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Preparation of SiC Ceramics by Laminated Object Manufacturing and Pressureless Sintering

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Abstract

In this paper, laminated object manufacturing (LOM) and pressureless sintering were applied to obtain complex-shaped SiC ceramic articles. The SiC green tapes measuring 0.15 mm in thickness used for LOM were prepared by means of organic-solvent-based tape casting. The influences of the binder content, plasticizer/binder ratio and solid loading on the tape properties were investigated and optimized. The binder removal and pressureless sintering processes were studied and optimized to obtain high-quality SiC samples. The relationships between the mechanical properties of the sintered body and the solid loading of green tape were studied. The final part with a relative density of 98.16 % can be obtained with a bending strength, hardness, toughness and elastic modulus of 402 ± 23 MPa, 19.86 ± 0.71 GPa, 3.32 ± 0.29 MPa m$^{1/2}$ and 393 ± 41 GPa respectively. The result shows that LOM is suitable for the manufacture of SiC products with complex shape and beneficial for the design and small batch fabrication of complex-shaped ceramic parts.

Keywords: LOM, SiC, tape casting, binder removal, pressureless sintering, mechanical properties

I. Introduction

Laminated object manufacturing (LOM) is a type of rapid prototyping technique [1–5]. Rapid prototyping (RP) is a new technology for automated production of complex 3D structures based on computer-aided design (CAD) files. This new technology alters the traditional "remove" method to an "add" method. Based on the RP fabrication process, it is possible to develop complicated shapes from CAD models without using any moulds or hard toolings, which significantly reduces the production time and cost. At present, RP techniques commonly include several technologies, such as SLA (stereolithography apparatus), SLS (selective laser sintering), FDM (fused deposition modelling), LOM (laminated object manufacturing) and 3DP (3D printing).

The LOM technique used paper as the starting material to manufacture models and prototypes by cutting, stacking, and bonding. This technology has been used to produce components from engineering materials such as ceramics and composites. LOM equipment is designed to deal with flat sheet materials such as tapes and papers for producing 3D objects at a high degree of automation with the aid of a CAD file.

The schematic LOM process is shown in Fig. 1 [4]. The basic machine elements consist of an elevator platform, a hot laminating roller, and a laser device. The first step is to draw a 3D CAD file, which can be converted to a standard format for manufacturing (STL file) and sliced into thin cross-sectional layers by means of appropriate software. The movement of the platform and laser device is controlled based on the sliced file. A laser beam is used to cut slices in a layer-by-layer model, then to laminate them to form samples with the required shape. After all layers have been stacked, the part block is removed from platform and any excess material is eliminated. After removal of the binder and sintering, a ceramic part with a complex shape is obtained.

There are some works in the relevant literature on preparing ceramics with LOM. Klosterman [1–2] prepared monolithic SiC ceramics and SiC/SiC ceramic matrix composites by means of LOM and reaction-bonding. During the tape lamination process, a solvent-assisted spray technique was used to improve layer adhesion and a seamless monolithic part was obtained after the LOM process. Y. Zhang [3] obtained Al_2O_3 ceramics by means of LOM and pressureless sintering, using polyvinyl acetate as paste to combine ceramic tapes. The three-point bending strength was 228 MPa. C. Gomes [6–7] prepared Li_2O-ZrO_2-SiO_2-Al_2O_3 (LZSA) glass-ceramics with LOM, to fabricate a 3-dimensional gear wheel geometry without distortion and cracking after heat treatment. A. Das [8] has studied the binder removal process in ceramic thick shapes made with LOM. B. Bitterlich *et al.* reported on the LOM of silicon nitride ceramics, a paste containing ceramic powder and precursor was developed and screen-

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printed to the tapes. After lamination of the silicon nitride tapes in a pressureless process, components with complex geometries can be produced [9]. S. Liu and F. Ye obtained components with complex shapes by means of a LOM process and subsequent pressureless sintering. The bending strength of the sintered silicon nitride bodies reached about 475 ± 34 MPa [10].

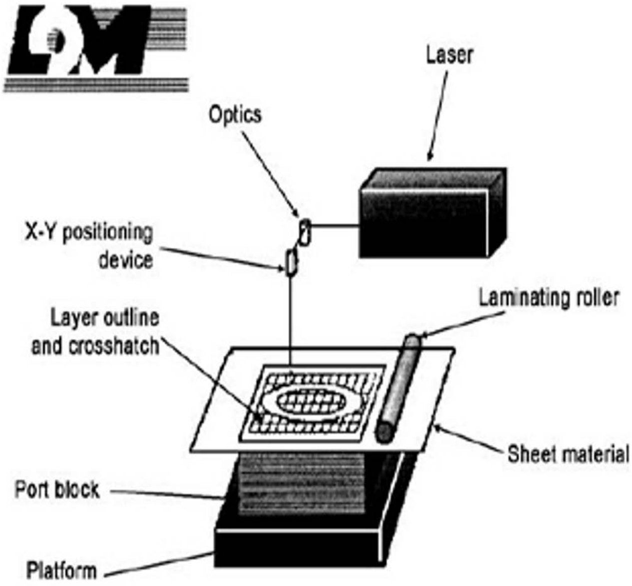

Fig. 1 : Schematic diagram showing the LOM process.

