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A screen-printing method for manufacturing of current collectors for structural batteries

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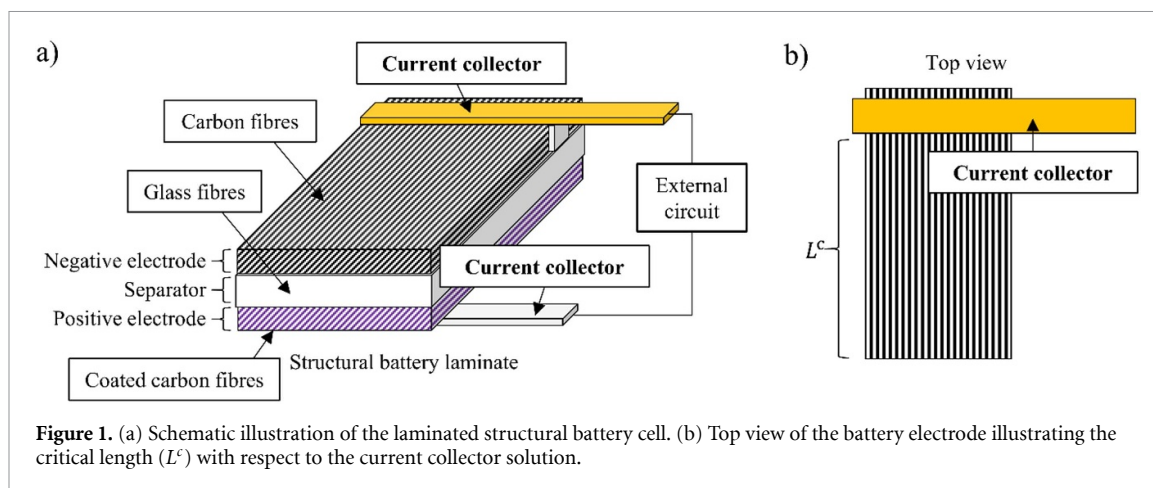
Abstract

Structural carbon fibre composite batteries are a type of multifunctional batteries that combine the energy storage capability of a battery with the load-carrying ability of a structural material. To extract the current from the structural battery cell, current collectors are needed. However, current collectors are expensive, hard to connect to the electrode material and add mass to the system. Further, attaching the current collector to the carbon fibre electrode must not affect the electrochemical properties negatively or requires time-consuming, manual steps. This paper presents a proof-of-concept method for screen-printing of current collectors for structural carbon fibre composite batteries using silver conductive paste. Current collectors are screen-printed directly on spread carbon fibre tows and a polycarbonate carrier film. Experimental results show that the electrochemical performance of carbon fibre vs lithium metal half-cells with the screen-printed collectors is similar to reference half-cells using metal foil and silver adhered metal-foil collectors. The screen-printed current collectors fulfil the requirements for electrical conductivity, adhesion to the fibres and flexible handling of the fibre electrode. The screen-printing process is highly automatable and allows for cost-efficient upscaling to large scale manufacturing of arbitrary and complex current collector shapes. Hence, the screen-printing process shows a promising route to realization of high performing current collectors in structural batteries and potentially in other types of energy storage solutions.

1. Introduction

Weight reduction of vehicles and aircraft is one crucial factor for reducing carbon-dioxide emissions. With the ongoing rise of electromobility, weight saving of vehicles is of particular interest. Substantial weight saving can be achieved by combining the energy storage of a battery with the load-carrying ability of a structural material in what is called structural batteries [1, 2].

Structural batteries can be realised by combining the technology of lithium-ion batteries with that of carbon fibre composites [3–12]. Carbon fibres are used to carry load, to host lithium and transfer electrons in the battery cell. A schematic illustration of the structural battery is presented in figure 1(a). In this design the carbon fibres in the negative electrode function as active electrochemical electrode materials. Further, carbon fibres coated with a lithium-based metal oxide [13] are used as positive electrode and a non-conductive glass fibre weave is used as separator. The electrolyte needs to enable ion conduction between the electrodes and



