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Optimization of 3D printing parameters for BaTiO₃ piezoelectric ceramics through design of experiments

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Abstract

Paste extrusion is an additive manufacturing technique that deposits paste through a nozzle in a layer-by-layer fashion to form a three-dimensional object. This technique can be used to fabricate piezoelectric ceramics with complex geometries and excellent material properties to fabricate sensors, capacitors, and energy storage devices with a high level of customization. Different factors can affect the quality of piezoelectric ceramics when using the paste extrusion technique, for example, the composition of the paste and printing parameters. This paper presents a study to optimize the piezoelectric coefficient, relative density, dielectric permittivity and dimensional accuracy of BaTiO₃ by using design of experiments with a $2^{k-1}$ design. Results indicated that the factors having a significant impact on the end-product outputs were the BaTiO₃ particle size and the binder concentration on the paste formulation. Using the optimal levels makes it possible to achieve a piezoelectric coefficient higher than 200 pC/N and a density of 90% of the theoretical value of pure BaTiO₃.

1. Introduction

Additive Manufacturing (a.k.a. 3D Printing) consists of the deposition of material in a layer-by-layer fashion to form a three-dimensional object. This technique is widely used for rapid prototyping, especially when the fabrication of complex geometries is required. Materials including metals, polymers, and ceramics are used for additive manufacturing processes [1]. Literature reports efforts in the fabrication of ceramics using different additive manufacturing techniques such as material jetting, material extrusion, direct energy deposition, binder jetting and stereolithography [2, 3]. The fabrication of ceramics is an area of interest due to their excellent thermal, mechanical, and electric properties, which are useful in several applications in the area of electronics and beyond [4, 5]. However, ceramics fabricated using additive manufacturing are hard to process because they must be sintered to remove the binder content to obtain a densified sample [3]. This debinding process can lead to nonconforming samples with high levels of porosity especially for powder-based methods, or inhomogeneity causing cracks and reducing the mechanical strength of the final product for slurry-based methods [6]. For this reason, the fabrication of ceramics with designed complex geometries and defects-free becomes a challenge.

Many research studies have been reported on the fabrication of technical or functional ceramic objects using free-forming techniques. Freeze-form Extrusion Fabrication (FEF), Ceramic Extrusion On Demand (CODE) and Robocasting are some examples of this technique [7]. For technical ceramics, researches commonly reported the use of Alumina (Al₂O₃) [3, 8, 9], Silica (SiO₂) [10] and Silicon Nitrate (Si₃N₄) [11]. For example, Huang et al used FEF to fabricate complex geometries with Al₂O₃ achieving a density of 90% with respect to the theoretical value [9]. For functional ceramics, researchers reported studies using ferroelectric ceramics [12] and magnetic ferrite ceramics [13, 14]. Ferroelectric ceramics are specifically of interest due to their piezoelectric properties.
properties. Piezoelectricity refers to the generations of an electric signal when mechanical loads are applied or vice versa [15]. Pb(ZrxTi1-x)O3 or PZT is a ferroelectric ceramic known to have good piezoelectric properties ($d_{33} \sim 500$–$600$ pC/N) [16], and it is widely used for sensors and capacitors applications [17]. However, the use of toxic materials such as lead oxide (PbO), especially for electronic devices could increase the environmental pollution and harm the human body [18–21]. Many efforts have been focused on lead-free ferroelectric ceramics, such as KNN, BT, BNT based ceramics [19, 20]. Barium Titanate (BaTiO3 or BTO), is known to have good piezoelectric properties ($d_{33} \sim 190$ pC/N) [21] and can be used as sensors and capacitors with comparable properties to PZT.

