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**Stability of High Temperature Dielectric Properties for  
(1-x)Ba<sub>0.8</sub>Ca<sub>0.2</sub>TiO<sub>3</sub> - xBi(Mg<sub>0.5</sub>Ti<sub>0.5</sub>)O<sub>3</sub> Ceramics**

Aurang Zeb and Steven J Milne,

Institute for Materials Research, University of Leeds, Leeds, LS2 9JT, UK

**Abstract**

Ceramics in the solid solution system, (1-x)Ba<sub>0.8</sub>Ca<sub>0.2</sub>TiO<sub>3</sub>-xBi(Mg<sub>0.5</sub>Ti<sub>0.5</sub>)O<sub>3</sub>, were prepared by a conventional mixed oxide route. Single phase perovskite-type x-ray diffraction patterns were observed for compositions  $x < 0.6$ . A change from tetragonal to single-phase cubic x-ray patterns occurred at  $x \geq 0.1$ . Dielectric measurements indicated relaxor behaviour for  $x \geq 0.1$ . Increasing the Bi(Mg<sub>0.5</sub>Ti<sub>0.5</sub>)O<sub>3</sub> content improved the temperature-sensitivity of relative permittivity  $\epsilon_r$  at high temperatures. At  $x = 0.5$ , a near plateau relative permittivity,  $835 \pm 40$ , extended across the temperature range, 65 °C to 550 °C; the permittivity increased at  $x = 0.6$  to  $2170 \pm 100$  for temperatures 160 °C to 400 °C (1 kHz). The corresponding loss tangent,  $\tan\delta$ , was  $\leq 0.025$  for temperatures between 100 °C and 430 °C for composition  $x = 0.5$ ; at  $x = 0.6$ , losses increased sharply at  $> 300$  °C. Comparisons of dielectric properties with other materials proposed for high temperature capacitor applications suggests that (1-x)Ba<sub>0.8</sub>Ca<sub>0.2</sub>TiO<sub>3</sub>-xBi(Mg<sub>0.5</sub>Ti<sub>0.5</sub>)O<sub>3</sub> ceramics are a promising base material for further development.

## Introduction

Ceramics with stable dielectric properties at temperatures  $>200\text{ }^{\circ}\text{C}$  offer opportunities for developing high-temperature capacitors for applications in electronic control systems for hybrid or electrically powered vehicles [1, 2]. The dielectric material must have high and uniform dielectric permittivity,  $\epsilon_r$ , coupled to low dielectric loss over a wide operating temperature range. Commercial X7R and X8R capacitors are rated to  $125\text{ }^{\circ}\text{C}$  and  $150\text{ }^{\circ}\text{C}$  respectively [3].

A range of lead-free ceramics have been investigated in the search for new high-temperature dielectrics, including compositions in the solid solution series:  $\text{BaTiO}_3$  -  $\text{Ba}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$  [4];  $\text{BaTiO}_3$  -  $\text{Ba}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$  -  $\text{BiScO}_3$  [5];  $\text{Na}_{0.5}\text{K}_{0.5}\text{NbO}_3$  -  $\text{LiTaO}_3$  -  $\text{BiScO}_3$  [6,8];  $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  -  $\text{KTaO}_3$  [7];  $\text{BiScO}_3$  -  $\text{BaTiO}_3$  -  $\text{K}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  [1];  $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  -  $0.06\text{BaTiO}_3$  -  $\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  [2];  $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  -  $\text{K}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  -  $\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  [9];  $\text{K}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  -  $\text{BiScO}_3$  [18];  $\text{BaTiO}_3$  -  $\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  [10,11]; and modified  $\text{BaTiO}_3$  formulations [12]. Most of these materials are relaxor dielectrics with diffuse, frequency-dependent dielectric peaks associated with interactions of polar nanoregions, and a high-temperature plateau permittivity above  $T_m$ , the temperature of peak dielectric permittivity. In some cases the temperature-stability in permittivity arises from overlap of two broad permittivity peaks [2,8,9,11]. Structural and compositional heterogeneity may contribute to a flattening in permittivity peaks, as has been demonstrated by core-shell grain structures identified in  $\text{BaTiO}_3$  -  $\text{BiScO}_3$  [13] and  $\text{Na}_{0.5}\text{K}_{0.5}\text{TiO}_3$  -  $\text{LiTaO}_3$  -  $\text{BiScO}_3$  [8, 14,19] ceramics.