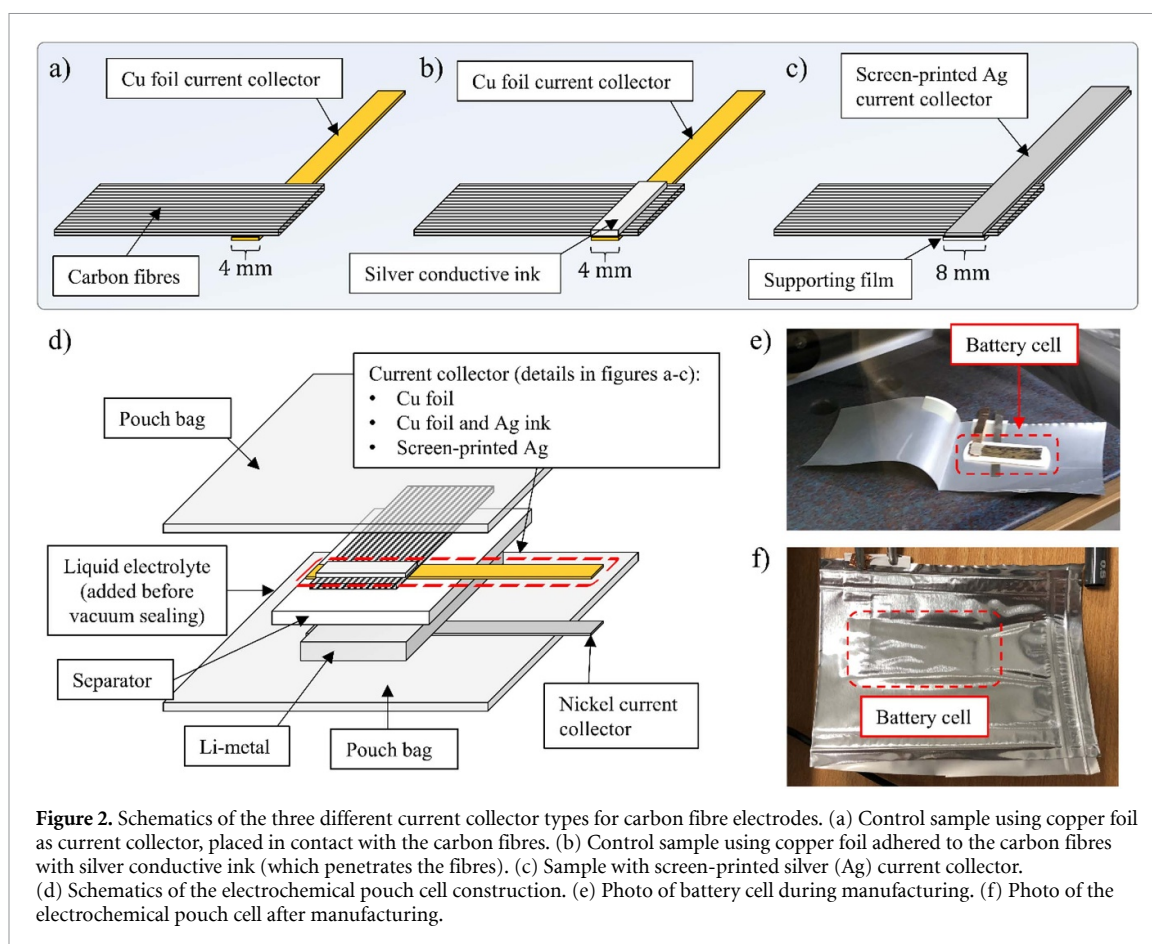
ensure mechanical load transfer between the constituents. These requirements are achieved with a structural battery electrolyte (SBE) which behaves like a solid polymer on the macro-scale and has a sub-micron interpenetrating network of liquid electrolyte for ion conductivity through the material [14–16]. It should be noted that the design of structural batteries is still in an early stage; nevertheless, it has been shown that structural batteries have the ability to outperform the monofunctional systems [1, 2, 12, 15].

A crucial part of structural batteries has so far been studied sparingly [17]; the current collectors (figure 1(b)). In previous work [12, 15], the current collectors have been made from metal strips adhered to the carbon fibres using silver conductive ink. Due to the fact that the electric conductivity of the carbon fibres [3, 18] is about three orders of magnitude lower than of copper and aluminium (commonly used for current collectors in conventional batteries), a substantial ohmic potential drop is expected for larger cells using longer carbon fibre tows. For example, an ohmic potential drop of approximately 0.14 V is estimated for a 0.2 m long carbon fibre electrode (L^c in figure 1(b)), discharged at a C -rate of 1 (i.e. a discharge time of 1 h)⁶. Consequently, larger structural batteries require a refined current collector design (in comparison with previous solutions) with additional extraction points for the electric current. The current collector must also allow for electron transfer between all fibres in the electrode, enable series and parallel connections of different electrodes and serve as a connection of the entire battery to a consumer. Moreover, for high-volume production of structural batteries, automated manufacturing methods of current collectors are required.

One technology which may suit the requirements for high volume production of current collectors is screen-printing; a fast and versatile printing process that allows for high volume manufacturing. Within the screen-printing process, screen-printing paste is deposited onto a substrate by being pressed through a partially opened screen with defined geometry. Today, screen-printing is a well-established process for manufacturing different kinds of (electro-)chemical [19, 20] and biosensors [21, 22], printed electronics [23–29] and electrodes [30]. Screen-printing allows for miniaturisation of electronic parts at reasonable cost and enables flexibility of the parts when flexible substrates are used [31]. Screen-printing is particularly cost efficient for high production volumes of identical print geometries on flat substrates. On the other hand, the cost and complexity of screen-printing increases considerably for variable geometries and curved substrates. Another current disadvantage of screen-printing is the high requirements for the surface quality of the substrates. There is little research on high-performance electronics screen-printing on rough surfaces.

In this paper, we demonstrate a proof of concept screen-printing process that enables printing of current collectors on spread carbon fibre tows for structural batteries, providing support of the carbon fibres and allowing flexible handling and electrical connection. The current collectors are directly screen-printed on the rough surface of spread carbon fibre tows using a commercial silver screen-printing paste. The electrochemical performance of carbon fibre vs lithium metal half-cells with screen-printed current collectors are evaluated during short term cycling and compared to half-cells using common copper foil current collectors. Half-cells are studied and silver additives are used as a model system focussing on the screen-printing technique.

⁶ Details on how this drop in voltage is approximated are presented in supplementary information (available online at stacks.iop.org/MFM/4/035002/mmedia).



2. Methodology

2.1. Current collector solutions

To evaluate the performance of screen-printed current collectors for structural batteries, three types of current collector solutions for carbon fibre electrodes are studied. The three solutions are schematically illustrated in figures 2(a)–(c). Schematics of control samples with current collectors made from copper foil are shown in figures 2(a) and (b). In the 1st case the copper foil (4 mm wide) is simply placed on top of the spread carbon fibre tow. The 2nd solution shows the current collectors used in previous works [12, 15] consisting of copper foil (4 mm wide) adhered to the carbon fibres using silver conductive ink [32]. Figure 2(c) shows the screen-printed silver current collectors (8 mm wide).

2.2. Electrochemical pouch design

To evaluate the electrochemical performance of the different current collector solutions (figures 2(a)–(c)), the carbon fibre electrodes are assembled into structural battery half-cells as illustrated in figures 2(d)–(f).

