Micro-powder injection molding: Investigation of powder-binder separation using synchrotron-based micro-tomography and 3D image analysis

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Abstract
Micro-powder injection molding (µ-PIM) is one of the most promising processes of mass production for the fabrication of small complexly shaped ceramic or metallic parts with high sintered density. However, dimensional accuracy of finished parts is difficult to achieve because of extremely high shear rates during the injection molding process. This promotes the separation of powder and binder even in highly homogeneous feedstocks leading to a particle density variation in the green part causing anisotropic shrinkage during sintering. The main objective of this study is to investigate the effect of the powder particle distribution in injection molded green metallic microparts with respect to the molding parameters using synchrotron micro-tomography (S-µCT) and three-dimensional (3D) image evaluation. Image analysis has been performed using the MAVI software package. To get information about the allocation of the metal particles along the sample the 3D CT-scans have been segmented and statistically analyzed via spatially resolved size distributions. Furthermore, the spatial arrangement of the particles has been investigated using the so-called summary statistics from the area of point process statistics. The results show that variations in the size distribution of the metal powder particles can be detected and give consistent evidence for a monotonic increase in particle size with distance to the injection point. In order to give recommendations for the choice of parameters as well as tool construction, knowledge about the causes for separation effects is essential. The presented work shows that S-µCT is a well-adapted analytical tool to investigate the powder-particle distribution in µ-PIM.

Keywords
Micro-powder injection molding, MIM, powder-binder separation, synchrotron radiation, micro-tomography, 3D image analysis

Introduction
Micro-powder injection molding (µ-PIM) allows for industry-related and low-cost production of small complexly shaped ceramic or metallic microparts with high sintered density. The process combines the advantages of plastic injection molding techniques with the traditional powder metallurgy [1]. It consists of feedstock compounding from fine powder and thermoplastic polymers, shaping to a green body via micro-powder injection molding and two post processing steps namely solvent and/or thermal debinding and sintering. These process steps have been extensively studied and are described in the literature in detail [1].

The main challenge for the production of micro-injection molded parts is to achieve dimensional accuracy at the end of the process chain. Due to the multi-phase character of feedstock systems the type of processing applied can induce inhomogeneity in green parts and therefore cause inhomogeneous binder extraction or anisotropic shrinkage during sintering, resulting in visual defects such as deformations, wrapping or cracks. Most of these defects may already appear when the molding compound is injected into the cavity, because during the injection process extremely high shears rates of up to 10^6 s^{-1} [2] arise due to the high speed and the high pressure applied. This leads to powder binder separations or orientation effects of the powder particles in the flow direction and directly affects all following processing steps [2,3]. Unfortunately, a direct correlation between the pre-cursor material and the final product is often impossible. Numerous experimental tests are required to find relationships between the influence of feedstock properties and processing parameters on the mold behavior during injection molding and the quality of resulting parts. Thus, it is essential to know the particle density distribution in the molded parts in order to optimize the process and/or to find the parameter window most suitable for the quality expected.
In this paper, the main focus is put on the phase segregation effects which are generated during the shaping step of the µ-PIM process. The influence of the machine settings on the resulting particle distribution of metal powder in the thermoplastic matrix was investigated by synchrotron micro-tomography and the data acquired was statistically analyzed using the MAVI software package [4-6].

Experimental Procedure and Methods

Material selection and feedstock preparation

The choice of suitable powder materials for µ-PIM depends mainly on the application [2]. Major attention is paid to powders with small particle size, which should be at least about one order of magnitude smaller than the minimal dimension of the finished micro-part [1]. Figure 1a shows the particle size distribution of stainless steel powder (17-4 PH, CARPENTER Technology Corp., USA) used for our research. The measurement was performed by a Microtrac X100 laser diffraction particle size analyzer. The particle size varies from about 3 to 50 µm with an average size of 14 µm. A Scanning Electron Microscopy (SEM) image of the metal powder is given in Figure 1b. The majority of the particles possess a spherical geometry close to perfection. These powder properties (large particle size, spherical shape) allow for a successful visualization of single particles by synchrotron-based X-ray micro-tomography and facilitate subsequent image analysis [7]

