A NEW SFF PROCESS FOR FUNCTIONAL CERAMIC COMPONENTS

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ABSTRACT

SRI International in collaboration with Saint-Gobain/Norton Industrial Ceramics Corporation and Allison Engine Company is developing a new SFF process, Direct Photo Shaping, for the fabrication of functional ceramic components. The process is based on the layer-by-layer photocuring of ceramic slurries containing monomers curable by visible light. Each layer is photoimaged by a liquid crystal display (LCD) or a digital light processing (DLP) projection system. After binder removal and sintering, the properties of silicon nitride tiles prepared according to the Direct Photo Shaping process were found to be comparable to properties of tiles formed by conventional methods.

INTRODUCTION

Rapid prototyping is presently being applied to the fabrication of nonstructural materials by means of computer-aided design/computer-aided manufacturing (CAD/CAM) technology, wherein computer files descriptive of the object are used to create parts made from materials such as UV-curable polymers. The components fabricated in this fashion are considered nonfunctional, and their main application is for iterative design evaluation in molding making. There is an increasing demand to extend rapid prototyping technology to the fabrication of functional components with engineering properties and dimensional tolerances comparable to those of conventionally produced components.

The development of solid freeform fabrication (SFF) manufacturing technology is expected to allow the fabrication of functional prototypes from advanced ceramic, metallic, and multiphase materials for structural and electronic applications. The SFF approach to net shape forming holds great promise for rapid prototyping of ceramic components through simplification of the processing cycle (e.g., elimination of the time consuming steps of pattern making and mold fabrication).

In this paper we describe a new SFF process, Direct Photo Shaping, and its use for the fabrication of silicon nitride components.

DIRECT PHOTO SHAPING PROCESS

The Direct Photo Shaping process is described in Figure 1. A liquid crystal display (LCD) or a digital light processing (DPL) projection system are used as an electronic mask to build lithographic images onto a photopolymerizable slurry containing ceramic powder. A visible light source is coupled with the projection system. The image to be projected is changed according to the CAD model information. The photopolymerizable slurry contains the ceramic powder, a dispersant, photocurable monomers, and a visible light initiator. The slurry is dispensed onto the build platform and leveled with a doctor blade. Each film is photoexposed by visible light projection. After each exposure, a new film of photopolymerizable dispersion is applied on the build platform. When the fabrication is complete, the green ceramic body is removed from the platform and rinsed with a suitable solvent to dislodge any uncured material.

The ceramic forming process is based on photo-gelcasting, since the green body is formed by photocuring of a slurry of the ceramic powder in a solution of photopolymerizable monomers. Gelcasting [1] has been shown to produce complex-shaped, near-net-shape parts. Gelcast green bodies are generally strong and machinable, and after sintering, highly dense ceramic parts are obtained.