In the electrochemical half-cell, the carbon fibre electrode functions as the working electrode while the lithium metal functions as the combined counter and reference electrode. The carbon fibre electrodes are made from ultra-thin spread tow unidirectional tapes of T800SC-12k-50C [33] PAN-based carbon fibres with a linear weight of the fibre tow of 0.52 g m^{-1} . The separator is made from a glass microfibre separator (Whatman GF/A, $260 \mu\text{m}$ thick, 90% porosity) and assures that the carbon fibres do not come in direct contact with the lithium metal (i.e. avoid short circuit). To extract the current from the lithium metal, a current collector made from nickel foil is used. Furthermore, to allow ion transport, the electrodes and separator are impregnated with 1.0 M LiPF_6 in EC:DMC 1:1 w/w electrolyte (all chemicals purchased from Sigma–Aldrich). All materials are used as received. After arranging the constituents as shown in figure 2(d), the electrochemical cell is placed inside a vacuum heat-sealed laminate bag (PET/Al/PE; $12 \mu\text{m}/9 \mu\text{m}/75 \mu\text{m}$ thick) to protect the electrochemical cell from air and moisture and to ensure good contact between the constituents inside the cell. The assembly is performed at ambient temperature inside a glovebox with an inert atmosphere with less than 2 ppm O_2 and H_2O .

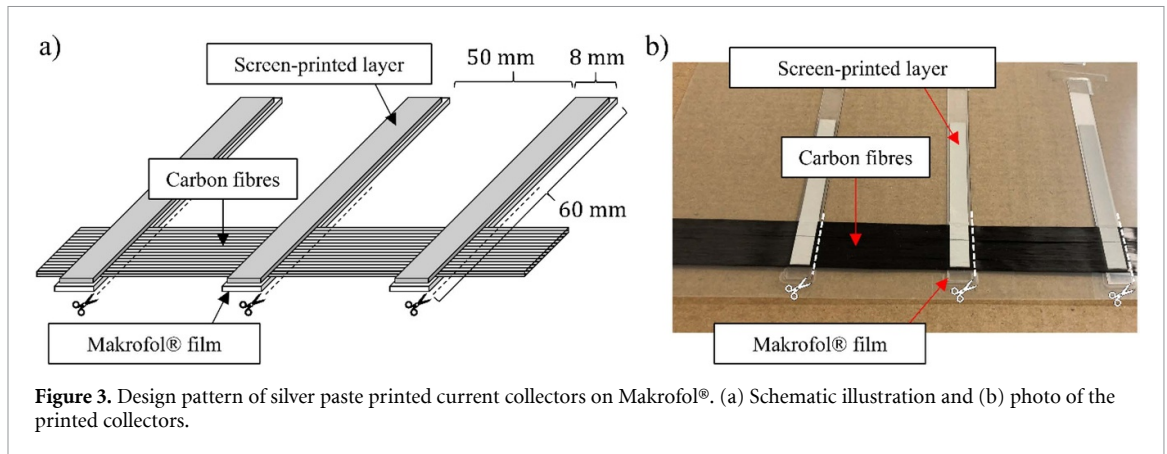


Figure 3. Design pattern of silver paste printed current collectors on Makrofol®. (a) Schematic illustration and (b) photo of the printed collectors.

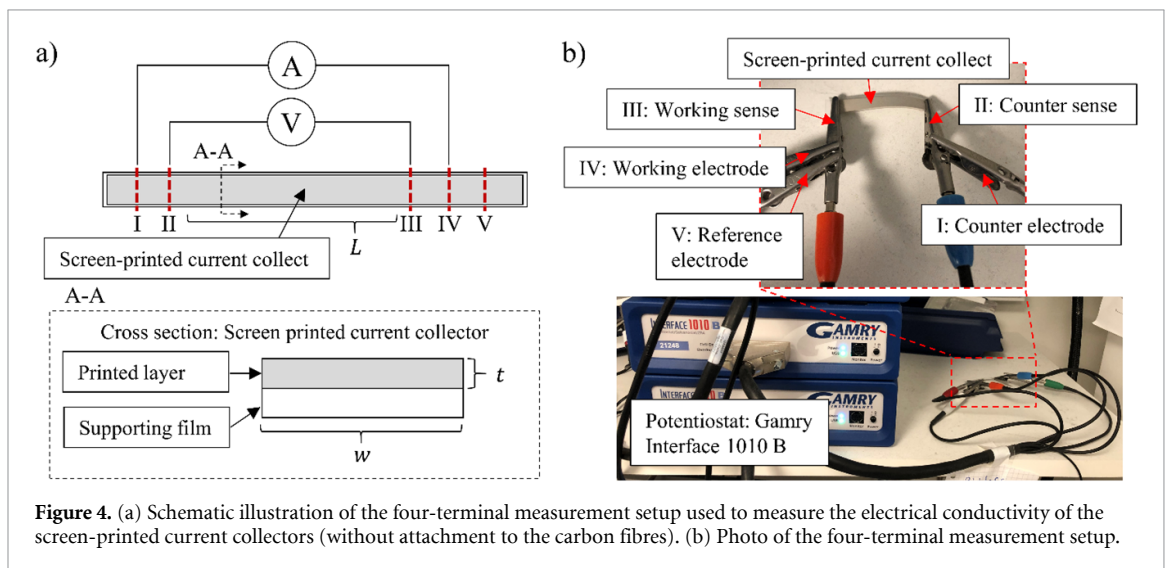


Figure 4. (a) Schematic illustration of the four-terminal measurement setup used to measure the electrical conductivity of the screen-printed current collectors (without attachment to the carbon fibres). (b) Photo of the four-terminal measurement setup.

2.3. Screen-printing

The screen-printed silver paste needs a carrier material to extend from the carbon fibres. For this purpose, a thin (125 μm) polycarbonate plastic film (Makrofol®, Covestro AG [34]) is used. Prior to screen-printing, the carbon fibres are held in place on the plastic film using tesafix® 4965. Figures 3(a) and (b) shows the screen-printing design, where two electrodes are manufactured in one print (the dashed lines in figure 3(a) illustrate how the electrodes are separated prior to assembly).

