Curved Layer LOM of Ceramics and Composites

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ABSTRACT

A novel rapid prototyping (RP) technology incorporating a curved layer building style has been developed. The new process, based on Laminated Object Manufacturing (LOM), is suited for efficient fabrication of curved layer structures made from ceramics and fibrous composites. A new LOM machine was developed that uses ceramic tapes and fiber prepregs as a feedstock and outputs a three dimensional green form. The green ceramic is then processed to a seamless, fully dense ceramic structure using traditional ceramic techniques. This report summarizes the new LOM process and necessary hardware. Also reviewed is the development of ceramic preforms and accompanying process technology for net shape fabrication of ceramic matrix composites (CMCs). Compared to making curved objects with the standard flat layer LOM process, the curved process affords the advantages of eliminated stair-step effect, increased build speed, reduced waste, reduced need for decubing, and the ability to maintain continuous fibers in the direction of curvature.

CURVED LAYER LOM

Based on LOM, a novel RP technology has been developed to provide an enhanced capability for fabricating structural ceramic and composite articles. A comprehensive research program was recently completed that involved materials and process research, machine design, and system software adaptation [1, 2]. The envelope of available RP materials was expanded to include structural composites. In addition, an entirely new building paradigm was implemented: the *curved* layer building style. Instead of being limited to building with flat layers, the LOM machine is now capable of building in a curved-layer manner. The new curved layer LOM process allows continuous fiber composites to maintain fiber continuity in the plane of curvature in order to achieve optimum mechanical performance.

The curved layer LOM process originated from the need to fabricate fiberreinforced structures containing sloping, curved surfaces, especially thin curved-shell components. An important factor in these structures is maintaining fiber continuity in the curved surfaces. All RP processes are capable of fabricating complex, curved geometries using thin flat layers in combination with post machining of the final part. However, flat layer RP processes are incapable of addressing the larger geometrical issues involved with fiber composite fabrication, namely fiber orientation and continuity. In addition to the technical incentives for implementing a curved layer build style there is an economic incentive. Such a process will have a favorable cost benefit payoff because of the reduced raw material costs compared to flat layer processes (see Figure 1). This results from a dramatic reduction in the amount of waste material generated during processing.

The curved layer LOM process is illustrated in Figure 2. It begins with production of a matched tool or mandrel for the intended part. The temperature and pressure requirements for this mandrel are not demanding, so it can be made with the standard flat layer LOM process using LOM paper. The finished mandrel is mounted to the flat building platform in the curved layer LOM machine. Sheets of the desired build material are loaded onto a rotatable feed table, picked up with a vacuum chuck, and transported to the mandrel. A flexible thermoforming mechanism laminates each new layer to the curved mandrel with steady, uniform pressure. A laser cuts each layer accounting for the sloped surface. The fiber orientation can be varied from layer to layer by programming the rotatable feed table. The process proceeds one layer at a time until the part is finished. The part is then removed from the mandrel and the excess material is manually stripped away ("decubed"). The advantages of the curved layer process are elimination of stair step effect and improved surface quality, increased build speed, reduced waste, and easier decubing.

Since the new process is a significant departure from the flat layer LOM process, substantial new hardware and software development was necessary. The software issues are summarized in a previous report [3]. New hardware components included the material sheet feeding and rotating mechanism, curved layer bonding apparatus, and curved surface laser cutting. An automatic sheet feeding system rather than a roll feed was desired for two main reasons. First, many commercially available advanced materials, such as prepregs (preimpregnated fiber preforms) and ceramic tapes, are stiff at room temperature and/or available only in sheet form. Second, there is more material waste associated with the automatic roll feed system. Although this waste may be affordable when building LOM-paper parts, efficient use of material is critical when working with costly feedstocks such as fibrous composites and ceramics.

The curved layer bonding mechanism is a flexible, resistively heated pad that is backed with a silicone rubber frame (Figure 3). The frame consists of a top and bottom horizontal surface connected by vertical walls. The placement of the internal walls was custom designed to provide even pressure to the curved layer parts under consideration in this study. Vacuum cups protrude vertically though the rubber frame in order to pick up a new material layer. A single layer is held just underneath the heating surface as the entire assembly is moved over the mandrel via a rail system. The new layer is laminated by elevating the build platform up into the thermoformer and holding it in contact for about one minute. The bonding pressure is approximately 2 - 10 psi, which is adequate for laminating typical tapes and prepregs. Figure 4 shows a ceramic part that has been layed-up on a LOM paper tool. Cutting curved layer parts required new computer algorithms for coordinating simultaneous control of the laser beam in the X, Y, and Z directions. In a conventional LOM machine, laser cutting is performed using a plotter system that transports a mirror and focusing lens over the X-Y envelope. The distance between the focusing lens and the top of the part is adjusted only once after placing each new layer, ensuring that the laser focus is maintained on the horizontal part surface. With the curved layer LOM machine, the build platform must be translated up and down dynamically in order to maintain the laser focus on the curved surface. An eight axis motion control card and new software algorithms were necessary to control this motion smoothly and quickly. A 225-Watt Diamond (pulsed) CO_2 laser was installed on the machine and proved effective in cutting SiC fibers and ceramic tapes. The quality of the fiber cuts was not as good as that obtained with a copper vapor laser as previously reported [1, 4], but the Diamond laser is more reliable, smaller, and less expensive than the copper vapor laser.

