Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

PPS025-05 Room:101 Time:May 23 15:15-15:30

Kinetics of spinel formation under circumstellar conditions

Mariko Harada[1*], Aki Takigawa[2], Shogo Tachibana[2], Hiroko Nagahara[2], Kazuhito Ozawa[2]

[1]Earth and Planetary Physics, Univ. Tokyo, [2]Earth and Planet. Sci., Univ. of Tokyo

Spinel ($MgAl_2O_4$) is one of the most abundant presolar oxides found in primitive chondrites with highly unusual oxygen isotopic compositions compared to solar-system materials. Presolar corundum and spinel grains are considered to be condensates in outflows from oxygen-rich AGB stars and/or supernovae. It has been reported that Mg/Al ratios of some presolar spinel grains are lower than the stoichiometric ratio (Choi B.-G.- et al., 1998). Spinel can form through the reaction between pre-existing corundum and Mg gas, and the non-stoichiometry of presolar spinel and the presence of presolar corundum may imply that corundum and Mg gas did not react completely. In order to discuss the origin of presolar spinel and evolution of refractory materials in circumstellar environments, it is important to understand the kinetics of spinel formation under low-pressure circumstellar conditions. In this study, we conducted spinel formation experiments through a reaction between corundum and Mg+O gas (Al_2O_3(s) + Mg(g) + O(g) = $MgAl_2O_4$(s) in vacuum.

An MgO pellet as a gas source of Mg and O was put in the bottom of an alumina crucible. The alumina tube was set in a vacuum chamber and evacuated to ˜10^{-3}-10^{-4} Pa, and then heated at desired temperatures (1640, 1590, and ˜1450 degree Celsius) for desired durations (6-695 h). The wall of the alumina tube was almost isothermal at 1640 and 1590 degree Celsius, and a small temperature gradient existed at 1450 degree Celsius, where it was 1470 and 1420 degree Celsius at the bottom and top of the tube, respectively. The inner wall surfaces and the cross sections of reacted alumina tubes were observed with FE-SEM, and their chemical compositions were determined by EDS and EPMA. Thicknesses of the reacted layer were measured at different heights from the bottom.

A spinel layer was formed on the inner wall of the alumina tube under all the experimental conditions. The thickness of the layer was largest at the bottom, and became smaller with increasing the distance from the bottom. The thickness of the spinel layer increases linearly with time. The Mg/Al compositional profile of the spinel layer showed that the layer was depleted in Mg, and the typical ratio of Mg and Al was Mg:Al = 0.72:2.18, 0.61:2.26 and 0.59:2.27 at 1450, 1590, and 1640 degree Celsius for O=4, respectively.

The linear increase of the thickness of the spinel layer with time suggests that the spinel formation rate is controlled by the surface chemical reaction, i.e., the reaction rate can be expressed by the Hertz-Knudsen equation. In order to obtain the condensation coefficient for spinel formation, we developed a model for steady-state diffusion of Mg gas inside the alumina tube under the molecular flow conditions. We fitted the growth rates of the spinel layer with the model, and obtained the spinel reaction coefficient of ˜0.02 and supersaturation ratio (S) of ˜5 at 1450, 1590, 1640 degree Celsius. Therefore, we conclude that only ˜2 percent of colliding Mg gas can react to form spinel in the reaction of Al_2O_3(s) + Mg(g) + O(g) = $MgAl_2O_4$(s) at a low super saturation ratio (S˜5),.

The obtained condensation coefficient was applied to spinel formation in an expanding mass-loss wind around a cool-low-mass evolved star with gas of the solar composition. The mass-loss velocity where spinel grains form may be much lower than the terminal velocity of the wind (˜10 km/s) and vary with time. The reaction efficiency between corundum and Mg+O gas changes with the wind velocity: corundum grains (1 micron) react almost completely with gas to form spinel in the case of wind velocity of 0.01 km/s, while little reaction occurs in the case of faster wind (1 km/s). Spinel formation under kinetic conditions in mass-loss winds with various velocities therefore may result in the variation of circumstellar oxides, which is consistent with the presence of both presolar spinel and corundum in the solar system materials.