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Phase, Microstructure, and Microwave Dielectric Properties of $NaCa_{4-x}Sr_xNb_5O_{17}$ (x = 0 to 4) Ceramics

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A series of $A_5B_5O_{17}$-type $NaCa_{4-x}Sr_xNb_5O_{17}$ (x = 0 to 4) compounds were processed through a solid-state mixed-oxide route. All the compositions formed dense single-phase ceramics within the detection limit of an in-house x-ray diffraction facility when sintered at 1300°C. The substitution of Sr for Ca changed the crystal symmetry from monoclinic (x = 0) to orthorhombic (x = 1 to 4) along with a slight increase in molar cell volume due to the relatively larger ionic radius of Sr. The relative permittivity (ε_r) and temperature coefficient of resonance frequency (TCF) increased from 46 to 84 and from −117 ppm/°C to +377 ppm/°C, respectively, while the quality factor ($Q \times f$) decreased from 11,063 GHz to 559 GHz with an increase in x from 0 to 4. Optimum properties were achieved for $NaCa_3SrNb_5O_{17}$, which exhibited ε_r = 57, $Q \times f$ = 4628 GHz, and TCF = −41 ppm/°C. Compounds in the $NaCa_{4-x}Sr_xNb_5O_{17}$ series exhibited high ε_r and $Q \times f$ with adjustable TCF; however, further work is required for simultaneous optimization of all three properties.

Key words: Ceramics, dielectric properties, microstructure

INTRODUCTION

The microwave communications industry developed rapidly due to the discovery of microwave dielectrics. Richtmyer[1] was the first to propose the idea of ceramic dielectric resonators. The main characteristics of a material to act as a dielectric resonator include high relative permittivity ε_r (>50), high quality factor $Q \times f$ (>10,000 GHz), and low temperature coefficient of resonant frequency (TCF) (~±5 ppm/°C).[2] High relative permittivity is necessary to reduce the size of a dielectric resonator. Obviously, a decrease in size enhances the portability of the relevant device, and reduces the amount of raw materials and hence the manufacturing cost of dielectric components. High quality factor is important to filter out noise during communication or fine-tuning, and near-zero TCF is essential for thermal stability. Several high-permittivity materials have been designed and processed, but they either are not stable thermally or have quality factors too low for commercial applications. Generally, high-relative-permittivity materials exhibit high TCF and vice versa, and high-ε_r materials have high dielectric losses; therefore, ceramists are constantly searching for new materials exhibiting all the above properties simultaneously.[3]

Materials with perovskite and related structures are of great interest due to their interesting electrical, magnetic, and optical properties. Layered perovskite-structured materials with general formula $A_nB_nO_{3n+2}$ are known to exhibit high permittivity, which is necessary for miniaturization of microwave dielectric resonators. The n = 4.5 and 5 members of the $A_nB_nO_{3n+2}$ series demonstrate interesting microwave dielectric properties; e.g., $CaLa_8Ti_6O_{18}$ (n = 4.5) and $CaLa_4Ti_5O_{17}$ (n = 5) were reported to exhibit ε_r = 48.6, $Q \times f_o$ = 19,345 GHz, and TCF = −6 ppm/°C and ε_r = 53.7, $Q \times f_o$ = 17,359 GHz, and TCF = −20 ppm/°C, respectively.[4] Substitution of Mg at A-site of $CaLa_4Ti_5O_{17}$, i.e., $Ca_{(1-x)}Mg_xLa_4Ti_5O_{17}$ with x = 0.01, has been reported[5] to give ε_r = 56.3, $Q \times f$ = 12,300 GHz, and TCF = −10 ppm/°C. Zhao et al.[6] substituted Zn for Ca at A-site and fabricated materials with ε_r = 57.6, $Q \times f$ = 17,100 GHz, and TCF = 4.9 ppm/°C. Similarly, Joseph et al.[7] reported ε_r = 44.9, $Q \times f_0$ = 17,600 GHz, and TCF = −112.9 ppm/°C for $Ca_5Nb_4TiO_{17}$. On the other hand, the substitution of

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