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Effect of Sb^{5+} substitution on the dielectric properties of $Ag(Nb_{0.8}Ta_{0.2})O_3$ ceramics

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Abstract

Sb_2O_5 were selected to substitute $(Nb_{0.8}Ta_{0.2})_2O_5$ and the effects of substitution on the dielectric properties of $Ag(Nb_{0.8}Ta_{0.2})O_3$ ceramics were studied. The dielectric properties of $Ag(Nb_{0.8}Ta_{0.2})_{1-x}Sb_xO_3$ ceramics were found to be improved by the substitution of Sb for Nb/Ta. The ε value of $Ag(Nb_{0.8}Ta_{0.2})_{1-x}Sb_xO_3$ ceramics sintered at 1060 °C increased from 430 to 825 with x increasing from 0 to 0.08, the tanδ value decreased sharply from 0.0085 to 0.0023 (at 1 MHz) with x increasing from 0 to 0.04, and then kept to a lower tanδ value ~0.0024 with x to 0.08. The TCC values decreased from +1450 ppm/°C for $x=0$ to −38.5 ppm/°C for $x=0.08$. The $Ag(Nb_{0.8}Ta_{0.2})_{1-x}Sb_xO_3$ ceramics with $x=0.08$ sintered at 1050 °C exhibited the optimum dielectric properties of ε ~854, tanδ ~0.0024 (1 MHz), and TCC ~36.86 ppm/°C.

Keywords: $Ag(Nb_{0.8}Ta_{0.2})O_3$; Ceramics; Dielectric properties

1. Introduction

New materials for microwave dielectric resonators should have a high dielectric permittivity ε, a high quality factor Q and a temperature coefficient of the resonant frequency τ_f close to zero [1,2]. Comprehensive study on the dielectric properties of microwave, submillimeter to infrared spectroscopy [3] has proved that silver niobate–tantalate solid solutions $AgNb_{1-x}Ta_xO_3$ (ANT) are potentially good microwave materials. In these materials, there is a negligible dielectric dispersion for a very broad frequency range from 1 kHz up to approximately 100 GHz [4–6]. The test performed in 1 GHz region indicated a permittivity of 430, a temperature coefficient of permittivity <50 ppm/°C and a Q value of 700 for the $AgNb_{0.65}Ta_{0.35}O_3$–$AgNb_{0.35}Ta_{0.65}O_3$ composite [7].

Partial replacements of Ag by K, Na and Li [8–10] in A site of $AgNbO_3$ were reported. However, only the Ta was used to substitute for Nb in B site of $AgNbO_3$ to modify the dielectric properties of $AgNbO_3$ ceramics [4,7,11]. For the similar radius of Sb^{5+}($r_{Sb}^{5+}=0.062$ nm) to that of Nb^{5+} ($r_{Nb}^{5+}=0.069$ nm) and Ta^{5+}($r_{Ta}^{5+}=0.068$ nm), the replacement of Nb ions by Sb ions in $(Bi_{1.8}Zn_{0.2})(Zn_{0.6}Nb_{1.4-x}Sb_x)O_7$ [12], $Bi(Nb,Sb)O_4$ [13] and $(Mg_{4-x}M_x)(Nb_{2-y}Sb_y)O_9$(M=Zn and Ni) [14] system have been

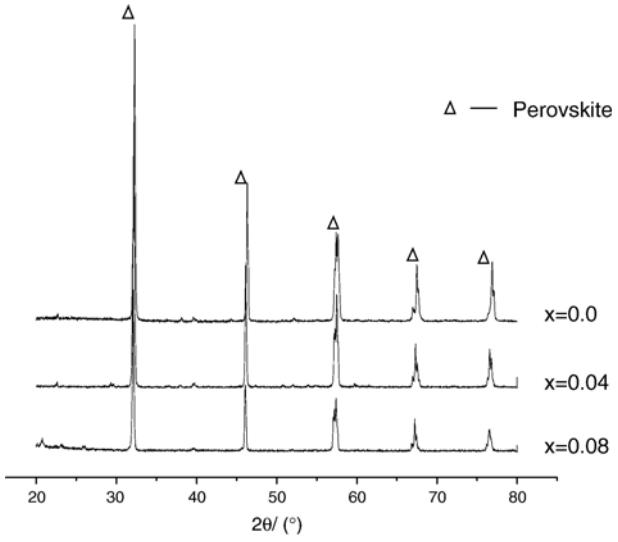

Fig. 1. XRD patterns of $Ag(Nb_{0.8}Ta_{0.2})_{1-x}Sb_xO_3$ ($x=0$, 0.04, 0.08) ceramics.