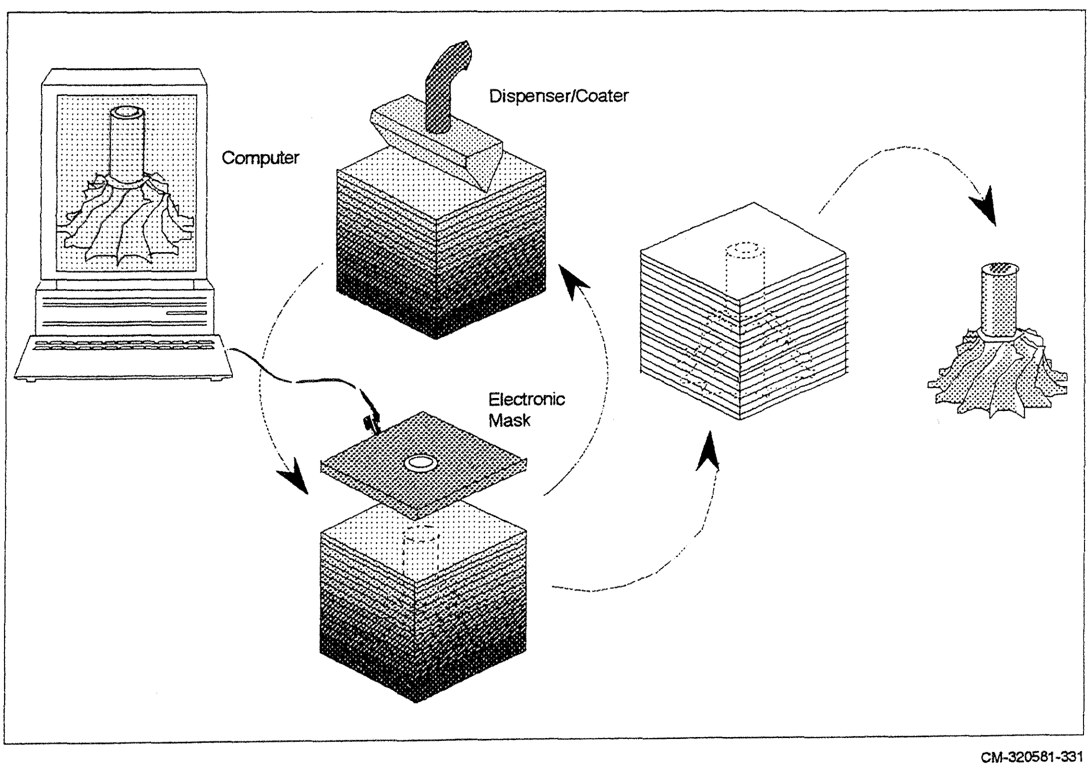

Figure 1. Schematic illustration of the Direct Photo Shaping process.

EXPERIMENTAL

Powder Production

Silicon nitride powder (Ube Industries, Yamaguchi, Japan) and 4 wt% Y_2O_3 sintering aid were coprocessed by aqueous milling to reduce the particle size and to homogeneously distribute the sintering aid. Milled slurries were passed through a magnetic separator to remove iron contamination and were then filtered and concentrated. The concentrated slurries were dried by freeze drying. Details of the powder processing have been previously described [2].

Ceramic slurry

Ceramic slurries were prepared by ball-milling the silicon nitride powder in the photocurable monomers, after addition of a suitable dispersant, a solvent/plasticizer and the photoinitiator. The viscosity of the ceramic slurries was determined with a Brookfield DV-II+ viscometer.

Component Fabrication

Tiles (3"x3"x0.5") were formed according to three different methods to compare their mechanical properties. Tiles were prepared by multilayer deposition and photoexposure to a quartz tungsten-halogen lamp. The photocurable slurry comprised of 68 wt% silicon nitride, a suitable dispersant, photocurable monomers, plasticizer, and a visible light photoinitiator. Each layer applied was 2 mil thick. Additionally, tiles were formed by either uniaxially pressing the dry powder followed by isostatic pressing (30 ksi) or by thermally curing (120°C) monolithic tiles formed from a slurry of the same composition as used for the SFF process.

Other complex shape samples were prepared by direct photoexposure using a 1000 watt xenon arc lamp from Oriel Corporation, Stratford, CT with a filter that allows only the transmission of radiation from 420 nm to 630 nm, and reduces thermal effects.

Binder solvent was removed from the photocured samples by heating in air to 450°C over 48 hours and then heating to 650°C. Samples were presintered to improve their handling strength after the binder removal and were then densified by HIPing using glass encapsulation.

Sample Characterization

Samples for microstructural evaluation were prepared by polishing fracture surfaces and then plasma etching using a 95% CF_4/5% O_2 carrier gas [3]. This etching technique removes the Si_3N_4 and leaves the intergranular phases. Sintered densities were measured in deionized water using the Archimedes method.

Specimens for flexure testing were machined either to 1.5 mm x 2 mm x 25 mm or 3 mm x 4 mm x 50 mm size. Specimens were cut both parallel and perpendicular to the deposition direction. In both cases the lamination direction was parallel to the long dimension. Flexure strength was measured by four-point bending in accordance with ASTM method C1161. Hardness was measured with a Vickers diamond indenter at a load of 0.5 kg. Fracture toughness was measured by both the indentation method and the controlled flaw method.

RESULTS

By pretreating the silicon nitride powder with a suitable dispersant, we were able to prepare slurries containing silicon nitride up to 55 vol%. These slurries were photocured as thin strong films. The typical working thickness of each layer used in our multilayer deposition process was about 2 to 4 mil. The slurry composition was optimized to improve interlayer adhesion and eliminate any possible delamination during debinderization. This was achieved by reducing the layer stiffness and using additives to produce a tacky top layer. This optimization allowed us to prepare freeformed tiles, that after debinderization and densification, were found to have mechanical properties comparable with that of silicon nitride pressed samples.

