SLM OF NET-SHAPED HIGH STRENGTH CERAMICS: NEW OPPORTUNITIES FOR PRODUCING DENTAL RESTORATIONS

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Abstract

Oxide ceramics yield excellent mechanical properties along with outstanding thermal and wear resistance. However, little work on additive Manufacturing (AM) of high-strength ceramics has been stated. In the present paper the current state of development in Selective Laser Melting (SLM) of pure ceramic specimens is reviewed. During the present approach the eutectic mixture of pure alumina (Al_2O_3) and zirconia (ZrO_2) powder is completely molten while crack formation is prevented by a high-temperature CO_2-laser preheating. This approach yields net-shaped, fully dense specimens reaching flexural strengths of above 500 MPa without post-processing. One potential application for this technology are fully-ceramic dental restorations frameworks, as the demanded maximum loads of above 1000 N are met. Alternative preheating strategies are presented to allow for manufacturing larger volumetric parts.

Introduction

Ceramics exhibit superior material strength, combined with high wear resistance and outstanding thermal and chemical resistivity which qualify this material for a growing range of applications. As conventional manufacturing technologies yield limited complexity and are costly due to high tool wear, attempts for rapid manufacturing, either indirectly[1-3] by additively manufacturing molds for gelcasting or directly[4,5] by utilizing laser processing routes have been conducted. However, all approaches request post-processing for reaching complete densification. Recent research has shown, that Selective Laser Melting (SLM) offers the possibility to manufacture fully dense, high-strength, complex, net-shaped oxide ceramics made from an alumina (Al_2O_3) / zirconia (ZrO_2) mixture using a high-temperature CO_2 laser-preheating[6-8]. Applications for this new technology comprise high-tech engineering ceramics, exploiting the possibilities of manufacturing complex geometries for the automotive or aerospace sector. Further applications can be found in the medical sector as SLM offers a cost-effective possibility to manufacture truly customized parts in lot sizes of one without any tool wear. Consequently, the feasibility of producing fully ceramic frameworks for dental restorations by SLM has been investigated and yielded promising results[9]. However, substantial deficits in view on surface qualities and dimensional accuracies persist. Furthermore, larger volumetric SLM parts of 10 x 10 x 10 mm^3 exhibit crack formation, which limits the technology to thin-walled specimens. Therefore, the present paper gives a review on the present research towards improving the surface roughness and introduces new preheating strategies, which ought to overcome limitations of the present setup in terms of functionality and cost-effectiveness.

SLM for Ceramics

Basic Principle

SLM is a powder-based AM technique during which functional parts are manufactured layer-wise using series material. In a repeating process (fig. 1 (a)) powder layers of less than 100 µm in thickness are deposited onto a substrate plate and selectively molten according to CAD data. Subsequent lowering of the building platform, followed by additional powder deposition provides new material for a consecutive melting step. These repeating steps allow for generation of complex, net-shaped parts. The consolidation during SLM is based on a complete melting of the powder layer alongside with the surface of the previous layer (fig. 1 (b)) yielding parts of up to 100% density for a broad range of applicable materials.

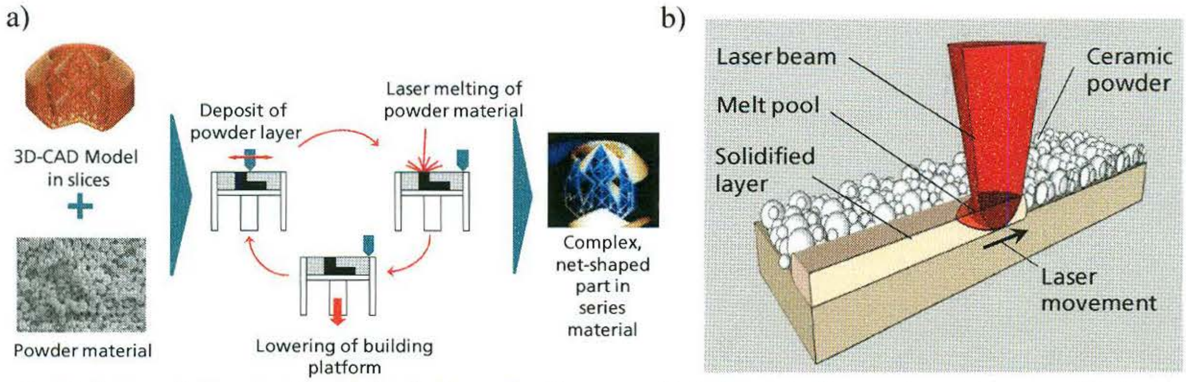

Fig. 1: (a) Schematic illustration of the SLM production steps (b) Schematic illustration of the consolidation process

The local heat input by the focused laser during SLM causes large thermal gradients accompanied by local stresses. As ceramics show little potential for plastic deformation due to the covalent ionic bonding, these stresses may not be relieved. Formation of micro-cracks throughout the SLM processed material limiting flexural strengths to ~ 10 MPa is the result. Employment of a high-temperature preheating reduces thermal gradients during laser processing and consequently crack formation may be avoided[6-8].

