Versatile 3D Porous Recycled Carbon Garments with Fully-Loaded Active Materials in the Current Collector for Advanced Lithium-Ion Batteries

Hyunjin Cho^{a,g,†}, Yeonho Kim^{b,†}, Yong Ju Yun^c, Kyu Seung Lee^d, Jaeho Shim^d, Chil-Hyoung Lee^d, Jin Won Seo^e, Won G. Hong^b, Hae Jin Kim^b, Hak Yong Kim^{a,f,*}, and Dong Ick Son^{d,h,†,*}

^aDepartment of Organic Materials and Fiber Engineering, Chonbuk National University, 567, Baekje-daero, Deokjin-gu, Jeonju, Jeollabuk-do, 54896, Republic of Korea

^bDivision of Analytical Science, Korea Basic Science Institute, 169-148 Gwahak-ro, Yuseong-gu, Daejeon 34133, Republic of Korea

^cDepartment of Energy Engineering, Konkuk University, 120, Neungdong-ro, Gwangjin-gu, Seoul, 05029, Republic of Korea

^dInstitute of Advanced Composite Materials, Korea Institute of Science and Technology, 92, Chudong-ro, Bongdong-eup, Wanju, Jeollabuk-do, 55324, Republic of Korea

^eDepartment of Materials Engineering, KU Leuven, 3001 Leuven, Belgium

^fDepartment of BIN Convergence Technology, Chonbuk National University, 567, Baekje-daero, Deokjin-gu, Jeonju, Jeollabuk-do, 54896, Republic of Korea

^gSecurity and Disruptive Technologies Research Centre, National Research Council Canada, 1200 Montreal Road, Ottawa, Ontario, K1A 0R6, Canada

^hKIST School, Nano and Information Technology, University of Science and Technology (UST), Gajeong-ro 217, Yuseong Gu, Dae-Jeon 34113, Korea

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† These authors contributed equally to this work.

* To whom correspondence to be addressed: khy@jbnu.ac.kr (H. Y. Kim), eastwing33@kist.re.kr (D. I. Son)

Abstract

We have developed a new and versatile three-dimensional (3D) porous and the conductive carbon spun fabric (CSF) structure and applied it to the current collector for advanced lithium-ion batteries (LIBs). The 3D porous CSF are manufactured from recycled oxidized polyacrylonitrile (Oxi-PAN) staple fibers via the spinning, the knitting, stabilization, and carbonization process in order. Furthermore, we have demonstrated the conductive T-shirts and gloves and investigated the structural, electrical, mechanical, and thermal properties of the CSF through various analytical methods including Joule heating simulation as well as the deformation simulation. The CSF with its 3D porous structure is applied as a current collector for advanced lithium batteries in order to replace the conventionally used metal-based current collector. During battery performances, the porous 3D network structure of the CSF provides effective diffusion pathway for lithium ions during the charge/discharge processes. Consequently, the CSF shows not only the improved cycling stability than that of the conventional aluminum current collector but also demonstrating high-rate performances at high percentage loading of active materials in the current collector. The pouch-type LIBs with the CSF/LiFePO₄ composites electrode exhibits excellent mechanical stability and flexibility with showing a discharge capacity of 148.7 mA h g⁻¹ at 2 C over 250 cycles over the 1200 times bending with a radius of 12 mm.

1. Introduction

Research on wearable electronics has currently focused not only on the development of flexible and stretchable materials that are smooth and durable but also on the integration of efficient energy storage [1-11]. In particular, electrode materials have been widely explored over the past decades as they essentially determine the overall performance of wearable electronics. Conductive fibers [6, 8, 12-15] and fabrics using various metals [6, 8, 13, 16] have been studied extensively. Conductive fiber and fabrics have been fabricated by physicochemical treatments, such as spin coating, sputtering, thermal evaporation [8, 13], and developed conductive inks [8, 13]. However, currently, it remains challenging to produce advanced electrodes for wearable electronics at a commercial level because of high production cost and low dimensional stability, although the conductive fibers, fabrics, and inks intrinsically show outstanding electrical conductivity and flexibility. Therefore, It is essential to improve the disadvantages of the previously reported electrodes [8, 13]. For these reasons, carbon materials, including carbon fibers, carbon nanotubes (CNTs), and graphene, have been extensively studied as one of the promising electrodes for wearable electronics, such as electrochemical energy-storage devices, due to their excellent mechanical and electrical properties [8, 13, 17-22].

Nevertheless, the production of carbon electrode materials for wearable electronics is far from reaching the commercially viable level due to their complicated and economically unfeasible process [18, 23-27]. Especially, for flexible LIBs in large-scale applications of wearable electronics, it is difficult to find a material which meets the essential requirements including high gravimetric energy density, rate capability, and long cyclic life combined with mechanical properties [7, 8, 28-33]. Various flexible carbon electrodes such as CNTs and carbon papers, surface modified metal electrodes, and conventional metal current collectors have not yet been able to meet the advanced requirements in terms of both excellent mechanical and electrical properties [34-38]. While it is challenging to use as an electrode because carbon

papers and CNTs are easily brittle, woven fabric electrode applied as a carbon material can be suitable for a high mass loading of active materials in the current collector with a tight contact to the whole area of the electrode. As to electrochemical performances, the woven fabric structure was utilized by a few research groups [34, 39].

In this work, we demonstrated a versatile and three-dimensional (3D) porous carbon spun fabric (CSF). Unlike the previous carbon fiber fabrics (woven structure, multifilament fibers) [34, 39], we developed the recycled ten-count oxidized polyacrylonitrile (Oxi-PAN) spun yarn manufactured from the defected and sliced Oxi-PAN multifilament fibers via the simple spinning process. After that, the Oxi-PAN spun yarns were knitted for fabricating the 3D porous structure. Finally, the 3D porous CSF was prepared via the stabilization and carbonization process. Furthermore, the structural, electrical, mechanical, and thermal properties of the CSF were investigated through various analytical methods, including not only heat insulation and flexibility testing under crumpled, folded, and twisted conditions, but also thermal image measurement under the standard, stretching, and bending conditions. In the thermal analysis, first of all, the experimental results of the CSF could be matched with the calculated thermal model results.

Secondly, we demonstrated that these knitted CSFs could be easily used for the fabrication of conductive carbon garments, such as T-shirts and gloves, illustrating their facile structural and functional integrity.

Last, the CSF was implemented as the current collector in a LIBs in order to replace the mostly used aluminum current collector. As a result of the experiment, we found that the electrochemical impedance of the battery decreases more than three times as compared with the reference. Furthermore, we investigated the mechanism of lithium-ion behaviors and confirmed the performance of LIBs via various analytical methods including nondestructive 3D X-ray microscopy and field emission scanning electron microscopy (FE-SEM)/atomic force microscopy (AFM) with a scanning kelvin probe microscopy (SKPM) hybrid system. In

particular, during the charge/discharge processes, the porous 3D network structure of CSF provides the effective diffusion pathway for Li-ions. Consequently, the CSF showed the improved cycling stability than that of the conventional aluminum current collector as well as demonstrating high-rate performances at high percentage loading of active materials in the current collector. Using the CSF electrode in LIBs, a discharge capacity of 157.1 mA h g⁻¹ and stable cyclability with 98.2% discharge capacity retention at 1 C during 300 cycles are measured. Also, the pouch-type LIBs with the CSF/LiFePO₄ composite electrode demonstrated outstanding mechanical stability and flexibility. The battery yields a discharge capacity of 148.7 mA h g⁻¹ at 2 C over 250 cycles over the 1200 times bending with a radius of 12 mm.

2. Results and discussion

2.1 The fabrication of conductive carbon garments from recycled staple fibers

Fig. 1 illustrates the manufacturing process of the Oxi-PAN spun yarn obtained from renewable wastes (i.e., defective or sliced Oxi-PAN fibers) and the scheme not only to fabricate conductive carbon garments, namely, a conductive T-shirt and glove but also to make the applications for the energy storage. Initially, the recycled staple fibers are short, approximately 40-50 mm (See Fig. 1a), similar to the typical length of cotton fibers. Staple fibers have the advantage of a relatively low manufacturing cost compared to multifilament fibers, owing to the recycling of oxidized multifilament fibers with defective or sliced parts during the manufacturing process. However, after the recycling process, they are typically heavily entangled, which makes the direct formation of a single yarn during the recycling process itself very difficult. We applied a simple spinning process typically used for cotton fibers to obtain the Oxi-PAN spun yarn (Fig. 1b and Fig. S1). Then, the Oxi-PAN spun fabrics for gloves and garments were produced by knitting process (Fig. 1c and Fig. S1). A detailed description of the manufacturing method of the Oxi-PAN spun yarn, including the spinning process, can be found in Fig. S1 (Supplementary Information). The knitted garments (T-shirts) and gloves became conductive after a

carbonization process at temperatures higher than 700 °C (Fig. 1d and 1e). Compared to the typical carbonization and graphitization temperature required for carbon fibers, the manufactured Oxi-PAN spun yarn and fabrics involve a lower temperature (approximately 1100 °C under the optimum conditions) [40]. The CSF-based heat sink, heater, and thermal insulation are demonstrated in Fig. 1e and 1f. The 3D-porous CSF was implemented as a flexible current collector in LiBs, namely in a coin cell and a pouch cell (Fig. 1 g).

