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Influence of Li-B-Si Additions on the Sintering and Microwave Dielectric Properties of Ba-Nd-Ti Ceramics

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Li_2O-B_2O_3-SiO_2 (LBS) synthesized via a solid-state reaction process was chosen as a novel sintering aid for tungsten-bronze-type $Ba_4Nd_{9.3}Ti_{18}O_{54}$ (BNT) ceramic. The effects of LBS additions on the sintering behaviors, microstructures, and microwave dielectric properties of the BNT ceramic have been investigated, indicating that LBS addition obviously lowered the sintering temperature of the BNT ceramic without damaging its microwave dielectric properties. BNT ceramic doped with 3 wt.% and 4 wt.% LBS addition could be well sintered at 975°C and 950°C for 3 h and had excellent properties: $\varepsilon_r = 65.99$, $Q \times f = 4943$ GHz ($f = 4.4$ GHz), $\tau_f = 19$ ppm/°C, and $\varepsilon_r = 64.56$, $Q \times f = 4929$ GHz ($f = 4.3$ GHz), $\tau_f = 11$ ppm/°C, respectively.

Key words: BaO-Nd_2O_3-TiO_2, LTCC, Li_2O-B_2O_3-SiO_2 (LBS), microwave dielectric ceramics

INTRODUCTION

The rapid progress in mobile and satellite communication systems has accelerated the development of dielectric materials for use at microwave frequencies. Moreover, extensive attempts have been made to reduce the size of electronic circuits and components to satisfy the requirements of portability, light weight, and miniaturization of communication terminals.[1–6] Medium to high relative permittivities ($20 < \varepsilon_r < 80$) are generally required, as the size of a dielectric component is inversely proportional to the square root of the dielectric constant ε_r, but this cannot be achieved at the expense of decreasing the quality factor, $Q \times f$, where $Q \approx 1/\tan \delta$ and f is the resonant frequency. Additionally, components must be temperature-stable with temperature coefficient of the resonant frequency τ_f close to zero. $Ba_{6-3x}R_{8+2x}Ti_{18}O_{54}$ (R = rare-earth element) ceramics have been reported to have good microwave dielectric properties.[7–10] High dielectric constant ε_r (80 to 85) with high $Q \times f$ value (9000 GHz to 10,000 GHz) as well as a tunable temperature coefficient of resonant frequency τ_f were obtained in $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$ ($x = 2/3$, $Ba_4Nd_{9.3}Ti_{18}O_{54}$) solid solutions. However, the sintering temperature is usually as high as 1350°C for these undoped solid-solution ceramics. As a result, development of low-temperature cofired ceramic (LTCC) materials has become of great interest, since they would offer low-cost processing by using cheap inner electrodes in multilayer microwave devices.[11–13]

Common methods to reduce the sintering temperature include adding sintering aids or using chemical routes to prepare nanopowders. On the one hand, addition of a sintering aid such as a low-melting glass or oxide with low melting point was considered to be a useful approach.[14–17] Zheng et al.[18] reported that $0.75(Ba_4Nd_{9.3}Ti_{18}O_{54})$–$0.25(BaLa_4Ti_4O_{15})$ ceramics with 10 wt.% B_2O_3-Bi_2O_3-ZnO-SiO_2 glass could be sintered at 1140°C, with $\varepsilon_r = 61$, $\tau_f = 38$ ppm/°C, and $Q \times f = 2305$ GHz. Park et al.[19] added 10 wt.% Li-B-Si-Ca-Al glass to MBRT90 (Fuji Titanium Industry Co. Ltd., Japan). The sintering temperature decreased from 1300°C to lower than 875°C, and dielectric properties of $\varepsilon_r = 61$, $Q \times f = 2500$ GHz, and $\tau_f = 18$ ppm/°C were obtained. Choi et al.[20] also used Li-B-Si glass to decrease the sintering

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