Laminated Object Manufacturing of Ceramic-Based Materials

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Since their inception, additive manufacturing (AM) techniques have been the go-to methods for obtaining highly complex-shaped rapid prototypes (RPs) and specialized parts, which were produced in small lot sizes. The AM technique of laminated object manufacturing (LOM) is an immensely convenient and cost-effective method for quickly producing millimeter-sized to meter-sized parts, while incorporating micrometer-sized constructive features. LOM machines offer an open work space, within which nontoxic and highly filled sheet materials can be processed at a high production velocity. The unique property profile of ceramic-based materials from LOM may be indispensable for applications calling for materials that unite high temperature resistance, mechanical strength, and light weight. Optionally, local material functionalization may engender the electrical conductivity, chemical stability, ferroelectricity, radiation shielding, or filter membrane stability of a limited portion of the material. Herein, a detailed evaluation of the applicability of LOM in the near net shaping ceramic-based materials is presented. Optional technical adjustments for the LOM process and extensions of the LOM machine configuration can improve the economic feasibility its operation. Previously successful LOM-printed ceramic-based materials are showcased within a comprehensive overview on the state of the art and potential novel composite materials are presented.

1. Introduction

As it has been laid out by Beaman,[1] the history of laminated object manufacturing (LOM) begins with the patent of DiMatteo,[2] which had been filled 1974. Within this patent, a computer-driven tool for cutting-out 2D shapes from layers a sheet material was presented. At each processing step, the shapes were cut from the topmost layer of a stack of sheets which had been continuously bonded onto each other. By doing so, highly complex of geometries for airfoils, propellers, or prototype objects should be achieved. DiMatteo did not specify the type of material. Historically, however, common sheet materials had been sheets of paper or wax. The LOM technology itself is partially based upon a preceding craft technique for building-up 3D topography relief maps, developed as early as in the 1890s. As patented by Blanther,[3] the topography relief maps were constructed from wax plates that each were impressed with desired topographical contour lines. Subsequently, the single wax plates were cut-out along the contour lines, and, the wax plate surfaces were smoothened. As a result, two interlocking positive and negative topographical surfaces were established, corresponding to the topology. By inserting and pressing moldable sheets of paper in between the two relief maps made from wax, the so-created 3D topography was transferred onto folded paper. For the birth of LOM yet a second crafts technique had to be developed. This second crafts technique was photosculpturing, as invented by the French sculptor Willème in the 1860s.[4] In his photosculpturing method, a person or an object was simultaneously photographed within a circular room, by means of 24 cameras at equal radial positions with respect to the person or the object. The sculptor used the person’s or object’s silhouette on each of the photographs to gradually carve out a 1/24th cylindrical portion from a sculpture in each iterative processing step. Later on, this tedious process has been simplified by Baese,[5] who exposed a cylindrical form of photosensitive gelatin to light which was gradually illuminated through floodlit photographic plates. The exposed gelatin expanded locally in volume, directly proportional to the incoming light intensity. The so-obtained annular rings within the gelatin, generated from the light exposure, served as markers for the later sculpture.

In the LOM process, a combination of the two aforementioned crafts techniques of photosculpturing and relief mapping together is replaced by a computerized part recognition or part design process and a subsequent automated multilayer part building process. Consequently, recognized or designed 3D structures are projected onto multilayers of a certain sheet material. The commercialization of LOM machines has been...
driven by the former Helisys Corporation (later continued as Cubic Technologies Inc.), starting from 1986. In the LOM process, layers can be either first laminated and then cut, allowing for a less error-prone parts production and for the usage of excess material as support structures, or, layers can be cut first and then laminated. Completed LOM parts are retrieved by peeling-off any support material from the part, a procedure referred to as “waste removal.” The two variations of LOM are hence defined as the “cut-then-bond” or “cut-off-the-stack” method, and, the “bond-then-cut” or “cut-on-the-stack” method. LOBM by the “cut-then-bond” method has also been referred to as computer-aided manufacturing of laminated engineering materials (CAM-LEM).[7] In publication about the CAM-LEM process, polymer tapes, partially filled with ceramic particles (casted green preceramic tapes), or metal foils, were rolled onto each other at roller temperatures up to 80 °C and compression forces of 0.34–0.68 MPa. The surface, on top of which the stack of laminated sheets was deposited was either plane, or had a curved surface (also referred to as “curved LOM,” see Section 2.2.2) which enabled an even higher variability in the part geometry. The excess material after cutting each single sheet layer was removed before the lamination. Using layers of 30–1300 μm in thickness, microfluidic ceramic or metal devices have been generated by CAM-LEM.[8–10] In regards to the types of sheet materials, papers, polymer tapes, or metal sheets all are viable types of material. In the case that papers are applied, each sheet of paper has to be coated with a selectively activatable adhesive, e.g., with a heat-activatable, thermoplastic adhesive layer.

Modern LOM machines differ from the set-up patented by DiMatteo in many ways. Most importantly, a continuous material feed is made possible, meaning a continuous strand of paper, polymer tape, or metal foil is run through instead of a tedious manual layer stacking. The cutting technique itself has vastly improved by the application of a CO₂ laser and multiple sensors in the machines, ensuring an optimal layer positioning. A basic schematic of the LOM process is shown in Figure 1. A part layer contour, or part cross-section outline, is cut into a layer of sheet material, prior or after its lamination onto a stack of already cut and bonded previous layers. In addition to this contour line, a pattern of cuts (dubbed “crosshatches,” see Section 2.2.1) has to be added to the topmost layer to enable the waste removal at the end of the LOM procedure. In the case that the support material, which has to be removed, is built as a cube structure, the waste removal is also referred to as “decubing.” The shape of cubes is chosen because it is the simplest-possible geometry for the support material. Optional additions to the basic LOM process, which have been presented throughout its history, enabled the freedom in parts design to increase substantially (Section 2.3).

Along with other additive manufacturing (AM) technologies, LOM is considered to be an integral part of the recent technological development, referred to as the “Fourth Industrial Revolution” or “Industry 4.0.”[12–14] AM enables the economic, material-saving, and environmentally friendly production of specialized components of varying sizes in between 1 μm and 10 m (Figure 2).[15] An increasing number of studies on the economic importance of AM technologies has been published over the years, including mentionable studies from Kleer and Piller,[17] Farayibi and Abiyou,[18] Wohlers and Gornet,[19] Tofail et al.,[19] as well as Ciobota.[20] With the advent of desktop-sized AM machines for home use, the minimum purchasing price for a machine decreased from $100 000 in 1990 to $500 in 2018.[15] Thereby, the production of AM parts from polymer and metals is far more common than AM parts from ceramic-based materials, glasses, hard metals, or smart materials. The reason for this is that the materials of the latter set typically require multistep production procedures, including a sintering process outside of the respective AM machine. Therefore, bringing the AM production of ceramic-based materials to industrial production levels may require an extensive development activity. A prognosis of the future significance of any AM can be derived from economic-mathematical models. One such model was presented by Thomas,[21,22] a technology diffusion model, known as the Chapman model, or extended Mansfield model, for estimating the proportion \( p_{\text{user}}(t) \) of all potential users to incorporate AM technologies in their production cycle by time (Equation (1))

\[
p_{\text{user}}(t) = \frac{\eta}{1 + \exp(\alpha - \beta \cdot t)}
\]

where \( \eta \) is the market saturation level, \( \alpha \) is the location parameter, \( \beta > 0 \) is shape parameter, and \( t \) is the time. This technology
A diffusion model can serve as an indicator for the impact of a technology on the industry, yield an S-shaped logistics curve. Expected values for the location and shape parameters are: $3 \leq \alpha \leq 5$ and $0.4 \leq \beta \leq 0.6$.\[23\] Economic-mathematical models, such as the Chapman models, are based on practical market data, such as AM machine sales figures or company investments in the development of new AM machine models. Nevertheless, relevant factors on the real market developments, such as the compatibility of certain AM technologies with the products from potential users, may be overlooked in the forecasts.

While metal parts obtained from AM have been considered in the commercial application such as motor parts for automobiles\[24,25\] or structural elements in airplanes,\[16\] the commercial applications of ceramic-based materials from AM include or have included cementitious materials extrusion\[14\] dental implants and microwave guides,\[27\] armament parts\[28\] and heat exchangers.\[29\] To ensure industry standards for AM processes, such as standards for the part quality or a sustainable, safe, and consistent production, Rodríguez-Prieto et al.\[14\] proposed a six-stage certification procedure (Figure 3). Such a certification procedure can be applied once a successful upscaling of the parts production has been implemented, so a sufficient production volume and a sufficient level of quality control are reached. Resulting AM parts have to fulfill certain desired property criteria, depending on the specific technical applications. These criteria are characterized by key performance indicators (KPIs).\[160\] A strategy for the design planning of AM products has been proposed by Perez et al.\[15\] and by Camburn et al.\[112\] the 4D Process, with its phases “discover,” “define,” “develop,” and “deliver.” In the first phase, efforts are undertaken to understand the needs of the user of an
AM machine and the current market demands. A certain identified needs and potential functions of the planned AM part are prioritized after project management tools are applied, such as the quality function deployment (QFD) method or Ishikawa diagrams. In the “develop” phase, a proposed part design is tested and validated. Once a successful rapid prototype (RP) has been selected for AM production, the “deliver” phase begins, where AM processes are iteratively improved to meet production standards. To start any AM process, a computer-aided design (CAD) file according to ISO 13567 is required. The CAD file includes the precise geometry of the AM part, information about the material texture, and its component structure in case the part is built-up from multiple components. The property constraints on the part, depending on the part material, and its performance within the expected engineering environment of the planned technical application can be determined by help of finite element method (FEM) simulations. This methodology of part planning has also been termed “design for additive manufacturing” (DfAM).[31] To transfer the data of the CAD model to corresponding machine of the AM process, the CAD file is converted into a standard tessellation language file or stereolithography tessellation file (STL file).[32] The STL file contains a triangular wire mesh that simplifies all rounded surfaces of the CAD model as multiple planar facets. This geometrical simplification is necessary for defining discrete edges for the AM part to be printed. During the printing process itself, the 3D shape stored within the STL file has to be sliced into multiple 2D layers. According to the Standard ISO/ASTM 17296, the LOM process is categorized as a sheet lamination (SL) which is one of the seven types of AM techniques. The other six AM technique types are listed as follows: material extrusion or extrusion-based additive manufacturing (ME or EBAM; which comprises 3D printing), vat photopolymerization (VP; also known as stereolithography), material jetting (MJ), direct energy deposition (DED; e.g., laser engineered net shaping), binder jetting (BJ), and powder bed fusion (PBF). By conventional definition, the term “direct AM” is assigned to AM procedures in which parts are fully finished just after being printed, whereas “indirect AM” describes AM procedures that solely produce precursors to fully finished parts. Essentially, direct AM is characterized in that the corresponding parts are finished immediately after their fabrication within the AM machine. Fabricated parts in an indirect AM technique, however, are adopted as molds or are to be used as preforms for a subsequent finishing step. One prominent example for indirect AM is the PBF of complex-shaped sand molds for steel casting (see ISO 52919). In addition to the more commonly known polymer-based and metal-based AM parts, ceramic-based AM parts may become of particular interest or even indispensable in various fields of applications, like transportation, defense-oriented applications, energy generation, as well as environmental and biomedical applications.[27,33] Within recent reviews by Gonzalez-Gutierrez et al.,[34] Deckers et al.,[7] Zocca et al.,[15] Yang and Miyataji,[36] and, by Colombo et al.,[37] the practicability of different AM techniques in achieving state-of-the-art ceramic-based components was discussed, including LOM. This work specifically focuses on progresses in the LOM production of ceramic-based materials and its economic potential.