Although Additive Manufacturing enables the fabrication of ceramics with complicated design, the optimization of the fabrication process is still required. Design of Experiments (DOE) is a statistical tool that evaluates different factors and their impact in the end-product output. DOE also allows to determine the optimal level of these factors to maximize the desired output. Several authors have used statistical analysis to optimize the fabrication process for different additive manufacturing techniques [22–25]. For example, Chen and Zhao investigated the process parameters that influence the surface quality of stainless steel powder manufactured with Binder Jetting technique using Signal to Noise ratio and ANOVA analysis. The factors considered in this study were layer thickness, printing saturation, heater powder ratio and drying time. They demonstrated that layer thickness was the most significant factor affecting surface roughness. The drying process showed the highest impact on the shrinkage of the samples [22]. Yan et al investigated the factors that affect the liquid phase migration of Al2O3 fabricated with FEF. The factors studied were extrusion velocity, extrusion interval time and nozzle length and analyzed with an orthogonal design of experiments. It was found that extrusion velocity and extrusion interval time have an impact on the liquid phase migration of FEF [25].

Even though a considerable amount of research has been dedicated to the optimization of ceramics, there is insufficient work reported on functional ceramics such as piezoelectrics. To obtain high-quality ceramics, techniques such as FEF have been developed to increase density and piezoelectric properties. FEF is an additive manufacturing technique that extrudes and deposits paste through a nozzle onto a substrate below the freezing temperature of the paste. This free-forming technique is an inexpensive slurry-based method that can fabricate complex designs of ceramics with better shape retention and high density. This paper aims to optimize the fabrication of BaTiO3 ferroelectric ceramics using FEF technique through DOE with a fractional factorial $2^{k-1}$ design. This study considers the evaluation of four different factors: particle size, binder amount, nozzle diameter, and printing speed.

2. Experimental details

2.1. Materials and paste preparation

BaTiO3 powder supplied from Inframat (Manchester, CT) was selected as solid loading of the paste. Polyvinyl Alcohol (PVA) powder (Mw=89,000–98,000) supplied from Sigma Aldrich (St. Louis, MO) was determined to be the binder in the ceramic suspensions. For the paste preparation, PVA powder was dissolved in deionized water using a stirring rate of 500 rpm and let to dwell for 20 min. Then, the solution was placed in a hot plate at 90 °C for 20 min to achieve a homogeneous dispersion of the PVA powder. After that, BaTiO3 powder was added to the solution and mixed until a homogenous paste was obtained. All the ceramics suspensions were fabricated using 70 wt% BaTiO3 and 30 wt% PVA solution. Lastly, the paste was deposited into a syringe tube and sealed immediately to prevent water evaporation.

2.2. Factors and levels

A two-level fractional factorial design $2^{k-1}$ was selected to optimize the fabrication of BaTiO3 ferroelectric ceramics. This statistical design considers the evaluation of $k$ factors with two levels of interest. However, when a large number of factors are evaluated, it is recommended that a $k-1$ design is used, as the number of runs per replications could overcome available resources [26]. Table 1 refers to the factors and levels considered for this study. BaTiO3 particle size was selected as a factor, using 300 nm as high level and 100 nm as low level. The particle size of BaTiO3 is a factor of interest as it may influence the mechanical properties of the final product [27]. The PVA concentration was also selected, since the binder content may affect the rheology of the ceramic suspensions [28]. The high level of PVA was set to 13 wt% in deionized water, and the low level was set to 9 wt% in deionized water. Another factor was printing speed; since different flow-rates can affect the relative density and the dimensional accuracy of the green bodies [29]. The high and low level for printing speed was set to 20 mm s$^{-1}$ and 10 mm s$^{-1}$ respectively. The last factor selected was the nozzle diameter; the high level was set to 1.2 mm while the low level was set to 0.8 mm. The nozzle diameter can possibly affect the dimensional accuracy of the green bodies due to the shear stress generated in the walls of the nozzle and the stress required for the piston of the machine to achieve a constant flow [30]. The fractional factorial design $2^{k-1}$ was created with
Minitab using the information provided in table 1. This study considers three replications and a randomized order to validate the results. Table 2 shows an example of the runs required, considering all the different runs for one replication.