Experimental Setup

The present experimental setup of the SLM process for ceramics is depicted in Fig. 2[7]. In addition to the conventional SLM setup a high-temperature preheating area of 30 x 40 mm² is established by a CO_2 laser (TRUMPF TLF 12000), let through homogenization optics. The build area of the powder bed is completely covered by the preheating area and thereby heated to temperatures above 1700 °C, not exceeding the melting point of the ceramic material. The selective and complete melting of the powder material is achieved by a continuous wave (cw) Nd:YAG laser beam (ILT production), which is let through a scanner (SCANLAB Hurry Scan 20). The spot size of the focussed laser beam is 200 µm with a top hat intensity profile. The temperature control of the preheating area is done with a two-colour pyrometer (KELLER Cella Temp PZ40AF7). Thermal images are taken with a thermal camera (JENOPTIC Vario Therm Head) and the according software (INFRATEC Irbis Professional). High-speed videography of the melting process is conducted with the MV-D1024 – 160 (Photonfocus) at a resolution of 256 x 256 pixels and a framerate of 2200 images per second.

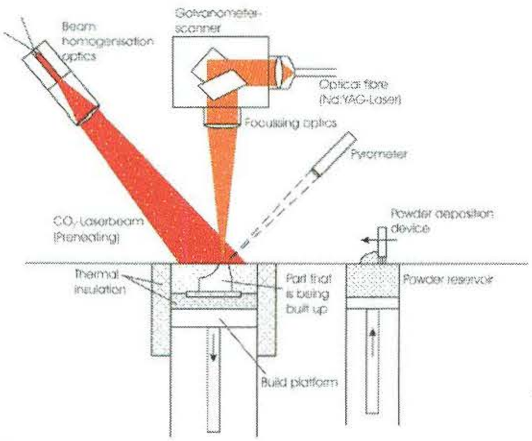

Fig. 2. Schematic illustration of the experimental setup of SLM for ceramics using a high-temperature CO_2 laser-preheating [7]

Utilized Material

The density of the SLM part strongly depends on the applied powder. Influencing factors are the particles' densities, shape, size and size distribution[7]. In fig. 3 (a) the rough alumina and the smooth zirconia particles of the drily mixed powder are depicted. The zirconia component is partly stabilized by 3 mol-% yttria (Y_2O_3). Both powders were supplied by INNALOX bv, Netherlands and do not contain any binder nor any other additives. All particles are dense and therefore no gas is trapped inside the SLM part while the particles are molten. The powder deposition is positively influenced by the monomodalic size distribution of 50 µm along with the spherical shape of each particle which also reduces the disposition to sintering while deposited onto the preheated area. All experiments are conducted using the eutectic material ratio of 58.5 wt.-% alumina and 41.5 wt.-% zirconia. This material ratio exhibits a considerably lower melting temperature of 1860 °C as compared to the single phases (fig. 3 (b))[10].

