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Process route adaption to generate multi-layered compounds using vibration-controlled powder nozzles in selective laser melting of polymers

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Abstract

Overcoming the inherent restriction of selective laser melting (SLM) of polymers to the processing of a single material remains a major challenge. Multi-material SLM requires more flexible deposition options to prepare arbitrary powder patterns. Besides the adaption of the recoating system, a suitable melting strategy must be developed. In this report, a previously described vibrational nozzle setup is used for the preparation of a multi-material powder layer. The irradiation strategy is modified to meet the requirements of processing multiple powders: Infrared emitters globally heat the building chamber and induce melting of the low-melting polymer shortly after its deposition. Subsequently, a scanned CO_2 laser beam irradiates the part area, thus melting the high-melting polymer and coalescing both materials. Flowability properties of different polymer powders are optimized for the nozzle-based deposition and analyzed accordingly. Using the adapted SLM process route, multi-layered compounds are generated and characterized regarding surface roughness and microstructure in the boundary zone.

Keywords: selective laser melting; polymers; multi-material; powder deposition; vibrational nozzles

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1. Introduction

Additive Manufacturing (AM) techniques such as Fused Deposition Modeling (FDM) and Selective Laser Melting (SLM) allow the generation of parts with larger geometrical complexity than what is achievable by traditional processes (e. g. machining, molding, casting) (Goodridge et al., 2012). However, SLM is restricted to the processing of one single powder material, in most cases polyamide 12 (PA12). Approaches to generate multi-material parts that combine different functionalities (e. g. hard-soft-compounds) from two or more polymers are still subject of current research (Laumer et al., 2015a; Laumer et al., 2015b; Laumer et al., 2016) and require the adaptation of the recoating system as well as the method of energy deposition.

Arbitrary powder patterns can be realized by vibrational nozzles which enable the selective deposition of fine lines and dots (Stichel et al., 2015; Stichel et al., 2016a; Stichel et al., 2016b). In such powder deposition systems, mass flow is initiated by vibration due to the break-up of powder bridges. These bridges are re-established as soon as the vibration is stopped, resulting in a valve-like start-stop-functionality of the nozzle system. Since the elevated temperatures during SLM processes strongly affect the flowability and therefore the mass flow of powder, precise control of the temperature within the nozzles is crucial. In this work, the temperature control is achieved by internal channels which allow the continuous flow of either distilled water (20 °C) or heat transfer oil (30 °C – 150 °C) through the nozzle.

A first approach to generate multi-material parts using vibrational nozzles was recently published by Stichel et al. (2018). In the presented process route, which employs three sources of irradiation, the temperature in the building chamber was set to the preheating temperature (T_{ph}) of the low-melting polymer. While this strategy was suitable for the generation of multi-material mono-layers, building more layers was not possible. Large temperature gradients led to an unwanted, early crystallization of the high-melting material, its contraction (i. e. curling) and thus did not allow the manufacturing of multi-layers. To overcome this obstacle and reduce the temperature drop during the process, we present a new process route using the same basic machine setup while altering the irradiation strategy.

2. Experimental

2.1. Modified SLM machine

Fig. 1a shows a schematic image of the recoating unit with integrated powder nozzles and a photograph of an additively manufactured metal nozzle for the selective powder deposition. The principal scheme of the nozzle control is displayed in Fig. 1b.

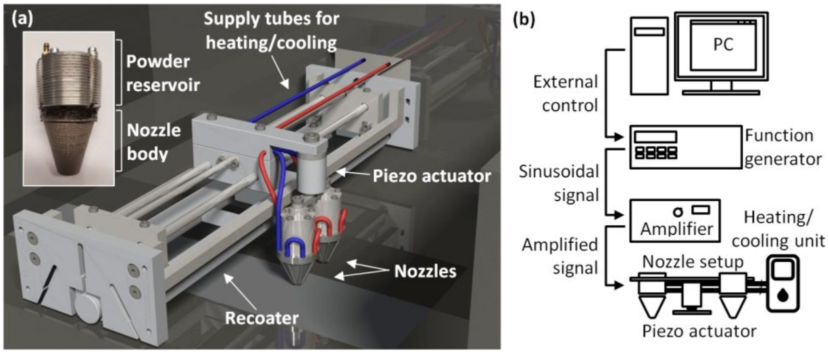

Fig. 1. (a) Recoating unit consisting of two attached vibrational nozzles, a piezo actuator and supply tubes for heating/cooling liquids, Inlet: photograph of an additively manufactured metal nozzle; (b) scheme of the nozzle control

