

Fabrication of barium titanate by binder jetting additive manufacturing technology

S.M. Gaytan^{a,b,*}, M.A. Cadena^{a,b}, H. Karim^c, D. Delfin^c, Y. Lin^c, D. Espalin^{a,c},
E. MacDonald^{a,d}, R.B. Wicker^{a,c}

^aThe W.M. Keck Center for 3D Innovation, El Paso, Texas University of Texas at El Paso, El Paso, TX, USA

^bMetallurgical and Materials Engineering Department, The University of Texas at El Paso, El Paso, TX, USA

^cMechanical Engineering Department, The University of Texas at El Paso, El Paso, TX, USA

^dElectrical and Computer Engineering Department, The University of Texas at El Paso, El Paso, TX, USA

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Abstract

Fabrication of barium titanate (BaTiO₃) specimens was accomplished with binder jetting additive manufacturing, and build parameters (e.g., binder saturation and layer thickness) and sintering profiles were modified to optimize the density achieved and the crystal structures obtained in the 3D printed parts. Surface and cross sectional grain morphology was characterized by scanning electron microscopy (SEM) revealing grain growth on localized areas of BTO fabricated specimens after sintering. Crystal structure was analyzed by X-ray diffraction (XRD) where the presence of a hexagonal phase was observed for BaTiO₃ only when sintered at 1400 °C. The dielectric constant of the fabricated BaTiO₃ specimens sintered at 1260 °C was obtained by using a K_u-band wave-guide and vector network analyzer setup in which the relative permittivity was measured from 8.6 to 6.23 for a frequency range of 12.4–18 GHz, respectively. When sintered at 1400 °C for 4 h, a density of 3.93 g/cm³ was obtained, which corresponds to 65.2% of the theoretical density. Piezoelectric properties exhibited a d_{33} value of 74.1 for specimens also sintered at 1400 °C. Results reported in this paper demonstrate the feasibility of BTO as a binder jetting material for 3D printed dielectric structures, ceramic capacitors and gas and pressure sensors.

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1. Introduction

Among several ceramics used for their ability for polarization is the dielectric BaTiO₃ (BTO) with applications varying from chip capacitors to dynamic random access memories [1], and embedded capacitance in printed circuit boards to piezoelectric devices like sensors, heaters and transducers [2,3]. Other applications that use BTO for ferroelectric and piezoelectric properties include semi-conductors with positive temperature coefficient of resistivity (PTCR) [4], dynamic access memories (DRAM), and IR sensors

[5]. BTO has a curie temperature at 120 °C and depending on its crystal structure, BTO can be para-electric (cubic; $a=4.031$ Å) or ferroelectric (tetragonal; $a=3.994$ Å and $c=4.038$ Å) [6,3], which are desired structures for sensor applications due to the properties provided (pyroelectric and piezoelectric). Ducharme [7] stated that BTO has the largest dielectric constant when being near the ferroelectric phase due to polarization from ion displacement or the alignment of permanent dipoles.

Some deposition methods used for BTO are vacuum evaporation, sputtering, laser ablation, hydrothermal synthesis, radio frequency plasma sputtering and sol–gel techniques used for BTO thin films preparation [8]. Some of these techniques still have drawbacks since cracking and peeling of the deposited layers occurs during the final heat-treating process, which is required to crystallize the as-deposited amorphous film [9].

*Corresponding author at: The W.M. Keck Center for 3D Innovation, El Paso, Texas University of Texas at El Paso, El Paso, TX, USA. Tel.: +1 915 747 6391; fax: +1 915 747 5019.

E-mail address: smgaytan@utep.edu (S.M. Gaytan).

The methods previously mentioned are designed for the fabrication of thin films where powder is in the nano-range or as a stable colloidal solution. One method used for the fabrication of undoped BaTiO₃ ceramics is by slip casting of slurries or sol-gel solutions to create thin dielectric layers [10–12]. Even when research has been pursued to improve dielectrics such as barium titanate, attempts have been made via 3D printing for this material like the use of inkjet printing to create capacitors with a low volume of polymer ink [13], and the fabrication of 3D piezoelectric BTO nanoparticles embedded in a polyethylene glycol diacrylate matrix by digital projection printing where d_{33} values of 40 pC/N were obtained for a maximum particle loading of 10 wt% [14,15]. Also, fabrication of ferroelectric photoactive suspensions by stereolithography has been reported for maximum BTO ceramic volume of 40% in hexanediol diacrylate, which proved to have poor curing behavior due to the difference on surface reflectance of the ceramic and the resin used [16].

