

# CARBON FIBRE COMPOSITES AS BATTERIES, SENSORS, ACTUATORS AND FOR ENERGY HARVESTING

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## ABSTRACT

Reduced mass for improvements in system performance has become a priority for a wide range of applications that requires electrical energy and includes load-bearing components. Use of lightweight materials has been identified as key for successful electrification of future transport solutions. Structure, energy storage and energy distribution are usually subsystems with the highest mass contributions but energy storage and energy distribution devices are structurally parasitic. One creative path forward is to develop composite materials that perform several functions at the same time – multifunctional materials. Combining functions in a single material entity will enable substantial weight savings on the systems level.

One such concept is a structural battery, a material that simultaneously carry load and stores energy like a battery. Structural batteries employ carbon fibres as structural reinforcement and negative electrode and can also be used as current collectors to save additional weight.

A number of new physical phenomena when using carbon fibres as battery electrodes have been found which allows for further multi-functionality. These are all based on the fact that carbon fibres intercalated lithium ions as an electrode material. The ion intercalation creates a reversible longitudinal expansion of the carbon fibres which could be used for actuation and morphing. A piezo electrochemical effect couples the electrical potential of the fibre to the strain acting on it, which can be used for sensing purposes. By combining the expansion and the piezo electrochemical effect one can convert mechanical energy to electrochemical energy, providing an energy harvesting function. The long-term vision of this work is to create a composite material that carries load, stores electrical energy, senses its own state, morphs and harvests energy.

## 1 INTRODUCTION

The use of lightweight composite materials is key for successful electrification of future vehicles. Structure and energy storage are subsystems with the highest mass contributions but energy storage devices are structurally parasitic. One possible remedy to this is to develop composite materials that perform several functions at the same. Combining functions in a single material entity will enable substantial weight savings on the systems level. To meet this challenge research aiming at developing so-called structural batteries, which are hybrid and multifunctional material assemblies, is being pursued [1]. In such a multifunctional composite material carbon fibres are used as the primary load-carrying material due to their excellent strength and stiffness properties and as active electrodes providing a battery function.

Several exciting phenomena have been discovered during the work on structural carbon fibre batteries which suggest that electrochemical intercalation of lithium can activate new intrinsic functionalities in carbon fibres: Carbon fibres expand in their length direction when lithium is intercalated into their micro-structure. This effect can be used to create actuation and morphing, i.e.,

transforming electrical work into mechanical work as an actuator, enabling shape-changing structures. Carbon fibres become piezo-electrochemical resulting in a change of the electrical potential of a lithium intercalated fibre versus its counter electrode when the fibre is subjected to mechanical loads, allowing direct sensing of forces within the material itself. This piezo-electrochemical effect can even be used to convert mechanical work into electrical energy, i.e., as an energy harvester.

It thus seems possible to add a number of very useful functions to a carbon fibre composite material that can both add functionality to the system and save weight, all intrinsically embedded in the material itself. The long-term vision of this work is to create a composite material that carries load, stores electrical energy, senses its own state, morphs and harvests energy.

## 2 CARBON FIBRE COMPOSITE STRUCTURAL BATTERIES

Multifunctional materials present an innovative concept to increase system efficiency as two or more functions are combined in one material [2,3]. A structural battery (SB) is one example of a multifunctional lightweight material that enables energy storage and structural integrity simultaneously [1]. A structural battery is a concept that resembles a standard fibre composite laminate. Carbon fibres are used as a high performance structural backbone and laid up arbitrary in angles in the sequence: negative electrode/separator/positive electrode. On the negative side the carbon fibres intercalate/insert ion (herein Li-ions) in just the same way as a state-of-the-art Li-ion battery electrode materials [4]. The separator could be a thin glass fibre weave or veil. On the positive side carbon fibres are coated with an active battery material, e.g.  $\text{LiFePO}_4$ . [5]. The laminate stack then needs to be embedded in a matrix/electrolyte material, the structural battery electrolyte, in order to achieve mechanical load transfer and ionic conductivity. The concept is illustrated schematically in Fig. 1.

