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Analysis of the influence of different flowability on part characteristics regarding the simultaneous laser beam melting of polymers

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Abstract

Powder based Additive Manufacturing technologies offer huge potential for building parts with almost no geometrical restrictions, but both the process controlling as well as the part properties are strongly dependent on different material characteristics of the material, like the flowability. In this work, different weight percentages of nano-scaled silica dioxide particles (Aerosil®) are admixed to pure polyethylene and polypropylene powder and the resulting flowability is determined. Besides using the Hausner ratio as standardized value, the degree of coverage is introduced as a new characteristic to quantify the powder flowability. The degrees of coverage are compared to the Hausner ratios to allow a discussion and evaluation about the different characteristic values. Additionally, tensile bars consisting of polypropylene are generated to determine the porosity by cross sections and the mechanical part properties by tensile testing. As mechanical part properties, the tensile strength and elongation at break are determined and the effects of different powder flowability on these properties are analyzed.

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Keywords: Additive Manufacturing; Laser Beam Melting of Polymers; Material Qualification

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1. Introduction

Additive Manufacturing technologies, like the selective Laser Beam Melting of polymers (LBM), allow the generation of polymer parts directly out of CAD data by a layer-wise melting of polymer powders without any tools. This enables quick adaptions of required geometry changes of parts and a high flexibility in the generation of individualized products. Besides these advantages, when compared to other technologies like injection molding or milling, however, several limitations exist. To name a few, the need for an improved repeatability for series production and the possibility to generate multi-material components needs to be mentioned. Another major limitation is the restricted amount of powder materials available. For more than 90 % of the parts built by LBM polyamide 12 powder material is used (Wohlers, 2014). Although new materials are available, these powders often need to be modified regarding their material properties to be used by LBM machines. Besides the modification of the absorptance (Laumer, 2013), a very important material characteristic is the flowability of the powder. The flowability has an important influence on the deposition by roller or recoater system and also on the resulting part properties. To determine the flowability of powders different methods exists. The most important are the Hausner ratio according to the VDI Norm 3405, the measurement of the repose angle (Amado, 2011) or the measurement of the powder tensile stress according to *Jenike* (Krantz, 2009). Each of these measurement techniques have advantages but also limitations if compared to each other. One common limitation is that the layer thickness, by which the powder is deposited in the process, is not considered by the former mentioned measurement techniques. Thus, in this work, the degree of coverage is introduced as a new characteristic number to quantify the powder flowability of different polymer powders in dependency of different layer thicknesses. The results are compared to the Hausner ratio of the different powder mixtures to allow an evaluation. Additionally, the dependency of the part porosity and the mechanical part properties on different degrees of coverage is analyzed.

2. Materials and experimental setup

In the following section the Simultaneous Laser Beam Melting (SLBM) process is introduced, before basic properties of the materials used are shown. Afterwards, the experimental setup and procedure to determine the powder flowability of different mixtures are presented. At last, single material specimens are built by SLBM to allow an analysis of a possible correlation between the flowability and the part porosity and mechanical part properties.

2.1. Simultaneous Laser Beam Melting of Polymers

The Simultaneous Laser Beam Melting (SLBM) process is based on a conventional LBM process. It allows the layer-wise generation of multi-material parts consisting of different polymers. In the process two different powder materials are deposited next to each other by a two-chamber recoater. Both powders are warmed by infrared emitters and a heated building platform to the preheating temperature of the polymer with the lower melting temperature, in this case polymer A. At the same time, polymer B, as polymer with the higher melting temperature, is locally warmed to its preheating temperature by CO_2 laser radiation ($\lambda = 10.60$ µm). To achieve a homogeneous energy and thus homogeneous temperature distribution on the powder surface, a diffractive optical element (DOE) is used for beam shaping. The DOE forms a rectangular beam cross section with a homogeneous intensity profile on the powder bed. After both polymers are warmed to its preheating temperatures, the specific layer geometry needs to molten. To achieve a simultaneous melting of complex layer geometries, a special beam shaping device is needed, which allows a simultaneous, and compared to a DOE, a geometrical flexible energy deposition as well. Therefore, a micro-mirror array is used. The array is homogeneously irradiated by the second laser source and acts as flexible mask for beam shaping. Because CO_2 laser radiation is absorbed by the front window of the micro-mirror array and therefore would damage the device, a thulium laser with a wavelength of 1.94 µm is used. Each of the nearly two million micro mirrors of the chip can be tilted between two angles with an individual tilting frequency. One tilting angle guides the incident beam onto the powder bed, whereas by the other tilting angle the beam is guided into a beam trap. By varying the tilting frequency, locally graded intensity profiles and therefore temperature distributions can be realized in the powder bed. After both materials are in molten state, as last process step, the building platform