The binder removal process is quite important for LOM processing. In general, the binder removal process for green parts was designed based on TG-DTA analysis [3] or on the theoretical analysis of diffusion models [8], which cannot ensure that there is no delamination or distortion in application. So, a novel, more practical method of debinding needed to be put forward. In addition, the development of ceramic articles with comparable mechanical properties to those made with a conventional sintering method was important. In previous reports, SiC ceramic articles were obtained by means of LOM and reaction bonding [1, 2], the 4-point bending strength of SiC specimens was about 150 MPa, which was lower than expected for this material system (400 MPa). In addition, the existence of residual silicon reduced the application range of the articles. Research on preparation of SiC ceramic articles with LOM and a pressureless sintering process has not been reported.

Good properties of laminated sheet material can ensure high bonding strength between the layers and reduce the processing time; the sheet materials are therefore important for LOM technology. Tape casting is a well-accepted method for fabricating ceramic green tape [11–14]. Basically, it consists of the preparation of a suspension of a ceramic powder in a solvent, with the addition of dispersant, binder and plasticizer. Organic-solvent-based tape casting systems are widely used mainly because improved quality of tapes can be obtained and because of easy and fast evaporation of solvents during the drying stage.

The objective of this work is to develop geometrically complex ceramic objects suitable for engineering applications directly with the LOM method and pressureless sintering. Non-aqueous tape casting was used to prepare SiC green tapes for LOM processing. The slurry properties were studied and optimized, the relationship between the slurry processing parameters and the properties of the green tapes was investigated. The relationships between the mechanical properties of the sintered body and the green tape properties were also studied.

II. Experimental Procedures

(1) Preparation of the SiC tapes

The quality of the green tapes determines most of the properties of the final product. Thus, green SiC tapes with high homogeneity were fabricated using a standard tape casting technique, tailored for the LOM process [15]. A high-purity, submicron alpha-phase silicon carbide powder (UFP PA, purity 99 %, mean particle size 0.5 μm, SYCERA New Materials Co., Ltd., Shanghai, China, D50 = 0.56 μm) along with boron carbide (purity 97 %, particle size 1.5 ¦Im, Mudanjiang Jingangzuan Boron Carbide Co., Ltd., PR China) and carbon black as sintering aids were used in this study to prepare the SiC tapes. The azeotropic mixture of butanone/alcohol at relative concentrations of 66/34 was selected as the solvent system, in order to avoid differential evaporation. The dispersant used for stabilizing the SiC suspensions was PVP (Polyvinyl Pyrrolidone). The optimum amount of this was 2 wt% relative to the SiC powders. Commercial PVB (Polyvinyl Butyral) and BBP (Butyl Benzyl Phthalate) were used as binder and plasticizer respectively. The binder and plasticizer content was optimized. A SiC suspension containing ceramic powders at approximately 23 vol% was prepared by dispersing ceramic powders in solvent systems with the dispersant for 24 h, then the binder and plasticizer were added and milled further for another 24 h. Rheological characterization of suspensions with different solid loading was performed using the parallel-plate system on a Universal Stress Rheometer SR5 (Rheometric Scientific, USA) at various shear rates. Finally, tape casting was performed on Procast Precision Tape Casting Equipment (Division of the International, Inc., Ringoes, NJ) with a gap height of 400 mm, at a speed of 100 mm/min. The green tape was stripped from the support polyester film after casting and dried for about 5 h.

(2) Processing with LOM

Ceramic tapes were laminated to form three-dimensional objects using a LOM machine (HRP-2, Wuhan Binhu Mechanical and Electrical Technology Co., Ltd.). Placement of tapes was performed manually. Before the process, the key machine parameters including the laser power, cutting speed, roller temperature, and roller speed were optimized to achieve the required results. In our experiment, a retract of 0.08 mm was used, which was related to the distance between the roller and the tape surface. Generally, the lower the retraction, the higher the pressure applied. The laser power was about 10 W. The cutting and roller speeds were 50 and 25 mm/s respectively. The roller temperature was kept constant at 60 °C. A self-made mixed solution, phenolic resin (Shandong Saint Spring Chemical Industry Co., Ltd.)/alcohol (volume ratio was 6:1), was used as adhesive agent to bind the ceramic tapes. Here, the low difference in weight loss between the adhesive and ceramic tapes gives rise to relatively small dimensional changes during pyrolysis. The pyrolysis residue of the adhesive ensures interfacial bonding of the pyrolyzed ceramic tapes. A professional airbrush was used to apply a fine spray of adhesive agent to the richest organic side of each tape (the side in contact with the support polyester film of tape casting equipment) before the subsequent one was stacked. Unlike with the solvent-assisted spray technique, which blurred the layer boundaries, with the adhesive agent, the interfaces remained relatively strong and distinct: phenolic resin was uniformly dissolved in alcohol, then sprayed onto the tape surface. The alcohol evaporated quickly because of high vapour pressure, leaving phenolic resin with a sufficient viscidity to bond the different tapes. The pyrolysis residue of phenolic resin ensures interfacial bonding of the pyrolyzed ceramic tapes. During the LOM process, it was thought that the interlaminar strength must be sufficiently weak for decubing but strong enough to provide acceptable strength in the final structure, which needs optimization of the machine parameters and the amount of adhesive agent sprayed with the airbrush [16, 17]. All these were determined by trial and error before the experiments. After all layers have been stacked, the part block is removed from the platform and any excess material is eliminated.

