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Process strategies on laser-based melting of glass powder

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Abstract

This paper presents the laser-based powder bed fusion (L-PBF) using various glass powders (borosilicate and quartz glass). Compared to metals, these require adapted process strategies. First, the glass powders were characterized with regard to their material properties and their processability in the powder bed. This was followed by investigations of the melting behavior of the glass powders with different laser wavelengths (10.6 µm, 1070 nm). In particular, the experimental setup of a CO_2 laser was adapted for the processing of glass powder. An experimental setup with integrated coaxial temperature measurement/control and an inductively heatable build platform was created. This allowed the L-PBF process to be carried out at the transformation temperature of the glasses. Furthermore, the component's material quality was analyzed on three-dimensional test specimen with regard to porosity, roughness, density and geometrical accuracy in order to evaluate the developed L-PBF parameters and to open up possible applications.

Keywords: 3D-printing; glass; additive manufactureing; laser based powder fusion;

1. Introduction

The laser-based powder bed fusion (L-PBF) process offers an alternative to already established manufacturing processes due to the comparable component properties, the freedom in geometric design and the production costs in small and medium series [1]. The focus of research work, especially for metallic powders, is on process and parameter development (such as laser power, scanning speed, focus position,

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focus diameter, track pitch/hatch and exposure strategy) as well as investigations into the influence on static and dynamic strength. More recent systematic investigations are concerned with the properties of the starting material, as these have a significant influence on process control and the resulting component properties. Only in this way, material and application-specific processing concepts can be successfully implemented. [3]

L-PBF is already an industrially established process for the manufacturing of three-dimensional components from polymers and metallic materials. Sintering of ceramics and glass is increasingly being investigated [4]. Laser sintering of glass powders is an effective technology for manufacturing products for micro-electromechanical systems or objects for medical applications [5]. Glass is a material with a wide variety of compositions. The characteristics of glasses can be adjusted very variably, which enables many applications of this material group [6]. The conventional production of glass powders involves the steps of melting the glass composition from a batch, glass fritting, grinding and sieving the glass powders into the required particle size and particle size distribution. This produces non-spherical, irregularly shaped particles. In addition to the geometric properties (particle size, particle shape) of the glass powders, the mechanical-physical properties (e.g. flowability and absorption/transmission versus the laser wavelength) are essential for processing by means of powder bed fusion with laser beams. Furthermore, the process control has to be adapted to the thermophysical properties of the glasses such as poor heat conduction or low thermal shock resistance depending on the thermal expansion coefficients.

2. Fundamentals

The absorption of the laser radiation in the powder provides the energy required to melt the powder by means of L-PBF. According to [7], the energy absorption on or in a powder layer is in general significantly higher compared to absorption on a compact solid of the same material. For different one-component powders, N. Tolochko carried out investigations on absorption with two different wavelengths [8]. In the investigations laser beams from a Nd:YAG and a CO_2 laser source were coupled into an integrating sphere. For SiO_2 glass powder an absorption for $\lambda = 1.06\ \mu m$ of $A = 0.04$ and for $\lambda = 10.6\ \mu m$ of $A = 0.96$ was determined. For a SiO_2 glass component, comparable absorption and transmission values are shown. On the other hand, compact soda-lime or borosilicate glasses, have a higher absorption for wavelengths smaller than 2 µm, due to their material composition, which probably also applies to the absorption for these glass powders.

Compared to crystalline solids (e.g. metals), solid glass components are amorphous. The glassy state is classically described as the frozen state of a super cooled liquid, whose property is characterized by the temperature-dependent viscosity behavior. Depending on the glass composition, characteristic fix-points [11] must be observed throughout the entire temperature and viscosity curve during glass production and glass processing. A simple method for observing this temperature-dependent behavior can be by means of high-temperature microscopy (HTM) images of glass powder compacts. The values thus determined provide information for conducting the L-PBF process.

