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3D printing high density ceramics using binder jetting with nanoparticle densifiers

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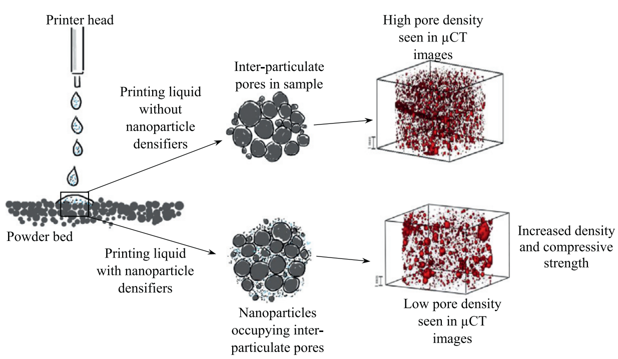
HIGHLIGHTS

- Alumina nanoparticles were introduced into alumina samples made via experimentally simulated binder-jetting process.
- Adding nanoparticles improved sample density and compressive strength while decreasing sample porosity.
- Printing liquid surface tension decreased due to nanoparticles which in turn decreased its penetration depth in powder bed.

GRAPHICAL ABSTRACT



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ABSTRACT

This study examines the effects of nanoparticle densifiers added to printing liquid on the mechanical performance and manufacturability of ceramics made using binder jetting. "Green" alumina samples were synthesized with filler particles of average particle size 40 μm embedded with nanoparticles of average size of 50 nm suspended in the printing liquid with varying concentration of 0–15 wt%. Samples were characterized for density, porosity, compressive strength, and printing liquid penetration depth in the filler powder layer assessed using surface tension testing. Results showed that the presence of the nanoparticle had a marked effect on the physical and mechanical properties of the samples whose relative density increased by about 30%. MicroCT imaging of the samples showed a decrease in interparticle pores with an addition of 15 wt% alumina nanoparticles. Compressive strength improved by 743%, from 76 kPa to 641 kPa as the densifier content was increased from 0 to 15 wt%. Surface tension of the printing liquid decreased from 44 mN/m to 23 mN/m with increasing densifier concentration from 0 to 15 wt% indicating that the penetration depth of the printing liquid would decrease with increasing densifier content. Implications of this approach on high density ceramic part printing efficiency are discussed in detail.

1. Introduction

Recent advances in the binder jetting printing (BJP) are making it a viable option for facile manufacturing of ceramic parts with complex geometry, due to faster manufacturing time and lower operation costs [1]. BJP is an additive manufacturing process where structural material in the form of powder, also referred to as fillers, is connected with the help of binder or printing liquid deposited by a print head resembling that of an inkjet printer in selective regions. The printed part can then be subjected to post processing to make the final part [1]. BJP can be used to manufacture parts made with a wide range of materials

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including, but not limited to, metals, ceramics, and polymers [2–5]. However, while BJP has been successful in producing complex ceramic parts, the applications of printed products are mostly limited to prototypes due to the inability of the printed parts to meet performance criteria. This is attributed to high volume of pores in the printed parts and consequent low density [6] due to the limited packing factor of the powder particles. Subsequently, high volume shrinkage is observed during post-processing resulting in dimensional inaccuracy and creep formation [5]. Also, the presence of porosity makes the printed parts brittle and has a negative effect on mechanical properties like compression strength, flexural strength, and bending strength [7–9].

Binder-jetted parts are typically post-processed for densification using techniques such as sintering, infiltration with a second phase material, and hot isostatic pressing [10, 11]. Sintering is the most common method of densifying binder-jetted ceramics during which the filler particles adhered by the binder are welded locally by heating the printed parts to temperatures below their melting point. Consequently, the filler particles coalesce at their points of contact owing to solid state diffusion [12]. This, however, leads to the post-processed ceramic part demonstrating shrinkage causing both dimensional inaccuracy and creep [13]. In addition, pores formed between the powder particles during the printing process (called inter-particulate pores) are not eradicated, leaving the 3D printed part with considerable amount of porosity [14].

Infiltration involves immersing the printed part in a solution containing a secondary material, called infiltrant, such that it fills the inter-particulate pores of the sintered binder-jetted part via capillary action. The infiltrant can be either a multi-phase fluid such as a solution with suspended particles, or a single-phase fluid such as molten metal or polymer [15]. Usually ceramics are subjected to infiltration to reduce the porosity, improve mechanical performance, as well as to manufacture cermets or alloys [16]. However, a major drawback of this method of part densification is the insufficient penetration of the infiltrant into pores in the binder-jetted part with size smaller than the average particle size of the solid loadings [9]. Similar problems are also noticed when the viscosity of the infiltrant increases either with the increase in the solid loadings or with the use of a viscous liquid metal or polymer. Additionally, the effectiveness of infiltration depends on the connectivity of the pore network developed in the printed part. Those pore clusters that are not connected to the outer surface of the printed part via channels remain unfilled with the infiltrant. All of these issues lead to having areas of inhomogeneity in the binder-jetted part due to pockets of pores [17].

