

HYDROGEN ISOTOPIC FRACTIONATION DURING IMPACT: SERPENTINE, ISOTOPICALLY ENRICHED SERPENTINE, AND MURCHISON; James A. Tyburczy¹, R.V. Krishnamurthy², Samuel Epstein², and Thomas J. Ahrens³, ¹Department of Geology, Arizona State University, Tempe, AZ 85287-1404, ²Division of Geological and Planetary Sciences 170-25, California Institute of Technology, Pasadena, CA 91125, ³Seismological Laboratory 252-21, California Institute of Technology, Pasadena, CA 91125.

Introduction. Planetary accretion, early atmosphere formation, and chemical weathering of planetary surfaces are influenced by impact-induced devolatilization of volatile-bearing rocks and minerals (1,2). In a previous report, we examined the hydrogen isotopic composition of the gaseous products of impact-induced devolatilization of serpentine, finding the gaseous products depleted in deuterium (that is, isotopically 'lighter') relative to the initial solid (3). This work has been extended to include studies of deuterium-enriched serpentine and Murchison carbonaceous chondrite. Deuterium fractionation between the bulk gas and the residual solid exhibits a smooth trend with devolatilization fraction. Thus, planetary interiors may be systematically enriched in deuterium over the planetary surface because of impact-induced isotopic fractionation during accretion.

Experimental Details. Gas-recovery experiments were performed using an assembly that retained the shock-evolved gases (3,4). Deuterium-enriched serpentine was prepared by sealing 'heavy' water and powdered serpentine in a fused quartz tube and heating at temperatures up to 600°C for a period of several weeks. The powdered product had a δD_{SMOW} value of +667 per mil ($\delta D_{SMOW} = \{[(D/H)_{Sample}/(D/H)_{SMOW}] - 1\} \times 1000$; SMOW, Standard Mean Ocean Water). The enriched serpentine was shocked as pellets of approximately 15% porosity. Meteorite samples were prepared as wafers as in previous studies (5); bulk δD_{SMOW} of Murchison is -51 per mil (6). After impact, the trapped gases were transferred to a vacuum system for manometric and isotopic analysis. An empty assembly was impacted in the same manner as the sample-bearing experiments. Manometric analysis of the recovered gases was performed using standard techniques (7). Recovered gases were separated into four fractions; H₂, other gases not condensable in liquid nitrogen, CO₂, and H₂O. The D/H ratios of the recovered H₂ and H₂O were determined by mass spectrometry. Shock pressures were calculated using a one-dimensional impedance match solution, assuming that the shock equation of state for Murchison is the same as that of serpentine (5).

Results and Discussion. Table 1 is a summary of the current and previous results on isotopic fractionation during impact. For both serpentine and Murchison, the lowest shock-pressure impacts ($P_{peak} = 17.5$ and 13.4 GPa, respectively) yielded only H₂O; no H₂ was recovered. Higher-pressure shocks yielded both H₂ and H₂O. The total H₂O driven from serpentine is calculated as the sum of the H₂O plus H₂ collected, less the blank.

The gaseous H₂O-H₂ fractionation factor $\alpha_{H_2O-H_2}$ and the bulk gas-residual solid fractionation factor $\alpha_{bulk\ gas-residue}$ are calculated from the isotopic composition of the starting material and recovered gases, the amount of each gas species recovered, and the total amount of H₂O present in the starting sample. The gaseous fractionation factors $\alpha_{H_2O-H_2}$ suggest equilibration between gaseous species at temperatures significantly greater than the calculated continuum temperatures (1100-1400 K based on equilibrium H₂O-H₂ fractionation (8) versus 400-700 K for continuum calculations). This result may reflect the initiation of devolatilization at 'shear bands' (9).

Figure 1 is a plot of bulk gas-residual solid fractionation $\alpha_{bulk\ gas-residue}$ versus fraction of H₂O devolatilized during the impact. As shock pressure increases, the bulk gas becomes isotopically lighter than the corresponding solid, that is, deuterium is partitioned into the solid residue. The striking feature about Figure 1 is the continuous, overlapping trend exhibited by the serpentine and Murchison data compared to the data for the enriched serpentine. Serpentine and Murchison have similar initial δD_{SMOW} values (-77 and -51 per mil, respectively) whereas the enriched serpentine has an initial value considerably greater (+667 per mil). This suggests that the bulk gas-residual solid deuterium-hydrogen fractionation during impact depends on the initial isotopic content of the sample.