In the modified SLM machine (P380, EOS GmbH, Germany), the nozzle setup consisting of the high-temperature piezo actuator and two additively manufactured nozzles is mounted to the standard recoater. For temperature control, the nozzles with orifice diameters of 0.7 mm (N1) and 1.0 mm (N2) are equipped with internal channels which are connected to a temperature control unit operating with heat transfer oil (150smart, Regloplas, Switzerland) and to a water cooler, respectively. To allow nozzle movement perpendicular to the movement of the recoater, a threaded rod and a stepping motor are added to the unit. A function generator creates a sinusoidal voltage signal which is amplified by a factor of 100 via an analog power amplifier. The amplified signal induces vibration of the actuator and therefore initiates powder deposition. The modified SLM machine furthermore consists of standard infrared (IR) emitters for global preheating of the building chamber to T_{ph}, a high-resolution thermal camera (Millenium 1310k M pro, IRCAM, Germany) for temperature control, a CO_2 laser (λ = 10.6 µm, P = 60 W, ti60, Synrad, USA) and a galvanometer scanner (MINISCAN II, Raylase GmbH, Germany). In the former works mentioned above, a second laser source (thulium, λ = 1.94 µm) was used for simultaneous melting of both materials.

2.2. Process route

Different strategies allow the melting of one or more multi-material powder layers. Two principal process routes are discussed here. Firstly, a third irradiation source (thulium laser) can be used for coalescence of both materials. Herein, the building chamber is heated by the IR heaters to T_{ph} of the low-melting material, the CO_2 laser selectively preheats the high-melting polymer and the thulium laser simultaneously melts both materials. As published by Stichel et al. (2018), this option was successfully used for the generation of single-layers of polypropylene (PP) and thermoplastic elastomer (TPE). Subsequent investigations to build more than multi-material mono-layers, however, revealed that the employed irradiation strategy is incompatible with the nozzle-based powder deposition. The temperature drop during the subsequent powder deposition of the second layer results in early and unwanted crystallization of the high-melting material. Several mechanisms are responsible for this undercooling (e. g. ongoing shadowing of the melted area by the nozzle setup during deposition, contact of the melted first layer with relatively cold powder). These lead to the contraction of the solidifying polymer (i. e. curling) and ultimately to the detachment in the boundary zone. Hence, a novel approach to generate multi-material parts of several layers is pursued, see Fig. 2.

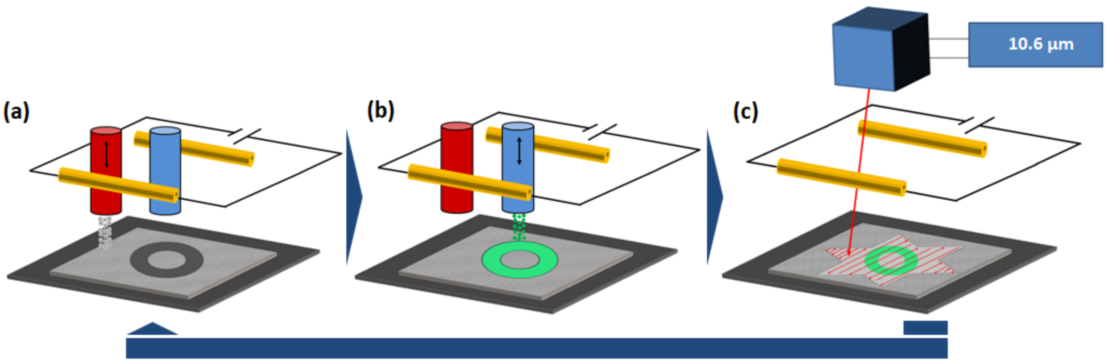

Fig. 2. Adapted process route for the generation of multi-material multi-layers: (a) deposition of the high-melting polymer (light grey); (b) deposition and – due to the high temperature in the building chamber – subsequent melting of the low-melting polymer (green); (c) quasi-simultaneous melting of the high-melting polymer by the scanned CO_2 laser radiation (red pattern)

