Conventional stereolithography is limited to the processing of one material only and cannot be applied for novel multi-material fabrication techniques. We developed a new approach based on the aerosol jet system as a spray coating device for the generation of a liquid film. The integrated dual spray generator is able to deposit two pure materials and mixed material compositions. Hence, the process was executed iteratively with a liquid material layer generation with a thickness of 50 µm and a following UV laser direct write induced polymerization. We investigated the feasibility of model adaptive curing depth control to minimize the effect of overcuring by 62%. The spray coating-based printing opens a wide range of applications in the field of multi-material additive manufactured products.

**Keywords:** Additive manufacturing, Adaptive control, Aerosol jet, Laser direct write, Multi-material

1. **Introduction**

In the last years, stereolithography (SLA) as a representative of polymer based additive manufacturing principles reaches the stage of commercialization. Parallel printing on large areas enables the access to markets of conventional manufacturing mechanisms in the field of specialized products such as mechanics, optics and bio-applications [1-4]. The layer-by-layer process inside a vat filled with the curing material allows the iterative building of 3D parts: first a liquid material layer is generated by moving the building stage from the irradiation position either bottom up or inverted. Then the resin is polymerized locally by an irradiation source such as a laser [5,6]. Consequently, the subsequent liquid layer is established and repeated until the total height of the structure is achieved. The next step to individualized printed parts is the handling of multi-materials to open the opportunity of multifunctional parts [7]. The vat of an SLA system is filled with one material only and cannot be exchanged easily. To overcome this drawback, we investigated an alternative deposition approach using a spray coating device [8]. Such an aerosol jet system (AJS) can deposit materials in the range from 10 to over 1000 mPa·s and was developed for printed electronics [9-11]. Adapted for UV curable materials, the layer thickness can be controlled [12]. We integrated a dual material spray generator for the deposition of pure materials and mixed conditions as sketched in Fig. 1. Besides multi-material printing devices, SLA has to compete with printing principles that rise in printing resolution. In the case of the one photon polymerization based SLA, overcuring is a well-known effect [13].

![Fig. 1. Sketch of the aerosol jet system including the dual atomizer system.](image-url)
In general a curing depth greater than the actual layer thickness is chosen to ensure a proper connection between the layers [14]. Therefore, different approaches of vertical process resolution optimization can be identified: material curing properties by modifying the chemical composition [15] or the polymerization procedure [16]. In this study, we optimized the process parameters and process strategy demonstrating a model based curing depth control. During the printing operation, the process parameters are adapted depending on the previous layer. Known from other laser based additive manufacturing principles such as selective laser melting, first concepts of model assisted temperature control were shown [17]. In the context of SLA, the approach is adapted: The actual layer is analyzed with the previous layer as shown in Fig. 2. If there is an overhang, the related area is extracted from the standard curing schedule and is cured independently with an alternative set of curing parameters.

2. Experimental
2.1. Data processing
For the scheduling of the machine, the input data has to be prepared including process parameters and shape information. Hence, the data preparation starts with the import of an STL file into Magics 23 (Materialise GmbH, Germany), shown in Fig. 3. Subsequently, a machine scene is created which corresponds to the properties of the real printer in which the part is placed and opens the opportunity to repair or edit the model. The geometric data is then transferred to the Build Processor (Materialise GmbH, Germany), which performs the slicing. For multi-material approaches, features depending on the mixing strategy are adjusted: If two materials are deposited on the same layer, set biting patterns supports the connection of these materials. Pure materials and mixed material compositions are defined in built direction to utilize the mixing option of the AJS.

Fig. 2. Model adaptive curing strategy for a bridge structure (a), showing the first layer with an overhang (layer (n) in (b)) and the split into two slides with modified process parameters (c).

