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Preparation of Highly Porous Carbonous Electrodes by Selective Laser Sintering

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Keywords: Porous Electrodes, 3D printing, Selective Laser Sintering, Conductivity, Graphite

Abstract

Selective Laser Sintering (SLS) 3D printing was utilized to fabricate highly porous carbonous electrodes. The electrodes were prepared by using a mixture of fine graphite powder and either polyamide-12, polystyrene or polyurethane polymer powder as SLS printing material. During the printing process the graphite powder was dispersed uniformly on the supporting polymer matrix. Graphite's concentration in the mixture was varied between 5 and 40 wt-% to find the correlation between the carbon content and conductivity. The graphite concentration, polymer matrix and the printing conditions all had an impact on the final conductivity. Due to the SLS printing technique, all the 3D printed electrodes were highly porous. By using polyurethane as the supporting matrix it was possible to produce flexible electrodes in which the conductivity is sensitive to pressure and mechanical stress. Physical properties such as graphite distribution, attachment and the overall porosity of the printed electrodes were studied using scanning electron microscopy (SEM), helium ion microscopy (HIM) and X-ray tomography. The results show that the combination of chemical design of the printing material and the

utilization of SLS 3D printing enables fabrication of highly customizable electrodes with desired chemical, physical, mechanical, and flow-through properties.

Introduction

Increasing demand for high performance batteries in wide variety of electronic applications from portable electronic devices to electric cars has generated a need for new approaches in battery design.[1,2] Especially the development of novel electrodes has gained a lot of interest. [3–5] Obviously, the most important property of an electrode is its conductivity but other features such as surface area and porosity can also have a significant impact on the electrodes' electrochemical properties and their applicability from more conventional systems to redox flow batteries.[6] When considering a typical graphite electrode, the achievable surface area consists only of the outer layer of the electrode the inner parts of the electrode are not accessible. Several methods to increase the surface area have been proposed, including the addition of nanomaterials into the graphite, the usage of different templating methods and the use of organic monolayered additives.[7–12]

During the last decade, three-dimensional printing has also been exploited in fabrication of highly customizable electrochemical devices.[13,14] Several papers have been published for example of 3D printed graphene-based electrodes and microbatteries.[15–20] In these studies, three-dimensional printing has enabled a rapid production of components and devices for electrochemical applications. However, in most cases the 3D printing technique used has been either Fused Deposition Modeling (FDM) or Direct Ink Writing (DIW), while other 3D printing techniques have received little to no interest. Both FDM and DIW produce surfaces that are not inherently porous and the methods often require specifically customized printing materials. In principle, a wide range of commercial materials are available but fine-tuning these printing materials for specific applications while simultaneously trying to match the requirements of a specific printing method can be laborious and demanding. From this point of view powder-based methods, such as Selective Laser Sintering (SLS), can provide a way to avoid some of these pitfalls. For example, with the SLS 3D printing technique, it is possible to produce objects that are inherently porous. In SLS printing, small particles with a typical diameter of 50–100 μm are fused together by laser which gives, at least up to a point, control over the physical

characteristics such as the porosity and mechanical strength of the material by fine-tuning the printing parameters such as laser power, exposure time and printing temperature.[21–24] When the particles are sintered in such a way that only their surfaces are partially melted, a solid structure containing accessible voids between the sintered grains is obtained. With powder-based methods, it is also relatively easy to add functional additives into the printed object by simply mixing the additive with the printable matrix. If the matrix particles are only partially sintered, the additive is dispersed on the surface of these particles and therefore accessible by fluids flowing through the printed object enabling applications such as porous flow-through columns for scavenging metal ions from aqueous solutions.[25] The SLS technique also allows the alteration of physical properties such as the density of the printed objects within the object itself. This means that different areas of the 3D printed object can be tuned to have different porosities and objects with well-defined permeable and impermeable areas can be made.[25] This in turn broadens the range of plausible applications for the printed objects.