In contrast to the first route, the building chamber is heated to T_{ph} of the high-melting material (i. e. above the melting point of the low-melting material) which leads to the melting of the low-melting polymer by the IR emitters directly after its deposition. This increased temperature in the building chamber is selected to prevent the abovementioned early crystallization since the temperature gradient between melted and powdery phase (i. e. the driving force for heat conduction) is reduced. The process route should therefore enable the fabrication of multi-layered compounds. The final part geometry is determined by both the IR-melted contour and the CO_2-scanned contour. Similarly to the standard SLM process, the non-melted powder acts as support material.

2.3. Powder materials

For the generation of multi-material parts (e. g. hard-soft-compounds), PP (PD0580 Coathylene, Axalta, Switzerland) and urethane-based TPE (TPE-U; Rolaserit PB, AM Polymer Research, Germany) were selected. Differential scanning calorimetry (DSC) measurements (from 25 °C to 200 °C with a heating/cooling rate of 10 K/min) and dynamic light scattering (DLS) analysis were conducted to initially characterize the powders with regard to melting temperatures (T_m) and particle size distribution (where per definition 10 %, 50 % and 90 % of the particles are smaller than the values of d_{10}, d_{50} and d_{90}, respectively). These powder properties are shown in Table 1.

Table 1. Melting temperatures and particle sizes of the as-received powder materials

	DSC	Particle size analysis		
	T_m in °C	d_{10} in µm	d_{50} in µm	d_{90} in µm
PP	167	54	118	204
TPE-U	120	32	89	138

Both powders were characterized and modified with respect to flowability which is essential for the nozzle-based processing during SLM. Furthermore, 0.5 wt% carbon black (Lamp Black 101, Orion Engineered Carbons, Luxembourg) was admixed to the TPE-U powder to allow the distinction between both materials and the microscopic analysis of the boundary zone.

2.4. Characterization methods

2.4.1. Flow behavior, deposition and building parameters

Flowability is crucial for the materials' use in nozzle-based powder deposition during SLM since it strongly affects the usability of the polymer powders. Flowability, which depends on temperature itself, needs to be adjusted to (i) prevent a permanent mass flow through the nozzle if there is no vibration stimulus and (ii) allow a reliable formation of powder bridges to instantaneously stop the vibration-induced mass flow as soon as the piezo actuator is turned off.

For evaluation of the flowability of both powders, the Hausner ratios (H_R) were determined according to VDI norm 3405 part 1.1. After analyzing the as-received powders, they were modified to fit the deposition method with nozzle N1 (connected to the oil heating unit) and nozzle N2 (connected to the water cooler). In case of PP (deposited by N1) which exhibits low flowability, a surface coating with flow enhancing agents was conducted by the institute of particle technology (LFG, Erlangen). Details on the functionalization process will be published elsewhere. TPE-U, on the contrary, required a reduction of flowability since it showed unreliable, delayed formation of powder bridges in N2 after external vibration. This was achieved by systematic fractionation and mixing of different ratios of fractions. The modified powders (termed PP* and TPE-U*) were again assessed with respect to H_R and then used for the nozzle-based SLM process.

The deposition parameters used throughout this work (see Table 2) were selected on the basis of former contributions concerning the thorough characterization of vibration modes and mass flow dependencies (Stichel et al., 2015; Stichel et al., 2016a; Stichel et al., 2016b) as well as on extensive parameter studies for the modified powders.