(3) Thermal treatment

Tape casting is a binder-assisted forming technique that requires the addition of a high volume percentage (often above 50 vol %) of organics prior to shape forming. Early practice revealed that the laminates were prone to delamination during the pyrolysis cycle. So, for defect-free manufacturing of complex shapes, the removal of such a substantial volume of organic processing aids from the green body is a very critical step [8, 18]. TG analysis of SiC blocks made with LOM was performed in Ar atmosphere in order to optimize the burnout cycle. In addition, because TG analysis cannot ensure that there was no delamination in the process, a novel, more practical method for optimizing the debinding process was put forward. To simulate the ideal state of weight loss, a round "seamless" SiC ceramic block (3 cm in diameter, 25 layers) was prepared at 80 °C and 5 MPa in an oil press. Then, it was used to measure weight loss and dimensional change under different temperatures, applied in a vacuum debinding furnace. At the end of every holding stage, the thickness of the round SiC ceramic block was measured, in order to ensure that there was no delamination during the previous pyrolysis process. It is generally believed that when the rate of decomposition of organic matter exceeds the rate of gas diffusion, delamination will occur, and the thickness will increase correspondingly. At the end of every holding stage, when the thickness no longer increases and there is almost no weight loss, it is possible to move to another holding stage at relatively higher temperature. Otherwise, the previous pyrolysis process should be repeated again. After debinding, the samples were further sintered by pressureless sintering in Ar atmosphere.

(4) Microstructure, mechanical and other properties

The density and the apparent porosity of the sintered samples were measured using the Archimedean method. The microstructures of both the green tape and the sintered body were observed with scanning electron microscopy (SEM, TM-1000, Japan). Tensile testing of the green tapes was performed at a constant load speed of 10 mm/min at a span length of 20 mm in a computer-controlled INSTRON universal testing machine (Model 5566, Instron Co., High Wycombe, UK). The three-point flexural strengths of sintered rectangular specimens (3.0 mm × 4.0 mm × 36.0 mm) were measured using a span width of 30 mm and a crosshead speed of 0.5 mm/min (universal testing machine, Model 5566, Instron Co.). Hardness and toughness were measured in an indentation test on a Wilson-Wolpert Tukon 2100B (Instron), the load and holding time were 49 N and 10 s respectively.

III. Results and Discussion

(1) Characterization of slurry and green tapes

In tape casting, high solid loading is required in order to obtain green tapes with high green density. However, a suspension with enough fluidity is required to ensure that green tapes have a uniform structure and only few defects such as bubbles and cracks. Therefore, it is important to obtain high solid-loading slurry with good fluidity. The influence of solid loading on the flow behaviour of SiC slurry is shown in Fig. 2. Here, the binder content was kept constant at 6.06 wt% relative to all components and the plasticizer/binder ratio was fixed at 1.2.

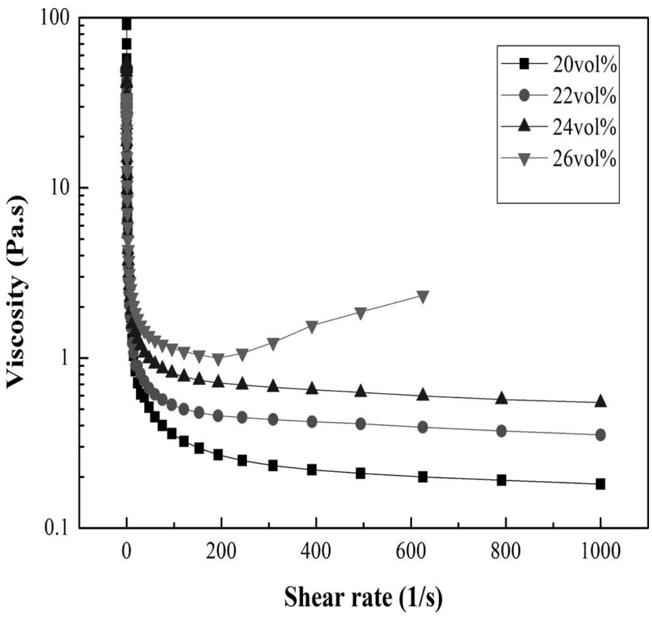

Fig. 2: Rheological behaviour of SiC slurry with different solid loading.

It can be seen that the slurries containing 20, 22 and 24 vol% SiC powder show shear thinning behaviour, which is suitable for tape casting. At lower solid loading (less than 18 vol%), the suspension viscosity was too low and high volumetric shrinkage can occur. With the increase of solid loading to 26 vol%, the slurry immediately shows a shear thickening behaviour, indicating that SiC particles in these slurries are in agglomerated state. When the solid loading is above 26 vol%, the slurry is too viscous to measure its rheology behaviour. In our experiments, 19 ~ 25 vol% SiC slurries were used for further tape casting and the LOM process. In order to study the relationship between slurry composition and the quality of SiC tapes, suspensions were prepared with 15 different compositions based on a change in the solid loading, binder content and plasticizer/binder ratio. First, the solid loading was kept constant at 20 vol% and the plasticizer/binder ratio was fixed at 1.2, the binder content was 4.92, 5.49, 6.06, 6.63 and 7.17 wt%, respectively. Fig. 3 shows the green density and thickness shrinkage (A); tensile strength and strain at failure (B) of green tapes with different binder contents.