Defined coating of the powders by means of a rubber lip or a metal blade on a building platform and their defined lowering after laser irradiation (exposure) perform the building process in L-PBF. The processability of powders during coating depends essentially on particle shape, particle size and particle size distribution. The processability can be determined experimentally by technological parameters bulk density and flow behavior. Bulk density directly relates to the compaction of a powder, i. e. how strongly it is compressed (solidification stress). As the solidification stress increases, the bulk density increases and the void volume between the particles decreases. With fine-grained bulk solids, the bulk density is usually more strongly influenced by the solidification stress. The flow properties of powder particles also depend on the particle shape, particle size distribution, the chemical composition of the powder particles, but also on the temperature as well as on the humidity. The particle shape has a decisive influence on the flow behavior of a powder. In theory, smooth, round particles larger than 0.5 mm flow more easily than rough, spherical particles. The adhesive forces between the particles are also responsible for the flow behavior of powders [2]. Due to their larger specific surface area, finer powders exhibit higher adhesive forces and show lower values of flowability [9]. The particle size distribution, in addition to the compaction and flow of a powder, also has a significant influence on the sintering or melting behavior in the L-PBF process. A homogeneous powder mass distribution ensures a homogeneous energy input during laser exposure and thus a homogeneous melting of the material. For this reason, a constant particle size should be achieved. In practice, an average diameter with a certain deviation is usually achievable [10].

3. Experimental investigations and results

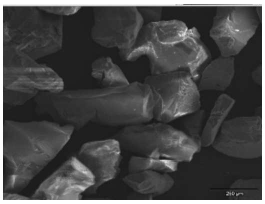
3.1. Characterization of the glass powders

Various glass powders with different compositions and geometric, thermo- and mechanical-physical properties were available for the investigations. First, an extensive analysis of the initial state of the glass powders was carried out to select glass powders with good processing properties for the experimental investigations in the L-PBF process. The following methods were used to determine the properties:

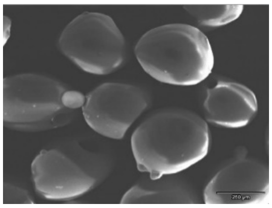
- Geometric properties
  - Powder geometry (Grain shape microscopically using a scanning electron microscope (REM) JSM-6300 with energy dispersive X-ray spectroscopy (EDX) (Fa. JEOL, Japan; EDX: NORAN Instruments, USA)
  - Particle distribution by means of laser diffraction LS230 (Fa. Beckman Coulter)
- Mechanical and physical properties
  - Flowability (according to DIN EN ISO 4490) with Hall flowmeter
  - Tab density (according to DIN EN ISO 3953)
  - Coating quality manually by means of defined doctor blade and
  - Coating system SLM 50 von ReaLizer
- Thermo-physical properties
  - Temperature-dependent viscosity behavior using a high-temperature microscope (HTM)
- Optical properties
  - Absorption measurement

Based on these investigations, two SiO_2 glass powders and one borosilicate glass powder were selected for their technological suitability. These are SiO_2 powders from the company Qsil with the designation "NC4A" (GP4) as well as a powder from the company Heraeus with the designation "Zandosil" (GP6). Both powders consist of more than 99.999 % silicon dioxide and have a low coefficient of expansion of $\alpha = 0{,}5*10^{-6}\,K^{-1}$. The manufacturer specifies further thermal properties for GP4 as follows: softening limit 1730 °C, transformation range from 1075 °C to 1210 °C, processing range from 1700 °C to 2100 °C. The thermal conductivity of this material is 1.38 W/m*K and is significantly lower than that of most metall. For powder materials, the thermal conductivities are correspondingly lower up to a factor of 100 [12].

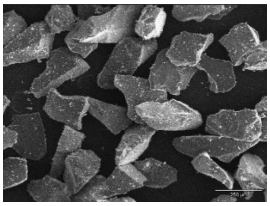
A borosilicate glass powder (GP10) from the company Schott AG is used, which was specially ground from borosilicate flat glass "BOROFLOAT® 33". The manufacturer specifies the characteristic viscosity dependent fix points as follows: Working Point (104 dPa*s) at 1270 °C, Softening Point (107.6 dPa*s) at 820 °C, transformation temperature (T_g) at 525 °C. The specific thermal conductivity of the material is 1.2 W/m*K and the thermal expansion coefficient of $\alpha = 3.25*10^{-6}\,K^{-1}$ is higher compared to SiO_2 and thermal stresses can be reduced less easily.