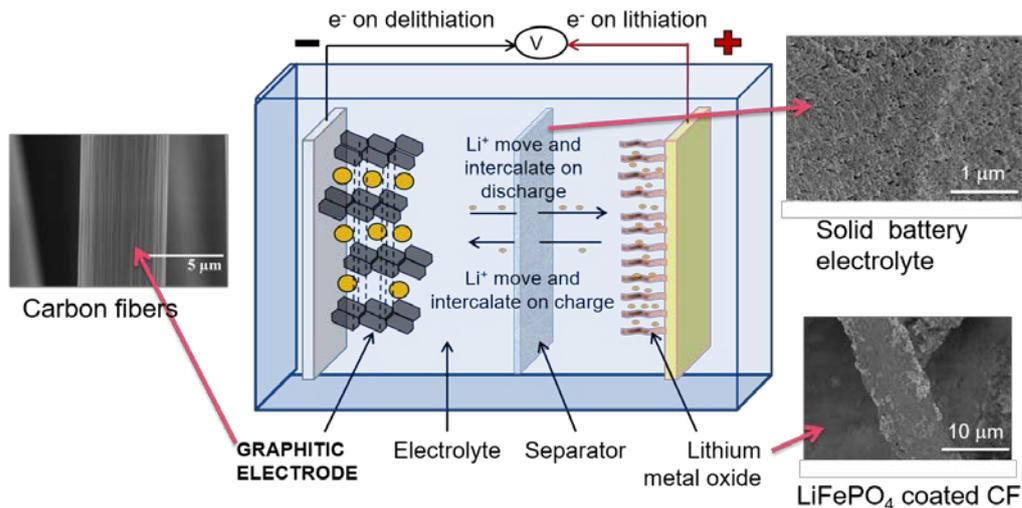


Figure 1: Principal concept for carbon fibre based structural batteries.

### 2.1 Carbon fibre properties

Commercial carbon fibres can be used as negative battery electrodes thanks to their excellent lithium storage capacity. Electrochemical cycling of some types of carbon fibres, particularly intermediate modulus fibres (e.g. Toray T800 and Toho Tenax IMS65) exhibit electrochemical capacities up to 350 mAh/g which is on par with commercial battery electrodes made from graphite [4,6]. They have also been shown to cycle stably and with high coulombic efficiency [7]. Results from typical galvanostatic cycling of T800 fibre and the coulombic efficiency of some carbon fibres are shown in Fig. 2.

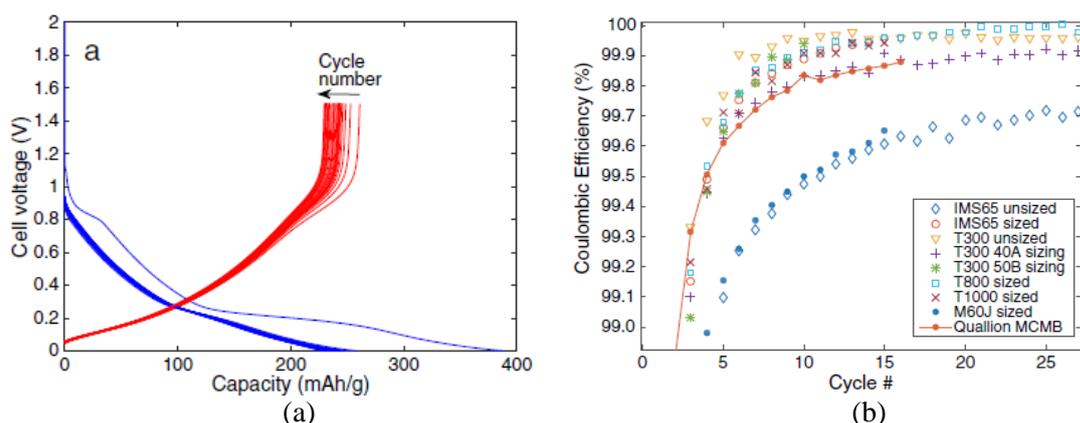


Figure 2: (a) Galvanostatic charge/discharge curves for T800 fibre and (b) coulombic efficiency vs cycle number for different carbon fibres compared to Quallion MCMB commercial graphite electrode material (solid line). From [7].