In SLS 3D printing, the most commonly used printing materials are simple polymers such as polyamides, polypropylene or polystyrene. These polymers are nonconductive

by nature and therefore not suitable as electrode materials. Obviously, conducting polymers could be used to obtain electrodes. However, the chemical nature of the electrode is also dependent of the material. Furthermore, the conductive polymers are often relatively expensive.[26] Therefore, use of a conductive component only as an additive seems like an appealing option. The SLS technique sets only minimal requirements for the choice of the functional additives as nearly any components can be mixed within the matrix powder. There is a wide range of different conductive components available, from conductive polymers to carbon nanotubes and metals, which could potentially be used as conductive additives. Simple graphite, however, is a good example of an additive that possesses conductivity and is still rather cost-efficient. In this work, graphite powder mixed into either polyamide-12, polystyrene or polyurethane matrix was used for preparation of highly porous carbonous electrodes using SLS 3D printing. The graphite content plays key role in the conductivity and therefore the impact of the graphite concentration was investigated to find the optimum graphite/matrix ratio.

Results and Discussion

The printing material for the fabrication of the electrodes was prepared by mixing either commercial polystyrene, polyamide-12 or polyurethane powder with synthetic bulk graphite powder. The graphite content was varied between 5 and 40 wt-% to find the optimum additive/matrix ratio. Electrodes with different shapes and sizes were designed for different analytical experiments. Square and rectangle-shaped electrodes (Fig. 1 a) were used for the resistance measurements and for performing the Helium Ion Microscopy (HIM)[27] and the Scanning Electron Microscopy (SEM) imaging, whereas cylinder shaped electrodes (Fig. 1 a) were prepared for the X-ray tomography analysis. Physical properties of the electrodes were fine-tuned by optimizing printing parameters such as layer thickness, laser power and printing speed.

The amount of graphite in the powder mixture affects the required printing parameters. To obtain electrodes with sufficient mechanical strength and durability while retaining the porous structure requires adjusting the printing conditions for each graphite/matrix system. The structure of the printed electrodes and the distribution of graphite were studied using SEM, HIM and X-ray tomography. The SEM images of the break surfaces of the 3D printed graphite/polystyrene electrodes are shown in Fig. 1 b, c and d. The set

of SEM images of the graphite/polyamide-12 electrodes also display similar structural characteristics (Figures S1-S4). The images reveal the highly macroporous structure of the material, in which the graphite powder is distributed evenly throughout the objects. As can be seen, the polymer particles have been only partially fused together by the sintering process, thus forming the porous structure with accessible voids between the particles.