The factors and levels selected in table 1 are justified with the rheology characterization of the paste formulation. Proper rheology can guarantee a constant flow and printability of the samples [30]. All the different pastes were measured in a DHR-2 rheometer (TA Instruments, New Castle, DE) at room temperature, using a parallel geometry with a 1.2 mm gap. Figure 1(A) shows the viscosity of each ceramic suspension as a function of shear rate in the ranges of 1 to 50 s$^{-1}$. While figure 1(B) shows a magnified version of the viscosity in a shear rate range of 10 to 30 s$^{-1}$. Figure 2 shows the stress profile at different shear rates in the ranges of 1 to 50 s$^{-1}$. All the ceramic suspensions presented a shear thinning behavior [30, 31] as low shear rates require higher force due to the initial viscosity of the paste. However, high shear rates require lower applied force. The initial high force is required to generate the flow through the nozzle. Also known as the dynamic yield stress of the fluid [30]. It was found that the suspensions with higher PVA concentration had a higher viscosity and thus required more initial force applied. Furthermore, it was observed that finer particle size of BaTiO$_3$ also affects the viscosity. The rheology of the different ceramic suspensions is in good agreement with literature according to the following. PVA behaves as a non-Newtonian fluid in aqueous solutions and exhibits different viscoelastic properties depending on the concentration [30]. The increment on the viscoelastic properties observed can be attributed to a stronger hydrogen-bonding interaction of PVA when increasing its content [30]. Literature suggest that particle size also

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affects the rheology of the suspensions. Finer size could increase the contact between particles dispersed in the suspension affecting the viscosity [31]. The increase in viscosity can be attributed to liquid trapped inside aggregates of powder within the paste [32]. Figure 2 shows the printability zone at shear rates of 30 to 50 s$^{-1}$ with ranges in viscosity between 40 to 10 Pa.s. Different viscosity values will affect the flow rate of the suspension during the fabrication of the samples and needs to be evaluated.

2.3. Fabrication process

The samples were printed using a Printrbot Simple Metal 3D printing machine, which was modified to extrude paste. A cylinder of 1-inch outer diameter, $\frac{1}{2}$-inch inner diameter, and $\frac{1}{2}$-inch height was selected as the standard sample for this study. Figure 3(A) shows the CAD design of the cylinder in Slic3r software. The printing process of the samples follows a randomized order. Additionally, the samples were identified immediately with the run number according to the DOE table (supplementary section 1–figure 9 is available online at stacks.iop.org/MRX/6/085706/mmedia). During the printing, an aluminum freezing plate was placed onto the substrate at $-40^\circ$C. The plate was covered with a thin film to allow for an easier detachment of the samples. After printing, the aluminum plate was removed from the bed and placed in a freezer at $-60^\circ$C (Thermo Scientific TSUTM Series $-86^\circ$C Upright Ultra-Low) for 12 h to dry some of the water content by sublimation. To prevent the generation of cracks, the samples were placed upside down in the freezer for another 30 min. The samples were also dried in an electric oven (Lab companion OF-01E) at 40 $^\circ$C for 24 h to remove any remaining water from the green body. A slow drying process under low temperatures prevents the generation of microcracks in the green bodies [33]. Figure 3(B) shows an example of an as-printed BaTiO$_3$ sample. The angle after drying was measured, comparing the displacement of the last printed layer with respect to the first layer (supplementary section 2–figure 10). Some of the samples presented an inferior ability to stack the material during the printing process. This issue was observed mainly on the pastes that contained a higher water content level. The density of the green bodies was measured using Archimedes’ principle and compared with the theoretical density of

![Figure 2. Stress behavior as a function of shear rate for each BTO-PVA ceramic suspension.](image2)

![Figure 3. (A) Cylinder design on Slic3r (B) BaTiO$_3$ green body ‘as-printed’.](image3)
BaTiO$_3$ of 6.02 g cm$^{-3}$ [34] (supplementary section 3-table 3). The density of the green bodies was 1.67 g cm$^{-3}$ on average before sintering. This relatively low-density value is attributed to the amount of water and binder content in the as-printed samples.