One of the most promising recent reports of a relaxor dielectric for high-temperature capacitor applications relates to  $0.5\text{BaTiO}_3 - 0.25\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3 - 0.25\text{BiScO}_3$  with  $\epsilon_r \sim$

1000, low resistivity at  $> 200\text{ }^{\circ}\text{C}$  and a near-flat  $\epsilon_r$  response for temperatures  $100\text{ }^{\circ}\text{C}$ - $500\text{ }^{\circ}\text{C}$  [5]. However the relatively high  $\text{Sc}_2\text{O}_3$  content in this optimum composition has cost implications that may be an obstacle to commercial implementation [9]. A number of other potential high temperature dielectrics also contain a significant proportions of scandium oxide [1,5,6,8,13,14] or indium oxide [15].

In this study we present a solid solution series between Ca-modified  $\text{BaTiO}_3$  and  $\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  that exhibits a high degree of stability at elevated temperatures, superior to unmodified  $\text{BaTiO}_3$  -  $\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  dielectrics in terms of stability in relative permittivity and operating temperature range [10,11]. Previous work on  $\text{BaTiO}_3$ - $\text{CaTiO}_3$  solid solutions indicates that Ca substitution on the  $\text{BaTiO}_3$  perovskite lattice lowers the temperature of the orthorhombic-tetragonal (and rhombohedral-orthorhombic) polymorphic phase transitions of  $\text{BaTiO}_3$ , with little variation in the tetragonal-cubic transition temperature, such that a tetragonal perovskite phase is stable over an extended temperature range for  $\sim 20$ - $30$  mol % Ca compositions [16,17]. Here we demonstrate properties of ceramics in the ternary  $\text{BaTiO}_3$  -  $\text{CaTiO}_3$  -  $\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  along the compositional join from  $\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3$  toward  $\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ . Properties of the optimal  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3$ - $x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  compositions compare favourably to other high-temperature dielectric ceramics.

## Experimental

Samples were prepared by conventional solid state processing using starting powders:  $\text{CaCO}_3$  (Sigma Aldrich, 99% purity),  $\text{TiO}_2$  (Sigma Aldrich, 99.9%),  $\text{BaCO}_3$  (Alpha Aesar, 99%),  $\text{MgO}$  (Alpha Aesar, 99.9%) and  $\text{Bi}_2\text{O}_3$  (Alpha Aesar, 99%). Reagents were dried at  $250\text{ }^{\circ}\text{C}$  overnight, before cooling to room-temperature in a desiccator and weighing according to stoichiometric ratios. The powders were then mixed in isopropanol and ball milled with stabilized zirconia grinding media for 24 h. The dried powders were calcined at

800 °C- 1100 °C for 3 h. The calcined powders were ball milled for 24 h and shaped into pellets by uniaxial pressing in a 10 mm steel die at 65 MPa, followed by cold isostatic pressing at 300 MPa. A binder (Ciba Glascol HA4) was introduced during the post-calcination milling stage. The pellets were embedded in powder of the same composition and sintered at 1000 °C - 1400 °C for 3 h in a closed alumina crucible to minimise loss of volatile bismuth oxide. Pellet densities were  $\geq 90\%$  of theoretical density calculated from lattice parameters. The sintered pellets were ground to a powder for phase analysis, performed using x-ray powder diffraction (XRD, Bruker D8, Cu,K $\alpha$ ~1.5406 Å); scan speed, 1°/min. Lattice parameters were calculated using a least square refinement method. For dielectric measurements, the sintered pellets were ground and polished to 0.8 mm thickness; silver paste (Agar Scientific) was applied to opposite parallel faces and coated pellets were fired in a furnace at 550 °C for 10 min to form the electrode. The dielectric relative permittivity and loss tangent were recorded using an impedance analyzer (HP Agilent, 4192A Hewlett Packed) in the temperature range, 20 °C to 550 °C.

## Results and Discussion

X-ray diffraction patterns of powders of crushed sintered pellets of  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3-x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  are shown in Figure 1. The  $x = 0$  ( $\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3$ ) and  $x = 0.05$  compositions appear tetragonal, but anomalies in relative intensity of 001/100 and 002/200 peaks at  $\sim 22^\circ 2\theta$  and  $45^\circ 2\theta$  respectively, Figure 1, suggest some perovskite phase heterogeneity [17]. A change to a (pseudo) cubic single-phase XRD pattern occurred at  $x = 0.1$ . Secondary peaks of similar d-spacings to bismuth titanate and bismuth oxide began to appear at  $x = 0.6$  and increased in relative intensity at higher  $x$  values [10,11,20,21]. Lattice