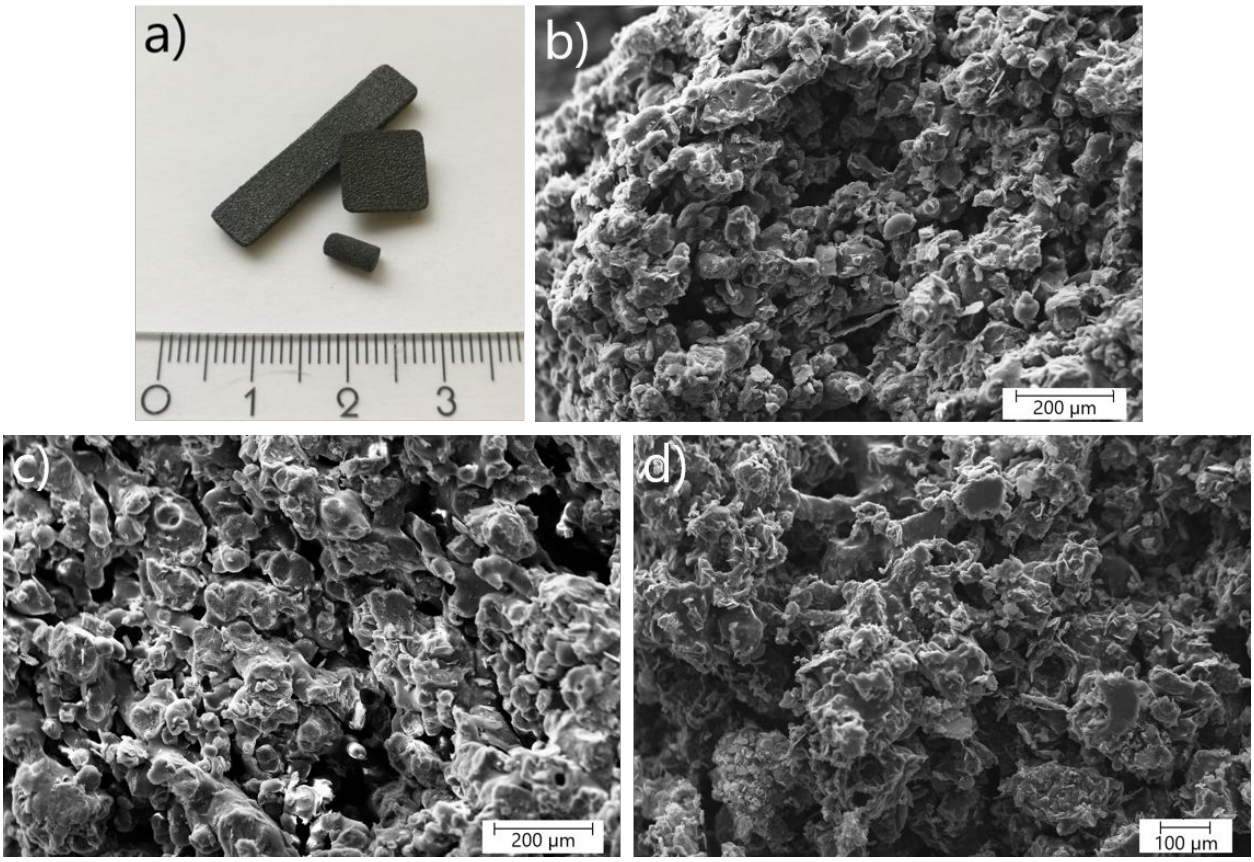

Figure 1. Rectangle, square and cylinder-shaped, polystyrene-based SLS 3D printed electrodes with 30 wt-% of graphite (a). SEM image of the break surface of 3D printed polystyrene electrode with 20 wt-% (b), 30 wt-%(c) and 40 wt-% (d) of graphite.

To avoid any problems caused by possible charging of the material during the SEM imaging, the 3D printed electrodes were also analyzed by Helium Ion Microscopy (Fig. 2). The HIM images confirmed the highly macroporous structure of the electrodes, but they also clearly show graphite's attachment on the surface of the polymeric three-dimensional network (Fig. 2 a). In the SLS 3D printing process, it is possible to obtain objects where the additive is not entirely capsulated by the matrix but it is firmly attached only on the matrix's partially melted surface of the polymer matrix. This means that the additive is not just loosely trapped within the three-dimensional structure, it is strongly anchored onto the polymeric matrix. On the other hand, the additive is achievable by fluids, gases or liquids, passing through the porous material.