The mechanical properties of carbon fibre are affected by electrochemical cycling [6]. The stiffness of the fibres remains unchanged with lithium content but the tensile strength drops when the fibres are charged (about 20-30%) but is then recovered when discharged to around 90% of their initial strength. The reason for not fully recovering their tensile strength at discharged state is that some lithium remains trapped in the carbon fibre microstructure after the first electrochemical cycle.

## 2.2 Positive electrodes

On the positive side of the structural battery the carbon fibres are not electrochemically active and some active battery material must be added. This could in principal be any lithium metal oxide compound but in our work we have so far used  $\text{LiFePO}_4$ . The idea is to coat the fibres with active material, thus use the coating as the positive electrode material while the carbon fibres act as the structural backbone and also as the current collector. One path to do this is to electrophoretically deposit a mixture of  $\text{LiFePO}_4$ , carbon black and polyvinylidene fluoride (PVDF) onto carbon fibers [5]. The mixture contained approximately (depending on composition) 90%  $\text{LiFePO}_4$  being the electrochemically active material, about 6% carbon in order to make the coating electrically conducting, and 4% PVDF as a binder. The coated fibres were cycled electrochemically providing quite good capacity of around 60-110 mAh/g of  $\text{LiFePO}_4$ . It was also shown that the coating adhered very well to the carbon fibres providing good mechanical integrity. Other methods to manufacture coated carbon fibre assemblies are being pursued using various method. One promising approach seems to be to use a layer-by-layer assembly to coat  $\text{LiFePO}_4$  onto carbon fibres using cellulosic nano fibrils as a binder material [9].

## 2.3 Structural battery electrolytes

In order to make a structural material a matrix material is of course needed. However, this matrix material must also be ionically conductive for the battery function. This is a great challenge since the best battery electrolytes are liquid and thus have no structural capacity, whereas the best matrix system does not conduct ions, and hence a trade-off has to be made. Several attempts to make such polymer systems have been made but never really reached high enough mechanical properties or ionic conductivity. However, recently a great step towards a well-functioning structural battery electrolyte (SBE) was developed by Ihrner *et al* [10] using an approach called polymerization induced phase separation. In short it is based on having a monomer system (in this case a Bis-phenol A based acrylate) that is fully soluble in a liquid electrolyte creating a homogeneous liquid mixture that phase separates during polymerization. This creates a two-phase system where one phase is a solid polymer phase that can transfer mechanical load and also a fully percolating material for the liquid phase that provides ionic conductivity. The phase separation is in sub-micron scale (see top right in Fig. 1) and

can be tailored to some degree with curing parameters and in-going monomer mixes. Depending on mix and choice of electrolytes these SBE's have an elastic modulus in the range from 500 MPa to almost 1 GPa and ionic conductivities around  $2 \times 10^{-4}$  S/cm. The first versions of these materials were cured using UV-light but recent developments now also allow for heat curing, which is imperative if a multi-layer stack structural battery will be made [11].

## **2.4 Structural negative carbon fibre half-cells**

Having the components, a UD carbon fibre lamina can be made by combining carbon fibres with the SBE. This was made using a vacuum infusion method of the SBE onto a dry UD-layer of spread carbon fibres [12]. Both the mechanical and electrochemical properties were measured on this single layer lamina. The longitudinal elastic modulus was around 52 GPa. This is low but is due to the low fibre volume fraction achieved when making one thin single layer and can be increased substantially when making multi-layer stacks. The transverse modulus was around 2 GPa and also strengths were measured. The electrochemical capacity was around 200 mAh/g of carbon fibres in the layer, which is very good. More importantly, it was found that the SBE adhered well to the fibres and remained adhered even after electrochemical cycling. Measurements of long term cycling of such half-cell laminae has also been performed showing good stability [13].