Screen-printing is performed on a flatbed parallelogram screen-printing system (Siri 5000pro, Bochonow GmbH). The screen is consisting of a polyester fabric (120/35 22.5 °) stretched on an aluminium frame. The screen-printed silver paste (Solamet PV410, Du Pont [35]) is chosen since it is a highly electrically conductive paste optimised for screen-printing. After printing the specimens are sintered at 110 °C for 10 min. Surface roughness measurements of the printed current collectors and the carbon fibre substrate are performed on a Keyence VHX 6000 digital microscope.

2.4. Conductivity measurement

A four-terminal measurement setup is used to measure the electrical conductivity of the printed current collectors. A schematic illustration of the setup is presented in figure 4(a) and photos of the setup are shown in figure 4(b). It should be noted that the measurement is performed on the printed current collectors only (i.e. without the carbon fibres).

The voltage (V) is measured over the gauge length (L) using a Gamry Interface 1010B potentiostat while applying a forced constant current (I). It is assumed that the isolating supporting film does not contribute to the electrical conductivity and that the resistance of the specimen follows Ohm's law. Hence, the effective conductivity (s) of the printed current collector layer is estimated as:

$$s = \frac{LI}{Vtw} \quad (1)$$

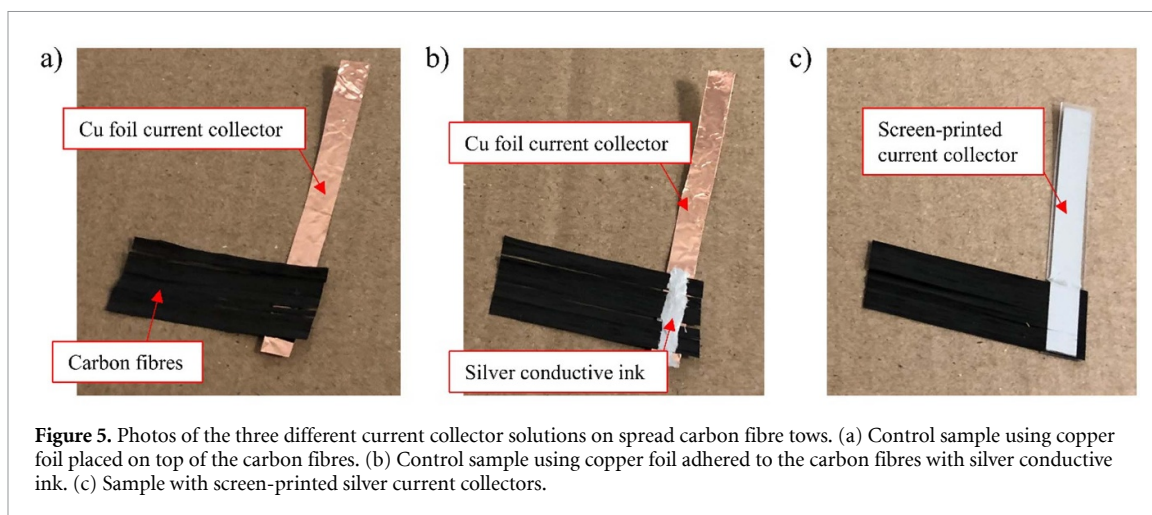


Figure 5. Photos of the three different current collector solutions on spread carbon fibre tows. (a) Control sample using copper foil placed on top of the carbon fibres. (b) Control sample using copper foil adhered to the carbon fibres with silver conductive ink. (c) Sample with screen-printed silver current collectors.

where L is the length, t is the thickness and w is the width of the printed layer, respectively (cf figure 4(a)). The length and width of the specimen are measured using a ruler while the thickness is measured using an optical microscope. The conductivity measurements are performed at room temperature (i.e. approximately 20 °C).

2.5. Electrochemical cycling

To investigate the electrochemical properties of screen-printed current collectors, electrochemical cycling is performed on the assembled battery half-cells utilizing the three current collector solutions presented in figure 2(a). The electrochemical capacity is measured with galvanostatic charge and discharge cycles using a Gamry 1010B potentiostat. The cells are cycled for ten cycles between 0.01 V to 1.5 V vs Li/Li⁺ using a current corresponding to 37.2 mA g⁻¹ of carbon fibres in the electrode (corresponding to an approximate C-rate of 1/10 with respect to the theoretical capacity of graphite as well as the actual capacity of the carbon fibre), and a 15 min rest between the charge and discharge cycles. The electrochemical cycling is performed at room temperature (i.e. same conditions as the conductivity measurements).

