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Accordion-like stretchable Li-ion batteries with high energy density

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ABSTRACT

High-performance stretchable batteries are key components for stretchable devices. However, it is challenging to have both high stretchability and high energy density simultaneously. Herein, we report a design of accordion-like stretchable lithium ion batteries, where the rigid energy storage units are connected by wrinkled and stretchable components. Simulation results show that such accordion-like design and the tape/metal/tape sandwiched structure reduces maximum stress of Al foil in the structure from 31.2 MPa to only 17.1 MPa, which significantly enhance the cell stability. Meanwhile, as the volume of rigid segments are larger than the stretchable part, such design can achieve stretchability of 29% while maintaining 77% (233 Wh L^{-1}) of volumetric energy density of that in conventional packing. Experimentally, the cell shows a high capacity retention of 95.4% even after stretching by 22% for 10,000 times, bending for 20,000 times, and 100 cycles at 0.5 C. It also provides steady power output during continuous dynamic mechanical tests. The corresponding average discharge voltage is only reduced by 1 mV. This accordion-like battery provides an alternative strategy to design stretchable batteries for stretchable devices.

1. Introduction

Stretchability is highly attractive for health care [1–4], sensing [5–7], displays and wearable devices [8–10], since stretchable devices can be conformably applied to human body and other surfaces with arbitrary shape. A successful stretchable device should operate steadily across large shape changes and mechanical stresses [11–17]. Stretchable batteries are highly desired as they can be seamlessly integrated with other stretchable components and provide steady power [8,16,18–24]. Such batteries should maintain reasonably high energy density so that the operation time is not significantly compromised [25–28]. Lithium-ion batteries (LIBs) are attractive to power electronic devices due to their high energy density [29–34]. In recent years, extensive efforts have been devoted into developing stretchable LIBs. PDMS and other stretchable polymers-based devices have been demonstrated with excellent stretchability, but they suffer from low energy density [29,35–38]. Buckled carbon structures (e.g. carbon nanofibers, carbon nanotubes) also have high stretchability, but corresponding energy densities are still not satisfactory [39–41]. Besides energy density, an intrinsic challenge in stretchable batteries is to develop stretchable packaging materials that perform as well as aluminum-plastic film regarding to air and moisture-proof capability in long term, which has not yet been demonstrated to the best of the authors' knowledge. To circumvent this challenge, wire-like designs have been proposed [42–44]. This highly stretchable structure creates new opportunities for cloth-based energy storage, but the energy density still needs to be improved. Recently, Liu et al. reported a wave-like device, where electrodes and packaging layers were shaped in a wavy structure together, so that the packaging material does not need to be directly stretched. An attractive energy density of 172 Wh L^{-1} was achieved [45]. However, no data was presented on the power stability when the cell undergoing dynamic deformation, which is crucial for stretchable batteries.

Here we report a new design inspired by the structure of an accordion (Fig. 1a), which can decouple the stretchable component and the energy storage component. Thus, high energy density and high

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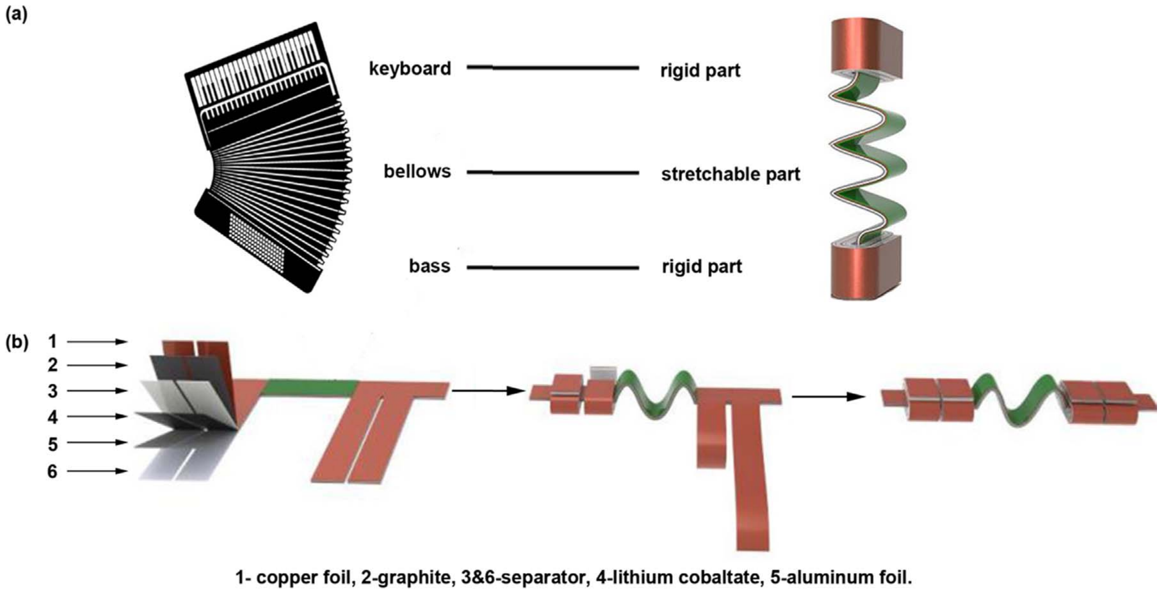

