Development of a new method for in situ cosmic-ray exposure age measurements using laser ablation

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On the surface of planets without thick atmosphere and magnetic field, including Mars, biosignatures and organic materials are degraded by cosmic and ultraviolet rays at a timescale of a few million years [1]. Thus, during lander missions, we should search for organic matters at places that had been buried and shielded from cosmic and ultraviolet rays until very recently. However, evaluating when a certain place/rock was exposed to the surface just by observing with a camera is challenging. Cosmic-ray exposure (CRE) ages can be an indicator of the duration for which the sample is subject to the severe environment. Thus, developing a method for in situ CRE age measurement would be important. In situ CRE age measurements are also useful for choosing samples that should be brought back to the Earth.

On Mars, K–Ar and CRE ages of powdered rocks were measured by the NASA’s Curiosity rover using APXS to derive the elemental composition and thus production rates, quadrupole mass spectrometer, and onboard furnace to derive the concentrations of noble gases [2]. Their method, however, had uncertainties in ages because (1) the production rates of cosmogenic nuclei were uncertain between the measured aliquots due to the heterogeneity of the samples and (2) the extraction of noble gases was incomplete due to insufficient heating temperature and duration [5]. Drilling rocks and keeping the onboard furnace at 900℃ for 30 minutes were among the technical challenges for the rover operation.

To resolve these problems and measure the cosmic-ray exposure ages of rocks, we develop a new method that combines laser-induced breakdown spectroscopy (LIBS) and quadrupole mass spectroscopy (QMS). The LIBS-MS approach potentially yields the spot-by-spot CRE ages by measuring cosmic-ray production rates with LIBS and concentrations of noble gases with MS. In this study, we assess the feasibility of measuring very small amount of cosmogenic noble gases (~10^{-15} mol) in a meteorite, as well as to investigate the accuracy of the CRE ages obtained with this method. This study focuses on the noble gas analysis with QMS because LIBS measurements for major target elements are more straightforward.

The CRE ages were measured in the following procedures: 5000 laser pulses (266 nm, 20 mJ, spot size 350 μm) were applied to a slab sample of the Camel Donga meteorite. Camel Donga is a eucrite that exhibits CRE ages of 11.9 ±1.3, 27.3 ±1.4, and 28.8 ±1.9 Ma for ^{3}He, ^{21}Ne, and ^{38}Ar, respectively [3]. The gases extracted by the laser were processed with a Ti-Zr getter and a charcoal trap before being introduced to the QMS. The abundances of noble gases, ^{3}He, ^{21}Ne and ^{38}Ar, were determined after correcting procedural blank and isobaric interferences. The volume of laser-ablation spots was measured with a microscope and was converted to ablated mass with the bulk density of the sample of 3.16 g/cm^3 [4]. The CRE ages were calculated from the concentrations of the cosmogenic noble gases (^{3}He, ^{21}Ne and ^{38}Ar) and published production rates [3].

In our preliminary CRE age measurements, noble gases of ~10^{-16} mol were detected although the signal-to-blank ratios were small (<5). The ablated mass was 5e-5 g. Isotopic ratios of noble gases, e.g., $^{38}Ar/^{36}Ar$ = 1.13 ±0.07, were more consistent with their cosmogenic origin (1.55) rather than the trap of the terrestrial air. By assuming the typical shielding factor ($^{22}Ne/^{21}Ne$ = 1.14), we obtained the CRE ages of

148 ±8 Ma for ^{3}He, 21.8 ±7.0 Ma for ^{21}Ne, and 60.4 ±91.7 Ma for ^{38}Ar. The CRE ages for ^{21}Ne and ^{38}Ar are consistent with those reported in the previous study, suggesting the validity of this approach. The much longer CRE age obtained with ^{3}He is most likely because of the isobaric interference by HD^{+} released from the meteorite. To obtain accurate ^{3}He-ages, HD^{+} should be removed more efficiently by adding a second getter near the QMS. We found that the reliable age for the Camel Donga meteorite is the ^{21}Ne-age under the assumption that the shielding indicator ^{22}Ne/^{21}Ne is 1.14, but otherwise the ^{38}Ar-age. Our noble gas analyses demonstrate that even very small amounts of cosmogenic noble gases can be measured with this method, suggesting the capability of in situ CRE age measurement based on laser ablation.

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