

NEW CONSTRAINTS ON THE PRODUCTION RATE OF COSMOGENIC DEUTERIUM AT THE MOON'S SURFACE. E. Füri¹ and E. Deloule¹, ¹Centre de Recherches Pétrographiques et Géochimiques, CNRS-UL, 54501 Vandœuvre-lès-Nancy, France (efueri@crpg.cnrs-nancy.fr, deloule@crpg.cnrs-nancy.fr)

Introduction: The Moon's surface lacks full atmospheric shielding and magnetic field protection against high-energy galactic (GCR) and solar cosmic rays (SCR). GCR and SCR particles can, therefore, penetrate lunar matter to depths of several meters and ~2 cm, respectively. Consequently, cosmic-ray induced spallation reactions are important sources of cosmogenic nuclides, such as deuterium (D) and noble gases (e.g., ³He, ²¹Ne, ³⁸Ar), in lunar rocks and soils with long cosmic ray exposure (CRE) ages. Given the recently emerged interest in the origin of lunar 'water', accurate knowledge of the cosmogenic D production rate (P_D) is critical for correcting measured D/H for the cosmogenic contribution, and, ultimately, for comparing lunar hydrogen isotope ratios with those of potential Solar System sources (i.e., solar, chondritic, cometary). Based on the D content of basalt 70215, Merlivat et al. [1] estimated the P_D value to be $9.2 \times 10^{-13} \text{ mol(g rock)}^{-1}\text{Ma}^{-1}$ at ≥ 2 cm depth. Reedy [2] derived a similar value for the GCR production of D for the top 40 g/cm² of shielding from theoretical calculations; however, the additional D production by SCRs (for a rigidity $R_0 = 100$ MV) results in a P_D value of 2 to $3.5 \times 10^{-12} \text{ mol(g rock)}^{-1}\text{Ma}^{-1}$ at the near surface. Here, we re-evaluate the lunar P_D value by coupling SIMS analyses of hydrogen isotopes with CO₂ laser extraction-static mass spectrometry analyses of noble gases in lunar olivines with variable exposure histories.

Samples and analytical methods: Eight olivine-rich mare basalts from Apollo 12 and 15, covering a wide range of CRE ages from ~50 to 300 Ma (Table 1), were selected for this study. Up to four olivine grains of each sample were mounted in indium together with four standards with known H₂O contents (as in [3]). The CAMECA 1280 HR2 ion microprobe at CRPG, Nancy, was used for hydrogen isotope analyses, and a CAMECA SX100 electron microprobe (GéoRessources, Nancy) was used to determine the major and minor element composition of olivine. Noble gas (He, Ne, Ar) abundances and isotope ratios are currently being determined by CO₂ laser heating and analysis on a VG5400 noble gas mass spectrometer in order to verify previously published CRE ages of the basalt samples.

Results: Typical hydrogen isotope count rates on the StHs6/80-G andesitic glass standard (250 ppm H₂O; $\delta D = -95\%$) were ~120,000 cps for H⁻ and 16.0±1.1 cps for D⁻. Count rates on synthetic 'dry' standards (Suprasil 3002 quartz glass and forsterite) averaged 2080±700 cps for H⁻ and 0.26±0.09 cps for

D⁻, corresponding to a water content of 4.2±0.5 ppm when compared to the StHs6/80-G standard. The majority (n = 28 out of 38) of the H-measurements on lunar olivine grains fall within the range observed for the two 'dry' standards, and there is no evidence for increasing H abundances with increasing exposure ages (Fig. 1). In contrast, the D⁻ ion intensities measured on the olivines are significantly higher than those of the synthetic standards and vary between 0.91 and 4.78 cps. Most importantly, the D⁻ signals, as well as the measured D/H ratios, show a clear correlation ($R^2 = 0.8$) with previously determined CRE ages of the basalts (Fig. 1). This is interpreted as evidence for the in-situ production of cosmogenic deuterium in olivines by cosmic ray-induced spallation reactions during exposure at the Moon's surface.

Discussion: The abundance of cosmogenic D in lunar olivines depends on the duration of cosmic ray irradiation and the D production rate, which itself is a function of the chemical composition and the shielding depth. The CRE age can be determined by quantifying the concentrations of cosmogenic noble gas isotopes, and by comparing them with well-established noble gas production rates [11, 12]. The studied olivines show a significant range in Fo contents (between Fo57 and Fo77); nonetheless, based on the model of Reedy [2], we estimate that 60 to 70% of the cosmogenic D in olivine is produced from the target element oxygen, and the observed variations in the MgO-FeO content do not significantly affect the P_D value. The mare basalt rocks targeted here were originally between ~4 and 10 cm in size. However, the location of the fragments allocated to us within their host rocks is unknown, and the orientation of the rocks might have varied over the course of their exposure histories [1]; hence, the shielding conditions of individual olivine crystals cannot accurately be constrained.

