Mechanical Properties of Interconnected Phase Alumina-Al Composites

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Abstract

Interconnected phase alumina-Al composites were fabricated by pressureless reactive metal infiltration of porous mullite ceramic preforms. Controlled porosity mullite ceramics were processed via the indirect fused deposition. Volume fraction, size and distribution of metals were varied in the composite to understand their effects on physical, thermal and mechanical properties. Mechanical testes were conducted under uniaxial compression and four-point bend loading. Fractographic analyses of these samples were performed to understand the mechanism of failure under different types of loading. The paper describes processing and characterization of structured alumina-Al composites.

Introduction

Processing of different types of metal-ceramic composites with numerous microstructures have been reported for the past three decades. These composites can be classified into two main groups namely, metal matrix and ceramic matrix based on the composition and distribution of the component phases. For the metal matrix composites, ceramic phase is added as a reinforcement to improve the properties such as strength, stiffness, and wear resistance. For ceramic matrix composites, metallic phase is added as a reinforcement primarily to induce toughness. Numerous processing techniques have been developed to produce these composites [1-6].

For the past couple of years, we are working on the development of interpenetrating phase alumina-Al composites using commercially available fused deposition modeling (FDM). The interpenetrating phase composites are also called the 3-3 composites where the matrix and the reinforcement phases are connected to itself in all the three directions. Researchers have already attempted to make 3-3 metal-ceramic composites using various processing techniques. Most of these techniques use a two-stage process where during the first stage, a ceramic preform is fabricated and then during the second stage, the preform is infiltrated using a molten metal. The final microstructure of the composite depends on type and structure of the ceramic preform. For the case of dense ceramic preforms, between 5-15% metal content in the final microstructure is most commonly reported. The total metal content depends on the amount of residual porosity in the starting ceramic preform. As the residual pore structure is random in terms of size and distribution, the metal distribution in these type of composites are random as well. For the case porous ceramic preforms, the control over the pore structure dictates the control over the final microstructure. In our approach, porous ceramic preforms were used to produce the interpenetrating phase metal-ceramic composites.

Processing of porous ceramic structures have been studied quite extensively due to their several applications. Porous ceramics have found several commercial applications including catalytic support, bone graft, and filters for molten metals. Most of these structures posses three dimensionally interconnected open cell porosity. The control over various porosity parameters such as pore size, pore shape and pore-pore interconnectivity is difficult to achieve using any one of the commercially available processes. Most of the processes produce structure with random three-dimensional porosity, except extrusion based processes in which controlled 2D honeycomb porous structures can be fabricated. In this work, controlled porosity three dimensionally honeycomb ceramics have been fabricated using the indirect fused deposition modeling (FDM). The FDM not only allow us to control the shape and size of the part, but also the internal microstructure and pore structure.

In this work, the indirect fused deposition process was utilized to produce controlled porosity ceramic structures as a preform for metal infiltration. Mullite ceramic is used for this work. In the indirect process, a polymeric mold is fabricated having the negative of the desired structure. The mold is then infiltrated with ceramic slurry or gel. The structure was then dried, and subjected to a binder removal and sintering cycle to form the sintered porous preform. The control over various porosity parameters can be achieved through CAD or changing various FDM build parameters. This method has been utilized to produce controlled porosity structures using other ceramic compositions as well. Controlled porosity mullite preforms were then infiltrated using Al metal to form the Al-alumina composites. After infiltration, pore were filled with Al and the metal assume the shape and the connectivity of the pores. In this method, by controlling the pore architecture, metal architecture and properties can be tailored. Four point bend flexural and uniaxial compression tests were carried out to understand the influence of different microstructural parameters on the mechanical properties of these novel composites.

Experimental Methods

Figure 1 shows a flow chart for the processing of these composites. The process starts with design of the composites using computer aided design (CAD) software. Once the design is complete, the file is saved in a standard *.stl* format. The part is then imported in to fused deposition modeling (FDM) software Quickslice™ to create the tool path for fabricating polymer prototype. A negative of the desired part is designed and built as a polymer mold. While the external shape and the size of the part are controlled via CAD, the internal architecture is varied by modifying FDM build parameters. The polymer molds are then infiltrated with water based mullite slurry. The structures are dried and subjected to a binder removal and sintering cycle in a muffle furnace to form the porous ceramic preform. During the initial heating stage, the mold polymer along with the binder leaves the part and creates the desired porosity. Details of the ceramic preform processing steps are described elsewhere for alumina ceramic [7].