Fig. 1. The design of the accordion-like battery. a) Analogy between the proposed design and an accordion. The rigid parts correspond to the keyboard and bass and the stretchable part mimics the bellow of accordion. b) The fabrication process of an accordion-like battery.

stretchability can be achieved simultaneously. The stretchable part acts as the bellows of an accordion, which allows the device to be extended readily, and the rigid part with high energy density behaves like the keyboard and bass of the accordion to store energy. The fabrication process is illustrated in Fig. 1b. Battery electrodes are first cut into comb-like structure, and tape is laminated to the backbone, which provides mechanical support to the thin metal foils and prevent them from fracture. Moreover, as the tape is only applied to the bellows-like region, it does not lead to redundant volume in the keyboard/bass part for energy storage, and has little effect on the volumetric energy density. Then the extended branches are wrapped around the backbone. After sealing in an Al laminated bag, the bellows-like part is manually pressed into curved structure to render the battery stretchable. Such an assembly strategy not only provides attractive stretchability and high energy density, such as 29% stretchability and 77% energy density of conventional cells with same parameters, but also has the potential to scale up and be compatible with conventional battery fabrication processes.

Moreover, compared with the structure of multiple small batteries linked by springs, our design can avoid packaging of multiple cells and fatigue at cell connections. In this report, we experimentally demonstrate this concept with steady electrochemical performance after stretching 10,000 times, bending for 20,000 times and charging/discharging for 100 cycles at 0.5 C. The corresponding capacity retention is as high as 95.4% in a 34.8 mA h pouch cell. The power output is also steady during dynamic stretching and the corresponding change in the average discharge voltage is only 1 mV at 0.5 C. In addition, the cell does not catch fire or generate smoke in the nail penetration test.

2. Structural designs

To have a theoretical understanding of the proposed design, we first analyze the trade-off between energy density and stretchability in our design, which depends on the relative dimension of the bellows (stretching length, L) to the keyboard and bass (energy storage length, a). With the design shown in Fig. 2a, given the bending radius r equals to 0.75 mm, and when the ratio of L/a is 0.30, the stretchability can reach 29% theoretically, and the corresponding energy density is 77% of a battery by conventional packaging but without the bellows-like part (Fig. 2b). When reasonable values in commercial batteries are used (e.g. 3 mA h cm^{-2}), the energy density of proposed stretchable batteries can reach 233 Wh L^{-1}. Details of calculation can be found in the Supporting Information (Fig. S1). Similarly, when $L/a = 1$, the stretchability can further increase to 120%, with a relative energy density of 50% (Fig. 2b) Moreover, compared with the structure of multiple small batteries linked by springs, our design can avoid packaging of multiple cells and fatigue at cell connections.