2. Detailed Description of LOM

2.1. Characteristics of LOM in Regards to Other AM Techniques

While under-represented by industrial standards, LOM can offer considerable advantages when compared with the other AM techniques. In contrast to the other AM techniques, which use powdery or liquid raw materials, a versatile range of raw materials, including fiberglass or ceramic fillers, can be processed by SL techniques in general. Within the technique of LOM, a subcategory of SL, nontoxic, relatively inexpensive, and continuous strands of paper or polymer tapes are laminated on top of each other, yielding complex-shaped, 3D multilayers. Compared with other AM techniques, LOM can accelerate the production of large-scale parts.[7,13,38,39] The open set-up of the basic LOM process can be put to use when adding further automated working processes to it. For instance, Bhatt et al.[40] combined with LOM with robotic sheet manipulation processes (see Section 2.3), enabling the incorporation of prefabricated components into a multilayer stack or the combination of layers from multiple sheet materials. When comparing the characteristics values for the printing resolution from different AM techniques, LOM provides a printing resolution that can compete with all other techniques in addition to MJ (Figure 2).

A potential benefit for the AM of ceramic-based materials in general are the near net shaping of customized parts without the requirement for tooling.[15] This advantage is especially important in the processing of often hard and brittle ceramics, as any added finishing procedures, such as grinding and polishing, may involve high costs and may significantly extend the overall production time. In contrast to other AM techniques, a wide variety of raw materials can be processed by LOM, neither involving toxic chemicals nor complex chemical reactions. The variability in part size and utilization of widely available paper has given rise to the commercialization of desktop LOM machines. Desktop LOM machines are useful for building paper or polymer models of implants, later to be reprinted by a medical biomaterial, or
Furthermore, the sheet materials can be coated, printed, embossed, or otherwise altered by dry-processing methods prior to be integrated into the built LOM part.

Despite the higher level of manufacturing flexibility, a disadvantage of LOM, as perceived by some authors, is its limited capability to produce AM parts with internal cavities. As shown by Au et al. and by Niels et al. however, microfluidic devices with channels widths as thin as 800 μm, at the channel height of four times the layer thickness or 100 μm, were successfully created by means of LOM, utilizing layers of poly(methyl methacrylate) (PMMA) and Mylar. Shulman and Ross built larger-scaled microfluidic devices with channels of 690 μm in width from casted tapes highly filled with AlN, mullite, and ZrO₂. The obtained parts were converted into fully ceramic composites by a subsequent heat treatment, which caused the channel width to shrink to 410 μm. An overview of the size ranges and part feature complexities for LOM and other AM techniques is shown within a Venn diagram in Figure 4. The complexity of the part feature can range from a planar feature to a fully 3D feature that is predictably and time-dependently altered by chemical or physical influences (left axis). The features of the highest complexity level are referred to a “4D features” or “smart features” (not to be confused with the 4D Process). One of the best-known examples for such a material feature is the shape memory effect, as demonstrated by a part made of methacrylated polycaprolactone (PCL) polymer (box on the top right). Smart materials, such as the shown PCL polymer parts (models of a bird and the Eifel tower) could be manufactured by 4D printing (4DP), an extended version of the 3D printing (3DP) process. The printed smart PCL parts could be deformed and reverted into their original shape at a temperature of 70 °C. In the commercial 3DP process (blue rectangle with rounded corners in the diagram of Figure 4), AM parts can be printed that contain prismatic or 3D features in the size range in between 100 μm and 10 m. Other AM techniques, namely, stereolithography (pink oval), material jetting by nanoparticle deposition (brown oval) or aerodynamically focused nanoparticle (AFN) deposition (purple oval) and DED by focused ion beam (FIB) or electron beam deposition (light green circle), are more limited in the assembly of AM part features.

While this Review focuses on ceramic-based materials obtained from LOM, it should be mentioned that the manufacturing flexibility of LOM allows for the production several types of materials that are significantly harder to obtain otherwise: 1) metal-based composites with adhesive interlayers made either from polymers, brazes, or solders, bonding metal layers which would be otherwise inherently difficult to be bonded to each other (e.g., aluminum sheet with an oxide passivation layer under certain circumstances); 2) cured resins or other types of polymers reinforced with fibers of any selected aspect ratio and inert material type (e.g., single- or multiwalled carbon nanotubes or E-glass fibers); 3) combinations of the described fiber-reinforced polymers and metal layers as mixed composites.

For bonding metal foils onto each other, an AM technique similar to LOM can be applied that does not require any adhesive layers. In ultrasonic consolidation (UC), a sonotrode causes the metal foils to vibrate within ultrasonic frequencies. Due to such vibrations, metal foil layers deposited onto a growing multilayer would be otherwise inherently difficult to obtain otherwise: 1) metal-based composites with adhesive interlayers made either from polymers, brazes, or solders, bonding metal layers which would be otherwise inherently difficult to be bonded to each other (e.g., aluminum sheet with an oxide passivation layer under certain circumstances); 2) cured resins or other types of polymers reinforced with fibers of any selected aspect ratio and inert material type (e.g., single- or multiwalled carbon nanotubes or E-glass fibers); 3) combinations of the described fiber-reinforced polymers and metal layers as mixed composites. Furthermore, Schmidt et al. mentioned resistance welding as a further possible bonding mechanism applicable for metal sheets in LOM. In the case that parts are designed as two-component laminates, made from fiber-reinforced composites and metal foils, the layers with reinforcing fibers could be oriented at certain angles with respect to other layers.

2.2. General Procedure

2.2.1. Input Data Processing

Following up on the basic working principle, as shown in Figure 1, further details of the LOM procedure are explained.

![Figure 4](https://www.advancedsciencenews.com/)  
**Figure 4.** An overview of the complexity and size of features in polymer parts and the means of manufacturing for achieving the part features by a Venn diagram. An area with a brown paper bag texture (partially transparent) has been added to the diagram, representing AM part features achievable by LOM. Two MJ techniques are indicated by the acronyms in the figure nanoparticle deposition system (NPDS) and “AFN” and one DED technique is indicated by “FIB.” Adapted with permission. Copyright Year 2019, Elsevier.
within this section. The LOM procedure starts with the conversion of a CAD file into an STL file. CAD files depict 3D objects by rounded geometrical shapes which may or may not include rounded surfaces, as specified by the Standard for the exchange of product model data (STEP). STL files, in contrast, display wire frames corresponding to objects from the CAD files. The file conversion process from CAD to STL file format is referred to as tessellation, within which continuous object surfaces are replaced by triangular meshes. The development of tessellation algorithms since the 1990s has led to more precise AM product geometries, concurring with the requirements of the standard for engineering tolerances of technical drawing, ISO 2768. An early tessellation method called the “butterfly scheme” has been presented by Dyn et al. and put into use by Rypl and Bittnar. In a more recent study by Ledalla et al., a modified version of the butterfly scheme was tested in the conversion of CAD files into STL files, and, the performance of the method was compared with the Loop’s subdivision scheme and the triangular midpoint subdivision scheme as alternative methods. From the performance comparison, it became clear that the triangular midpoint subdivision was the most shape-accurate with respect to the original CAD object. The tessellation results can be optimized by repeating the mesh building, which iteratively yields a more and more accurate tessellation. The triangular mesh can be smoothened by a subsequent numerical operation, such as the Laplacian smoothing method (Figure 5). In the Laplacian smoothing method, undulating contour lines of the triangular mesh are straightened by moving mesh vertices toward the weighted means of the neighboring mesh vertices. As the local mesh size determines the tessellation accuracy, the triangular mesh may be graded additionally. For this purpose, a higher density of mesh vertices, and thus a finer mesh grid, can be chosen to draw object surfaces at bends and folds. Methods for narrowing the tessellation mesh at these complex features of CAD objects have been described by Hao et al. The authors explained the following three mesh partitioning methods: vertex-based methods, edge-base methods, and face-based methods.