## **2.5 Multifunctional performance**

Even though a full cell structural battery of the kind has not been realized yet experimentally one can still predict its performance based on the properties of the constituents. There are many design parameters for a structural battery and they impact both the electrochemical and mechanical properties. The mechanical behaviour and electrical energy storage of the structural battery can be matched to the mechanical behaviour of a conventional carbon fiber composite, and the electrical energy storage of a standard lithium ion battery. The latter are both monofunctional and have known performance and mass. In order to calculate the advantage of using structural batteries, the mass of the structural battery can be compared to that of those two monofunctional systems. Performing this analysis [14] shows it would be possible to achieve a 26% mass saving compared to state-of-the-art composite laminates and lithium ion batteries with only slight improvements in constituent properties. However, comparing with other types of structural components, e.g. metal sheets used in electric cars, much larger mass savings are possible.

## **3 CARBON FIBRES AS SENSORS**

Carbon fibres exhibit a piezo-electrochemical response when intercalated with lithium ions resulting in a potential change versus the counter electrode when the carbon fibre (electrode) is subjected to a tensile strain (load) [15]. Carbon fibre bundles were tabbed with glass fibre composite tabbing material, then placed in an electrochemical pouch cell with a thin glass separator, lithium metal as counter electrode and then soaked in liquid electrolyte. The carbon fibre bundle was then subjected to electrochemical charge/discharge cycles and at various points the cycling was stopped, the circuit opened preventing electrical currents to run and a mechanical load was applied. The load created a change in the cell potential - a piezo electrochemical effect. The response seems to be immediate as shown in Fig. 3. The potential response depends on the state of charge (amount of lithium intercalated) and peaks at approximately 50% state of charge (about 150 mAh/g capacity for the two types of fibres tested) [16].

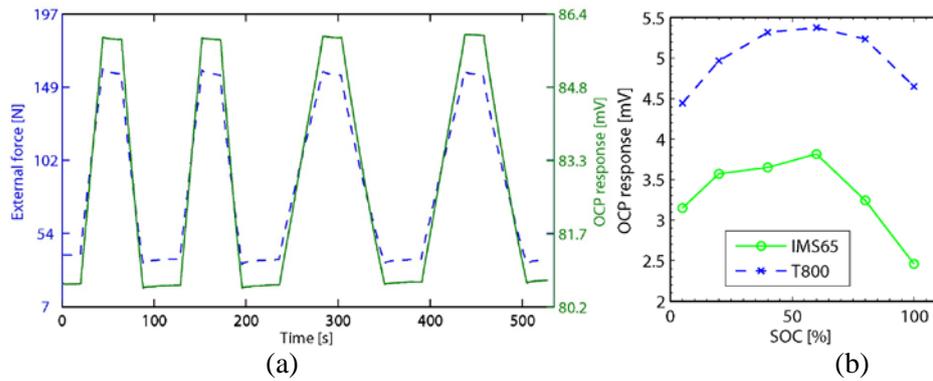


Figure 3: (a) Potential response (open circuit potential response) of a T800 fibre at 80% state of charge subjected to a tensile strain of 0.37% and (b) piezo-electrochemical response of T800 and IMS65 fibres as function of state of charge for an applied strain of 0.37%. From [16].

Similar experiments were recently performed using Sodium as the insertion ion instead of Lithium [17]. A piezo electrochemical response was found again but with a significantly smaller potential change.

A consequence of this piezo-electrochemical effect is that charged carbon fibres creates a change of the electrochemical potential in the order of several mV's when an external load is applied. A carbon fibre could thus also be used to sense the load by measuring the electrochemical potential changes in the material. Our measurements indicate that the response is fast, but this needs to be further studied. With the sensing capability we have a functionalised material, which could be used in applications where the level and spatial distribution of loads in structures are of interest to monitor, e.g. aircraft wing spars. This would facilitate low-cost sensing of both small and large structures.