3. Results and discussion

3.1. Screen-printing

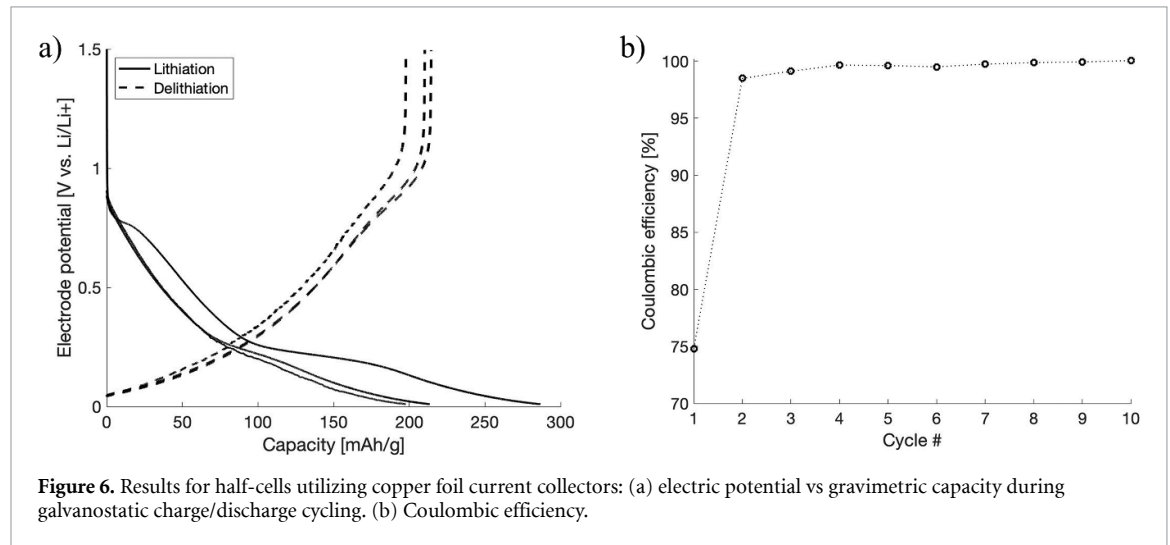
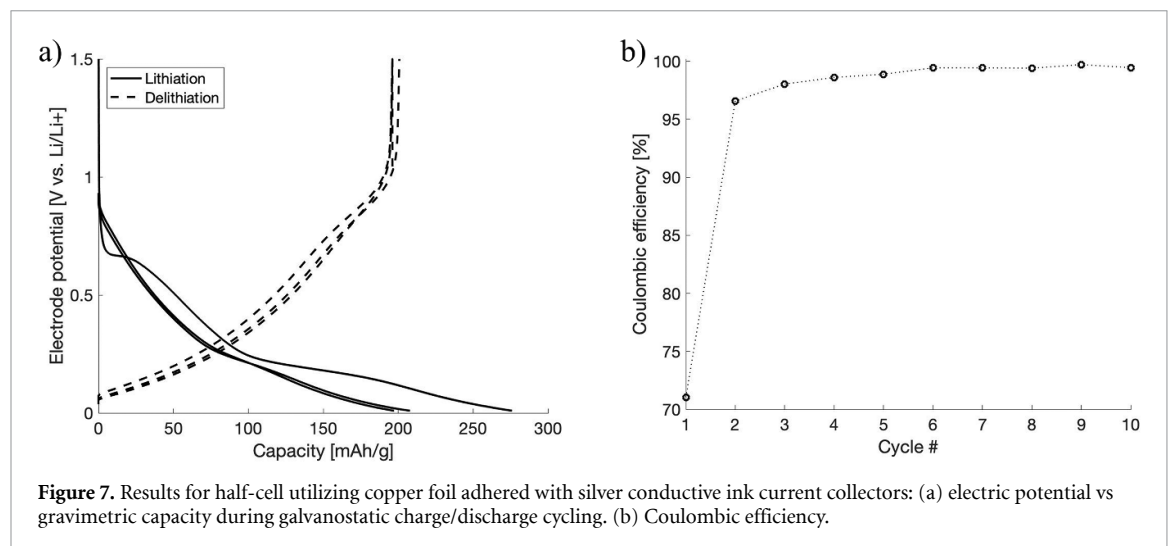
Figure 5 shows examples of the three types of current collector solutions. The screen-printed current collectors have a homogeneous print quality and show a sufficient adhesion to the polycarbonate foil. The edges of the current collectors are sharp and show a high dimensional accuracy compared with the screen-printing stencil. The reproducibility of the process is high when comparing different current collectors. The achieved printing quality would allow for smaller and more complex shapes if required, but the combination of small complex shapes with appropriate conductivity will need to be subject for future research.

The surface roughness of the carbon fibre substrate was averaged over at least five measurements in various locations. Locally, the arithmetic mean surface roughness determined by line measurement is $R_a = 7 \mu\text{m}$ which is close to the filament diameter of 5 μm (according to the data sheet [33]). However, when measuring a larger distance, the overall arithmetic mean surface roughness of the substrate is $R_a = 23.3 \mu\text{m}$ and the average peak-to-valley height $R_z = 136.5 \mu\text{m}$. The surface roughness is higher due to gaps in the spread carbon fibres. Despite the high surface roughness and the strong fluctuations, the printed current collectors are homogeneous and continuous. The current collectors compensate for much of the surface roughness of the fibres and have a surface roughness of $R_a = 4.6 \mu\text{m}$ and an average peak-to-valley height of $R_z = 26 \mu\text{m}$.

The screen-printed current collectors enable flexible handling of the fibre electrode without breaking. In contrast to initial experiments performed by the authors, in which another silver paste was tested (CRSN2442, Sun Chemical [36]) which was more brittle and fragile and had larger silver particles. Due to the brittleness, the current collectors printed with the alternative silver paste were damaged during sample handling and insertion into the pouch cells. Therefore, no further electrochemical tests were carried out with the alternative material. The samples with silver conductive ink are also fragile but can be integrated in the pouch cells when handled with enough care in order not to break off the current collector from the fibres. The utilized screen-printed collectors can easily be handled and incorporated in pouch cells without losing the electrical connection between the collector and the fibres. Consequently, fragility and brittleness are

Table 1. Measured electric conductivity (σ) and thickness of the printed layer (t), of the screen-printed current collector.

Constituent	Electric conductivity (σ)	Thickness of the printed layer (t)
Screen-printed collector	$6 \times 10^5 \text{ Sm}^{-1}$	0.01 mm

**Figure 6.** Results for half-cells utilizing copper foil current collectors: (a) electric potential vs gravimetric capacity during galvanostatic charge/discharge cycling. (b) Coulombic efficiency.**Figure 7.** Results for half-cell utilizing copper foil adhered with silver conductive ink current collectors: (a) electric potential vs gravimetric capacity during galvanostatic charge/discharge cycling. (b) Coulombic efficiency.

additionally to conductivity and printability (viscosity, thixotropy) important factors when choosing a suitable material for current collector printing.