The requirements for input files in AM processes have been formalized within the Standard ISO/ASTM 52915, which introduces the new file format additive manufacturing file format (AMF). While STL files can only convey the part design information as a triangular mesh, the new format contains additional information. Within the AMF files, accurate information about the tessellated object surface mesh is stored in addition to further specifications regarding color, texture, substructure, and material for the desired parts. AMF files enable the introduction of

![Image 1](image1.png)

![Image 2](image2.png)

**Figure 5.** The tessellation of a mechanical part and the underlying measure for the tessellation accuracy, given by the chord height $b_\Delta$. The tessellation was achieved by means of the butterfly subdivision scheme and Laplacian smoothing: the working principles of both mesh processing methods are illustrated on the right-hand side of the figure top part. The triangular mesh from the primary tessellation (on the left) is refined by repeated tessellation steps, whereas additional mesh vertices are added to edges and corners at complex object features (vertex-based method), allowing for a more accurate representation of strongly bent or folded surface parts. As shown on the bottom of the figure, the deviation of the triangulation mesh from the original curved surface can be captured by the chord height, or sagitta, $b_\Delta$, as the characteristic geometric parameter. For determining the chord height spherical triangles are overlaid on top of the tessellation triangles, with curvatures following the part surface along the projected length $L_\Delta$. Adapted with permission. Copyright Year 2006, Elsevier.
curved triangles as facets in the tessellation mesh of STL files. This measure increases the shape accuracy to a significant degree in comparison with a tessellation mesh with planar faces (Figure 5 and 6). The chord height $h_b$ in between the local object curvature and straight-line approximations of the curvature by a triangular mesh with planar faces is reduced drastically by curved faces\cite{60,61}. During the LOM part production, images of the 2D layers at the current building position are extracted from an input STL or AMF file. The 2D object cross-sections are either stored as common layer interface (CLI) files or directly transferred to the machine as information written in G-code\cite{62}. In each layer building step, the cutting pattern on each layer has to contain the accurate outline of the cross-section at the respective vertical position of the LOM part. An additional cutting pattern, in addition to the cross-section outline, enables the extraction of finished LOM parts. In the bond-then-cut method of LOM, a pattern of square-shaped crosshatches has to be added to each layer, in addition to the part’s cross-section outline. The crosshatches are placed in close proximity to the outline of the part cross-section, onto the outside area of each sheet layer. This outside area surrounds the part’s cross-section, which is defined as inside area. The crosshatch patterns, acting as supporting structures that hold LOM parts in place, may consist of equal-sized squares, or, may be patterns with squares in an adapted size-range. A crosshatch-size adaptation can be achieved by positioning the smaller-sized crosshatches closer to the cross-section outline and larger-sized ones further away. In this way, the LOM part decubing is facilitated in comparison with a uniform pattern of equal-sized crosshatches. Smaller-sized crosshatches in the adaptive pattern are less likely to become wedged within recesses of winding portions of the LOM part surface\cite{63}. A feasible computing approach to obtain an adaptive crosshatch pattern has been described by Pitayachaval et al.\cite{64} In their approach, a bounding box was defined, roughly enclosing an area sufficiently large to contain the entire inside area of the part. The bounding box was divided into four quarters, and a basic uniform crosshatch pattern was inserted into it. Within the inside area and its direct neighborhood, the basic crosshatches were subdivided into smaller-sized tiles, whereas basic crosshatches were fused into larger-sized tiles at greater distances from the cross section.

While the waste removal by cubic crosshatches can only be implemented in the bond-then-cut LOM method, the creation of bridge supports is preferred in the cut-then-bond LOM method. For this purpose, contiguous areas of the outside area can be removed from each layer prior to bonding, whereas a supporting bridge structure at each single layer is left intact, holding the inside area of the layer in place. The amount of material allocated to the bridge supports is kept to a minimum. A comparison between both cutting methods and resulting LOM parts is shown in Figure 7. Within this figure, hollow LOM parts from thermoplastic paper with a layer thickness of 0.1 mm are depicted. In the case of the shown calabash and ball-within hollow cuboid workpiece, the waste removal was only made possible by bridge supports, whereas the removal of regular-sized cubes from fixed-size crosshatches proved to be cumbersome, at best. Especially for hollow parts with narrow openings, as the calabash shown in Figure 8, the waste removal by decubing may not be possible. In the alternative bridge-supporting method, cut-out waste material is removed from the LOM workpiece already during

**Figure 6.** The results from different tessellations performed by different 3D design modules commercially distributed in 2016: the presented programs apply various tessellation algorithms on the same exemplary CAD input file, leading to the displayed results for the obtained STL output files. The resulting STL files can be either files in American standard code for information interchange (ASCII) format or in binary format. The resulting triangular meshes include tessellation triangles of varying sizes and vary therefore in accuracy in the object surface outline and have different file sizes. To compare the tessellation accuracies, the minimum triangle edge lengths at a certain circular feature of an AM part was measured by Hallgren et al. who used the commercial programs Dassault SolidWorks 2015, Siemens NX 9, Dassault Catia V5-6 2012, and Materialise Magics 18 at manually selected accuracy settings. Adapted with permission.\cite{60} Copyright Year 2016, Elsevier.
its build-up. In this setup, an optional additional continuous strand of paper or polymer tape coated with an adhesive may pick-up waste from the built-up multilayer. According to Liao et al., the amount of waste material which had to be removed after finishing LOM parts with the bridge supporting method was 30–80% less than for decubing.

2.2.2. Adjustments of the LOM Machine Setup

When working on a workpiece, to become integrated within a complete part, several different machine parameters may be fine-tuned to ensure a reduction of manufacturing errors. The different possible machine adjustments in LOM are explained in the following, starting with the layer cutting technique. For layer cutting either a tungsten carbide-cobalt (WC–Co) blade or by a CO₂ laser is utilized. A further alternative was presented in the study of Butt et al., which mentioned the water jet cutting of metal foils as sheet material. However, this cutting technique is unlikely to be applicable for paper sheets, as the paper may lose its structural integrity completely while being macerated by a stream of water. Depending on the sheet material, each way of layer cutting has advantages and disadvantages. On the bright side for laser cutting, a laser beam does not have to be reoriented for every cutting step, as a knife blade would have to be oriented to always face in the cutting direction. Moreover, a laser beam can cut through a more extensive variety of sheet materials in comparison with a metal knife. However, the generation of heat by the laser beam irradiation may pose a fire hazard, which becomes more pronounced the higher the laser energy setting becomes.

Effects of the LOM machine setup for the laser cutting, as the laser energy setting and the number of runs for each cut, have been examined by several authors. A passing laser beam leaves behind a typical parabolic cutting profile, shaping the outer edges of the cut-out layers (Figure 8). When a laser beam is utilized, the depth of the laser cutting profile is set equal to the thickness of one single layer. This is done to keep layers below undamaged, at a given laser cutting speed and laser energy. A complete cut may be achieved either in one or in multiple runs of the passing laser beam. Preventing a local overheating may be a reason to increase the number of runs, while reducing the laser energy, or vice versa, increasing the laser energy and decreasing the number of runs may be desired to heat local areas up. If the outline of LOM parts is separated by single cuts, waste material may still have to be torn away from exposed horizontal surface
portions at sloped surfaces in between layers, or at overlaps (Figure 8). To counteract the bonding of waste material onto the overlaps, one of two strategies for laser cutting may be applied: either the laser energy is set high enough to burn-off any adhesive bonds at the overlaps, or a pattern of tightly neighboring crosshatches is placed on top of the overlaps. The first method, applied to burn-off the adhesive, is also known as “burnishing” or “burning rule,” and the other method is simply described as “adaptive crosshatching” (see Section 2.2.1).\textsuperscript{[27,66–71]}

The surface roughness of LOM parts can be characterized in an analysis by means (ANOM). When considering the stepwise surface profile, typically leftover from a laser cutting procedure, the surface has to be characterized for any orientation on the part surface in separate. For each orientation on the part surface, a surface angle $\theta$ resembles the position of the normal vector $\hat{\mathbf{z}}$ with respect to the fabrication direction $\hat{\mathbf{B}}$. The schematic in Figure 9 shows this spatial relation.\textsuperscript{[11,69,72,73]} A further significant influence on the geometry of finished LOM parts, in addition to the layer cutting, is brought along by the bonding procedure. In many cases, a heatable roller is utilized for this purpose, however, applying a heatable planar pressing head can be a viable alternative. The main difference in regards to bonding by one of the two methods is the spatial distribution of induced mechanical stress and heat into an already built-up multilayer. When applying a roller for layer bonding, the mechanical stress is transferred as a line load, whereas a pressing head can compress the multilayer uniaxially across its entire cross section. The latter layer bonding method of uniaxial compression is highly recommended for a cut-then-bond LOM process, in which bridge supports are built-up instead of cubes.

During layer bonding, the parallel and flat deposition of new layers and the homogeneous attachment of the layers onto the multilayer are the most important criteria to avoid warping for the LOM part. In the case that the layer bonding is accomplished by a roller, a thorough analysis of the induced mechanical stress into the multilayer stack may be beneficial. As shown in Figure 10, the rolling step can be analyzed by means of FEM.\textsuperscript{[66]} By doing so, the local compression and adjacent relaxation of freshly bonded layers is simulated, which allows an assessment of the structural stability and any potential deformations of newly bonded layers. Sonmez and Hahn\textsuperscript{[75]} formulated a general expectation for the rolling process: bigger rollers enable a potentially more uniform lamination, whereas smaller rollers may allow faster roller speeds to achieve the lamination due to the greater concentration of mechanical stress at the roller position. A semi-analytical equation for the reaction time of the adhesive layer, $t_{ad}$, during the rolling process has been presented by Knyazeva and Travitzky (Equation (2))\textsuperscript{[76]}

$$t_{ad} = \frac{c_{\text{glue}} \cdot \rho_{\text{glue}} \cdot RT_{\text{surf}}^2 \cdot \exp \left( \frac{E_{\text{glue}}}{RT_{\text{surf}}} \right)}{z_0 \cdot E_{\text{glue}} \cdot Q_{\text{glue}}}$$  \hspace{1cm} (2)
where \( c_{\text{glue}} \) is the heat capacity of the adhesive topmost layer, \( \rho_{\text{glue}} \) is the density of the adhesive topmost layer, \( T_{\text{surf}} \) is the temperature value at the contact interface in between the topmost adhesive layer and the highly filled layer surface below, \( E_{\text{glue}} \) is the activation energy for curing the adhesive, \( Q_{\text{glue}} \) is the heat release during adhesive curing, and \( z_0 \) is a pre-exponential factor. For the sake of simplicity, it is assumed that the adhesive cures under adiabatic conditions. Furthermore, Lin and Sun\(^{[77]} \) have formulated a simplified relation to calculate the additional lateral displacement force \( F_{n+1} \) acting laterally on the layer \( n \), just below the freshly added topmost layer \( n+1 \), after each lateral movement step of the roller, potentially causing a layer stretching, and therefore, warping of the built-up multilayer (Equation (3))

Figure 9. The expected surface roughness of multilayer parts from the LOM process and a typical parabolic laser power profile (bottom). An scanning electron microscopy (SEM) image of a real LOM part, cut-out by a laser is shown on the right-hand side. Adapted with permission.\(^{[69,74]} \) Copyright Year 2003, Elsevier and Copyright Year 2008, Wiley.