GP4, broken grain, angular and angular, no fines



GP6, round grain



GP10, broken grain, various angular geometries, high fine grain content

Fig. 1. REM images of various glass powders

Glass powders are usually present as broken grains due to their production by crushing and grinding (Fig. 1). Spherical glass powders can be produced by using complex technologies and are therefore more expensive and not available for all glass compositions and grain sizes. Powders GP4 and GP10 are characterized by an irregular, angular broken grain. Powder GP10 also has a high proportion of fines, sticking on a larger surface, which can be seen in the SEM image by the small particles on the powder grains. Within the scope of powder characterization, a powder size distribution for GP4 of 137 µm to 340 µm and for GP10 of 115 µm to 250 µm was determined. The SiO_2 glass powder GP6 was produced by a special process, is spherical and amorphous with a grain size of 141 µm to 434 µm. The particle distribution of the powders mentioned corresponds to a Gaussian-like distribution, which is also typical for metallic powders in the L-PBF process. Based on these results, the minimum layer thickness to be applied was determined experimentally (see Table 1). It is assumed that the higher the flowability and bulk density of the powder, the smaller the layer thickness that can be set.

Table 1. Geometrical and technological properties of the used powders

Sample number	Particle size				Flowability		Tap density	Applied coating thickness
	MW [µm]	D 10 [µm]	D 50 [µm]	D 90 [µm]	Funnel Ø 2.5mm [s]	Funnel Ø 5mm [s]	[g/ml]	[µm]
SiO2 (GP4)	232	137	228	340	95.4	18.42	1.39	300
SiO2 (GP6)	277	141	264	434	194.4	24.90	0.81	400
B3.3 (GP10)	176	115	175	250	148.2	21.30	1.15	200

For the technological processing of the glass powders in the powder bed, the spreading and the layer thickness is of great importance. Therefore, within the scope of the experimental investigations, the glass powders were first examined with a hand operated blade and adjustable gap size, homogeneity of the coating and the coating thickness. The SLM 50 coating system by ReaLizer was then used to mechanically process relevant powders and to evaluate the quality of the result, depending on the layer thickness by means of visual assessment. Fig. 2 shows an example of the homogeneous coating application for the glass powder GP10 with a layer thickness of 200 µm, which can be rated as good for this material. A homogeneous coating thickness is achieved for all three powders with 1 to 1.5 times the average particle size (see Table 1).

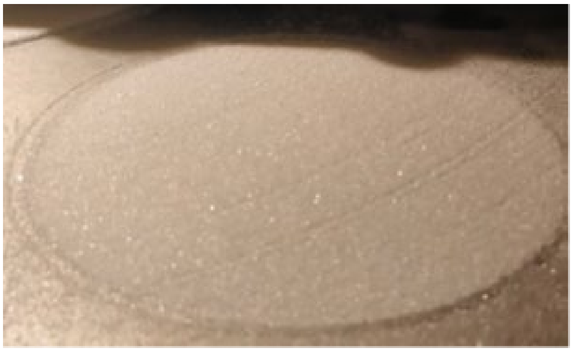


Fig. 2. Coating glass powder GP10 with SLM 50 from ReaLizer

Furthermore, the respective processing temperatures are relevant for the thermal processing. Compared to pure quartz glass, these temperatures are significantly lower for borosilicate glass. Thermal behavior of pressed borosilicate glass powders is shown in Fig. 3 at typical processing temperatures. They show the compression or the start of melting of the glass powder. It can be seen that above the softening point of 820 °C at 900 °C there is a clear shrinkage of the powder compact and above 975 °C melting can be observed. In the L-PBF process these temperatures determine the adjusted quality of the glass component (sintered or melting) and its later application.

