Al_2O_3 Ceramics Made by CAM-LEM (Computer-Aided Manufacturing of Laminated Engineering Materials) Technology

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Abstract

The concept of CAM-LEM technology is presented and discussed in the context of the fabrication of Al_2O_3 ceramics. Particular attention is paid to the interplay of green tape characteristics and the unit operations involved in CAM-LEM. Examples of ceramic shapes difficult to form by conventional methods are described.

Introduction

Ceramic materials offer a wide range of attractive engineering properties. For example: their thermal conductivity can be very low (e.g., vitreous SiO_2) or very high (e.g., AlN); some are excellent electrical insulators (e.g., Al_2O_3), others are semiconductors (e.g., SiC) or show metallic conductivity (e.g., TiB_2). Structurally, many show good corrosion resistance, and, increasingly, ceramics are being developed with high fracture toughness and strength (e.g., partially-stabilized ZrO_2 and Si_3N_4). In particular, the properties of advanced ceramics have been markedly improved in the last decade as the result of intensive research into the relationships between powder synthesis, processing and properties [1].

Nevertheless, shape forming remains a critical issue that is always challenging. Conventional ceramic forming methods are either of limited precision or require a prohibitive investment in tooling, or both. Diamond machining can produce many shapes, but is expensive [1] and generally compromises mechanical properties [1,2]. It is also true that many desirable shapes cannot be fabricated using conventional methods.

CAM-LEM technology is a novel Solid Freeform Fabrication (SFF) method developed to allow direct fabrication of components of nearly arbitrary complexity using engineering ceramics of arbitrary composition without the need for tooling. The roots of the CAM-LEM approach lie in the fabrication of multi-layer ceramic substrates for microelectronic packaging [3]. In such substrates, complex internal wiring is created by stacking thin sheets of ceramic powder distributed in a porous polymer matrix (so called "green tape") that have been punched and screen printed with powdered metal inks. The processing of such substrates, while representing some of the most advanced ceramic processing, is geared toward mass production of a single

design. CAM-LEM is a true SFF technique capable of producing arbitrary one-of-a-kind components. (While attention is restricted to ceramics in this paper, we are currently extending the technology to the production of both metallic and engineering plastic components.)

The CAM-LEM Process for Engineering Ceramics

The feedstock for the CAM-LEM process included two types of sheet material: green ceramic tape and a fugitive tape. Green ceramic tape is widely available, as it is the basis of a multi-billion dollar microelectronic packaging business. As green tape, the properties are dominated by the polymer matrix; the ceramic powder acts, for the most part, as an inert filler. This allows a wide variety of ceramics to be formed using one of several tape forming processes (tape casting, roll compaction, etc.). Fugitive tape is manufactured to exhibit viscoelastic properties that are similar to those of the green ceramic tape, but is completely burned out of the 3D structure during firing.

The CAM-LEM system generates a green ceramic component that must be post-processed and sintered to form a dense monolith. The details of the machine design will be presented elsewhere [4]. One distinguishing feature of CAM-LEM technology is that a "cut then stack" approach is used. This allows sections cut from multiple materials to be used within a given layer of the stack, if desired. It also allows pieces of complex internal geometry to be fabricated, as shown in Fig 1.
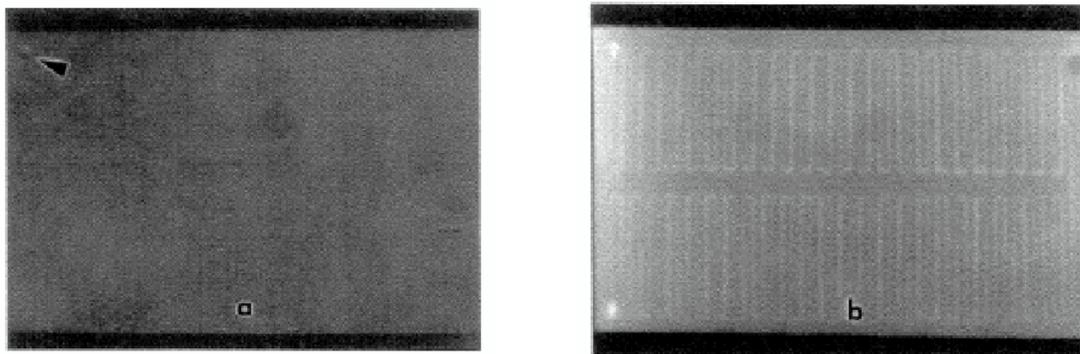