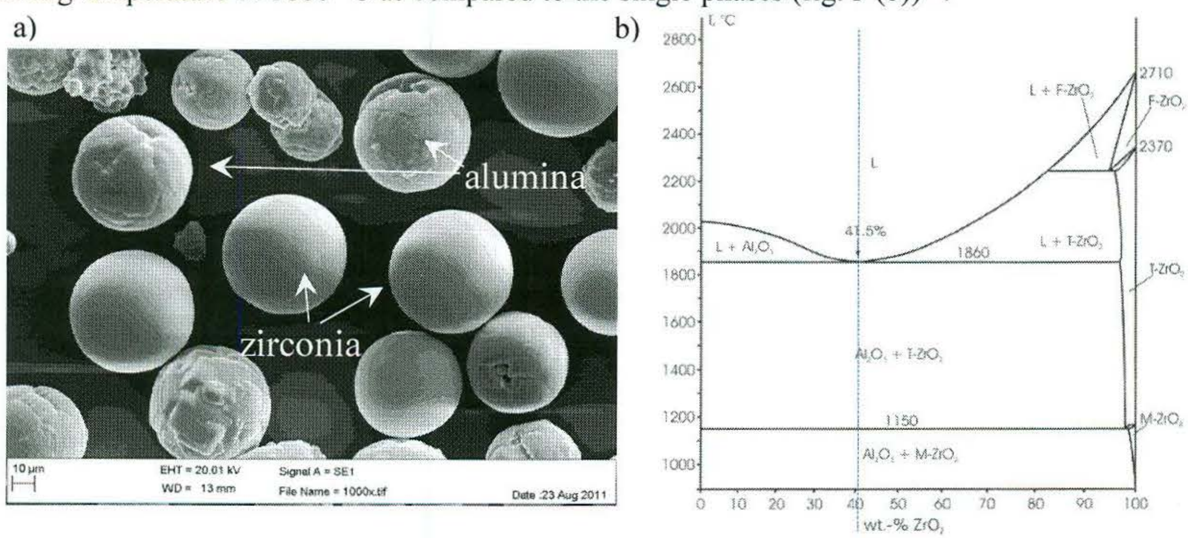

Fig. 3: (a) SEM image of the dry-mixed alumina / zirconia powder (b) Phase diagram of the system alumina and zirconia[10]

Consequently, preheating temperatures, diminishing thermal gradients during SLM may be kept below 1800 °C. Furthermore, simultaneous crystallization during solidification at this material ratio yields a fine-grained microstructure[6] potentially allowing for superior mechanical properties due to grain boundary strengthening.

Experimental Conduction

The process parameters comprise a preheating temperature of 1730 °C, a layer thickness of 50 µm, a scanning velocity of 200 mm/s, a laser power of 60 W and a scanning offset of 50 µm. Due to the preheating temperatures close to the melting point of the eutectic powder ratio (1860 °C) a large melt pool evolves, which, on the one hand, positively influences the density of the obtained part. On the other hand, a negative influence on the surface quality is examined since the low viscous melt pool exceeds the boundaries of the scanned part and wets the surrounding powder. Fig. 4 (a) shows a thermal image of the SLM process[7]. The solid SLM part is fully covered by the preheating area and exhibits a different thermal radiation coefficient than the surrounding powder. Also depicted are the laser spot and the melt pool, which is flowing into the surrounding powder, exceeding the boundaries of the specimen. Improving surface qualities, accordingly, necessitates a control of the melt pool size at the immediate surface. This may be achieved by utilizing altered process parameters for the contour and introducing a hatching delay between two subsequent scans, allowing for solidification of precedent scanning tracks. This strategy considerably decreases the melt pool size. Typically, these delays are chosen to be < 100 ms as each delay increases the manufacturing time of the specimen. Utilization of high-speed videography allows for analyzing the evolution of the melt pool and allows for improving surface qualities. Fig. 4 (b) shows an online process image of the SLM process taken by high-speed videography. For improving the surface qualities, a contour / hatch scanning strategy during which the contour is scanned first was chosen. By doing so, the contour forms a barrier which keeps the melt pool of the hatch from exceeding the specimen's boundaries. The contour was scanned at a scanning velocity of 120 mm/s and a laser power of 40 W resulting in an even, dense melt track of < 300 µm in width.

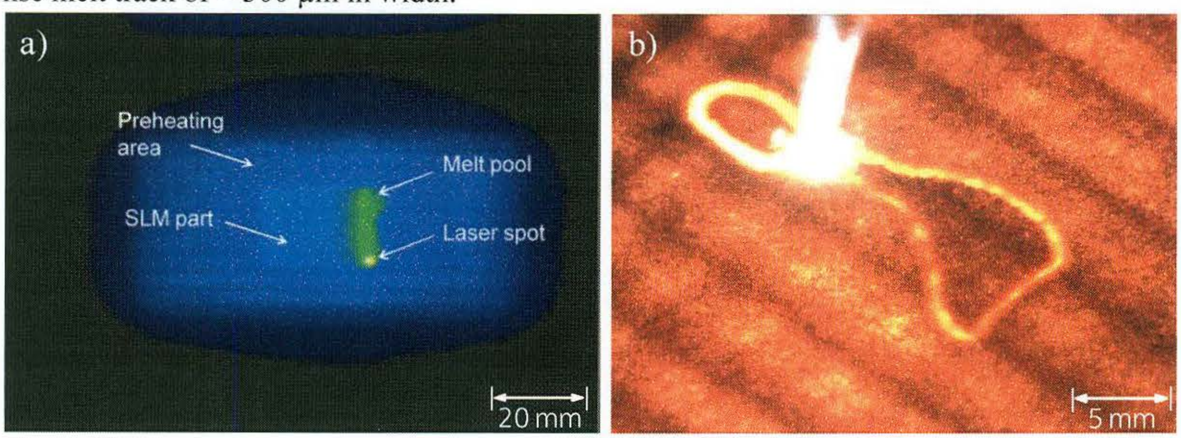

Fig. 4: (a) Thermal image for the SLM process of ceramics[7] (b) Process image taken by high-speed videography