Figure 10. Illustration of the bonding step in LOM: either achieved by a heated roller (left) or by a heated pressing head. An illustration of the “curved LOM” method, which may require modifications of the laminating roller or printing head, respectively, is shown on the top right. Critical influences on the quality of layer bonding are shown at the bottom of the figure: these can be the spatial distribution of the heat and mechanical stress induced by a roller, or, the deflection of thin features in the layers of sheet material during uniaxial pressing. Moreover, the optional features of LOM machines are depicted on the right-hand side: a nozzle to spray an adhesive coating and a rotatable working stage. Adapted with permission.\(^{[69,74]} \) Copyright Year 2003, Elsevier and Copyright Year 2008, Wiley.
where \( L_k \) is the vertical length of the \( n \)th layer, built-up at the iteration time step denoted by the index \( k \). \( E_{n,0.5} \) is the connection intensity which quantifies the bonding strength in between the layers \( n-1 \) and \( n \) \((k/k +1\): behind and in front of the roller). \( E^\circ \) is the critical value for the connection intensity for an added new layer, which is connected to the layers below by the area \( A_{layer} \). For a more accurate and thorough analysis of any potential deformations, FEM models are created for representing the planned LOM part. Such FEM models help to clarify the influences from mechanical stresses acting on a workpiece during laminating and influences from inflows of heat into a workpiece during building. Heat inflows and mechanical stresses can cause undesired delaminations, layer tears, or bulging of the sheet material in front of a heatable roller. Based on realistic FEM models, measures can be taken to avoid critical thermal and mechanical loads.[74,78]

The risk of layer warping may be reduced using a pressing head, instead of a heated roller, during laminating. In this case, the mechanical stress in homogeneously applied onto the laminated sheet area. In this case, using an optimized layer design with sufficiently wide supporting bridges can reduce potential deflections of thin features within the multilayer (see right-hand side of Figure 10). For each new layer added in the LOM process, the potential mechanical deflection \( \delta \) can be estimated by the mechanical law for bar bending (Equation (4)).

\[
\delta = \frac{F_b(L) \cdot L^4}{8 \cdot E_{mod}(T) \cdot I_A}
\]

with

\[
I_A = \frac{h^3 \cdot w}{12}
\]

where \( F_b(L) \) is the bending force, acting on the portion of feature furthest from a fixed mechanical fulcrum (e.g., the outer rim of a layer). \( L \) is the length of the bent thin feature, such as a bridge support in a freshly added layer, \( E_{mod}(T) \) is the temperature-dependent Young’s modulus of the feature, \( I_A \) is its area moment of inertia, in turn depending on the width of the feature \( w \) and height of the feature \( h \). The possibility of deflections in the layers is especially important if LOM parts with bridge supports are generated.

In addition to adjustments to the cutting and bonding of layers, further machine adaptations may allow for a desired orientation of each single layer within LOM parts. For doing so, the machine may be equipped with a rotatable working stage. In this way, single layers may be rotated prior to bonding. A further degree of freedom in LOM can be acquired by intentionally bending the layers of the sheet material prior to bonding (curved LOM). This is achieved by prefabricating a multilayer that acts as a mandrel (shown as brown layers in Figure 10), transferring a certain profile onto any sheet materials that are deposited on top of it. In the case of curved LOM, a certain mechanical deflection of each single layer is desired (Equation (4)).[78–80]

2.2.3. Potential Postprocessing Steps

The post-processing of LOM parts strongly depends on the very nature of the parts themselves. As mentioned by Cvetković et al.,[41] as well as Zhang and Liu,[25] paper products obtained from LOM typically display wood-like properties and may have to be moisture-protected by lacquer. For parts from curved LOM, undergoing a curing process for the adhesive or a compression step to improve the layer bonding may be necessary, as the curved part shapes may bring about layers that bend away from each other.[53,79–82] In addition to a waste removal or a coating step, an additional downstream heat treatment may be required. This is of particular interest for the LOM process on ceramic-based materials. Typically, LOM parts for ceramic-based materials are built-up from paper or polymer sheets highly filled with ceramic particles.[37,74,82–89] A heat treatment step at temperatures below 600 °C is required to remove any organic components from such a preceramic LOM part, either by debinding in an oxygenated atmosphere or by pyrolyzing the organic components under the exclusion of oxygen. After this first heat treatment step, a green body is generated that has to be consolidated by sintering. In the case that a debinding has been conducted, pore spaces are left-over from burnt-off organic material, such as interconnected pore channels left-over from burnt-off pulp fibers in highly filled paper sheets. As a feasible further pathway to generate ceramic-based materials by LOM, while reducing the content of organic material in the green bodies, Zhang et al.[90] used a ceramic slurry with Al₂O₃ particles as sheet material, which was applied by a doctor blade and then frozen onto an already built-up multilayer (FS-LOM). The lamination step with a frozen slurry is comparable to the process of freeze casting, a technique also explained by Li et al.[91] Parts from the FS-LOM process can be freeze-dried, instead of an otherwise required debinding or pyrolysis step. In this way, any potential boiling of the green bodies caused by combustion gases is avoided at the heat treatment. The applicability of FS-LOM, however, may be restricted to a limited number of applications due one significant drawback of this method: ice crystals grow until they reach a certain size distribution, with a limited maximum size value. The ice crystal size distribution is influenced by temperature, pressure, and the powder concentration in the applied slurry. After the freeze-drying step, any ice crystals are sublimated and the size distribution of pores left behind within the resulting FS-LOM green bodies is fixed. Any dry-processing steps to modify the pore size are not applicable on FS-LOM green bodies. One such dry-processing step, calendering, a rolling process that can compress layers of highly filled papers, has been applied to reduce the average pore size in paper-derived ceramics.[83,87,92–95]
An entire heat-treatment procedure is shown in Figure 11. As shown for LOM parts built-up from polymer tapes filled with carbide ceramic powders, a stepwise heat treatment of to derive ceramic-based materials from LOM parts may involve the infiltration of the green body by a liquid melt such as silicon melted at 1500 °C. Because sintering is almost always accompanied by a shrinkage in the dimensions of the sintering green body, any surface roughness features may shrink as well, in turn causing...
the surface of the sintering product to become smoother. Furthermore, the sintering shrinkage causes the pores in ceramic-based materials to contract and the infiltration by a liquid melt causes the narrowing or closing-up the pores within the ceramic. As for additional postprocesses that are adequate to smoothen the surfaces of sintered ceramic-based materials, vibratory bowl abrasion, manual optical polishing, micromachining, and laser micro-machining can be mentioned.\[96\] Thereby, laser micro machining is comparable with the aforementioned laser cutting technique, with the main difference being the laser energy setting adjusted for carving submicrometer-sized grooves into the part surface.

### 2.3. Optional Process Optimizations for LOM

Upgrades of the LOM machine can be either achieved by adding further machine components, or by pursuing certain methodologies. Simple machine modifications may be installing a nozzle into the LOM machine which can spray an adhesive coating on the single layers, as shown in the right side of Figure 10. In contrast to technical modifications, which are specific to the LOM machine, one simple methodology possible may be to conduct a careful part design planning. In the part design planning, engineering-driven and economically driven considerations can be made, as explained in the following.

Technical challenges and economic considerations can play an important role in the introduction of the LOM of ceramic-based materials. While the adaption of the surface roughness of finished parts at a given part building speed is a typical technical aspect, the efforts to shorten the processing time and reducing production costs is a typical economic consideration. One measure to reach these economic objectives can include appropriate savings in the used raw materials and in the energy consumption. As stated by Yim and Rosen,\[97\] the costs of a LOM machine, constitute from the machine purchase price, costs for raw materials, as well as, costs for machine operation and labor. The costs for operation can be calculated by the product between the operation rate and the build time. Costabile et al.\[98\] presented an equation to determine the total cost for each build \(C_{\text{build}}\) as follows (Equation (6))

\[
C_{\text{build}} = (C_{\text{indirect}} \cdot t_{\text{build}}) + (w_{\text{part}} \cdot P_{\text{mat}}) + (E_{\text{build}} \cdot P_{\text{energy}})
\]

where \(C_{\text{indirect}}\) is the indirect cost per machine hour, \(t_{\text{build}}\) is the total build time, \(w_{\text{part}}\) is the weight of all printed material during the build, typically the part itself and support material, \(P_{\text{mat}}\) is the price per kilogram of raw material, \(E_{\text{build}}\) is the total energy consumption per build, and \(P_{\text{energy}}\) is the mean price for electricity. To satisfy technical and economical demands at once, any LOM process can be combined with an elaborated part design, in which entire part designs are partitioned into multiple subparts. The feasibility of a part partitioning is often determined by the savings in production time, which depends on the build time for all subparts and the time it takes for assembling the parts. As clarified by Equation (6), the production time for LOM parts directly correlates to the costs of building a part. A modified manufacturing quantities diagram, originally used in the production planning for traditional subtractive manufacturing (SM) processes, can serve to visualize the range of production volume at which LOM may be economically most viable. The diagram for the cost estimation for parts from SM is converted into a part production time diagram (Figure 12)\[99,100\]. In contrast to any SM processes, LOM as an AM technique may render the storage and transportation of parts obsolete. The desired parts may be produced directly on demand and at the target location, which dramatically increases the importance of the part production time in the cost structure of LOM.