Table 2. Parameters for powder deposition

	Frequency f in Hz	Amplitude voltage U in V	Travel speed v in mm/s	Nozzle temperature T_{Nozzle} in °C
PP*	350	4	5	50 (N1)
TPE-U*	350	4	10	20 (N2)

With the selected parameters for powder deposition in combination with the adapted process route, multi-material specimens of five layers were manufactured. While a simple rectangular shape (30 mm x 18 mm) was selected for the subsequent topographic analysis, scaled (1:4) tensile bars (Type 1A based on DIN EN ISO 3167) were built to demonstrate the feasibility of the adapted process route for the generation of complex multi-material parts. These tensile bars consisted of two PP* regions at the ends and TPE-U* in the middle.

Since TPE-U* melts due to the high temperature in the building chamber, the scanning parameters only depend on the processing characteristics of PP*. Thus, these scanning parameters were found by preliminary melting tests of PP* with a fixed T_{ph} of 160 °C (selected based on the DSC values), varying CO_2 laser power (P_{CO2}), scan speed of the galvanometer scanner (v_{scan}) and hatch distance between the scan lines (h). Based on the qualitative evaluation of these PP* layers with respect to the degree of melted material compared to residual powder particles, suited parameters for energy deposition were selected (see Table 3).

Table 3. Parameters for energy deposition

Preheating temperature T_{ph} in °C	CO_2 laser power P_{CO2} in W	Scan speed v_{scan} in mm/s	Hatch distance h in µm	Scan pattern
160	18	200	100	Parallel lines

2.4.2. Microstructure

Microstructure of the boundary zone between both materials was evaluated by means of optical microscopy of thin films. Thin films of thicknesses between 30 µm and 50 µm were prepared using a microtome and subsequently analyzed.

2.4.3. Part topography

Surface topography of rectangular, five-layered multi-material specimens was analyzed by means of laser scanning microscopy (LSM) and laser profile sensor measurements. Quantitative values of areal surface roughness (i. e. areal arithmetic mean S_a) were determined based on DIN EN ISO 25178 and evaluated. The specimens were measured three times at different locations across the surface to allow the calculation of the mean value and the standard deviation (SD).

3. Results and discussion

3.1. Flow behavior

Fig. 3 shows the values of H_r of the analyzed powder materials prior to and after modification. By functionalization of the PP powder, a considerable decrease of H_r, i. e. increase in flowability, is achieved. An increase in surface roughness and thus particle-particle distances lead to reduced adhesive forces within the PP* powder (Li et al., 2004). For TPE-U, sieving fractions < 63 µm and > 100 µm and mixing these in a weight percentage ratio of 50:50 results in the desired slight increase of H_r. Here, the reduced powder flow of very fine particles as a result of strong van der Waals forces (Li et al., 2004) is used to deliberately decrease overall flowability in TPE-U*.

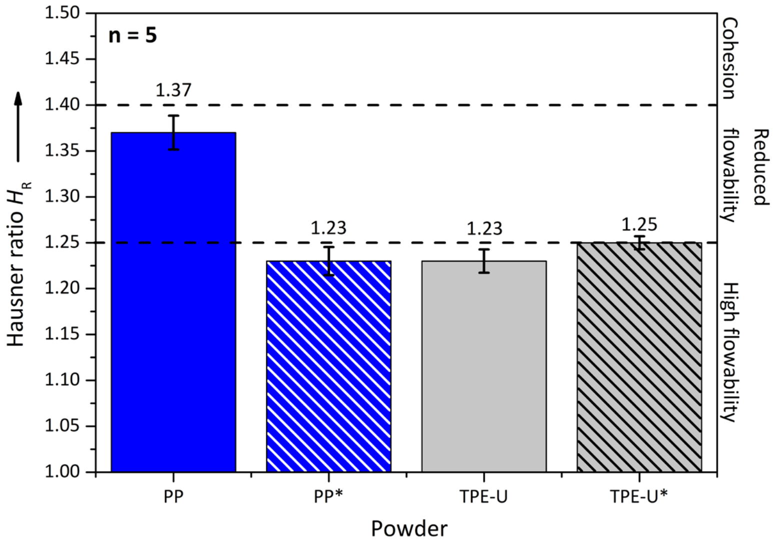

Fig. 3. Mean values of H_r of the as-received (PP, TPE-U) and the modified (PP*, TPE-U*) powders; error bars indicate SD; dashed horizontal lines separate the regions of high flowability (H_r < 1.25), reduced flowability (1.25 < H_r < 1.40) and cohesion (1.40 < H_r) based on VDI 3405 part 1.1

