Additive Manufacturing of Ceramics by Composite Microextrusion

D. Penner, A. Fassbind, R. Henke, St. Mauchle, Y. de Hazan

A concept for Additive Manufacturing (AM) of ceramics by a modified Fused Deposition Modeling (FDM) process is presented. Instead of feeding a 3D-printer with a prefabricated filament, the feedstock consists of a ceramic powder/polymer composite granulate. The granulates are fed to a pressure and temperature controlled twin screw extruder where the material is extruded continuously to fine filaments through a small nozzle. These filaments are deposited and arranged to pre-determined 3D-structures by controlled 3-axis movement of the extruder. Printed structures ranging from dense monolithic bars to honeycombs with varying filling levels to shallow angle thin wall dishes are demonstrated. Detailed studies of the combination of varying process parameters are required to yield optimized print results comparable to those of commercial FDM printers.

Introduction

Additive Manufacturing (AM) of ceramics could be realized by different processes, which typically adapt procedures initially developed for polymers or metals. Currently stereolithography [1, 2] and powder printing [3] are the main techniques applied in an industrial context. Furthermore processes like laser sintering, fused deposition modeling, paste extrusion or direct ink jet printing were intensively investigated but not implemented yet on a wide scale into industrial processes.

The main reason is the discrepancy between achievable technical properties, tolerances and production speed in comparison to established ceramic production processes. Travitzky et al. [4] and Zocca et al. [5] reviewed available techniques for AM of ceramics, their applications and characteristics.

Fused Deposition Modeling (FDM) is the most common AM process, mainly by using the two well suitable polymers Acrylonitrile Butadiene Styrene (ABS) or Poly Lactic Acid (PLA). This process is used in a range from home consumer market up to professional applications. There are hundreds of different models available and low cost desktop devices are sold for less than EUR 1000. They are all based on the principle of microextrusion of a thin molten polymer filament which is line- and layerwise deposited and fused by software controlled movement to achieve the predefined structure. Where software parameters are properly set and devices are well constructed, the results of these polymer printing procedures are often somehow impressive.

As a consequence it is obvious to try to apply the basic principle of FDM to produce ceramics via a composite route by analogy to materials used in ceramic injection molding [6]. Since more than twenty years, e.g. Bandyopadhyay et al. [7] (and references therein) or Jafari et al. [8] demonstrated successfully the feasibility of such an approach.

In the past few years many low cost or free software and control optimizations became available, driven by the mass distribution of polymer FDM printers, which could help to improve the ceramic composite printing process and reduce its cost.

Since some years, students of materials and process engineering and mechanical engineering at the ZHAW Zurich University of Applied Sciences investigated such processes during student projects and bachelor thesis. Initially, alumina polymer composite filaments to feed standard FDM printers were prepared, characterized and investigated. It turned out, that it is difficult to achieve composite filaments having both sufficient strength and sufficiently high solid loading. Although this might be still a straightforward approach appropriate for home use and principal feasibility was demonstrated we encountered some general drawbacks. Due to the necessary high solid loading the prepared composite filaments were too brittle, tend to clog the nozzles, did not melt in a proper range, had a too high viscosity or were difficult to debind.

As a consequence a print head was developed based on the principle of microextrusion similar to the approach described by Bellini et al. [9] and mounted on a 3-axis movement system. It should be noted, that this general approach might be used as well

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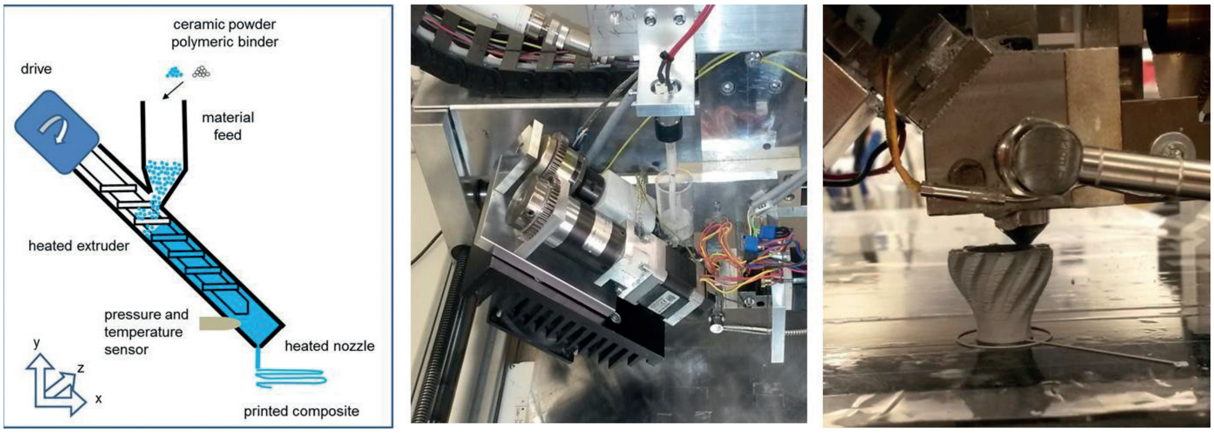

Fig. 1 3D-printer via microextrusion filament printing: concept (l.), print head (middle), and material ejection through nozzle (r.)

for any other material which is extrudable in the available temperature range of the instrument.