2.2 Characteristics of the CSF through various analytical methods

The characteristics of the carbon spun yarn (CSY) and CSF are measured using various analytical methods, including FE-SEM, X-ray photoelectron spectroscopy (XPS), and X-ray diffraction (XRD), **Raman spectroscopy**, to determine the physical, chemical, electrical, thermal, and mechanical properties. The results are summarized in Fig. S2-S6. We also compared the CSF (along with the wale and course directions) before and after washing ten times in a washing machine by using commercial laundry detergents to verify the durability of the CSF. The results obtained from the pristine and washed CSFs are denoted by CSF (Wale & Course) and CSF (Dry), respectively. Visually, CSFs did not show a significant difference as a function of the carbonization temperature. However, based on the significant drop of the sheet resistance at about 900 °C (Fig. S3a) and the noticeable increase of electrical conductivity at > 1000 °C (Fig. S3b), we selected 1100 °C as the optimum carbonization temperature. The fabrics carbonized at temperatures >1100 °C behaved very similar to those carbonized at 1100 °C (For more information, please refer to Supplementary Information Fig. S2-S6). Consequently, the demonstrators, namely a heat sink, heater, and electrodes for wearable electronics, were fabricated using CSF (Dry) carbonized at 1100 °C.

The directional nature of the thermal conductivity of CSF (Dry) carbonized at 1100 °C was observed (Fig. S3c). In particular, the in-plane thermal conductivity was higher than the isotropic-plane and through-plane thermal conductivities. The obtained values correspond to

those reported for other graphitic structures [41]. In carbon materials, heat is efficiently transferred by electrons and acoustic phonons in a crystal lattice [42-44]. To identify the dominant carrier between electrons and acoustic phonons for thermal conduction in CSF (Dry), we used the equation $[K=K_p+K_e]$ [45]. K_e and K_p are the electron and phonon contribution, respectively. Additionally, we used the equation $[K_e/(\sigma T) = \pi^2 k_B^2 / (3e^2)]$ with the Wiedemann-Franz law [45], where σ is the experimentally measured electrical conductivity value of CSF (Dry) carbonized at 1100 °C. Due to the knitted structure of CSF (Dry), we used the average value of the wale and course electrical conductivities and the average value of the in-plane, isotropic-plane, and through-plane thermal conductivities. k_B is the Boltzmann constant, and e is the charge of an electron. Based on the calculation, we determined that the K_e of CSF (Dry) is about 0.52% (1100 °C) of K (total). The results indicate that phonons dominate over electrons in CSF (Dry) heat conduction. The calculated values are slightly higher than the values reported in previous works [46], but the dominant phonon role in CSF (Dry) is in agreement with that observed for other carbon materials [41, 47].

2.3 The flexibility, durability, resistance and thermal performance of CSF and thermal application

The tensile strengths of the raw Oxi-PAN spun fabric and the CSFs (Wale & Course Dry) carbonized under various temperature conditions were measured (For more information, please refer to Supplementary Information Fig. S3, Fig. S6, Video S1, and Video S2 of the measurement process of the tensile strength of CSF (Dry)). We found the tensile strength of the samples carbonized at high temperature decreased compared to the tensile strength of the raw sample, and we found the CSF has different tensile strengths depending on Wale & Course direction. We believe that those results were due to the carbonization effects and the unique

knitted structure. Consequently, we confirmed that the CSFs have sufficient strength for handling via the additional flexible tests.

Fig. 2a-2c show the results of the 10,000 cycle tests performed to evaluate the flexibility, durability, and resistance of CSF (Wale & Course Dry). The resistance of CSF (Wale & Course Dry) was maintained during the repeated tests, and no deformation was seen. However, little difference was observed in the electrical conductivity in the wale and course directions. We assume that this difference is due to the knitted configuration. In knitted CSFs, the individual yarns are continuous in the course direction, whereas the CSYs are not directly connected in the wale direction. We also carried out cycle tests with an individual CSY (without knitting) prepared under the same 1100 °C carbonization conditions (shown in Fig. S7). The CSY also showed no deformation and constant resistance during the 10,000 repeated cycles. Therefore, we can conclude that most probably the difference in the wale and course directions originates from the knitting.

Fig. 2d-2f show the temperature variation within a CSF (Wale & Course Dry) under an applied voltage of 1 to 6 V. The photographs presented in Fig. 2g-2i illustrate the experimental bending tests performed during the thermal analysis, extracted from Video S3. The temperature of CSF (Dry) gradually increased from the base temperature to a maximum of 82.2 °C (wale direction) and 188.3 °C (course direction) until the voltage reached 6 V, as shown in Fig. 2e-2i. The specific response times were 60 s (0 to 6 V), 20 s (6 V), and 40 s (0 V). In contrast, the temperature of CSF (Wale & Course Dry) sharply decreased after the applied voltage was terminated. Fig. 2g-2i shows the thermal analysis images for the base and maximum temperature when the applied voltage was 0 V, 3.7 V, and 6 V, respectively.

A remarkable difference was observed in the temperature variation of CSF (Wale & Course Dry). As shown in Fig. 2jA-2jB and 2kA-2kB, CSF (Wale & Course Dry) showed uniform heat properties, such as heat dissipation and generation, in most regions of the samples at voltages varying from 0 to 6 V. However, the samples prepared at different carbonization

temperatures performed differently. The heat dissipation and generation effects found with the CSF carbonized at 1100 °C could not be observed in CSF (Dry) carbonized at 700 °C ~ 900 °C, as shown in Fig. S8. Those CSF (Wale & Course Dry) samples have relatively high sheet resistances (approximately 100 ohms or more) compared to CSF (Wale & Course Dry) samples carbonized at >1000 °C, with the lowest calculated sheet resistances of less than 30 ohms. The temperature data measured as a function of the different voltages (1 V ~ 6 V) applied to the CSF samples carbonized at various temperatures are shown in Fig. S8a-S8d along with the consumed power energy, which was derived from the measured current based on the equation [$W = I \times V$]. Indeed, the power consumption is significantly higher for carbonization temperatures of 1000 °C ~ 1500 °C; for the CSF carbonized at 1100 °C, a power of approximately 1 W was obtained at 6 V.

Based on these results, a heater was fabricated using CSF (Dry) carbonized at 1100 °C. Its characteristics are summarized in Fig. 2j and 2k. CSF (Wale & Course Dry) yields a remarkably stable and efficient heat, with the temperature reaching 109.4 °C and 48.2 °C for the course and the wale direction in the extended configuration, respectively, at a low voltage of 3.6 V to 3.8 V. Upon bending, the temperature slightly decreased, to 96.3 °C and 46.7 °C, for the course and wale directions, respectively. Based on the analysis data, it is due to the structural form between Wale & Course direction of the CSF like the different value of electrical conductivity and tensile strength. The consumed power and current in the CSF (wale/stretching, Dry) were slightly higher than those for other CSF conditions (Fig. S9-S11). We assume that the CSF (wale direction, Dry) had the best contact among the carbon staple fibers during the stretching test. It should be noted that the stable heating effect occurred in the entire area, in particular at temperatures higher than 60 °C, and at this low voltage (3.6 V to 3.7 V). Additionally, temperature-dependent Joule heating simulations were carried out for a single loop of the course-standard (0%) and the course-stretching (40%) conditions of the CSF. Deformation of the sample was simulated using finite element analysis (FEA) based on the

SEM images of Fig. 3a and 3b. Fig. 3c-3d and Fig. S12 show the distributions of temperature changes in the FEA models. We found that the Joule heating simulations data (average 114.4 °C) of CSF course-stretching (40%) in the FEA almost corresponded to the experimental data (average 116.1 °C) of the CSF heating at 3.7 V. Moreover, the tendency of the Joule heating simulation data was also in agreement with the experimental data, as shown in Fig. S12. The slight difference between Joule heating simulation data and experimental data could be due to additional experimental factors, such as heat sink from the air and the defected CSY. Additional stress in the FEA and Joule heating simulations of the CSF heater for each of the flat and strain types, depending on the voltages, is described in detail in Fig. S12e-S12j.

Fig. 3e-3g demonstrate multiple applications of wearable electronics, such as flexible electrodes, heat insulation, heat sinks, and heaters, using the conductive CSF. Fig. 3e shows the outstanding properties and possible applications of a flexible electrode based on CSF (Dry) carbonized at 1100 °C. No difference was observed in the resistance of CSF (Dry) even when it was severely crumpled, folded, and twisted, as shown in Video S4. Additionally, Fig. 3f shows the excellent heat insulation properties of CSF (Dry) carbonized at 1100 °C. In the heat insulation experiment shown in Video S5, we confirmed little change in the resistance and surface morphology of CSF (Dry) after subjecting it to the intense flame of a gas torch.