It must be mentioned that H_r is only an indicator of flowability and several factors (such as ambient temperature, humidity, triboelectric charging, etc.) influence the actual flow behavior during the very sensitive nozzle-based powder deposition. Hence, despite showing the same values of H_r, PP* and TPE-U do flow differently. When poured into N2, PP* shows a permanent mass flow without vibration, while powder bridges are formed in TPE-U in the same nozzle. The bridging in as-received TPE-U, however, occurs with a certain delay and as a result, residual mass flow can be observed even after stopping the vibration. Fractionation and mixing lead to a more reliable and faster formation of powder bridges of TPE-U* in N2. From this observation it can be concluded that, in addition to the conduction of standardized characterization methods (e. g. determination of H_r), the powder qualification for nozzle-based deposition requires the evaluation of the actual flow behavior during the process. This inspection of the deposition characteristic can be conducted by, for instance, a high-speed camera and/or a weighing cell, as published by Stichel et al. (2014) for a similar setup.

3.2. Prevention of early crystallization and inter-layer detachment using the adapted process route

Using the adapted process route successfully prevents early crystallization and curling between the layers, allowing the fabrication of multi-layered specimens. In contrast to the former process route, the temperature drop from T_{ph} (160 °C) is sufficiently small to avoid reaching of the crystallization onset temperature of PP* (approx. 132 °C). Cutting of the five-layered parts and optical analysis of the cross section confirms sufficient melting and inter-layer bonding. This is also a prerequisite for the preparation of thin films and the subsequent microscopic evaluation of the boundary zone.

For enhanced distinction between the structurally different polymers, a suitable polarization filter is used. A microscopic image of the boundary zone in a five-layered rectangular test part is depicted in Fig. 4.

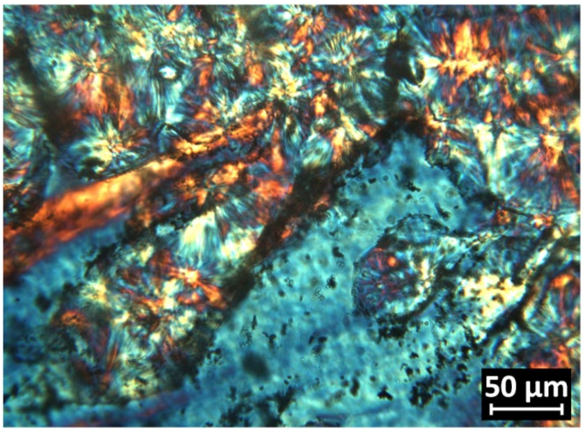

Fig. 4. Cross section of the boundary zone between PP* (multicolored) and TPE-U* (blue) in a five-layered test part

PP* (multicolored region) exhibits a fine microstructure with spherulites smaller than approx. 50 µm. It is suggested that the surface coating serves as a nucleating agent during the SLM process leading to the fine polymeric structure. Besides the dispersed and partially agglomerated carbon black particles, no conclusive evidence regarding the TPE-U* microstructure (blue region) can be extracted from Fig. 4. Most notably, however, the two polymers do not show a discrete border but a pronounced interlocking via the formation of microsized undercuts. This interlocking is suggested to result from the adapted process route (i. e. high temperature in the building chamber) allowing TPE-U* melt to flow into the pores in-between the PP* powder particles before laser irradiation. The subsequent melting of PP* then establishes a mixed zone of both melted polymers which provides mechanical adhesion in the joint of the solidified parts. Thus, a bonding based on mechanical interlocking between PP* and TPE-U* is achieved despite the fact that these materials are thermodynamically immiscible (Wang et al., 2006). Mechanically stable undercut-based boundaries between two incompatible polymers have been reported in former works that employed a simple two-chamber recoater in combination with the original irradiation strategy at low temperatures in the building chamber (Laumer et al., 2016).

