WATER/ROCK INTERACTIONS IN CARBONACEOUS CHONDRITES: POSSIBLE FINGERPRINTS FOR IN-SITU COMET-NUCLEUS ANALYSIS

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Introduction. In the "dirty snowball" model for cometary nuclei [1], carbonaceous chondrite meteorites are regarded as useful analogs for the rocky component [2]. Accordingly, preparation for both in-situ chemical analyses of comet-nucleus materials and cometary sample-return missions [3] should benefit from chemical-thermodynamic studies of ice/chondrite mixtures. Toward that end, we report preliminary results of calorimetric experiments with two-component systems involving carbonaceous chondrites and water ice.

Samples and Methods. Two series of experiments were performed in a differential scanning calorimeter (DSC) with homogenized whole-rock powders of the Allende (CV3), Murchison (CM2), and Orgueil (CI) meteorites. In Series 1, 1-5 mg of chondrite was blended with a comparable mass of deionized water and crimp-sealed in air at room temperature (295-298 K) as mud on the bottom of an aluminum container of 20 μl volume; a physically separate drop of pure water (0.5-1 mg), held to the inside of the lid by surface tension, was included as an internal standard. The sample container was placed in a Perkin-Elmer DSC-2C instrument and cooled at 10 K/min to 200 K (including some runs to 100-110 K), followed by re-heating at 10 K/min, under a continuous gas purge of 20 cm^3 Ar/min. DSC heat-flow data were acquired during multiple freeze/thaw cycles. Series 2 used the same procedures except that the chondrite was loaded dry in the bottom of the sample pan and was exposed to water only through vapor released by the overhanging water droplet. Certified mercury (National Institute of Standards and Technology SRM-2225) was used for DSC primary calibration.

Results. *SERIES 1 (LIQUID WATER).* Because freezing of water depends heavily on heterogeneous nucleation, and smooth aluminum is a relatively poor ice nucleator, liquid water undercools substantially before freezing in the absence of a mineral sample. Heterogeneous nucleation of ice reduces undercooling and can be quantitatively expressed as the extrapolated-onset temperature differences, ΔT_m and ΔT_f (Fig. 1). Onset temperatures of melting peaks are more reproducible than those of freezing peaks although the latter offer better peak resolution. With water alone (i.e. no chondrite), the two separate freezing peaks (a_1, c_1) reflect the temperature gradient within the sample container. With a chondrite present, the initial freezing peak (a_1 or chon) shifts to systematically higher temperatures whereas the internal standard peak (c_1 or std) oscillates about its statistically average position (245-250 K). Mean ΔT_m and ΔT_f values ($\pm$ 1σ), respectively, distinguish the three chondrites: 19.5 ± 2.3, 8.3 ± 1.6 (Allende); 16.3 ± 0.7, 11.3 ± 2.0 (Murchison); 5.6 ± 1.6, 12.1 ± 1.6 (Orgueil). The increasing effectiveness of ice nucleation from Allende through Murchison to Orgueil can be understood in terms of the dominant minerals in each meteorite; olivine and pyroxene are poorer nucleators of ice than is serpentine [4]. A second minor "melting" peak in frozen Orgueil, and a systematic decrease in the onset of melting in both Orgueil and Murchison (Fig. 1), further suggest interactions of water with hygroscopic minerals. The previously reported water-complex decrepitation peak at 280-285 K in wet Murchison [5,6] could not be consistently reproduced in the current work and remains enigmatic. *SERIES 2 (WATER VAPOR).* Peaks controlled by the chondrites grew during successive freeze/thaw cycles (Fig. 2), presumably as water vapor was progressively adsorbed and condensed on the initially dry samples. Characteristic signatures are well established after 3-5 cycles. Effects are seen in all three chondrites but are most pronounced for Orgueil, probably as a consequence of its abundant hygroscopic phyllosilicates and sulfates.

Meteoritic and Cometary Applications. Calorimetric fingerprints can be used to distinguish one major type of carbonaceous chondrite from another in the presence of water; further work might extend these fingerprinting capabilities to ordinary chondrites and achondrites. An in-situ DSC experiment on an asteroid or comet could positively identify water ice (and other volatile ices not discussed here) and might also reveal whether the bulk rocky component resembled carbonaceous chondrites or other meteorites.

References: [1] Whipple F. L. (1978) Moon and Planets, 19, 305-315. [2] McSween H. Y. Jr. and Weissman P. R. (1989) Geochim. Cosmochim. Acta, 53, 3263-3271. [3] European Space Agency (1987) Rosetta, SCI(87)3, Noordwijk, The Netherlands, 64 pp. [4] Gooding J. L. (1986) Icarus, 66, 56-74. [5] Gooding J. L. (1986) Lunar Planet. Sci. XVII, Lunar and Planetary Institute, Houston, 269-270. [6] Gooding J. L. et al. (1990) In I. R. Harrison (Ed.), Proc. 19th NATAS Conf., Vol. 1, 9-15.