The amount of heat emitted by the wearable electronics, including light emitting diodes (LEDs), affects potential malfunctions and reduces their lifetime [48]. The CSF has an excellent heat dissipation effect because of its carbon mesh with sufficient space for air convection [49-52]. Therefore, the CSF can be considered as suitable for the replacing copper (Cu), aluminum, or graphite, which are heat dissipation materials currently used in wearable electronics and various LED devices. Fig. 3g compares the heat dissipation of a commercial LED chip using the CSF with that of a Cu foil. The results demonstrate that the temperature of the LED chip using the CSF as a heat sink was approximately 7 °C lower compared to that of the Cu foil. (For more information, please refer to Supplementary Information Fig. S13)

2.4 Characteristics of the Carbon spun fabrics/LiFePO₄ (CSF/LFP)@C composite electrode

The schematic overview in Fig. 4a illustrates the coin cell assembly using the flexible and durable CSF as the current collector. The CSF replaced the conventional aluminum and the brittle carbon paper as a current collector. Effectively, the cathode material is impregnated into the CSF, as schematically illustrated in Fig. 4b, and also the electrolyte can easily penetrate the 3D porous structure of the CSF. Fig. S21 shows the electrochemical stability window of the CSF current collector, which stably works in the voltage range of 1.5 – 4.9 V without showing any electrochemical behaviors. We used a 3D X-ray microscope (XRM) in order to directly observe the 3D porous structure of CSF (Fig. 4c and S14) from a cross-sectional sample. XRM analysis provides quantitative information about critical features such as the layer interface, pore morphology, volume percentage, presence of voids, and microcracks. According to virtual cross-sectional data of pristine (only CSF) in the top image (A) of the Fig. 4c (see also Fig. S14a and Video S6), the porous structure contains many voids. The bottom image (B) of Fig. 4c (see also Fig. S14c and Video S8) indicates that the composites structure of CSF/LFP@C is filled in the voids after charging and discharging for about 200 times. In the image, the traces of the slurry containing cathode materials of LFP@C, conductive carbon of Super P, and binder of polyvinylidene fluoride (PVDF) can be clearly observed homogeneously distributed both inside and on the surface of the CSF. Fig. 4d-4f show in color-code the distribution of the components of the sample after the charge/discharge process. The area occupied by LFP@C excluding the CSF is estimated to 48.29% and the remaining 4.06% void which corresponds to the empty space between the LFP active material and the CSF. **During 3D XRM measurement, the exact mass loading of active materials (LFP@C) in CSF is 5.6 mg/cm².** The coverage of the cathode active material over the whole area of the CSF porous structure is 92.24%, as deduced from Fig. 4d-4f. For the clarity, we also examined cross-sectional morphologies with their

elemental maps of CSF/LFP@C and Al/LFP@C composites electrode by SEM analysis (Fig S22). The results above can demonstrate the high percentage loading of active materials in the CSF current collector. When comparing the XRM data obtained from the CSF/LFP@C composites electrode before and after charging/discharging (see also Fig. S14a-S14c and Video S6-S8), it becomes clear that the cross-section SEM images show the direct evidence of 3D packing nature of CSF/LFP@C composites electrode before charge/discharge. It is considered to be the most crucial factor to confirm the damage and durability of the current collector, due to the behavior of lithium ions caused by the electrolyte in the battery elements. Therefore, we investigated the damage of the CSF using the XRM analysis. As shown in Fig. S14 and Video S9, we finally confirmed not only that the morphology of the CSF was relatively uniform but also that the durability of the CSF was excellent. We utilized a hybrid SEM/AFM system equipped with SKPM functionality in order to measure the surface potential and topography of the CSF/LFP@C composites electrode. This measurement enables simultaneous analysis under vacuum. Fig. 4g-4l summarize the results of SEM, AFM, and SKPM analysis of the LFP@C mounted on the CSF before/after cycles. The scanning probe can be seen in the SEM images (Fig. 4g and 4j). Consequently, the position of the sample can directly be followed during the measurement with the AFM tip (Fig. 4h and 4k) and the simultaneous SKPM analysis (Fig. 4i and 4l). The topographic information and surface potential distribution obtained by AFM and SKPM, respectively, clearly demonstrate a morphologic change of the surface of the CSF/LFP@C composites electrode after 200 times charge/discharge cycles. Notably, the average grain size in the film increased almost from ~40 nm to ~200 nm, as can be observed in Fig. 4h and 4k (detailed measurements can be seen in Fig. S15a, S15b, and S15e-S15f). We assume that the change in grain size originates from the change in the lattice structure of the LFP due to Li^+ intercalation/deintercalation during the charge/discharge processes. The surface potential derived from SKPM measurements (Fig. 4i-4l, see also Fig. S15c, S15d, S15g, and S15h) also showed a significant change after the cycles. It significantly decreased with

increasing the cycle number. As the number of cycles increased, the surface potential shifted by a maximum of 200 mV, and a significant reduction was found in the rectangular profile analyzed by the SKPM system (Fig. S15c, S15d, S15g, and S15h). The potential energy reduction in the CSF/LFP@C composites electrode after cycling could be explained as follows: Firstly, it could be related to the transition between LiFePO_4 and Li_xFePO_4 phases during the charge/discharge processes. These two phases exist simultaneously in the CSF/LFP@C composites electrode due to the irreversibility of lithium-ions intercalation and deintercalation processes, and the surface potential decreased because the work function of LiFePO_4 is higher than that of Li_xFePO_4 . Secondly, it could be associated with the disintegration of the electrolyte and some impurities on the surface of the film caused by surface electrochemical reactions. Lastly, the roughness of the grain size could become larger, causing an increase in the surface electrical resistance. Therefore, the surface conductivity is reduced, which leads to a decrease of the surface potential [53-57]. Furthermore, Fig. S16 shows the SEM and Energy-dispersive X-ray spectroscopy (EDX or EDS) image of the CSF/LFP after the cycles, and Fig. S17-S19 indicate the analyzed data, including XRD, STEM, EDS mapping, and XPS data of the LFP@C nanoplatelets.

2.5 The performance and application of lithium-ion batteries.

Based on the experimental and analytical results in Fig. 4 (and also Fig. S14-S19), we can conclude that the CSF was successfully applied as a current collector for the cathode electrode in a LIB by substituting the metal-based current collector of aluminum. Fig. 5 summarizes the electrochemical performances including galvanostatic charge/discharge profiles, rate capability, and long-term stability of LFP@C coated onto the different current collectors (aluminum and CSF). In Fig. 5a the charge/discharge profiles of the SF/LFP@C composite electrode are shown with a different current rate (0.5 C and 5 C). A typical voltage plateau at 3.45 V (vs. Li/Li^+) can be observed, which corresponds to the $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox processes. This result indicates that the

CSF was effectively functioning as the current collector without any electrochemical side-reactions during charge/discharge processes in the voltage range of 2.4 – 4.2 V. In comparison with the conventional aluminum current collector, the CSF/LFP@C electrode delivers comparable discharge capacity of 157.9, 155.2, 152.4, 147.2 mA h g⁻¹ in each 6th-cycle at 0.2, 0.5, 1, 2 C, respectively. The CSF/LFP@C composites electrode shows a slightly higher capacity than the Al/LFP@C composites electrode. Especially for the high rate of 5 C with high percentage (92.4%) loading of the active material, the CSF/LFP@C significantly shows superior performance with a discharge capacity of 133.9 mA h g⁻¹ which is about 8.6% higher than that of the Al/LFP@C composites electrode (123.3 mA h g⁻¹).

Moreover, the value of the initial discharge capacity of 157.9 mA h g⁻¹ at 0.2 C was equivalent to 93% of the maximum theoretical capacity with the mass loading of electrodes (5.6 mg cm⁻²). When a mass loading increases to 11 mg cm⁻², the initial discharge capacity and cycling performance at a rate of 5 C remained comparable after 100 cycles (see Fig. S20). To evaluate the electrochemical stability further, the long-cycling performance of both employed electrodes was measured at 1 C after the initial five cycles at 0.2 C (Fig. 5c). During the cycling experiment, both electrodes of Al/LFP@C and CSF/LFP@C were set at the same mass loading of 5.6 mg cm⁻². While CSF/LFP@C revealed high stability at 1 C during 300 cycles, retaining 98.2% capacity at the last cycle, Al/LFP@C showed significantly lower stability over 300 cycles with retaining of about 96.1% capacity. Especially between 250 and 300 cycles a notable drop can be seen for the Al/LFP@C whereas the CSF/LFP@C shows a constant linear decrease with the number of cycles. The representative Nyquist plots of the Al/LFP@C and CSF/LFP@C electrodes are shown in Fig. 5d before/after 300 cycles. The first semicircle in the high-frequency region can be ascribed to the charge transfer resistance (R_{ct}). Accordingly, the CSF/LFP@C composite electrode yields remarkably decreased R_{ct} value (decreased to 93.5%) compared to that of the fresh CSF/LFP@C cell, which can be ascribed to the efficient electrode activation during cycling. Moreover, CSF/LFP@C shows much smaller diameter than that of

Al/LFP@C, indicating an effective contact established between the uniformly distributed LFP@C and the electrolyte owing to the excellent electrical properties of CSF as well as its porous 3D network structure, which provides an excellent diffusion pathway for lithium ions during the charging/discharging processes.

Our experimental data demonstrate that the novel current collector based on the CSF provides effective diffusion pathways for lithium ions throughout the whole surface area. Moreover, the slurry containing LFP@C, conductive carbon, and PVDF binder can easily and effectively be pasted on both sides of the CSF as well as into the pore channels homogeneously, providing advantageous electrical properties to the composite electrode of CSF/LFP@C. Thus, even at a high mass loading of active materials ($> 11 \text{ mg cm}^{-2}$), the CSF/LFP@C composite electrode shows stable performances and improved kinetics in the electrochemical reaction compared with the Al/LFP@C composite electrode system. In addition, it can also be used as an anode current collector replacing for the Cu-based metal electrode (Fig. S23a). The current collector of the CSF shows electrochemical behavior in the voltage range of 0.1 – 1.5 V, which also contributes to the capacity of CSF/Fe₂O₃. Thus, the discharge capacity is 2.7 times higher than the theoretical capacity of Fe₂O₃ (1007 mA h g⁻¹). However, the assembled cell of CSF/Fe₂O₃ is unstable after 30 cycles at a rate of 0.2 C (Fig. S23), which should be improved further research. To demonstrate the commercial application potential of the CSF/LFP@C composite electrode for flexible LIBs, pouch-type LIBs were assembled with a size of 3 cm × 4 cm (For more information, please refer to Video S10 in Supplementary Information). All the assembly procedures were conducted in a dry room with a dew point below -50 °C. The rate capability of CSF/LFP@C with a mass loading of 5.8 mg cm⁻² is presented in Fig. 5e. The discharge capacity of CSF/LFP@C was 158.9 mA h g⁻¹ at 0.2 C, which is slightly higher than that of Al/LFP@C. The small difference is comparable with that of the coin cell. The relatively large area (12 cm²) electrode of CSF/LFP@C also shows the capacity of 148.7 mA h g⁻¹ at 2 C with stable cycling performance. The assembled battery was bent with a radius of 12 mm for

300 times. The discharge capacity and the Coulombic efficiency remained nearly unchanged even after bending 1200 times (see Fig. 5f for cycles up to 250), demonstrating the outstanding flexibility of the CSF/LFP@C pouch-type cell. Fig. 5g shows the cell bent and folded without affecting the lighting of the light-emitting-diode (LED) lamps.