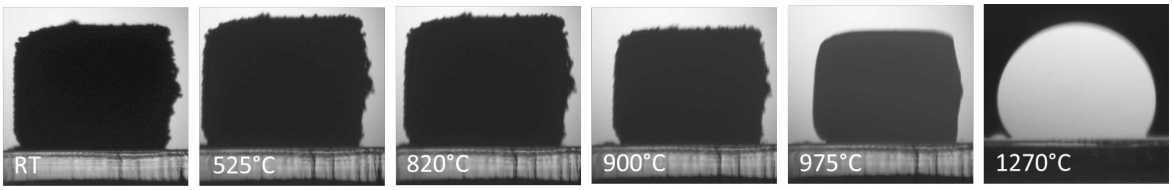


Fig. 3. HTM images – glass powder GP10

3.2. Experimental investigation of the L-PBF process

3.3. Systems engineering

The experimental investigations were carried out on two different systems, a commercial SLM 50by ReaLizer with a Nd:YAG laser (manufacturer IPG Laser GmbH) of the wavelength λ = 1.064 µm and an experimental CO_2 laser plant with different sources (SYNRAD 57-1 series P_{max} = 100 W, FEHA P_{max} = 1200 W) of the wavelength λ = 10.6 µm.

The SLM 50 is an encapsulated system with a heatable copper platform, which can be preheated up to 190 °C during the process, and a coater, which distributes the powder on the platform.

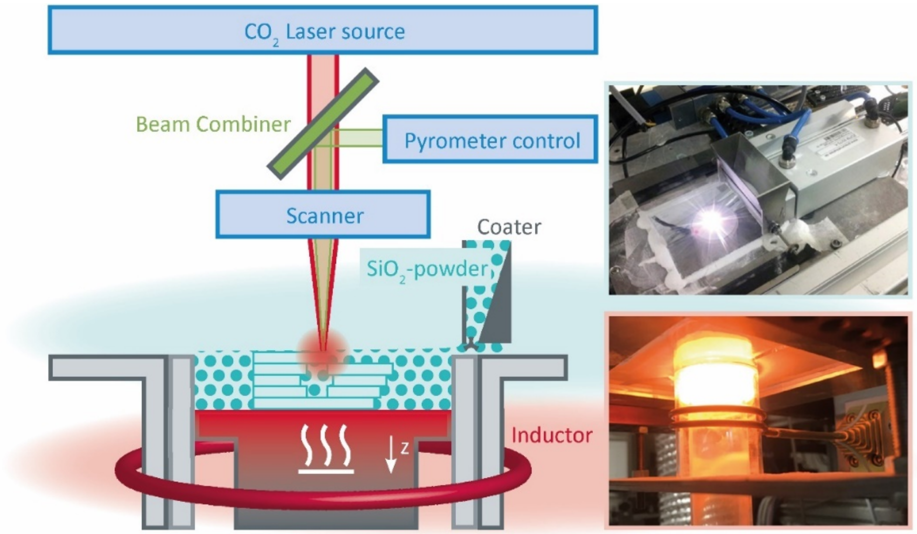


Fig. 4. Schematic setup of the experimental CO_2 laser plant, coater and inductive heating

The experimental CO_2 laser plant was further developed during the investigations to meet the requirements of powder processing of glass. Fig. 4 shows the schematic principle. To ensure a constant glass temperature during processing, a pyrometric control system was integrated into the plant. For this purpose, the measuring beam of the pyrometer is coaxially superimposed on the laser beam so the glass temperature is recorded directly in the laser action zone. This prevents partial evaporaion of the glass and ensures a constant temperature during the entire construction period. The powder is applied via a pneumatically movable hopper, through whose slotted opening the powder can flow to the building platform. This slot is sealed with a temperature-resistant lip all around. A challenge with L-PBF processing of glass powders is the required heating of the building platform to the range of the transformation temperature of the glass. For GP4 approx. 1000 °C, for GP10 approx. 500 °C are required. In the experimental plant heating is carried out indirectly by induction. A conductive ceramic under the building platform provides the necessary temperature transfer. This allows high temperatures to be quickly achieved without contact but leads to an extremely high load on the entire plant. Therefore, the construction of the plant and the selection of suitable materials is of great importance.

3.4. Investigations and results for melting glass powder

The aim of the investigations was to melt the glass powders GP4, GP6 and GP10 using the laser wavelengths 1.064 µm and 10.6 µm in the L-PBF process and to produce compact glass components. Laser and process parameters such as laser power, beam diameter (focus position), scan speed, track pitch and scan strategy were varied. In the investigations with the experimental station, the temperature of the platform was adjusted depending on the transformation temperature of the glass powders. The temperature-dependent laser power control was adjusted to the melting temperature of the glass powders that was determined in the HTM investigations. Based on the results of the coating tests for powders GP4, GP6 and GP10, the layer thickness was also adjusted to a constant value.