parameters are plotted as a function of composition in Figure 2, revealing a slight contraction in  $c$  and increase in  $a$  tetragonal parameters between  $x = 0$  and  $0.05$ , thereafter a progressive increase in cubic  $a$  parameter is observed for  $x > 0.1$ . Plots of relative permittivity,  $\epsilon_r$ , versus temperature for the  $x = 0$  end-member showed a sharp Curie peak, typical of a normal ferroelectric with Curie point,  $T_c \sim 130$  °C, Figure 3a; the peak became more diffuse and  $T_c$  decreased to  $\sim 110$  °C for composition  $x = 0.05$ , Figure 3b. A change to relaxor dielectric behaviour occurred at  $x = 0.1$ , with a diffuse, frequency-dependent dielectric response below the temperature of peak permittivity,  $T_m$ . Values of  $T_m$  increased with increasing  $x$ , as shown in Figure 5. For  $x = 0.2-0.7$ , the  $T_m$  values increased from  $\sim 40$  °C to  $170$  °C, Figure 5; for  $x = 0.1$ ,  $T_m$  at  $1$  kHz fell below room temperature and was not recorded.

The peak  $\epsilon_r$  values decreased from  $> 2000$  at  $x = 0.1$  (estimated) to  $875$  at  $x = 0.5$ , then  $\epsilon_r$  increased to  $\sim 2270$  for  $x = 0.6$ . A gradual decrease in the temperature-sensitivity of relative permittivity at  $T > T_m$ , occurred with increasing  $x$ , with a minimum temperature-dependence at  $x = 0.5$  and  $0.6$ . For  $x = 0.5$ , the permittivity values were  $835 \pm 40$  in the temperature range  $65$  °C to  $550$  °C. For  $x = 0.6$ , the maximum  $\epsilon_r$  values (at  $> T_m$ ) were higher,  $2270$ , with an overall consistency,  $2170 \pm 100$ , for temperatures from  $160$  °C to  $400$  °C. Hence the variation in permittivity for  $x = 0.5$  and  $0.6$  was within  $\pm 5\%$  of the mid-point ( $\epsilon_r \text{ max} - \epsilon_r \text{ min}$ ) value. In terms of  $\pm 10\%$  variation, the operating temperature range of  $x = 0.5$  extended from  $45$  °C to  $550$  °C and  $140-420$  °C for  $x = 0.6$ . Composition  $x = 0.5$  has a wider temperature range of operation but lower dielectric permittivity than  $x = 0.6$  ceramics. The values of  $\tan \delta$  as a function of temperature for  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3-x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  are shown in Figure 4. The relaxation in  $\tan \delta$  values around  $T_m$ , is typical of a relaxor dielectric. This loss tangent maximum is followed at higher temperatures by a relatively constant and low  $\tan \delta$  value  $\leq 0.025$ , extending from  $\sim 100$  °C and  $430$  °C for  $x =$

0.5, and 170 °C to 300 °C, for  $x = 0.6$  (1 kHz). Above these upper limiting temperatures,  $\tan \delta$  rose sharply in the manner usually associated with electronic conduction. Losses also rose sharply at higher frequencies,  $> 1$  kHz, in the  $x = 0.6$  sample, similar to the trends for high temperature losses in  $x < 0.1$  samples, and for  $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3 - \text{BaTiO}_3 - \text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  and  $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3 - 0.06\text{BaTiO}_3 - \text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  high - temperature dielectrics [2,9].

At temperatures  $> 300\text{-}400$  °C relative permittivity measured at 1 kHz and 10 kHz increased as dielectric losses (conduction) rose sharply- except for compositions  $x = 0.2$  and 0.5, Figures 3-4. Because of the possible contribution of loss mechanisms to the apparent stability in permittivity of  $x = 0.6$  at upper-limiting temperatures, the temperature-stability of  $\epsilon_r$  at 100 kHz was investigated, since at this frequency,  $\tan \delta$  remained  $\leq 0.025$  for temperatures from 250 °C ( $T_m$  100 kHz) to 400 °C, Figure 4(f). The resulting  $\epsilon_r$  consistency was  $\pm 5\%$  over the temperature range 200 °C to 400 °C at 100 kHz similar to the 1 kHz precision, indicating that losses were not the dominant factor in the near-flat permittivity response of  $x = 0.06$  at 1 kHz. For  $x = 0.5$ ,  $\tan \delta$  remained  $\leq 0.025$  until 430 °C, and there was very little frequency-dependence in high-temperature permittivity, Figure 3(e) and 4(e).