#### 4 ACTUATION AND MORPHING USING ION INTERCALATED CARBON FIBRES

Electrode materials in batteries expand and shrink (change volume) as the battery is charged or discharged. Graphite, being used in commercial lithium ion batteries, increase its volume by as much as 10% when fully charged with lithium ions [18]. The longitudinal expansion of carbon fibres was measured in-situ, using carbon fibre bundles specimens as a structural electrode in a pouch cell in a similar setup as for the measurements of the piezo electrochemical effect. The stiffness of the carbon fibre bundle is unaffected by the lithium intercalation and the amount of intercalated lithium (state-of-charge) [6]. Carbon fibre bundles in pouch cells were subjected to a constant tensile deformation in a micro-tester. The carbon fibres were then intercalated and deintercalated with Li-ions causing the fibres to expand and contract due to the volume change. The longitudinal expansion strain was then measured indirectly via the force variation on the carbon fibre bundle [19]. The expansion [%] was calculated as the variation of the tensile load divided by the tensile stiffness of the CF specimen, measured in a tensile test. The fibres do not contract fully during discharge due to trapped lithium in the fibres. The longitudinal expansion of two different fibres are shown in Fig. 4. For high specific capacities (high ion content) the measured expansions are quite large, in the order of 1%, and the irreversible expansions (strain difference between charged and discharged states) were around 0.7%.

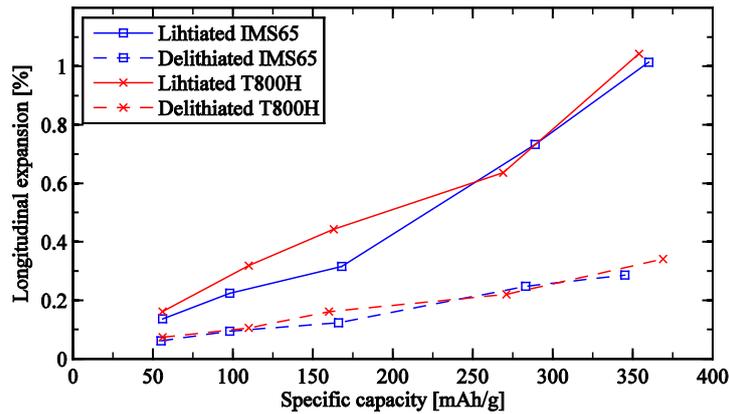


Figure 4: Longitudinal expansion of IMS65 and T800 CFs for different capacities measured after 5 electrochemical cycles.

A problem with such large expansions during charge/discharge cycles in a structural battery would be the shape changes in the structural materials. The expansion strains for lithium intercalation is orders of magnitude larger than those from temperature changes. A model was developed to predict these changes [20]. It was based on classical lamination theory and extended to compute free edge stresses analytically. It was found that both global deformations and free edge stresses can be suppressed by clever lay-up sequences of the negative and positive layers, however they cannot be completely avoided.

One could also use such a model to design laminate sequences that create the largest deformation changes, thus aiming for actuation or morphing structures. Since carbon fibres are very stiff, even a small expansion strain will create very large actuation energy enabling morphing of stiff load carrying structures. A result from the model developed in [20] is shown in Fig. 5 where large bending deformations can be achieved by charging the structural battery.

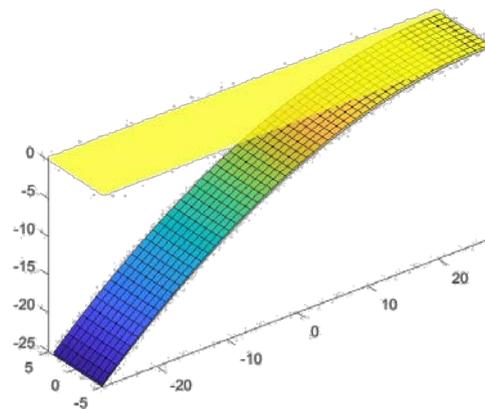


Figure 5: Model of carbon fibre composite actuator

There are several conceivable ways to construct such an actuator by alternating negative and positive layer and their lay-up angles. For example, by placing negative layers top and bottom at +10 and -10 degrees, respectively, very large twisting deformations are possible [20].