As derived from the modified manufacturing quantity, an optimal number of parts can be determined for which a subdivision into subparts becomes economically viable due to a reduction of the assembly time. Completed and potentially postprocessed subparts have to be assembled by means of one of four methods\[100\]:

1. discrete fasteners may be inserted;
2. an integral attachment can occur if the subparts interlock;
3. bonding can be achieved by energy deposition, e.g., in case the subparts are heated up to connect;
4. an adhesive coating in between the subparts can glue the parts together.

Practical guidelines for an optimized part subdivision have been described by Karasik et al.\[101\] (Figure 13). To avoid having to print sharp protrusion angles, concave angles, and large overhanging part surfaces, the parts are rotated and segmented in such a way that the surface inclinations have an angle of less than 135° with respect to the building direction. The authors implemented their subpart design rules within C++ and included programming functions to simplify the geometric complexity of the subparts.

---

*Figure 12.* The diagram with manufacturing quantities curves for SM processes after being adapted for AM processes: the primarily cost-related diagram is transformed into a diagram related to built time (according to refs. [33,99]).
Further process improvements than part partitioning are well-documented in literature. A recent approach for LOM is to include a robotic manufacturing cell, to enable the incorporation of prefabricated structures into LOM parts or the sequential application of two different sheet materials (“robotic cell LOM”). This approach has been limited to metal foils and polymer composite materials up to now. By including the prefabricated structures within robotic cell LOM multilayers, inclined faces and overhangs can be obtained without the need for support structures or a part partitioning step and an assembly step (Figure 14).

Other fabrication techniques in addition to LOM, in addition to the robotic manufacturing cell, have been included within a US patent from 2015 by Feygin, founder of the former Helisys Corporation. Some potential extensions to the LOM process mentioned in the patent are listed as follows: 1) the incorporation of a magnetized component within sheet materials, which may facilitate the waste removal during a magnetic separation step. 2) A printing apparatus for depositing a material in the form of a powder, liquid, or plasma on single layers prior to or after being, which enables the application of a refilling material onto undesired surface cavities in a LOM part during building. 3) A peeloff roller for the removal of any excess refilling material. 4) A sensor-controlled repositioning mechanism for inaccurately placed layers. 5) The application of an ultraviolet (UV) curable adhesive or UV curable resin, potentially highly filled with ceramic or metallic particles, either for applying a surface coating or for building-up an entire layer. 6) The installment of a mobile sand blasting device that allows for localized and selective removal of material or the smoothening of layer surfaces directly after lamination. 7) The utilization of protective masks to obtain a localized pattern from any sand blasting, coating, or printing procedure.

3. Types of Ceramic-Based Products Obtained from LOM

3.1. General Overview

The development of highly filled papers, preceramic polymer tapes, and frozen slurries, extended the manufacturing freedom...
of LOM dramatically. By the introduction of new sheet materials or frozen slurry layers, ceramic-based materials with microstructure of functionally graded material (FGMs), with targeted porosity distributions or with reinforcing fibers or particles, to generate ceramic matrix composites (CMCs). While FGMs are materials that contain a gradation in between two different material phases, e.g., an Al$_2$O$_3$ multilayer with an increasing content of interspersed ZrO$_2$ within layers along the top or bottom direction, CMCs contain distinctly separated second-phase materials dispersed throughout the material, e.g., an Al$_2$O$_3$ multilayer with reinforcing ZrO$_2$ fibers or particles. CMCs can be homogeneous composites, if the secondary components are distributed uniformly, e.g., reinforcement fibers with one single orientation direction. In CMCs which are heterogeneous composites, the secondary components are oriented within multiple directions, e.g., in the case of a fabric of reinforcement fibers. One prominent application of CMCs in thermal barrier coatings (TBCs) that withstand thermal shocks in high temperature environments.

Since the LOM had been established, casted preceramic tapes were used for obtaining oxide ceramics and silicon-based ceramics, whereas papers could be pyrolyzed and infiltrated by melt silicon to obtain silicon-based ceramics, exclusively. In the case of curved LOM, paper sheets were only used to generate the base support for desired curvatures. Glass-ceramics were included into the variety of LOMed ceramic-based materials by the application of preceramic tapes filled with LZSA (Li$_2$O-ZrO$_2$-SiO$_2$-Al$_2$O$_3$) glass. From 2007 onward, preceramic papers were used in LOM, as an additional sheet material in addition to preceramic tapes. Equivalent to green bodies from LOMed preceramic polymer tapes, the LOMed preceramic papers were subjected to a debinding or pyrolyzing procedure. In contrast to other types of paper that had been included within LOM parts before, preceramic papers were highly filled with ceramic particles (with typical filler contents of 70–95 vol%). Optionally, ceramic fibers could be included in the filler content. The type of ceramic filler in preceramic papers could be an oxide or a nonoxide ceramic. In optional dry-processing steps, preceramic papers could be compressed to generate narrower pores from burnt-off pulp fibers, or, could be printed or coated by a ceramic slurry, closing-up pore spaces within the papers.

Preceramic papers could be prepared by instruments on laboratory scale, such as by a dynamic handsheet former, either to be tested prior to a potential application in LOM or to be used in first RP parts from LOM. For larger-sized continuous strands of preceramic papers, a Fourdrinier machine in the industrial size-scale is required.

As a novel alternative to papers and preceramic polymer tapes, frozen slurry layers have been successfully tested in LOM by Zhang et al. By a freeze-drying postprocessing step, subsequent to LOM, a potentially fully ceramic green body is created. At the time this Review was written, the freeze lamination method was limited to Al$_2$O$_3$ ceramics. The size distribution and shape of the filler particles, as well as, dissolved species within the initially liquid medium may determine the speed of ice crystal growth and may cause the freezing temperature to be set to a certain value. After the initially liquid medium is removed by freeze-drying, the filler particles are fixed within a microstructure with laminar pores, left-over from sublimated ice crystals. A representative selection of different LOM parts from ceramic-based materials that have been presented in previous studies is shown in Figure 15. In this figure, the manufacturing methods for the obtaining the corresponding raw materials are illustrated as schematics.
3.2. Monolithic Ceramic Materials

A more detailed description of the ceramic-based parts, which are shown in Figure 15, is given in the next two sections. Significant KPIs of the parts are mentioned, as well. Several oxide and nonoxide monolithic ceramic parts were created by LOM. Zhang et al.\(^\text{123}\) obtained their finished Al\(_2\)O\(_3\) parts by sintering LOMed tape-casted multilayers with an open porosity of \(\approx 2.9\%\), a sintered density of \(\approx 3.62\ \text{g cm}^{-3}\), and a Vickers hardness of \(\approx 391\ \text{MPa}\). The flexural strength of the Al\(_2\)O\(_3\) multilayer, determined in three-point bending tests, was \(\approx 145\ \text{MPa}\) for loads in parallel to the thickness direction and \(\approx 228\ \text{MPa}\) for loads in perpendicular to the thickness direction. In addition to Al\(_2\)O\(_3\) parts, monolithic LOM SiC parts generated from LOMed preceramic polymer tapes were presented by Klosterman et al.\(^\text{28,53,80,106}\) within several studies. For the generation of curved LOM parts as viable materials for body armor plates, SiC-filled tapes were deposited onto a paper mandrel, shaped into the desired curvature. Within the studies of Klosterman et al.,\(^\text{53}\) it was noted that curvature was adapted to the part design, facilitating a later on waste removal by minimizing overlap areas. The tape casting slurry was composed of a mixture with bimodal SiC, carbon black, and graphitic powder dispersed within a polymeric binder. The binder content in the tapes was 15–20 wt\%, whereas the tape thickness was 250 mm. At first, the LOMed green bodies were pyrolyzed under pressure up to 325 °C to avoid delamination, after which the pyrolysis was completed at 700 °C in argon atmosphere. In a silicon infiltration step at 1600 °C, the pyrolyzed green bodies were converted into dense SiC parts. Flexural strength values for the monolithic SiC parts of 142–165 MPa were determined in four-point bending tests.\(^\text{53,106}\)