Results and Discussion

Density and Crack Formation

Using the above mentioned process parameters yield fully dense, crack-free specimens as depicted in fig. 5[7]. The specimen has the dimensions of ø 18 mm x 2.5 mm and the ceramic material is fully molten. The surface quality of the specimen's boundaries is poor, as the low-viscous melt pool exceeds the specimen's boundaries and wets the surrounding powder. (see section Experimental Conduction).

Fig. 5: Polished cross section of an alumina / zirconia compound, manufactured by SLM. Dimensions: ø 18 mm x 2.5 mm[7]

Larger volumetric parts of 10 x 10 x 10 mm^3 yield severe crack formation, as depicted in fig. 6 (a). This phenomenon is explained by thermal gradients over the heights of the SLM part, as the high-temperature CO_2 laser-preheating is applied in a top-down manner. FEM simulation as depicted in fig. 6 (b) shows the temperature distribution of a SLM part during high temperature preheating. As the material's absorbance is also considered, the maximum temperatures are observed right beneath the surface of the preheated powder bed. For reasons of experimental control of the simulation's accuracy, preheating temperatures of 1600 °C have been chosen as this represents the maximum working temperature of the applied thermal sensors which have been incorporated into the SLM part. At these temperatures, a temperature loss of 400 °C is observed over the height of a 10 mm specimen. A bottom up preheating (see section Alternative Preheating Strategies) bears the potential of reducing thermal gradients and, thus, allow for manufacturing larger volumetric parts.

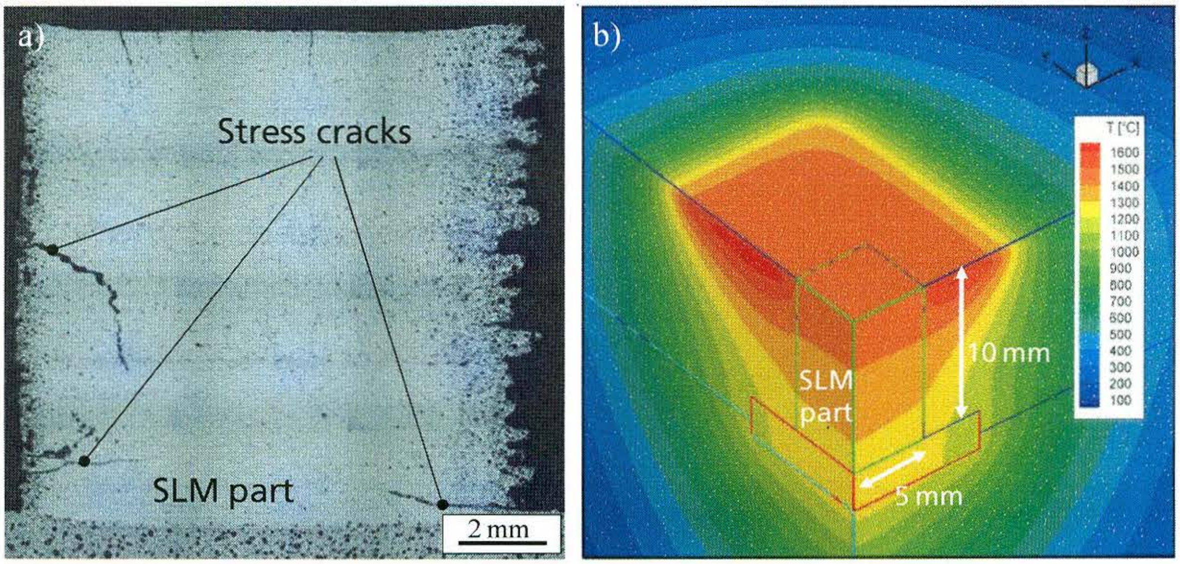

Fig. 6: (a) Polished cross section of an alumina / zirconia compound, manufactured by SLM. Dimensions: 10 x 10 x 10 mm³ (b) FEM simulation of the temperature distribution during top down high-temperature CO_2 laser-preheating

Mechanical Properties

Testing of the mechanical properties of the ceramic specimens has been conducted by BEGO Bremer Goldschlägerei (GmbH). The flexural strength of the SLM specimens has been tested according to DIN EN ISO 6872:2009 – 01 (dentistry – ceramic materials) as depicted in fig. 7 (a). Specimens of ø 18 mm x 2.5 mm have been manufactured, removed from the substrate and tested with the last solidified SLM layer facing downwards, yielding flexural strength of above 500 MPa. According to the quoted standard this strength is sufficient for manufacturing three-unit dental restorations for the posterior region. However, analyses on ceramic bars manufactured from the same material and resolidified after melting reveal a potential flexural strength of above 1600 MPa[11]. To which extend, the testing setup, surface qualities or phase transformation of the zirconia phase influence the strength of the SLM part is subject to present research.

