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High-Dielectric-Constant and Low-Loss Microwave Dielectric in the (1-x)$Nd(Zn_{1/2}Ti_{1/2})O_3$-x$SrTiO_3$ System with a Zero Temperature Coefficient of Resonant Frequency

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Miniaturization of dielectric devices for volume efficiency is a major requirement in modern microwave telecommunication technology such as 3G and GPS systems. Furthermore, the unique electrical properties of ceramic dielectric resonators have revolutionized the microwave-based wireless communications industry by reducing the size and cost of filter and antenna in circuit systems. Miniaturized GPS antennas, in particular, have become more and more popular in these few years. Materials with dielectric constant 90~100 can cut the antenna size to 12mm×12mm, however, it also possesses a much higher equivalent capacitance. In order to achieve appropriate capacitance, the antenna thickness is therefore increased. The mentioned problem can be solved by using dielectrics with k~50. Still, it suffers from its high dielectric loss. Consequently, low loss would then play a more prominent role instead. Previous report indicated that $Nd(Zn_{1/2}Ti_{1/2})O_3$ exhibits a mixture of Zn and Ti showing 1:1 order in the B-site and possesses excellent dielectric properties. In this work, $SrTiO_3$ (~ 1700 ppm/ ˚ C) was chosen as a compensator for . The studied system turned out to show a much better combination of microwave dielectric properties.

Fig. 1 shows the room temperature XRD patterns recorded for (1-x)$Nd(Zn_{1/2}Ti_{1/2})O_3$ (hereafter referred to as NZST) ceramic system at 1330 ˚ C for 4 h. No second phase was detected and the XRD patterns showed a two-phase system with a monoclinic structured $Nd(Zn_{1/2}Ti_{1/2})O_3$ and a cubic structured $SrTiO_3$. The measured lattice parameters of $Nd(Zn_{1/2}Ti_{1/2})O_3$ (a = 5.4652 ± 0.0005 •, b = 5.6399 ± 0.0007 •, c= 7.7797 ± 0.0008 • and β = 90.01 ± 0.01 ˚) were not changed with different $SrTiO_3$ contents, confirming the formation of two-phase system. The SEM images of 0.48$Nd(Zn_{1/2}Ti_{1/2})O_3$-0.52$SrTiO_3$ (hereafter referred to as 48NZST) ceramics sintered at 1330 ˚ C for 4 h are illustrated in Fig. 2. The grain morphology of well developed 48NZST ceramics could be grouped into two types, which were large grains identified as $Nd(Zn_{1/2}Ti_{1/2})O_3$ and small ones represented $SrTiO_3$ phase. A rapid grain growth resulted in an inhomogeneous grain morphology of pure $Nd(Zn_{1/2}Ti_{1/2})O_3$ ceramic was observed at temperatures higher than 1330 ˚ C. In this experiment, further increase in the sintering temperature would only lead to a gradual grain growth. The microstructure development of 48NZST was different from that of pure $Nd(Zn_{1/2}Ti_{1/2})O_3$. It indicates that rapid grain growth was hold back by an appropriate addition of $SrTiO_3$, which has a much smaller grain size than that of $Nd(Zn_{1/2}Ti_{1/2})O_3$.

