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## ACCEPTED MANUSCRIPT

Optimising dopants and properties in BiMeO<sub>3</sub> (Me = Al, Ga, Sc, Y, Mg<sub>2/3</sub>Nb<sub>1/3</sub>, Zn<sub>2/3</sub>Nb<sub>1/3</sub>, Zn<sub>1/2</sub>Ti<sub>1/2</sub>) lead-free BaTiO<sub>3</sub>-BiFeO<sub>3</sub> based ceramics for actuator applications

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#### **Abstract**

A crystallochemical framework is proposed based on electronegativity difference  $(e_n)$  and tolerance factor (t) to optimise the BiMeO<sub>3</sub> dopants and therefore the piezoelectric and electrostrictive response in BaTiO<sub>3</sub>-BiFeO<sub>3</sub> based ceramics. Compositions in the series 0.05Bi(Me)O<sub>3</sub>-0.25BaTiO<sub>3</sub>-0.7BiFeO<sub>3</sub> (BMe-BT-BF, Me: Y, Sc<sub>1/2</sub>Y<sub>1/2</sub>, Mg<sub>2/3</sub>Nb<sub>1/3</sub>, Sc, Zn<sub>2/3</sub>Nb<sub>1/3</sub>, Zn<sub>1/2</sub>Ti<sub>1/2</sub>, Ga, and Al) were fabricated using solid state synthesis and furnace cooled. Scanning electron microscopy and X-ray diffraction revealed that only Bi(Mg<sub>2/3</sub>Nb<sub>1/3</sub>)O<sub>3</sub> and BiScO<sub>3</sub> dopants, which lie in a narrow range of e<sub>n</sub> vs. t, form homogeneous ceramics, free from secondary phases reflected in their superior piezoelectric coefficients (d<sub>33</sub> ~145 pC/N). All other BiMeO<sub>3</sub> additions exhibited either secondary phases (Y) and/or promoted a two-phase perovskite matrix (Zn, Ga and Al). The promising initial properties of BiScO<sub>3</sub> doped compositions prompted further studies on  $0.05BiScO_3$ - $(0.95-x)BaTiO_3$ - $(x)BiFeO_3$  (BS-BT-BF, x = 0.55, 0.60, 0.625, 0.65, and0.70) ceramics. As x increased the structure changed from predominantly pseudocubic to rhombohedral, resulting in a transition from a relaxor-like to ferroelectric response. The largest  $d_{33}^*$  (465 pm/V) was achieved for x = 0.625 under 5 kV/mm at the crossover from relaxor to ferroelectric behaviour. BS-BT-BF with x = 0.625 showed >0.3% strain under 6 kV/mm up to 175°C, demonstrating its potential for actuator applications.

Keywords: Lead-free piezoelectric ceramics

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