To understand the strain/stress distribution in such accordion-like design and how applied tape release the mechanical stress, numerical mechanical simulations were carried out. Two configurations were compared with each other: metal foils without tape protection (Fig. 2c) and metal foils with tape laminated on both sides (Fig. 2d). In the simulations, stretchable layers are laterally compressed (Fig. S2, Supporting Information) into buckles with a half-wavelength of 5 mm, resulting in the smallest bending radius of ~1.5 mm at peaks, similarly to that in accordion-like batteries we fabricated. As shown in Fig. 2d, the separator and tape layers can effectively undertake the strain in the battery, and mitigate stress in metal foils. Without tape protection, the maximum stress in Cu and Al foil are 33.4 and 31.2 MPa, respectively. Once tape is applied, the maximum stress in Cu and Al foil are reduced to 19.9 and 17.1 MPa, respectively. Such low stress in the stiff metal foil attributes to two reasons. First, the local wrinkling of the stiff layer of metal foil sandwiched between soft tape layers can release film stress. Additionally, the strong adhesion of tape increases the effective bending stiffness of tape/metal/tape sandwiched structure compared with bare metal foils, thus reducing the bending curvature. Importantly, the tape protects metal foils from randomly harsh bending and plastic deformation. Therefore, the laminated tape allows to stretch our cells for thousands of cycles.

Parallel experiments were conducted to further demonstrate the significance of supportive tape. As shown in Figs. 2e and 2f, after only 1000 stretches of the bellows-like part, both bare Al and Cu foils without tape were broken. However, when laminated by the tape, neither Al nor Cu was broken after 10,000 stretches, and no obvious cracks were observed in Scanning Electron Micrographs (SEM, Figs. 2g and 2h; Fig. S3, Supporting Information). The foils also looked similar to pristine foils without stretching in Fig. S4 (Supporting Information). In addition, we also examined the folding edges of the rigid parts in both electrodes (Fig. S5, Supporting Information). As expected, this section showed no delamination without seeing metal substrate after mechanical deformations described above, as it had not been stretched at all.