Conclusions. The continuous fractionation trend of Figure 1 allows calculation of the effect of impact-induced isotopic fractionation on the hydrogen isotopic evolution of an accreting planet. Employing an extremely simple accretion scenario in which there is no back-reaction once a planetesimal has impacted the growing planet, the planetary interior acquires a cumulative D/H ratio of 5 to 30 per cent greater than that of the incident material. Thus, this mechanism may not be sufficient to explain the observed 6- to 10-fold enrichments in the D/H ratios of the atmospheres of Mars and Venus relative to the Earth (10,11). However, this previously unrecognized process can cause different reservoirs within an accreting planet to develop D/H ratios significantly different than those of the material from which the body was assembled. Furthermore, the dependence of the impact-induced isotopic fractionation on the value of the initial D/H ratio may be large enough to significantly influence bulk planetary isotopic ratios.

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Table 1. Isotopic results of gas-recovery experiments

Shot Number	Initial Density g/cm ³	Peak Shock Pressure GPa	Total Recovered H ₂ + H ₂ O as % H ₂ O in Initial Sample	$\alpha_{\text{H}_2\text{O-H}_2}$	$\alpha_{\text{bulk gas - residual solid}}$
<u>Serpentine</u> $\delta\text{DSMOW} = -77$ per mil					
867	2.09	17.5	4.2	-	1.01
911	1.98	23.2	12.7	1.15	0.94
915	1.99	26.4	11.9	1.19	0.90
<u>Enriched Serpentine</u> $\delta\text{DSMOW} = +667$ per mil					
994	2.06	21.0	3.8	1.11	0.75
995	2.05	22.5	5.6	1.11	0.79
<u>Murchison</u> bulk $\delta\text{DSMOW} = -51$ per mil					
959	2.45	13.4	2.1	-	1.01
958	2.48	17.7	28	1.18	0.85
949	2.54	20.5	44	1.15	0.87

$$\alpha_{\text{H}_2\text{O-H}_2} = (\text{D/H})_{\text{H}_2\text{O}} / (\text{D/H})_{\text{H}_2}$$

$$\delta\text{DSMOW} = \{[(\text{D/H})_{\text{sample}} / (\text{D/H})_{\text{SMOW}}] - 1\} \times 1000; \text{SMOW, Standard Mean Ocean Water}$$

- Indeterminate, no H₂ in recovered gases

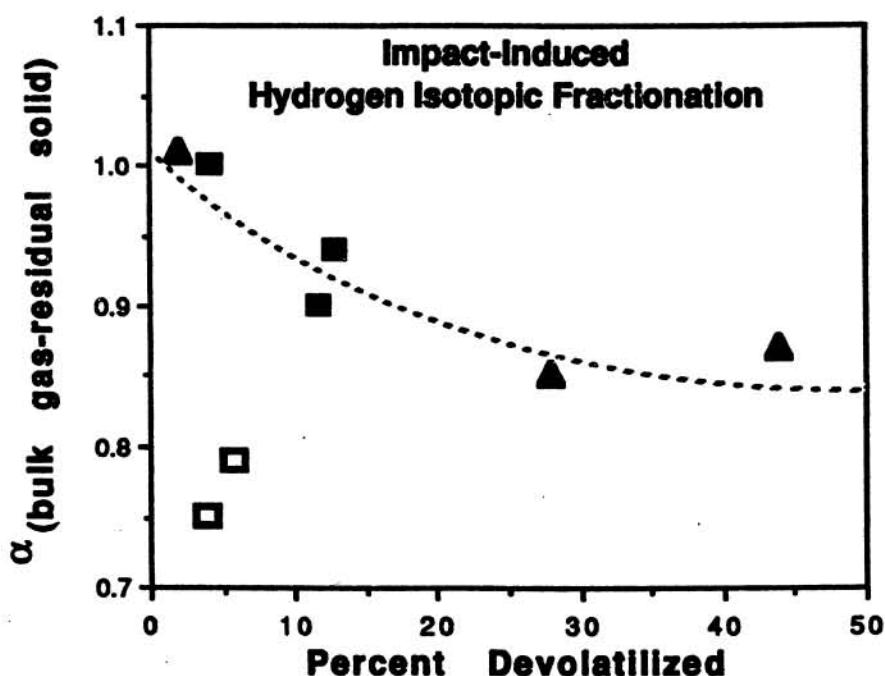


Figure 1. Bulk gas-residual solid fractionation factor $\alpha_{\text{bulk gas-residual solid}}$ versus per cent devolatilized. Filled triangles, Murchison; filled squares, serpentine; open squares, deuterium-enriched serpentine. Data from Table 1. Dashed line is polynomial fit to the data.