Experimental

The granulate material used consisted of Al_2O_3 (ALTEO P172LSB, D50 0,4 µm), stearic acid as dispersion aid and ethylene vinyl acetate (EVA) polymer. Generally, any other ceramic raw materials like e.g. zirconia could be used as basis of a feedstock. All components were compounded in a Haake Process 11 twin screw extruder and afterwards ground in a simple kitchen blender under liquid nitrogen. The appropriate particle size fraction of 200–400 µm was separated by sieving. This granulate material serves as feed for the printer.

The granulate material is feed into the print head through a slowly stirred vertical storage vessel. The print head (Fig. 1) consists of a micro twin screw extruder, driven by a stepper motor and equipped with a temperature and pressure sensor. The extrusion unit is heated by a heating cartridge. The exchangeable nozzle has variable diameters in the range of 100–450 µm. The print head is mounted on a commercial 3-axis milling machine and the whole system is controlled and driven by an Arduino Rambo interface card.

The software selection for print control and slicing of objects plays an important role for achievable print results. A choice of commercial software solutions is available (e.g. Slic3r, Craftware, Kissslicer, Simplify3D, Cura), either as freeware or under licensing conditions. Some programs have better options for infill parameters but problems with printing of sharp objects such as corners, others print unwanted lines or are generally unreliable. All programs allow the variation of countless numbers of parameter combinations which all could significantly influence the printing results. Adjustable parameters beside print speed and extrusion rate are e.g. z-offset, overlap, bottom- and top-layer, perimeter, infill etc.

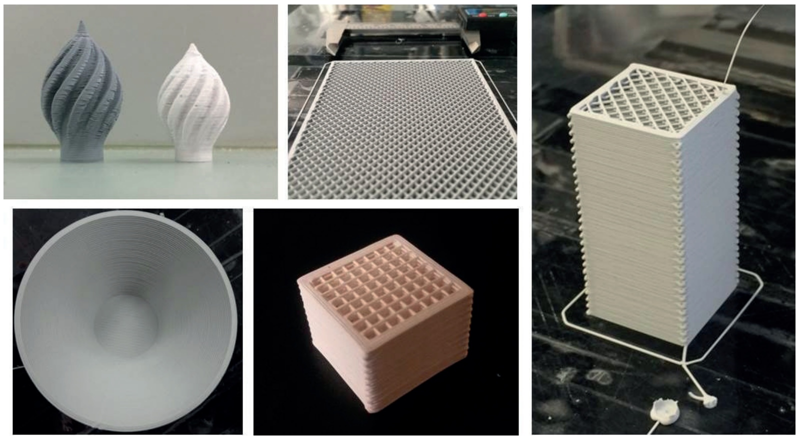

Fig. 2 Selection of fired and unfired samples

Finally, the printed composite has to be transferred into a ceramic article by debinding and sintering. Currently, a debinding and firing schedule of nearly 100 h and a maximum sintering temperature of 1500 °C are used.

Results

After numerous software and process parameter and optimization studies the developed printer is able to generate articles which are comparable to those achievable with standard ABS polymer FDM printers. Fig. 2 shows examples of articles printed by microextrusion of feedstock granulate. Either scaffold structures or partly to fully filled structures were produced. As a process inherent feature, the surface layer and infill structure remain visible. To reduce this visibility and to achieve fully dense articles, detailed parameter studies have to be continued and optimization of hardware has to be implemented.

Fig. 3–4 show examples of optimization studies. Fig. 3 shows two different V- or bridge tests [10] to find a suitable process window for extrusion temperature, speed, pressure and lateral extruder movement velocity. Larger distances might be bridged by the introduction of (sacrificial, e.g. water soluble) support structures. Fig. 4 shows test results for the determination of process parameters to achieve minimum wall inclination angles. In this example, angles low as 25° were achieved for green bodies. It is worth mentioning that such thin structures need support by a powder bed during debinding and sintering, and that dimensional precision after debinding and sintering of thin walls could be an issue. The

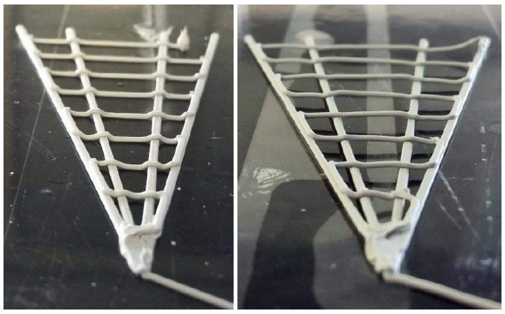

Fig. 3 **V- or bridge test: 14 bar extrusion pressure (l.), and 12 bar extrusion pressure (r.)**