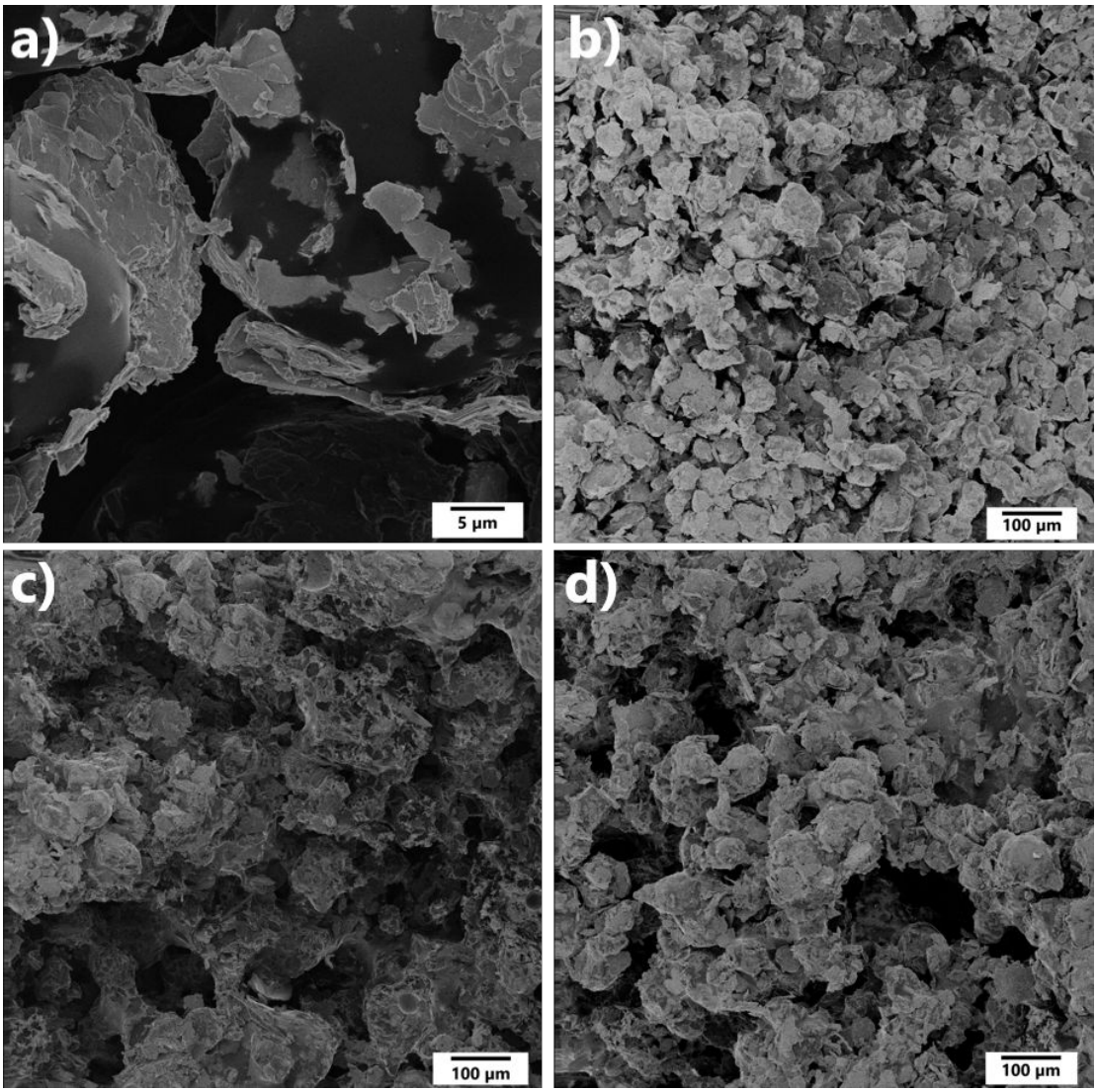

Figure 2. Helium Ion Microscope (HIM) image of the break surface of the 3D printed polystyrene electrode having 20 wt-% of graphite, showing that the graphite flakes are covering the surface of the partially melted polystyrene matrix (a). HIM image of the break surface of the 3D printed polystyrene electrode with 20 wt-% (b), 30 wt-%(c) and 40 wt-% (d) of graphite.