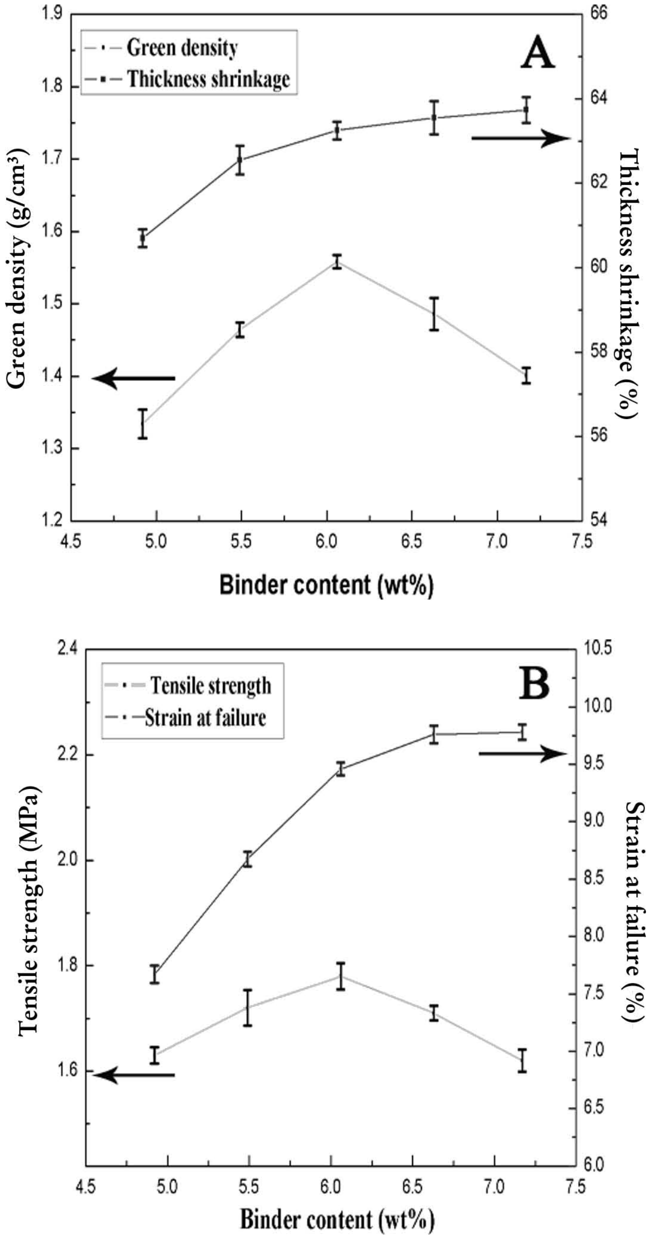

Fig. 3: (A) Green density and thickness shrinkage and (B) tensile strength and strain at failure of green tapes with the change of binder content.

From Fig. 3(A), it can be seen that as the binder content increases, initially the density of the green tapes increases, reaching its high point at the binder content of 6.06 wt.% and then decreasing. The tape thickness decreases continuously with the binder content. This might suggest that a binder content above 6.06 wt% is too much and might separate ceramic particles from each other, leading to the decrease in the tape density.

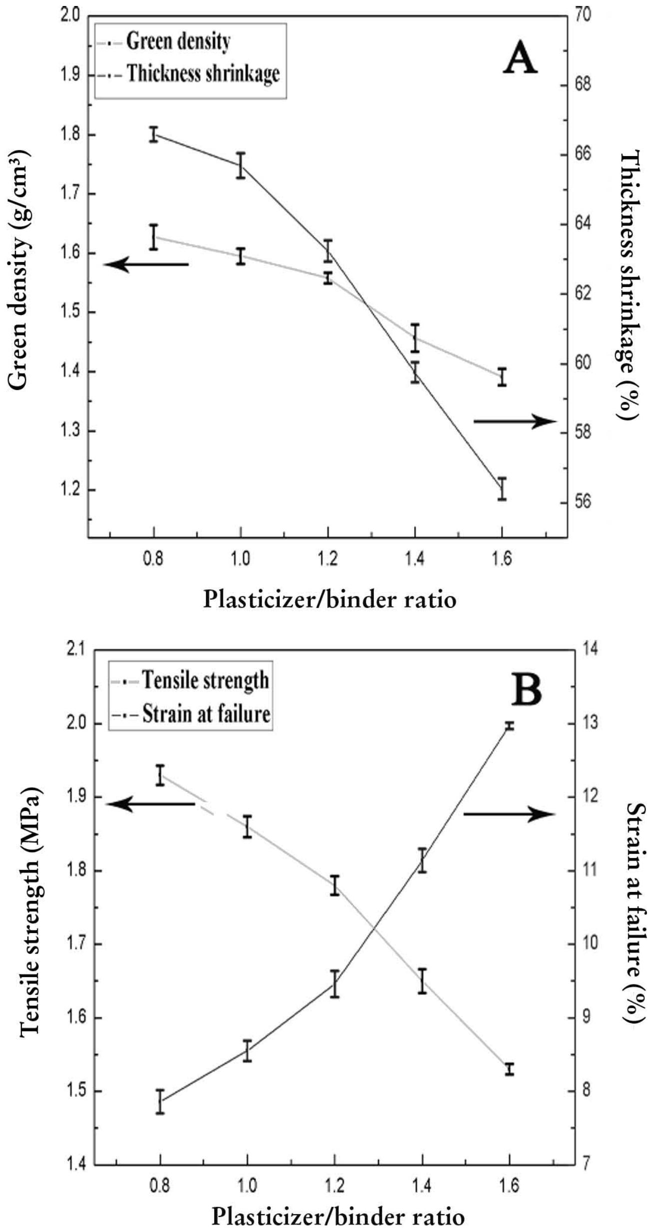

Fig. 4: (A) Green density and thickness shrinkage and (B) tensile strength and strain at failure of green tapes with the change of plasticizer/binder ratio.

