance Ionization Noble Gas mass spectrometer). The conventional MS is mainly used to measure light noble gases (He, Ne, and Ar) in micro- or submicro-gram samples enriched in solar-wind noble gases, such as mineral separates from regolith breccia meteorites, Antarctic micrometeorites [1], lunar soils, and the Itokawa dust particles returned by the Hayabusa spacecraft [2]. Extraction of noble gases is performed using infrared radiation lumps for 50-200 ℃ and a resistant furnace (called Pot-pie furnace) for 200-2000 ℃. In addition, the MS system is equipped with a Nd:YAG laser that produces energy of 200 mJ/pulse (with a 10 nsec width) at 1064 nm and a spot with $\sim$50 μm in diameter. In the ablation of samples, plasma lights are emitted due to the high energy density of the laser pulse, and have wavelengths depending on the chemical composition of the fused area. The plasma emission spectra are measured using a LIBS (Laser-induced Breakdown Spectroscopy, e.g., [3]) system prepared for the laser extraction system. Combination of the conventional MS and LIBS enables to determine K-Ar radiometric ages for the specific areas (e.g., shock-melt veins or feldspathic phases) of polished sections. Experimental analyses using terrestrial rocks containing K-rich phases (e.g., K-feldspar) and equilibrated ordinary chondrite samples are now on going.

The RING mass spectrometer is designed to measure extremely small amounts of Kr and Xe ($>$100 atoms), as modeled after RELAX (Refrigerator Enhanced Laser Analyzer for Xenon) developed by the Manchester group (e.g., [4] and [5]). In our system, a dye laser equipped generates 3.5 mJ/pulse at $\sim$216 and $\sim$256 nm of wavelengths that ionize resonantly Kr and Xe, respectively. At this moment, we have got mass spectrum of Xe isotopes but not those of Kr yet.

In addition to the improvements and developments of instruments for noble-gas mass spectrometry, sample preparation methods have also been developed for combinational (multidiscipline) analyses for individual samples with micro- and submicro-gram masses. For example, we have performed TEM and noble gas analyses for individual micrometeorites [1] and the Itokawa dust particles [2].

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Keywords: noble gas, extraterrestrial material, resonance ionization mass spectrometry, laser-induced breakdown spectroscopy, K-Ar dating

Japan Geoscience Union Meeting 2015
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SGC51-09 Room:A04 Time:May 24 17:30-17:45

Relationships between morphological features and isotopic signatures of noble gases of four Itokawa particles

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Introduction: Fine-grained particles of the asteroid Itokawa preserve the effects related to the processes on and near the surface of an asteroid. Various types of surface modification were found on the very surface ($<$100 nm) [1-6]. The modification textures suggest that irradiation by solar wind (SW) plays an important role for their formation. Number density of solar flare tracks in them ranges from a few × 10^9 to a few x 10^{10} tracks/cm^2 [2, 5], which are considerably lower than expected. These data suggest the mechanisms and periods of the surface modification of Itokawa grains are still unresolved. In addition to the SW irradiation, there seems to have been another mechanism that modified the morphology and surface features of the grains. Matsumoto et al. [3] found some Itokawa grains were aggregates composed of subgrains with stepped surfaces, which suggests evaporation and/or condensation. Noble gas mass spectrometry is another tool to infer the individual history of each Itokawa grain. Nagao et al. [7] argued that a grain experienced multiple processes: exposure to SW, burial in the regolith layer, and removal of its external surface. We intend to investigate the relationships among morphology, surface modifications, and noble gas isotopic signatures to discuss the history of each Itokawa particle from various perspectives.

Samples and methods:. We fixed four Itokawa particles on tantalum plates by acetone-soluble glue. We observed the surface of the particles and prepared thin foils by FIB-SEM at Ibaraki University. After preparation of FIB foils, all the Ta plates with samples rinsed by acetone in a N_2 filled glove box. They were transferred to Kyushu University and noble gas mass spectrometry was performed for all the noble gas elements using a modified VG-5400. Noble gas extraction from each sample was conducted stepwise at 50, 100, 200, 300, 600 and 1400 ℃.