As a third type of monolithic ceramic, Si\(_3\)N\(_4\) was derived from LOM parts made from casted polymer tapes. In contrast to monolithic Al\(_2\)O\(_3\) or SiC, the preparation Si\(_3\)N\(_4\) requires a higher manufacturing effort. The reason for this is the complex phase diagram of Si\(_3\)N\(_4\) phases and their corresponding oxidized derivatives. Two phases of the ceramic are stable at room temperature, the trigonal \(\alpha\)-Si\(_3\)N\(_4\) phase and the hexagonal \(\beta\)-Si\(_3\)N\(_4\) phase, from which the latter phase often more desired because of its epitaxial crystal growth, and therefore, self-reinforcing effect in the Si\(_3\)N\(_4\)
microstructure. For the production of β-Si₃N₄ parts, a filler or starting power of α-Si₃N₄ intermixed with Y₂O₃, Al₂O₃ or other oxide ceramics, acting as sintering aids, is typically utilized. Upon heat treatment and sintering at 1600 °C, α-Si₃N₄ is transformed into β-Si₃N₄ phase. At this temperature, a liquid phase can be formed within the Si₃N₄ microstructure, facilitating a “dissolution–precipitation” process. Simultaneously formed or initially abundant whiskers of the β-SiAlON phase (Si₆₋₆nAl₆O₁₆N₄₋₄ᵣ, with 0 ≤ n ≤ 4.2) are promoted by sintering aids like Y₂O₃ or Al₂O₃ and act as seeds for growing hexagonal β-Si₃N₄ crystallites. During sintering, an oxidation of the fine Si₃N₄ filler powder, which degrades its sinterability, is to be avoided, preferably. In the presence of oxygen at high temperatures, Si₃N₄ is prone to be oxidized in one of two ways: either passively or actively. If Si₃N₄ oxidizes passively, whereas nitrogen gas is released and a SiO₂ layer is formed on the outside of the ceramic, which is also referred to as “scale layer.” In the active oxidation process of Si₃N₄, however, the material can decompose into gaseous SiO (silicon monoxide) and nitrogen. The active oxidation process may occur at temperatures above 1300 °C and can be assisted by a low oxygen pressure in the furnace atmosphere or the abundance of metal ions, e.g., Al³⁺ or Y³⁺ originating from the sintering aids. The aforementioned generation of β-Si₃N₄ parts from tape-casted LOM multilayers has been successfully conducted by Rodrigues et al.,[65] who tested the infiltration of the LOMed green body by a polyurea–silazane-based resin (Figure 15, on the left), and by Park and Cho,[134] who added β-Si₃N₄ whiskers to the filler powder. Both groups of authors sintered the LOMed green bodies at temperatures of at least 1750 °C and in nitrogen atmosphere. For the resulting β-Si₃N₄ parts with a density of 97%, Rodrigues et al.[65] determined Vickers hardness value of ≈14.3 GPa, which decreased to ≈13.5 GPa for the polymer-infiltrated part, a flexural strength at room temperature of ≈918 MPa, which decreased to ≈707 MPa, and, a fracture toughness of ≈7.45 MPa·m¹/₂, which decreased to ≈5.42 MPa·m¹/₂. The Vickers hardness and the fracture toughness values in parallel and in perpendicular of the layer planes were ≈13.9 and ≈14.1 GPa, respectively, whereas the fracture toughness values were ≈6.1 and ≈5.2 MPa·m¹/₂. In addition, the fabrication of Si₃N₄, Klosterman et al.[79] described the fabrication of AlN multilayer from LOM parts. As alternative sheet materials for monolithic ceramic parts, in addition to the preceramic tapes, preceramic papers were used. Travitzky et al.[74] coated Al₂O₃-filled preceramic papers with a thickness of 200 μm x 40 μm thick Al₂O₃-filled thermoplastic adhesive layer. The flexural strength values of the sintered Al₂O₃ LOM part were ≈96 MPa in parallel to the layer plane and ≈104 MPa in perpendicular to it. Other monolithic oxide ceramic materials achieved by sintering LOMed preceramic papers were β-tricalcium phosphate (β-TCP) and hydroxyapatite (HAP), ceramics similar to the ceramic component in bones. Respective β-TCP and HAP multilayers with rectangular cross-sections were built-up for testing purposes. According to Lorenz et al.,[117] these monolithic multilayers displayed physical properties which are compatible with requirements for bone replacement materials. They compared the mechanical properties of the multilayers with ranges of the Young’s modulus for trabecular bones in between 0.05 and 0.5 GPa, and, ranges of the corresponding flexural strength in between 7.6 and 20.7 MPa. The open porosity of the β-TCP multilayer was 58–64 vol%, depending on filler content of the papers in between 50 and 70 vol% and the dry-processing of the corresponding preceramic papers by calendaring prior the LOM procedure. The β-TCP multilayer offered a flexural strength of ≈7.6 MPa for a filler content of 70 vol%, whereas a Young’s modulus of ≈0.33 GPa was measured. For the HAP multilayer, the range in open porosity lay in between 51 and 56 vol%, whereas a range in Young’s moduli in between 0.75 and 1.53 GPa and a range in flexural strengths in between 20 and 28 MPa.

3.3. Glasses and Glass Ceramics

In addition to paper-derived monolithic ceramics, paper-derived glass and glass ceramics were applied in LOM to serve in biomedical and mechanical applications. In the aforementioned study on potential LOM bone replacement materials by Lorenz et al.,[117] a bioactive glass (BaG) with the following oxide composition was tested: 6 wt% Na₂O, 12 wt% K₂O, 5 wt% MgO, 20 wt% CaO, 4 wt% P₂O₅, and 53 wt% SiO₂. The corresponding multilayers from the LOM procedure and adjacent heat treatment had an open porosity in the range of 37–47 vol%, a range of Young’s moduli in between 0.33 and 0.9 GPa and a range of flexural strengths in between 20.7 and 33.1 MPa. Before BaG was applied as filler material, LZSA glass (Li₂O–ZrO₂–SiO₂–Al₂O₃) had been incorporated into preceramic tapes. A maximum filler content, at which the desired fluidity of the tape-casting slurry was still ensured has been determined to be 27 vol% (72 wt%). A minimum green tape tensile strength of ≈4.39 MPa guaranteed the applicability of the LZSA tapes in the LOM process.[84] After LOM, the LZSA tapes could be debindered at 750 °C and sintered at 850 °C, which also caused the precipitation of the crystalline phases β-spodumene (LiAlSi₃O₈) and lithium metasilicate (Li₂SiO₃). By changing the orientations of LZSA tapes within the LOM part, determined by the direction at which the tapes had been pulled in the tape-caster, the flexural strength of the sintered part could be increased from 70 to 120 MPa.[114] By an addition of ZrSiO₄ to the LZSA glass, an almost constant thermal expansion coefficient up to 600 °C could be achieved. While at porosities >10%, the flexural strength reached values up to ≈130 MPa, further process optimizations could make LOMed and sintered LZSA tapes viable components for low temperature co-fired ceramics (LTCC) in electronic parts.[109] Inversely, Schindler and Roosen[110] presented commercial LTCC green tapes as potential raw material for the LOM devices. With its SiO₂–Al₂O₃–RO composition, the LTCC green tapes required a firing temperature of 870 °C to be sintered into solid layers. The joining technique of “cold low-pressure lamination” (CLPL), with a pressure of 10–30 MPa at a temperature of 60–80 °C, which are similar parameters as for the lamination during LOM, proved to be sufficient for providing a defect-free layer bonding.

3.4. Ceramic Composite Materials

Similar to the silicon infiltration of green bodies from LOMed preceramic papers, which yielded SiC monoliths, a further liquid melt infiltration of LOM green bodies from paper-derived oxide ceramics had been successfully conducted. A paper-derived
Al₂O₃ ceramic multilayers were infiltrated by a Cu—O alloy (oxygen content: 3.2 wt%). While the preceramic paper multilayers had been debinded at temperatures up to 700 °C and sintered at 1600 °C for 1–4 h, the copper alloy infiltration was conducted at 1320 °C in an argon atmosphere at normal pressure, during a time interval of 4 h. Due to the copper alloy infiltration, a metal–ceramic composite was obtained, with an electrical conductivity of 0.6–2 MS m⁻¹, depending on the copper alloy content that ranged from 14.3 to 25.1 vol%. For increasing temperatures in between 25 and 800 °C, a decreasing thermal conductivity of 10.3–36.6 W (m·K)⁻¹ was determined for this metal–ceramic composite. Furthermore, Vickers hardness values of 0.89–1.55 GPa and fracture toughness values of 5.4–6.0 MPa m⁰.₅ were determined, again depending on copper alloy content. By combining the preceramic paper sheets, filled with Al₂O₃, with a tape-casted and fully dense Al₂O₃ ceramic layer, an Al₂O₃ FGM part with varying layer porosity was obtained after the heat treatment. The so-obtained Al₂O₃ FGM was exposed to the same copper alloy infiltration as the multilayers derived from preceramic papers only, yielding a potential electronic structure with two conductive sides and an insulating fully dense Al₂O₃ layer in between.²⁸,³⁸

Based on the same manufacturing procedure as for monolithic SiC derived from LOMed preceramic tapes, fiber-reinforced SiC/SiCm composites were created as part for aerospace applications. Other relevant applications for this material may be attracting material in fusion reactors²¹ or cladding material in fission reactors.²³,²⁴ Unidirectional prepregs of a furfural–phenolic thermosetting resin with continuous SiC fibers were used as sheet material layers carrying the unidirectional ceramic fibers. In repetitive order, a prepreg layer was cut-out and bonded onto a tape-cast SiC sheet, and then, a tape-cast SiC sheet was bonded on top of the prepreg, in the next step (manufactured in cut-then-bond method). The same heat treatment as for monolithic SiC parts from LOMed preceramic tapes, was applied on the green bodies for the SiC/SiCm composites. These green bodies were pyrolyzed a under pressure up to 325 °C and then at 700 °C in argon atmosphere, thereupon, the silicon infiltration step at 1600 °C was conducted. Other SiC-containing ceramic composites were obtained from LOMed and then pyrolyzed. For this purpose, pyrolyzed filtering papers, originally 230 μm in thickness, were covered by a polymeric slurry with phenolic resin, polyvinyl butyral, and benzyl butyl phthalate prior to lamination. The lamination was performed by LOM in a bond-then-cut method with a knife blade instead of a laser. Thereby, a phenolic resin acted as adhesive coating. The LOM parts were pyrolyzed once again and infiltrated by liquid silicon at a temperature of 1500 °C. For the resulting Si/SiC composites, a flexural strength of ≈123 MPa was measured for a 7-h long silicon infiltration and of ≈130 MPa for a 1-h long silicon infiltration. These value ranges for the flexural strength comparable with the monolithic SiC ceramics derived from LOMed tape-cast sheets.²⁸,³⁸ A higher flexural strength value increased bending strength up to ≈315 MPa was obtained by applying 240 μm-thick papers loaded with 76.8 wt% of a SiC filler. A 160 μm-thick layer of a thermosetting polymer was applied as adhesive on the SiC-filled papers, which were bonded at a temperature of 140 °C. LOMed laminates underwent a pyrolysis step and an infiltration by liquid silicon, yielding a SiC content of up to 48.6%.²⁷,³⁸ A Si/SiC gear wheel (diameter: 50 mm) with a reflecting, smooth surface finish, which had been obtained from a LOM part made from SiC-filled preceramic papers, is shown in Figure 16, placed in front of a corresponding raw material paper roll.