To confirm that the images of the outer layer and the break surfaces obtained via HIM and SEM imaging represent the overall internal structure of the electrodes, X-ray tomography of the electrodes was carried out. For these analyses, electrodes with 30 wt-% of graphite were used. The tomography images (Fig. 3) show that the whole electrode is porous and that the structure observed from the HIM and the SEM images describe the structure well. The structural analyses confirmed that porous electrodes can be obtained with both polystyrene and polyamide-12. The detailed analysis of the X-ray tomography results showed that the macroporous structure of the polystyrene electrode has 41 % of empty space inside the electrode whereas the polyamide-12 based electrode has 49 % of empty space. Analysis of the average pore diameter indicated that the polystyrene and polyamide-12 electrodes have average pore diameters between 10-55 μm and 10-75 μm, respectively (Fig. S5 and S6).

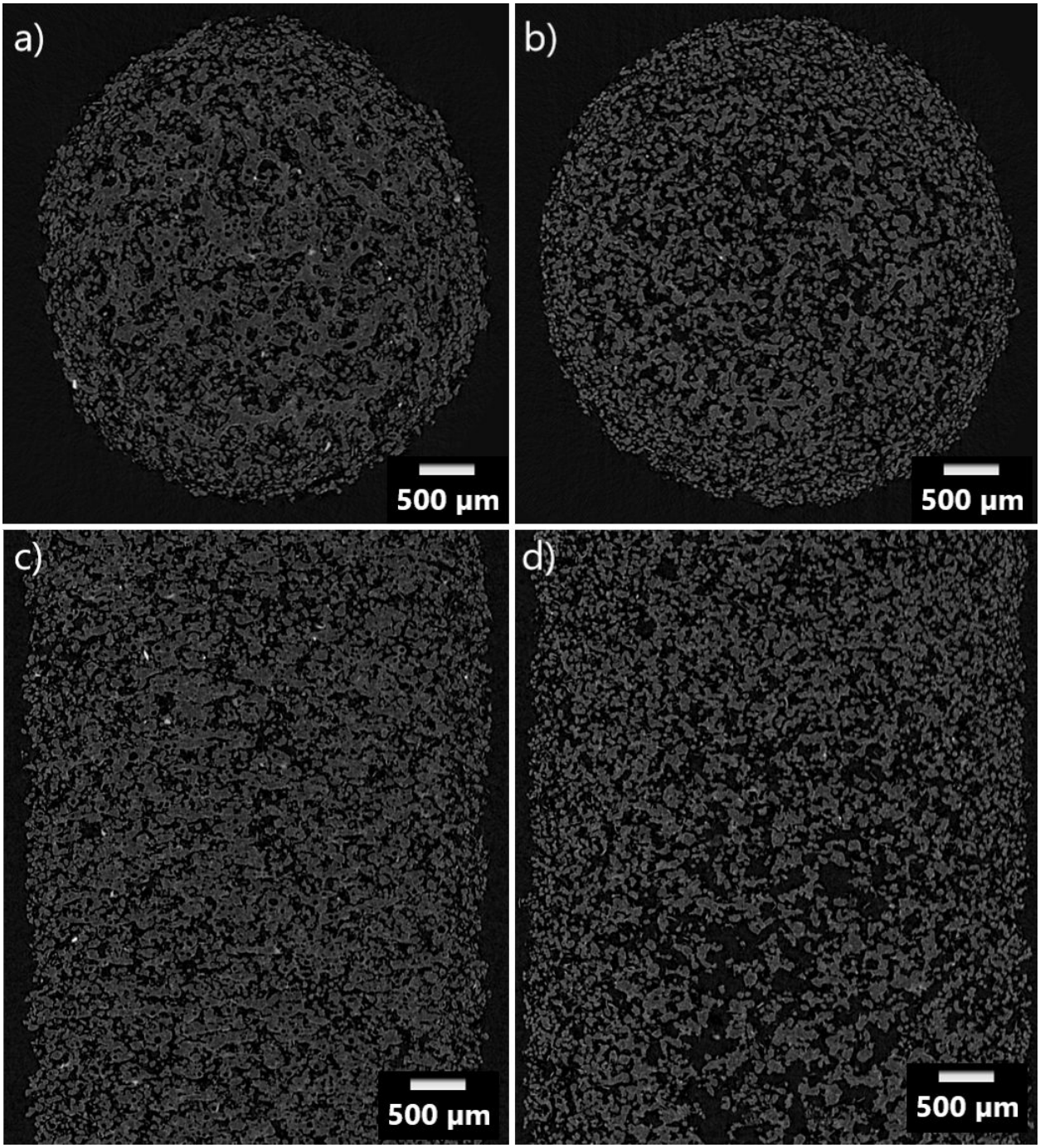

Figure 3. Horizontal slice of the structure of the cylinder shaped SLS printed electrode from X-ray tomography analysis of polystyrene (a) and polyamide-12 (b) based electrodes

having 30 wt-% of graphite. Vertical slices of the X-ray tomography images of the polystyrene (c) and polyamide-12 (d) based electrodes having 30 wt-% of graphite.

The printed electrodes can be used as electrodes for example for conventional electrolysis. However, in such a case the electrochemical processes take place on the outermost surface of the electrode (Fig. S7 and S8). The full advantage of the printed electrodes can be obtained when they are used as flow-through electrodes for example in flow batteries (Fig. S9). By printing, it is possible to fine-tune the flow properties by adjusting the printing conditions or by printing optimized flow channels with desired diameter and shape throughout the electrodes.