3. Conclusions

We developed an innovative and multifunctional 3D porous CSF structures using Oxi-PAN spun yarns manufactured from recycled Oxi-PAN staple fibers. These yarns can be knitted to textile, or even to T-shirts and gloves followed by application to the current collector in a LIB. We systematically investigated the structural, electrical, mechanical, and thermal properties of the CSF through various analytical methods and also demonstrated that the CSF could be efficiently used as heat insulation and heat sink. Moreover, the CSF was used as the current collector in LIB for replacing the conventional aluminum current collector. Our results clearly indicate that the versatile 3D porous CSF can provide a high surface area to incorporate LFP@C. The CSF/LFP@C composites electrode showed a stable cycling and high-rate capability than the Al/LFP@C composites electrode. Although the mass loading increased to 11 mg cm^{-2} , it still exhibits the discharge capacity of $156.4 \text{ mA h g}^{-1}$ at 0.2 C and cycling stability up 100 cycles. In the Nyquist plot, the composite electrode of CSF/LFP@C also showed much smaller diameter than that of Al/LFP@C, indicating the formation of effective contact between the uniformly distributed LFP@C and electrolyte, as well as providing efficient lithium-ion pathway during cycling. In a pouch-type flexible LIB, the CSF/LFP@C demonstrated excellent mechanical stability and flexibility. Over 1200 times bending with a radius of 12 mm hardly affect the performance of the flexible battery: a discharge capacity of $148.7 \text{ mA h g}^{-1}$ at 2 C over 250 cycles was measured. Moreover, during the bending and folding of the flexible battery, it still sustained the lightning of the LED lamps. Our results clearly demonstrate that the CSF with its excellent electrical, mechanical, thermal properties and low process-cost is promising for various applications in flexible and wearable devices, e.g. a heater for the people who work in the space (polar and cold area), a heat sink for the commercial electronics, flexible electronics, the protective clothes for firefighters, and an electrode for lithium batteries.

4. Methods

4.1 Preparation of the Oxi-PAN spun yarn, Oxi-PAN spun fabrics, and knitted T-shirt made of Oxi-PAN spun fabrics

The 40-50mm Oxi-PAN staple fibers cut from the defected Oxi-PAN multifilament fibers was bought from ZOLTEK Corporation. The ten-count Oxi-PAN spun yarn was manufactured by the cotton spinning process in SINHAN Spinning Co., Ltd., including mixing, blowing, carding, drawing, roving, spinning and winding [49, 58]. Then, the Oxi-PAN spun yarn was utilized to manufacture Oxi-PAN spun fabrics by a knitting process [49, 58]. In the final step, the prepared Oxi-PAN spun fabrics were used to fabricate products such as T-shirts and gloves. More information about the spinning and knitting processes is presented in Fig. S1 [49, 58].

4.2 Carbonization of the Oxi-PAN spun fabrics and knitted T-shirt made of Oxi-PAN spun fabrics

The knitted Oxi-PAN fabrics, T-shirts, and gloves prepared by the spinning and knitting processes were carbonized at various temperatures from 700 °C to 1500 °C to compare various characteristics. The experiments were carried out in a thermal furnace. The ramping rate was 5 °C per minute up to the required temperature, and after the holding time (60 min.), the samples were naturally cooled down to room temperature. All carbonization processes were performed in an argon gas atmosphere.

4.3 Synthesis of LiFePO_4 (LFP) nanoplatelets

LFP was synthesized by the solvothermal process according to the reported method [59]. After the reaction, the solution was cooled to room temperature, and the resulting precipitate was isolated by filtration, washed with deionized water, and vacuum dried at 80 °C overnight. The as-synthesized LFP was mixed with sucrose (Sigma-Aldrich, 99.5%) at a weight ratio of 5:1

and placed in a tube furnace. Then, it was sintered at 700 °C for 4h under a hydrogen-argon mixture (4 vol% H₂). The carbon-coated LFP sample is designated LFP@C nanoplatelets. The carbon contents in LFP@C nanoplatelets was 3.7%, which was determined by thermogravimetric analysis.

4.4 Characterization of LFP nanoplatelets.

Scanning transmission electron microscopy (STEM) images with energy-dispersive X-ray (EDX) elemental mappings were analyzed with a ZEISS Libra 200 HT Mc Cs (200 kV). X-ray photoelectron spectroscopy (XPS) was carried out with an AXIS Ultra DLD model with a monochromatic Al K α line at 1486.69 eV. Thermogravimetric analysis (TGA) was conducted with a PerkinElmer Pyris 1 TGA at 50-700 °C and a heating rate of 10 °C min⁻¹ in air flux.

4.5 Flexible tests of the CSY and CSF

All samples were placed in a computer-controlled, homemade, motorized actuating system for the repeated bending-releasing test. One side of the sample was the fixed stage, and the other side was the movable stage. The electrical resistance of both sides was simultaneously measured using a 34461A digital multimeter (Keysight Technologies). The reliability test was performed for up to 10,000 cycles of bending from flat to a 1 mm curvature radius (1R).

4.6 Thermal analysis of the CSF (Wale & Course Dry)

For the thermal analysis of the CSF (Wale & Course Dry), the samples were tested for 120 s between 0 and 6 V. The thermal images and analysis results for the heater and heat sink tests, including the bending and stretching tests, were performed using a heater/heat sink performance test system (Synwiz Corporation, SY-HRD-18010)

4.7 Fabrication of the LED electrical circuit on the CSF and Cu foil

To fabricate the samples for application as a heat sink, we prepared commercial LED chips along with the CSF (4 cm×4 cm) and Cu foil (4 cm×4 cm×125 μm). The LED chips were mounted on the center of the surface of the two samples with Pb, silver paste, and electrical wires. The injected voltage was 5 V DC for 90 s. The thermal images and temperatures of the LED chips were measured by an infrared thermal imaging camera (FLIR T-420).

4.8 Characterization of the CSF

The sheet resistance and electrical conductivity of the CSF after the carbonization process were measured by an automatic sheet resistance measurement system based on a four-point probe measurement (ARMS-600, Robot arm X, Y, Z axis weight system, 4 point probes, interval 1.59mm, radius 350 μm, force 160g). The surface observations of the CSF Dry and CSF Wet were performed by FE-SEM, MERLIN (ZEISS). The tensile strength of the samples was measured by a high loading machine (Instron 5985) according to the test method (KS K 0642) of the Korean Standards Association. A conductivity-measuring instrument (TPS 2500S, Hot Disk ab, Gothenburg, Sweden, according to the ISO 22007-2) and differential scanning calorimeter (DSC, DSC 200 F3 Maia, NETZSCH) were utilized to measure the thermal diffusivity and specific heat capacity of the CSFs carbonized at 1100 °C. XRD was carried out by a high-resolution X-ray diffractometer (Panalytical Instruments). The elemental analysis of the materials was conducted by XPS (K-Alpha, Thermo Scientific). **The ratio of I_D/I_G in Raman spectra of CSFs at different carbonization temperatures was measured by Raman spectroscopy (a Horiba Jobin Yvon-Labram HR UV visible NIR Raman microscope spectrometer with a 514 nm Ar laser.).**

4.9 Finite element analysis (FEA) of the CSF heaters

Two-dimensional FEA (2D-FEA) was used to calculate the thermal and strain distributions of the CSF heaters during various deformations of course-stretching (40%), as shown in Fig. 3c-

3d and Fig. S12. Two-dimensional modeling and numerical simulations were performed using COMSOL Multiphysics 5.3. In detail, the Joule heating module and structural mechanics module were used to simultaneously obtain an electrothermal profile and mechanical stress profile of the CSF heaters. In the model, the diameters of single CSF were set to 100 μm , based on observation of the FE-SEM results shown in Fig. 3i and 3j. The density, heat capacity, electrical and thermal conductivity, relative permittivity, Young's modulus, and Poisson ratio of the heater are listed in Table S1.

4.10 Electrochemical characterization of lithium-ion batteries

For the measurement of the electrochemical properties of LFP, the electrolyte was 1 M solution of LiPF_6 (Sigma-Aldrich, 99.99%) dissolved in a solution of ethylene carbonate (EC, Sigma-Aldrich, 99%) and diethyl carbonate (DEC, Sigma-Aldrich, 99%) with 1:1 by volume. The 2032 coin-type cell, aluminum pouch for pouch-type cell, and lithium metal are purchased from Wellcos Corporation. Carbon black (Alfa Aesar, Super P, metal basis, >99%), polyvinylidene fluoride (PVDF, MTI Corp.), polypropylene separator (Celgard 2400), and N-methyl pyrrolidone (NMP, Sigma-Aldrich, >99.5%) were used as received without further purification. The coin-type (CR2032) and pouch-type cells were assembled using an LFP positive electrode, an electrolyte containing a 1 M LiPF_6 in EC:DEC (1:1), and a Li metal negative electrode. The positive electrode was prepared by mixing the active materials, Super P carbon, and PVDF in NMP at a weight ratio of 80:10:10. For the comparison, current collectors of aluminum foil and the CSF have been used throughout the experiments. The slurry was then spread on the above-described current collectors and dried under vacuum overnight. The cells were galvanostatically charged and discharged in a voltage range of 2.4 – 4.2 V by a WBSC3000 automatic battery cycler system (WonAtech Corp.). The electrochemical impedance measurements were conducted using a Gamry Reference 600 potentiostat with a 5 mV AC voltage signal in the frequency range of 0.01 Hz – 100 kHz.