A remaining limitation of the adapted process route, however, is a strong geometrical deviation of the generated multi-material parts from the specified shape. The reason for this is the increased processing duration (powder deposition times in the range of several minutes) which leads to thermal conduction from the melted area to the surrounding powder bed and hence to the observed blurring of the contour. To optimize part geometry and reduce the observed blurring of the contour, a reduction of deposition time is necessary to minimize lateral heat conduction to the surrounding powder bed. By increasing flowability, this process acceleration can only be achieved within a certain range, where powder bridges are still reliably formed after stopping the vibration. Another approach could be the parallelization of more nozzles to decrease the time necessary for layer preparation.

3.3. Part topography

A representative three-dimensional topographic image obtained from laser profile sensor analysis is shown in Fig. 5. Here, the displayed boundary zone between the materials clearly reveals a step of about 1 mm (i. e. height difference 200 µm per layer) due to the increased mass flow of TPE-U* compared to PP*. Furthermore, suitable irradiation (i. e. sufficient energy input) results in the vanishing of the groove-like topography characteristic for the nozzle-based deposition.

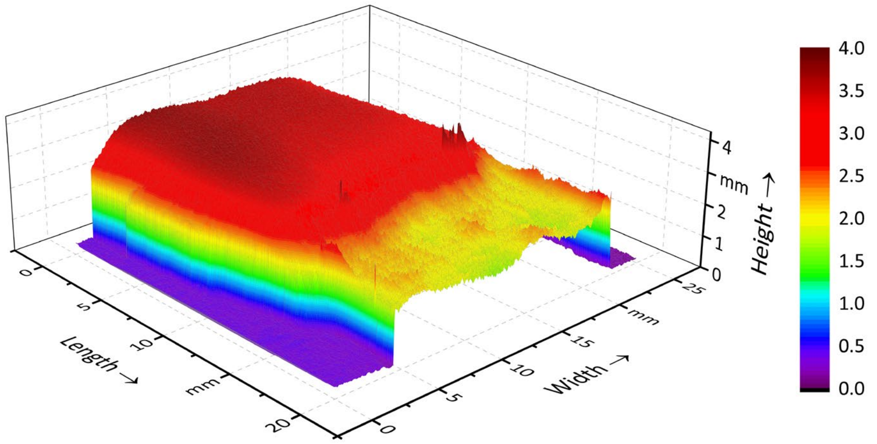

Fig. 5. Three-dimensional part topography in the boundary zone between PP* (front) and TPE-U* (back) obtained from laser profile sensor measurements

LSM analysis shows that the parts exhibit a smooth surface in both the PP* (S_a = 3.69 ± 0.63 µm) and the TPE-U* (S_a = 4.64 ± 1.65 µm) region whereas the roughness is slightly increased in the boundary zone (S_a = 13.11 ± 8.11 µm). The reason for this is the difference in mass flow between PP* and TPE-U* resulting in a rather pronounced height difference observable in the five-layered specimens. However, the overall roughness of parts generated by the adapted process route using the nozzle-based deposition is comparable to that of parts manufactured by conventional SLM machines that use blades or rollers as powder deposition systems (Delfs and Schmid, 2017).

4. Conclusion

In this work, an adapted process route for multi-material SLM of polymers deposited by vibration-controlled powder nozzles is presented. The former melting strategy, which led to large temperature gradients during SLM, undercooling of the melted polymer below its crystallization temperature and finally its detachment in the boundary zone, was modified by increasing the temperature in the building chamber. This approach successfully reduces temperature gradients and prevents an uncontrolled early crystallization of the high-melting polymer as well as its interfacial detachment. As demonstrated, the adapted process route allows the generation of multi-layered compounds with stable joints and complex geometries. However, the parts lack geometrical accuracy (i. e. exhibit blurred contours) as a result of the long processing times and heat conduction to the surrounding powder bed, respectively. In following works, this problem will be tackled by accelerating the deposition process which can be achieved, for instance, by parallelization of the powder nozzles. Furthermore, mechanical properties will be statistically investigated to quantify the strength of the joint and analyze the failure behavior.

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