Results and discussion:. SEM observation of the four Itokawa grains investigated shows various morphology and surface features. Edges observed on the surfaces of three grains vary from sharp to dull. Additionally, one grain is composed of two subgrains with stepped surfaces. Morphology and surface features are related to the He release patterns. The three particles have similar ^{4}He release patterns; they release most ^{4}He below 200 ℃ fractions and a small amount of ^{4}He above 600 ℃. Although the gas-release profiles are similar to each other, there is a difference in the bulk concentrations of ^{4}He; one of the three grain having dull edges contain more abundant ^{4}He than the others having sharp edges. Because bulk ^{3}He/^{4}He ratios in the three grains are 2.5 to 6.5 x 10^{-4}, which is comparable with that in the solar wind of 4.57 x 10^{-4} [8], it is likely that the implanted He in the grains increased as the surfaces were degraded by solar wind sputtering. On the other hand, the grain composed of subgrains with stepped surfaces shows a different ^{4}He release pattern; ^{4}He was released only below 200 ℃. This fact suggests that deeply implanted He, which is probably related to He released above 600 ℃, was completely reset by severe heating. The steps on the surfaces of this grain may have been formed by evaporation and/or evaporation and subsequent recondensation by a heating event, which released He from the interior of the grain. These results strongly suggest that micrometer-size surface features are related to the amounts and the release patterns of the implanted He.

References: [1] Noguchi et al. (2011) Science 333, 1121-1125. [2] Noguchi et al. (2014) Meteorit. Planet. Sci. 49, 188-214. [3] Matsumoto et al. (2013) 44th Lunar Planet. Sci. Conf. abstr. #1441. [4] Thompson et al. (2014) Earth Planet. Space 66, 89-99. [5] Keller and Berger (2014) Earth Planet. Space 66, 71-78. [6] Noguchi et al. (2014) Earth Planet. Space 66, 124-134. [7] Nagao et al. (2011) Science 333, 1128-1131. [8] Heber et al. (2009) Geochim. Cosmochim. Acta 73, 7414-7432.

SGC51-P01 Room:Convention Hall Time:May 24 18:15-19:30

Present status for in-situ noble gas analysis by Sputtered Neutral Mass Spectrometry with tunneling ionization

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LIMAS (Laser Ionization MAss nanoScope) was installed in 2011. In-situ analysis for trace elements in nanometer-scale is a concept of this apparatus. I have conducted mechanism of solar wind (SW) implantation into extra-terrestrial materials such as Itokawa particles. Because depth profile of SW-He have not been determined by using conventional noble gas mass spectrometer (NG-MS) in a quantitative way, a depth profiling of the SW noble gas contributes to the great progress for understanding implantation and removing processes of the SW noble gases. The processes imply the space-weathering history of the materials on its asteroid surface.

LIMAS is mainly composed Ga focused ion beam (FIB) for primary ion, femtosecond (fs) laser for tunnel ionization of sputtered particles, and time-of-flight mass spectrometer. Depth profiling procedure is as follows. The FIB system is carried out high spatial resolution with large primary current. The smallest beam size was 8 nm at 3 pA, and the largest size was 600 nm at 30 nA. The fs laser can ionize the all element even He because of high energy density. The energy density is 8E15 W/cm^2 because the pulse energy and width was 6.3 W and 30 fs, respectively, and the laser pulse was focused on a volume of 50 x 50 μm^2. This laser pulse induces tunneling ionization (Delone and Krainov, 1998). Helium depth profile method in a few micrometer has been developed with LIMAS and atomic force microscope for crater depth measurement.

LIMAS can detect tens ppma He from sub-micrometer area on solid surface. In presence useful yield of He is 0.02% which should be up to 1% by optimize laser focusing and irradiation position. The yield of 0.02% corresponds to 4000 He atoms for single He ion count. The back ground of He is much less than that of the NG-MS because LIMAS is used by dynamic operation under the ultra-high vacuum.

LIMAS can measure locally concentrated noble gas because the sputter rate of the pulsed FIB is low, which indicates that this measurement is almost non-destructive isotope analysis. This feature can play a role for analysis of tiny and precious samples such as the Itokawa particles. U,Th-He and K-Ar dating for single grain can be applied by the in-situ noble gas analysis. The in-situ dating may obtain a new insight of thermal history of igneous rocks by comparing conventional in-situ dating method such as U-Pb age.

Keywords: noble gas, in-situ analysis, Sputtered Neutral Mass Spectrometry

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SGC51-P02 Room:Convention Hall Time:May 24 18:15-19:30

New preparation apparatus for noble gas isotope analyses to measure submarine hydrothermal fluids and renewed GVI-5400

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Hydrothermal activity is an essential phenomenon to drive geochemical differentiation on the Earth's surface, which should form polymetallic massive sulfide ores. Noble gas isotopes are regarded to be a powerful tracer even for the geochemical study on hydrothermal activity; however, there are abundant interference volatiles, e.g. halogens and hydrogen sulfide. Here, we designed a new preparation system to remove such volatiles using non-traditional gettering materials. This new preparation system consists of following components: the water preparations system, composite gettering system (halogen getter, sulfide getter and Ti-Zr getter), cryogenic pump, Saes-getter pump and charcoal trap to analyze the all noble gases. The new preparation system has compact volume, which needs small amount of seawater sample of 2-5 cc in volume. It is almost 1/20 compared to the typical requirement for He isotope measurement.