Fig. 3(B) shows the variation of the tensile strength and strain at failure of tapes with different binder contents. It can be seen that with the increase of binder content, strain at failure increases continuously, but slows down after the binder content is higher than 6.06 wt.%. While the tensile strength shows the same trend as the green density of the sheets. In consideration of the relative density, strength and flexibility, the binder content was selected at 6.06 wt% for the green sheet preparation.

Fig. 4 shows the green density and thickness shrinkage (A); tensile strength and strain at failure (B) of green tapes with the different plasticizer/binder ratio. The solid loading was kept constant at 20 vol% and the binder content was fixed at 6.06 wt%, the plasticizer/binder ratio was selected as 0.8, 1.0, 1.2, 1.4 and 1.6, respectively. It can be seen that with the increase of the plasticizer/binder ratio, the green density and thickness shrinkage decrease gradually. This can be corresponded to the decrease in strength and the increase in strain at failure owing to the nature of plasticizer. The addition of plasticizer could effectively reduce the PVB glass transition temperature. The low molecular weight of BBP could improve the flexibility but reduced the strength of tapes. In consideration of these comprehensive factors, in order to guarantee enough flexibility and strength of green tape, the final plasticizer/binder ratio was selected at 1.2.

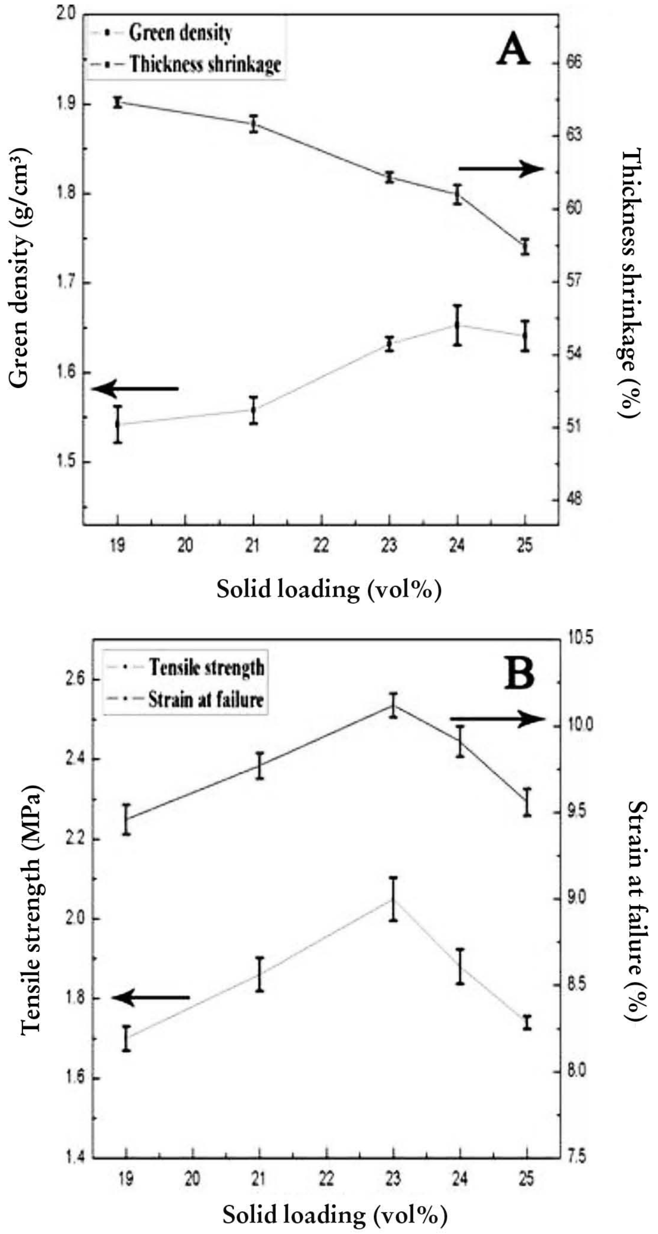

Fig. 5: (A) Green density and thickness shrinkage and (B) tensile strength and strain at failure of green tapes with different solid loading.

Fig. 5 shows the green density and thickness shrinkage (A); tensile strength and strain at failure (B) of green tapes with the different solid loadings. The binder content was fixed as 6.06 wt%, the plasticizer/binder ratio was kept constant at 1.2, the solid loading was 19, 21, 23, 24 and 25 vol%, respectively.