4.11 3D X-ray microscopy measurement

To measure the nondestructive microstructural characterization of the CSF/LFP composites, we acquired unique values for extracting volumetric and quantitative information of the layer interface, pore morphology, and volume percentage and voids from X-ray microscopy (ZEISS Xradia 520 Versa, Carl Zeiss X-ray Microscopy Inc., Pleasanton, CA). Tomography projections of specimens up to 120 μm thick with a $360\ \mu\text{m} \times 240\ \mu\text{m}$ field of view were obtained at the 0.3713 μm pixel size and 40 kV X-ray energy.

4.12 SEM-AFM (SKPM)-based setup

To measure the surface potential and topography of the CSF/LFP composites, we used a unique combined SEM-AFM setup with SKPM from ZEISS MERLIN FE-SEM. SEM imaging and SPM measurements can be performed simultaneously in this combined system, enabling characterization without breaking the vacuum. Both the surface morphology and SKPM experiments were performed in tapping mode using a conductive probe (PR-E75) with a force constant of $3\ \text{N}\cdot\text{m}^{-1}$ and a resonance frequency of 75 kHz. During the SKPM scanning process, an AC voltage ($V_{AC} = 4\ \text{V}$) was applied to the tip. All SKPM measurements were conducted under a vacuum.

Author contributions

H.C., D.I.S., Y.K., and H.Y.K. conceived and designed the experiments. H.C., D.I.S., Y.K., Y.J.Y., and C-H.L. performed the experiments. H.C., D.I.S., Y.K., Y.J.Y., C-H.L., J.S. and K.S.L. measured the analytic data using various analysis tools. H.C., D.I.S., Y.K., Y.J.Y., W. G. H., H. J. K., and J.W.S interpreted and analyzed the data. H.C., Y. K., H.Y.K., and D.I.S. prepared the manuscript.

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Competing interests

The authors declare no competing financial interests.

Additional information

Correspondence and requests for materials should be addressed to D.I.S. and H.Y.K.

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Figure captions

Fig. 1. Schematic of the preparation and application of conductive knitted fabrics. **a** Oxi-PAN staple fibers. **b** Oxi-PAN spun fibers manufactured by the spinning process. **c** Preparation of the Oxi-PAN spun fabrics, T-shirt, and gloves by the knitting process. **d** Preparation of the CSF, T-shirt, and gloves by the carbonization process. **e** Fabrication of the conductive T-shirt and gloves and their application as heat sinks and heaters using the CSF. **f** Application to heat insulation using the CSF. **g** The CSF with 3D porous structure applied to a coin cell and pouch cell for energy storage.

Fig. 2. Repeated resistance test of the CSF, heat management of the CSF and heater applications. **a** Photo images of the 10,000 cycle repeated bending test of the CSF (Dry, 1100 °C). **b** Resistance data for the CSF (Dry, 1100 °C) during the bending tests. **c** Variation analysis of the CSF (Dry, 1100 °C) data during the bending tests. **d** Photo images of the CSF (Dry, Wale & Course) during the experiments (bending and stretching). **e** Thermal analysis data of the CSF (Dry, 1100 °C) over time (120 s, 1 to 6 V). **f** Repeated thermal analysis data of the CSF (Dry, 1100 °C) over time (600 s, 1 to 6 V). **g, h, i** Thermal images of the CSF (Dry, Wale & Course, 1100 °C) as a function of voltage (1 V, 3.75 V, and 6 V). **(j) A-B, (k) A-B** Thermal images of the CSF (Dry, Wale & Course, 1100 °C) at the voltages (3.6 V and 3.8 V) used during the bending test.

Fig. 3. Theoretical thermal diffusion calculations, heat dissipation, heat insulation and stability of the CSF. **a, b** The scheme and SEM images of the course standard and course stretching (40%), respectively. **c A-B, d** The Joule heating simulations data in finite element analysis (FEA) of the CSF course (standard (0%) and stretching (40%)) and the temperature comparison between the modeling & simulations of CSF and the experiments. **e** Photographs

of the flexible, crumpled, folded, and twisted CSF as an electrode for wearable electronics. **f** Photographs of testing the heat insulation of the CSF. **g** A-D Photographs of the LED electrical circuit on the CSF (Dry, 1100 °C) and commercial Cu foil as a heat sink along with heat analysis data from the infrared thermal imaging camera.

Fig. 4. Analysis of LiFePO₄/carbon fabric composites for applying lithium-ion energy storage. **a** The schematic of the coin cell. **b** The schematic of the only CSF (pristine) and the CSF/LFP composites applied to the coin cell. **c** Virtual cross-section photograph of the nondestructive 3D XRM of the only CSF (pristine) with 3-dimensional porous structure (top image, A). Virtual cross-section photograph of the structure of the CSF/LFP composites after charge/discharge Nc=200 (bottom image, B). **d** XRM image showing the total area of the CSF/LFP composites after charging/discharging Nc=200. **e** Estimation of the area of LFP active material excluding CSF. **f** Pore area excluding the LFP active material and CSF. Structural and electrical analysis of the CSF/LFP by SEM-AFM (KPFM). **g** SEM image of AFM tip approaching the CSF/LFP composites before the charge/discharge cycle. **h** Surface topography and **i** Surface potential data before the charge/discharge cycle, scan size of 1.5 μm × 1.5 μm. **j** SEM image of AFM tip approaching sample after the charge/discharge Nc=200 cycle. **k** Surface topography and **l** Surface potential data after the charge/discharge cycle, scan size of 1.5 μm × 1.5 μm.

Fig. 5. Applications for lithium-ion energy storage. **a-c** Electrochemical properties of coin-type cells. **a** Charge-discharge profiles of the CSF/LFP@C electrode at different rates. **b** Rate capability of the Al/LFP@C and CSF/LFP@C composite electrode. **c** Cycling performance of the Al/LFP@C and CSF/LFP@C composite electrode at a rate of 1 C after 10 cycles at a rate of 0.2 C. **d** Nyquist plots of Al/LFP@C and CSF/LFP@C electrodes before and after cycles. **e-g** Electrochemical properties and the corresponding digital images of the pouch-type cell. **e**

Rate and cycling performance of the Al/LFP@C and CSF/LFP@C composites electrode. **f** Cycling performance with the Coulombic efficiency at a rate of 2 C after bending cycles (each arrow indicates bending of battery for 400 times) of the CSF/LFP@C electrode. **g** The corresponding digital images of the pouch-type cell with LED light.

Figure 1. Cho *et al.*

Figure 2. By Cho *et al.*

Figure 3. By Cho *et al.*

Figure 4. By Cho *et al.*

Figure 5. By Cho *et al.*

Versatile 3D Porous Recycled Carbon Garments with Fully-Loaded Active Materials in the Current Collector for Advanced Lithium-Ion Batteries

Hyunjin Cho^{a,g,†}, Yeonho Kim^{b,†}, Yong Ju Yun^c, Kyu Seung Lee^d, Jaeho Shim^d, Chil-Hyoung Lee^d, Jin Won Seo^e, Won G. Hong^b, Hae Jin Kim^b, Hak Yong Kim^{a,f,*}, and Dong Ick Son^{d,h,†,*}

^aDepartment of Organic Materials and Fiber Engineering, Chonbuk National University, 567, Baekje-daero, Deokjin-gu, Jeonju, Jeollabuk-do, 54896, Republic of Korea

^bDivision of Analytical Science, Korea Basic Science Institute, 169-148 Gwahak-ro, Yuseong-gu, Daejeon 34133, Republic of Korea

^cDepartment of Energy Engineering, Konkuk University, 120, Neungdong-ro, Gwangjin-gu, Seoul, 05029, Republic of Korea

^dInstitute of Advanced Composite Materials, Korea Institute of Science and Technology, 92, Chudong-ro, Bongdong-eup, Wanju, Jeollabuk-do, 55324, Republic of Korea

^eDepartment of Materials Engineering, KU Leuven, 3001 Leuven, Belgium

^fDepartment of BIN Convergence Technology, Chonbuk National University, 567, Baekje-daero, Deokjin-gu, Jeonju, Jeollabuk-do, 54896, Republic of Korea

^gSecurity and Disruptive Technologies Research Centre, National Research Council Canada, 1200 Montreal Road, Ottawa, Ontario, K1A 0R6, Canada

^hKIST School, Nano and Information Technology, University of Science and Technology (UST), Gajeong-ro 217, Yuseong Gu, Dae-Jeon 34113, Korea

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[†] These authors contributed equally to this work.

* To whom correspondence to be addressed: khy@jbnu.ac.kr (H. Y. Kim), eastwing33@kist.re.kr (D. I. Son)

Conflict of interests

The authors declare no competing financial interests and conflict interests.