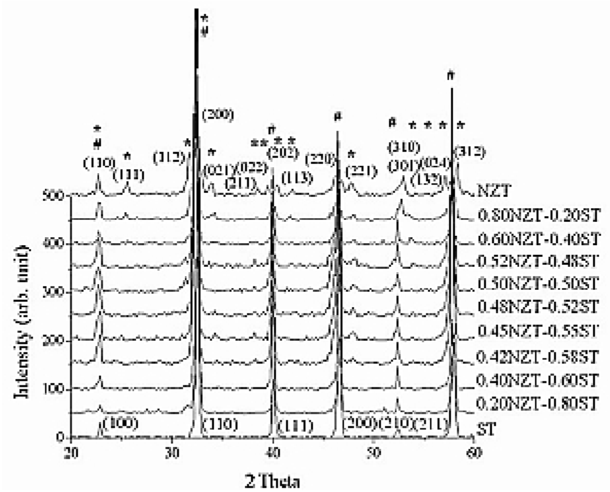

Fig. 1

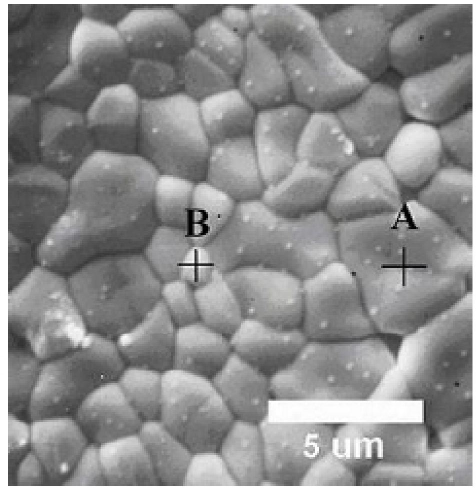

Fig. 2

Fig. 3 shows the apparent densities of NZST ceramic system at different sintering temperatures for 4 h. With increasing sintering temperature, the apparent density increased to a maximum value at 1330 ˚C and thereafter it slightly decreased. The increase in density was a result from grain growth. The degradation of apparent density at temperatures above 1330 ˚C, however, was owing to the evaporation of Zn. The dielectric constant of NZST ceramic system as a function of its sintering temperature for 4 h is illustrated in Fig. 4. The dielectric constant increased with increasing sintering temperature due to a denser specimen. After reaching its maximum at 1330 ˚C, it decreased. A maximum value of 54.2 was obtained for 48NZST ceramics sintered at 1330 ˚C for 4 h. It indicates further increase in the sintering temperature does not certainly lead to a higher dielectric constant. Moreover, frequencies in microwave regime had no significant influence on since the contribution from the interfacial/surface polarization is minimized at frequencies above 100 kHz.