In addition to more rigid polystyrene and polyamide electrodes, polyurethane based graphite electrodes were 3D printed to obtain flexible and fully bendable, but still porous and conductive objects (Fig. S10). The flexible electrodes were printed using 30 wt-% of graphite mixed with polyurethane powder. The inherent porosity of the SLS printed material and the elastic nature of the polymer led to an object which conductivity could be

reversibly changed by applying pressure to it (Fig. S11). When the flexible material is compressed, the graphite particles, scattered on the surfaces of the polymer beads, are pressed together, which lowers the resistance and increases the conductivity. The change is considerable and can be easily measured. Such behavior opens up the possibility of using the material as a simple pressure sensor or a sensor for mechanical stress. Even though closely related piezoresistive materials are known and widely reported,[28–31] SLS 3D printing could provide an alternative way to prepare objects with pressure/conductivity correlation.

Conductivities of the rigid electrodes were studied in detail by using a high resistance meter. From the conductivities of the graphite/polystyrene electrodes (Fig. 4), it can be seen that the conductivity increases rapidly when graphite concentration is raised from 7.5 wt-% to 15 wt-% indicating that in these samples the conductivity jumps after the graphite concentration is high enough to produce firm graphite-graphite contacts in the material. It is likely that only certain chains of graphite-graphite contacts participate in the actual conduction paths, as stated in the percolation theory.[32] However, the printed electrodes contain enough of these chains to generate uniform behavior throughout the

object. The increasing trend in conductivity then continues as the graphite concentration is increased further beyond 15 wt-%. Naturally, the highest conductivity of 4.3 S m^{-1} is observed with the highest concentration of graphite. Similar trend can be seen in the conductivities of the graphite/PA12 electrodes as they also seem to possess a rather sharp threshold for the conductivity between 10 and 15 wt-%. Highest observed conductivity was 0.9 S m^{-1}, which is considerably less than that observed for the PS based electrodes.