Supplementary Information

Versatile 3D Porous Recycled Carbon Garments with Fully-Loaded Active Materials in the Current Collector for Advanced Lithium-Ion Batteries

Hyunjin Cho^{a,g,†}, Yeonho Kim^{b,†}, Yong Ju Yun^c, Kyu Seung Lee^d, Jaeho Shim^d, Chil-Hyoung Lee^d, Jin Won Seo^e, Won G. Hong^b, Hae Jin Kim^b, Hak Yong Kim^{a,f,*}, and Dong Ick Son^{d,h,†,*}

^aDepartment of Organic Materials and Fiber Engineering, Chonbuk National University, 567, Baekje-daero, Deokjin-gu, Jeonju, Jeollabuk-do, 54896, Republic of Korea

^bDivision of Analytical Science, Korea Basic Science Institute, 169-148 Gwahak-ro, Yuseong-gu, Daejeon 34133, Republic of Korea

^cDepartment of Energy Engineering, Konkuk University, 120, Neungdong-ro, Gwangjin-gu, Seoul, 05029, Republic of Korea

^dInstitute of Advanced Composite Materials, Korea Institute of Science and Technology, 92, Chudong-ro, Bongdong-eup, Wanju, Jeollabuk-do, 55324, Republic of Korea

^eDepartment of Materials Engineering, KU Leuven, 3001 Leuven, Belgium

^fDepartment of BIN Convergence Technology, Chonbuk National University, 567, Baekje-daero, Deokjin-gu, Jeonju, Jeollabuk-do, 54896, Republic of Korea

^gSecurity and Disruptive Technologies Research Centre, National Research Council Canada, 1200 Montreal Road, Ottawa, Ontario, K1A 0R6, Canada

^hKIST School, Nano and Information Technology, University of Science and Technology (UST), Gajeong-ro 217, Yuseong Gu, Dae-Jeon 34113, Korea

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† These authors contributed equally to this work.

* To whom correspondence to be addressed: khy@jbnu.ac.kr (H. Y. Kim), eastwing33@kist.re.kr (D. I. Son)

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1. The manufacturing process of the recycled oxidized polyacrylonitrile (Oxi-PAN) spun yarn and carbon spun fabric (CSF).

Fig. S1. Illustration of the manufacturing process of the recycled Oxi-PAN spun yarn and CSF. **a** Preparation of the initial Oxi-PAN staple fibers (length of 40–50 mm) **b** The mixing and blowing processes untie the entangled fibers and remove dust and impurities. **c** The carding process removes impurities and aligns the slivers. **d** The drawing process doubles the number of slivers (6–8 ea) and repeatedly draws the slivers. **e** The roving process makes a thin yarn with a twist. **f** The spinning process draws and twists the Oxi-PAN roved yarns. **g** The winding process winds the Oxi-PAN spun yarn and distributes the same weight as a cone-type product. **h** The knitting process knits the Oxi-PAN spun yarns and manufactures knitted fabrics with the wale and course directions. **i** The Oxi-PAN T-shirt and glove prepared by an additional manufacturing process.

The manufacturing process of commercially available carbon fibers typically includes a wet spinning process, stabilization, carbonization, graphitization, surface treatment, and sizing process [1, 2]. The raw material of the carbon fibers was polyacrylonitrile (PAN). PAN multi-filament fibers were generally prepared via a wet spinning process [2]. During the subsequent stabilization process, cut and defective PAN multi-filament fibers could be generated. Therefore, the PAN staple fibers with lengths ranging from 40 to 50 mm were prepared by regularly cutting. However, it is difficult to directly manufacture the prepared carbon staple fibers into one yarn due to loose, untied and entangled nature of the carbon staple fibers. Therefore, in order to fabricate a yarn, we utilized a cotton spinning process, which is a well-known technique for producing cotton staple fibers in yarns.

First, the entangled carbon staple fibers were untied through a mixing and blowing process, which included a step that removed mixed dust and impurities. Second, through the carding process, the carbon staple fibers were aligned in one direction, and the remaining impurities were removed. Then, the Oxi-PAN slivers were manufactured. Third, the Oxi-PAN slivers were collected into two or more Oxi-PAN slivers during the pulling process, which occurred concurrently as the drawing process. Fourth, the collected Oxi-PAN slivers were twisted into a thin yarn via the roving process. Fifth, the thin roving yarns were additionally twisted to manufacture the yarn with the desired thickness via the spinning process. Sixth, the Oxi-PAN spun yarn was distributed and wound in the same weight unit through the winding process. Seventh, Oxi-PAN spun fabrics were produced from the Oxi-PAN spun yarn by using a knitting process, and they were manufactured in the wale and course directions. Finally, the manufactured Oxi-PAN spun fabrics were used to produce an Oxi-PAN T-shirt and glove by a manufacturing process commonly used for commercial applications.

2. SEM images of the CSF (Dry) and CSF (Wet) carbonized at various temperatures.

Fig. S2. SEM images of the CSF (Dry) and CSF (Wet) carbonized at various temperatures from 700 °C to 1500 °C. a-r The low- and high-magnification SEM images.

3. Various characteristics of the CSF carbonized in the range from 700 °C to 1500 °C.

Fig. S3. **a** Data for the sheet resistance of the CSF (Dry & Wet, Wale & Course, 700 $^{\circ}C$ to 1500 $^{\circ}C$) and data for the sheet resistance of the CSF (the inset) **b** Data for the electrical conductivity of the CSF (Dry & Wet, Wale & Course, 700 $^{\circ}C$ to 1500 $^{\circ}C$) and the thickness of the samples (the inset) **c** Analysis data for the thermal conductivity (in-plane, isotropic-plane, and through-plane of the CSF (1100 $^{\circ}C$)) **d** Tensile stress data (MPa) for the CSF (Dry, Wale & Course, 700 $^{\circ}C$ to 1500 $^{\circ}C$) **e** The measured dimension (width) of the raw (Oxi-PAN) spun fabric and CSFs carbonized at different temperatures.

Fig. S3a and S3b compare the data of the sheet resistance with the electrical conductivity of the CSF (Wale & Course Dry and Wet). The SEM results in Fig. S2 show that

the surface and morphology of CSF (Dry) did not change much after washing. However, the surfactants used in the washing process most probably remain on the surface, even after rinsing times, resulting in the increased sheet resistance and reduced electrical conductivity of CSF (Wet), as shown in Fig. S3a and S3b and the inset of Fig. S3a. Furthermore, we determined that the carbon contents increased with the increasing carbonization temperature between CSF (Dry) and CSF (Wet), as shown in the X-ray photoelectron spectroscopy (XPS) data in Fig. S4. Those results are due to the different quality of the graphitic structure. We assume that the residual surfactants could also affect the difference between the CSF (Dry) and CSF (wet) samples.

The inset of Fig. S3b shows the thickness of the CSF (Dry) gradually decreasing with carbonization temperature. This trend can be explained by a higher graphitization ratio and by the fact that the interlayer spacing and the void content in the CSFs get significantly reduced, finally leading to a reduced thickness of the sample. We also measured the dimension (width) of all samples as shown in Fig. S3e to compare the difference among them. As a result, we found that after the carbonization process, the dimension was slightly decreased with increasing the temperature of carbonization. However, the carbon content of CSF (Wet) was lower than that of CSF (Dry), as shown in Fig. S4a, 4b, and 4c.

The results presented in Fig. S2 and S3 show that the properties measured were not much different after carbonization at temperatures higher than 1100 °C, (especially from 1200 °C to 1500 °C). For the sake of the low production cost must be considered, a lower carbonization temperature is preferential, and it is vital to reduce the manufacturing process for commercial applications. It is supposed that the manufacturing cost of the CSF in the optimum condition is lower than that of typical carbon multifilament fibers because the samples prepared at the optimum condition have the sufficient electrical conductivity and flexibility despite the elimination of the additional process like the graphitization [3]. Consequently, the carbonization at 1100 °C was selected as the optimum condition.

Fig. S3d shows the tensile strength of the raw Oxi-PAN spun fabric and the CSF (Wale & Course Dry) carbonized at the different temperature conditions. As a result, we found the tensile strength in the wale direction was higher than the course direction because of the unique structure in the knitting process [4]. We believe that the loops in the wale direction became tighter when the tensile stress was applied to the CSF (Dry). On the other hand, the CSY in the course direction became loose with untied fibers. The detailed mechanical response and shape of knitted fabrics were reported through the theoretical study of the fundamental framework [5]. The details of the tensile strength measurements are highlighted in Fig. S6. The tensile strength values of the CSF (Wale Dry) almost linearly decreased with the increasing carbonization temperature. This gradual degradation (approximately 5%) can be explained by the CSY in CSF (Wale Dry) becoming more brittle with the increase of the carbonization temperature.

In contrast, the tensile strength along the course direction (CSF (Course Dry)) remained almost constant despite the increasing the carbonization temperature. We believe that the CSY in CSF (Course Dry) was loose and torn because of the unique structure formed by the knitting process. (Video S1 of the measurement process of the tensile strength of CSF (Dry))

4. XPS analysis data of the CSF (Dry) and CSF (Wet) carbonized at various temperatures.

Fig. S4. XPS analysis data of the CSF (Dry) and CSF (Wet) carbonized at various temperatures ranging from 700 °C to 1500 °C. **a-c** Carbon, nitrogen, and oxygen contents determined by analyzing the XPS data. **d** XPS spectrum of the CSF (Dry) carbonized at 1100 °C. **e** The carbon spectrum of the CSF (Dry) carbonized at 1100 °C. **f** The nitrogen spectrum of the CSF (Dry) carbonized at 1100 °C. **g** The oxygen spectrum of the CSF (Dry) carbonized at 1100 °C. The fits of the data are shown in red in **e-g**.

Fig. S4 indicates XPS analysis data of the CSF (Dry) and CSF (Wet) carbonized at various temperatures. In particular, the carbon peak of the CSF (Dry) carbonized at 1100 °C was considerably higher than the nitrogen and oxygen peaks as shown in Fig. S4d. As a result, the central peak at 284.5 eV was due to C-C and C-H bonds [6-9]. The small peak at 286.3 eV was due to C-O and C-N bonds [6-9]. The other peak at 288.8 eV was due to the O=C-O bond,

as shown in Fig. S4e. The three nitrogen peaks are shown in Fig. S4f. The highest peak at 399.8 eV was for the pyridonic and pyrrolic position, like the N-C-O bond [6-8]. The small peak at 401.2 eV was due to quaternary position nitrogen[6-9]. The other small peak represented the pyridinic position, like the C-N-C bond, at 398.4 eV [6-9]. Lastly, the small oxygen peak is shown in Fig. S4g. The other individual bonds, such as the H-O-C and C-O bonds, were observed at 532.4 and 534.3 eV, respectively [6-9].

