Journal of ELECTRONIC MATERIALS, Vol. 42, No. 6, 2013
DOI: 10.1007/s11664-013-2543-x
© 2013 TMS

MnO_2-Modified $Ba(Ti,Zr)O_3$ Ceramics with High Q_m and Good Thermal Stability

P. ZHENG,[1,3] J.L. ZHANG,[2] H.B. QIN,[1] K.X. SONG,[1] J. WU,[1] Z.H. YING,[1] L. ZHENG,[1] and J.X. DENG[1]

1.—College of Electronics and Information, Hangzhou Dianzi University, Hangzhou 310018, China. 2.—School of Physics, Shandong University, Jinan 250100, China. 3.—e-mail: zhengpeng@hdu.edu.cn

MnO_2-modified $Ba(Ti_{0.9625}Zr_{0.0375})O_3$ ceramics have been prepared by the conventional solid-state reaction technique at different sintering temperatures. Room-temperature piezoelectric properties, thermal stability, and crystalline structures were investigated. It was found that both the MnO_2 additive and sintering temperature significantly influence the piezoelectric properties of the MnO_2-modified $Ba(Ti_{0.9625}Zr_{0.0375})O_3$ ceramics. The sample sintered at 1400°C exhibited the best room-temperature piezoelectric properties of Q_m = 1907, d_{33} = 205 pC/N, and k_p = 40.5% with tan δ = 0.46%, and its k_p remains larger than 35% in the broad temperature range from −38°C to 65°C. The results indicate that MnO_2-modified $Ba(Ti_{0.9625}Zr_{0.0375})O_3$ ceramics are promising lead-free materials for frequency device and power device applications.

Key words: Ceramics, lead free, piezoelectric properties, high Q_m, thermal stability

INTRODUCTION

Lead zirconate titanate (PZT) and PZT-based ceramics have been popularly utilized for more than 50 years in actuators, sensors, resonators, transducers, and transformers because of their excellent piezoelectric and electric properties.[1] However, due to the toxicity of lead oxide used in the production process, there are increasing demands to replace these ceramics with environmentally benign lead-free alternatives. $BaTiO_3$-based ceramics are famous materials that were widely used as piezoelectric materials before the discovery of PZT.[1] However, their current main technical applications are not as piezoelectric but as dielectric materials, largely because of their inferior piezoelectric activities and weak thermal stability in comparison with those of PZT. Surprisingly, high d_{33} values have been successfully obtained in the past 4 years in $BaTiO_3$-based ceramics by many investigators.[2–5] In our previous study, we also fabricated a high-d_{33} $BaTiO_3$ ceramic with 419 pC/N through a conventional solid-state reaction route using ordinary $BaCO_3$ and TiO_2 powders as the starting raw materials.[6] However, subsequent investigation confirmed that the piezoelectric properties of the recently obtained $BaTiO_3$ ceramics with high d_{33} values are also strongly temperature dependent around room temperature, which is the temperature range in common applications, similar to the conventional $BaTiO_3$ ceramics. To overcome this problem, we recently succeeded in obtaining $BaTiO_3$-based ceramics with both outstanding piezoelectric activities and good thermal stability by a combination of partial Ti substitution with Zr or Sn and a small amount of CuO additive.[7,8] This progress brings great hope that $BaTiO_3$-based ceramics might be revitalized as popular low-cost lead-free piezoelectric materials.

Although larger d_{33} and good temperature stability have be achieved in $BaTiO_3$-based ceramics, the low mechanical quality factor Q_m (usually not more than 300) and high dielectric loss tangent of these ceramics are not suitable for frequency device and power device applications. Therefore, developing

(Received November 4, 2012; accepted February 8, 2013; published online April 3, 2013)