It can be seen from Fig. 5(A) that with the increase of solid loading, the amount of ceramic powders in unit volume increases, resulting in the increase in density. But the density decreases when the solid content is higher than 24 vol%, maybe due to the higher slurry viscosity and agglomerated state of the powders. On the other hand, with the increase of solid loading, tensile strength and strain at failure both reach an optimal value at about 23 vol%, then decrease thereafter, may be due to tiny internal defects result from the inhomogeneous state. Practice shows that slurry with about 6.06 wt% binder (relative to all components), 1.2 for plasticizer/binder ratio at the solid loading of 23 vol% is suitable for tape casting. Tapes produced with this composition possess superior comprehensive performance, green density, thickness shrinkage, tensile strength and strain at failure, which are appropriate for LOM process: tapes can withstand the temperature and pressure applied by the hot laminating roller without cracking, are suitable for laser cutting, facilitate waste stripping after the LOM process, and possess good dimensional stability after debinding and sintering. The tapes were homogeneous in their microstructure, Fig. 6.

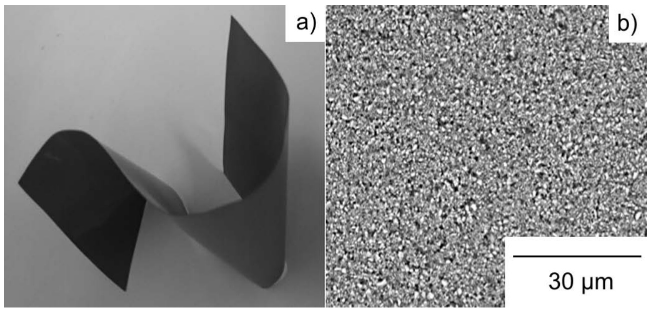

Fig. 6: (A) The flexible tape with optimized component and (B) the SEM image of the upper surface.

(2) Binder removal and sintering process

As there are quite a lot of organic elements in the green tapes, the binder removal process without delamination presents a problem. During the binder removal process, as the temperature increased, the organic component decomposed and was released from the samples. Practice has proven that the laminates were prone to delamination during the pyrolysis cycle. This was mostly because the rate of decomposition of organic matter exceeded the rate of gas diffusion, the gas cannot be released on time, resulting in the delamination problem [6, 7, 19]. TG analysis (performed in Ar) of the SiC block made by LOM is shown in Fig. 7. Fig. 8 shows weight loss of the round crack-free SiC ceramic block at different temperatures under well controlled conditions. Fig. 9 shows the average thickness (three different points) of the block at the end of every heat preservation stage.

Fig. 7 and Fig. 8 show roughly the same trend, but there are two obvious differences between them. First, compared with Fig. 7, the weight loss showed in Fig. 8 moves as a whole towards low temperature; second, the TG curve shows that there was only about 1.5 % of weight loss before 180 °C, whereas Fig. 8 shows a considerable weight loss before 180 °C (more than 15 %), and with a high weight loss rate. The cause of the difference lies in that TG was performed at a relatively fast heating rate (10 K/min), which cannot guarantee that the organic matter decomposes completely; while the weight loss in Fig. 8 for round SiC disks at different temperatures was quite slow and near the equilibrium condition. The method of developing an expedient heating cycle for LOM samples according to the weight loss curve of the round seamless SiC ceramic block is a method of general, which is a good solution to the delamination problem. From Fig. 9, it can be seen that when the temperature was between 80 and 150 °C, larger volume expansion occurred and the thickness was almost the same from 200 to 300 °C, then, thickness shrunk significantly after 300 °C. It was proposed to optimize the heating cycle by carefully measuring the weight loss and dimensional change during binder removal at quite a low heating rate with a long holding time at different temperature steps. It was found that at the lower temperature range (50 ~ 150 °C), samples should be placed in the oven with a long enough holding time, in order to eliminate the residual mechanical and thermal stress imparted during the original LOM cycle. To eliminate the delamination problem in our experiment, in addition to the expedient temperature schedule, another method is to put samples inside sand to ensure a homogenous temperature and to apply a certain amount of pressure so as to maintain their shape during the pyrolysis step. After the binder removal process, the samples remained the same shape and size as the green parts without distortion and cracking. Fig. 10(A) shows an SEM micrograph of the fracture surface of SiC ceramic made with LOM after debinding, it can be seen that the SiC ceramic layers made by LOM were bonded to each other tightly without delamination after the binder removal process. Fig. 10(B) and Fig. 10(C) show SEM micrographs of the fracture surface of SiC ceramic made by means of LOM after sintering. It can be seen that after sintering, the gaps between the layers disappear (B), and the structure of the final product is uniform (C).

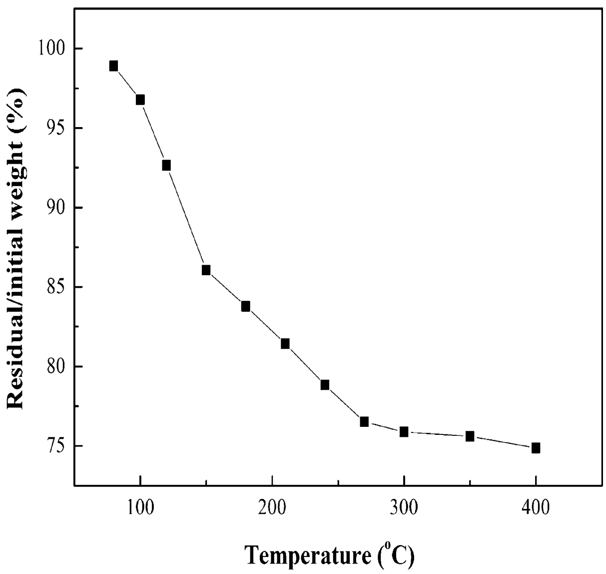

Fig. 8: Weight loss of the round "seamless" SiC ceramic block at different temperatures without delamination.