Additionally, three-unit dental restorations have been built up via SLM and tested according to the testing setup in fig. 7 (b)[9]. Two frameworks were tested and yielded a maximum load of 1435 N. This value exceeds the required initial minimum load for all-ceramic three-unit dental restorations of 1000 N as postulated by literature[12].

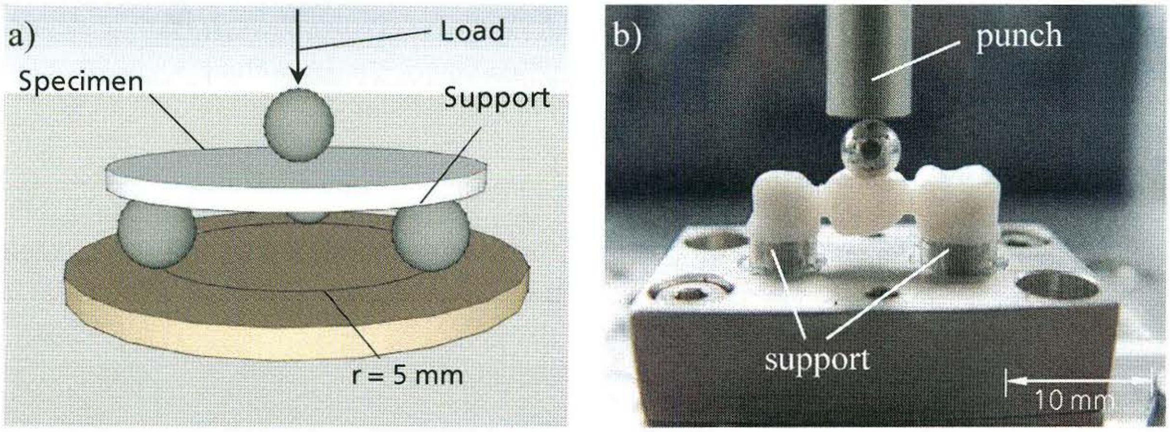

Fig. 7: (a) Testing setup for the flexural strength of ceramic specimens according to DIN EN ISO 6872:2009 – 01 (dentistry – ceramic materials) (b) Testing setup for the maximum load of three-unit dental restoration frameworks[9].

Dimensional Accuracy and Surface Roughness

As high preheating temperatures cause a large melt pool size, poor surface qualities are encountered for ceramic specimens (see section Experimental Conduction). This has a decisive impact on the mechanical strength as rough surfaces may induce stress peaks upon mechanical loading which result in premature failure. Furthermore, dimensional accuracies of the specimens are limited to the surface roughness. In fig. 8 a target-performance comparison of dental restoration frameworks manufactured conventionally (top) and by SLM (bottom) in view of the corresponding CAD data is illustrated. Accordingly, the dimensional accuracy of the conventionally milled part yields ~ 50 µm, while the SLM part reaches ~ 150 µm. This value is not sufficient for the final application as cavities may occur after insertion. Consequently, an improvement of the maximum surface roughness to values of Rz ~ 50 µm is demanded.

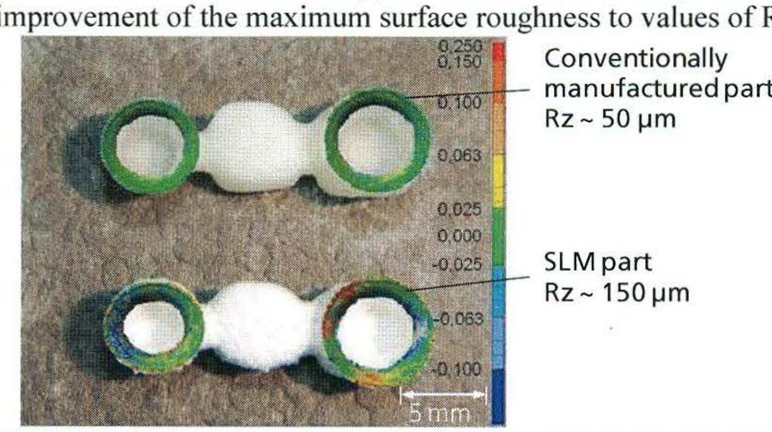

Fig. 8: Target-performance comparison of a conventional manufactured dental restoration framework (top) and a SLM framework (bottom)

In fig. 9 (a) a topographic measurement of the surface quality of a vertical structure (fig. 9 (b)) yielding a maximum surface roughness of Rz ~ 60 µm is depicted. The surface roughness may be further improved by decreasing the particle size from 50 µm to 30 µm.