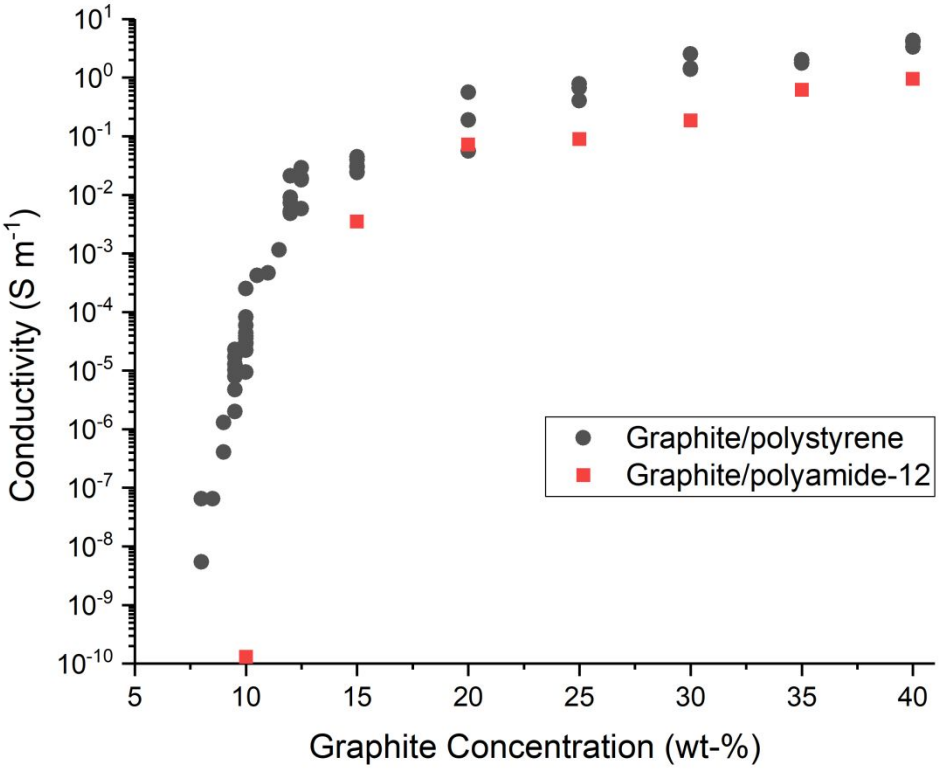

Figure 4. Conductivities of the 3D printed PS and PA12 electrodes as a function of the graphite concentration measured at 300 K.

To obtain a reference point and a limiting maximum conductivity for the results, tightly packed compressed pellets were made of the graphite powder using a hydraulic press. The conductivities of the pure graphite pellets ranged between 55.1 and 70.8 S m^{-1}. The conductivities observed for the 3D printed electrodes are an order of magnitude lower than those observed for the pure graphite pellets, but obviously, the controllable porosity, shape, sizes, and flow properties are not achievable by the compressed pellets.

The results highlight the usability of the SLS 3D printing technique in the preparation of highly customizable porous carbonous electrodes. It should be noted that the electrodes fabricated here are not optimized to obtain the maximum conductivity. Low-cost carbon additive with rather small inherent conductivity was utilized for the study. Meaning that the electric properties of the electrodes could be significantly increased by customization of the material itself. However, even these simple electrodes are already fully functional

and can be used in electrochemical processes. It is more than likely that the full advantage could be achieved in processes where the porosity and flow-through properties can be utilized. One of such potential application could be flow batteries.

Discussion

The SLS 3D printing provides a way to easily fabricate highly customizable electrodes even by using low-cost materials. The 3D printing technique gives the possibility to fine-tune the mechanical parameters such as the porosity and the flexibility of the electrodes by altering the supporting matrix material and the printing conditions. The technique is able to produce durable electrodes with high porosity and sufficient conductivity compared to the bulk graphite. Even though the study focused on graphite as the additive and common polymers as matrices, the method is not limited to these to materials and could be expanded to use different additives and polymers.

By using other additives such as conductive polymers or other carbon sources, or maybe even metals as well as choosing other types of supporting matrices, including conductive materials, the properties of the printed electrodes could be tuned and

enhanced further. The possibility to tailor and design highly porous electrodes with well-defined conductivity and flow-properties could open up a whole new way of fabricating highly efficient electrodes.

Experimental Section

Materials

The graphite used for preparation of the starting material was synthetic graphite powder (<20 μm) purchased from Sigma Aldrich. The polymer powders with average particle diameters of 50 μm were purchased from ADVANC3D Materials. All chemicals were used as received.