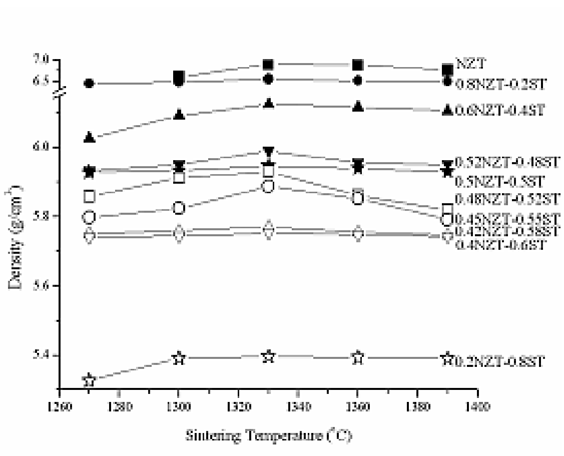

Fig. 3

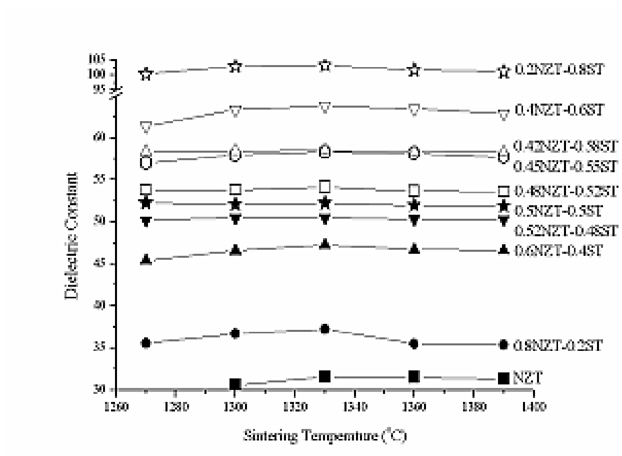

Fig. 4

The quality factor values ($Q \times f$) of NZST ceramic system at different sintering temperatures for 4 h are demonstrated in Fig. 5. Since the variation of $Q \times f$ was consistent with that of density, it suggested $Q \times f$ was most likely associated with the corresponding variation in apparent density. A maximum *Q*×*f* value of 84,000 GHz was obtained for 48NZST ceramics sintered at 1330 ˚C for 4 h. The *Q*×*f* was also a function of compositional ratio, and decreased with increasing $SrTiO_3$ content since $SrTiO_3$ possesses a much lower *Q*×*f*. Moreover, uniform morphology was observed for specimen at 1330 ˚C, which would certainly also benefit to a loss reduction. Fig. 6 shows the temperature coefficients of resonant frequency (   ) of NZST ceramic system at different sintering temperatures for 4 h. With x=0.52 (48NZST), zero   value was achieved for $(1\text{-}x)Nd(Zn_{1/2}Ti_{1/2})O_3$ -x$SrTiO_3$ ceramic system sintered at 1330 ˚C for 4 h. Moreover,   was almost independent of sintering temperature since there was no significant compositional change observed. It provides a wide process window which would be benefit to commercial applications.

Investigations on the microwave dielectric properties of two-phase system $(1\text{-}x)Nd(Zn_{1/2}Ti_{1/2})O_3$-x$SrTiO_3$ have shown that these materials possess relatively high *Q*×*f*, high dielectric constant and tunable   . A two-phase system was confirmed by the XRD patterns, the EDS analysis and the measured lattice parameters. The proposed system exhibits the best currently available combination of dielectric properties (= 54.2, *Q*×*f* = 84,000 GHz,   = 0 ppm/ ˚C at x = 0.52), which can be as a suitable candidate material for today's 3G passive components and small-sized GPS patch antennas.

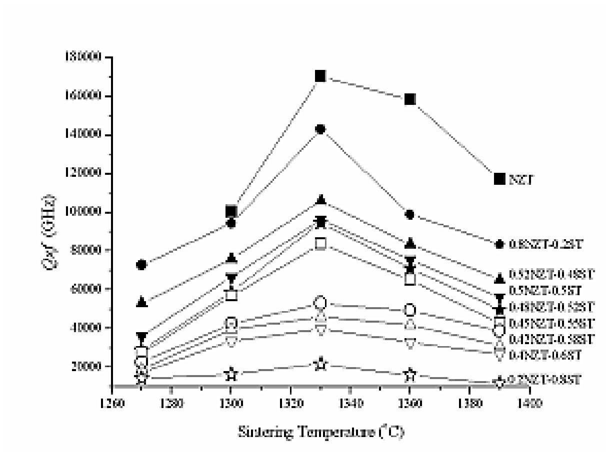

Fig. 5

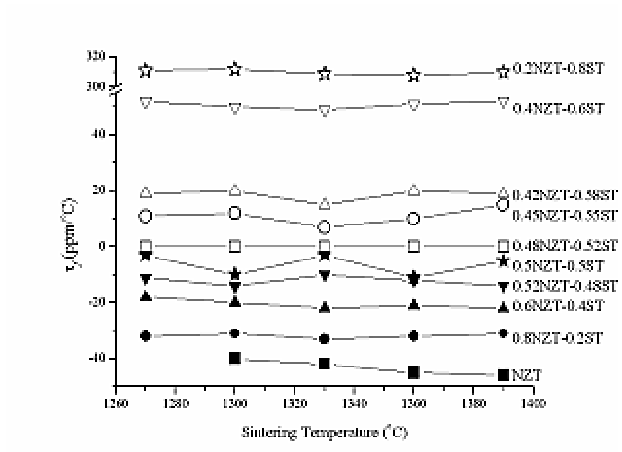

Fig. 6