

STRUCTURES OF AMORPHOUS SILICATE DUSTS SIMULATED BY MOLECULAR DYNAMICS METHOD

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ABSTRACT. Atomic structures of amorphous silicate dusts with the MgSiO_3 composition were simulated by molecular dynamics method as a function of the dust density based on the assumption that the density corresponds to cooling rate of dust formation. The SiO_4 tetrahedra are more polymerized with decreasing density, suggesting phase separation between SiO_2 -rich and MgO-rich components in less dense dusts formed by rapid condensation. A mode of atomic vibration probably due to the Si-O bending is different in the amorphous silicates with different densities. This may cause changes of the 20 μm bands of IR spectra of silicate dusts with different cooling rates.

1. INTRODUCTION

It has been proposed that amorphous silicates are present as interstellar and circumstellar dusts based on IR observations of broad and structureless bands at 10 and 20 μm wavelengths (*e.g.*, [1]). There were extensive laboratory works on amorphous silicates with different compositions to reproduce these IR features (*e.g.*, [2]). However, different IR features are expected for amorphous silicates with the same composition if their structures, which are related to conditions of dust formation, are different.

In the present study, structures of amorphous silicates were simulated by molecular dynamics (MD) method (*e.g.*, [3]) to discuss relation between the structures and IR spectra of the silicate dusts. Special attention was paid on the density of the silicate dust because the density is related to the cooling rate of dust condensation, *i.e.*, the density is expected to be smaller with increasing cooling rate.

2. MOLECULAR DYNAMICS (MD) CALCULATION

The composition of the amorphous silicates was chosen as the enstatite composition (MgSiO_3) based on the cosmic abundance of major

refractory elements ($\text{Mg}/\text{Si}=1.06$) and the inferred SiO_2 content for astronomical silicates (about 50 wt.%; [4]). Calculations were done in the system with a basic cell containing 800 particles with the ionic pair potentials of Kawamura [5]. In the calculations, initial random structures were equilibrated at 4000 K and 3000 K for 2000-3000 steps

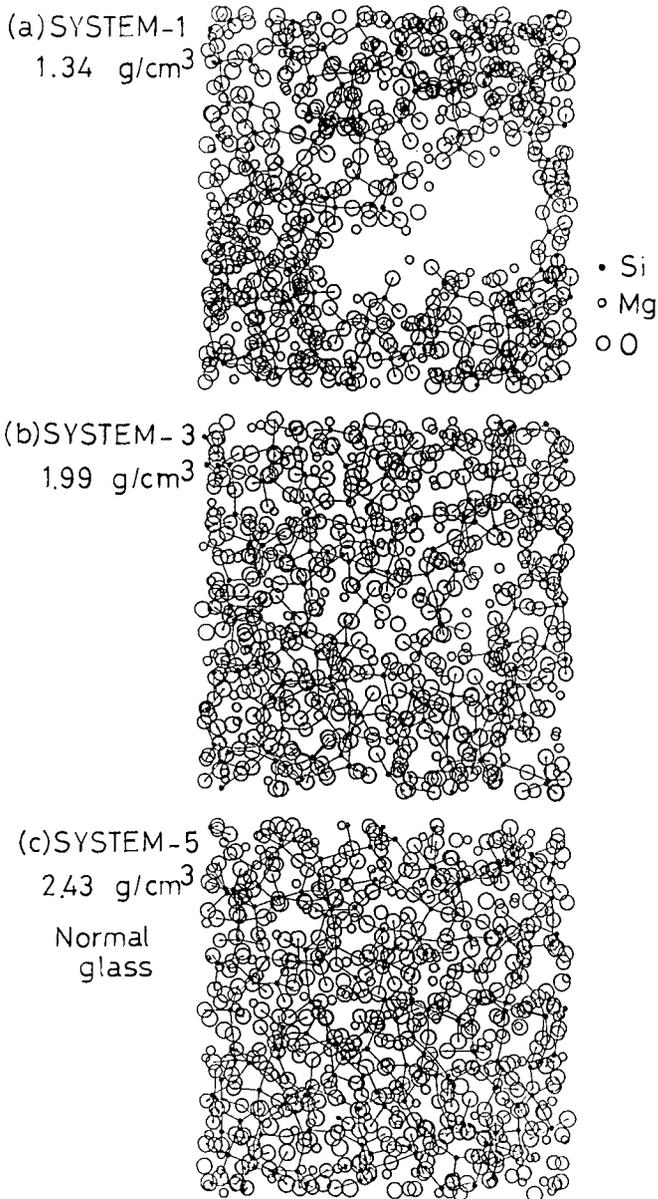


Figure 1. Instantaneous structures of the amorphous MgSiO_3 at 3000 K.

(2×10^{-15} sec/step), and structural properties were obtained at 300 K and 1 bar. Five different systems with different densities (initial density = 1.0, 1.5, 1.8, 2.1 and 2.5 g/cm^3) were examined. The densities of the systems were changed during the calculation by relaxation at the constant pressure (final density = 1.57, 2.02, 2.04, 2.25 and 2.52 g/cm^3 , respectively, at 300 K and 1 bar). The MD calculations were also done for crystals of enstatite and forsterite (Mg_2SiO_4) as references.