After binder removal, the samples showed no blistering or discoloration and tiles prepared with the optimized slurry composition showed minimal delamination. Carbon analysis showed no residual carbon. After HIPing the samples were >98% dense, as shown in Table 1, and no evidence of the original layers was observed by SEM examination of cross-sectional surfaces of the SFF tile. Figure 2 shows representative etched surfaces for a sample prepared by the SFF method and for a tile formed by dry pressing. Both samples show a pronounced bimodal distribution of grain sizes and as is typical of *in situ* reinforced Si_3N_4 show a pronounced acicular microstructure.

Figure 3 shows two representative silicon nitride green parts, SRI and DARPA logos, that were initially selected to demonstrate the feasibility of Direct Photo Shaping to build multilayers with variable cross-section and contours.

Mechanical Properties

Table 1 summarizes the mechanical property data for tiles formed by dry pressing, thermal gel casting a monolithic tile and layered tiles formed by the solid free form method. The three forming techniques were used to assess the effect of the forming technique on the mechanical properties and to verify that the solvent and polymers could be removed without deleteriously affecting the densification and the mechanical properties. In many cases, the gelled monolithic tiles could be processed to near full density and the mechanical properties were comparable to those measured for tiles formed by dry pressing. This indicated that the organic solvents/polymers could be removed without affecting the mechanical properties of the Si_3N_4. In some cases the monolithic samples showed lower densities and strengths than the dry pressed and layered samples; this was attributed to slight differences in the HIPing cycles which were used for these samples. The sample formed by the solid free form technique showed similar properties to the dry pressed samples with MOR values slightly in excess of 1 GPa. The flexure strengths for the layered tiles were not dependent on the orientation relative to the casting direction. Fracture toughness measured by Vickers indentation was 4.0 MPa $m^{1/2}$ for the dry pressed samples and 4.5 MPa $m^{1/2}$ for the solid freeform sample. Fracture toughness as measured by controlled indentation was slightly higher, as is typically observed, and was comparable to values previously reported on optimized slip cast material of the same composition [1].

Table 1

SUMMARY OF MECHANICAL PROPERTY DATA FOR TILES FORMED BY UNIAXIAL PRESSING, THERMAL GELATION AND LAYER-BY-LAYER FABRICATION METHOD

		Density	Hardness	K_{IC}(MPa $\sqrt{m}$)		MOR (MPa)		Weibull parameters	
Sample	Forming	(g/cc)	(GPa)	Indent	Indent Fracture	3x4x50 mm	1.5x2x 25 mm	beta	n
C012	Press	3.22	17.0	4.0	5.9	844	937	23	30
L08	Press	3.22	15.5	4.0	6.1	832	976	24	30
L02	Gelled	3.22	NA	NA	5.2	839	NA	NA	NA
L08	Gelled	3.10	15.8	3.8	4.1	580	779	13	13
#202a	SFF	3.21	16.5	4.5	NA	NA	1018	16	15
#202b	SFF	NA	NA	NA	NA	NA	1052	25	15

Note: Specimens for 202a were cut so that the casting planes were parallel to specimen faces; specimens for 202b were cut so that the casting planes were perpendicular to the specimen faces. In both cases the lamination direction was parallel to the long dimension of the specimen.

CONCLUSIONS

We have described a new SFF process, Direct Photo Shaping, based on the additive fabrication of layers photoimaged by a liquid crystal display (LCD) or a digital light processing (DLP) projection system. By optimizing the slurry composition we fabricated layers that adhered to each other and showed minimal delamination after removing the binder. Mechanical properties of silicon nitride tiles prepared according to the Direct Photo Shaping process were comparable to properties of tiles formed by dry pressing and were slightly in excess of 1 GPa.