is lowered by the specific layer thickness, a new powder layer is deposited and the process steps are repeated. The energy deposition and resulting temperatures on the powder surface are controlled by a thermal imaging system. The system offers a high resolution with 1280x1024 pixels and is calibrated with thermocouples in a temperature range between room temperature and the respective melting point of the materials. The calibration is necessary because temperature-dependent changes of the emissivity of the polymers would otherwise lead to a wrong temperature measurement. Figure 1 shows the schematic process cycle.

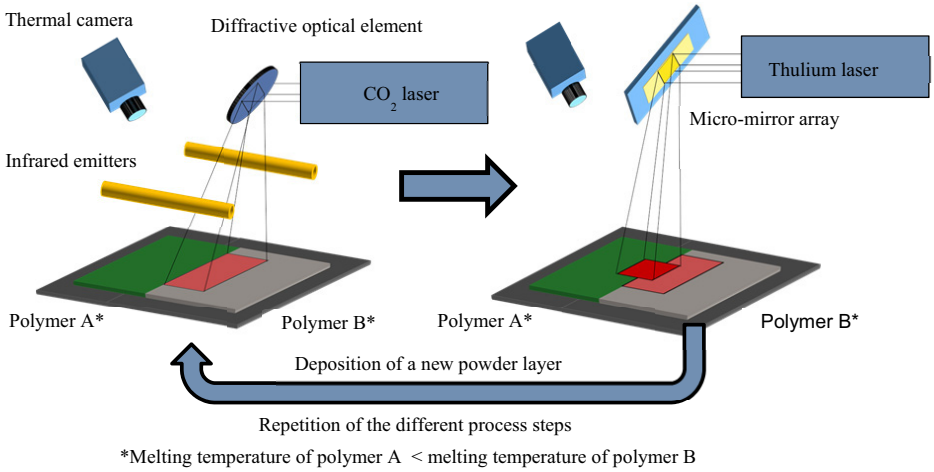

Fig. 1. Schematic process cycle of SLBM.

2.2. Materials

For the experiments in this paper, high density polyethylene (PE-HD, NB5374 Coathylene, Axalta) and polypropylene (PP, PD0580 Coathylene, Axalta) powders are used. Table 1 shows the particle size distribution and the thermal material properties of both powders. The particle size distribution was determined by a Mastersizer Hydro 2000S (Malvern) and the thermal material properties by a Differential Scanning Calorimety (DSC) measurement by using a DSC822e (Mettler-Toledo). As shown in former works of the authors, the absorptance of both materials needs to be improved because of the lower absorptance of the thulium laser radiation by the polymer powders. Thus, to both powders 0.25 wt.-% carbon black is admixed as absorption intensifier to increase the absorptance of 200 µm thick powder layers to values in the range of 0.70 (Laumer, 2013).

Table 1. Particle size distribution and thermal material properties of PE-HD and PP.

	PE-HD	PP
d_{10} [µm]	35	60
d_{50} [µm]	57	100
d_{90} [µm]	100	150
Melting temperature [°C]	129	165
Crystallization temperature [°C]	117	129

To allow a qualitative analysis of the particle geometry, scanning electron microscope images of both materials are shown in figure 2. Both powders have rough surfaces and an elliptical geometry. By comparing both images, the different particle size distribution of the powders is visible.