CMC PREFORM DEVELOPMENT FOR LOM

SiC/SiC was used as a focus "model" material system for demonstrating curved layer, net shape CMC fabrication. A novel approach was used to fabricate CMCs that involved lay-up of separate, alternating layers of ceramic tape and fiber prepreg [1]. Green parts containing this alternating layer architecture were made using LOM, subsequently pyrolyzed, and densified through reaction bonding.

Fiber prepregs were fabricated with unidirectional SiC fiber tows (Nicalon[™], Dow Corning, carbon coated grade) and a furfural-phenolic thermosetting resin (FurCarb[®] UP440, QO Chemicals, Inc). The furfural resin served a dual role: as a binder and adhesive during the part fabrication and as a carbon source during the subsequent reaction bonding process. Descriptions of the reaction bonding process and monolithic SiC tapes are given in a previous report [1]. Alternating layers of the fiber prepreg and ceramic tape were delivered to the LOM machine as one single sheet, referred to as a "tape-preg" [1]. Tape-pregs, made by pre-adhering a prepreg layer to a tape layer, were stiff, non-tacky, and board like at room temperature, becoming soft and pliable at higher temperatures (130°C). The tape-preg contained approximately 25% fibers by weight, and overall it proved to be a robust sheet material suitable for the LOM process.

The advantages of the alternating layer technique are the relative ease of preparation of the preforms, elimination of fiber abrasion from the ceramic particles, reasonably high fiber volume fraction in the final CMC, and low cost resin binder. The tape-preg embodies several important characteristics of other commercially available CMC preforms. Key among these characteristics is the use of a fiber prepreg that contains a thermosetting, ceramic precursor resin. Based on this similarity, it is anticipated that LOM will accommodate other commercially available CMC preforms as easily as the "tape-preg".

Several parts were manufactured with curved layer LOM using SiC/SiC tapepregs. Acceptable process parameters (e.g., temperature, pressure, time) were determined through initial experimentation with a manual operation, mock-up system at the University of Dayton [4], and finally through operation of the actual curved layer LOM machine. A bonding time of 1 minute at 130-150°C was adequate for the tapepregs; the bonding pressure was estimated to be 5 psi. Several curved layer parts were produced and successfully decubed. These green composites were not fully cured, but they were stiff and self-supporting. Curved layer LOM was also used to fabricate monolithic SiC parts by laminating only the ceramic tapes (no fibers). The temperature in this case was 90°C, and butanol was sprayed between the layers to act as a tackifier. Thus, the LOM process was successfully used to produce various types of green ceramic forms that were robust and handleable. Additional processing steps were required to convert them to full density.

POST-LOM PROCESSING

It is not possible to execute all of the necessary ceramic processing steps within the LOM machine. The LOM machine is used only to produce the green form. Thus, post processing steps are needed. The overall process used for SiC/SiC is illustrated in Figure 5.

The first post-processing step combines binder burnout, pyrolysis, and pressing into a single operation. Although green LOM parts are sufficiently laminated to survive LOM processing, decubing, and handling, they require additional layer consolidation prior to the final densification step. Pressure must be applied during binder burnout and pyrolysis to counteract delamination and bloating that result from several sources such as outgassing and relaxation of residual stresses imparted during the lamination step. In order to maintain the complex geometry of green parts during pressure application, a technique involving quasi-isostatic powder pressing was developed.

Green parts from the LOM process were placed in a cylindrical chamber which was subsequently back filled with powder and fitted with a ram. Monolithic SiC was used as the powder medium in the chamber. For LOM parts that could be fitted with a porous Teflon bag, silica was used as the powder medium (i.e., sand pressing). A heated, programmable, uniaxial press was used to apply a heated pressure cycle to the enclosed chamber. The powder medium enabled pressure to be evenly distributed while allowing volatile degradation products to escape through a permeable bed. The various SiC/SiC and monolithic SiC parts were pressed at 60 psi and 30 psi, respectively, and at temperatures up to 325° C. Shrinkage of SiC/SiC and monolithic SiC parts during this step was $7\% \pm 2\%$ and $5\% \pm 2\%$ respectively, in the z direction (parallel to press movement), and zero in other directions. Appropriate scaling of the computer graphic file for part building in the z direction by 7% or 5% will compensate for this dimensional change due to compression.

An additional, freestanding binder burnout and pyrolysis step up to 700°C in argon was needed to achieve full chemical conversion of the organic resins to carbon. Because most of the total expected binder burnout and resin pyrolysis occurred during the powder pressing step, no part damage or shrinkage accrued during freestanding burnout.