To design new noble gas measurement system in JAMSTEC, another critical problem is to be solved. One of widely used noble gas mass spectrometers, GVI-5400, is now not listed in commercial production models. Thus, all successor models of VG-5400 series have entirely finished their production support since several years ago. However, the VG-5400 series and its successor models still show satisfactory sensitivity and precision to determine noble gas isotope ratios. If we decide to use them much longer, we should maintain their control units, such as power supply, emission control or source control and so on, which requires replacement of essential electrical parts, including vacuum tube (!), regularly. At present, some of these parts are not supplied anymore and we are forced to choose between not replacing them with the new/second-hand electrical parts while they still worked, or renew their electrical circuits with currently manufactured electrical parts. By chance we had started the project to re-use old model of power supply unit. The first trial was applied to the very original VG5400 that was moved from Univ. Tokyo to Yamagata Univ. As the second case, we renewed the power supply and the emission control unit equipped for the JAMSTEC-2 noble gas mass spectrometer (GVI-5400He).

Here we report the better performance of new version of the renewed power supply unit, noiselessness and better stability, with the new noble gas preparation system.

Keywords: noble gas, getter pump, hydorothermal activity, mass spectrometer, extraction system

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High sensitive noble gas mass spectrometer equipped with a Giese-type ion source

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Although noble gas isotopes are powerful tracers in geosciences, their extremely low abundances in mineral and rock samples make their analysis quite difficult. For example, concentration of ^{3}He, which is a good indicator of mantle-derived component because of its primordial origin, is as high as 0.01 parts-per-trillion in volcanic rocks and mantle-derived materials. Such scarce noble gas isotopes are detected with a special mass spectrometer operated in static mode. We have made it possible to detect 10^3 to 10^4 atoms of noble gas isotopes by modifying a commercial sector-type single focusing noble gas mass spectrometer (VG5400), which is equipped with a double collector system to detect ^{3}He and ^{4}He simultaneously with a secondary electron multiplier and Faraday cup, respectively [1]. Here we report an attempt of further improvement of sensitivity of the mass spectrometer by installation of a new ion source (Giese-type source).

The Giese-type electron ionization (EI) ion source is equipped with two electrostatic quadrupole lenses [2]. This source has been reported to have up to two orders of magnitude higher sensitivity than conventional Nier-type EI source because of the absence of a beam defining slit to collimate the ion beam and thus high transmission [3]. We designed a Giese-type source to have an adequate resolution to separate $^3\mathrm{He}^+$ from HD^+ and $\mathrm{H_3}^+$, to have the source housing volume as small as possible, and to be bankable at up to 300 ℃ to reduce outgas from the source materials. The ion and electron optics were based on a calculation by Lu and Carr [4] and refined using SIMION-3D software [5]. Prior to the installation on the mass spectrometer, the ion beam profile emitted from the source was monitored by a microchannel plate and phosphor screen to optimize the configuration of the quadrupole lens.

A sufficient mass resolution over 500 essential for ^{3}He/^{4}He analysis has been achieved with an improved sensitivity approximately three times higher than the previous condition. The amount of helium required to obtain a precision with ^{3}He/^{4}He ratio is two orders of magnitude smaller than that with the condition installed by the manufacture. However, total ion transmission is estimated to be about 30%, suggesting further refinement of the source condition is required to obtain the maximum sensitivity.

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Keywords: Noble gas, Mass spectrometer, Quadrupole lens, Ion source

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Ar-Ar dating by fluid inclusions in hydrothermal quartz, compared with its field and microscopic observations

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The chemical evolution history of the ocean must have been one of the most critical factors to unravel the evolution of life on the Earth. However, this topic has not fully understood yet. Recently, de Ronde et al. (1997) and some studies try to estimate seawater composition during Archean and Proterozoic using fluid inclusions trapped in hydrothermal quartz correlated with pillowed basalt, which is expected to erupt in sub-seawater condition. Lowe and Byerly (2003) claims that the hydrothermal minerals used by de Ronde et al. (1997) were formed at far younger age (Quaternary) than expected age of eruption of the matrix lavas (3.2 Ga), based on the only field and microscopic observations. This problem must be solved by dating of the fluid inclusions trapped in hydrothermal quartz and comparing the age of pillowed basalt as a matrix in addition to field and microscopic observations. We carried out Ar-Ar dating of the fluid inclusions with stepwise crushing techniques at Guangzhou Institute of Geochemistry, Chinese Academy of Sciences.