A carbon fibre with a Young's modulus of 300 GPa that can expand 0.5% when intercalated with lithium would have a gravimetric energy density of more than 2000 J/kg, i.e. 100 times higher than for

PZT! Such an actuator would create very large forces per unit weight, and at much lower voltage than for PZT's. For morphing structures, the layers must be protected from moisture and covered by some layers of non-active materials, e.g. ordinary composite layers. This will lower the response. Actuation will be slow since the intercalation process in carbon fibres is slow, just as in any ordinary battery. However, this can be achieved with low electrical potentials, a few Volts only. It will also provide a zero-power hold, i.e. when the currents are cut-off the carbon fibres remain expanded, as opposed to a PZT-actuator. New approaches to create morphing structures (structures that change shape), e.g. airplane wings, are thus feasible.

## 5 ENERGY HARVESTING

Applying a mechanical force to Li-intercalated carbon fibres results in a response of the open circuit potential (OCP), i.e., of the carbon fibre electrode potential when disconnected from any circuit as discussed above.

A battery generally delivers less electrical energy during discharge than provided during charge because of a higher cell potential at charge than at discharge (electrochemical hysteresis), as shown in Figure 5a. This is due to the total cell overpotential, an induced voltage difference caused by the electrochemical processes and originating from the charge transfer processes, resistances and transport limitations in the battery cell. However, if the OCP response to a mechanical force carried by the carbon fibre electrode (piezo-electrochemical effect discussed above) overtakes the induced overpotential, more electrical energy will be delivered from the cell than provided, as schematically shown in Figure 5b. This can be achieved by lowering of the charge rate (low C-rate) making the electrochemical losses decrease, i.e. the overpotential decrease.

The carbon fibre has a higher electrode potential than the Li metal and are in essence used as positive electrodes in our setup. That leads to a higher cell potential during delithiation (cell charge) than lithiation (cell discharge) of the carbon fibre electrode, opposite to the behaviour of a normal Li-ion battery cell where the carbon material acts as the negative electrode. However, the electrode potential of the counter electrode is not important for demonstration of the concept.

The tests were performed on tabbed carbon fibre bundles in a pouch cell with liquid electrolyte and a lithium metal counter electrode. Loads were applied with a micro-tester.

The procedure for harvesting energy is to carry out a 4-step harvesting cycle according to the following route [16],

1. In step 1 the applied strain is increased at OCP, which results in an increase of the OCP from an initial potential  $E_0$  to  $E_1$ ;
2. In step 2 the cell is discharged at a constant current density while maintaining the applied strain;
3. In step 3 the discharge is stopped and the applied strain is decreased, which results in a decrease of the OCP to  $E_3 < E_0$ , since the fibres have been lithiated during step 2;
4. In step 4 the cell is charged at the same current density and duration as for the discharge. Thus, the final cell potential is then back to  $E_0$  since the applied strain and state of charge are the same as before step 1.