In this work, -325 mesh mullite (CE Minerals, PA) powder was used to form the ceramic preform. Mullite powders were mixed with 1-butanol (Antifoaming agent, Fisher Scientific) and Darvan-821 (Dispersant, R. T. Vanderbilt, CT) and ball milled for one hour. The B-1000 (Rohm and Haas, PA) binder was then added to the slurry and milled for another five minutes before pouring into the polymer mold. For every batch of 20 gms of water, 85 gms of mullite powder, 1 gm of 1-butanol, 1 gm of Davan 821A and appropriate amount of binder (typically 12wt% of the powder) was added. After pouring into the mold, parts were dried for three to four days. Binder removal and sintering of the structures were carried out in two stages. During the first stage, parts were heated up to 1200 ^{o}C. In this stage, mold polymers left the system and porous mullite parts were partially densified. In the second stage, parts were heated up to 1600 ^{o}C and held for 3 hours. At the end of the two-stage sintering process, parts were densified up to ~94% theoretical density, leaving a 6% residual microporosity. The two-stage process helped us to avoid any warping in the ceramic preforms occurring during the densification process. The total binder removal and sintering cycle time was 36 hours for parts up to 25mm x 25mm x 25mm.

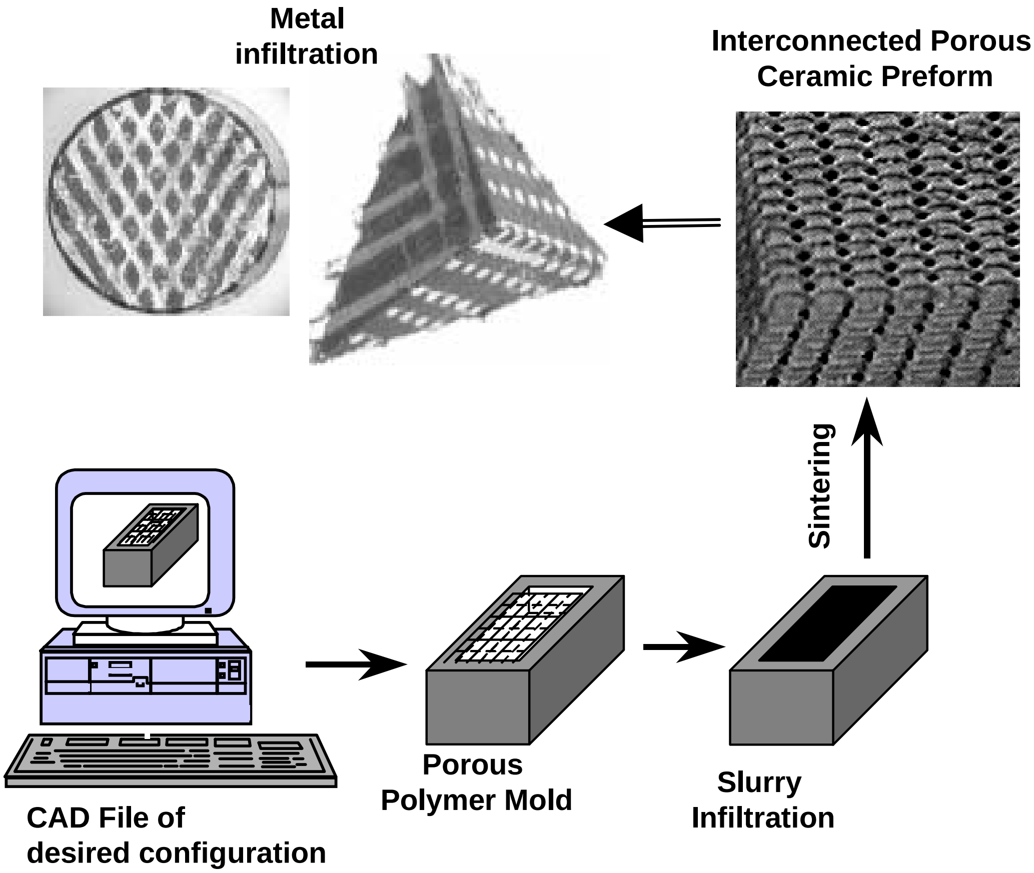

Figure 1: Process flow chart for the interpenetrating phase composites

Mullite ceramic preforms processed via the indirect FD have a continuous three dimensional network porosity. The polymeric mold design determines the size, distribution and the connectivity of the pores in these three-dimensional honeycomb structures. Using this approach to process porous structures allows the flexibility in design and fabrication of parts where both the macro as well as the micro-structures can be designed simultaneously to tailor properties. These controlled porosity ceramic structures were then infiltrated with Al metal to form metal-ceramic composites, where the controlled pore network becomes the interconnected metal network in the composite.