A recent approach to improving green density of binder-jetted parts has been to use multimodal filler particles. Particles with an average size greater than 20 μm are easy to spread while fillers smaller than 5 μm occupy the interstitial positions between the larger particles, decreasing porosity [18]. Gonzalez et al. studied the effect of filler particle size on the density and compressive strength of binder-jetted parts synthesized with aluminum oxide powders of three different sizes: 53 μm, 45 μm, and 30 μm which were 3D printed, heat-treated for 2 h at 195 °C to cure the binder, and then sintered at two different time intervals of 2 h and 16 h. They showed that samples made with a combination of powder sizes had a relative density of 96.51% while samples printed with 53 μm particles alone had a lower relative density of about 64%. Binder -jetted samples with multimodal filler particle distribution also showed the highest compressive stress value of 146.6 MPa [19]. They hypothesized that the smaller particles tend to occupy inter-particulate pores in powders dispersions and improve the density of the green printed parts, resulting in mechanical properties after post processing.

An extreme instance of this strategy is to use nanoparticle powders as both densifiers (occupying inter-particulate pores between the fillers in the powder bed) and sintering aids [56], [57]. In such situations, the fillers are assumed to have a bimodal particle size distribution with the larger micron size particles remaining in the powder bed and the smaller nanoscale particles acting as the densifiers. Nanoparticles may be introduced as an infiltrant after a binder-jetted part is printed and sintered as discussed before, or by suspending them within the binder itself and introducing them into the part during the printing process. The ideal method of including nanoparticles is via the binder itself since that precludes the disadvantages of infiltration post part-sintering. One of the first reports of using nanoparticles to densify binder-jetted parts was by Crane et al. who infiltrated sintered 410 stainless steel parts made using BJP with iron nanoparticle suspensions to achieve 60% reduction in part shrinkage and 95% decrease in the creep deflection compared to the samples printed without nanoparticles [22]. Similarly, Bai et al. showed that silver parts binder-jetted with printing liquid containing 20 wt% silver nanoparticles showed increased sample green density from 2.116 g/cm^3 to 2.184 g/cm^3 and an increased engineering tensile strength from 46 to 55 MPa after sintering at 850 °C for 20 min [20]. Zhao et al. used a 10 wt% zirconia nanoparticle suspension in the printing liquid to print zirconia ceramic samples [23]. They showed that by increasing the binder content (amount of printing liquid dispensed into each layer of powder bed) from 50% to 125%, the relative density of the printed parts increased from 75.2% to 86.8% while linear shrinkage decreased from 22.3% to 10.6% after sintering [23].

Most of the current research focused on improving density and related mechanical properties of binder-jetted specimens by including nanoparticles in the printing liquid examined the effects of binder saturation level and post processing parameters such as sintering profiles on mechanical performance of the samples. However, the effects of nanoparticle densifier concentration in the printing liquid on porosity, density, and mechanical performance are still largely unexamined. There is a critical need to bridge this knowledge gap in order to effectively design additive manufacturing process such that manufactured parts demonstrate optimal performance parameters desired for variegated applications. It is also essential to assess the effects of nanoparticle densifiers on the manufacturability and performance of binder-jetted parts to complement existing literature. As a preliminary step, in this study, we explore the effect of nanoparticle densifiers suspended in the printing liquid on the density, porosity, compressive strength, and part manufacturability of binder-jetted, green alumina parts. We hypothesize that the density, porosity, and compressive strength of binder-jetted, green alumina samples will be influenced by the nanoparticle densifier content suspended in the printing liquid in that higher content will lead to improved bulk performance. Additionally, we also anticipate that the nanoparticles will affect the penetration depth of the printing liquid in the filler particle layers in the powder bed, which determines the effective binding between the alumina particles in the powder bed, thus ultimately affecting the printing efficiency. We explore manufacturability by evaluating the surface tension of the printing liquid with nanoparticle densifiers.

2. Experimental

2.1. Materials

Alumina (Al_2O_3) powder (AdValue Technology, USA, 99.9% purity) with mean particle diameter of 40 μm was used as the filler particles which form the principal structural component of the 3D printed part. Alumina nanoparticles (Sigma-Aldrich, USA) with a particle size less than 50 nm were used as nanoparticle densifiers suspended in the printing liquid. Polyvinyl alcohol (PVA) (Sigma Aldrich, USA, M_w: 9000–10,000 g/mol, 80% hydrolyzed) was used as both binder and dispersant/surfactant. Deionized (DI) water was used as the solvent in the printing liquid. The filler powder was sieved through 150 mesh, and the rest of the materials were used as received.