論　文　の　要　旨

題　目: FINE PARTICLE SYNTHESIS IN TUBULAR FLAME SYSTEMS

（　管状火炎システムによる微粒子合成　）

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In this dissertation, we apply a tubular flame to the reaction field in gas-phase synthesis of fine particles. Flames have been used to fabricate various functional fine particles and devices, and it is important to predict the temperature and gas concentration to control particle characteristics with high energy efficiency even in the so-called "dirty" gas phase generated by combustion. Tubular flame combustion, a new combustion technology, has high thermal and aerodynamic stability, and the flame temperature and gas composition can be controlled. In addition, the tubular structure is convenient and can be easily integrated into various gas-phase processes. As a first step toward the development of a particle synthesis process using tubular flame combustion, we developed a new tubular flame burner, investigated the effects of various combustion parameters on particle formation, and clarified the detailed flame structure by spectral analysis of chemiluminescence. A brief description of each chapter in this dissertation is given below.

Chapter 1 introduces the current research background for flame aerosol synthesis of nanostructured particles. A review of gas-phase combustion synthesis and burner types in previous research is also presented in this chapter.

In Chapter 2, we describe the development of a tubular flame burner for particle synthesis and investigate the synthesis of tungsten oxide nanoparticles by efficient use of combustion energy. When synthesizing fine particles using the flame-assisted spray pyrolysis method—which is one of the flame aerosol synthesis methods—submicron-sized particles are easily obtained owing to the size of the raw material droplets. However, by using a high-temperature tubular flame, energy can be supplied to the particles efficiently. As a result, the gasification of the particles is accelerated and they renucleate in the gas phase, resulting in the formation of tungsten oxide nanoparticles with a primary particle size of 5-20 nm.

Chapter 3, describes the successful preparation of tungsten metal nanoparticles using fuel-rich methane/air tubular flames due to the effect of reducing species in the combustion gas. Because the tubular flame structure has high-temperature combustion gas inside and low-temperature unburned gas outside, the produced particles are not affected by the unburned gas and react in the combustion gas with a controlled composition and temperature. When the composition of the combustion gas was examined under various equivalence ratio (Φ) conditions, the oxygen concentration approached zero for $\Phi > 1.0$, while the concentration of CO, a reducing species, increased significantly. Under the condition $\Phi > 1.0$, tungsten trioxide was synthesized as described in the previous chapter. In addition to tungsten trioxide (WO_3), the crystalline phases of tungsten suboxide ($WO_{2.72}$) and tungsten metal (W) were precipitated. Furthermore, increasing the residence time of the particles in the tubular flame accelerated the reduction effect and caused the WO_3 and $WO_{2.72}$ phases to disappear, and only the W phase was observed. The particle size decreased with increasing residence time, and the primary particle size of the tungsten metal particles was 5--10 nm. It was shown that the oxidation state and particle size of the flame-made particles could be widely controlled using the controlled reaction atmosphere of tubular flame combustion and by adjusting the residence time.

In Chapter 4, a direct spray type tubular flame burner was developed and its flame structure was analyzed to establish a particle synthesis system by liquid fuel combustion using a tubular flame burner. Liquid fuel (ethanol) was sprayed into the tubular flame burner from the axial direction using a two-fluid nozzle capable of transporting liquid at a high flow rate, and the characteristics of the resulting flame were evaluated. When ethanol was sprayed onto the burner with a tubular flame, a uniform tubular flame was observed. The flame appearance was observed while varying the overall equivalence ratio, and it was shown that combustion was possible for a wide range of equivalence ratios. Temperature measurements showed that the flame structure comprised high-temperature gas inside and low-temperature gas outside, and exhibited the temperature distribution characteristics of tubular flames.

Furthermore, the detailed flame structure and the effect of tubular flame combustion were investigated by measuring the intensity distribution of the chemiluminescence of the flames. It was found that when the equivalence ratio of the tubular flame was outside the combustible range, the base of the flame was lifted even when the overall equivalence ratio was in the combustible range. In contrast, if the tubular flame was in the combustible range, a stable flame could be formed from the burner base.

In Chapter 5, we summarize the results obtained in this study and detail the prospects of the tubular flame system for particle synthesis.