5. XRD and Raman spectra of the pristine Oxi-PAN spun fabric and the CSF (Dry).

Fig. S5. Comparison of the XRD and Raman spectra of the pristine Oxi-PAN fabric and the CSF (Dry) at various carbonization temperatures ranging from 700 °C to 1500 °C **a** The XRD spectra of the pristine Oxi-PAN fabric and the CSF (Dry) (carbonized at temperatures ranging from 700 °C to 1500 °C) **b** The Full width at half maximum (FWHM) of XRD spectra of the pristine Oxi-PAN fabric and the CSF (Dry) carbonized at the various temperatures from 700 °C to 1500 °C. **c** I_D/I_G ratio of the pristine Oxi-PAN fabric and the CSF (Dry) carbonized at the various temperatures from 700 °C to 1500 °C. **d** The table of I_D/I_G ratio of the pristine Oxi-PAN fabric and the CSF (Dry) carbonized at the various temperatures from 700 °C to 1500 °C.

Fig. S5a-S5b shows the XRD results with integrated azimuthal profiles (2θ scans) of the raw Oxi-PAN fabric and the CSF carbonized at different temperatures. Typically, the (100) plane of pure PAN fibers leads to diffraction peaks at 17° (2θ) and 29.3° (2θ) [10]. According to the previous study, the intensity of these two pure PAN fiber peaks gradually decreased depending on the operation time of the stabilization process [2]. Moreover, the broad peak at

26.2° (2 θ), which corresponds to the (002) plane, was observed as a result of the prolonged stabilization process. The peak indicates that the PAN fibers have a pre-graphitic structure [10]. Thus, the XRD data of the raw Oxi-PAN spun fabric we manufactured is in agreement with that of stabilized PAN fibers [2]. We also found that the raw Oxi-PAN fabric was partly crystallized during the stabilization process.

Fig. S5a-S5b indicates the normalized XRD spectra (Based on (002) plane) of the CSFs carbonized at different temperatures. As a result, we found the intensity of the peak at 25.2~25.6° (2 θ) near the (002) plane sharply increased with increasing the temperature from 700 °C to 1500 °C. As to the peak at 17° (2 θ), we found that it was almost disappeared in contrast to that of the raw Oxi-PAN spun fabrics. We found the peak positions of our CSFs were in accordance with that of commercial carbon fibers [2, 10]. Thus, we believe that the CSFs could be consist of a turbostratic carbon structure.

Furthermore, we confirmed that the decreasing trend of the width at half maximum (FWHM) of the peaks at 25.2~25.6° (2 θ) near the (002) plane, based on the normalized XRD spectra of the CSFs carbonized at different temperatures. The FWHM trend, which is decreased with increasing the carbonization temperature, indicates the increase of the degree of graphitization [11]. Consequently, the degree of graphitization in XRD spectra based on FWHM value calculated from the various CSFs carbonized at the different temperatures correspond to the trend of the degree of graphitization in XPS and Raman analysis data.

Fig. S5c-S5d shows the ratio of I_D/I_G in Raman spectra of CSFs at different carbonization temperatures corresponded to those of XPS results in Figure S4 and XRD results in Figure S5a-b. In order to confirm the ratio of I_D/I_G , we measured the Raman spectra of CSFs at different carbonization temperatures from 700 °C to 1500 °C. Then, we normalized them based on the I_G peak of the Raman spectra of CSFs. At last, we calculated the ratio of I_D/I_G of the Raman spectra, respectively. As a result, we found that the I_D/I_G ratio decreased with increasing the carbonization temperature, as shown in Figure S5c and S5d. The ratio of I_D/I_G

which is decreased with increasing the carbonization temperature indicates the number of the defect sites in the samples[12, 13]. Therefore, it can be considered as the degree of graphitization. In our results, the trend of I_D/I_G ratio in Raman spectra corresponds to those of XPS and XRD analysis data.

6. Images of measuring the mechanical property of the CSF (Wale & Course direction, dry) obtained after carbonization at 1100 °C

Fig. S6. Measurement of the mechanical properties of the CSF (Dry) produced at various carbonization temperatures ranging from 700 °C to 1500 °C by means of the tensile strength tester (wale and course directions) **A** The picture showing the tensile strength tester **B** Structure of the CSF (wale and course directions) **C** Pictures (a-i) taken during the measurement process of the mechanical property of the CSF (Wale Dry) carbonized at 1100 °C **D** Pictures (a-i) taken during the measurement process of the mechanical property of CSF (Course Dry) carbonized at 1100 °C.

7. Repeated bending test of the carbon spun yarn (CSY, dry) carbonized at 1100 °C

Fig. S7. Repeated bending test of the CSY (Dry) carbonized at 1100 °C **a** Pictures demonstrating the 10,000 cycles repeated bending test of the CSY (Dry, 1100 °C) **b** Analysis of the resistance of the CSY (Dry, 1100 °C) during the bending tests ranging from 0 to 10,000 cycles **c, d** Variation data of the CSY (Dry, 1100 °C) during the bending test from 0 to 1,000 cycles **e, f** Variation data of the CSY (Dry, 1100 °C) during the bending test from 9,000 to 10,000 cycles.

8. The thermal analysis and calculated power (SI unit: W, consumed power energy) of the CSF obtained from various carbonization temperatures.

Fig. S8. Thermal analysis and calculated values of the power (SI unit: W, the consumed power energy) of CSF **a** Thermal analysis of the CSF (Course Dry) at various carbonization temperatures ranging from 700 °C to 1500 °C against voltages from 1 V to 6 V **b** Power analysis of the CSF (Course Dry) at various carbonization temperatures ranging from 700 °C to 1500 °C against various voltages (1 V to 6 V) **c** Thermal analysis of the CSF (Wale Dry) at various carbonization temperatures ranging from 700 °C to 1500 °C against various voltages (1 V to 6 V) **d** Power analysis of the CSF (Wale Dry) at various carbonization temperatures ranging from 700 °C to 1500 °C against various voltages (1 V to 6 V) For **b** and **d**, the power (W) was calculated by using the formula $W = V \times A$, based on the current (A) measured throughout the experiment.

9. The measured resistance (Ω), temperature ($^{\circ}\text{C}$), current (A), and power (W) under the standard, stretching (+40%), and bending conditions (-40%) of the CSF (wale and course direction-Dry) carbonized at 1100 $^{\circ}\text{C}$.

Fig. S9. Data obtained under the standard, stretching, and bending conditions as a function of voltage for the CSF carbonized at 1100 $^{\circ}\text{C}$ **a** Resistance data of the CSF (Wale & Course direction, dry) against the voltage (1 V to 6 V) **b** Thermal analysis of the CSF (Wale & Course direction, dry) against the voltage (1 V to 6 V). **c** The current analysis of the CSF (Wale & Course direction, dry) against the voltage (1 V to 6 V) **d** Power analysis of the CSF (Wale & Course direction, dry) against the voltage (1 V to 6 V) derived from the formula $W = V \times A$ (based on the current (A), which was additionally measured throughout the experiment).

10. The measured thermal images under the standard, stretching (+40%), and bending conditions (-40%) of the CSF (course direction-Dry) carbonized at 1100 °C.

1100 °C (Course-Standard)

1100 °C (Course-Stretching) (+40 %)

1100 °C (Course-Bending) (-40 %)

Fig. S10. The thermal analysis of the CSF (Course direction, dry) carbonized at 1100°C under the standard, stretching, and bending conditions against the voltage A (a-g) Thermal analysis of the CSF (course direction, under the standard and dry conditions) at 1100 °C against the voltage (1 V to 6 V) **B (a-g)** Thermal analysis of the CSF (course direction, under the stretching and dry conditions) against the voltage (1 V to 6 V) **C (a-g)** Thermal analysis of the CSF (course direction, under the bending and dry conditions) against the voltage (1 V to 6 V).

11. The measured thermal image under the standard, stretching (+40%), and bending conditions (-40%) of the CSF (wale direction-Dry) carbonized at 1100 °C.

1100 °C (Wale-Standard)

1100 °C (Wale-Stretching) (+40 %)

1100 °C (Wale-Bending) (-40 %)

Fig. S11. The thermal images obtained under the standard, stretching, and bending conditions of the CSF (Wale direction) carbonized at 1100 °C against the voltage A (a-g) Thermal analysis of the CSF (wale direction, under the standard, and dry conditions) against the voltage (1 V to 6 V) **B (a-g)**, Thermal analysis of the CSF (wale direction, under the stretching, and dry conditions) against the voltage (1 V to 6 V) **C (a-g)**, Thermal analysis data of the CSF (wale direction, under the bending, and dry conditions) against the voltage (1 V to 6 V).

12. The results of the Joule heating simulations and stress simulations of the CSF course-stretching (40%) in FEA.

Fig. S12. The Joule heating simulations and stress simulations in FEA of the CSF heater (course direction) for both, the standard and 40% stretching a-d The Joule heating simulations and stress simulations in FEA of the CSF standard (0%) and stretching (40%) c-d The calculated temperature of the CSF course stretching (40%) at 3.7 V. e-j The Joule heating simulations data of CSF course stretching (40%) from 1 V to 6 V.

13. The heat dissipation of the CSF and Copper (Cu) foil.

Fig. S13. The heat dissipation of the CSF and Cu foil a-b Photographs of the LED electrical circuit on the CSF (Dry, 1100 °C) c A heat sink and the heat analysis data from the infrared thermal imaging camera on the CSF (Dry, 1100°C) d-e Photographs of the LED electrical circuit on the CSF (Dry, 1100 °C) on the commercial Cu foil f A heat sink and the heat analysis data from the infrared thermal imaging camera on the commercial Cu foil.