2.3.1. Post-processing and material characterization

All the samples were debinded at 600 °C for one hour and sintered at 1250 °C for two hours at 5 °C min$^{-1}$ ramp. After sintering, the samples were polished using sandpaper 240-grit. Conductive silver paint was applied in the top surface, then dried at 400 °C for 15 min. The same process was repeated for the bottom surface of the samples. Each sample was exposed to thermal poling on silicon oil for 2 h for dipoles alignment. The electrical field and the temperature were set to 5.4 kV cm$^{-1}$ and 90 °C respectively [35]. The piezoelectric coefficient was calculated using a d$_{33}$ meter (APC YE2730A). Dielectric permittivity was calculated with an LCR meter (1920 Precision, IET lab).

The grain morphology characterization of the samples after sintering was observed using a scanning electron microscope (SEM, TM-1000 Hitachi). Figure 4(A) shows the cross-section of a 300 nm BaTiO$_3$ and 9% PVA sample after sintering. The density for this sample was 4.03 g cm$^{-3}$, which represents 66.95% of the theoretical value. It was also observed that the grain size was $\phi \approx 0.9 \mu$m on average. Figure 4(B) presents the cross-section of a 100 nm BaTiO$_3$ and 13% PVA sample after sintering. The density for this sample was 5.22 g cm$^{-3}$, which represents 86.75% of the theoretical value. The 100 nm BaTiO$_3$ sample showed bigger grain sizes of $\phi \approx 1.8 \mu$m on average. There is an evident increase in density for finer particle sizes. Researchers suggest that finer particle size tends to achieve a higher grain growth due to high curvature of the ceramic powder, which requires a lower change in energy during sintering [36].

Figure 4. (A) Cross section of a 300 nm BaTiO$_3$ 9 wt% PVA sample after sintering (B) Cross section of a 100 nm BaTiO$_3$ 13 wt% PVA sample after sintering.

Figure 5. XRD of 100 nm and 300 nm BaTiO$_3$ particle size in range 2θ from 20° to 80°.

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Crystal structure of the samples was analyzed by x-ray diffraction (XRD) on a D8 discover diffractometer (Brucker). Figure 5 shows the XRD for 100 nm and 300 nm BaTiO$_3$ particle size after sintering using a range $2\theta$ from 20° to 80°. The peaks splitting at (002) and (200) for both particle size index for a tetragonal crystal structure [37, 38]. Previous studies report that a transition from cubic to tetragonal phase can be observed in BaTiO$_3$ using heat treatments above 1000 °C [39–41]. A higher intensity of (002) and (200) peaks was observed for the 100 nm BaTiO$_3$ sample, which can be attributed to a higher amount of crystals in the tetragonal phase. Researchers reported that the growth of BaTiO$_3$ crystallite strongly depend on the heat temperature and exposure time during the sintering process [40].

3. Statistical results

This study considered the piezoelectric coefficient, dielectric permittivity, density, and dimensional accuracy as critical outputs to fabricate BaTiO$_3$ ferroelectric ceramics. All the experimental results were collected (supplementary sections 4–7, tables 4–7) and analyzed in Minitab. The normal probability plots of the standardized effect provided in figure 6 presents the factors that have a statistical significance with respect to every individual output at a 95% confidence level. The interaction plot (supplementary section, figures 11–13; figure 7) evaluates the effect of two variables and their impact on the end-output at a 95% confidence level. The interaction plot is significant when the effect of a factor with a specific output depends on another factor [42]. Lastly, the main effect plots shown in figure 8 presents an analysis of factors and levels that maximize every output.