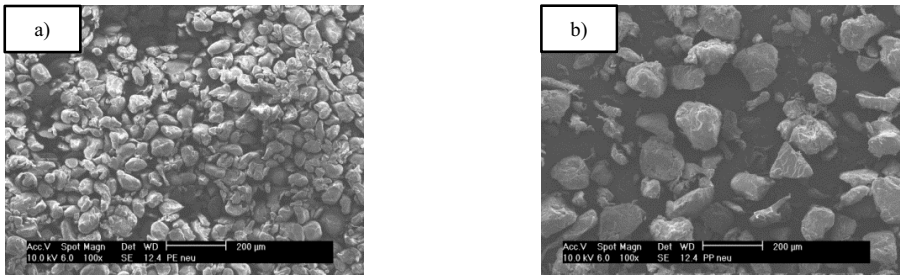

Fig. 2. Scanning electron microscope images of (a) PE-HD powder and (b) PP powder.

2.3. Determining the Hausner ratio and the degree of coverage as characteristic number for powder flowability

Former investigations have shown that the flowability of PE-HD and PP is not sufficient to allow a homogeneous powder deposition by recoater (Laumer, 2015). Thus, aerosil (Aerosil® R106, Evonik), nano-scaled silicon dioxide particles, have to be admixed to both powders to increase the flowability. In this work, the pure powders and the powders with an admixture of 0.1, 0.25, and 1.0 wt.-% aerosil are analyzed. For the mixing process, a turbula mixer is used with a mixing time of 60 minutes. To start, the Hausner ratio of the different powder mixtures is determined according to VDI norm 3405. A defined volume of 50 ml of powder is poured into a beaker glass with an inner radius of 20 mm and weighed. By knowing the volume and the mass of the powder the bulk density ρ_{bulk} can be calculated. Subsequently, by shaking the beaker glass, the powder is compacted until no further volume reduction occurs. By dividing the constant mass by the volume of the compacted powder the tamped density ρ_{tamped} follows. The Hausner ratio H_r itself is calculated as the quotient of the bulk and tamped density. A Hausner ratio smaller than 1.25 represents good flowability, a value between 1.25 and 1.4 reduced flowability and a value higher than 1.4 describes cohesive powder. The Hausner ratio allows the comparing of the flowability of different powders but gives no information about the dependency of the powder flowability and the used layer thickness on the quality of the deposited powder layer.

Thus, in addition to determining of the Hausner ratio, the degree of coverage is introduced as a new characteristic number for evaluating the powder flowability. To determine the degree of coverage a defined amount of powder in the range of 10 g is given into the chamber of a film applicator (Erichsen) and a powder layer is deposited onto a paper as surface. The distance between the surface and the blade of the film applicator is varied between 100, 150, 200, 250 und 300 µm. Therefore, depositing different layer thicknesses is possible. The width of the powder layer is 40 mm. Because of a possible influence of the paper as surface material, the same kind of paper is used for all experiments. The surface roughness of the paper is measured by a laser scanning microscope Lext OLS4100 (Olympus) and shows a S_a-value of 5 µm. After the deposition of the powder layer, an image of the layer is taken by a digital SLR-camera. With the help of image processing software, the ratio between area covered with particles and the uncovered area is determined. The percentage of covered surface represents the degree of coverage. To detect and discuss any correlation between the Hausner ratio and the degree of coverage the different values are compared to each other. In figure 3 the film applicator and a deposited powder layer of PP is shown.

Fig. 3. Film applicator with deposited PP powder layer.

2.4. Generation of tensile bars and determination of mechanical part properties

After determining the flowability, PP tensile bars are built by SLBM. Thus, a possible correlation between the powder flowability, the part porosity and the mechanical part properties, like the tensile strength and elongation at break, can be analyzed. The building parameters are listed in table 2. For each powder mixture, five tensile bars are built to allow a statistical interpretation of the mechanical properties.

Table 2. Building parameters for the generation of the tensile bars.