The indirect fused deposition process can also be utilized to fabricate non-uniform or functionally gradient porous ceramic structure where the size, shape and connectivity of porosity can be different at different height of the structure. This can easily be achieved due to layered manufacturing process, which is the inherent concept behind any solid freeform fabrication (SFF) or rapid prototyping (RP) techniques such as FDM. During mold making step, internal architecture for the mold at different height can be tailored differently to create a gradient porosity structure in the ceramic preform.

The metal infiltration process was carried out in one step. Al metal pieces were placed in an alumina crucible and heated in a muffle furnace up to 700 ^{o}C. Porous mullite preforms were heated up simultaneously at the same temperature inside the same furnace. At 700 ^{o}C, mullite ceramic preforms were placed inside the alumina crucible into the molten Al metal. The crucible is then heated up to different infiltration temperature from 850 to 1150 ^{o}C. At the infiltration temperature, the assembly was kept for one hour and then cooled back to 700 ^{o}C. Metal-infiltrated mullite preforms were taken out from the molten Al metal. During this infiltration process, liquid metal fills all the designed pores in the ceramic preform and forms a three dimensional network of metal in the ceramic. For this reason, these composites can also be referred to as interconnected or 3-3 metal ceramic composites where both metal as well as ceramic phases are connected to itself in all three directions.

One of the problems in the pressureless metal infiltration with Al is the oxide layer formation. Due to the Al_2O_3 layer formation at the exposed surface of the liquid metal, infiltration of liquid metal into the designed pores may be hindered or may not happen at all without an external pressure. It was observed that during the insertion of the ceramic preform into the molten metal at 700 ^{o}C, it is important to remove the oxide layer from the top of the molten metal and then place the sample into it.

Road width, road gap and slice thickness were three FDM build parameters that were varied to create different types of porosity. By changing those three parameters, structures having different volume fraction metals, with various road width and road heights were fabricated. Once fabricated, four point bend samples were machined and tested using an Instron screw driven closed loop machine under stroke control mode. Preliminary uniaxial compression tests were also carried out with some of these samples. Results are compared to understand the influence of various processing parameters on the mechanical properties of these composites.

Results and Discussion

Using the FDM build parameters, road width, road gap and slice thickness, composites microstructures were varied keeping the external geometry constant. Due to the variation of road width, the metal thickness was changed, road gap changed the ceramic width and slice thickness changed the metal road height. Using these three parameters, it was possible to design composites having different volume fraction metal content with same metal road width and thickness or composites having the same volume fraction of metal content with different road width and thickness. This capability allowed us to study the influence of the meso-scale structure parameters on the physical, mechanical and thermal properties of these composites.

Figure 2 shows influence of ceramic width on the flexural strength of the alumina-Al composites. Only the road gap was varied to vary the ceramic width. In these structures metal