The complexity of material compositions has been further increased by Krinitcyn et al.,³⁸ who were successful in producing ceramic-based materials containing the MAX-phase ceramic Ti₃SiC₂. This carbide ceramic component had been synthesized by the SiC and TiC filler particles within novel preceramic tapes that were used as raw material for LOM. The tapes had thicknesses within a range of 350–550 μm. The temperature of the roller during green tape lamination was set to 80 °C, whereas the roller speed was set to 60 mm s⁻¹ and the laser cutting speed to 30 mm s⁻¹. The pyrolysis of the LOMed green tapes was conducted at 900 °C in argon atmosphere and for a duration of 1 h. The eventual sintering step was conducted at 1600 °C, during a dwelling time at 2 h, after which an option infiltration by liquid silicon at 1500 °C and under a medium vacuum of <100 Pa. An X-ray diffraction (XRD) analysis on sintered and silicon-infiltrated LOM parts revealed that a higher content of the MAX-phase Ti₃SiC₂ within the sintered ceramic was achieved within the sintered ceramic using green tapes with a higher the relative content of TiC. However, the creation of the Ti₃SiC₂ phase was accompanied by changes in volume. A defect-free gear wheel resulted from green tapes with a filler ratio of 30 vol% TiC and 70 vol% SiC, however, no Ti₃SiC₂ was abundant within this gear wheel microstructure. For this defect-free ceramic-based composite the overall shrinkage was determined to be 3% and a Young’s modulus of ≈224 GPa was determined, at a flexural strength of ≈180 MPa and a Vickers hardness (HV 10) of ≈9.8 GPa. Moreover, Travitzky et al.,³⁸ demonstrated a fabrication of a composite containing the ceramic components SiC and FeSiCr, which had been derived from with LOMed polysiloxane-based green tapes with SiC and FeSiCr fillers. Due to the polysiloxane content within the corresponding green tapes, a nitridic surface reaction layer appeared during sintering at 1400 °C in nitrogen atmosphere. The flexural strength of the composite containing SiC/FeSiCr was compared with the flexural strength of a laminated Si–SiC–SiOC–N composite which had been obtained by an SM process from a pressed multilayer of polysiloxane–polysilane-based green tapes with SiC, SiC fillers. The pyrolysis and sintering at 1400 °C were the same for the laminated Si–SiC–SiOC–N, as for a composite derived from polysiloxane/SiC/FeSiCr, resulting in a flexural strength of ≈250 MPa, counterposed to a flexural strength of ≈400 MPa for the SiC/FeSiCr composite.³⁸ The economically viability of ceramic-based composites from ZrO₂-toughened mullite and AlN layers, derived LOM green parts, has been demonstrated by Shulman and Ross³⁸ for the application as microfluidic devices.

### 3.5. Potential Future Ceramic-Based Products

In addition to the presented portfolio of ceramic-based materials derived from LOM parts, further sheet materials that matched the specifications of LOM machines, have been tested as multilayer structures. To manufacture refractory-grade CMCs, preceramic papers filled with Al₂O₃ and ZrO₂, as well as Al₂O₃ and MgAl₂O₄, have been combined with preceramic polyvinylbutyral (PVB) tapes filled with Al₂O₃, MgAl₂O₄ or MgO, and MgAl₂O₄.
A polyvinylacetate (PVA) adhesive with a ZrO$_2$ was used within interlayers for multilayer made-up of the mentioned preceramic papers and preceramic tapes, stacked in an alternating sequence. The stacking could be achieved either manually without pressure or by CLPL at pressures up to 5 kPa. The obtained FGM multilayer after debinding at up to 600 °C and sintering at 1700 °C for 5 h, could withstand a corrosion by a CaO–Fe$_2$O$_3$–SiO$_2$–slag at high temperatures up to 1390 °C$^{[113,137]}$. Further design freedom for paper-derived oxide ceramic refractories may has been attained by including Al$_2$O$_3$ or ZrO$_2$ fibers within the ceramic microstructure. Gutbrod et al.$^{[138]}$ obtained paper-derived CMCs with Al$_2$O$_3$ as matrix material and ZrO$_2$ fibers, upon sintering preceramic papers at 1600 °C for 2 h. In the corresponding preceramic papers, a content of up to 73% of the total pulp fibers had been replaced by the ZrO$_2$ fibers. The porosity in the paper-derived ceramics could be limited to a range of 28–32% by pressing the preceramic papers at 40 MPa prior to heat treatment. Without pressing, the paper-derived fiber-reinforced ceramic had porosities in the range of 34–44%. In mechanical tests, Young's moduli in between 55 and 127 GPa and moduli of rupture in between 125 and 210 MPa were determined for the paper-derived CMCs. For a different type of CMCs, paper-derived Al$_2$O$_3$ matrix ceramics with Al$_2$O$_3$ fibers, Dermeik et al.$^{[83]}$ investigated sintering strain rates and compared it with expected sintering strain rates from numerical models. The sintering strain rate at 1600 °C, determined by an optical dilatometer, was $\approx 9.4 \times 10^{-4}$ s$^{-1}$ for ceramics derived from paper with monomodal Al$_2$O$_3$ particles and Al$_2$O$_3$ fibers and was $\approx 2.3 \times 10^{-5}$ s$^{-1}$ for ceramics derived from paper with monomodal Al$_2$O$_3$ particles and Al$_2$O$_3$ fibers. The sintering densities for these paper-derived CMCs, reinforced by Al$_2$O$_3$ fibers, were $\approx 58.6\%$ and $\approx 40.8\%$, respectively.

A different type of oxide ceramic, BaTiO$_3$, had been incorporated into preceramic paper layers, so that the multilayer part after LOM, debinding and sintering had an open porosity in between 13.0% and 30.3% and ferroelectric properties. This material may be useful for underwater sonar detection or medical ultrasonic imaging. One characteristic quantity, the relative electric permittivity at room temperature varied in between 660.8 and 1514.1, increasing with increasing sintering density. The permittivity displayed increased steeply up to $\approx 4800$, at the material’s Curie temperature of $\approx 120$ °C, decreasing by a negative exponential curve at higher temperatures. The piezoelectric coefficient, which is another characteristic quantity in ultrasonic and sonar applications, was determined for orientations in parallel and in perpendicular to the layer plane. Determined piezoelectric coefficient values varied in between 39.1 and 77.3 pC/N for the direction in perpendicular to the layer plane and varied in between $-15.5$ and $-49.7$ for directions within the layer plane.$^{[119]}$ The general orientation of pulp fibers inside the previous preceramic papers with the BaTiO$_3$ filler influenced the in-layer-plane piezoelectric coefficient significantly. Thus, a finetuning of the overall
piezoelectric coefficient for parts derived from LOMed preceramic papers may be possible by to individually orienting the paper layers prior to bonding.

Further electric applications may require printed circuits inside of single layers from the CMCs derived from LOM parts. One way to attain this may be by direct ink-jet printing or screen printing a carbide ceramic slurry on preceramic papers filled with an oxide ceramic filler prior to being processed by LOM. In direct ceramic ink-jet printing (DCIJP), the desired result from direct inkjet printing may be the fine distribution and accurate application a ceramic ink. Patterns of ceramic pastes from screen printing are expected to yield a defect-free and dense printed layer on top of the paper-derived oxide ceramic. It was shown by Carrijo et al.\textsuperscript{[111,137]} that an aqueous dispersion with 0.03 vol% of Ti₃SiC₂ particles \((d_{50} = 0.16 \mu m)\), 2 wt% polyethyleneimine (PEI), and 40 wt% glycerol could serve as ceramic ink in DCIJP, and, a paste with terpinene as the solvent, 30 vol% of Ti₃SiC₂ particles \((d_{50} = 1.5 \mu m)\), 3–5 vol% of ethyl cellulose, 2 vol% of a cationic polymer dispersant could serve as ceramic ink in screen printing. While in DCIJP with the Ti₃SiC₂ dispersion, a droplet size of \(\approx 53 \mu m\) could be achieved, defect-free ceramic layers on paper-derived Al₂O₃ could be achieved after sintering screen-printed patterns at 1600 °C for 1 h in argon. The screen-printed layer density increased with increasing ethyl cellulose content within the ceramic paste. Electronically functionalized layers within CMCs derived from LOM green bodies may be classified as smart materials, similar to ceramic-based materials derived from 3D or fused freeform fabrication (FFF) green bodies, as presented by Scheithauer et al.\textsuperscript{[115]} and Abel et al.\textsuperscript{[140]} These ME processes are comparable with the screen-printed pattern on paper-derived ceramic layers, with the difference that 3D structures with a high ceramic or metallic filler content can be deposited onto preceramic papers or preceramic polymer tapes in FFF or 3DP. These FGM and metal–ceramic structures may be incorporated within future LOMed green bodies. Printed ceramic layers of the Si-O-CN system can be derived from precursor resins, such as the gelatinous ceramic polymer precursors patented by Hill and Easter.\textsuperscript{[141]} The patented resins may offer one important advantage, as opposed to highly filled polymer sheets or pastes. Gelatinous beads are embedded within the resins, which form pore channels upon pyrolysis, helping with the outgassing of combustion gases.