Parameter	Value	Unit
Preheating temperature	150	°C
Building platform temperature	120	°C
Irradiation time of thulium laser	20	s
Intensity of thulium laser	8	mJ/mm²s
Cooling rate	1	K/min

As specimen geometry 1:5 scaled Campus tensile bars are used. The parts are five layers thick, whereas the deposited layer thickness is 200 µm. All bars are tested with a tensile testing machine. The detailed geometry of the specimen is shown in figure 4. To determine the part porosity, untested tensile bars are embedded in resin and prepared as cross section.

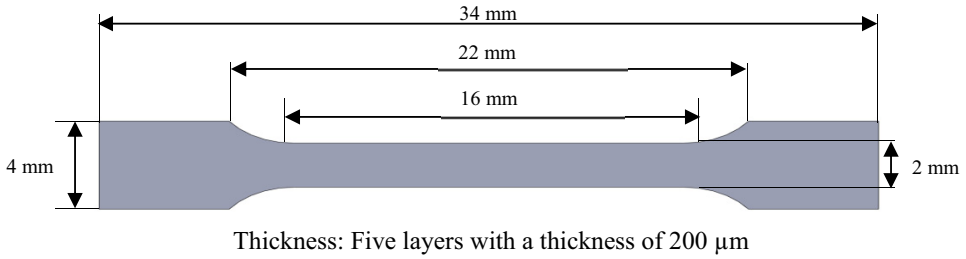

Fig. 4. Geometry of the tensile bars.

3. Results and Discussion

3.1. Hausner ratio and degree of coverage

In table 3 the bulk and tamped density and the resulting Hausner ratio of the different powder mixtures are listed. The standard deviation based on five measurements of the Hausner ratio is also added. By admixing 0.1 wt.-% aerosil to the pure PE-HD powder, the Hausner ratio is significantly decreased, which indicates a higher flowability. With an increasing amount of admixed aerosil the Hausner ratio increases, but is still below the value for the pure powder material. Thus, according to the Hausner ratio, the flowability is overall improved by admixing aerosil, but reaches its optimum at an amount of 0.1 wt.-% aerosil. PP shows the same behavior. Although by admixing aerosil the Hausner ratio is reduced compared to the pure powder and thus, the flowability is increased, the minimal Hausner ratio is reached at an admixed amount of aerosil of 0.1 wt.%. After surpassing this optimum, the Hausner ratio starts to increase and the flowability decreases. Both pure powders have a Hausner ratio higher than 1.40 and thus should show a cohesive powder behavior. By admixing aerosil, the Hausner ratio can be reduced to values between 1.27 and 1.39, thus the different mixtures should show a reduced but sufficient flowability.

Table 3. Density and Hausner ratio of the different powder mixtures.

Material	Bulk density [g/cm³]	Tamped density [g/cm³]	Hausner ratio	Standard deviation
PE-HD + 0.0 wt.-% aerosil	0.35	0.50	1.43	0.012
PE-HD + 0.1 wt.-% aerosil	0.37	0.47	1.27	0.019
PE-HD + 0.25 wt.-% aerosil	0.37	0.49	1.33	0.018
PE-HD + 1.0 wt.-% aerosil	0.39	0.52	1.35	0.037
PP + 0.0 wt.-% aerosil	0.34	0.50	1.47	0.013
PP + 0.1 wt.-% aerosil	0.38	0.52	1.37	0.019
PP + 0.25 wt.-% aerosil	0.38	0.53	1.39	0.017
PP + 1.0 wt.-% aerosil	0.39	0.55	1.40	0.023

Besides the Hausner ratio the degree of coverage is determined. In figure 5, the coverage is shown for a 200 µm thick powder layer of PP. On the left side of the image pure PP is deposited, whereas on the image on the right side a mixture of PP and 0.25 wt.-% aerosil is deposited. The difference in the coverage resulting of different flowability is clearly recognizable.

