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Densification of BTS Functionally Graded Materials during Sintering

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Introduction

Functionally graded materials (FGMs) belong to an attractive class of materials in which it is possible to obtain a gradient of properties cannot be attained in any monophase materials. Continuous changes in the properties of these materials for instance: *composition*, *grain size*, *porosity*, etc., result in the gradient of their features such as mechanical strength and thermal conductivity. During the years, FGMs have found applications in various functional materials, such as piezoelectric ceramics, thermoelectric semiconductors, and biomaterials.

Fabrication of FGMs by powder technology brings significant problems of anisotropic densification and consequently shape distortion of the components. An important processing goal for FGMs is to obtain high quality microstructure with desired grain size and density. During the thermal treatment, different graded layers in FGM show different shrinkage kinetics, i.e. different shrinkage rates and extents of shrinkage during sintering, as well as different final density. This phenomenon can lead to excessive shape distortion, warping, delamination, developments of cracks and micro-structural damage in the sintered FGMs. Therefore, it is desirable to predict the sintering process (shrinkage and anisotropy) for every graded layer in FGM and design sintering strategies to achieve high quality FGM without any form of deformation.

BTS ($BaTi_{1-x}Sn_xO_3$) powders are very important in ceramic industries. BTS powders improve the dielectric behavior of barium titanate ceramics, i.e. increasing of Sn content (up to 15 mol%) in barium titanate ceramics decrease temperature of phase transformation, also, increase dielectric constant. Furthermore, BTS powders are important for practical applications in ceramic capacitors, as well as for FGMs (in the form of monolithic ceramics with an uniaxial gradient of piezoelectric and/or dielectric coefficients). BTS FGMs are very useful because they have a broad transition temperature and high dielectric constant in a wide temperature range. Width of transition temperature range for BTS FGM depends on number of the layers, as well as on tin content in each of a graded layer.

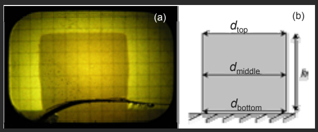
At the same thermal conditions, BTS powders with different tin content show different shrinkage rates and different extents of shrinkage during sintering, as well as different final density, which can lead to excessive shape distortion, warping, delamination, developments of cracks and microstructural damage in the sintered FGMs. Here, we chose BTS2.5/BTS15, BTS2.5/BTS5/BTS7, BTS15/BTS5/BTS7 and BTS2.5/BTS5/BTS7/BTS10 FGMs as model systems for prediction of the sintering processes (shrinkage and anisotropy) for every layer in FGMs and to design sintering strategies to achieve high quality FGMs without any form of deformation.

Experimental part

The initial BTS powders were prepared by a conventional solid state reaction between $BaCO_3$, TiO_2 and SnO_2 at 1100 °C during 2 h. FGMs were prepared by powder stacking method The BTS powders with different compositions were stacked sequentially in die; they were uni-axial pressed into cylindrical compacts (∅ 4 mm and $h \approx 2$ mm) under a pressure of 300 MPa. Combinations of the layers were 2.5-15, 2.5-5-7, 15-5-7 and 2.5-5-7-10 (numbers denotes mol% of Sn). The samples were sintered in a heating microscope (E. Leitz, Wetzlar, Germany) in order to determine the sintering shrinkage. The experiments were performed in air up to 1420 °C, using a heating rate of 10 °C/min. The sintering shrinkage of cylindrical compacts was recorded in axial and radial directions. A shrinkage anisotropy factor was determined based on experimental measurements.

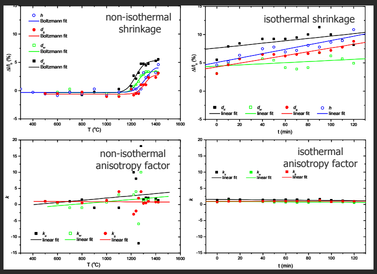
$$shrinkage(\%) = \frac{\Delta l}{l_o} \times 100$$

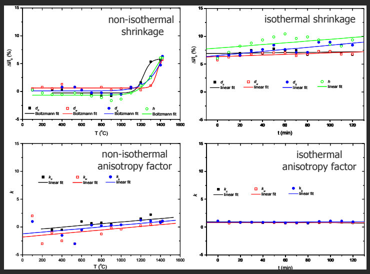
$$anisotropy \quad factor = k = \frac{\Delta d}{\Delta h} = \frac{do - d_i}{ho - h_i}$$

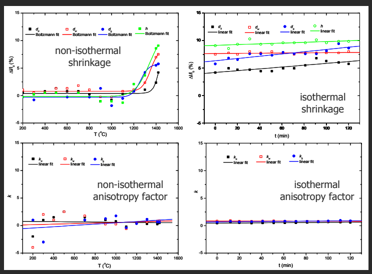


(a) image of sintering cylindrical FGM as observed in thermal microscope
(b) scheme of the geometrical parameters measured during sintering

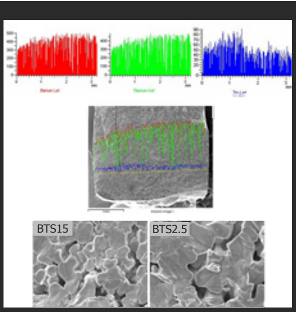
Scheme of FGMs samples



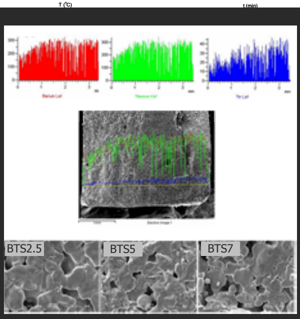




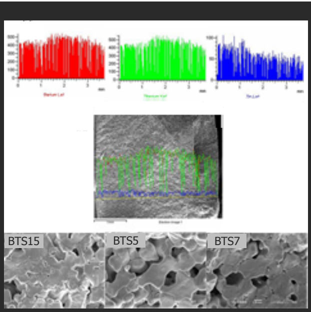
BTS2.5/BTS15



BTS2.5/BTS5/BTS7



BTS15/BTS5/BTS7



Shrinkage parameters during sintering of FGMs

FGM	shrinkage h			shrinkage d_{bottom}			shrinkage d_{middle}			shrinkage d_{top}		
	T (°C)	$(dl/l_o)_{ni}$ (%)	$(dl/l_o)_i$ (%)	T (°C)	$(dl/l_o)_{ni}$ (%)	$(dl/l_o)_i$ (%)	T (°C)	$(dl/l_o)_{ni}$ (%)	$(dl/l_o)_i$ (%)	T (°C)	$(dl/l_o)_{ni}$ (%)	$(dl/l_o)_i$ (%)
2.5_b-15_t	1270	5.0	10.9	1140	5.6	11.3	1200	3.4	6.3	1220	3.2	9.0
2.5-$_b$-5_m-7_t	1100	6.2	10.4	1100	5.8	7.8	1250	5.8	7.2	1080	6.4	9.4
7_b-5_m-15_t	1150	9.0	10.0	1300	4.3	6.9	1200	7.5	8.0	1120	5.8	8.0

Conclusion

It is shown that shrinkage and anisotropy factor of the FGMs depend on the ceramic's composition i.e. concentration gradient. Here, a smart choice of combination of the BTS layers and heating rate during sintering enable a preparation of high-quality FGMs, without any form of defects: shape distortion, warping, delamination, cracks and/or micro-structural damage.