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SILICATE DUST: FROM OBSERVATIONS TO THE LABORATORY

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Abstract. Silicate dust is ubiquitous in the Interstellar Medium (ISM). It is thought to be, primarily, the product of the mass loss phase of oxygen rich Asymptotic Giant Branch (AGB) stars and to be transported into the ISM by stellar winds. In the ISM, dust is subjected to many processes which could explain its observed structural forms. In order to explain the different observed structures (amorphous, crystalline and often both), we have developed in the laboratory an experimental protocol simulating the different stages in the dust life-cycle (including formation, annealing and irradiation experiments). The synthesized and processed samples are studied by infrared (IR) spectroscopy and electron microscopy in order to infer their chemical, structural and micro-structural characteristics. The results are compared to astronomical observations and to Interplanetary Dust Particles (IDPs) collected in the upper Earth atmosphere by NASA.

1 Introduction

Silicate is one of the major components of cosmic dust. Before the results from the Short and Long Wavelength Spectrometer instruments (SWS, LWS) on board the Infrared Space Observatory (ISO) it was largely thought that all silicate dust in space is amorphous. The extensive results provided by ISO revealed the existence of different forms of silicates (crystalline, amorphous, and/or both of them and a variety of different crystalline minerals e.g. forsterite, enstatite and diopside) depending on the astrophysical environment where they are detected and thus on the physical conditions to which they are subjected.

Crystalline silicates have been observed in evolved star outflows (Waters et al. 1996, Molster et al. 2002), some comets (Crovisier et al. 1997; Wooden et al. 1999, 2004) and young stellar environments (Malfait et al. 1998, 1999). The co-existence of both amorphous and crystalline silicate phases in AGB M-type stellar envelopes is also observed (Kemper et al. 2001). An upper limit of 1% on the abundance of crystalline silicates in the ISM was determined (Kemper et al. 2004). Recently the Spitzer Space Telescope (SST) IR Spectrometer recorded spectra few minutes after the impact of Deep Impact onto the comet 9P/tempel 1 and revealed the existence of crystalline olivine and pyroxene in the ejecta of the excavated material from the comet's interior (Lisse 2006). Finally, with the Mid-IR interferometer instrument MIDI on the Very Large Telescope Interferometer (VLTI), well suited to the study of stellar disks at high spatial resolution, it will be possible to study the dust properties as function of the distance to the central star (Van Boekel et al. 2006).

Numerous experimental studies have been devoted to an investigation of the structural modification of silicate samples after ionic irradiation (Demyk et al. 2001, 2004; Carrez et al. 2002; Jäger et al. 2003; Brucato et al. 2004). These studies have shown that ion irradiation of crystalline silicates, performed at energies and fluences similar to those expected in interstellar shocks, rather efficiently leads to the amorphization of the samples.

Other experimental investigations have been devoted to the thermal annealing effects on the structural modification of silicate samples (Hallenbeck et al. 1998; Fabian et al. 2000; Brucato et al. 2002). These studies led to the determination of the activation energy for crystallization and find a value of $\sim$ 40 000 K.

Currently little has been done on the further evolution of irradiated silicates, in particular when this dust is involved in the formation of new stars. During this stage, dust temperature may significantly increase and lead to crystallization of the amorphous precursor, the silicates coming from the ISM. The role of previous irradiation in

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modifying the thermodynamical properties of the grains has only recently been considered (Djouadi et al. 2005). For this study, we developed an experimental protocol aiming to evaluate the influence of ion irradiation on the value of the activation energy of crystallization of a silicate (hereafter called the recrystallization activation energy). Such a parameter can help to constrain the silicate grain evolution in the ISM.

2 Experiments

Our experimental protocol had as a first aim, the determination of the recrystallization activation energy of amorphous silicate samples produced by the ion irradiation of crystalline olivine. We used the San Carlos olivine as a precursor ($Mg_{1.8}Fe_{0.19}Ni_{0.01}SiO_4$) and we evaporated it using an electron beam and deposited it onto a diamond substrate (for the IR spectroscopy). We simulated different stages in the life-cycle of dust, from its formation around evolved stars to its incorporation into the stellar disks around new stars. The annealing experiments were performed in a tubular furnace under vacuum at temperatures, typically between 600° and 1000°C, in order to simulate processes at high temperatures in the inner disks of new stars.

All the steps of our protocol (i.e. synthesis, thermal annealing, irradiation) are monitored by IR spectroscopy in order to follow the induced structural modification. The micro-structural state of the samples are investigated by electron microscopy. We coupled these two analytical techniques in order to derive the maximum possible information on the evolution of the samples under treatment. The configuration of the sample (thin films on a diamond substrate) is not suitable for the TEM analyses. For this we have to scrape off a part of the film and transfer it to a suitable substrate (carbon) for the TEM analyses.

The details of the experimental protocol and analytical techniques are given in our papers (Djouadi et al. 2005, Davoisne et al. 2006).

3 Some principal results

3.1 IR spectroscopy results, Djouadi et al. 2005

Figure 1 shows the typical IR spectra obtained. We can see the IR signatures due to the Si-O bonds around 10 μm (stretching) and 20 μm (bending) and their evolution with annealing time at a temperature of 750°C.