Figure 1. Three layer part fabricated using CAM-LEM technology; a) is a room lit view and b) is a back lit view. The piece is designed as a flow channel with an inlet in the top left corner of the upper surface (visible in a) connected by a complex flow channel in the central sheet (visible in b) to an outlet in the bottom left corner of lower sheet.

The central steps in the process associated with fabrication of a complex part are schematically illustrated in Fig. 2. There are two critical components in the apparatus (which is discussed elsewhere in these Proceedings [4], a cutting station and a stacking platform. An arbitrary component is built using the following steps: 1) a sheet of green ceramic tape is picked up robotically and placed on the cutting station; 2) the desired outline is cut using a CO_2 laser; 3) waste is selectively removed and the cut piece is transferred robotically onto the stacking table; 4) a sheet of fugitive tape is similarly processed to yield a complementary piece (either inside, outside, or both) and the fugitive is robotically added to the stack and its waste removed. The cycle is repeated until the 3-D object is finished, complete with the fugitive supports.
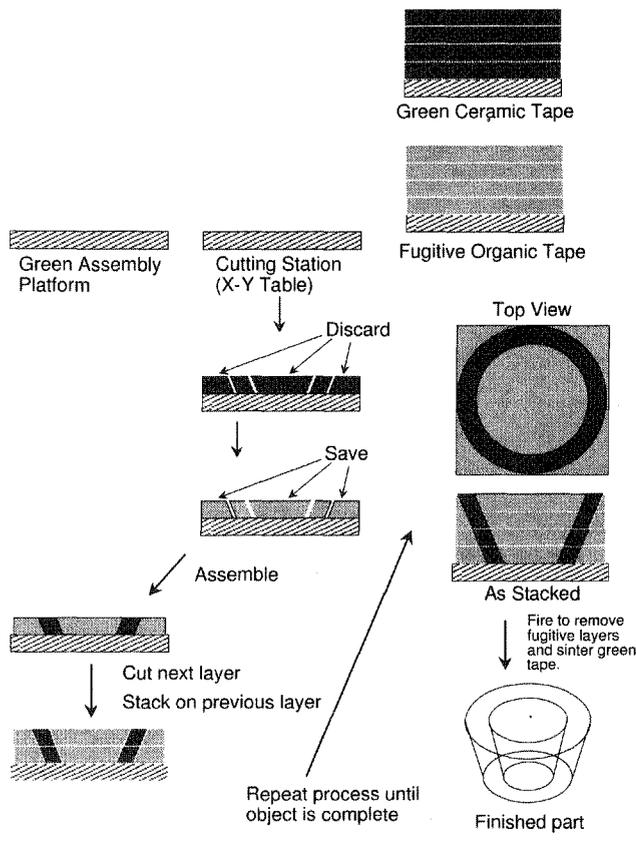
Figure 2. Schematic outline of the CAM-LEM process building a green ceramic component surrounded by, and cored with, a fugitive tape that burns out during firing.

The finished part is 5) laminated to fuse the individual layers, 6) heated to burn out both the fugitive and the binder in the green tape and finally, 7) the component is sintered to complete densification.

