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From Molecules to Cosmic Silicate Grains: An Experimental Study of Cluster Intermediates and Growth Mechanisms

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Abstract. It has long been advocated that interstellar silicate grains are amorphous with non stoichiometric elemental ratios, and circumstellar outflows from evolved stars are the primary contributors to transitions from molecules to such type of silicates. Currently there is a paucity of experimental studies of fast chemical reactions which may enable initiation of the direct vapor phase growth of silicates pertaining to processes operative in interstellar clouds. Based on laboratory smoke condensation experiments and analysis of end products, it was conjectured that formation of pure $(SiO)_x$ cluster should provide the first surface for the kinetics of condensed phase growth of amorphous unequilibriated silicates in the circumstellar envelopes surrounding M-type giants. Evidence was presented that growth mechanisms involved formation of silicates through the loss of Si atoms during the nucleation and growth of SiO polymers. SiO is one of the major reactive molecular species comprised of cosmically abundant mineral forming elements. An experimental study of the kinetics of the elementary association reactions underlying the nucleation process of SiO is critical in order to understand the formation processes of silicate grains.

A preliminary study of the growth mechanisms and associated chemical evolution has recently been undertaken in our laboratory. Samples of silicon and its oxides have been laser ablated under a series of different ambient environments, and during the course of laboratory experiments an unexpected chemically anomalous composition of $(SiO)_x(SiO_2)_y$ was observed in both neutral and ionic clusters. Our findings are consistent with a mechanism in which these complex molecular species evolve upon the clustering of SiO_2 to nucleation centers comprised of $(SiO)_x$ clusters.

The present findings offer an expectation that a quantitative understanding of the chemistry of formation of silicate grains of widely varying composition in astronomical regions may be reached through laboratory experiments of the type reported here. Such expectation awaits future experimental studies of the interplay between structure and dynamics in small gas phase clusters.

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