The resulting porous structures were densified through reaction bonding. The final parts are shown in Figure 6.

The microstructure of flat LOM SiC/SiC panels prior to pyrolysis illustrates the alternating tape/fiber layer arrangement, excellent compaction, and minimal porosity (Figure 7, left). The photomicrograph of a specimen that has undergone reaction bonding indicates that there is some porosity, although much of this may be attributable to particle and fiber pull-out that occurred when the specimen was polished. It is clear, however, that the flatness, continuity, and integrity of the layers have been well maintained. The level of porosity is comparable to that of commercially available CMC systems.

Upon closer examination of the LOM SiC/SiC microstructure, it is evident that although the fibers are intact, there is fiber damage particularly at the fiber interface (Figure 8). This damage occurs as a result of the instability of Nicalon fibers at the temperatures encountered in the reaction bonding process (up to 1600°C). There are several potential solutions to this problem, such as developing superior fiber coatings and using silicon alloys that will infiltrate the composite at lower temperatures. However, the physical mating of the layers is intimate and the overall infiltration efficiency is quite good relative to commercial CMC systems.

Testing of final SiC/SiC specimens in four-point bend was not possible due to damage to specimens during cutting. The brittle behavior of the samples was expected, since the fiber coating problem has not been solved. The major result to report here is that the entire process, starting with fiber preforms and ending with a near net shape, densified part, has been fully demonstrated. Densified samples such as the flame holder and body armor are handleable, and photomicrographs illustrate that the microstructure compares quite favorably to commercial CMC systems. Further work with other material systems is being carried out to fully characterize and assess the overall process reproducibility and accuracy.

SUMMARY AND CONCLUSIONS

The feasibility of using LOM for net shape, freeform fabrication of monolithic ceramics, continuous fiber CMCs, and curved layer composites has been fully demonstrated. Monolithic SiC and continuous fiber SiC/SiC were used as demonstration material systems. Commonly available preforms such as ceramic tapes and fiber prepregs are entirely suitable as feedstocks to the process. The overall process methodology involves use of a LOM machine to produce dimensionally accurate green forms directly from CAD files, followed by post processing steps to bring the part to full density. Both flat layer and curved layer laminates can be built with this technology. The curved layer paradigm is critically important for fabricating continuous fiber, curved shell composites in order to maintain fiber continuity. Curved layer SiC/SiC composites were produced with good microstructure, although mechanical properties are not expected to be as high as commercial CMC systems due to ineffective fiber coatings. Inherent to the process of making the CMCs was the production of a precursor composite structure, consisting

essentially of a polymer matrix composite (PMC), which was subsequently converted to a CMC. Thus, the results demonstrate that PMC fabrication is also viable with the new LOM process. This also was demonstrated in our previous LOM processing studies using epoxy resin prepregs [5].

Although the parameters of the various processing steps are expected to change with different materials systems, the overall LOM process is expected to be generically applicable to a wide range of high performance materials. Thus, an entirely new fabrication capability is now available for designers and manufacturers. The new LOM process holds promise for lowering the cost of fabricating monolithic ceramic, CMC, and polymer matrix composite parts by virtue of its automation, which reduces fabrication time and eliminates the need for tooling. The process can best be applied by industry as a product development tool for fabricating molds, tools, or testable prototypes, or used directly for small lot production.

This is an ongoing effort. The immediate following steps are to improve the laminator design to achieve higher pressures and more flexibility, switch to working with commercially available CMC material systems, further investigate lasers that combine the advantages of the copper vapor laser and Diamond CO_2 laser, and investigate low cost tooling materials which would provide more robust mandrels for the curved layer process than LOM paper. Also, the laminator size envelope will be expanded to near or equal to that of a standard LOM2030 machine (32" x 22").

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Figure 1: comparison of the flat layer and curved layer LOM processes for building a curved shell. Due to the mode of building, the curved layer process will require fewer layers of material to build the same part.



Figure 2: curved layer LOM process schematic.



Figure 3: curved layer bonding apparatus (prior to installation in LOM machine). The vacuum cups, which protrude though the structure normal to the heater surface, are not installed on the unit in this picture.



Figure 4: curved layer, monolithic SiC body armor panel, immediately after LOM processing. The piece has been fully decubed and placed back on the LOM-paper mandrel for illustration. Notice the smooth surface and lack of stair steps.



Figure 5: overall process flow chart for fabricating net shape CMCs with curved layer LOM process.



Figure 6: Reaction bonded SiC parts made with curved layer LOM process: three SiC/SiC aircraft engine flame holders of various curvature, and one small-scale monolithic SiC body armor panel.

Figure 7: Polished cross section of (left) eleven-layer SiC/SiC LOM composite prior to pyrolysis, and (right) after pyrolysis and reaction bonding. From top to bottom the layers are: ceramic tape, 90° fiber layer, ceramic tape, 0° fiber layer, ceramic tape, etc.

Figure 8: Photomicrograph of reaction bonded SiC/SiC microstructure showing mating of various layers, infiltration efficiency, and fiber condition.