

Powder Characteristics and Sintering Behavior of SiO₂ Coated BaTiO₃

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Abstract

The Powder characteristics and sintering behavior of SiO₂ coated BaTiO₃ were studied. Silica coated BaTiO₃ powders were prepared by sol-gel method. The particle size of the BaTiO₃ powders were ~35 nm and the thickness of the SiO₂ coating layer was ~5 nm. As the SiO₂ content increased, the SiO₂ layers improved the powder dispersion. The Zeta potential of SiO₂ coated BaTiO₃ was getting close to that of pure silica with a more negative charge, compared with that of the uncoated BaTiO₃. The onset temperature of shrinkage curves shifted to higher temperatures with increasing SiO₂ contents

Keywords: BaTiO₃, SiO₂, Nano, Coating, Zeta potential

1. Introduction

Ferroelectric BaTiO₃ is the material of choice for multilayer ceramic capacitor (MLCC) manufacturing. To fabricate thinner dielectric layers for increasing volume efficiency, nano-sized BaTiO₃ powders are essentially required. As the BaTiO₃ particle size gets smaller, the uniform distribution of additives becomes more difficult. Chemical coating techniques are effective in enhancing the uniform distribution of additives along the grain boundaries. The advantages of coating methods include improvements in the dispersability of powders, the introduction of a uniform incorporation of additives and microstructure control [1-2]. Hence, various coating techniques have been developed [3-5]. SiO₂ coating is an important way to improve the sintering behavior of BaTiO₃ powders because SiO₂ is known to be an effective sintering aid [6]. The sol-gel method is widely used to form a silica layer coating on powders using silicon alkoxides. In this paper, the effects of silica coating on the powder characteristics and sintering behavior of nano sized BaTiO₃ powders will be discussed.

2. Experimental and Results

Nano sized BaTiO₃ powders were prepared using the liquid mix method developed by Pechini [7]. The silica coating on barium titanate powders was achieved via sol-gel method and the amount of SiO₂ addition was 0.5 and 5.0 wt%.

The primary BaTiO₃ particles were heavily agglomerated with uniform particle sizes of about 30~50nm. The average size of BaTiO₃ particles was observed to be about 35nm. Figure 1 show TEM micrographs of bare BaTiO₃ powders and SiO₂ coated BaTiO₃. The thickness of the SiO₂ coating layer was about 5nm. The uniform coating of silica on

BaTiO₃ was formed by hydrolysis and condensation reactions of TEOS.

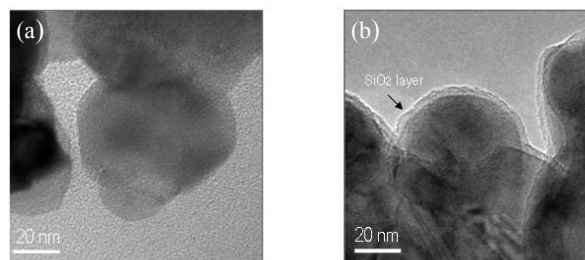


Fig. 1. TEM image of uncoated (a) and 5.0wt% SiO₂ coated BaTiO₃ (b).

Figure 2 shows zeta potentials of BaTiO₃, SiO₂ and SiO₂ coated BaTiO₃ powders as a function of pH value. Above pH 3.0, the zeta potentials of SiO₂ coated BaTiO₃ powders are more negative than the uncoated BaTiO₃. This confirms that the surface of BaTiO₃ particles was coated by SiO₂ layers, which caused the change of the surface characteristics of BaTiO₃ powders and improved the dispersion of BaTiO₃ primary particles. The disappearance of powder agglomeration with increasing SiO₂ content is thus believed to be due to the increase in electrostatic repulsions between the coated SiO₂ layers on the surface of the BaTiO₃ particles.

Figure 3 exhibits the shrinkage curves of BaTiO₃ as a function of temperature for various silica contents. The SiO₂ coated BaTiO₃ powders began to shrink at higher temperatures than the uncoated BaTiO₃. The SiO₂ coating layer could prevent BaTiO₃ particles from contacting directly, resulting in higher sintering temperatures. Dukhin et al. reported a deactivated sintering by the coating of particles [8].