BTO has a melting temperature of 1654 °C. Sintering temperatures of 1200 and 1300 °C, for 1 mm-thick specimens from powder particle size of 35 nm have been reported with a maximum grain growth of 2.2 µm (6185.71% grain growth) [17]. Sintering temperature of 800 °C applied for 10 h using 30 nm diameter BTO powders provided a theoretical density of 90% [2]. Density increase and sintering behavior for BTO ceramics is a continuously researched topic; one reason is the need for porous structures with gas and liquid permeability for humidity sensors [18]. For simple 3D printed parts, cold isostatic pressing is used to increase the density, but when the geometry is complex, the internal enclosed cavities have to be supported to prevent the collapse of the structure [19].

It is foreseen that fabrication of BTO by 3D printing will enhance current fabrication methods for pressure sensors and ultracapacitors. The dielectric material can be customized to the required geometries and mass produced, therefore, a cost reduction is also foreseen. Additionally, porous structures that have gas and liquid permeability can be fabricated for humidity sensors applications [18]. Additive manufacturing provides the advantage of modifying porosity size and porosity homogeneity as necessary for optimum properties of the sensor. In this study, a binder jetting technology was used to fabricate robust geometric specimens of 12 × 12 × 3 mm³ by using commercial BTO powder. Coupon specimens were fabricated with different binder saturation percentages and sintered at different temperatures in an attempt to obtain fully dense specimens. Part shrinkage was calculated and used to obtain accurate dimensions for dielectric constant measurements. XRD data showed no signs of contamination after sintering and corroborated the maximum sintering temperature allowed for BTO to remain in the cubic and tetragonal phases. Dielectric constant values were obtained and presented in this paper for a frequency range of 12.4–18 GHz. Since it is known that dielectric constant increases with smaller grain size or higher density [20], it is expected that dielectric constant values obtained in this project can be further increased if grain growth can be further minimized. It is expected that by increasing the density and, as previously mentioned, reducing the grain size, the dielectric and piezoelectric properties can be enhanced sufficiently to enable the manufacture of geometrically

complex high-K dielectrics necessary for ultra-capacitors or conformal piezoelectric structures for customized energy harvesting applications.

BTO fabricated by 3D printing can provide the necessary means to create dielectrics for ultracapacitors in the bulk size, where an ultracapacitor with a capacitance of up to 5000 F can measure up to 5 × 5 × 15 cm³. Ultracapacitors can be used in a number of functions for a car, such as the air conditioning, door locks, seats and windows, the seat adjustment mechanism, and in the acceleration and braking modules [21], the generator action of the piezoelectric ceramic can also be used in fuel-igniting devices, and solid state batteries. Additionally, BTO by binder jetting can be the next fabrication method for vibration control by the creation of piezoelectric systems with coupled actuators [22]. The freedom of 3D printing can enable custom, conformal, geometrically-complex piezoelectric devices for next generation energy harvesting in the consumer, automotive, bio-medical, aerospace and defense industries. One transformative example includes the possibility of an energy-harvesting active bio-medical implant the shape of which could be customized for a unique human anatomical geometry (e.g. a bone implant for a specific patient that will eliminate the need for battery replacement and resulting follow-on surgeries).

2. Materials and methods

2.1. Materials

Spherical barium titanate (IV) (BaTiO₃) powder of +325 mesh (0.85–1.45 µm) (Fisher Scientific, USA) was used for fabrication by binder jetting. Fig. 1(a) shows as-received powder where the presence of agglomerations and a binding agent can be visualized. Since powder sifting is not a feasible path to remove powder agglomerations due to the small powder size of BTO, it was determined to heat the BTO powder for 1 h to a maximum temperature of 1000 °C with a ramp up rate of 10 °C/min after fabrication for recycling purposes throughout the length of the project. Prior to heating the powder, it was noted that in order to achieve a uniform spread layer of BTO, it was necessary to level the powder bed and add ~10 extra layers of powder in order to avoid unfilled areas. Fig. 1(b) shows powder after heating, and even though the powder continues to display some signs of agglomerations, the spreading behavior in the printing system was improved when compared to the as-received powder. The flowability of the powder was determined to have improved after the heating cycle since the extra layers of powder were not necessary to obtain a uniform layer of powder. For this reason, it was determined that in order to reuse the powder, and remove any excess of binder and agglomerations, a heating cycle was implemented prior to each fabrication. Thermal gravimetric analysis (TGA), as shown in Fig. 2, was performed on the fabricated samples prior to sintering to determine the debinding temperature (necessary to remove the binder used during fabrication; the process is explained in detail below). BTO spherical powder shows a pronounced change in mass at a temperature range between 375 and 425 °C. TGA analysis demonstrates that the last change in mass occurred at 450 °C

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