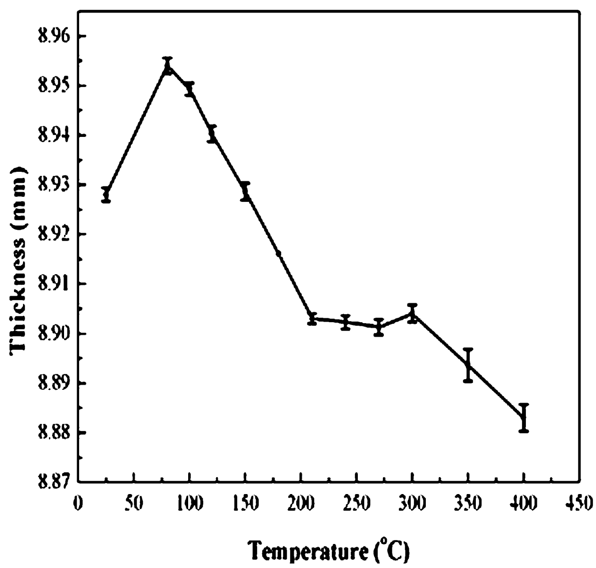

Fig. 9: Average thickness of the round "seamless" SiC ceramic block at the end of every heat preservation stage.

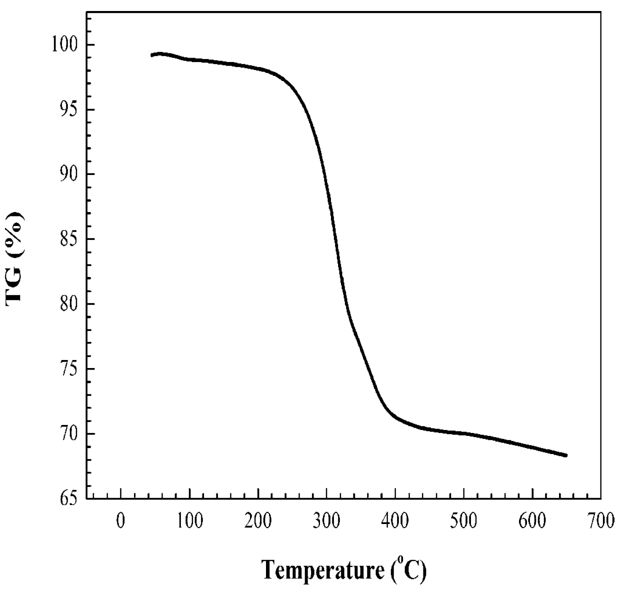

Fig. 7: TG analysis (performed in Ar) of a SiC block made with LOM.

At high temperatures during the sintering process, there was mass diffusion among the ceramic grains, which caused larger volume shrinkage (about 30 %) after sintering. The higher the solid content, namely the lower the organic matter, the smaller volume shrinkage caused by mass diffusion. Therefore solid content should be increased as high as possible so as to ensure that slurry exhibits appropriate rheological behaviour. Distortion and cracking often occurred during the sintering process, especially for large parts during the cooling process. Therefore, the rate of heating and cooling should be controlled and optimized.

(3) Mechanical properties

Tapes with different solid loading (21, 22, 23 and 24 vol%) were laminated by means of LOM, a retract of 0.08 mm being used. The mechanical properties are shown in Table 1 (all the results are the average of five samples), it can be seen clearly that tapes with a solid loading of 23 vol% and 24 vol% are suitable for LOM. It was shown that the mechanical properties of ceramics made of tapes with 23 vol% solid loading were optimal compared with those of other samples. The final part with a relative density at 98.16 % and a bending strength of 402 ± 23 MPa was obtained. The hardness, toughness and elastic modulus were 19.86 ± 0.71 GPa, 3.32 ± 0.29 MPa $m^{1/2}$ and 393 ± 41 GPa respectively. The mechanical properties of SiC ceramics prepared by means of LOM with a pressureless sintering process were comparable to those of SiC products prepared with the conventional forming process. LOM is indeed suitable for the manufacture of ceramic products with complicated shapes and is beneficial for the design and small batch fabrication of complex-shaped ceramic parts because no mould is required.

It is well known that the microstructural differences in the tapes and microstructural defects between the tapes can lead to direction-dependent properties [9]. The mechanical properties parallel and perpendicular to the stacking directions were measured and are shown in Table 2. ("23vol—parallel" means the samples fabricated from tapes with 23 vol% solid loading laminated with LOM and tested in the direction parallel to the stacking direction). The strength shows a significant difference depending on the direction of testing: Taking "23vol", for example, the flexural strength decreases from 408 ± 16 MPa (parallel) to 314 ± 48 MPa (perpendicular), similarly the fractural toughness decreases from 3.86 ± 0.17 MPa $m^{1/2}$ (parallel) to 3.48 ± 0.28 MPa $m^{1/2}$ (perpendicular). The mechanical properties in the stacking direction were significantly higher than those in other directions, which may be due to the layered structure though no obvious interface was observed between the individual sintered layers.