Fig. 1 a) Particle size distribution of the stainless steel powder used (17-4 PH d90 < 22 µm, CARPENTER Technology Corp., USA) and b) SEM image of the particles

A proprietary binder system based on polyethylene and wax [8] was used for feedstock preparation. The compounding of the binder components and 63 vol.% metal powder was performed at 125°C in a kneader machine (W50 EHT, Brabender, Germany). The mixing rotation frequency was set to 30 rpm, the compounding time lasted 60 min. After compounding the material was subsequently granulated for further processing.

Injection molding

Shaping experiments were carried out using a Microsystem 50 injection molding machine (Battenfeld GmbH, Germany) using either isothermal or variothermal process control. A flexural micro-specimen geometry was chosen. In Figure 2, a 3D model of this microstructure together with the gating system is illustrated. The bar type micro-structure shows a quadratic

cross section with an edge length of 260 µm and an overall length of 3250 µm (Figure 2). An intended break is designed at the beginning of the bar type test structure that is located at the end of the flow direction. An overview of the samples studied and the corresponding injection parameters are compiled in Table 1.

Fig. 2 3D model of the flexural micro-specimen. The feedstock enters the cavity at the gating system, the investigated microstructure is located at the end of flow length

Table 1 List of the different injection molding parameters. The feedstock material contained 63 vol.% of metal powder in a polyethylene-wax-based binder system

Sample ID	Processing	nozzle temp. [°C]	mold temp. [°C]	plunger velocity [mm/s]
A2, A3	variotherm	50/45	50/45	500
A8, A9	isotherm	45	45	500
A12	isotherm	45	45	50

Synchrotron-based X-ray micro-tomography

Micro-tomographic scans were performed at the beamline ID19 of the European Synchrotron Radiation Facility (ESRF) [9]. Specific details about hard X-ray micro-tomography using laboratory-based or synchrotron light sources are published in [4,5]. For this experiment, a high resolution indirect pixel detector as frequently used for synchrotron micro-imaging was employed [10]. The system consisted of a visible light microscope operating with an effective 20x magnification (NA 0.3 of the front objective) which projected the luminescence image of a 6.2-µm-thick GGG:Eu (Eu-doped $Gd_3Ga_5O_{12}$, grown on top of an undoped gadolinium gallium garnet substrate) single-crystal thin film scintillator onto the CCD chip of a FReLoN camera (type 2k14-e2v) [11-12]. The detector operated with an effective pixel size of 0.7 µm. Based on the experiences from a previous experiment, the detector's corresponding resolving power is considered to be sufficient to depict the small features within the samples [7]. A X-ray photon energy of 27 keV was selected via a multilayer monochromator. 2000 projection images were recorded per 180 degree scan. Reconstruction of the volume images by means of filtered back-projection was performed via the ESRF software package PyHST [6, 13].

Image processing

Image processing on the reconstructed tomographic volume data sets was performed using the MAVI software package developed by the Fraunhofer ITWM [14]. The volume images were first smoothed using a 3x3x3 pixel median filter and binarized using a manually chosen, global gray value threshold. In order to separate single powder particles in the image, the watershed transform was applied to an inverted Euclidean distance map computed on the binarized volume images [15]. This well-established strategy often produces inaccurate segmentation results due to noise and artifacts in the preceding binarization step(s). Therefore, so-called pre-flooding was employed, see e.g. [15,16], using a volume threshold of 10 pixels to prevent oversegmentation. These image processing steps are displayed in Figure 3 using an example slice showing a cross-section in the flexural specimen.