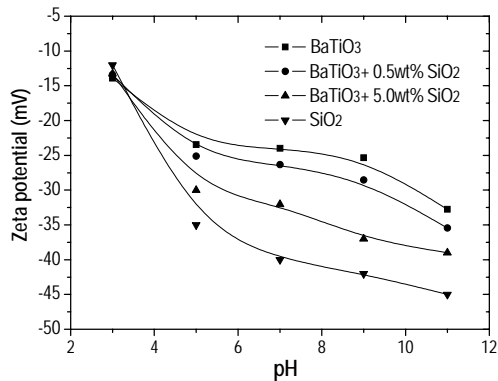


Fig. 2. Zeta potentials of BaTiO₃, SiO₂ and SiO₂ coated BaTiO₃ as a function of the pH value.

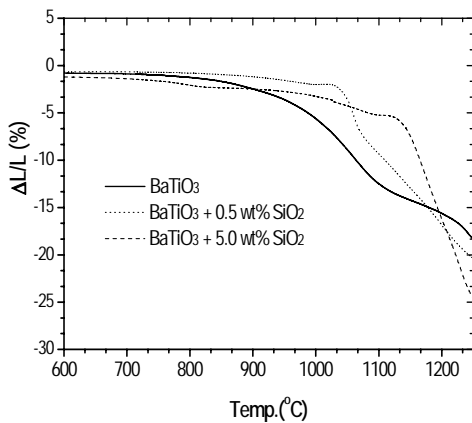


Fig. 3. Shrinkage curves of BaTiO₃ and SiO₂ coated BaTiO₃.

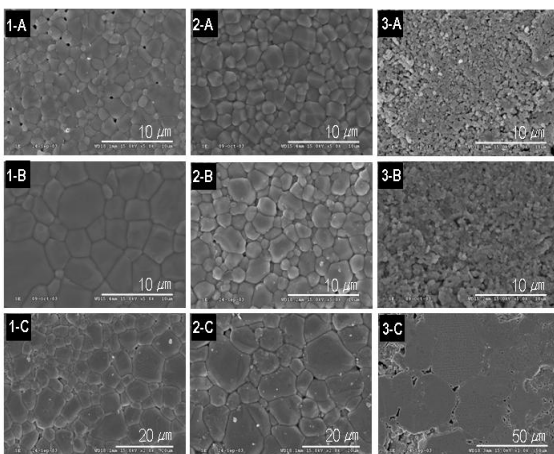


Fig. 4. Microstructures of BaTiO₃ and SiO₂ coated BaTiO₃ with various sintering temperatures. 1: Uncoated BaTiO₃, 2: 0.5% SiO₂ coating, 3: 5.0% SiO₂ coating A: 1150°C, B: 1200°C, C: 1250°C

Figure 4 shows the microstructures of uncoated and SiO₂ coated BaTiO₃ sintered at 1150, 1200 and 1250°C. The uncoated and 0.5 wt% SiO₂ coated BaTiO₃ samples made some densification at 1150°C and the average grain sizes increased with increasing sintering temperature. However, the specimen with 5.0 wt% SiO₂ exhibited poor densification and small grains up to 1200°C. At 1250°C, the microstructure of 5.0 wt% SiO₂ developed exaggerated grain growths. It was reported that the dominant mechanism for the exaggerated grain growth in the BaTiO₃ and SiO₂ system is Ostwald ripening due to the formation of inter-granular SiO₂ rich liquid phases [9].

3. Summary

BaTiO₃ powders were prepared by the Pechini process and coated with silica by the sol-gel process. The zeta potential of the SiO₂ coated BaTiO₃ is more negative than the uncoated BaTiO₃. The zeta potentials of 5.0 wt% SiO₂ coated BaTiO₃ approached that of pure SiO₂ particles. As the SiO₂ content was increased, powder dispersion improved due to the electrostatic repulsion between the coated silica layers on the surface of BaTiO₃ particles. The onset temperature of shrinkage curves is dependent on SiO₂ contents and shifts to higher temperatures with increasing SiO₂ because the SiO₂ coating layer prevents the direct contact between BaTiO₃ particles and retards the sintering rate at lower temperatures.

4. References

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