Homogeneous Nucleation from Vapor in Al-O Binary System

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A gaseous molecule can have a chemical formula that is inconsistent with its ability to act as a building block for crystals; for example, hot AlO and AlO_2 vapors cannot solidify directly to crystals. The contributions of such gases to homogeneous nucleation processes are unknown. Using in situ IR spectroscopy on nucleating nanoparticles, we show that a liquid-like behavior of nucleating nanoparticles in the Al–O system induces immiscible phase segregation. In an oxygen-deficient atmosphere, aluminum oxide nanoparticles formed with a unique shape composed of an Al metal head and an anisotropic Al_2O_3 crystalline tail. The anisotropic nanoparticles are larger than Al_2O_3 nanoparticles formed in a sufficient oxygen condition. Fewer nuclei may be available in the supersaturated gas, indicating that homogeneous nucleation is initiated by oxygen-bearing species. The nuclei grow by incorporation of Al-bearing species, changing the composition of the particle as a whole toward an Al-rich material. In situ IR measurements revealed that the degree of anisotropy continues to increase, suggesting that oxygen-bearing species also formed by oxidation on the surface of the molten Al-rich head. The scenario is supported by quantum-chemical calculations indicating that oxygen-deficient $(AlO)_n$ clusters as few as 16 molecules induce aggregation of Al atoms. Because Al metal has a significantly lower melting temperature than Al_2O_3, the immiscibility of the nucleating nanoparticles leads to vapor–liquid–solid growth. Astronomical implication will be given in the presentation.

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