Table 3. Overview of essential laser and process parameters of the test facilities

Heading level	SLM 50, ReaLizer	Experimental station	
		SYNRAD 57-1	FEHA 1200
Laser wavelength λ [µm]	1.064	10.6	10.6
Laser power maximum P_{max} [W]	100	100	1200
Laser power used P_L [W]	80–100	10–60	200–800
Scanning speed v_s [mm/s]	2–50	1–10	800–1200
Temperature Building platform T [°C]	190	500–1000	500–1000

The investigations with SLM 50 by ReaLizer show that both SiO_2 powders (GP4, GP6) cannot be processed regardless of the powder geometry. Due to the solid-state laser (Nd:YAG) with a wavelength of 1.064 µm, which is only poorly absorbed in the SiO_2 powder, and the low preheating temperature of the substrate plate (190 °C), it was not possible to produce any solids.

Although the borosilicate glass powder (GP10) also has low absorption at the laser wavelength used in this system, the powder with its significantly lower softening temperature (820 °C) could be processed with the SLM 50 [13]. Significant fusions of the powder are achievable by a volume energy density of about 200 J/mm³ to 250 J/mm³. Evaluations of these experiments showed that there is a conflict of objectives between achieving a melt line and maintaining geometric accuracy. With the aid of higher volume energy densities, better fusions can be achieved. However, this increases the geometric deviations as a result of higher amount of particles sticking on the surface powder. This is due to the low thermal conductivity of borosilicate glass powder and the resulting heat accumulation in the powder bed. In addition, tests with different scanning strategies showed that the best results can be achieved by scanning the outer contour and the hatch. In the tests, parameters could be determined to produce test specimens for materials testing (Fig. 5). Their quality (porosity, surface, tightness) improved by a subsequent heat treatment.

Fig. 5. Test specimen of GP10

Investigations with different CO_2 laser systems at the experimental plant showed that the two quartz glass powders GP4 and GP6 can be processed due to their almost complete absorption for the wavelength of a CO_2 laser. Compared to metal powders, glass powders have a lower thermal conductivity by a factor of 100. This makes it more difficult to bond the upper powder layers to the layers below. For this reason, the glass powder must be converted to its molten state with high energy densities in order to achieve fusing. Depending on the exposure strategy, beam diameters of 2 to 3 mm were used to achieve the necessary processing temperature of 1700 °C to 2100 °C and to establish the fusing. Two exposure strategies were investigated:

- Slow, progressive exposure due to small beam movement:
    - Speed: 1 to 10 mm/s
    - Laser power: 10 to 60 W
    - Volume energy density: 5 to 7 kJ/mm²
    - Number of scans: 1
- Quasi-simultaneous exposure through high beam movement:
    - Speed: 800 to 1200 mm/s
    - Laser power: 250 to 800 W
    - Volume energy density: > 30 kJ/mm³.
    - Number of scans: approx. 1000

The construction rate for circular geometries could be increased considerably by the quasi-simultaneous exposure. However, this requires an energy density that is 5 times higher. Furthermore, the experiments showed that SiO_2 solids (Fig. 6) can be produced, which have a density of approximately 2.2 g/cm³ and are comparable with literature values. The component itself has a milky appearance, because during the construction process, in addition to the melting trace, other particles adhere and do not melt completely.