Three-dimensional printing

The 3D-models of electrodes were designed using FreeCAD v.0.16 software after which they were sliced into two dimensional, 0.1 mm thick, slices using Slic3r v. 1.2.9. Electrodes were printed with Sharebot SnowWhite SLS 3D printer. For preparation of the polystyrene based electrodes, laser speed was varied between 200 mm s^{-1} and 1200 mm

s^{-1}, depending on the amount of graphite in the mixture, with powder temperatures between 106 and 108 °C and CO_2 laser power of 14 W. For polyurethane, similar settings were used but temperature was set to 121 °C. Polyamide-12 based electrodes were fabricated using laser speeds varying between 360 mm s^{-1} and 2560 mm s^{-1} and temperatures between 160 and 167 °C with laser power varying between 4 W and 14 W. The 3D printing settings were adjusted for different graphite contents to obtain sufficient mechanical properties. The printed objects were carefully cleaned of all unsintered powder before using them in experiments.

Helium Ion Microscopy and Scanning Electron Microscopy

Helium Ion Microscopy was performed using Carl Zeiss ORION NanoFab. The beam energy used was around 30 kV with beam current ranging from 0.242 to 0.303 pA. Scan dwell time of 0.2 µs was used. Flood gun was used to counteract the charging effect. For HIM imaging samples did not receive any additional pretreatment after the 3D printing process. Scanning electron microscopy was done using Zeiss EVO-50XVP with

accelerating voltage of 25 kV. Prior to SEM imaging the 3D printed electrodes were coated with gold for optimal picture quality.

X-ray tomography

X-ray tomographic imaging of the electrodes was carried out using SkyScan 1172 microtomograph. Two cylinder shaped samples of 4-5 mm in diameter were imaged with 3.0 μm pixel size. The scanning parameters for both samples were identical. The X-ray source parameters were 29 kV and 100 μA and no filter was used. A total of 1200 projection images were taken using 0.3 degree step size over 360 degree of rotation. Projection images were averaged over 4 exposures of 4712 ms resulting in a total exposure time of 18.85 s for each of them. A total scan duration was 8 hours. Tomographic images were reconstructed using NRecon software, which is based on Feldkamp algorithm.[33] One pixel post-alignment was needed for ideal reconstruction. No ring artifact correction or beam hardening correction was used. A total sample height that was reconstructed was 5.6 mm for both samples. To measure the relative porosity of the samples, the volumes of the pore structures and the total volumes of the samples were

analyzed from the X-ray tomography images similarly, using ImageJ software. Additional information about the analysis is presented in supplementary information.

Conductivity measurements

Keithley 6517A Electrometer/High resistance meter was used to measure the resistance at room temperature. An attempt to minimize contact resistance was done by connecting copper plates to the sample with carbon black paste. Xylene was used as a solvent in carbon black paste. Copper wires were soldered to the plates and the wires were ran into the measuring apparatus. Copper plates were scratched with sand paper to get rid of oxide layer and to increase the contact area. High resistance samples were also soldered inside a shielded box so that coaxial wires came out of the box and were connected to the apparatus. Carbon black paste was applied as viscous as possible to prevent the paste from entering the pores of the material.

Supporting information. Detailed description of X-ray tomography analysis as well as Pore structure distribution graphs, Additional SEM images, Images of 3D printed

electrodes being used for electrolysis and in a flow cell, Images of flexible 3D printed electrodes.

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Author Contributions

M.H. conceptualized the idea of the SLS 3D printed electrodes. E.L. and E.K. did the designing and the 3D printing of the electrodes. J.J. performed the conductivity experiments for the electrodes. M.A. supervised the conductivity measurements. J.P. and J.V. performed the X-ray tomography and the analysis of the results. L.K. assisted in the design and fabrication of the flexible electrodes. E.L. wrote the initial manuscript which was jointly revised.

Competing Interests

The authors declare no competing interests.

Data Availability

The data supporting the findings of this study is available from the corresponding author upon reasonable request.

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