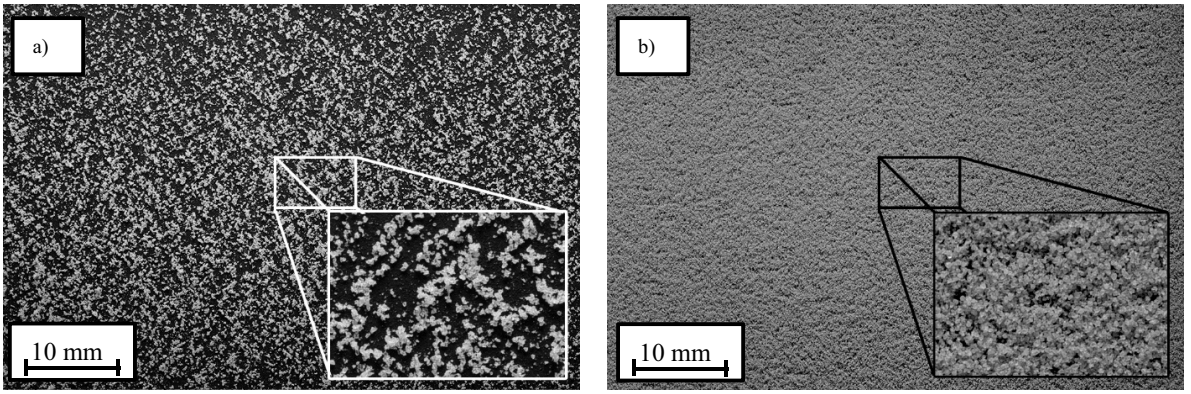

Fig. 5. Different degrees of coverage for different powder flowability: (a) PP powder without aerosil (b) PP powder with 0.25 wt.-% aerosil.

In figure 6 the degree of coverage for the different powder mixtures at different layer thicknesses is shown. The standard deviation is below 3 % and therefore is not added in the diagram. In accordance with the Hausner ratios, the admixture of aerosil leads to higher degrees of coverage due to an increase of the powder flowability for both powders. The pure PE-HD powder shows a low coverage of below 0.3 for layer thicknesses in the range between 100 and 300 µm. The admixture of 0.1 wt.-% aerosil leads to a significant increase of the coverage to nearly 0.8 for a layer thickness of 100 µm. The maximal coverage at a layer thickness of 100 µm is reached for PE-HD with 0.25 wt.-% aerosil. For a higher amount of aerosil the coverage decreases again. Thus, between 0.25 and 1.0 wt.-% admixed aerosil a saturation threshold is reached. After surpassing this threshold, the forming of agglomerations of aerosil particles can be expected. This runs counter to the basic principle of the flowability increase by aerosil particles because the particles stick together instead of adhering themselves onto the polymer particles to reduce the inter-particular interaction forces. Additionally, the agglomerations can reach sizes up to or even bigger than the polymer particles itself (Blueml, 2015). If these agglomerations are in the size range of the layer thickness they are tagged along by the recoater blade during the powder deposition of a new layer and are resulting in streaks in the powder bed.

Overall, the degrees of coverage of the powders with admixed aerosil increase with increasing layer thickness. At a layer thickness of 300 µm all powder mixtures have the same degree of coverage of 0.95. For high layer thicknesses in comparison to the average particle size of PE-HD of 50 µm, as for 300 µm, the probability that particles are not only positioned next to each other but also on top of each other increases. This effect superposes the influence of the flowability so that a higher degree of coverage is automatically measured. Therefore, the degree of coverage is especially suited for comparing different powders at smaller layer thickness between 100 and 200 µm, which are typically used layer thicknesses for the SLBM and LBM process.