The related system  $(1-x)\text{BaTiO}_3 - x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  is a relaxor dielectric, but there are conflicting reports regarding the temperature-stability of permittivity. Wada et al. report dielectric peak temperature,  $T_m$ , increased from  $\sim 50$  °C at  $x = 0.1$ , to  $\sim 360$  °C at  $x = 0.5$  and showed a strong temperature dependence in  $\epsilon_r$  at  $x = 0.5$ . Dielectric peaks were most diffuse for  $x = 0.2\text{-}0.3$ , varying in the range  $\epsilon_r \sim 800 \pm 200$  (estimated) from 50 °C to 450 °C (estimated)[10]. Xiong et al. report a transition from tetragonal to pseudocubic in  $(1-x)\text{BaTiO}_3 - x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  at  $x = 0.05$ , similar to the changeover composition in  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3 - x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ . Diffuse and double overlapping permittivity peaks gave a flatter dielectric response at higher temperatures 200-500 °C than reported in reference 10, with a peak permittivity of  $\sim 1500$  at  $T_m \sim 200$  °C decreasing to a minimum of  $\epsilon_r \sim 1300$  at

~450 °C (estimated) [11]; dielectric loss was < 2% from 200 °C - 400 °C. Hence the present Ca-modified material ( $x = 0.5$ ) shows improved temperature stability in terms of temperature coefficient of dielectric permittivity ( $\pm 5\%$ ) and a wider temperature range of stability (65 °C - 550 °C) than the corresponding  $(1-x)\text{BaTiO}_3\text{-}x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  parent composition. However absolute values of permittivity are lower in Ca-modified  $x = 0.5$  ceramics. The permittivity of Ca-modified  $x = 0.6$  is higher than the optimum unmodified  $\text{BaTiO}_3\text{-Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  temperature-stable composition, but the temperature range of stability is lower in the new material, Table 1.

Incorporating  $\text{Nb}_2\text{O}_5$  into  $0.85\text{BaTiO}_3\text{-}0.15\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  produced a flatter permittivity-temperature response than in the undoped material, but at the expense of a reduced  $\epsilon_r$  value,  $\leq 1000$ , and a lower upper temperature range, < 150 °C ( $\epsilon_r \pm 15\%$ ) as opposed to 550 °C and flatter permittivity,  $\epsilon_r \pm 5\%$ , for the new Ca-modified  $x = 0.05$  ceramics [11].

The dielectric properties for  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3\text{-}x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ ,  $x = 0.5$  and  $0.6$ , are summarised and compared to a representative selection of other relaxor-based high temperature lead-free dielectric ceramics in Table 1. Another approach to exploring high temperature dielectrics is to investigate the properties of normal ferroelectrics with high temperature Curie points,  $T_c$  and with invariant relative permittivity to below room-temperature [6,8,14].

There appear to be two categories of dielectric ceramics with diffuse permittivity peaks and reasonably stable temperature response: one group such as unmodified  $\text{BaTiO}_3\text{-Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  exhibits overlapping twin diffuse permittivity peaks, [2, 9-11]; the other grouping shows a single plateau at  $T > T_m$  [5] as found here for  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3\text{-}x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ . The reasons for either behaviour are not well understood at this stage,



although in some cases, twin dielectric peaks are associated with intra-granular chemical segregation, for example in  $\text{Na}_{0.5}\text{K}_{0.5}\text{NbO}_3$  -  $\text{LiTaO}_3$  -  $\text{BiScO}_3$  where a near-flat relative permittivity response  $\epsilon_{\text{rmax}} \geq 1300$  over a wide temperature range, 50 °C- 450 °C [6] is associated with core-shell chemical segregation within individual grains [6,8,14,19]. Alternatively, the double diffuse permittivity peaks in  $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ - $\text{K}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  -  $\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  have been attributed to two types of polar nanoregions [9].