The critical issues for fabricating ceramic components using CAM-LEM technology are: i) suitability of the tape for laser cutting; ii) development of an efficient method for "tacking" during assembly (necessary to preserve registration during handling); iii) post-assembly lamination to create a monolithic green part; and iv) preservation of dimensions during sintering. Our observations are consistent with recent reports describing the effect of the organic phase on the physical and mechanical properties of green tape [5], and that flaws in and between tapes can be healed during thermocompressive lamination [6].

Experimental Results

Materials

Two types of green ceramic tape were employed in our experiments, a commercial 94% Al_2O_3 tape (Coors Electronic Materials, Chattanooga TN) and an in-house formulation based on A16-SG alumina (Alcoa, Pittsburgh PA). The Coors tape was cast from a nonaqueous slurry with polyvinyl butyrate as the primary binder. The in-house tape was cast from an aqueous slurry with a combination of acrylic emulsion binders (Rohm and Haas, Springhouse, PA); in some cases, polypropylene was added as a plasticizer. The compositions of several in-house tapes are given in Table 1.

The principal difference between the two types of green ceramic tape was their respective glass transition temperatures. These were determined by a dynamic mechanical analyzer to be ~ 40°C for the Coors tape and -20°C for the in-house tape.
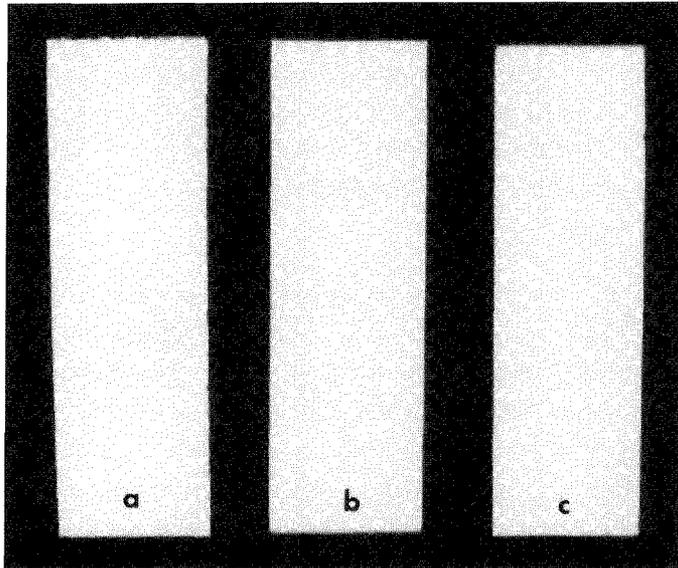
11

Figure 3. Green tapes used in experimental trials; a) Coors 94% Al_2O_3 tape, b) In-house Al_2O_3 tape, and c) fugitive tape based on corn starch.

In addition, a novel fugitive tape was developed based on the use of corn starch as a filler. Chemical grade corn starch (Sigma Chemicals, St. Louis, Mo.) has a reported loss on ignition (or ash content) of <0.4%; thus it can be expected to burn cleanly from the component during firing. Aqueous-based slurries were initially employed, but the dried tape adhered strongly to the mylar, presumably because the starch is slightly soluble in water and served as a glue during drying. Nonaqueous slurries using toluene and polyvinyl butyrate released easily from the mylar. Samples of the three tapes are shown in Fig. 3.

Table 1: Al_2O_3 Slurry Formulations Used in Tape Casting Trials

Component (g)	Baseline	AS-3	AS-3-3	AS-4	AS-7
Al_2O_3	100.00	100.00	100.00	100.00	100.00
DI-water*	21.00	21.00	21.00	21.00	21.00
D-05**	0.86	1.15	1.15	1.15	1.15
B-1000***	10.91	10.91	10.91	-	7.28
B-1035***	10.91	10.91	10.91	21.82	3.7
PPG****	-	-	4	-	3.7

*Deionized water
**Dispersant (Rohm and Haas)
***Acrylic Emulsion Binders (Rohm and Haas)
****Polypropylene Glycol

Laser Cutting

The effectiveness of laser cutting is a complex function of the nature of the surface supporting the green tape and the composition of the green tape (both the type of polymer and the composition of the ceramic).