The Build Processor itself is linked to the printer and functions as the printer driver providing the machine data. In this study, the exported data contain the material composition information and the adjusted process parameters. The set number and size of pixels of the exported picture file for each slice determines the accuracy of the resolution. With an additional configuration file, the process parameters are saved and are read by the control program. It prepares the printing schedule, while the iterative layer-by-layer process is divided into four steps, shown in Fig. 4: (1) Deposition of the materials, (2) fixing the material film at a transparent layer in sandwiched condition, (3) UV polymerization, (4) release. The steps belong to the three main modules of the system: the aerosol jet executes the material deposition, the laser unit induces the chemical photoreaction, the controlled stage performs the fixing and releasing of the substrate at the transparent layer, shown in Fig. 5.

Fig. 3. Data handling: STL-file modification (blue), data preparation for printer (orange), printing control units (purple).

Fig. 4. Iterative layer-by-layer procedure with aerosol jet based material deposition and UV laser direct write.
2.2. Aerosol jet material deposition

The spray coating device (Aerosol Jet, Optomec Inc., Albuquerque, USA) realizes the material deposition [18]. Two atomizers and two virtual impactors generate the mist flow for each material independently. The generated droplets inside the mist flow show a typical droplet size of between 0.2 and 5.0 µm [19,20]. Tubing transports the mist flow through a mixing module and towards the nozzle that enables the spray coating function, shown in Fig. 5.

The atomizer gas flow rates in the range of between 500 and 4500 cm³/min control the mist flow. After that, each mist flow enters the virtual impactors. Inside these devices, an outlet gas flow extracts surplus gas, while the droplets are kept in the mist flow. After leaving the virtual impactor, the two independent generated liquid droplet mist flows, also called push flows (difference of the atomizer flow rate and virtual impactor flow rate) are combined in the mixing module. The push flows are set equal to avoid backflow. The combined push flow is guided to the nozzle and is deposited onto the substrate. The substrate is moved meander shaped ensuring a homogeneous layer thickness [21]. The push flows for both materials are set to 500 cm³/min, while the atomizer gas flow rates are changed in the range of between 600 and 3600 cm³/min. The investigated deposition rate is recorded by measuring a glass substrate that collects the material deposition for three minutes. The investigated temperatures cover the range of 60 °C to 80 °C. The materials used are the methacrylate compound FotoTec DLP.A (Dreve Otoplastik GmbH, Germany) further called material A, and material B consisting of (5-ethyl-1,3-dioxan-5-yl) methacrylate, aliphatic difunctional urethane acrylate, urethane acrylate, and 2-hydroxy-2-methylpropiophenon (6 wt%) as a photoinitiator.

2.3. UV laser direct write

The one photon polymerization reaction is initiated by an UV laser source (Zouk, Cobolt AB, CW, λ = 355 nm) that is controlled and shaped by an acoustic-optical modulator, a beam expander, a galvanometer scanning system, and an F-Θ lens. The scanning device controls the laser beam movement on the substrate. In this SLA approach, a local curing is processed by moving the laser beam meander shaped with a fixed line-to-line distance of 4 µm. A more detailed description can be found in [21]. The laser fluence determines the curing depth [22]:

\[ F = \frac{P}{v \cdot h} \]  

where \( P \), \( v \) and \( h \) is the laser power, the laser scanning speed and the line-to-line distance, respectively.

The model-adapted approach is executed by characterizing the materials first for the layer-by-layer printing by polymerizing a bridge shaped structure, see Fig. 2. The material was deposited manually and the layer thickness of 50 µm in the sandwiched condition was executed with the stage system, step 2 to step 4 in Fig. 4. This enabled the penetration of the already polymerized areas with a fresh material film for the next irradiation step. A two-dimensional pattern enables the single layer curing. After curing the base layer and 10 more layers for the pillars, the overhanging connecting part of the bridge was conducted for 10 more layers. The fluence was varied by changing the laser scanning speed between 1000 and 5200 mm/s at a laser power of 2 mW. The sample was then washed in a bath of isopropanol for three minutes. The sample was depicted and laid on aside to quantify the multilayer overhang with a light microscope (Leica Microsystems GmbH, Germany).

3. Results and discussion

3.1. Material deposition rate

The achievable material deposition rates of the two materials using the aerosol jet are depicted within Fig. 6 as a function of the atomizing gas flow rates at varied temperatures. The temperature increase causes a viscosity decrease.