The new  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3$ - $x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  materials compare well in specific respects to  $0.8(0.4\text{BiScO}_3$ - $0.6\text{BaTiO}_3)$ - $0.2\text{K}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$  [1];  $(1-x)0.94[\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ - $0.06\text{BaTiO}_3]$ - $x\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  [2];  $0.85(\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ - $\text{K}_{0.5}\text{Bi}_{0.5}\text{TiO}_3)$ - $0.15\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  [9];  $\text{BaTiO}_3$ - $\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$  [4] and  $0.5\text{BaTiO}_3$ - $0.25\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ - $0.25\text{BiScO}_3$  [5]. The  $x = 0.5$  and  $0.6$   $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3$ - $x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  materials are generally similar in terms of reagent costs and availability of component oxides to  $0.85(\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ - $\text{K}_{0.5}\text{Bi}_{0.5}\text{TiO}_3)$ - $0.15\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  [9] and  $(1-x)0.94[\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ - $0.06\text{BaTiO}_3]$ -  $x\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  [2], temperature stability of dielectric properties compare favourably to these materials, Table 1, although relative permittivity for  $x = 0.5$  is lower. To aid comparisons with other high-temperature dielectrics [1,5], Table 1 includes values of the temperature coefficient of dielectric permittivity  $\text{TC}\epsilon_r$  expressed in  $\text{ppm}/^\circ\text{C}$  for both  $x = 0.5$  and  $0.6$ , calculated as described in reference 5. The  $0.5\text{BaTiO}_3$ - $0.25\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ - $0.25\text{BiScO}_3$  material, compositionally engineered to contain a sub-stoichiometric content of barium to increase electrical resistivity, offers one of the most consistent high-temperature permittivity values with  $\text{TC}\epsilon_r = 182 \text{ ppm}/^\circ\text{C}$  over the temperature range 100 °C -500 °C and peak  $\epsilon_r \sim 1100$  [5]. Corresponding values for the Sc-free high temperature dielectric  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3$ - $x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$   $x = 0.5$  are  $\text{TC}\epsilon_r = 200 \text{ ppm}/^\circ\text{C}$ , temperature range of stable permittivity 65 °C -550 °C, and  $\epsilon_r \text{ max} = 875$ . For  $x = 0.6$ , values are  $400 \text{ ppm}/^\circ\text{C}$ , range 160-400 °C and  $\epsilon_r \text{ max} = 2270$  °C, Table 1.

## Conclusions

Ceramics in the solid solution system,  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3-x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ , have been fabricated by a conventional mixed oxide route. Single-phase perovskite-type x-ray diffraction patterns were observed for  $x < 0.6$ . For  $x \leq 0.05$  no frequency dispersion in relative permittivity was observed, although  $x = 0.05$  peaks were more diffuse than for a normal ferroelectric. A change to relaxor dielectric behaviour occurred for the (pseudo) cubic ceramics,  $0.1 \leq x \leq 0.6$ . Diffuse, frequency-dependent permittivity-temperature plots exhibited near flat responses in the frequency independent region  $T > T_m$  for compositions  $x = 0.5$  and  $0.6$ . For  $x = 0.5$ , relative permittivity (1 kHz) was  $835 \pm 40$  for temperatures extending from  $65^\circ\text{C}$  to  $550^\circ\text{C}$ . Permittivity values were much higher for  $x = 0.6$ , in the range  $2170 \pm 100$  for temperatures  $160^\circ\text{C}$  to  $400^\circ\text{C}$ . The loss tangent (1 kHz) values were  $\leq 0.025$  for temperatures  $100^\circ\text{C}$  -  $430^\circ\text{C}$  for composition  $x = 0.5$ ; losses were generally higher for  $x = 0.6$  and increased sharply at  $> 300^\circ\text{C}$ . Properties compare well to a range of other high-temperature dielectrics.

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### Figure List

Figure 1. X-ray powder diffraction patterns of crushed sintered pellets for (1-x)Ba<sub>0.8</sub>Ca<sub>0.2</sub>TiO<sub>3</sub>-xBi(Mg<sub>0.5</sub>Ti<sub>0.5</sub>)O<sub>3</sub> 0 ≤ x ≤ 0.8. Secondary peaks of bismuth titanate [11,20] ( or bismuth oxide [10,21] ) are indicated by symbol, ♦.

Figure 2. Lattice parameters for (1-x)Ba<sub>0.8</sub>Ca<sub>0.2</sub>TiO<sub>3</sub>-xBi(Mg<sub>0.5</sub>Ti<sub>0.5</sub>)O<sub>3</sub> as a function of composition x.

Figure 3. Relative permittivity versus temperature for  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3-x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ :  
(a)  $x = 0$ ; (b)  $x = 0.05$ ; (c)  $x = 0.1$ ; (d)  $x = 0.2$ ; (e)  $x = 0.5$ ; (f)  $x = 0.6$ .

Figure 4 Dielectric loss tangent versus temperature for  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3-x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ : (a)  $x = 0$ ; (b)  $x = 0.05$ ; (c)  $x = 0.1$ ; (d)  $x = 0.2$ ; (e)  $x = 0.5$ ; (f)  $x = 0.6$

Figure 5. Variation in  $T_m$  of  $(1-x)\text{Ba}_{0.8}\text{Ca}_{0.2}\text{TiO}_3-x\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$  as a function of composition  $x$  at 1 kHz