3.2. Conductivity measurements

The measured electric conductivity of the screen-printed current collector is presented in table 1. The electric conductivity is found to be approximately $6 \times 10^5 \text{ Sm}^{-1}$. This corresponds to roughly one order of magnitude higher than the electric conductivity of carbon fibres [3] and two orders of magnitude lower than copper and aluminium. Hence, the conductivity is sufficiently high to allow for more complex shapes with longer electric current paths. A thickness of the printed layer t (cf figure 4(a)) of 0.01 mm is measured using an optical microscope (table 1). This thickness was used to calculate the conductivity of the printed current collector (equation (1)).

3.3. Electrochemical cycling

Representative charge–discharge curves and Coulombic efficiency curves of cells with the different collector types are shown in figures 6–8. The gravimetric capacity is calculated based on the total length of the carbon fibre tow used in the sample. The charge and discharge curves show stable cycling, however the overpotentials are larger for the silver conductive ink samples and screen-printing samples.

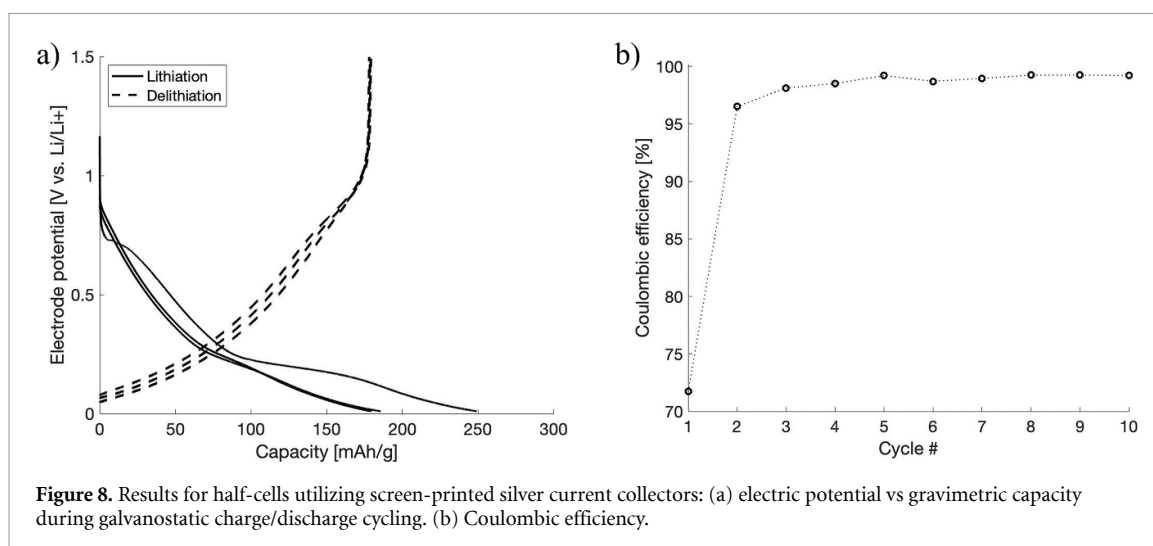


Figure 8. Results for half-cells utilizing screen-printed silver current collectors: (a) electric potential vs gravimetric capacity during galvanostatic charge/discharge cycling. (b) Coulombic efficiency.

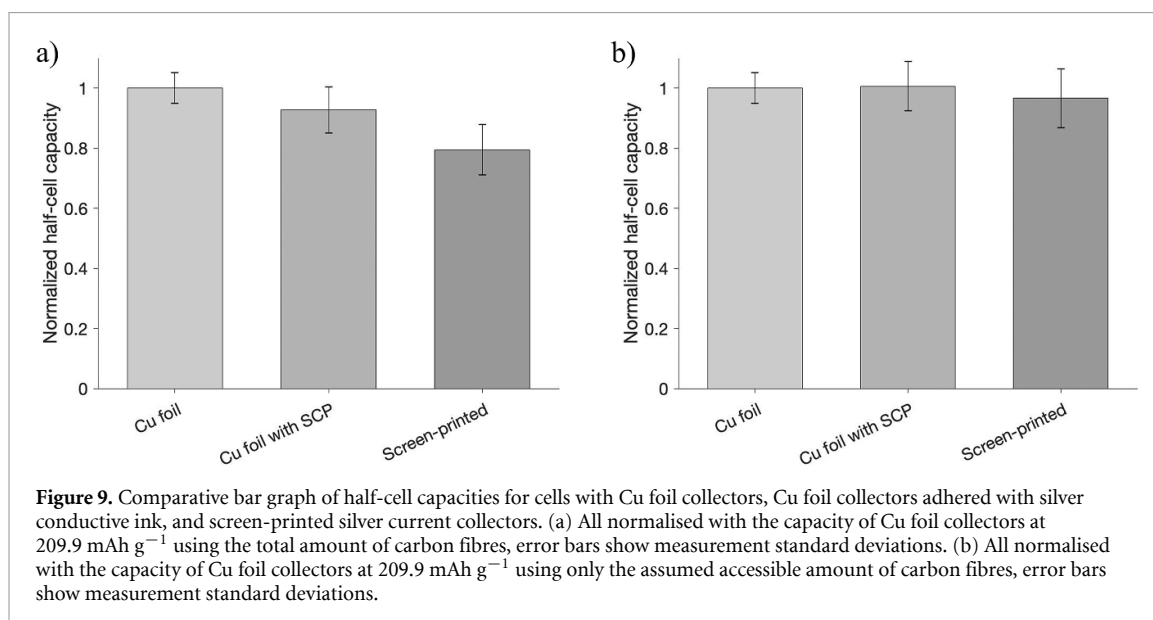


Figure 9. Comparative bar graph of half-cell capacities for cells with Cu foil collectors, Cu foil collectors adhered with silver conductive ink, and screen-printed silver current collectors. (a) All normalised with the capacity of Cu foil collectors at 209.9 mAh g^{-1} using the total amount of carbon fibres, error bars show measurement standard deviations. (b) All normalised with the capacity of Cu foil collectors at 209.9 mAh g^{-1} using only the assumed accessible amount of carbon fibres, error bars show measurement standard deviations.