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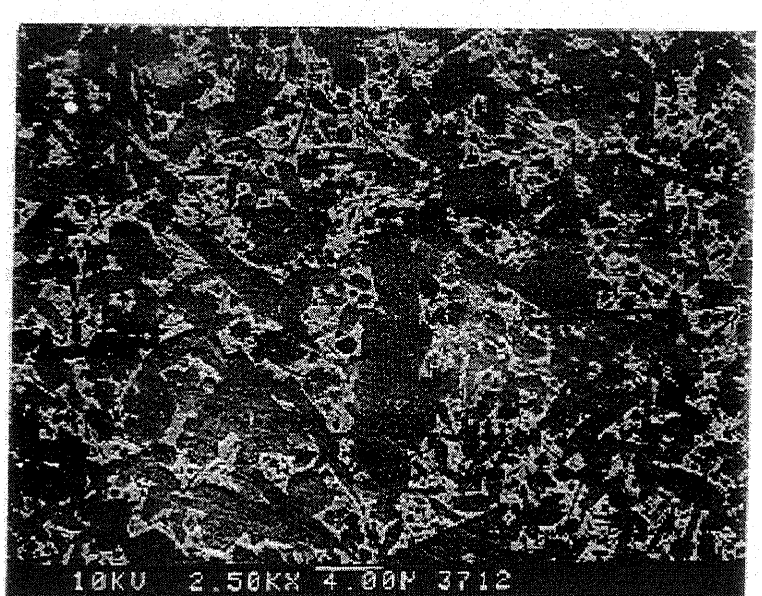
(a)



(b)

Figure 2. Representative microstructures of (a) a sample prepared according to Direct Photo Shaping process and (b) a sample prepared by uniaxially pressing.

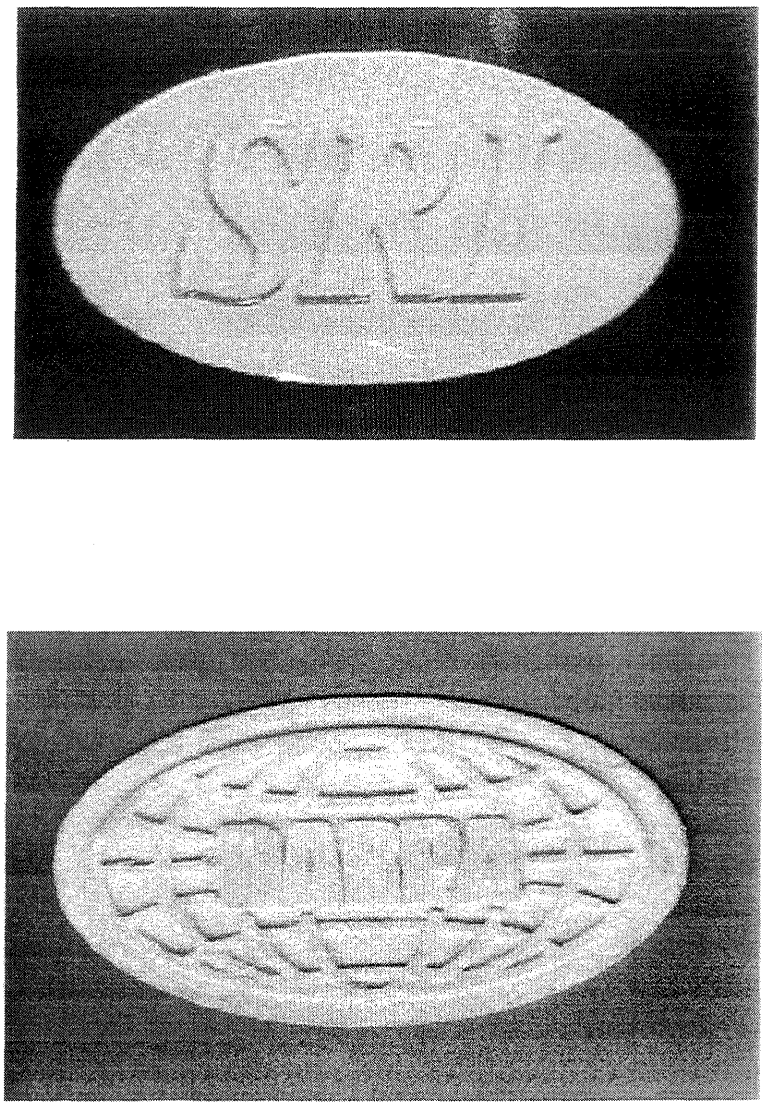


Figure 3. Representative green silicon nitride parts built by Direct Photo Shaping.