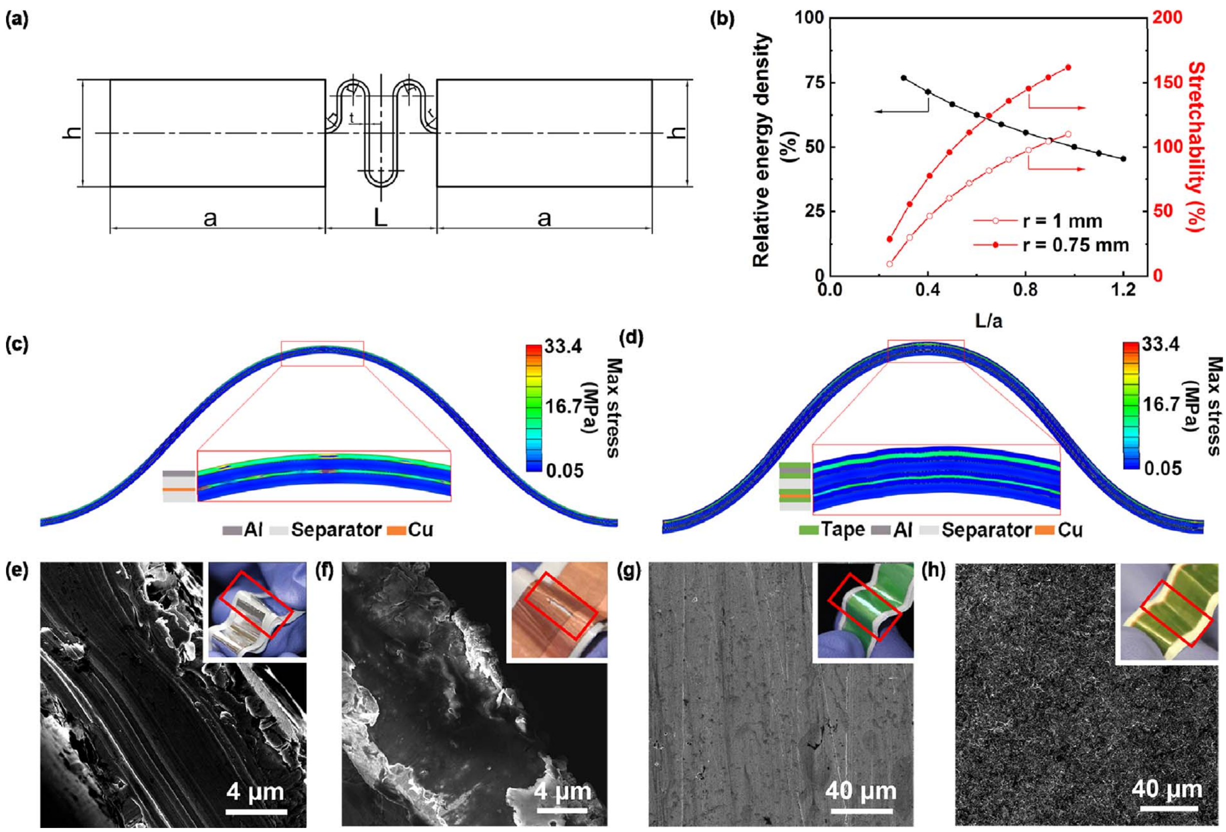

Fig. 2. The relative energy density and numerical mechanical simulations. a) The schematic of the geometry of an accordion-like battery for calculating relative energy density and stretchability. b) The calculated relative energy density and stretchability of the accordion-like battery as a function of *L*/*a* for different radii. c) and d) Stress contours of the stretchable layers without and with tape protection, respectively. e) and f) SEM images of Al and Cu substrate, respectively, in the bellows-like region after stretching 1000 times without protective tape (cross-sectional areas). g) and h) SEM images of Al and Cu substrate, respectively, in the bellows-like region after stretching 10,000 times with protective tape. The insets in c-f) are corresponding optical images.

3. Electrochemical performances

The mechanical analysis suggests high mechanical stability of our accordion-like design. To demonstrate that such mechanical stability can be transferred to excellent electrochemical performance, a $LiCoO_2$ (LCO)/graphite cell was tested at 0.5 C (1 C = 145 mA g^{-1}) at different states for 100 cycles. This cell had a capacity loading of 0.96 mA h cm^{-2}, a total capacity of 34.8 mA h, and stretchability of 22%. During the test, the cell was first cycled in the flat condition (Fig. S6, Supporting Information) for 20 cycles, and the specific capacity slowly changed from 147.6 to 144.3 mA h g^{-1} (Fig. 3a). Then the cell was stretched 10,000 times (Fig. 3b), followed by tests in the fully stretched state for the next 50 cycles (region II). The specific capacity slightly increased from 144.3 mA h g^{-1} before stretching to 146.4 mA h g^{-1} after stretching. Moreover, the capacity remained at 140.5 mA h g^{-1} after 50 cycles in region II, corresponding to a slow fading of 0.08% per cycle. Similarly, after 90° and 180° bending (Fig. S6) for 10,000 times, the capacity remained almost constant under both bent (120°) and pressed states, respectively, as illustrated in cycle 70–100 in Fig. 3a. The capacity remained at 140.8 mA h g^{-1} after 100 cycles. During the 100 cycles, the Coulombic efficiency (CE) was highly robust, and the average CE reached 99.70%, indicating that the mechanical deformation did not cause noticeable damage to SEI and no significant side reactions occurred. As shown in Fig. 3c, the change in capacity over 100 cycles was only 6.7 mA h g^{-1}, corresponding to a fading of 0.046% per cycle. The cell's robustness was further proved by the impedance measurement at the fully discharged state, presented in (Fig. S7, Supporting Information). From the flat state in cycle 20 to the stretched state in cycle 21, the charge transfer resistance changed slightly from 2.94 to 2.99 Ω, and the overall absolute value of impedance at 0.1 Hz only increased slightly from 38.1 to 38.5 Ω (Fig. S7). This suggests no noticeable delamination of materials or other forms of damage exists during the stretching process. The energy density of such cell depends on the thickness of electrode coating. When high mass loading electrode material (e.g. 3 mA h cm^{-2}) is used, an attractive energy density of 233 Wh L^{-1} can be obtained, including all components and packaging.

The mechanical stability is further examined by the voltage fluctuation during manual stretching , 90° bending, and 180° bending at ~0.2 Hz. Regardless of stretching or bending, the maximum voltage fluctuation is less than 10 mV in both discharge and charge at 0.5 C (Fig. 3d and Fig. S8, respectively), which indicates the resistance change is less than 5% (see Supporting Information for detailed calculations). To further illustrate the stretching stability of our design, the cell was stretched, bent 90°, and bent 180° throughout the whole charge/discharge cycle (Fig. S9, Supporting Information), and there was still no obvious change in overpotential, which indicates that such mechanical operations have almost no impact on the electrochemical performance of our cells.

