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Si₃N₄ EMISSIVITY AND THE UNIDENTIFIED INFRARED BANDS

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INTRODUCTION

Infrared spectroscopy of warm (about 150-750 K), dusty astronomical sources has revealed a structured emission spectrum which can be diagnostic of the composition, temperature, and in some cases, even size and shape of the grains giving rise to the observed emission. The successful identifications of silicate emission in oxygen-rich objects and SiC in carbon-rich objects are two examples of this type of analysis. Cometary spectra at moderate resolution (e.g. Merrill, 1974, and Campins and Tokunaga 1987) have similarly revealed silicate emission, tying together interstellar and interplanetary dust.

However, Goebel (1987 and 1988) has pointed out that some astronomical sources appear to contain a different type of dust which results in a qualitatively different spectral shape in the 8-13 micron region. Furthermore, the association of the unidentified infrared (UIR) bands at 3.3/3.4, 6.2, 7.7/7.9, 8.6, and 11.3 microns with regions of thermal dust emission and regions of reflection nebulosity has led several authors to attempt to relate that emission to structure in the wavelength dependent emissivity curve of a grain or molecular cluster material. Gas phase species seem to be unlikely, in view of the constancy of central wavelength of the features in such a variety of source types and environments. It is crucial that the wavelength dependence of the emissivity (not extinction, which usually includes significant scattering effects that can qualitatively change the appearance of the spectrum) of proposed dust components be measured in the laboratory over the entire range of wavelengths available for the celestial sources before a positive identification can be made. This poster presents part of an ongoing effort in our laboratory to obtain such data for proposed or likely celestial dust constituents.

THE SAMPLES

Goebel (1987 and 1988) suggested that silicon nitride might be a viable candidate for at least one component of celestial grains, based on the wavelength and the width of the bands at 9-12 microns in the spectra of the celestial sources Nova Aql 1982 and NGC 6572 and the absorption shape in the laboratory spectra of this material. This poster reports the results of an emissivity study of both crystalline and amorphous samples of silicon nitride prepared at the Los Alamos National Laboratory.

The samples were prepared by G. J. Vogt by injecting SiH₄ and NH₃ into a thermal RF coupled plasma which dissociates them. The Si₃N₄ condenses in the cooler plasma flame and is collected in a cyclone separator for analysis. Electron diffraction patterns obtained in the Material Sciences Lab at Aerospace showed a

mix of alpha and beta silicon nitride with some silicon particles in the crystalline sample. The amorphous sample, as expected, gave no diffraction peaks. Energy dispersive analysis of x-rays (EDAX) showed the crystalline sample to be all Si and N with a ratio of about 2:3 to 1:1. The results for the amorphous sample were consistent with this range, but might possibly be consistent with up to 5 % oxygen. No silicon oxide or silicate electron diffraction peaks were seen, however.

The samples were spread thinly on copper blocks and studied in the same vacuum emissivity chamber (see Figure 1) used in earlier studies of interstellar and cometary dust (Stephens and Russell, 1979, Cohen et al., 1980, and Hecht et al., 1986). The spectra were obtained with a circular variable filter (CVF) wheel spectrometer with a resolving power of about 50. The spectra were sampled at about two points per spectral resolution element to better define the structure in the emissivity curve. The samples were heated to 350 K for these studies, although we do not expect the emissivity to exhibit much temperature dependence in this regime. Spectra of black felt and 3M black velvet were used as blackbody references. DISCUSSION

Figures 2 and 3 present the emissivity curves for amorphous and crystalline silicon nitride. Three points should be emphasized regarding the application of these data to the task of identifying the celestial grain components responsible for the UIR bands and the emission features seen in the spectra of Nova Aql 1982 and the planetary nebula NGC 6572. First, the spectra of the amorphous and crystalline samples of silicon nitride are remarkably similar, in direct contrast with the behavior of the spectra of amorphous and crystalline silicates. The only significant difference seen here is that the amorphous silicon nitride spectrum exhibits a long wavelength tail not present in the spectrum of the crystalline emission. Note that although the EDAX data suggested that only the amorphous sample might have had some oxygen (less than or about 5 %) and thus possibly some silicon oxide which could contribute to a 9 micron peak, that peak appears quite the same in the spectra of both samples. Also, the 12.5 micron silicon oxide peak is absent from both spectra, strengthening the contention that silicon oxide is not present in any significant amount.

Secondly, the ratio of the 9 micron to 11 micron emissivity is much higher here than that seen in published absorption spectra (e.g., Nyquist and Kagel, 1971). This effect could be due to particle size effects or the scattering contribution in the extinction data. This makes the spectra reported here look quite different from the Nova spectrum (Gehrz et al. 1984), which shows much less emissivity at 9 microns than at either 11 or 12 microns. The spectrum of NGC 6572 (Willner et al. 1979) exhibits an even higher ratio of 11 to 9 micron fluxes, but the temperature of the grains is much more difficult to determine making it hard to derive the ratio of emissivities. Thus, no strong quantitative statement can be made, but qualitatively the slopes in the lab sample spectra are very different from the slopes in these two astronomical spectra.