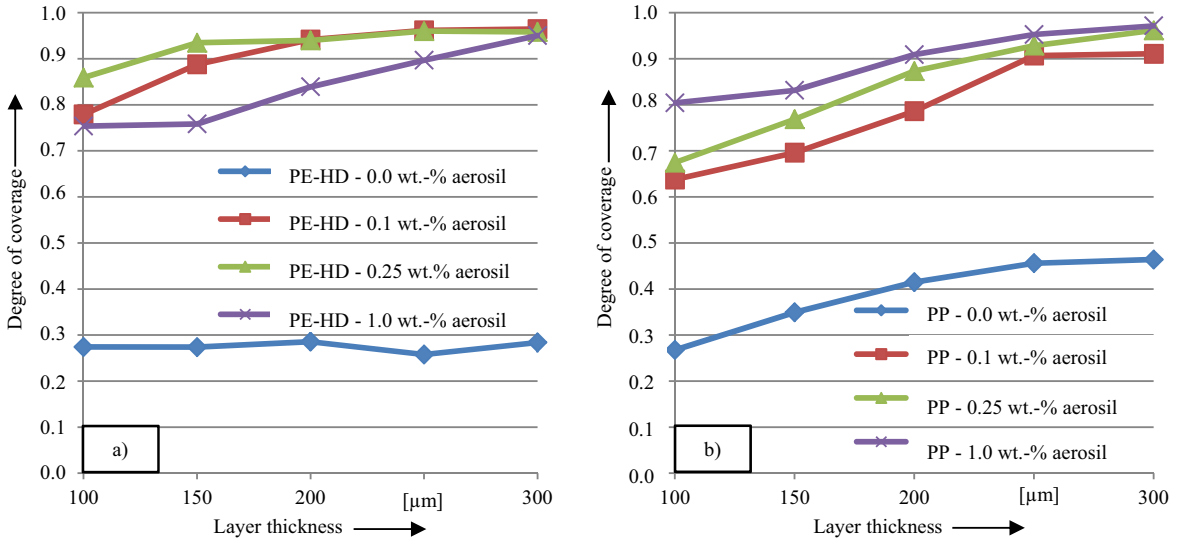

Fig. 6. (a) Degree of coverage PE-HD; (b) degree of coverage PP.

In the case of PP, the degree of coverage of the PP powder continuously increases with the increase of admixed aerosil. Thus, the saturation threshold of the PP is not reached within the range of 0.1 and 1.0 wt.-% aerosil as in the case of PE-HD. Possible reasons can be different intermolecular interaction forces of both polymer molecules or the different particles sizes of both powders. A further explanation can only be given with a more detailed analysis in future works.

3.2. Porosity and mechanical part properties

Before the results of the tensile testing are presented, the porosity is determined by cross sections of the tensile bars. With the pure PP powder no sufficient powder deposition and thus no building of specimens is possible. Therefore, the cross sections and mechanical tests are only possible for the different mixtures of PP with aerosil. In figure 7 an example for the cross section of a tensile bar built of PP with 1.0 wt.-% aerosil is shown. Next to the cross section the porosity for all powder mixtures is given.

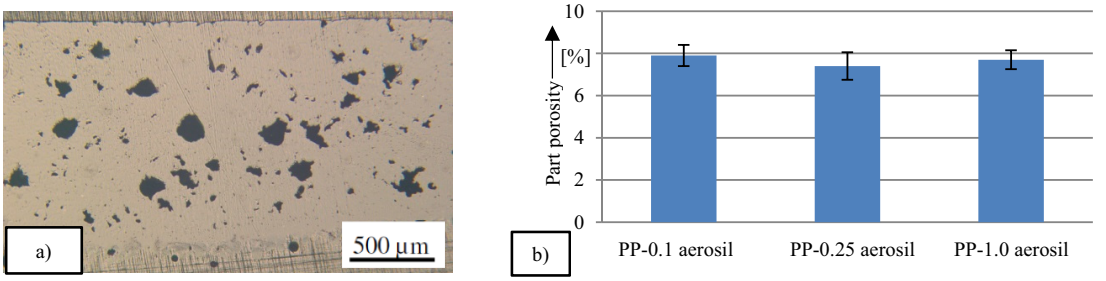

Fig. 7. (a) Cross section of PP tensile bar; (b) part porosity of different powder mixtures.

