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Low-Temperature Sintering of $Ba_{0.5}Sr_{0.5}TiO_3$-$SrMoO_4$ Dielectric Tunable Composite Ceramics for LTCC Applications

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A sintering-aid system using melting of B-Li glass for barium strontium titanate (BST)-based compositions to be used in low-temperature cofired ceramic (LTCC) layers is introduced. The effects of the sintering aid on the microstructure, dielectric properties, and application in LTCC were investigated. The composition $Ba_{0.5}Sr_{0.5}TiO_3$-$SrMoO_4$ with 3 wt.% B-Li glass sintered at 950°C exhibits optimized dielectric properties, including low dielectric constant (368), low dielectric loss (0.007), and moderate tunability (13%, 60 kV/cm) at 10 kHz. At 1.44 GHz, it possesses a dielectric constant of 218 and Q value of 230. LTCC multilayer ceramic capacitors fabricated by the tape-casting process have steady relative tunability of 12% at 300 V, suggesting that BST50-$SrMoO_4$-B-Li glass composite ceramic is a promising candidate for electrically tunable LTCC microwave device applications.

Key words: LTCC, tunability, microwave properties, BST, tape casting

INTRODUCTION

Barium strontium titanate (BST) is one of the most important ferroelectric materials, being widely used in many applications.[1] It is well known that BST materials have high dielectric constant and high tunability. BST materials have been intensively investigated for microwave applications because of their excellent dielectric tunable property. Recently, research on use of microwave tunable BST materials in low-temperature cofired ceramic (LTCC) modules for miniaturization has become a topic of interest for many researchers and engineers working in the fields of materials science and microelectronics.[2,3] However, the design of tunable microwave-generation devices requires materials that have sufficiently low sintering temperature (ideally <960°C), good dielectric properties at microwave frequencies, and cost-effectiveness. The major challenge in designing materials for tunable devices is the simultaneous requirement for high relative tunability $[(\varepsilon_0 - \varepsilon_E)/\varepsilon_0 > 10\%]$, low dielectric loss, and moderate dielectric constant at microwave frequencies.[1,4] Therefore, studies of BST-based ceramics have focused on improving the dielectric properties and decreasing the sintering temperature. Researchers have already attempted to decrease the dielectric constant and suppress the dielectric loss by adding nonferroelectric phases (MgO,[5] Mg_2TiO_4[6]) and to decrease the sintering temperature of BST with lithium salts (Li_2CO_3, LiF)[7–11] and B_2O_3.[12–14] The phase diagram of the Li_2O-B_2O_3 binary system[15] also shows that Li_2O and B_2O_3 can produce a low-melting-point phase. Veres et al.[16] investigated B_2O_3/LiF-codoped $Ba(Mg,Zn)_{1/3}Nb_{2/3}O_3$ ceramics that can be successfully sintered at a low temperature of 940°C. Codoping with B_2O_3 and Li_2CO_3 was suggested as a promising sintering aid for densification at relatively low sintering temperatures.[17–19]

Furthermore, addition of a large amount of B_2O_3 as a sintering aid generally causes an additional problem when applying the tape-casting process for LTCC applications: slurry preparation is difficult because the B_2O_3 cross-links with the polymer binder.[20] For these reasons, sintering aids or sintering-aid systems for BST-based LTCC compositions are needed. Our previous publications[21,22] presented systematic study on the sintering behaviors and dielectric properties of $Ba_{1-x}Sr_xTiO_3$ ceramics with

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