

THE INHIBITION OF PYROXENE FORMATION DURING LOW TEMPERATURE ANNEALING OF COSMIC SILICATES.

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Introduction: Infrared observations indicate that a dominant fraction of the condensed matter found in the interstellar medium (ISM) is amorphous. On the other hand, similar spectroscopic studies show that crystalline silicates are abundant in protoplanetary disks. Because the ISM dust is the precursor material of protosolar cloud, it appears that structures and compositions of the silicate dust are strongly thermally processed in such environments.

Experiments: Glass powders and sol-gel materials were used as analogs of cosmic dusts. Various compositions in the systems CaO-Al₂O₃-SiO₂ (CAS), CaO-MgO-SiO₂ (CMS) were studied in order to derive systematic trends for cosmochemically-relevant compositions (pyroxenes, plagioclase, melilite, olivine). Different starting materials allowed us to assess the effects of the surface/volume ratio and of the structure of the amorphous precursors on microstructures and mineralogy of the crystalline phases formed. Samples were heated in a tubular furnace, in air, for 30 min to 15 days at temperatures ranging from 40°C below to 100°C above the glass transition temperature of starting glasses (T_g's). After experimental runs, samples were analyzed by Scanning Electron Microscopy (SEM), Electron Microprobe (EMP) and Transmission Electron Microscopy (TEM).

Results: All our experimental data show that sub-solidus crystallization is accompanied by a significant redistribution of cations, especially magnesium and calcium. In the CAS ternary diagram, crystals are essentially enriched in calcium while the Al/Si ratio approaches that of the parent liquid. With increasing crystallization temperature (T), the transition to phase composition expected near the solidus takes place via a gradual change of Al/Si, this change being a systematic function of (T-T_g). This feature extends to other systems. It may be explained by the mobility decoupling of cations, of which the network modifiers (Ca and Mg) becomes several orders of magnitude faster than that of the network formers Si and Al around T_g. In addition this crystallization is kinetically favored because the enrichment of the nuclei in low-field strength cations decreases the relative number of strong bonds (i.e. Al-O and Si-O), which need to be reordered to reach a crystalline structure.

Discussion: A critical consequence of this diffusion decoupling is the inhibition of pyroxene formation during annealing of amorphous silicates. From enstatite and diopside starting materials, forsterite, SiO₂, merwinite, akermanite and non-stoichiometric diopside are respectively formed. These phases are indeed enriched in CaO and MgO compared to the starting materials. These results are reminiscent of condensation studies that demonstrated that equilibrium condensation could not account for the formation of enstatite. They may also be consistent with some presolar mineral assemblages recently observed. Comparing our results to previous works on condensates, smokes and gels, it appears that pyroxenes are extremely difficult to produce whereas they are dominant minerals in protosolar disks together with olivines. One way to reconcile experiments, calculations and IR observation is to assume that the average annealing temperature was significantly higher than those commonly proposed to favor equilibrium crystallization of silica-rich phases.