The porosity for all powder mixtures lies between 7 and 8 %. Thus, no significant influence of the different amount of admixed aerosil can be detected. Although the coverage for a 200 µm thick PP layer varies between 0.78 and 0.91 for different amounts of admixed aerosil, the different flowability is not influencing the porosity of the generated bars. Compared to the porosity of parts built of polyamide 12 (PA2200, EOS), which have typical porosities of below 2 % (Wegner, 2015), the porosity is very high. The reason is the particle size distribution of the powder. According to *Alscher* a powder mixture of bigger particles with a smaller amount of fine particles reaches higher bulk densities compared to a powder with a broad particle size distribution. Because the bulk density correlates directly with the part density and thus the porosity, a higher bulk density leads also to a higher part density and a smaller porosity (Alscher, 2000). Unlike PA2200, which has very round particles and a small particle size distribution around 60 µm with an additional amount of small particles below 10 µm, for powders manufactured by grinding, like the PP used in this work, the particles size vary broadly (Rietzel, 2011). Therefore, the smaller bulk density of PP compared to PA12 explains the higher porosity of the PP specimens. The relation between bulk density and porosity explains also the similar porosities of the different PP powder mixtures with aerosil. Because the bulk density of the different PP powder mixtures is similar and varies only between 0.38 und 0.39 g/cm³, also the resultant part porosity is almost the same.

In summary, the degree of coverage does not allow a prediction of the part porosity, because the bulk density cannot be determined by the measurement. Like schematically shown in figure 8 the relation between the summed up volume of all powder particles and the total volume of the deposited layer, which is defined by the covered area of the substrate multiplied with the layer thickness, represents the bulk density. To determine the volume of all powder particles, a three-dimensional measuring of the whole layer would be necessary.

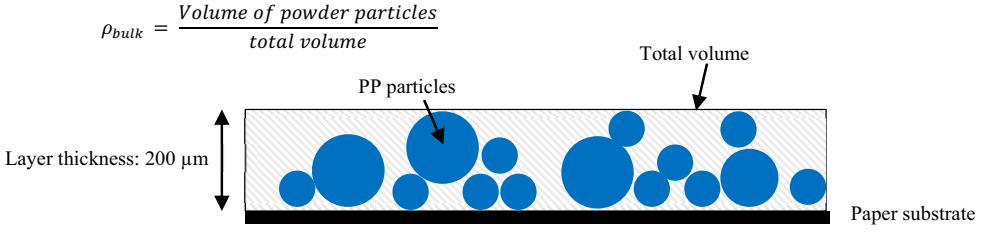

Fig. 8. Difference between coverage and bulk density.

Additionally, the tensile strength and the elongation at break are determined. In figure 9 the results for the mechanical testing are shown for the three different powder mixtures of PP and aerosil. The standard deviation is also included in the figure.

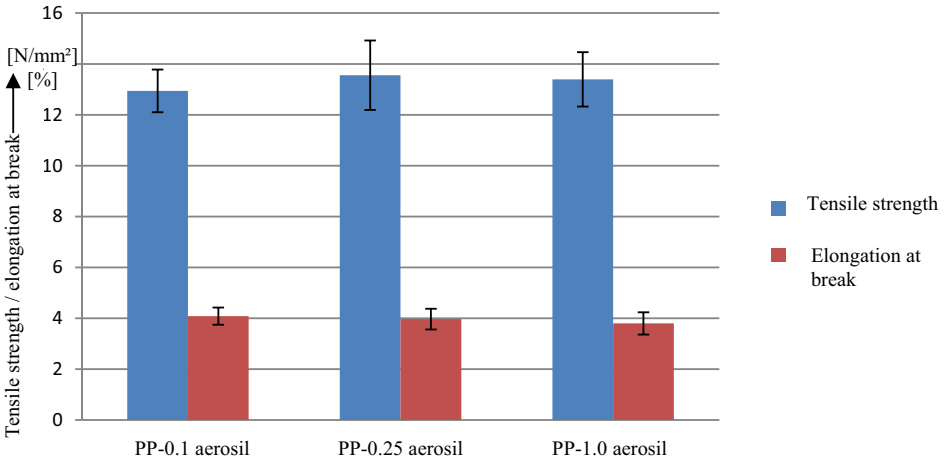

Fig. 9. Tensile strength and elongation at break of the tensile bars consisting of PP.