3. RESULTS

The structures are almost homogeneous except for the least dense system, where vacant parts are clearly present (Fig.1). The amorphous silicates are composed of SiO_4 tetrahedra and Mg ions. The former are polymerized by sharing their corners. Populations of SiO_4 -dimer and non-bridging oxygens decreases while those of SiO_4 -chains or rings, SiO_4 -sheets and bridging oxygens increase with decreasing density. These results indicate that the SiO_4 tetrahedra are more polymerized in the

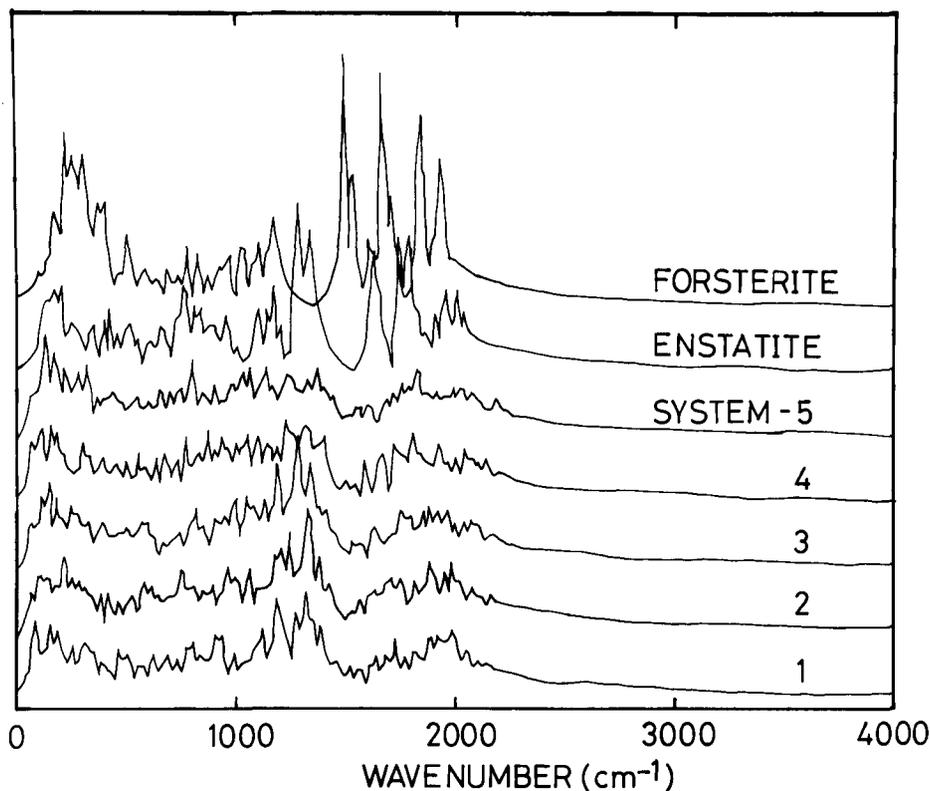


Figure 2. Total vibration spectra of Si atoms in forsterite, enstatite and amorphous MgSiO_3 with different densities (1.57, 2.02, 2.04, 2.25 and 2.52 g/cm^3 for systems 1, 2, 3, 4 and 5, respectively).

less dense silicates than the normal glass (2.5 g/cm^3). The polymerization suggests separation between SiO_2 -rich (and thus MgO -poor) and SiO_2 -poor (MgO -rich) portions in the amorphous silicates.

Total vibration spectra of the Mg, Si and O atoms were obtained to discuss IR spectra (Fig.2). Peaks for O and Si spectra at $1600\text{--}2000 \text{ cm}^{-1}$ are considered to correspond to the Si-O stretching modes because the correlation between the O and Si spectra is good. In fact, shift of the locations of the $1600\text{--}2000 \text{ cm}^{-1}$ peaks of the enstatite and forsterite crystals is consistent with that of the IR observations. The deviation of the wave numbers from the real Si-O stretching modes ($800\text{--}1200 \text{ cm}^{-1}$) is due to the steep potentials used in the present calculation.

Peaks at $800\text{--}1400 \text{ cm}^{-1}$ are considered as the Si-O bending modes. A peak at about 1300 cm^{-1} is distinctive only in the systems with the density of about 2.0 g/cm^3 or less and enstatite crystal (Fig.2).

4. DISCUSSION

The present MD simulation shows that the structures of the amorphous MgSiO_3 and their time variations (vibration spectra) are different with different densities. If amorphous silicate dusts with low densities were formed by rapid condensation, their structures and thus their IR spectra must be different from those of normal glass. If the $800\text{--}1400 \text{ cm}^{-1}$ peaks (Fig.2) are due to the Si-O stretching and every vibrational peaks are active for the IR adsorption, the peak location of the $20 \mu\text{m}$ band shifts towards higher wave number with decreasing density or increasing cooling rate. This is consistent with implication from the experiments of Koike and Tsuchiyama (1991).

References

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