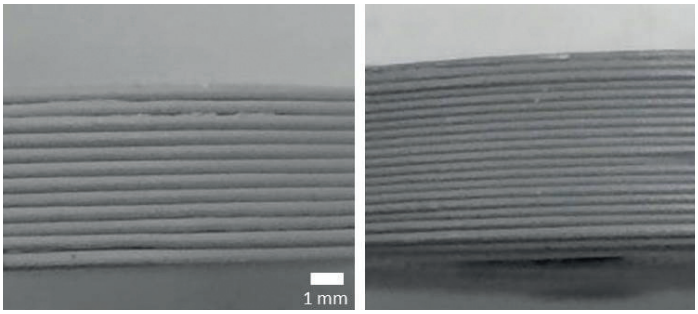

Fig. 5 **Comparison of resolution with nozzle opening diameter of 0,45 mm (l.), and 0,38 mm (r.)**

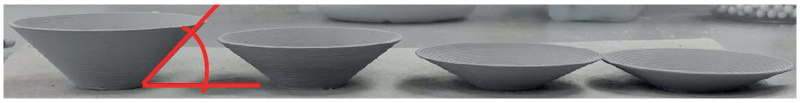

Fig. 4 **Print of dishes for determination of minimum wall inclination angle**

resolution of the printer can be increased to a certain degree by using nozzles with smaller diameter.

Fig. 5 shows examples for the use of 450 µm and 380 µm nozzles. In any case, the nozzle opening is subjected to abrasion due to the hard ceramic particles, and smaller nozzles are prone to clogging. Another source of dimensional errors is unwanted adhesion of material on the nozzle surface partially blocking the nozzle tip, which may occur if the distance between build platform and nozzle is significantly lower than the filament diameter.

Finally, debinding and sintering result up to now in articles with around 92 % of theoretical density. This relatively low value is a consequence of incomplete filling and voids between the deposited filaments. As mentioned above, further detailed studies of printing parameters in relation to rheological properties of the compounds are likely to yield bodies with higher densities. Articles with larger volumes may suffer from cracks and pores due to internal pressure and stresses developed during debinding procedures.

All known approaches for improvement from the field of debinding of ceramic injection molding parts may apply here as well. Debinding could be supported by wick powder beds, partly soluble binders, catalytic debinding, and optimized debinding temperature profiles.

Summary

Additive Manufacturing by microextrusion or Fused Deposition Modeling of ceramic composites can be used for the production of ceramic articles following the general principle of Fused Deposition Modeling. Architectures ranging from dense monolithic bars to honeycombs with varying filling levels to shallow angle thin wall dishes could be printed successfully.

Nevertheless, the print results reveal inherent process characteristics, especially a wavy surface structure due to layerwise filament deposition, limitations in geometrical resolution and limitations of spanning distances. Nevertheless, optimization potential is obvious as polymer fused deposition modeling demonstrates the capabilities of the process. Finally, post shaping processes rely on the optimization approaches originally developed for ceramic injection molding.

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References

[1] Griffith, M.L.; Halloran, J.W.: Freeform Fabrication of Ceramics via Stereolithography. J. of the Amer. Ceram. Soc. **79** (1996) [10] 2601–2608

[2] Schwentenwein, M.; Homa, J.: Additive manufacturing of dense alumina ceramics. Int. J. of Applied Ceram. Technology **12** (2015) [1] 1–7

[3] Sachs, E.; Cima, M.; Williams, P.; Brancazio, D.; Cornie, J.: Three Dimensional Printing: Rapid Tooling and Prototypes Directly from a CAD Model. J. of Engineering for Industry 114 (1992) [4] 481–488

[4] Travitzky, N.; Bonet, A.; Dermeik, B.; Fey, T.; Filbert-Demut, I.; Schlier, L.; Schlordt, T.; Greil, P.: Additive Manufacturing of Ceramic-Based Materials. Advanced Engineering Materials **16** (2014) [6] 729–754

[5] Zocca, A.; Colombo, P.; Gomes, C.M.; Günster, J.: Additive manufacturing of ceramics: issues, potentialities, and opportunities. J. of the Amer. Ceram. Soc. **98** (2015) [7] 1983–2001

[6] Penner, D.: Technische Keramik – Eine neue Herausforderung für den 3D-Druck. Swiss Engineering – Materials Now! **1** (2013) 12–13

[7] Bandyopadhyay, A.; Panda, R.K.; McNulty, T.F.; Mohammadi, F.; Danforth, S.C.; Safari, A.: Piezoelectric ceramics and composites via rapid prototyping techniques. Rapid Prototyping J. **4** (1998) [1] 37–49

[8] Jafari, M.; Han, W.; Mohammadi, F.; Safari, A.; Danforth, S.C.; Langrana, N.: A novel system for fused deposition of advanced multiple ceramics. Rapid Prototyping J. **6** (2000) [3] 161–175

[9] Bellini, A.; Shor, L.; Guceri, S.I.: New developments in fused deposition modeling of ceramics. Rapid Prototyping J. **11** (2005) [4] 214–220

[10] Smay, J. E.; Cesarano, J.; Lewis, J. A.: Colloidal inks for directed assembly of 3-D periodic structures. Langmuir **18** (2002) [14] 5429–5437