Fig. S13a-f demonstrates the heat dissipation of a commercial LED chip, which was mounted on the center of the CSF (Dry) carbonized at 1100 °C and on a copper foil for comparison. Fig. S13b and S13e show photographs of the fabricated electric circuit, and Fig. S13a and S13d are images of light emission. When the Cu foil was used as a heat sink, the temperature of the LED chip was 56.8 °C after 90 s.

In contrast, the CSF (Dry) yielded a temperature of 49.8 °C, i.e., approximately 7 degrees lower. This comparison highlights the excellent heat radiation and heat dissipation properties of CSFs at elevated temperatures. In particular, the heat dissipation in carbon materials, like graphene, is known to be superior to that of Cu because graphene has a better thermal emissivity

than Cu [14, 15]. These results corresponded to the observation reported in a previous paper [16].

The experimental results also showed that, although the heat dissipation effect in the CSF was less than that of typical carbon materials (e.g., graphene), the CSF (Dry) can replace metal heat sinks. Notably, the CSF (Dry) can be directly applied as a flexible electrode substrate, which can be used in wearable electronics, as well as a heat dissipation material. For these reasons, we expect that the CSF and CSY will have a significant impact on the development of wearable electronics in the future because the CSF has outstanding properties such as excellent flexibility, remarkable electrical conductivity, and effective heat dissipation. Due to the low power consumption, it also can be used as a flexible and portable heater in extreme environments (e.g., polar regions or space) where the highly efficient heater is required.

14. Virtual cross-section photograph of non-destructive 3D XRM of the CSF with 3-dimensional porous structure and the structure of the CSF/LFP composites before and after Charge/Discharge.

Fig. S14. **a** Virtual cross-section photograph of non-destructive 3D XRM of CSF with 3-dimensional porous structure. **b, c** Virtual cross-section photograph of the CSF/LFP composites structure before and after Charge/Discharge $N_c=200$. **d** Another virtual cross-section photograph of the structure of the CSF/LFP composites after Charge/Discharge $N_c=200$.

15. The results of AFM and KPFM analysis of the CSF/LFP composite before/after the charging and discharging.

Before Charge/Discharge cycle

After Charge/Discharge $N_c=200$

Fig. S15. The CSF/LFP film images and line profile data by AFM **a** Surface morphology and **b** Line profile data before the charge/discharge cycle, the scan size is $1.5 \mu\text{m} \times 1.5 \mu\text{m}$. The CSF/LFP images and line profile data by KPFM: **c** Surface morphology and **d** Line profile data before the

charge/discharge cycle, the scan size is $1.5\ \mu\text{m} \times 1.5\ \mu\text{m}$. **e** Surface morphology and **f** Line profile data after the charge/discharge cycle, the scan size is $1.5\ \mu\text{m} \times 1.5\ \mu\text{m}$. The CSF/LFP images and line profile data by KPFM: **g** Surface morphology and **h** Line profile data after the charge/discharge cycle, the scan size is $1.5\ \mu\text{m} \times 1.5\ \mu\text{m}$.

16. The SEM-EDS image of the CSF/LFP composite before/after the charging and discharging.

Before Charge/Discharge cycle

After Charge/Discharge $N_c=200$

Fig. S16. **a** SEM image of the CSF/LFP composites and **b-e** The EDS elemental mapping of its corresponding compositions of C, Fe, O and P, respectively, before the charge/discharge cycle **f** SEM image of CSF/LFP composites and **g-k** The EDS elemental mapping of its corresponding compositions of C, Fe, O and P, respectively, after the charge/discharge cycle.

17. XRD data of LFP nanoplatelets.

Fig. S17. XRD analysis of LFP@C nanoplatelets.

Fig. S17 indicates the XRD pattern of LFP@C nanoplatelets. All the diffraction patterns can be indexed to an orthorhombic space group without any impurity peaks, *Pnma* (ICSD Card No. 98-016-2282), which indicates the perfect crystalline structure of olivine LFP. Fig. S18 shows that the EDS elemental mapping and the corresponding composition analysis, the elements of Fe, P, and O are distributed homogeneously and their composition is almost the same as the theoretical stoichiometry of LFP.

Moreover, the presence of C atoms in elemental maps represents a successful carbon coating onto the LFP. The Fe 2p spectrum in Fig. S19 shows the two major peaks whose Fe 2p_{3/2} and Fe 2p_{1/2} binding energies were measured to 711.1 and 724.7 eV, respectively. The binding energy separation can be calculated to 13.6 eV, which matched well with the value of Fe²⁺ in LFP.

18. EDS mapping and composition analysis of LFP@C nanoplatelets.

Fig. S18. **a** STEM image of LFP@C nanoplatelets and **b-e** EDS elemental maps of Fe, P, O, and C, respectively, of LFP@C nanoplatelets. **f** EDS profiles spectrum and their composition analysis of LFP@C nanoplatelets.

19. High-resolution XPS spectra of LFP@C nanoplatelets.

Fig. S19. **a** Full survey and **b** Fe 2p XPS spectra of LFP@C nanoplatelets.

20. Electrochemical performance of the coin cell with CSF as a current collector.

Fig. S20. Cycling performance of the CSF/LFP@C electrode with a mass loading of 11 mg cm^{-2} at a rate of 5 C.

21. Electrochemical stability windows of the CSF as a current collector and Cyclic-voltammetry profiles of CSF.

Fig. S21. Electrochemical stability windows of the CSF as a current collector and Cyclic-voltammetry profiles of CSF in the voltage range of 0.1 – 4.9 V at a scan rate of 20 mV/s.

Figure 1 displays EDS elemental maps for Al/LFP@C and CSF/LFP@C. The figure is organized into two rows of panels. The top row (a-f) corresponds to Al/LFP@C, and the bottom row (g-k) corresponds to CSF/LFP@C. Each row includes a main map (a, g), an overlay map (b, h), and individual elemental maps for Al (c), Fe (d, j), C (e, i), and O (f, k). Technical SEM/EDS data is provided at the bottom of each map.

Al/LFP@C Data:

- Panel (a):** Al/LFP@C map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: Al. Color: Blue. Scale: 10.0 μm .
- Panel (b):** Overlay map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: Al. Color: Blue. Scale: 10.0 μm .
- Panel (c):** Al map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: Al. Color: Blue. Scale: 10.0 μm .
- Panel (d):** Fe map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: Fe. Color: Red. Scale: 10.0 μm .
- Panel (e):** C map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: C. Color: Green. Scale: 10.0 μm .
- Panel (f):** O map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: O. Color: Blue. Scale: 10.0 μm .

CSF/LFP@C Data:

- Panel (g):** CSF/LFP@C map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: Al. Color: Blue. Scale: 10.0 μm .
- Panel (h):** Overlay map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: Al. Color: Blue. Scale: 10.0 μm .
- Panel (i):** C map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: C. Color: Green. Scale: 10.0 μm .
- Panel (j):** Fe map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: Fe. Color: Red. Scale: 10.0 μm .
- Panel (k):** O map. Scale bar: 20 μm . Magnification: $\times 10,000$. Date: 12 Apr 2017. Time: 14:14:00. Voltage: 15.00 kV. Beam: 10.0 nA. Detector: SDD. Filter: 10.0 μm . Element: O. Color: Blue. Scale: 10.0 μm .

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23. Charge-discharge profiles of the Cu/Fe₂O₃ and CSF/Fe₂O₃ electrode at a rate of 0.1 C and cycling performance of the CSF/Fe₂O₃ electrode at rate of 0.2 C.

Fig. S23 **a** charge-discharge profiles of the Cu/Fe₂O₃ and CSF/Fe₂O₃ electrode at a rate of 0.1 C and **b** cycling performance of the CSF/Fe₂O₃ electrode at a rate of 0.2 C.

24. TGA curves of LFP and LFP@C.

Fig. S24. TGA curves of LFP and LFP@C.

25. The parameters of the Joule heating simulations in the CSF course stretching (40%).

Table S1. CSF properties used for simulations.

	Course
Density	0.481 g/cm ³
Electrical Conductivity	1157 S/m
Thermal Conductivity	1.060 W/m.K
Heat Capacity	852 Kg/K
Relative permittivity	10 (Grphite 10-15)
Young's modulus	0.360 Mpa
Poisson ratio	0.428

*** Video data**

- S1. Video data of the measurement process of the tensile strength of CSF (Wale-Dry)
- S2. Video data of the measurement process of the tensile strength of CSF (Course-Dry)
- S3. Video data of the thermal analysis data for the standard, stretching (+40%), and bending conditions (-40%) of CSF (Wale & Course direction-Dry) carbonized at 1100 °C
- S4. Video data on the flexibility (crumpled, folded, and twisted) test of CSF (Dry) carbonized at 1100 °C
- S5. Video data on the heat insulation and flexibility (crumpled, folded, and twisted) tests of CSF (Dry) carbonized at 1100 °C
- S6. Video data on virtual cross-section photograph of non-destructive 3D XRM of CSF with the 3-dimensional porous structure.
- S7. Video data on virtual cross-section photograph of non-destructive 3D XRM of CSF/LFP@C composite electrode before charge/discharge
- S8. Video data on virtual cross-section photograph of non-destructive 3D XRM of CSF/LFP@C composite electrode after charge/discharge $N_c=200$.
- S9. Video data on another virtual cross-section photograph of non-destructive 3D XRM of CSF/LFP@C composite electrode after charge/discharge $N_c=200$.
- S10. Video data on CSF/LFP@C applied flexible LIB with repeated bending test.

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