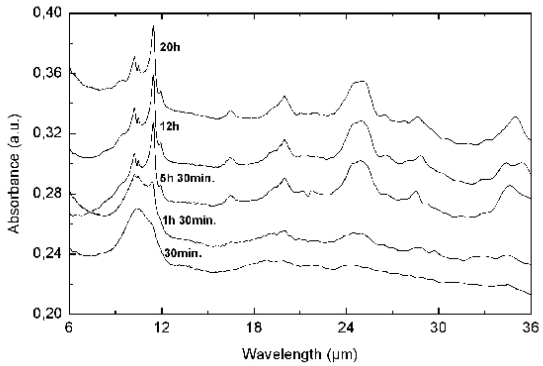

Fig. 1. Evolution of the bands with annealing time for a fixed temperature of 750°C

For the determination of the recrystallization activation energy, we used an Arrhenius law relating the crystallization time t to the activation energy E_a via the temperature T (given in K).

$$t = \nu^{-1} exp\left(\frac{E_a}{kT}\right) \tag{3.1}$$

where k is the well known Boltzmann constant and ν is the vibrational frequency of the lattice.

Using a qualitative analysis of the IR spectra, we extracted a value of the recystallization activation energy of the pre-irradiated silicates. The determined value in this work is compared below in table 1 to the activation

energies of crystallization which have been determined by other authors who did not consider any irradiation step.

Table 1. Comparison of the recrystallization activation energy, denoted as E_a/k (K), extracted from our work along with the crystallization activation energies determined by other workers

Reference	E_a/k (K)
Hallenbeck et al., 1998	45500
Fabian et al., 2000	39100
Brucato et al., 2002	40400
our work, Djouadi et al, 2005	41700 ± 2400

From table 1 we can see that no significant difference is observed, our value is similar to the other derived values; an expected modification of the crystallization enthalpy due to the irradiation is not observed. The silicate recrystallizes as the same temperature as crystallization from the amorphous phase without irradiation. This result suggests that the IS grains when incorporated into protostellar disks have to be processed at high temperatures in order to crystallize and for this, they must be in the inner regions (in the vicinity of the star, i.e. some AU distant). Finally, in order to explain the presence of crystalline silicates in cold environments we have to consider the redistribution of matter between the inner and cooler (outer) regions, possibly due to turbulence as proposed in the model of Bockelée-Morvan et al. (2002).

3.2 TEM analyses results, Davoisne et al. 2006

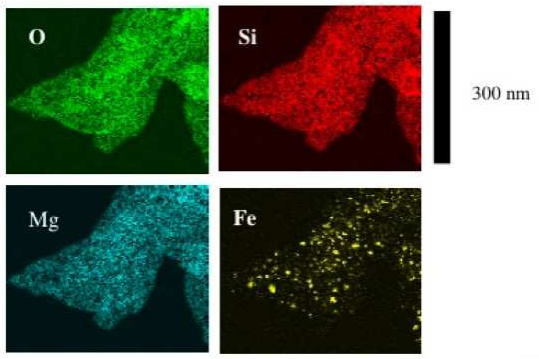

Fig. 2. Distribution of the elements in the matrix of the analyzed sample

TEM analyses have revealed the presence of nano-particles of pure iron (2-50 nm in diameter, see figure 2) enclosed in an amorphous matrix when the amorphous silicate sample is heated at relatively low temperature 600 ° C. These iron beads are also observed in crystallized or partially crystallized samples when the experiments are performed at higher temperatures. We interpret the formation of these inclusions in terms of a redox reaction occurring during heating in vacuum and due to the carbon in the atmosphere due to the pumping system. The presence of these metallic inclusions implies that the remaining amorphous silicate matrix becomes free of iron. Figure 3, obtained from X-ray energy dispersive technique (EDX), shows that no iron is found inside the matrix. The totality of the initial iron has been reduced to form the metallic nano-particles.

From these results we conclude that the observed microstructures closely resemble those of GEMS (Glass Embedded with Metal and Sulfides) found in chondritic IDPs for none crystallized samples. Since IDPs contain abundant carbonesceous matter, a solid-state reduction may have occurred during heating in the hot inner regions of the protosolar disk, leading to the formation of metallic nano-sized precipitates.

Locking iron as metallic particles within a silicate matrix may explain why astronomical silicates mostly appear, observationally, to be Fe poor. Finally, and on account of these metallic inclusions, we can assume that

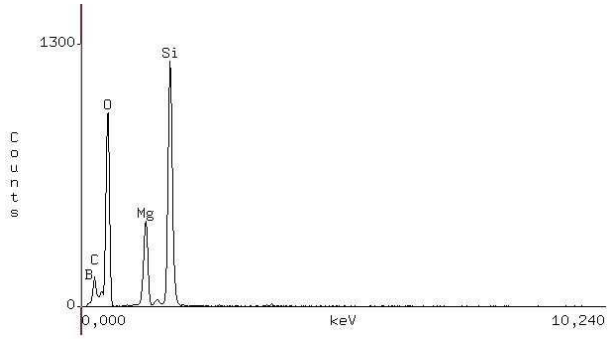

Fig. 3. Chemical analysis of the silicate matrix: No iron is found inside it

the samples will have magnetic properties and could be at the origin of the grains responsible for the polarization of starlight due to their alignment with the interstellar magnetic field. For this, we are currently analyzing the data obtained from magnetic susceptibility measurements on our samples. Results will be discussed in a forthcoming paper (Djouadi et al. in preparation)

4 Conclusion

We have developped laboratory experimental protocols aimed at simulating various stages in the life-cycle of dust. The combination of two complementary analytical techniques, IR spectroscopy and TEM, led us to access to the structure of the lattice, the micro-structure of the samples as well as their chemistry. The synthesized samples exhibit strong similarities with natural material, in particular the presence of metallic nano grains embedded in an amorphous matrix like the GEMS in IDPs. However, for better simulations we have to consider the coexistence of carbon phases with the minerals in the analogues as well as the role of gas-grain interaction on the microstructural evolution of silicates. These require modifications of the experimental set up and could be envisaged in the longer term.

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