Thus, an increase of the deposition rate is observed until an asymptotical limit inhibits a further increase. This limit is depending on the replacement of the material inside the Venturi nozzle. The atomizing threshold is correlating with the temperature and it is observed that material B needs a higher atomizing gas flow rate compared
to material A. The system conditions, namely the heating power and the temperature control, limit a stable material deposition for higher temperatures greater than 80 °C. With this and the lower atomizing threshold of material B, the material deposition rate was set to 10 mg/min for a reproducible and uniform material deposition for all material compositions.

With the determined AJS deposition parameters a free-form body is built performing the layer-by-layer process, shown in Fig. 7. The material composition is changed from the bottom to the top at material increments of 25 wt%. The size of the part is $19.75 \times 17.01 \times 16.97 \text{ mm}^3$ at a layer thickness of 50 µm.

### Table 1. Atomizer flow rate table for mixed material deposition (material A and material B).

| Material ratio Mat. A - Mat. B (wt. %) | Atomizer Flow Rate (cm³/min) Material A | Material B |
|---------------------------------------|----------------------------------------|------------|
| 100 – 0                               | 1900                                   | 500        |
| 75 – 25                               | 1850                                   | 1500       |
| 50 – 50                               | 1600                                   | 1850       |
| 25 – 75                               | 1350                                   | 2050       |
| 0 – 100                               | 500                                    | 2350       |

Fig. 6. Material deposition rate of material A (a) and material B (b) as a function of the atomizer flow rate and the temperature.

The slow temperature change inhibits this parameter as a control variable. Hence, the temperature is fixed to 60 °C and 80° C for material A and material B, respectively. The atomizing flow rate for the mixed material conditions were adapted on the set material temperatures and are identified to the values shown in Table 1.

The degree of overpolymerization is shown in Fig. 8. The increasing fluence, modified by a decreasing set scanning speed, causes an increasing overcuring (transition in Fig. 8 (b) to (c)). The curing depth increases with the logarithm of the laser fluence and is following the Lambert-Beer law of absorption [22].

In Fig. 8 (a) it can be seen that the fluence was not reaching a threshold for a connection of the pillars. For the model adapted curing control, the utilized shape was split as shown in Fig. 2 (c). The pillars are polymerized with a fluence of 10.05 mJ/cm² for a curing depth of 70 µm. The first layers of the connecting part are irradiated with the fluences 9.43, 9.63, and 9.84 mJ/cm² for adapted layer thicknesses of 40, 50, and 60 µm, respectively.

![Fig. 6](image6.png)

![Fig. 7](image7.png)

![Fig. 8](image8.png)
Then, the following layers are irradiated with a fluence of corresponding the curing depth of 70 µm again. As expected, the adapted layer thickness of 50 µm results in the best sample, shown in Fig. 9 (b) with an overpolymerization of 38 µm. If the curing depth is too low, the overhanging structure is tapered and a proper built is less accurate (Fig. 9 (a)), in the opposite case overcuring is significant with 91 µm (Fig. 9 (c)). The model-based adaption is reducing the overcure length by 63 µm or 62%, while the default curing depth with 10.05 mJ/cm² ensures an interlayer connection.

4. Conclusion
In this study, we have demonstrated the adapted material deposition with a new approach for a multi-material stereolithography principle using the aerosol jet system. The temperature adjustment increased the material deposition significantly (material A: 60 °C, material B: 80 °C), so that a constant deposition rate of 10 mg/min was achieved for the pure materials and for three mixed conditions. The UV laser initiated polymerization was adjusted in terms of a model based curing adaption for material A at a layer thickness of 50 µm. Hence, the fluence was optimized to 9.63 mJ/cm² for overhanging areas at the actual layer in the layer-by-layer procedure and to 10.05 mJ/cm² for the remaining areas that are connected to the previous polymerized layers resulting in a successful interlayer connection. Consequently overcure length was reduced by 62%.

Acknowledgements
The authors want to thank Linda Thies and Hendrik Stürmer. This work was supported within the Project 3D Polysprint (FKZ: 13N13567), funded by German Federal Ministry for Education and Research (BMBF).

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