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Low-pressure clathrate-hydrate formation in amorphous astrophysical ice analogs

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In modeling cometary ice, some researchers have called upon the properties of clathrate hydrates to explain anomalous gas release at large radial distances from the sun, and the retention of particular gas inventories at elevated temperatures.¹ Clathrates may also have been important early in solar system history.² However, there has never been a reasonable mechanism proposed for clathrate formation under the low pressures typical of these environments. We show for the first time that clathrate hydrates can be formed by warming and annealing amorphous mixed molecular ices at low pressures. The complex microstructures which occur as a result of clathrate formation from the solid state may provide an explanation for a variety of heretofore unexplained phenomena.

We have modified the vacuum and imaging systems of an Hitachi H-500H Analytical Electron Microscope to study mixed molecular ices at temperatures between 12 K and 373 K.³ The resulting ices are characterized by low-electron dose Transmission Electron Microscopy (TEM) and Selected Area Electron Diffraction (SAED). Amorphous H₂O-CH₃OH ices are prepared by vapor-deposition of a 2:1 H₂O:CH₃OH gas mixture onto a thin (<10 nm) carbon film substrate maintained at a temperature of 85 K inside the TEM. On stepwise heating at ~1 K per minute, the 2:1 H₂O:CH₃OH sample remains in the amorphous state until about 120 K at which time a series of powder diffraction rings begin to appear in the SAED pattern. Bright field TEM micrographs in the 120-130 K temperature range reveal the development of a tightly intergrown microstructure of two components, of which one is crystalline. By the time the sample temperature has reached 130 K, 7-8 powder diffraction rings are observed which can be indexed to the clathrate II hydrate structure (space group Fd3m)^{4,5} using an *a*-parameter of 16.28 Å.⁶ All of the diffraction maxima present fit the clathrate II hydrate structure reasonably well, though some maxima are absent. This slight mismatch is likely due to hydrogen bonding between the guest molecules and the H₂O of the clathrate cages.⁶ As the temperature of the ice reaches ~145 K, the amorphous component sublimates away leaving a microporous architecture. The powder diffraction rings in the SAED pattern from the remaining material can be indexed as hexagonal H₂O ice. Laboratory IR spectra from these ices show temperature-dependent band changes consistent with the above interpretation.

The implications of these results for the mechanical and gas release properties of comets will be discussed. Laboratory IR data from similar ices will be presented which suggest the possibility of remotely observing and identifying clathrates in astrophysical objects.

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OBSERVATIONS OF OH IN COMET LEVY (1990c) WITH THE NANCAY RADIO TELESCOPE; D. Bockelée-Morvan, P. Colom, J. Crovisier, E. Gérard, and G. Bourgois (Observatoire de Paris-Meudon).

From mid-June to September 1991, the radio lines of OH were monitored in comet Levy (1990c) with the Nançay radio telescope. The signal was strong, due to favourable excitation of the OH radical, a gas production rate exceeding 10^{29} s^{-1} , and a small earth-comet distance. At the beginning of September, the signal was still enhanced when the comet was observed against the continuum of the galactic centre region; at that moment, the OH lines exceeded 1.5 K in antenna temperature and were the strongest ever recorded in a comet.

The exceptionally good signal-to-noise ratio of these observations allows us to make an analysis of the following topics:

- a) Line shapes and study of the kinematics of the cometary atmosphere: expansion velocity and anisotropic outgassing.
- b) Spatial variation of the line shapes and intensities, in relation with anisotropic outgassing, scalelength of the OH radical, and collisional quenching of the OH maser.
- c) Relative intensities of the four hyperfine components of the OH lines and test of the excitation model.
- d) Zeeman effect in the OH main lines and corresponding limits on the magnetic field averaged over the line of sight in the cometary atmosphere.

Coma Imaging of Comet P/Brorsen-Metcalf at Calar Alto
in Late July to Mid August 1989

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The coma of the periodic comet P/Brorsen-Metcalf was monitored from late July to mid August 1989 at the Calar Alto Observatory/Spain. On 1989/07/28+30 broad-band B and R filter CCD images of the inner coma were obtained with the 3.5m telescope. Large-scale phenomena were observed on 1989/08/04+12+14 with the 80cm Schmidt telescope in B and R filter exposures as well as in integral light. The coma regions of the Schmidt plates were digitized (pixel size 0.86x0.86 arcsec) and calibrated using intensity spots on the plates. In that way they can be directly compared with the calibrated CCD observations (pixel size 0.25x0.25 arcsec). To all CCD frames and to the calibrated scans of a R filter plate on 1989/08/04 and of a B filter plate on 1989/08/12 further image processing (radial renormalisation of the coma brightness distribution) was applied in order to enhance faint structures in the cometary coma.

A narrow ion tail, pointing into anti-solar direction, can be clearly identified in the calibrated and in the processed images. It is particularly prominent in the R filter exposures which might be due to the H₂O⁺ ion emission in this spectral region. While on 1989/07/28 and on 1989/08/04+12+14 a single ion tail of about 5 deg opening angle originated from the cometary nucleus, the triplicate tail of in total about 20 deg opening angle on 1989/07/30 may resemble the convolution of cometary tail rays. A fan-like brightness core of about 7000 km extension is located in the tail vertex on the anti-solar side of the coma.

The CCD images and the B filter Schmidt plate on 1989/08/12 show prominent asymmetries in the isophote patterns of the coma. Radial renormalization of the coma images reveals a broad brightness enhancement (slightly asymmetric in North/South) in the sunward coma region on 1989/07/28 and 1989/08/12. A strong curved jet feature was detected in the coma on 1989/07/30. The brighter sunward coma hemisphere may indicate the higher nuclear activity of gas and dust emission because of the solar illumination. The coma jet (extending at least 30000 km into the coma) may arise from a temporarily, but highly active emission source releasing a significant amount of gas and dust into the coma. The jet curvature can be interpreted as due to the rotation motion of the cometary nucleus. More coma structure observations and jet feature modelling are needed to derive the characteristics (period, axis orientation) of the nuclear rotation of P/Brorsen-Metcalf.