Materials Issues in Laminated Object Manufacturing of Powder-Based Systems

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Abstract

Laminated object manufacturing of fine ceramic and powder metallurgy components can be carried out using either the cut-then-stack or stack-then-cut motif. With either approach, it is necessary to effect laser cutting to define layer geometry and lamination to fuse the stack into a seamless monolith. The relationship between the microstructure of the green tape used as feedstock on the relative ease of each process is reviewed.

Introduction

In general, the production of components from either ceramic or metal powders requires shaping and densification to be carried out in separate unit operations. That is, first a green powder compact is formed that has the desired shape; subsequently, the compact is fired to cause densification. This is no less true when freeform fabrication is used. The shrinkage that occurs during densification is anticipated and designed into the green shaping process so that the final fired part is true to the desired dimensions. Difficulty in meeting dimensional specifications is the result of part shrinkage that deviates from expectation. Such deviations can be either systematic or random. In both cases, the source of nonuniform shrinkage is ultimately inhomogeneous packing of particles; areas of local low-density particle packing tend to shrink more than regions associated with high packing.

Laminated Object Manufacturing

There are two motifs for producing green powder compacts via laminated object manufacturing; the Helisys process, which is a stack-then-cut process, and the CAM-LEM process, which employs the cut-then-stack approach. Regardless of which is employed, the two key unit operations are: laser cutting to define the geometry of the outline corresponding to each layer and lamination to produce a powder compact that is compatible with subsequent binder burnout and sintering without delamination.

Material Systems

The material systems are summarized in Table 1. For CAM-LEM, tape has been produced two ways; tape casting and compression molding. Tape-cast materials are characterized by a relatively high volume fraction of porosity (as high as 20 vol. %) whereas compression molded sheets are essentially pore-free. Another important

distinction is that the polymers used in tape-casting (polyvinyl butyral-based) are precipitated from solution during casting and are amorphous, whereas the compression molded sheets used in this work are based on the BASF commercial injection molding feedstock that employ a highly crystalline polyacetal-based binder, which is melted and resolidified during forming.

These two systems have proven very flexible as carriers for a variety of inorganic powders. Tapes have been produced that yield: alumina, zirconia-toughened alumina, partially stabilized zirconia, silicon nitride, stainless steel, and iron-nickel alloys.

Laser Cutting

Laser cutting in LOM processes typically employs a low-power carbon dioxide laser that couples strongly to the polymeric binder, which is melted and removed from the kerf through the action of a gas jet that is coaxial to laser. In the cutting of arbitrary outlines, it is necessary for the average cutting speed to be low (to avoid inertial effects on the cutting trajectory) and to have the speed vary, for example, to slow down when going around corners. Even if the same amount of energy in deposited per unit line length, low-speed/low-power cutting differs from the standard industrial process of high-power/high-speed because lateral heat conduction plays a larger role. In fact, experience has shown that controlling heat conduction away from the kerf region is the key to producing cuts that have the desired narrow and smooth kerfs. Machine variables are important, but it appears that a great deal of control can be effected by tailoring tape composition and microstructure.

An example of a deleterious effect of heat conduction is shown in Fig. 1a & b. This is a kerf cut in a commercial green tape that is produced using a combination of a polyvinyl chloride binder and highly volatile plasticizer. The presence of the chlorine ion, with its large radius, causes the structure to be open and mass transport is relatively easy. Therefore the plasticizer is "boiled out" of the tape on a time scale that is rapid compared to the cutting. The result is that the material immediately adjacent to the kerf is embrittled and it breaks away under the gas jet. This gives the very wide kerf and ragged edge seen in Fig. 1b. Tapes that employ polyvinyl butyral and low volatility plasticizers do not show this effect.

Given that a binder system with favorable properties is chosen, cutting characteristics can be further improved by controlling the amount of binder in the tape. Shown in Fig. 2 are scanning electron microscopy images of top and bottom of cut tapes that are similar in all respects except in the relative amounts of binder. The lower binder content corresponds to a matching increase in porosity. In both cases, the kerf is narrow and straight, but the lower binder content gives a distinctly smoother cut. Using optimized binder content also can minimize the amount of kerf taper, see Fig. 3. In general, increasing porosity of green tape can be advantageous with respect to laser cutting. This is attributed, at least in part, to the decrease in thermal conductivity of the tape. Further evidence of the importance of binder can be seen by comparing the results of laser cutting of sheet stock based on the BASF binder to those observed with the green alumina tape. For comparable cutting conditions, e.g., velocity of 19.1 mm/s (0.75 in/s) and laser power of 10 W, the kerf width measured on the top surface is 160 μ m for the alumina feedstock versus 350 μ m for the steel feedstock. Furthermore, the taper angle of the kerf is roughly 7-8° for the alumina, but nearly 21° for the steel. Clearly, the high thermal conductivity of the dense crystalline polymer matrix creates a less favorable set of conditions for cutting.