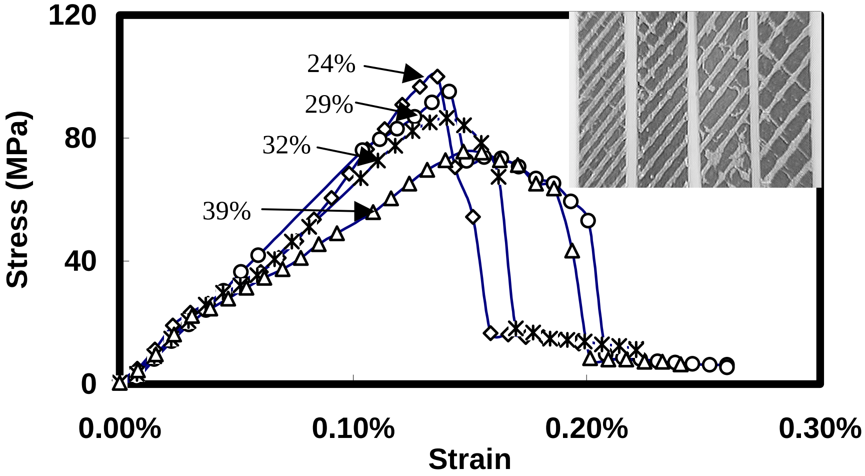

Figure 2: Variation of flexural strengths as a function of ceramic width

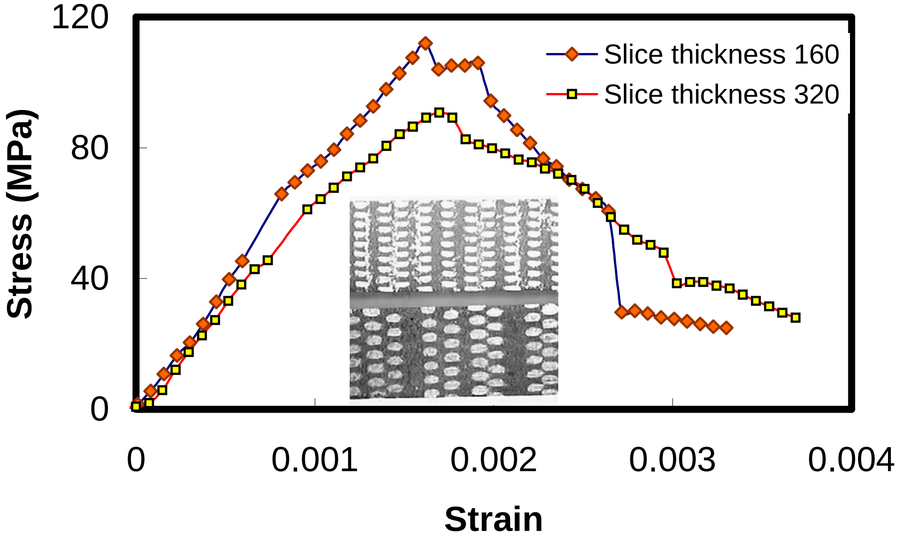

Figure 3: Variation of flexural strength as a function of metal height

width and height were kept constant at 300 and 250 microns, respectively. As the ceramic width increased, volume fraction metal content decreased and the composites became more brittle. The result is obvious from the plots in which samples with 24% metal content showed an abrupt failure after reaching the maximum failure stress, whereas samples with higher metal content, such as 32% and 39%, showed a gradual failure with a higher strain to failure. Moreover, failure strength and stiffness decreased with increasing metal content. In general, it can be seen that increasing metal content by lowering the ceramic width increases the toughness in the composites, but lowers the failure strength. Figure 3 shows the influence of slice thickness. Reduced slice thickness decreased the metal height, while keeping the ceramic and metal width