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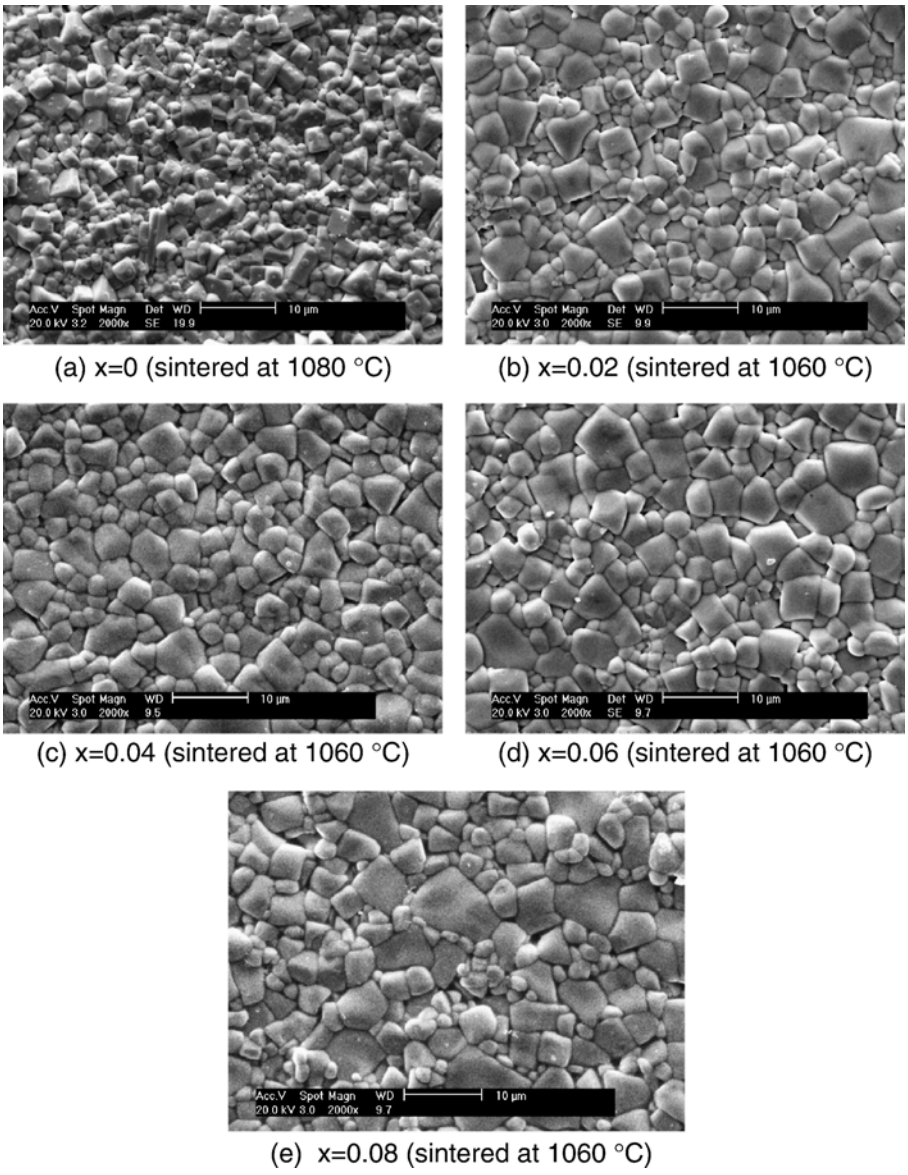

Fig. 2. SEM micrographs of $Ag(Nb_{0.8}Ta_{0.2})_{1-x}Sb_xO_3$ ceramics. (a) $x=0$ (sintered at 1080 °C). (b) $x=0.02$ (sintered at 1060 °C). (c) $x=0.04$ (sintered at 1060 °C). (d) $x=0.06$ (sintered at 1060 °C). (e) $x=0.08$ (sintered at 1060 °C).

investigated. We suggest that the substitution of Nb^{5+}/Ta^{5+} by Sb^{5+} in perovskite $AgNb_{1-x}Ta_xO_3$ might cause a slight modification of crystal structure, resulting in a change on dielectric properties. In our previous works, we have systemically researched the influences of doping, sintering atmosphere and process of preparation on $Ag(Nb_{0.8}Ta_{0.2})O_3$ ceramics [15–17]. So, here, $Ag(Nb_{0.8}Ta_{0.2})O_3$ ceramics was used as the host material and Sb_2O_5 was selected to substitute for $(Nb_{0.8}Ta_{0.2})_2O_5$. The aim of this work is to obtain perovskite $Ag(Nb_{0.8}Ta_{0.2})_{1-x}Sb_xO_3$ ceramics of low loss and with near zero temperature coefficients.

2. Experimental

A traditional solid-state reaction method was applied to prepare $Ag(Nb_{0.8}Ta_{0.2})_{1-x}Sb_xO_3$ ($x=0$, 0.02, 0.04, 0.06, 0.08) solid solutions using reagent-grade Nb_2O_5, Ta_2O_5, Ag_2O and Sb_2O_5. First, Nb_2O_5 and Ta_2O_5 powders were mixed on the basis of the stoichiometric composition $(Nb_{0.8}Ta_{0.2})_2O_5$ and calcined at 1200 °C for 10 h, after which Ag_2O and Sb_2O_5 were added in the appropriate molar ratio. And the mixture was milled for 4 h in distilled water, then dried and fired at 950 °C for 10 h to form the ANT's precursors. The prereacted powders were then milled again for 4 h and dried. Then, 6 wt.% organic binder was added to the dried powders, and samples were pressed into disks of 10 mm in diameter and 1 mm in thickness under a pressure of 10 MPa. The disks were sintered at 1000 °C–1100 °C in air for 2 h.

The crystal phases of samples were examined with an X-ray diffractometer (Model 2038X, Rigaku Co.) with Cu *K*a radiation. The microstructure investigations were conducted via Scanning Electron Microscopy (SEM, Model Hitachi X-650).

The dielectric properties were measured at 1 MHz. The loss tangent and capacitance of the samples were measured by HP4278A RLC and the temperature coefficient of capacitance