Machine variables also play an important role. The key dimension group for lateral heat conduction away from a moving line source is the Peclet number, $Pe = Vx/2\alpha$, where V is the cutting velocity, x is some reference position (in our case, a position near to the kerf where temperature rise is important) and α is the thermal diffusivity. Conduction effects get small when the value of Pe gets large. Given that α is fixed for a given feedstock, the question becomes how large does V need to be for conduction effects to be modest enough to be neglected. One method to assess the relative role of heat conduction is to experimentally determine to minimum laser power, P*, necessary to produce a through-cut as a function of table velocity. When conduction effects are negligible, the ratio of P*/V, which has units of joules deposited per unit line length, should be constant. This is because it corresponds to the amount of energy necessary to remove equivalent volumes of material. Figure 4 shows such a plot for the case of an alumina-based tape. It can be seen that at high values of V, and thus Pe, the ratio does approach a constant value, whereas at low values very high relative power is required because conduction bleeds heat away from the illuminated region on a time scale that is competitive with the cutting speed. For practical cases, heat conduction effects were noted at speeds below 6.35 mm/s (0.25 in/s), but were never observed at cutting speeds above 10 mm/s.

Lamination

The second key issue is lamination, which must serve two independent roles. Firstly, it must provide "tacking" to preserve registration during stacking that is sufficiently strong to allow subsequent handling. Secondly, the process must eventually lead to complete fusion of the stack, that is, the final part must not have any memory of the original stacked motif. It is important to note that preservation of geometry only requires the former, and therefore the latter may be achieved with supplementary processing after assembly is complete.

In prior work, an adhesive joining process was described that has proven very successful in terms of laminating a wide variety of green tapes. A variety of adhesive formulations have been developed for different feedstocks, but all involve volatile solvents. Although eminently workable on the lab scale, this can cause a problem due to potentially harmful or flammable vapors collecting in high concentration when production quantities of parts are being produced. Thus, it would be a great advantage to design a feedstock that could be thermally laminated. Recently, such a system has been developed. A technique has been developed to apply a thin coating of a heat sensitive adhesive to the surface of green tape. Tacking is then achieved be either serial application of heat and pressure (e.g., a heat lamp followed by cold rolling) or simultaneous application of heat and pressure (e.g., a heated roller). The latter is of particular interest since it means that the tape can be directly laminated by a technology that has already been proven by the Helisys system.

The results of laboratory experiments are shown in Figs. 5a-d. These figures present scanning electron microscopy images of the cross sections of samples that each had a one-mil adhesive film and were laminated using a very small applied load at different temperatures. It can be seen that as higher temperatures are used, the film diffuses away leaving the continuous particle network necessary for good sintering behavior.

More importantly, it has been shown that if tacking is completed at a low temperature, e.g., 125°C, then subsequent annealing more elevated temperatures will cause complete fusion of the tapes, see Figs 6 a & b. This means a heated roller need only induce tacking, it need not dwell for a long time to allow complete migration of the adhesive.

To determine the feasibility of using a heated roller for laminating this type of feedstock, a set of two 3"x2"rectangular blocks was produced using a Helisys LOM2030. The following conditions were used for the roller: temperature = 140° C; speed = 1 in/s; compression = 0.015; and number of passes = 2. The feedstock laminated without difficulty and was later observed to fire to full density without delamination.

Equal success, though with a different method, has been demonstrated for steel feedstock.

Summary

The key technical challenges of laminated object manufacturing, laser cutting and defectfree lamination, can both be engineered through design of the feedstock on the microstructural level.

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Table 1. Green Tape Characteristics					
<u>Tape</u> Alumina Silicon nitride Stainless steel Graphite	Thickness 600 μm 300 600 300/600**	Solids <u>Loading</u> 56.6 vol.% 51.6 64.0 42.8	Binder <u>Content</u> 20.5 vol.% 29.9 36.0 21.9	Binder <u>Type</u> PVB* PVB Polyacetal PVB	Porosity 22.9 vol.% 18.5 none 35.3

*Plasticized polyvinyl butyral

**Fugitive tape for temporary support, thickness varies to match persistent material



Fig. 1a & b. Scanning electron micrographs of laser-cut commercial PZT tape that contains a polyvinylchloride-based binder. The kerf width is nearly twice the diameter of the beam, it has a ragged edge, and there is cracking both ahead of the moving laser (see Fig. on right) and along the edges of the cut part.

21 vol. % Binder / 29 vol % Porosity



37 vol. % Binder / 13 vol % Porosity







200 µm

Fig. 2. Scanning electron micrographs of the top (upper row) and bottom (lower row) faces of a laser cut silicon nitride tape that were identical expect for the relative concentrations of binder and porosity.



Fig. 3. Cross section of the laser cut silicon nitride tape (21% binder) shown in Fig. 2.



 $(a=2540 \text{mm/s}^2)$

Fig. 4. Plot showing the minimum power necessary to cut through either a single or double layer of alumina-based tape. At high velocities, the power consumption reached a plateau that is directly related to the thickness being cut.



(a) 75°C

(b) 125°C



(c) 150°C

(d) 175°C

Fig. 5. Scanning electron micrographs of the cross section of alumina laminates made from polymer-coated green tape (2.5kPa for 2 hrs at specified temperatures). It can be seen that at higher laminating temperatures, the polymer layer dissolves into the base tapes giving rise to a uniform green microstructure.



(a) As-laminated sample at 125°C



(b) Heat as-laminated sample to 175°C for 10hrs

Fig. 6. Scanning electron micrographs showing the cross section of polymer-coated alumina tack laminated at 125°C, then annealed at 175°C for 10hrs. These micrographs indicate that, provided intimate contact is established during tacking, it is not necessary to apply pressure to induce the redistribution of the polymer coating.