Fig. 3 Image processing steps, from left to right: a) example slice taken from the gray-scale volume images, b) the corresponding binarized image of the powder particles, c) the separated and individually labeled particles

From the resulting labeled image, geometric characteristics of the single particles can be estimated using MAVI's "ObjectFeatures" [14] functionality. In the following sections, the particle volumes and 3D-coordinates (centers of circumscribed cuboids) will be used. In order to remove noise and artifacts of the particle separation from the data, only those particles with a diameter larger than 3 µm will be included in all following analysis results. This is valid because it is known from the particle size distribution measurement (Figure 1), that the smallest particles are larger than this value. In Figure 4 the spatial arrangement of powder particles inside the investigated section of the flexural micro-specimen is visualized.

Fig.4 Longitudinal cut through the volume data of the separated particles after image processing. The dashed line represents the position of the intended break. The different color values symbolize the segmentation result, similar to Figure 3c

In order to investigate changes in the spatial arrangement of the particles along the flow direction, each dataset was divided into 10 stacks of slices, 130 µm thick each, arranged along the injection direction (Figure 5). Within each stack, the maximal cuboid which is completely filled by material was determined such that each particle centered in the cuboid is completely contained within the image. The following statistics were computed for all particles centered in the respective cuboid ("minus sampling" [17]).

Fig. 5 Longitudinal section along the flexural micro-specimen indicating the analyzed ten stacks. The distance in the flow direction between each vertical line is 130 µm, corresponding to the thickness of each stack

Particle size distribution

In order to measure influences of injection parameters on the spatial particle size distribution, each particle was characterized in terms of its diameter. The diameter d of a particle with volume v is computed as the diameter of an equivalent sphere with the same volume v, i.e., $d = (6v/\pi)^{1/3}$. The mean values of the diameters in each stack are plotted in Figure 6.

Point process statistics

For further analysis, we assume that the point patterns of particle centers in each sub-region are realizations of a stationary and isotropic point process, i.e., invariant under translations and rotations of space. A variety of well-established summary statistics [17] to characterize such point processes is available. Here, we consider the nearest-neighbor distance distribution function $G(r)$. Further distribution functions were applied but are not described here to ensure clearness.

The nearest-neighbor distance distribution function is the distribution function of the distance from a point of the point process to its nearest neighbor, i.e., $G(r)$ is the probability that the distance from a point to its nearest neighbor is smaller than r.

In practice, $G(r)$ is estimated from the empirical distribution function. However, edge correction techniques have to be applied since the nearest neighbor of a point might fall outside the observation window. We use the so-called nearest-neighbor estimator ([17]) which takes only points into account whose nearest neighbor is closer than the boundary of the observation window.

The G-functions for each of the 10 stacks were estimated. From the trend in the particle sizes it is evident that the whole sample is not stationary. However, by subdividing the sample into sub-regions the deviation from the static state in each sub-region is small and justifies the stationarity assumption. The results are shown in Figures 7 and 8.

Results and discussion

To determine the expected powder-binder separation, the average particle sizes were determined. The particle size distributions in all specimens are slightly increasing (Figure 6). However, a standard deviation of ± 4 µm for the analysis has to be considered.

Fig. 6 Metal particle size distributions in flexural micro-specimen along the flow direction: average diameter of the corresponding sphere as a function of the distance from injection gate. The dashed line represents the position of the intended break

To verify the origin of the increase in particle size, we additionally consider the nearest-neighbor distance distribution function *G(r)* in each stack and its variation between the 10 stacks. As expected, the estimated nearest-neighbor distance distribution functions show significant deviation from complete spatial randomness towards a more regular spatial distribution (Figure 7). The change in the summary statistics with increasing distance from the injection gate is consistent with the increase in the particle diameters observed. By considering the shift of *G(r)* along the specimen in the flow direction the increasing trend for the particle size can be ascertained. This trend was observed in each specimen, but depending on the considered sample it was differently expressed. Figure 7 exemplarily shows the *G(r)* behavior for two specimens molded with different plunger velocities (Table 1).