The Coors tape was not designed for laser cutting and proved a challenge. Two problems were encountered. Firstly, detritus that recondensed after the laser was employed for cutting caused undesirable adhesion of the cutout piece to the cutting station platen. Secondly, when cutting thick tape (e.g., nominally 24 mil, or 600 μm), a significant taper was observed. Both problems were reduced in severity through a redesign of the cutting station platen, [4] and components could then be automatically fabricated using this tape.

The in-house tape proved to be better suited for laser cutting. Adherence to the platen does not occur and edge taper is much less severe. For these reasons, development of the in-house formulation continues. (This tape also shows promise in its ease of lamination.) However, the fabrication experiments reported here concentrate on results obtained with the Coors tape.

Tacking

The glass transition temperature of the Coors tape is 40°C and the recommended lamination temperature for this tape is 80°C. The use of thermal tacking during build would require complex thermal management and such high temperatures might significantly soften the entire assembly, to the point where gross flow is likely. Instead a solvent-based approach was employed.

The principle of solvent lamination is to locally and temporarily dissolve the polymer on the surfaces to be mated, so that polymer chains in adjacent tapes become entwined. With porous tapes, the solvent is redistributed by capillary action, but in all cases it is ultimately evaporated and the polymer returns to the solid state.

Often the solvent used is the same as that employed in the original slurry. In our experiments, however, ethanol (rather than toluene) was most effective. Ethanol wets and spreads rapidly on the Coors tape, so that application using a dropper was entirely adequate.

The solvent was applied to the top surface of the existing stack prior to the placement of each cutout. Timing and quantity of solvent proved important, as excessive solvent can migrate into the cutout, causing adherence to the robot, or insufficient solvent will not be present to soften the surface of the cutout. With correct timing, tall stacks (e.g., 40 layers) were easily produced. This tacking procedure produced robust pieces, and several components were directly fired after tacking and yielded sound ceramics. Nevertheless, for strength critical parts, it is desirable to post-process to assure complete lamination efficiency.

Lamination

Isostatic pressing is widely employed in ceramic forming. Typically, the part is placed in an impermeable bag, which in then evacuated, sealed, and submerged in a hydraulic pressure vessel. Often the fluid is at room temperature (so called cold isostatic pressing), but it can be heated (warm isostatic pressing).

Experiments using a commercial warm isostatic press (National Forge Co., Andover, Ma.) to fabricate bend specimens confirmed that lamination efficiency is easily achieved.

Isostatic pressing can be very cumbersome if the piece is delicate or of complex shape. One important role for the fugitive tape is to assist in pressure transmittal to complex components during isostatic pressing; absent the fugitive component, isostatic pressing might lead to deformation of delicate portions of complex structures.

Firing

The two issues arising during firing are the ease of binder removal and the extent to which shrinkage can be controlled. Standard firing schedules were used to successfully remove the binder. No special procedures were required.

Shrinkage issues are comparable to those encountered using powder compacts derived from conventional ceramic forming operations. For example, a part of relatively complex geometry was cut from a single green tape; after firing, the relative dimensions were preserved to 1%, see Fig 4.
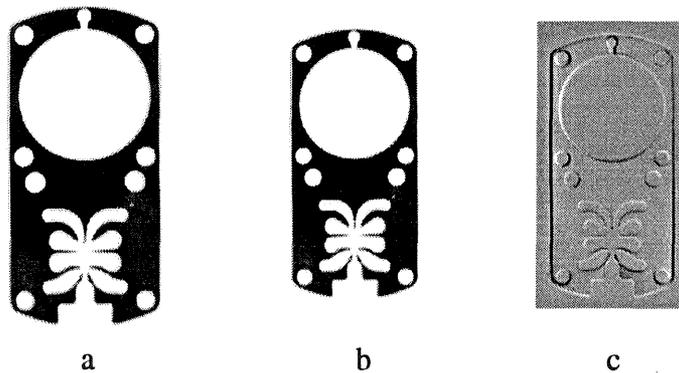