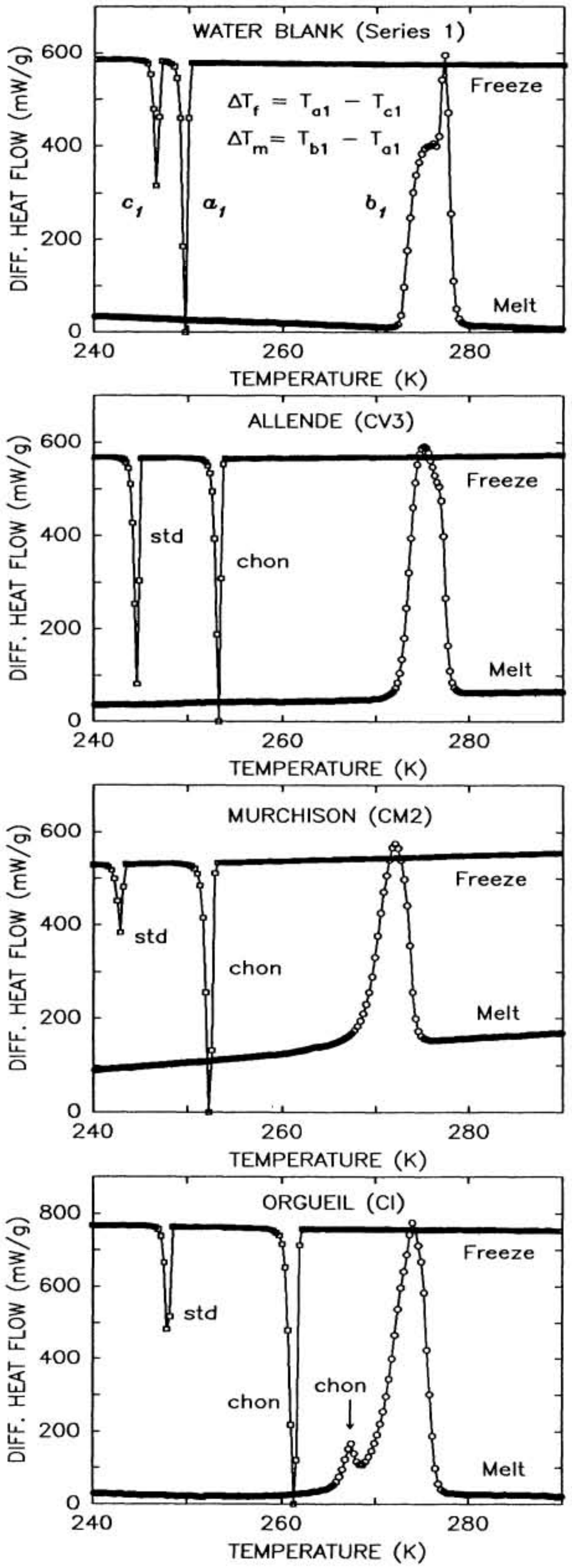

Figure 1. DSC first-cycle freezing and melting of ice in chondrite mud (normalized to total mass of water), with chondrite (chon) and internal standard (std) peaks.

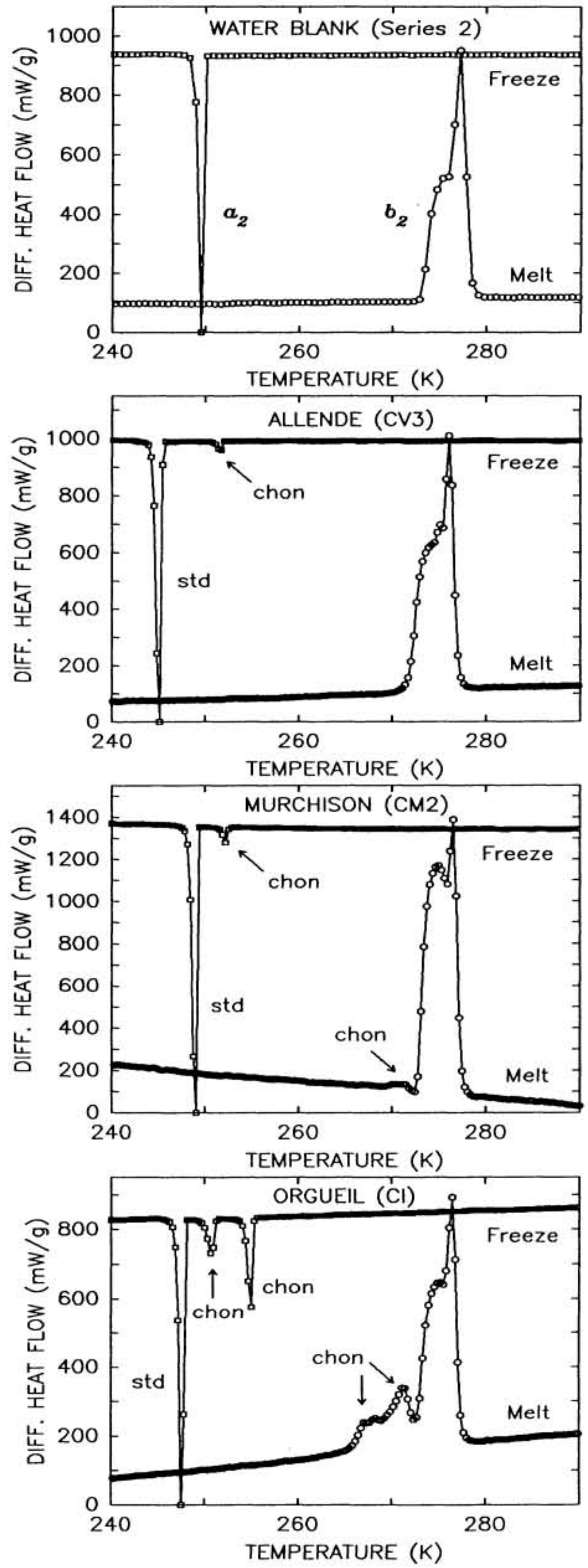

Figure 2. DSC fourth-cycle (fifth cycle for Orgueil) freezing and melting of ice in (initially) dry chondrites exposed to water vapor. Definitions same as for Fig. 1.