The Coulombic efficiencies for all collector types reach a plateau after the 2nd cycle. For comparison, an average of all cycles excluding the 1st is calculated. Samples with only copper foil reach an average Coulombic efficiency of 99.7% ($\pm 0.16\%$, four samples), with silver conductive ink 98.7% ($\pm 0.67\%$, five samples) and screen-printed 99.0% ($\pm 0.46\%$, six samples) after ten complete charge/discharge cycles. The Coulombic efficiency of the screen-printed samples are slightly higher than the samples using silver conductive ink and copper foil, but lower compared with the samples using only copper foil. It is known that silver conductive ink is not very good for high capacity and stable cycling of lithium half-cells [37] due to side reactions, and these results indicate that side reactions might occur in the cells utilizing silver paste/ink. In summary, the results show that screen-printing is a viable method for making current collectors for carbon fibre electrodes, but there is scope for more research on screen-printing conductive particles and binders that are more chemically stable.

Figure 9(a) shows a bar plot comparing the average capacities of the different current collectors, where the gravimetric capacity is calculated by the total length (i.e. total mass) of carbon fibres available in the sample. All averages are normalised by the capacity of the control sample with copper foil ($210 \pm 11 \text{ mAh g}^{-1}$). A decreasing trend of capacity is visible to the control sample with silver conductive ink ($195 \pm 13 \text{ mAh g}^{-1}$), and to the screen-printed silver current collectors ($167 \pm 15 \text{ mAh g}^{-1}$).

Calculating the gravimetric capacity based on the total length of carbon fibres can be misleading, since the section of the fibres covered by silver conductive ink or screen-printed paste might be inaccessible to lithium ions (i.e. not able to contribute to the capacity). A more direct comparison of the electrochemical performance is to base the gravimetric capacity on the assumed accessible length of carbon fibres in the



Figure 10. Complex shapes made with screen-printed silver paste.

sample (i.e. the visible part of carbon fibre tows in figures 5(a)–(c)). Figure 9(b) shows a bar plot comparing the average capacities of the different current collectors, where the gravimetric capacity is calculated by the accessible length of carbon fibres. The average capacities are normalised by the capacity of the control sample with copper foil ($210 \pm 11 \text{ mAh g}^{-1}$). The capacity of the control sample with silver conductive ink is roughly the same ($211 \pm 13 \text{ mAh g}^{-1}$), and the screen-printed silver current collectors have a slight decrease in capacity compared to the other ($203 \pm 18 \text{ mAh g}^{-1}$). The silver conductive ink seems to display higher capacity than expected, which may be linked to a partial coverage of silver ink as seen in figure 5(b). The results highlight that the electrochemical performance is promising using screen-printed current collectors. Further, the wide confidence interval shows that there is scope for more research on screen-printing and binders that are more chemically stable.

The fact that a section of the carbon fibres may become inaccessible by the screen-printing paste is still a concern for high gravimetric capacity structural batteries. For example, the thickness, width, and pattern of the printed section should be controlled to assure high gravimetric performance. No optimisation has been made of this relationship. It should also be noted that the density of the utilized screen-printing paste is approximately 60%/70% of that of pure silver/copper, respectively. Normalizing the average capacity of the screen-printed silver current collectors, using only the assumed accessible amount of carbon fibres while accounting for the difference in density, gives an effective improvement in capacity of approximately 50% compared with the reference samples. Hence, significant mass savings can be envisioned by utilizing smart design of the screen-printed current collectors.

3.4. Opportunities with screen-printing

The simple layout shown for current collectors in this research is a 1st step towards showing the potential of screen-printed current collectors in structural batteries. Screen-printing is a very versatile manufacturing method that can achieve complex shapes and electrical circuits, see figure 10.

Thus, more complex screen-printed current collectors can be envisioned, e.g. by printing current collectors in an architecture according to figure 11(a); where multiple strips of current collectors are printed in a transverse direction relative to unidirectional carbon fibres with a specified distance to overcome the ohmic potential drop over the length of the carbon fibres. This can be made in different fibre directions and shapes. All carbon fibres in one layer are then electrically connected, but also held together by the screen-printed frame, which could significantly improve handling and layout of the fibres in the manufacturing. Also, containing the fibres may help prevent short circuits from stray carbon fibres. Making both positive electrodes and negative electrodes in the same way may be possible, which would facilitate a layout as shown in figure 11(b). This layout and layer design would make it possible to connect positive and negative layers through thickness, making external connections simpler.

To evaluate the performance of different layout options several design parameters need to be considered. The most critical design parameters for the electrical performance are: (a) the distance between the printed strips along the fibres (corresponds to L^c in figure 1); (b) the electric conductivity of the printed substrate; (c) the width (w) and thickness (t) of the printed strips (cf equation (1)). Hence, it is possible to improve the design in terms of its electrical performance by altering these parameters to minimize the total electrical loss within the system (in accordance with the procedure for estimating the voltage drop along the fibres described in supplementary information). It should be noted that due to the multifunctional nature of the material system, modified design parameters will alter its multifunctional performance. For example, the electric conductivity of printed substrates is often improved by increasing the volume fraction of the conductive medium (in this case silver particles). This is often associated with a more brittle material, subsequently resulting in an inferior mechanical performance. Also, increased width (w) of the printed strips in combination with reduced distance between the strips along the fibres reduces the specific capacity of the