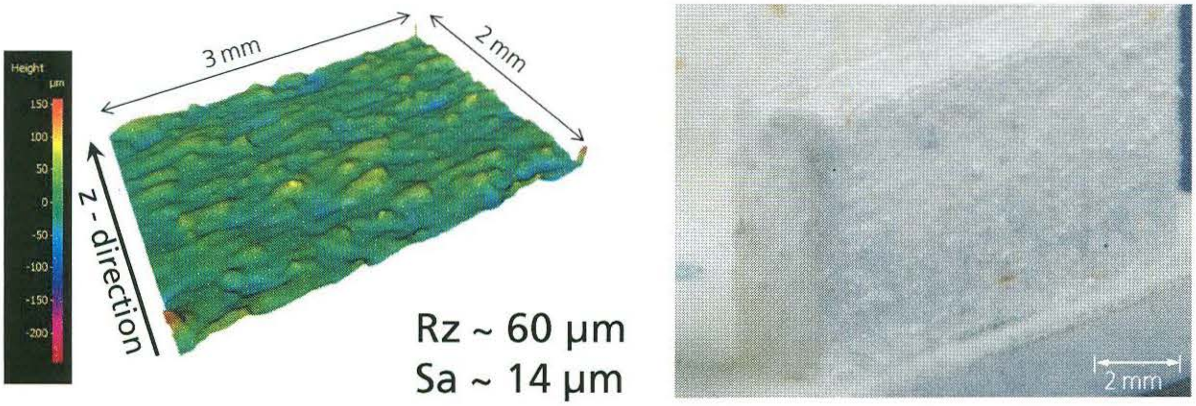

Fig. 9: (a) Topographic measurement of the surface quality of a vertical ceramic structure (b) Corresponding single track vertically ceramic structure

The surface quality of a SLM part is also influenced by the actual contour, as the powder surrounding the solid SLM part yields altered thermal conduction, which again influences the size of the melt pool. As a consequence, surface qualities of complex parts are limited and may be improved by an online process control, monitoring and diminishing the size of the melt pool. The setup of such process control requires an in-depth understanding of the complex mechanisms behind SLM for an extensive range of different materials and is subject to present research.

Demonstration Object

In fig. 10 a demonstration object of a fully ceramic dental restoration is depicted[9]. The maximum surface roughness was measured to reach Rz ~ 100 µm and represents the present state of the art in processing high-strength oxide ceramics via SLM.

Fig. 10: Demonstration object of a fully ceramic dental restoration framework[9]

Alternative Preheating Strategies

As larger specimens with a height > 3 mm may not be processed by the present setup (see section Density and Crack Formation) an alternative experimental setup has been established. Thermal gradients over the part's height are diminished by employing a bottom up preheating embodied by an inductive heating (fig. 11 (a)). As oxide ceramics may not be heated inductively due to insufficient electrical conductivity, a metallic substrate mount is inductively heated, which again heats the ceramic part conductively. The setup of the inductive heating is depicted in fig. 11 (b). Active water cooling is essential for protecting the surrounding machinery equipment from overheating. A thermal sensor measures the induced temperature in the substrate mount and allows for online controlling of the temperature.