Sample	CRE age (Ma)	Reference	Fo#
12004	45-60	[4, 5]	67-69 (n=2)
12009	112-160	[4, 6, 7]	71-76 (n=3)
12018	170-210	[4-7]	77 (n=1)
12020	40-80	[4]	72-74 (n=3)
12075	240-300	[4]	75-76 (n=3)
12076	220-240	[4]	75-76 (n=3)
15016	285-335	[8, 9]	67-70 (n=2)
15535	110	[10]	57-69 (n=4)

Table 1: Previously determined CRE ages of the studied mare basalts, and forsterite contents of the olivine grains targeted for hydrogen isotope analyses.

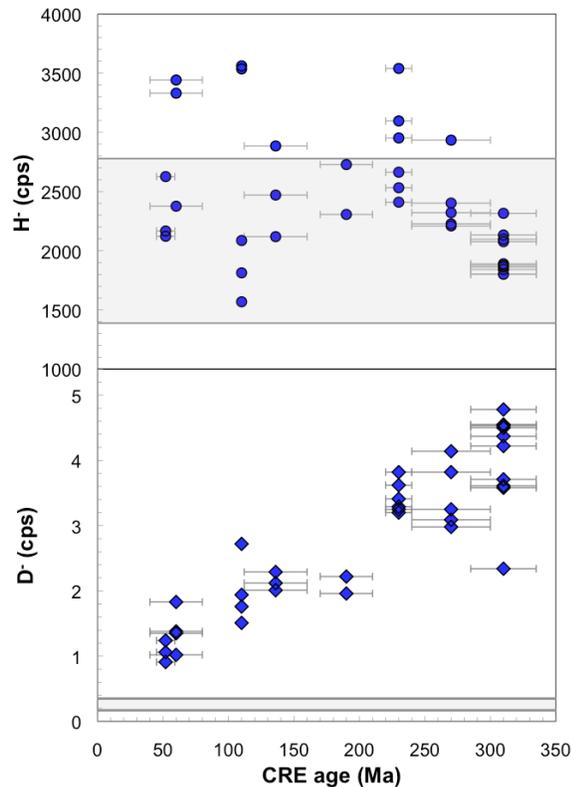


Fig. 1: Measured H^+ and D^- ion intensities in counts per second (cps) as a function of previously published CRE ages. Grey lines delineate the signals determined for the ‘dry’ quartz glass and forsterite standards.

Based on the D content ($3.9 \times 10^{-9} \text{ mol(g rock)}^{-1}$) and the measured D^- ion intensity (16 ± 1.1 cps) of standard StHs6/80-G, the D concentration of the lunar olivines is estimated to vary between 2.2×10^{-10} and $1.2 \times 10^{-9} \text{ mol(g rock)}^{-1}$. Furthermore, the observed linear relationship between the D^- signal and the CRE age shown in Fig. 1 is consistent with an average P_D value on the order of $3 \times 10^{-12} \text{ mol(g rock)}^{-1} \text{ Ma}^{-1}$. This value is three times higher than the estimate of Merlivat et al. [1] but it is in good agreement with the data of Reedy [2] when both GCR and SCR production of cosmogenic D are considered.

Conclusion: While traces of H-bearing species have been discovered in lunar volcanic glasses [3, 13], olivine-hosted melt inclusions [14], and apatites [15], there is no clear evidence for the presence of indigenous ‘water’ in the Apollo 12 and 15 olivines studied here. Furthermore, there is no increase of the H content with increasing exposure durations, indicating that the production of cosmogenic H in lunar samples is negligible. The D abundance, in contrast, is correlated with the exposure age, demonstrating that the contribution of cosmogenic D *must* be carefully evaluated for samples with low ‘water’ (H_2O or OH) abundances and/or

long CRE ages before lunar D/H ratios can be compared to potential ‘water’ sources (i.e., solar, chondritic, cometary). Strikingly, the cosmogenic D production rate derived here is significantly higher than the value proposed by Merlivat et al. [1]; however, these authors concluded that the upper ~ 2 cm of basalt 70215 have been eroded during space exposure, thereby preventing the accumulation of cosmogenic D produced by SCRs. Our new results are consistent with the model of Reedy [2], provided that all olivines analyzed here originate from the near-surface of rocks collected on the Moon and are, therefore, affected by SCR-induced spallation reactions. Our findings indicate that the use of a P_D value of $9.2 \times 10^{-13} \text{ mol(g rock)}^{-1} \text{ Ma}^{-1}$ is not appropriate in all cases for correcting the measured D/H ratios of lunar samples for the cosmogenic D contribution. For samples that are poor in H-bearing species and were exposed to cosmic irradiation within the upper ~ 2 cm below the lunar surface, corrected D/H ratios have likely been overestimated.

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