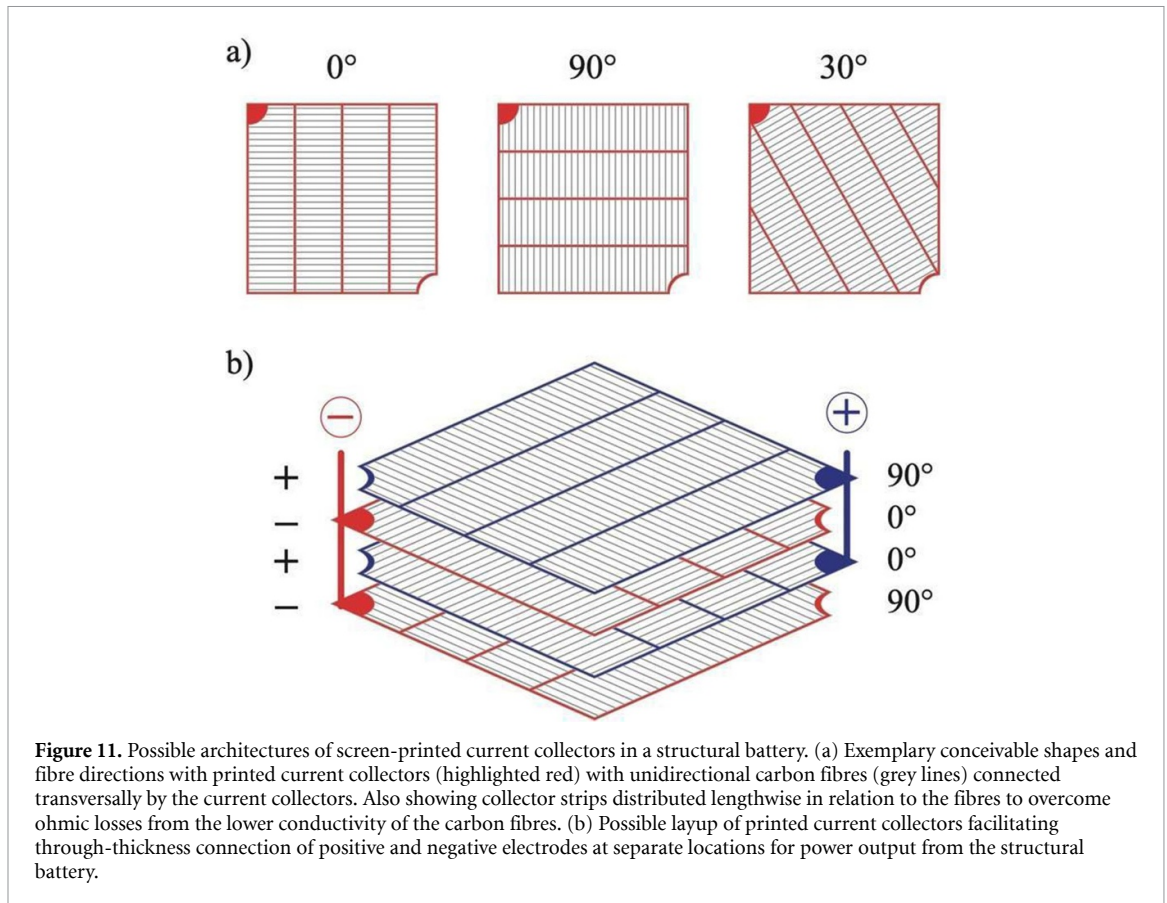


Figure 11. Possible architectures of screen-printed current collectors in a structural battery. (a) Exemplary conceivable shapes and fibre directions with printed current collectors (highlighted red) with unidirectional carbon fibres (grey lines) connected transversally by the current collectors. Also showing collector strips distributed lengthwise in relation to the fibres to overcome ohmic losses from the lower conductivity of the carbon fibres. (b) Possible layout of printed current collectors facilitating through-thickness connection of positive and negative electrodes at separate locations for power output from the structural battery.

device, as discussed previously. Hence, the combined electrochemical and mechanical (i.e. multifunctional) performance needs to be considered when optimizing the collector design.

Finally, in this study silver paste was used as model material. For a complete battery cell, different conductive additives are needed for the different electrodes (linked to the stability of the material at different potentials). This can be achieved by altering the screen-printing paste (e.g. copper screen-printing pastes are available [38, 39]).

4. Conclusion

In this paper, a proof-of-concept screen-printing method to manufacture current collectors for structural batteries is presented. The silver paste collectors are screen-printed directly onto spread carbon fibre tows and a carrier polycarbonate film in areas without fibres. Promising results are found when comparing the electrical and electrochemical performance of the screen-printed current collectors with reference current collectors made from copper foil and copper foil adhered to the fibres using silver conductive ink. The experiments demonstrate that the screen-printed collectors adhere to the carbon fibres and make the current collector connection less fragile, improving the manufacturability and should allow for large scale manufacturing. The mechanical adhesion between fibres and screen-printing paste is not quantified, and more research is needed on the mechanical properties and requirements.

The measured conductivity of the printed collectors is roughly one order of magnitude higher than the electric conductivity of carbon fibres [2] and two orders of magnitude lower than copper and aluminium. Hence, in combination with the possibility for printing complex shapes, the conductivity is considered high enough to allow for realizing large scale structural batteries or battery modules.

The electrochemical capacity and Coulombic efficiency of carbon fibre electrodes, utilizing the manufactured screen-printed collectors, are shown to be similar to those utilizing metal-foil collectors (commonly used in literature [12, 15]). The printed collectors are also found to be electrochemically stable in carbon fibre vs lithium metal half-cells, which is linked to the electrical potential window. In the case of full-cell application, galvanic corrosion of the silver is identified as an issue. Hence, in future work, other metal particles in the printing paste need to be studied as well as screen-printing paste binders need to be further investigated for side reactions.

Finally, we demonstrate how screen-printing can be used for complex patterns and allow for optimization (mass, ohmic loss, etc) of current collectors for structural battery electrodes.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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Conflict of interest

The authors declare no competing interests.

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