a   b   c

Figure 4. Digital image of a) a green part cut from a single sheet of green tape and b) the fired piece. The firing shrinkage was nominally 17%. Image c is a difference image obtain by subtracting the green image scaled to 83% from the image of the fired piece. (Black and white shading reveal differences in the two images.) The degree of registration is overall very good, less than 1% variation being independently measured for the width as a function of position along the length. The small anisotropy (1%) in shrinkage is also visible; note the white fringes lie outside left-to-right, but are inside up-and-down.

Test Component

As a test of the CAM-LEM method, the software and machine design, as well as the compatibility of CAM-LEM technology to ceramics, a virtual part was rendered in the 94% Al_2O_3 tape. The test piece was a CAD file taken from the AutoCAD (AutoDESK, Sausalito CA) tutorial. The CAD file was translated to STL and computationally sliced. Sheets of Coors tape were then laser-cut and robotically stacked using ethanol tacking, yielding the green compact

shown in Fig. 5a. This green compact was fired using the following schedule: 5 hour ramp to 600°C, 4 hour at 600°, 3 hour ramp to 1560°C, 0.5 hours hold at 1560°C, and power off cool. The fired piece is shown in Fig. 5b.
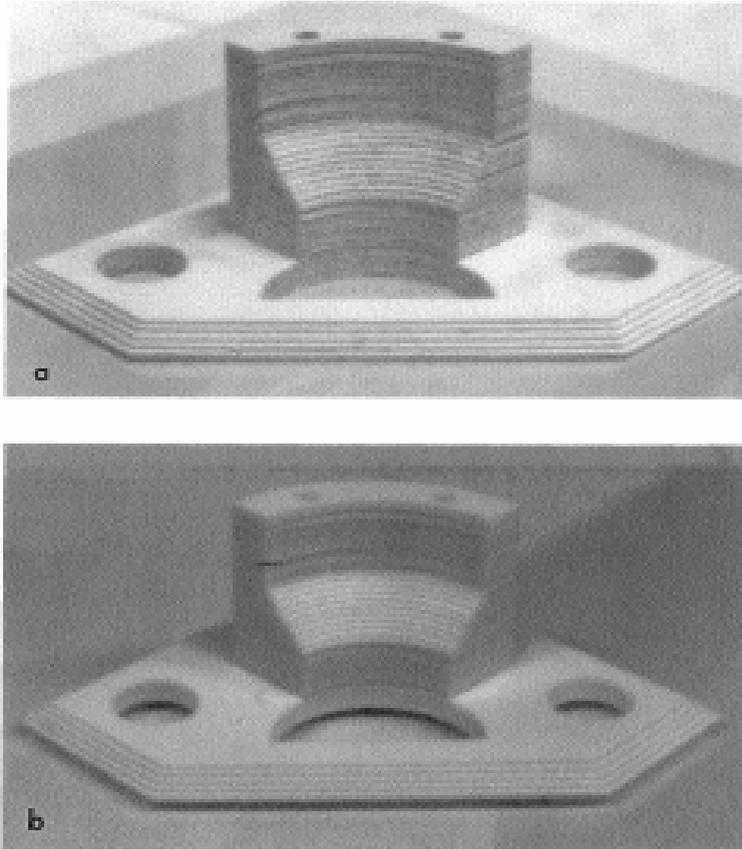

Figure 5. Component derived from an AutoCAD file. Image a) was taken of the green assembly after tack lamination and image b) was taken after firing. The edge length of the base on the fired piece is nominally two inches.

Summary

CAM-LEM, a novel SFF technology optimized for the direct fabrication of components from engineered ceramics, has been conceptualized, developed, and demonstrated.

Acknowledgements

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