Analyzed samples in this study are from 2.2 - 2.4 Ga Ongeluk Formation, Transvaal Supergroup, South Africa. Above Ongeluk Formation, two younger volcanic layers exist, one is mafic intrusions of the Malopo Farms complex formed at 2044 ± 24 Ma, the other is porphyritic andesite of the Hartley Formation formed at 1928 ± 4 Ma.

The analyzed samples are hydrothermal quartz with growth zoning texture, filling the primary shaped drainage cavities and interstitial spaces of lava flows of the Ongeluk Formation without quartz vein crosscut pillow of lava, suggesting the quartz had precipitated soon after eruption of the lava. We analyzed four of the most primary-rich samples (GU84, GU91, GU103a and GU103b), in terms of microscopic observations. However, secondary fluid inclusions are not excluded completely, in other words, argon gas extracted the quartz samples including some secondary fluid inclusions origin in the analysis.

Three samples, GU 84, GU103a and GU103b, yielded Ar-Ar isochron ages of 2688 ± 584 Ma, 1952 ± 279 Ma and 1967 ± 64 Ma Ar-Ar isochron ages, respectively. GU91 was also dated in the same manner as well, but failed to obtain a tight line of isochron with a diffused age (611 ± 517 Ma).

Comparing the age of the Ongeluk lava and younger two volcanisms with the results, the age from GU84 has a wide range of error, but is consistent with Ongeluk lava, GU103a has also a wide range of error, but is consistent with all three volcanisms, and GU103b has the narrowest range of error and is consistent with Malopo Farms complex and Hartley Formation, especially match with Hartley. Based on field and microscopic observations, it is unlikely to precipitate quartz with filling cavities 100-200 Myr after eruption of Ongeluk lava. Two probable mechanisms to explain the results, one is that fluid inclusions were re-equilibrated at younger volcanisms in terms of the argon with preserving primary textures. The other is that secondary fluid inclusion formed by younger volcanism driving hydrothermal circulations and that extracted argon gas in the analysis is mainly composed of the secondary inclusions. The secondary fluid inclusions have argon ratio suggesting younger age than primary fundamentally, suggesting that GU84 obtaining the oldest argon age is the most primary rich samples. Assuming that obtained younger ages than Ongeluk lava are resulted in argon gas from secondary fluid inclusions, the consistency with Ar-Ar age of GU84, the most primary-rich sample, and Ongeluk lava support that our field and microscopic observations are robust and that the primary fluid inclusions formed soon after Ongeluk lava eruption.

Keywords: fluid inclusion, Ar-Ar dating, quartz, seawater

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Possibility of K-Ar age mapping on the moon using cosmogenic ^{39}Ar

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Rapid and wide range of age survey on the moon surface requires in situ measurement. ^{39}Ar of cosmogenic origin from ^{39}K in meteorite has been noticed since the early stage of noble gas research, and became a cue to the ^{40}Ar/^{39}Ar method. If the production rate of ^{39}Ar is considered uniform, the production and decay of ^{39}Ar reaches to equilibrium after a long time, since ^{39}Ar has a half life of 293 years. This half life is long enough to detect ^{39}Ar in meteorites and moon rocks, and short enough to reach equilibrium of the isotope. Thus, such rocks or minerals possess a certain amount of ^{39}Ar proportional to their potassium content. Using meteorites or rocks on moon surface under the same exposure condition, ^{40}Ar/^{39}Ar age can be determined. For a start, a precise date of one of those samples in a laboratory and ^{40}Ar/^{39}Ar ratio measurement of the same sample in the field are required to obtain J-value in the field. ^{40}Ar/^{39}Ar ratios measurement of the rest of unknowns in the field allow us to calculate ages. Interfering isotopes from calcium will not affect much as long as Ca/K ratio is low. Samples in a depth or a shadow from exposure are not suitable for the measurement since the neutron density attenuates with depth. However, no need for atmospheric contamination, mass measurement and ^{36}Ar measurement may provide us more mobility and rapid measurement.

Keywords: ^{39}Ar, Cosmogenic, age distribution, ^{40}Ar/^{39}Ar age, moon surface