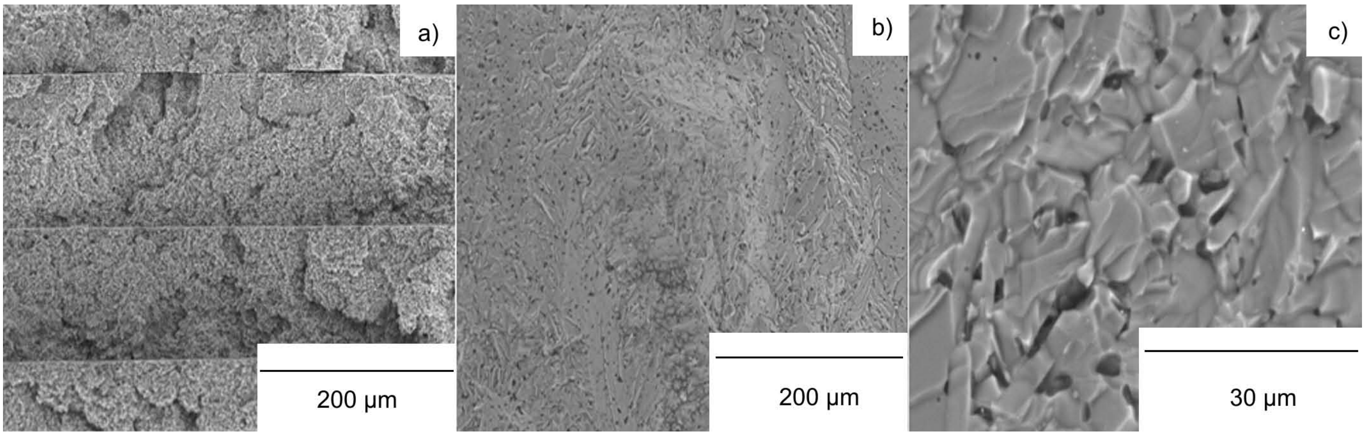

Fig. 10 : SEM image of fracture surface of SiC ceramics made by means of LOM (A) after debinding, and (B) (C) after sintering.

Table 1: The mechanical properties of sintered bodies fabricated from tapes with different slurry solid loading (21, 22, 23 and 24 vol%) laminated by means of LOM with a retract of 0.08 mm.

Solid loading (vol%)	Sintered density (g/cm³)	Relative density (%)	Porosity (%)	Flexural strength (MPa)	Vickers hardness (GPa)	Fractural toughness (MPa $m^{1/2}$)	Elastic modulus (GPa)
21	3.06	97.21	0.30	343 ± 14	16.87 ± 0.45	2.48 ± 0.36	337 ± 26
22	3.07	97.53	0.28	350 ± 43	17.23 ± 0.54	2.74 ± 0.21	355 ± 46
23	3.09	98.16	0.16	402 ± 23	19.86 ± 0.71	3.32 ± 0.29	393 ± 41
24	3.08	97.84	0.20	397 ± 65	19.07 ± 0.46	2.56 ± 0.34	396 ± 16

Table 2: The mechanical properties of sintered bodies depending on the different direction of testing (parallel and perpendicular to the stacking direction).

Sample number	Flexural strength (MPa)	Vickers Hardness (GPa)	Fractural toughness (MPa $m^{1/2}$)	Elastic modulus (GPa)
23vol—parallel	408 ± 16	19.67 ± 0.26	3.86 ± 0.17	394 ± 12
23vol—perpendicular	314 ± 48	19.23 ± 0.43	3.48 ± 0.28	315 ± 43
24vol—parallel	390 ± 69	19.78 ± 0.71	3.55 ± 0.46	403 ± 28
24vol—perpendicular	327 ± 35	19.16 ± 0.65	3.37 ± 0.21	348 ± 19

Fig. 11 presents two different gear wheels fabricated by means of LOM from tapes with the optimized components after heat treatment. Each part consists of approximately 80 laminate tapes. After sintering, the linear shrinkage in the stacking direction (20 %) was less than that in the perpendicular direction (25 %), which might be due to the orientation during tape casting and the LOM process. The initial size of the sample can be designed in advance according to the difference in shrinkage rate. The sintered sample kept the curved edges and internal profile without distortion and cracking after heat treatment.

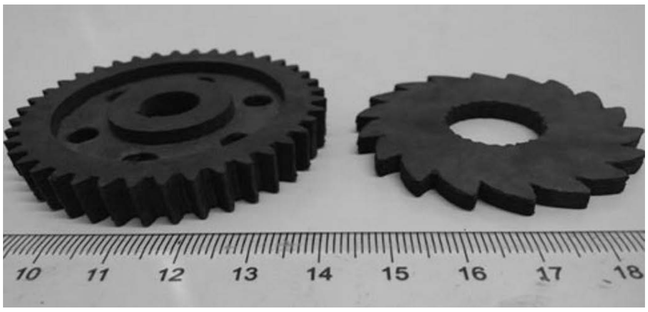

Fig. 11 : The 3-dimensional-shape gear wheels fabricated by means of LOM after heat treatment.

IV. Conclusions

SiC ceramics were prepared by means of the LOM technique and pressureless sintering. The tape casting slurry formulation was optimized and SiC green sheets suitable for LOM process were prepared. After binder removal and pressureless sintering, SiC parts with relative density of 98.16 % could be fabricated. Results showed that LOM is suitable for the manufacture of SiC products with complicated shapes and is beneficial for the design and small batch fabrication of complex-shaped ceramic parts because no mould is required.

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