In addition to its application within ceramic inks, the MAX-phase ceramic Ti₃SiC₂ has been derived from preceramic papers, as well. Porous Ti₃SiC₂ parts with a high oxidation resistance up to 1100 °C, a high mechanical stability due to pseudoplasticity, as well as a good electrical and thermal conductivities, may find their applications as soot particle filters, fast moving electrical contacts, light weight heating elements, or heat exchangers. The pseudoplastic behavior, which allows for a high creep resistance and good machinability of MAX-phase ceramics in general, is enabled by the formation and movement of kink bands within the grain structure and move upon mechanical loading.\textsuperscript{[132–144]} In contrast to sintering printed powder layers of this tertiary carbide onto a fully ceramic Al₂O₃ substrate, the conversion of papers filled with Ti₃SiC₂ bears an additional challenge: the disproportionation reaction of Ti₃SiC₂ and carbon from pyrolyzed papers, yielding TiC and SiC, has to be avoided during sintering. Therefore, the first evolutionary step in the development of paper-derived Ti₃SiC₂ has been establishing successful sintering processes to acquire sufficiently pure paper-derived Ti₃SiC₂. At first, Schultheiß et al.\textsuperscript{[144]} converted papers highly filled with Ti₃SiC₂ \((d_{50} = 2.4 \mu m)\) into a ceramic with a considerable abundance of TiC₃ phases. After sintering the preceramic papers at 1600 °C in argon, a paper-derived Ti₃SiC₂ with a density of \(\approx 1.6 \text{ g cm}^{-3}\) and a flexural strength of \(\approx 12.6 \text{ MPa}\) were obtained. By calendering the preceramic papers, the sintering density of the paper-derived could be increased to \(\approx 2.9 \text{ g cm}^{-3}\) and the flexural strength to \(\approx 82.4 \text{ MPa}\). Lorenz et al.\textsuperscript{[118]} improved the purity of paper-derived Ti₃SiC₂ using a mixture of three parts each of metallic titanium \((d_{50} = 2.5 \mu m)\), TiC \((d_{50} = 2.5 \mu m)\), and elemental silicon \((d_{50} = 3 \mu m)\), as well, one part of carbon black \((d_{50} = 4 \mu m)\) as filler material. The sintering process was further developed by Kashkarov et al.\textsuperscript{[145]} and by Sedanova et al.\textsuperscript{[146]} who introduced spark plasma sintering (SPS) for preceramic papers with SiC filler and preceramic papers with Ti₃SiC₂ filler. During SPS, the papers were heated by electric pulses under a controlled atmosphere, in this case in a gasless environment, while mechanical punches exerted a uniaxial pressure onto them. Applying a pressure of 100 MPa at 2100 °C onto preceramic papers with SiC, resulted in a porosity of \(\approx 13.4\%\) and a Young’s modulus of \(\approx 292 \text{ GPa}\) for the corresponding paper-derived ceramics. For paper-derived Ti₃SiC₂, which had been subjected to a temperature of 1200 °C and a pressure of 50 MPa, a porosity of \(\approx 11.1\%\) and a Young’s modulus of \(\approx 195 \text{ GPa}\) was determined. As mentioned by Sedanova et al.\textsuperscript{[146]} one technical application for paper-derived carbide ceramics, in addition to that in soot particle filtration, may be in nuclear technologies, as the materials may be sufficiently resistant to an exposure to \(\gamma\) radiation, high-speed nuclear fission fragments and neutrons.

Preceramic papers as fabric sheet material that can be used for LOMed green bodies of ceramic-based materials, can be replaced SiC, glass, or carbon fiber tissues, as well. This has been shown already by Klosterman et al.\textsuperscript{[28,53]} as well as Ortona et al.\textsuperscript{[147]} in the case of the transformation of phenolic resin layers with SiC fibers into a SiC/SiC₃m dense or foam-like CMCs. The suitability of such CMC materials in aerospace\textsuperscript{[147]} or nuclear applications, among others for the replacement of traditional zirconium alloys in nuclear fuel rod claddings, has been discussed by Cinciz et al.\textsuperscript{[148]} and by Colombo et al.\textsuperscript{[135]} Unlike the planar- or simply curved parts already printed by LOM, the discussed radiation shielding materials would require AM parts with a rounded, cylindrical geometry. As frozen slurries are already successfully established as a possible sheet material for FS-LOM, it may be considered to apply liquid slurries at room temperature, which necessitates the addition of a hardening polymer for providing slurry layers with sufficient mechanical strength to maintain the geometric shape and to be cut. In the AM technique of layerwise slurry deposition (LSD), 20–200 µm thin slurry layers are applied by doctor blade and are dried subsequently.\textsuperscript{[149]} As Zocca et al.\textsuperscript{[136]} stated, polymer binder may be printed onto the dried slurry layers to make them more resistant against washout (LSD-print). The authors produced the green bodies for Si/SiC composites in the LSD-print method. These green bodies were pyrolyzed up to 800 °C, sintered at 1400 °C and infiltrated by liquid silicon at a higher temperature, yielding Si/SiC composites with a sintering density of above \(\approx 3.05 \text{ g cm}^{-3}\) and a maximum mechanical strength of \(\approx 479 \text{ MPa}\). For the LSD-print green bodies,
a preferential orientation of its irregular-shaped SiC particles due to the mechanical drag by the doctor blading was expected and confirmed. This mechanical orienting mechanism was often used for ME processes to deposit short fibers, as well as, in-nozzle impregnated continuous strands of fibers.\[13,151\]

4. Conclusions

4.1. Potentials of Novel LOM Machine Functionalities

LOM offers unique characteristics, not given by the other AM techniques: The SL techniques enable the rapid building of large-scaled multimaterial, FGM or CMC parts. The open working space of LOM machines enhances the multitude of possible process adjustments, as outlined by the proposed machine configuration extensions (see Section 2.3). The first generation of ceramic-based materials from LOMMed green bodies was composed of lightweight materials mainly exploited for their mechanical properties, such as structural components and body armor. Over time, variety in part designs and property profiles has significantly increased for ceramic-based materials obtained from LOM. More recent ceramic-based materials from LOM include bioactive ceramics as potential bone replacement materials, metal–ceramic composites, and ferroelectric ceramics as potential electronic components, as well as, electrically conductive carbide ceramics as potential heat exchangers, soot particle filters, or even radiation shielding. Some of the technical applications for ceramic-based composites require the incorporation of fibers into the materials, whereas others require functionalized printing patterns or 3D-ordered structures. A production of such composites, in turn, necessitates extensions in the LOM machine configuration. Upon a careful survey of the current developments in LOM technology, the following parallel processes can be identified as feasible upgrades to LOM (Figure 16): 1) a robotic arm to extract, transport, and curve or fold multilayered workpieces or prefabricated workpieces for a later LOM part; 2) an overhanging rotatable spindle for fixating freeform parts of rounded or fully round geometries; 3) a nozzle to spray UV-curing resins or adhesive layers, expanding the set of available work processes for the layer build-up within workpieces and for the coating of workpiece surfaces, or, enabling a higher number of attachment methods for workpieces, respectively; 4) a movable sand blaster for localized polishing procedures, as described by Feygin,\[102\] which promotes an increased shape accuracy for LOM parts by an immediate removal of excess material.

Some of the proposals have already been implemented within published studies. The integration of a robotic cell within LOM machines has been successfully established in the fabrication of metal foil composites and adhesive spraying nozzles have been used for papers and polymer sheets (see Section 2.3). The installation of a rotatable, overhanging spindle can open-up the opportunity to produce rounded cryogenic containers from woven fabric layers.\[152\]

4.2. Economic Viability and Market Potential

All aforementioned extensions to the LOM process may imply an upcoming leap in LOM technology. Smart ceramic-based materials from LOM with the proposed extensions may become important to an ever-growing field of technical applications. The augmenting number of technology-related standards has already led to the establishment of basic procedures for preselecting economically viable engineering tasks and the matching part designs for ceramic-based LOM parts. As Yeh and Chen\[153\] pointed out, the future demand for LOM machines, enabling the fabrication of ceramic-based materials by an AM process, depends on a few key factors related to technology, organization, environment, and cost. The aforementioned process capability for each LOM process would fall under the technology category. The organization category describes, among others, the management considerations of interested companies and their leading mindset. Furthermore, the market developments relevant to materials obtained from LOM would be assigned to the environment category. The last of the categories, costs, stands in relation to the choice of materials, and shape complexity of LOM parts. Costs involved with the part design procedure and the operational complexity of the LOM process, compared with alternative SM processes adequate for the ceramic-based materials production, may define a “frontier of convenience”: a maximum cost value for a given LOM part production volume that would break even with the costs for the alternative SM part production.\[154\] During the past decades, the focus on the LOM part types moved away from laminates of only one class of materials, such as the ceramic laminates fabricated by LOM machines (e.g., LOM machines of the former suppliers CAM-LEM Inc. and Cubic Technologies Inc.).\[156\] The more recent commercial LOM solutions target fiber-reinforced multimaterial composites, such as metal foils with fiber-reinforced polymer layers.\[40,48\] Companies, such as EnvisionTEC Inc. and Impossible Objects LLC, still active to the time this Review has been written, have promoted the more recent development of LOM parts consisting of thermoplastics and elastomers reinforced by multidirectionally oriented carbon fibers, glass fibers, or aramid fibers. In all the presented cases, the suppliers of LOM machines provided the raw materials for their machines, as well. With the growing potential of LOM to achieve ever increasingly complex part microstructures with functionally graded porosities and improved surface finishes for novel FGM and CMC products, many new fields of applications may become unlocked for ceramic-based materials from LOM: applications in medical technology, aerospace, high-performance electronics, energy generation, defense armament, and security. Therefore, upcoming market entries of new ceramic-based materials from LOM are expected to be favored by unique selling points in the production of large-scaled functionally graded multimaterials by LOM and existing synergies in between machine suppliers, product designers who are responsible for DFAM, materials science engineers and the end users of targeted products.

Dedication

The authors would like to dedicate this article to the memory of Prof. Dr. Elazar Gutmanas, a very good friend and colleague who, just two days short of his 80th birthday, sadly passed away due to a tragic laboratory accident. He will always be remembered in the Materials Science and Engineering society for his pioneering works such as “Cold Sintering” technique, “Powder Immersion Reaction Assisted Coatings” (PIRAC) or his works in the field of Self-Propagating High-Temperature Synthesis (SPS).
Conflict of Interest

The authors declare no conflict of interest.

Keywords

additive manufacturing, ceramic matrix composites, ceramic-based materials, laminated object manufacturing, paper-derived ceramics

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