For the piezoelectric response shown in figure 6(A) the only significant factor was the BaTiO$_3$ particle size. Figure 8(A) indicates that the 100 nm particle size is the level that will maximize the piezoelectric coefficient of the samples. This result is a good agreement with literature, which indicates that an increase on piezoelectric properties is observed when decreasing particle size of BaTiO$_3$ [36, 37]. Figure 6(B) indicates that the factors affecting the dielectric permittivity were the PVA concentration and the interaction (figure 7) of PVA and BaTiO$_3$. Figure 8(B) shows that 13% PVA is the level that will increase the dielectric permittivity. Results in the interaction plot indicate that the combination of PVA and BaTiO$_3$ that maximize the output is 100 nm BaTiO$_3$ and 13% PVA concentration. PVA is a carbon base binder, and although PVA was burnout during sintering, literature indicates that carbon residues may be encapsulated affecting the permittivity of ceramics [38, 43]. Figure 6(C) shows that the only significant factor that has an impact on the density was the BaTiO$_3$ particle size.
Figure 8(C) shows that the 100 nm BaTiO3 particle size will achieve a higher density. The influence of the ceramic particle size is consistent with previous studies [35] suggesting that finer particle size normally achieve a higher grain growth due to high curvature of the powder. For dimensional accuracy, it was found that the PVA concentration was the only significant factor as shown in figure 6(D). Figure 8(D) indicates that 13% PVA is the level that minimizes the angle of deformation and enables a better stack of material. This result is in good agreement with the rheology of the paste (figures 1–2), which indicates that there is an increase in viscosity for higher binder contents.

Based on the results obtained in the factorial 2^k-1 design, it was found that the parameters that positively affect the end-output are 100 nm BaTiO3 and 13% PVA concentration. There is no evidence to claim that the printing speed and the nozzle diameter affects the output evaluated in this study. Therefore, to optimize the fabrication of BaTiO3 ferroelectric ceramics, the printing speed and nozzle diameter can be neglected and use either level of said factors. From the experimental results, a predictive probability model can be created by minimizing the uncertainty on the estimated effect for different design points [42]. The probability cube plot (supplementary section 9, figures 14–17) considered a fitted means analysis, which is useful to predict the outputs in a balanced design [26]. The fitted means analysis indicates that 100 nm BaTiO3 and 13% PVA concentration enables the fabrication of ferroelectric ceramics with a piezoelectric coefficient higher than 200 pC/N. While the 300 nm BaTiO3 can achieve a piezoelectric coefficient of only 135 pC/N. The mean predicted piezoelectric coefficient represents 48% higher value compared to any other combination levels analyzed in this study. Dielectric permittivity is predicted to be higher than 2200. Additionally, using the optimal interaction levels from the probability cube for PVA and BaTiO3 makes it possible to achieve 33% higher permittivity. The density is predicted to be about 90% with respect to the theoretical value of pure BaTiO3, which is 33% higher than any other combination of levels. Additionally, a low angle of deformation of about 10 degrees can be achieved, representing a 44% increase in dimensional accuracy. These predicted outputs are comparable with the BaTiO3 theoretical values, proving its capabilities to use FEF 3D printing technique for the fabrication of complex geometries for commercial applications.

4. Conclusion

A DOE for BaTiO3 ferroelectric ceramics using FEF 3D printing technique was presented. This paper aimed to optimize the formulation of the ceramics suspensions to produced defects-free samples. This study considered the evaluation of BaTiO3 particle size, PVA concentration, printing speed, and nozzle diameter. Results showed
that the BaTiO$_3$ particle size positively affects the piezoelectric coefficient, dielectric permittivity and density of the samples. In another hand, it was found that the PVA concentration has an impact on dielectric permittivity and dimensional accuracy. The optimal levels are 100 nm for BaTiO$_3$ and 13% PVA concentration. The printing speed and nozzle diameter do not influence the desired end-product outputs. Using the optimal levels in the paste formulation, it is possible to fabricate samples with 90% density. A piezoelectric coefficient higher than

![Figure 8. Main effect plot for (A) piezoelectric coefficient (B) dielectric permittivity (C) relative density (D) dimensional accuracy.](image)
200 pC/N and a dielectric permittivity higher than 2200 can be obtained. Additionally, a low angle of deformation of about 10 degrees can be achieved, which is a good parameter for dimensional accuracy. These results correspond to the full theoretical properties normally achieve by traditional manufacturing techniques for BaTiO₃ ceramics. Therefore, this past extrusion technique can be used in the fabrication of more complex geometries and extend the applications of piezoelectric ceramics for sensors and actuators with designed geometry.

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