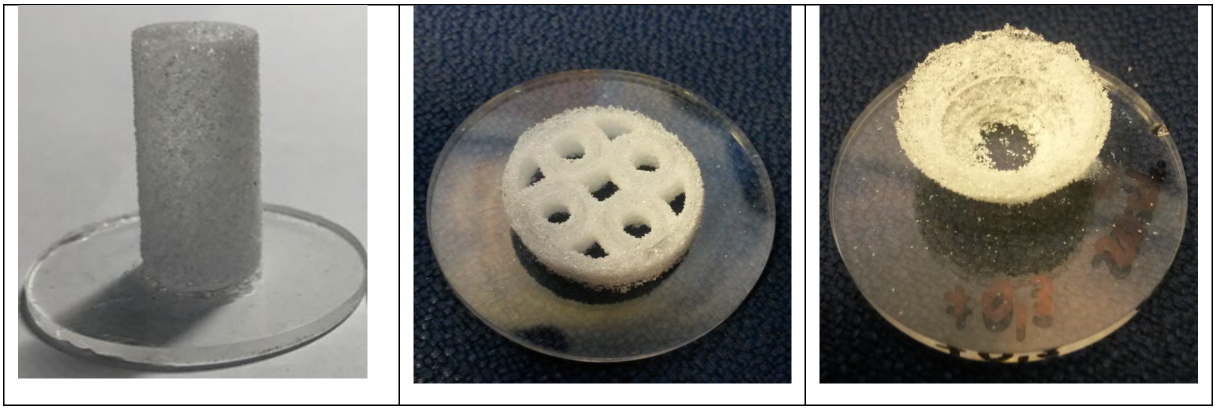


Fig. 6. Test specimen from GP4 (platform diameter Ø 30 mm)

Fig. 7 shows the influence of the platform heating by means of melted monolayers (10 x 10 mm²). The exposure parameters were identical in both cases. The considerably higher melt content in investigations with preheating is clearly visible.

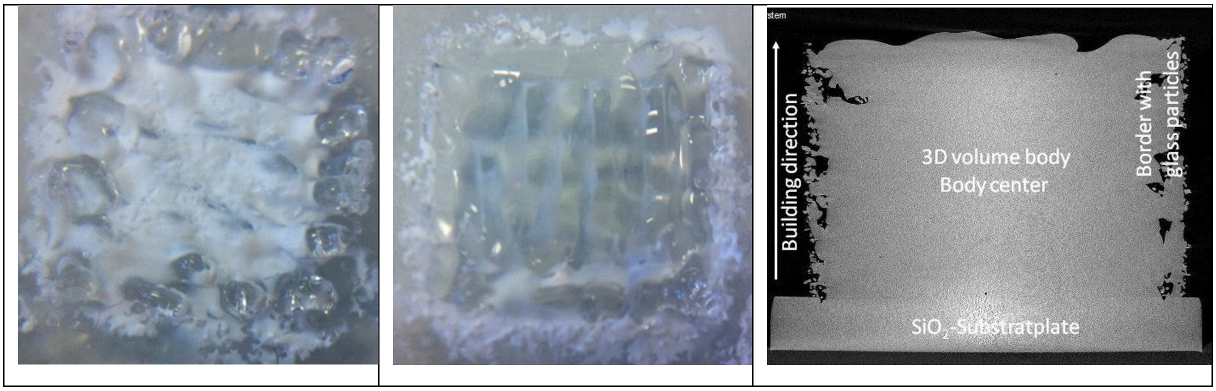


Fig. 7. Fused monolayers of GP4 (platform temperature at room temperature (left) and T = 600 °C (right)) and radiographic inspection of a volume body

4. Summary and outlook

This paper presents investigations for laser beam powder bed fusion of borosilicate and quartz glass powders. Different glass powder classes were characterized with respect to their geometric, thermo- and mechanical-physical properties and the technological processability in the PBF process using different laser wavelengths was investigated. The plant technology was adapted to the different requirements of glass processing, especially for quartz glass powders with CO_2 laser radiation. For the production of test specimens from the different glass powders, process parameters were determined, and initial quality evaluations were carried out. In the tests the basic processability of the glass powders was proven. Glass specimen could be manufactured successfully from borosilicate and quartz glass powder by laser radiation.

In future investigations, however, the process and experimental setup must be further optimized. To improve the component quality, further investigations on post heat treatment (e. g. stress relief) are to be carried out. Actually a new test setup was build, where all experimental results will be included. Highlight is an adjustable optic, which offers to produce a better surface quality of the printed parts. In interaction to the laser and process parameters transparent glass parts should be generated with the L-PBF process. Furthermore, the analysis of the material component quality with regard to porosity, roughness and tightness in relation to the developed L-PBF parameters is to be continued and possible further fields of application are to be shown.

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