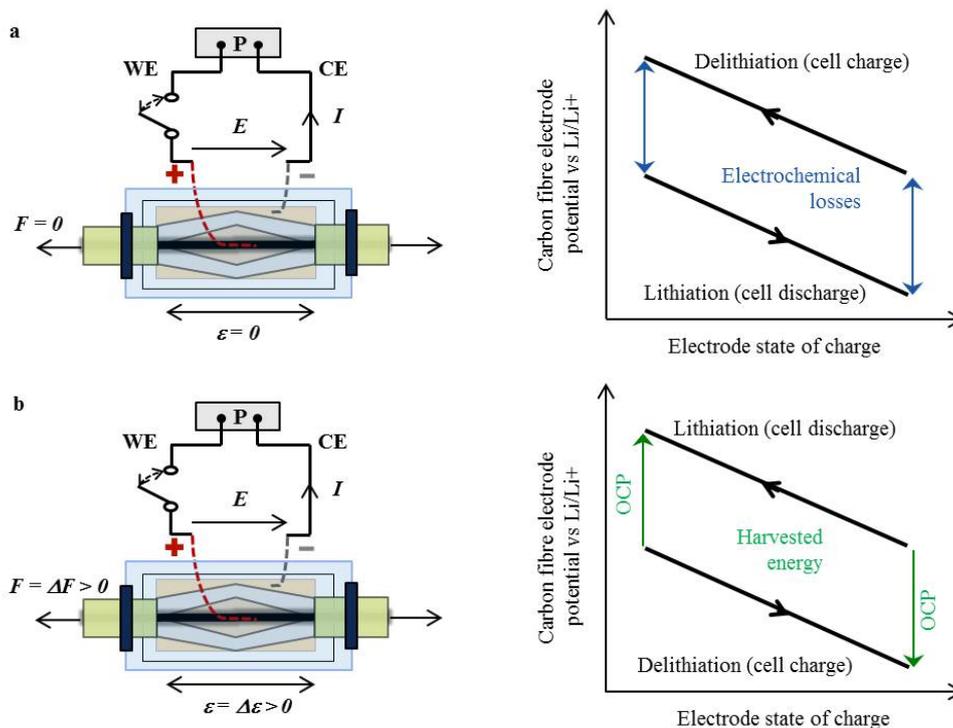


Figure 6: The working electrode (WE) and the counter electrode (CE) of the cell are connected to the potentiostat (P). a) Electrochemical hysteresis when the carbon fibre electrode carries no mechanical force. b) Harvested electrical energy when the carbon fibre electrode subjected to an applied strain  $\epsilon$  and carries a mechanical force  $F$  during lithiation.

The specific electrical energy harvested per cycle is the difference between the electrical work delivered during lithiation (cell discharge) and the electrical work provided during delithiation (cell charge) of the carbon fibre electrode (the area of the encircled charge/discharge cycles shown in Figure 5b marked with “Harvested energy”). The harvester specific power is the useful energy per cycle divided by the cycle time. In the experiments lithiation of the carbon fibre electrode was consistently performed at higher potential than delithiation which clearly demonstrates the concept of piezo-electrochemical energy harvesting. A maximum of  $0.85 \mu\text{W/g}$  was measured for a current density of  $1.2 \text{ mA/g}$ .

Considering a density of  $7.5 \text{ g/cm}^3$ , the typical specific power of MEMS PZT piezoelectric energy harvesters are in the order of  $100 \mu\text{W/g}$ . However, the frequencies are typically  $100\text{-}1,000 \text{ Hz}$  which leads to a specific energy per cycle in the order of  $0.1\text{-}1 \mu\text{J/g}$  per cycle. The specific energy per cycle of the present harvester is much higher,  $100\text{-}350 \mu\text{J/g}$  per cycle at a frequency of  $2.5 \text{ mHz}$  (for  $400 \text{ s}$  cycle time), and it could be even higher for higher cycle times. The energy should therefore be harvested from mechanical motions of very low frequencies. There are, however, several paths to significantly increase the frequency and power. Thinner electrode layers would allow for higher current densities while still having low overpotentials. Hold times between charge and discharge cycles were quite long in these experiments and could be shorted considerably. Yet another way to increase the power is to increase the applied strain on the fibre which increases the OCP response, however, this is of course limited by the failure strain of the carbon fibres which is in the order of  $1.5\text{-}2 \%$ . By embedding the fibres in the SBE it would be possible to make stiff composite layers that can carry bending loads. If so, compression strains can also be applied potentially doubling the piezo electrochemical response and thus the strain regime. This has not been tested yet.

## 6 CONCLUSIONS

We have shown that carbon fibres can be used to create a multitude of functions to a composite material. Carbon fibres can be used as negative electrodes in structural batteries thus providing two functions simultaneously, carry load and store lithium ions. They can also be used as structural backbone for positive battery electrodes by coating them with active battery materials, acting as the main load carrying material and as current collectors. Combining these with a multifunctional matrix system, a structural battery electrolyte, creates a load carrying composite material that can store electrical energy as a battery – a structural battery.

Intercalating ions into the micro-structure of carbon opens up for other functionalities. Carbon fibres become piezo electrochemical allowing for direct sensing of strain in the fibres. They also expand (and shrink) during ion intercalation which can be utilised to create actuation and morphing. Combining the piezo electrochemical response with the expansion allows for converting mechanical energy to electrical energy, i.e. energy harvesting.

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