The tensile strength as well as the elongation at break are showing no significant difference for the different powder mixtures. The tensile strength varies between 13.0 and 13.6 N/mm² and the elongation at break lies between 3.8 and 4.1 %. Due to the use of the same building parameters, the same energy amount is deposited into the powder layers during the melting process and because the bulk density and thus the porosity is similar as shown before, these results are expected. The results confirm that in the analyzed parameter field no correlation between the degree of coverage and the porosity and mechanical properties is possible for PP.

4. Conclusion and outlook

The degree of coverage allows the evaluation of the flowability of different polymer powders. Compared to the Hausner ratio, by the degree of coverage the relation between the flowability and the layer thickness can be also analyzed. It is shown, that the quality of the deposited powder layer is significantly influenced by the layer thickness, which is not considered by the Hausner ratio. Thus, the degree of coverage is a good characteristic number, which allows comparing the flowability of different polymer powders in dependence of the layer thickness. Further it is shown, that by the degree of coverage the bulk density cannot be determined, because only a two-dimensional information about the layer geometry is given. Due to the direct relation between the bulk density and the resulting part density and porosity, the degree of coverage is not suited for analyzing the relation between the flowability and the resulting mechanical part properties.

In future works the degree of coverage will be determined for additional materials and analyzed regarding any correlation with other material properties like the optical material properties. It is expected that there is a correlation between the coverage and the transmittance of single powder layers. A smaller degree of coverage leads to more blank spaces within a powder layer, thus possibly resulting in a direct transmission of the incident laser radiation in these areas and a higher effective transmittance of the powder layer.

Acknowledgements

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References

Alscher, G., 2000. Das Verhalten teilkristalliner Thermoplaste beim Lasersintern. Berichte aus der Kunststofftechnik, Dissertation,Universität Essen, Shaker Verlag, Aachen.

Amado, A.; Schmidt, M.; Levy, G.; Wegener, K., 2011: Advance in SLS powder characterization. Proceedings of Solid Freeform Fabrication Symposium, Austin, Texas.

Blümel, C.; Sachs, M.; Laumer, T.; Winzer, B.; Schmidt, J.; Schmidt, M.; Peukert, W.; Wirth, K.-E., 2015: Increasing flowability and bulk density of PE-HD powders by a dry particle coating process and impact on LBM processes. In: Rapid Prototyping Journal, Vol. 21, Issue 6.

Krantz, M.; Zhang, H.; Zhu, J., 2009: Characterization of powder flow: static and dynamic testing. Journal of Powder Technology, Issue 194.

Laumer, T.; Stichel, T.; Amend, P.; Schmidt, M., 2015: Simultaneous Laser Beam Melting of multi-material polymer parts. In: Journal of Laser Application, Vol. 27.

Laumer, T.; Stichel, T.; Sachs, M.; Amend, P.; Schmidt, M., 2013: Qualification and modification of new polymer powders for laser beam melting using Ulbricht spheres. In: High Value Manufacturing, pp. 255-260..

Laumer, T.; Wudy, K.; Drexler, M.; Amend, P.; Roth, S.; Drummer, D.; Schmidt, M., 2014. Fundamental investigation of laser beam melting of polymers for additive manufacture. In: Journal of Laser Application, Vol. 26, Issue 4.

Rietzel, D., 2011: Werkstoffverhalten und Prozessanalyse beim Lasersintern von Kunststoffen. Dissertation, Friedrich-Alexander-Universität Erlangen-Nürbnerg, Lehrstuhl für Kunststofftechnik.

Wegner, A., 2015: Theorie über die Fortführung von Aufschmelzvorgängen als Grundvoraussetzung für eine robuste Prozessführung beim Laser-Sintern von Thermoplasten. Dissertation, Universität Duisburg-Essen.

Wohlers, T., 2014: Wohlers Report 2014 - State of the Industry, Fort Collins,Colorado, Wohlers Associates Inc..