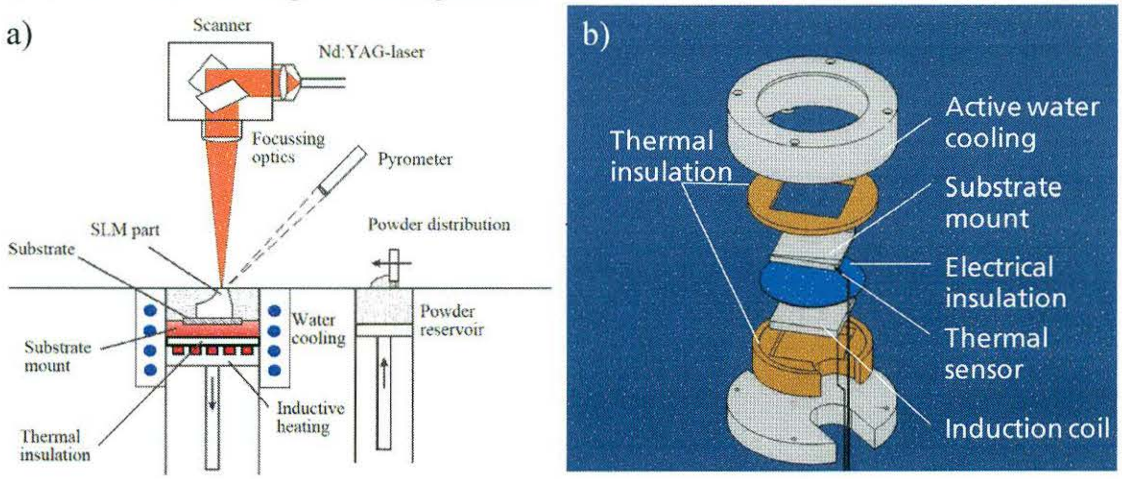

Fig. 11: (a) Schematic illustration of the experimental setup of SLM for ceramics using an inductive preheating (b) Schematic illustration of the induction heating

The electrical energy for the inductive heating is provided by a 7 kW generator (ELVA Minimac). The present setup allows for preheating temperatures of 1400 °C in the substrate mount, reaching temperatures of ~ 1200 °C on the surface of the ceramic specimens. The limited maximum temperature arises from the tendency of oxidization of the metallic substrate mount. As most refractory metals form a porous oxide layer upon extensive heating which does not protect the remaining material. As a consequence, the present substrate mount is manufactured from chromium as this metal forms a dense oxide layer. Still, chemical reaction between the chromium and the ceramic powder prevent reaching temperatures > 1400 °C. In fig. 12 (a) a photographic illustration of the SLM process while preheated in a bottom up manner by inductive preheating is given. The ceramic substrate shows severe crack formation after selective melting. This phenomenon is due to insufficient preheating temperatures. Consequently, efforts are undertaken to increase the preheating temperature while keeping oxidization of the metallic substrate to a minimum. Towards this end, two strategies exist:

1. Employment of an encapsulated process chamber, allowing for utilization of inert gas and, thus, protecting the metallic substrate from oxidization. Following this strategy, preheating temperatures of above 1700 °C have been achieved, as the melting temperature of chromium lays at ~ 1907 °C[13].
2. Utilization of an alternative material for the substrate mount, capable of reaching high temperatures, while electrically conducting and resistant to oxidization. One potential material for this application is molybdenum disilicide ($MoSi_2$) utilized for high temperature heating elements, capable of reaching temperatures of up to 1900 °C.

Both approaches are presently investigated.

In addition to the inductive preheating a selective preheating strategy is depicted in fig. 12 (b). This setup foresees employment of two laser sources, in which a fiber laser with a focused spot size of ~ 200 μm is utilized for selective melting while a diode laser-beam is coaxially superpositioned by a dichroidic beam splitter, forming a selective preheating with a focused spot-size of ~ 5 mm. This setup aims at supplying preheating temperatures in the immediate proximity of the molten material. A combination of inductive preheating and selective preheating decreases thermal gradients, while being suited for producing larger specimens which exceed the dimensions of the present setup using static CO_2 laser-preheating.

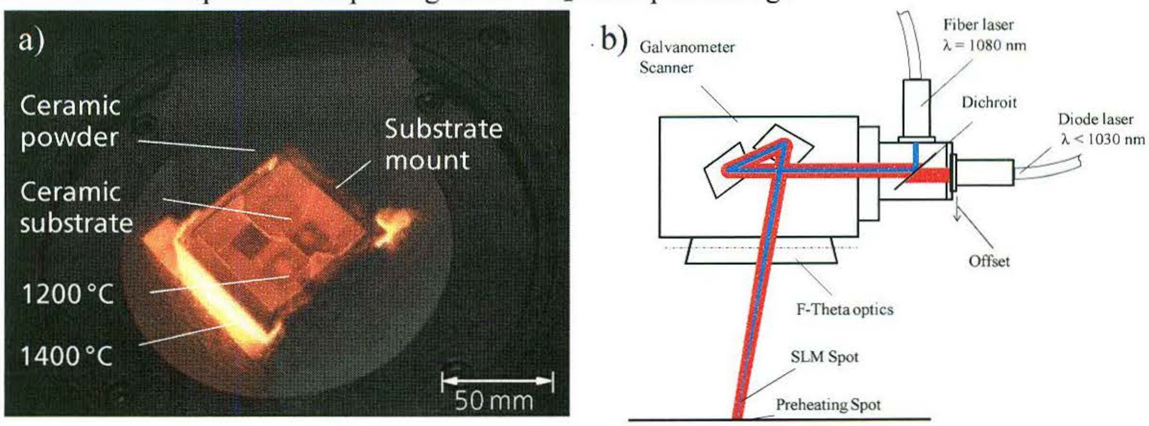

Fig. 12: (a) Photographic illustration of the SLM process for ceramics using bottom up inductive preheating (b) Schematic illustration of the setup for selective preheating using a diode laser.