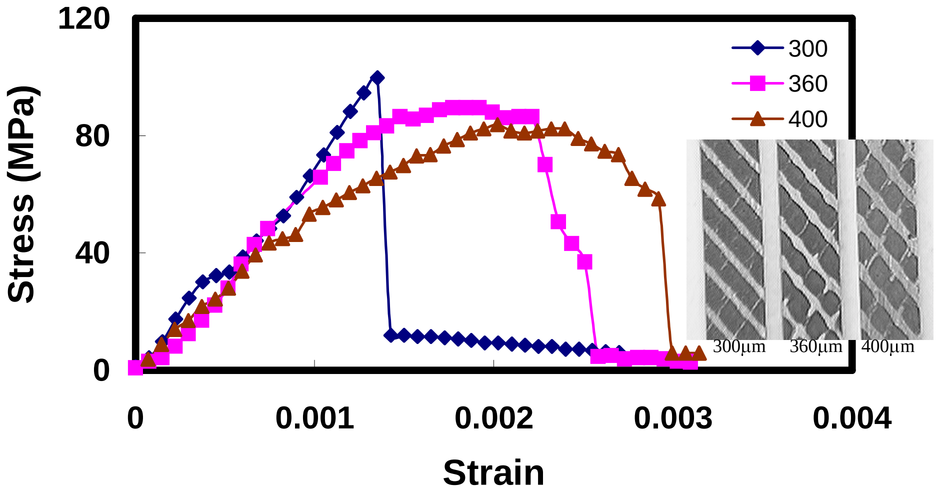

Figure 4: Variation of flexural strengths due to the change of metal thickness

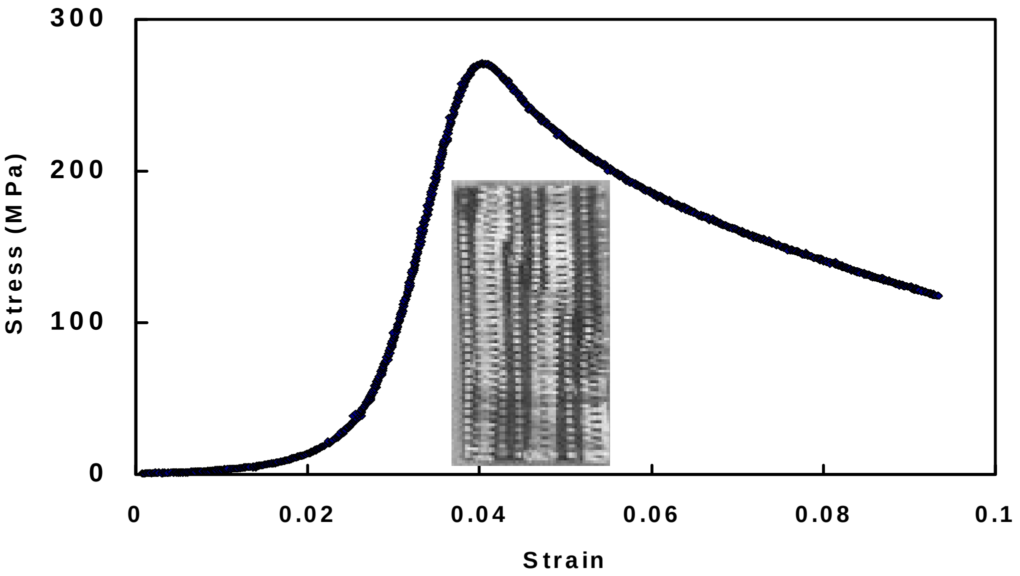

Figure 5: Cross-section image of a compression sample and a typical compression test curve

constant. It can be observed that by changing the slice thickness, the shape of the curve does not change, though failure strength values does. Lower slice thickness produces composites with higher failure strength. It is believed that finer microstructures help to increase the mechanical properties in these composites. Figure 4 shows the influence of metal width in which metal height (250 micron) and ceramic width (750 micron) were kept constant. As the metal width increased, the metal content in the composite increased as well, resulting a higher toughness and failure strain with lower strength and stiffness.

Preliminary compression tests were carried out with some of these samples. Figure 5 shows a cross-section image of a compression test sample and a typical plot of a stress-strain curve. The

Orientation		α Heating $x10^{-6}$	α Cooling $x10^{-6}$	Average α $x10^{-6}$
+/-30		4.65	8.48	6.57
+/-45		4.06	5.6	4.83
+/-60		5.02	6.02	5.52
+/-75		4.6	4.75	4.68
0-90		4.04	4.01	4.03

Table 1: Variation of the CTE as a function of metal road orientation

strength varied between 200 to 480MPa for composites having metal content between 24 to 39%. As usual, increasing metal content decreased the strength and stiffness of the composites. All the tests were carried out with samples in which metal content was varied by varying the ceramic width, keeping the metal height and width constant. The cross-sectional image showed that the ceramic had fractured completely, though the sample remained in one piece due to the interconnected metal network in the composites.

The measurement of the coefficient of thermal expansion of these composites was carried as a function of the orientation of the metal road in the composite. In these cases, road width, road gap and slice thickness were kept constant, but the orientation of the polymer roads were varied. The composite structure showed different metal orientation due the change in orientation in the polymeric molds. In general, there was no significant change in the CTE values due to the change in orientation. The +/-30^{o} composite showed a hysteresis in which significantly different values during heating and cooling were measured. This may be due to some experimental error. All other values remain in the range of 4-6 ppm for samples containing 32% metals. The CTE values increased as metal content increased in the composites.

Summary

Alumina-Al composites were processed via reactive metal infiltration of controlled porosity mullite preforms. The mullite preforms were fabricated using the indirect fused deposition process. The porous preforms had three-dimensionally interconnected porosity that was filled with Al during metal infiltration. The resulting structure had an interconnected metal and ceramic phases in all three directions. Metal width, metal height and ceramic width were varied in the composites to understand their influence on the mechanical properties. In general, increasing metals content due to increasing metal width or decreasing ceramic width increased failure strain and lowered the strength and the stiffness of the composites. Decreasing metal height increased the failure strength of the composites. The CTE of the composites did not change significantly due to the variation of the orientation of the metal roads in the composites.

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