Third, the rise toward 3 microns needs to be investigated over a wider wavelength range and at a higher temperature to permit a valid comparison of the silicon nitride emission with the 3.3 micron UIR emission. Again, the shape of the spectrum at these wavelengths in the extinction data is usually dominated by a

scattering component, and a peak such as this would be hard to detect against the scattering continuum.

SUMMARY

The spectra shown here make it appear unlikely that silicon nitride can be identified as the source of the 8-13 micron emission in either NGC 6572 or Nova Aql 1982. The similarity between the general wavelength and shape of the 10 micron emission from some silicates and that from the two forms of silicon nitride reported here could allow a mix of cosmic grains which includes some silicon nitride if only the 8-13 micron data are considered.

A feature is seen near 3.3 microns in the spectra of silicon nitride which requires further study. It might help explain the UIR emission at this wavelength. However, if the spectral shape at shorter wavelengths is different from that seen in the astronomical data, it would permit strong upper limits on the amount of silicon nitride present in spite of the potential ambiguity due to the similarity between the silicate and silicon nitride 10 micron features.

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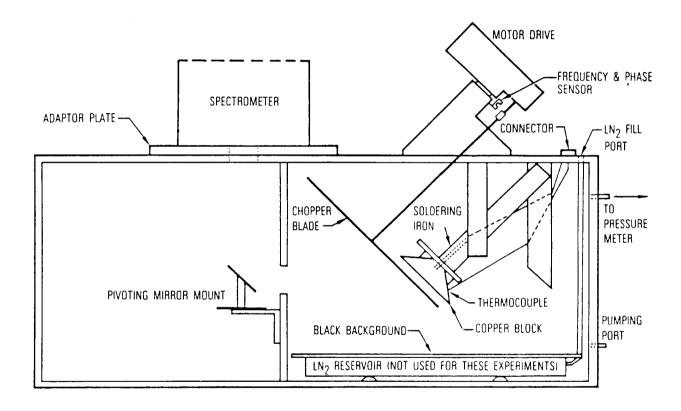


Figure 1. A top schematic view of the emissivity chamber used to make the emission measurements.



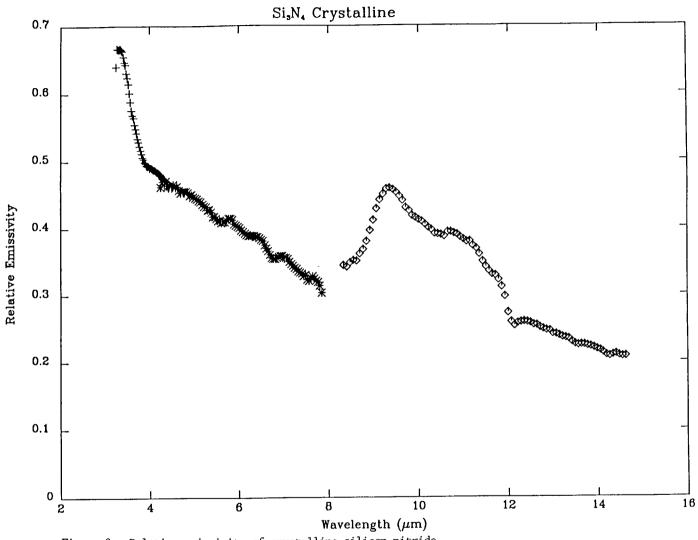


Figure 2. Relative emissivity of crystalline silicon nitride.

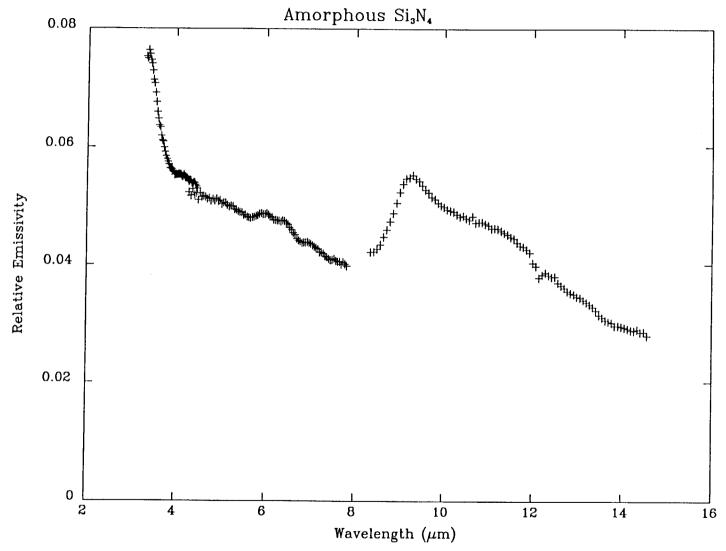


Figure 3. Relative emissivity of amorphous silicon nitride.