Thus, this advanced preheating strategy bears the potential of overcoming present drawbacks and consequently allows for manufacturing larger specimens as demanded for by various applications for engineering disciplines and medical implants.

Conclusion

The present paper covers the current state of the art in processing alumina / zirconia oxide ceramic compounds by means of SLM. Fully dense specimens, yielding flexural strengths of above 500 MPa are produced. The qualification for the technology for producing dental restorations is shown as minimal loads of above 1000 N for three-unit dental restoration frameworks are exceeded. The influence of the surface quality of the specimens on the mechanical strength and the fit is discussed and strategies for decreasing the surface roughness are presented.

In the second part of the paper, alternative strategies for preheating aiming at allowing for manufacturing larger ceramic specimens with dimensions above 10 x 10 x 10 mm³ are presented. These strategies foresee utilization of a bottom up inductive preheating which may decrease thermal gradients over the heights of the part, avoiding formation of cracks. Additionally, a selective laser preheating concept is introduced. This setup allows for reaching the desired preheating temperatures while scanning and should, therefore, allow for manufacturing specimens exceeding the dimensional limitations of the present static CO_2 laser-preheating.

References

1 J. Stampfl, H.C. Liu. Rapid Prototyping and Manufacturing by Gelcasting of Metallic and Ceramic Slurries. *Materials Science and Engineering* 334 187 – 192, 2002

2 H. Wu, L. Dichen. Rapid Fabrication of Alumina-based Ceramic Cores for Gas Turbine Blades by Stereolithography and Gelcasting. *Journal of Materials Processing Technology* 209 5886 – 5891, 2009

3 A. Bertsch, S. Jiguet, P. Renaud. Microfabrication of Ceramic Components by Microstereolithography. *Journal of Micromechanics and Microengineering* 14 197 – 203, 2004

4 P. Bertrand, F. Bayle, C. Combe, P. Goeuriot, I. Smurov. Ceramic Components Manufacturing by Selective Laser Sintering. *Applied Surface Science* 254 989 – 992, 2007

5 I. Shishkovsky, I. Yadroitsev, P. Bertrand, I. Smurov. Alumina-Zirconium Ceramics Synthesis by Selective Laser Sintering/Melting. *Applied Surface Science* 254 966 – 970, 2007

6 J. Wilkes, Y.-C. Hagedorn, S. Ocylok, W. Meiners and K. Wissenbach. *Rapid Manufacturing of Ceramic Parts by Selective Laser Melting*, 137 – 148. John Wiley & Sons, Inc., 2010.

7 J. Wilkes, Selektives Laserschmelzen zur generativen Herstellung von Bauteilen aus hochfester Oxidkeramik, RWTH Aachen, 2009

8 Y.-C. Hagedorn, J. Wilkes, W. Meiners, K. Wissenbach, R. Poprawe. Net – Shaped High Performance Oxide Ceramic Parts by Selective Laser Melting. *Physics Procedia*, 5 (2) 587 – 594, 2010

9 Y.-C. Hagedorn, S. Dierkes. Generative Manufacturing of All-Ceramic Frameworks. *Digital Dental News*, 5 10 – 22, 2011

10 S. M. Lakiza and L. M. Lopato. Stable and Metastable Phase Relations in the System Alumina-Zirconia-Yttria. *Journal of the American Ceramic Society*, 80 (4) 893 – 902, 1997

11 J. Y. Pastor, J. Llorca, P. Poza, I. de Francisco, R. Merino, J.I. Peña. Mechanical Properties of Melt-Grown Al_2O_3-$ZrO_2(Y_2O_3)$ Eutectics With Different Microstructure. *J. Eur. Ceram. Soc.* 25 1215 – 1223, 2005

12 J. Tinschert, G. Natt, B. Doose, H. Fischer, R. Marx. Seitenzahnbrücken aus Hochfester Strukturkeramik. *Dtsch. Zahnärztl. Z.* 54 545-550, 1990

13 M. Hämäläinen, K. Jääskeläinen, R. Luoma, M. Nuotio, P. Taskinen, O. Teppo. A Thermodynamic Analysis of the Binary Alloy Systems Cu-Cr, Cu-Nb and Cu-V. *Calphad* 14 125 – 137, 1990