Our design also shows excellent high rate performance under mechanical deformation. When it was in the 180° bent state, the discharged capacities at 0.5, 1 and 2 C were 144.2, 138.4, 123.9 mA h g^{-1}, respectively, and the capacity was recovered after returning to 0.5 C (Fig. 3e). The capacity at 2 C was as high as 86.0% of that at 0.5 C, which proves that our design functions well at high current rates. In contrast, when a conventional stacking cell with a capacity of 33.4 mA h was cycled under 180° bent state, its discharge capacity quickly dropped from 123.1 at 0.5 C to 97.4 mA h g^{-1} at 2 C. This represented only 79.1% capacity retention. Such improved rate performance attributes to the less damage to the interfacial contact

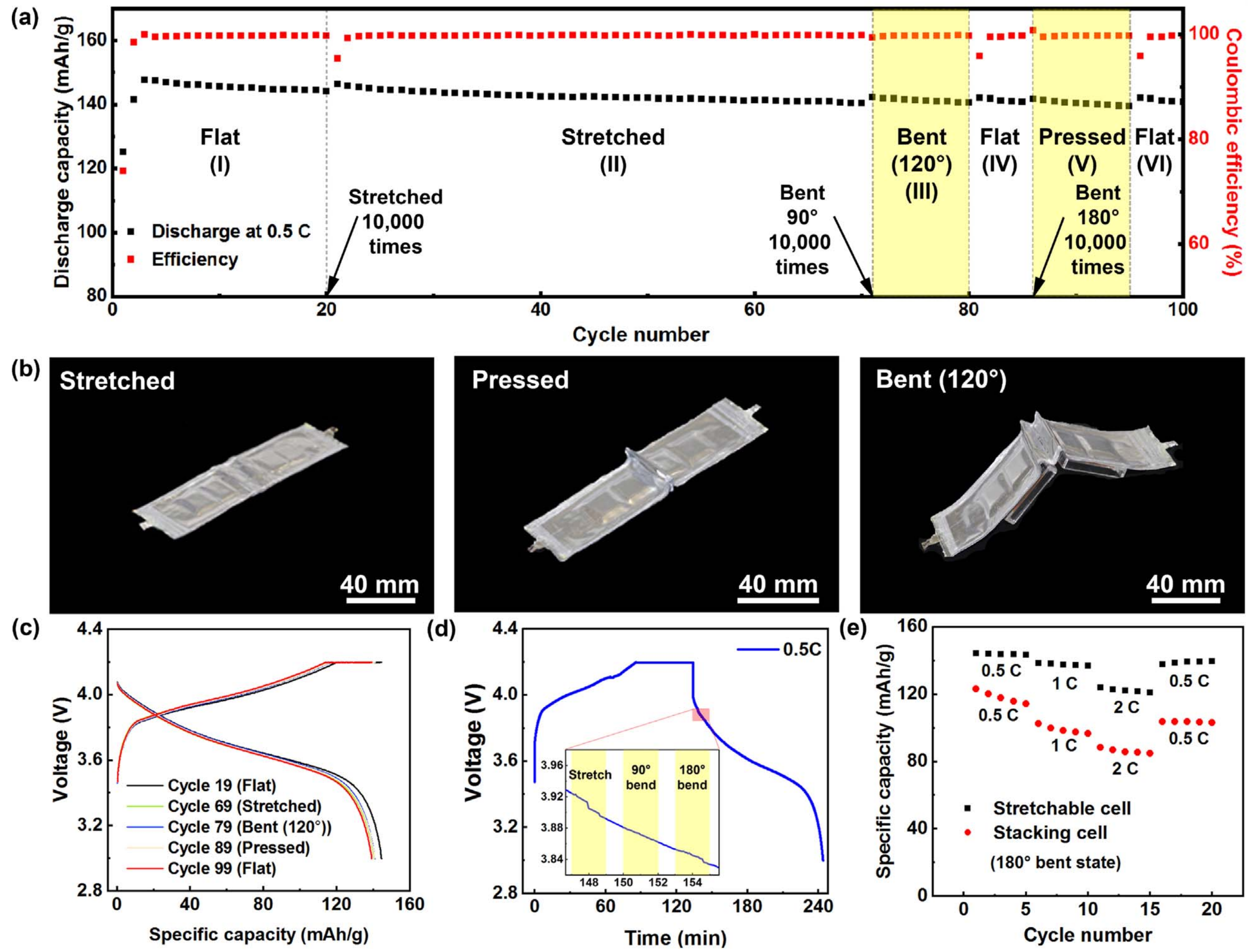

Fig. 3. Electrochemical performance of an accordion-like $LiCoO_2$/graphite battery under different states of mechanical deformation. a) Galvanostatic cycling in various deformation states for 100 cycles. The voltage range is 3.0–4.2 V, and the current rate is 0.5 C. A constant voltage step with cut-off of 0.05 C was added at the end of charging. b) Optical images of the accordion-like battery in stretched, pressed and bent (120°) states. c) Galvanostatic charge/discharge curves at different states of deformation and representative cycles (16th, 69th, 79th, 89th, and 99th). d) Galvanostatic charge/discharge curves at 0.5 C in the flat state; during this period, the battery was continuously stretched, 90° and 180° bent and the inset shows the voltage fluctuation under different mechanical deformations. e) Rate performance at 0.5 C, 1 C, and 2 C for accordion-like battery and a stacking cell with similar size and capacity.

between the electrode and the current collector in the accordion-like structure.

Practical operation of stretchable batteries not only requires steady electrochemical performances under static configurations, but also needs stable power output upon dynamic operations. To evaluate the dynamic performance, a stretchable battery was charged/discharged with continuous stretching. The cell was first cycled in the flat state for 5 cycles at 0.5 C, and the discharge capacity was 133.4 mA h g^{-1} in cycle 1 and 132.9 mA h g^{-1} in cycle 5, showing a steady performance. The two ends of the cell were then fixed to two linear actuators