THE SiH STRETCH AS AN INDICATOR OF THE OXIDATION STATE OF A COMETARY/ ASTEROIDAL REGOLITH, J. A. Nuth,[1] M. H. Moore,[1,2] T. Tanabe[1,3] (1. Code 691, NASA/GSFC; 2. Chemistry Department, University of Maryland; 3. Institute for Astronomy, University of Tokyo)

We have measured the infrared spectra of amorphous silicate grains and films formed in a variety of laboratory experiments and have found that the energy of the SiH fundamental vibration is extremely sensitive to the chemical environment of the grain in which the silicon is bound. The position of this stretching vibration varies from a high of 2270 cm^{-1} in oxidized grains to only 2110 cm^{-1} in a reduced solid. We have previously[1] suggested that SiH may be an important diagnostic indicator of the chemical state of interstellar grains and here suggest that the position of the SiH fundamental could serve a similar function for asteroidal and cometary regolith materials as well.

In the case of a cometary regolith, it is possible that SiH may be retained in relatively pristine interstellar grains. Spectra in the 2.5–5 micron region of Comet Halley obtained by the IKS-Vega instrument[2] during its fly-by through the coma showed several small bumps in the region between 4.4–4.6 microns which would be consistent with the presence of SiH groups. Unfortunately, no firm conclusions can be reached from these observations due to the poor signal-to-noise ratio of the small features.[2] Another possible mechanism by which regolith grains might pick up hydrogen would be by ion-implantation of the solar wind into regolith silicates. It has already been suggested that ion-sputtering by the solar wind leads to the partial reduction of exposed silicates[3] and it is possible that this gradual chemical reduction could increase the efficiency with which hydrogen is implanted. Laboratory studies of the ion-implantation of H^+ and D^+ into silicon at energies between 70 and 400 keV showed the development of infrared features between 4.5–5.5 and 6.2–7.5 microns due to the formation of SiH and SiD groups, respectively.[4] Although many of these bands could be destroyed by annealing at ~575 K for twenty minutes, a band at 4.63 microns required annealing at 975 K before it disappeared. One might expect these features to survive for very long times at the temperatures expected in the asteroid belt or in comets.

Although the infrared spectra of mixtures are usually equal to the sum of the spectra of the various components in the mixture, more detailed analysis[5] shows that intramolecular factors such as the electronegativity of adjacent atoms or groups could affect the energy of the SiH fundamental.[6] In order to investigate this possibility we produced simple silica grains using two different methods. In the first system, silicon containing residues were prepared from low-temperature films of silane plus water, methane or ammonia which were exposed to 700 keV protons (10^{15} cm^{-3}) at 10 K and then slowly warmed to room temperature.[1] In the second system, silane plus O_2 diluted in a hydrogen flow were allowed to react in a furnace at nominal temperatures between 470–1370 K.[1] Infrared spectra of both types of samples were obtained immediately after formation and at intervals thereafter as the samples were allowed to "age" in various environments; ambient air, vacuum at 370 K and water vapor at 370 K.

Our experiments have unequivocally demonstrated that the frequency of the SiH fundamental is sensitive to the chemical environment in which the silicon atom resides; the higher the electronegativity of the surroundings, the higher the frequency of the fundamental. Furthermore, as the carbon-rich samples oxidize, the shape of the SiH fundamental slowly changes and the peak centroid moves to higher frequency (see Figure 1). With an observable variation from ~2110 cm^{-1} in reducing environments to ~2270 cm^{-1} in oxidizing systems the SiH system should prove to be an excellent indicator of the oxidation state of regolith grains. In addition, it may be possible to use the relative strength of the feature on asteroidal surfaces as a measure of the exposure age of the regolith to the solar wind. In view of the possible utility of this feature in characterizing the state of asteroidal and cometary regoliths, we suggest that an observational search for the SiH feature might prove to be worthwhile.

References

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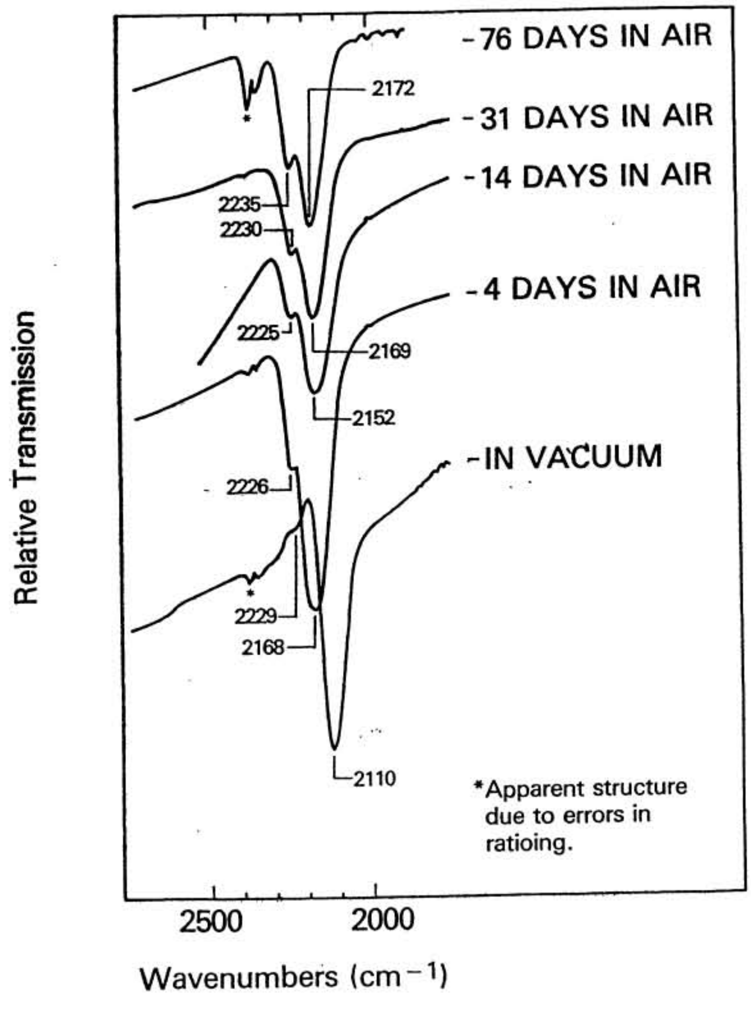

Figure 1. Infrared spectra showing the evolution of an irradiated SiH-CH_4 residue at room temperature in air. Notice the shift to higher energy as the residue oxidizes and the silicon bound to the hydrogen becomes surrounded by an increasing number of oxygen (rather than carbon) atoms.