Fig. 7 Distribution functions *G(r)* of the nearest-neighbor distance *r* for four selected stacks in two specimens representing different plunger velocities

To determine the influence of different processing parameters on the spatial particle arrangement in the defined sub-regions, the distribution functions of all specimens were compared with respect to the position. Figure 8 displays the results for four different sub-sections. Only very small differences between the specimens, i.e. different processing parameters, are detectable.

Fig. 8 Comparison of the distribution functions *G(r)* of nearest-neighbor distance r in the specimens for four selected stacks

Conclusion

The results presented in this article highlight that synchrotron-based micro-tomography combined with 3D image analysis is a powerful analytical probe to detect and quantify powder binder separation effects. However, concessions are necessary regarding the minimum particle size: the spatial resolution of established micro-tomography systems is currently not suited to resolve particle sizes below one micrometer in combination with a field of view large enough to depict our samples. Nevertheless, variations in the size distribution of metal powder particles can be analyzed. Evidence for a monotonic increase of the particle size with its distance to the injection gate was found using the MAVI software package and confirmed by point process statistics using the nearest-neighbor distance distribution function. Further methods from spatial statistics are being applied to validate the current results. One possibility is to use second-order summary statistics such as Ripley's K-function *K(r)* which are believed to be more powerful than *G(r)*. The S-µCT experiments presented refer to a limited section of the whole flexural micro-specimen due to limitations introduced by the amount of data and available synchrotron beam time. In future works, the complete flow length will be investigated. This is required in order to optimize the ratio between the values for the particle size which increase in the flow direction and the standard deviation of the distribution in each stack. Additionally the particle size and particle density distribution

vertically to the flow direction and the density distribution along the flow direction are currently under investigation.
This paper is presenting the first step for defining a measurement procedure. The final aim of the research activities are guidelines for tool construction and the choice of parameters for the production of defect free, dense micro-parts produced by powder-injection molding.

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References

[1] RM German (1990) Powder Injection Moulding. Metal Powder Industries Federation, Princeton
[2] R Heldele, S Rath, L Merz, R Butzbach, M Hagelstein, J Hausselt (2006) Nucl Instrum Meth B 246: 211. Doi:10.1016/j.nimb.2005.12.030
[3] JC Gelin, T Barriere, J Song (2010) J Eng Mater-T Asme 132. Doi:10.1115/1.2931155
[4] J Banhart (2008) Oxford Univ. Press, Oxford
[5] SR Stock (2008) MicroComputed Tomography: Methodology and Applications. CRC Press, London, New York, Boca Raton
[6] A Mirone, R Wilcke, A Hammersley, C Ferrero (2010) ESRF, Grenoble
[7] A Rack, T Weitkamp, SB Trabelsi, et al. (2009) Nucl Instrum Meth B 267: 1978. Doi:10.1016/j.nimb.2009.04.002
[8] R Heldele (2008) PhD-Thesis, Department of Microsystems Engineering, University of Freiburg, Freiburg
[9] T Weitkamp, P Tafforeau, E Boller, et al. (2010) X-Ray Optics and Microanalysis, Proceedings 1221: 33.
[10] A Koch, C Raven, P Spanne, A Snigirev (1998) J Opt Soc Am A 15: 1940.
[11] T Martin, A Koch (2006) J Synchrotron Radiat 13: 180. Doi:10.1107/S0909049506000550
[12] JC Labiche, O Mathon, S Pascarelli, et al. (2007) Rev Sci Instrum 78. Doi:10.1063/1.2783112
[13] AC Kak, M Slaney (2001) Principles of computerized tomographic imaging. siam-Verl., Philadelphia
[14] (2010) Fraunhofer IWTM, Department of Image Processing, Kaiserslautern
[15] J Ohser, K Schladitz (2009) 3d images of materials structures - Processing and analysis. Wiley-VCH, Weinheim
[16] FB Tek, AG Dempster, I Kale (2005) Comp Imag Vis 30: 441.
[17] J Illian (2008) Statistical analysis and modelling of spatial point patternsWiley, Chichester [a.o.], Section 4.2.2 and 4.2.6