for dynamic stretching (Fig. 4a). During the dynamic stretching, one end was fixed and the other end moved back and forth with a speed of 12 mm s^{-1} and an amplitude of 24 mm, corresponding to a stretching range of 22%. The frequency was 0.04 Hz (Video S1 and S2, Supporting Information). The dynamic test was carried out for stretching 5,200 cycles within ten charge/discharge cycles, followed by another five cycles in the flat state. During the ten cycles of continuous stretching, the capacity was reduced only slightly from 132.7 to 129.6 mA h g^{-1}, indicating the high stability during dynamic stretching (Fig. 4c). The voltage profiles before, during, and after stretching coincided with each other. The average discharge and charge voltage in cycle 8 (during stretching) were only 1.1 mV less than and 0.9 mV larger than those in cycle 3, respectively, which means that there is no obvious increase in overpotential (Fig. 4b). These results together prove that the accordion-like design offers highly stable electrochemical performances even under dynamic stretching.

To further demonstrate the design's potential in real applications, a stretchable battery was used to power 17 LEDs in a “CU” pattern under both stretched and pressed states (Figs. 5a and b, respectively). The cell had a capacity of 35 mA h and the operating current was 23 mA. The cell can provide steady power at both stretched and pressed states, and during continuous deformations (Video S3, Supporting Information). The safety of our battery, a critical factor for wearable devices, was also examined. After a 35 mA h cell was charged to an open circuit voltage (OCV) of 4.11 V, the rigid part was punctured by a nail and the OCV dropped quickly to 0 V (Video S4, Supporting Information). No sign of fire or smoke was observed, as shown in Figs. 5c and d. These results show promising power and safety performance for practical applications in wearable devices.

Supplementary material related to this article can be found online at doi:10.1016/j.ensm.2018.11.019.

4. Conclusion

In conclusion, we have demonstrated a simple strategy to fabricate accordion-like stretchable batteries. Such design can realize attractive energy density, excellent cycling performance, reasonable stretchability, and mechanical robustness. A high energy density of 233 Wh L^{-1}

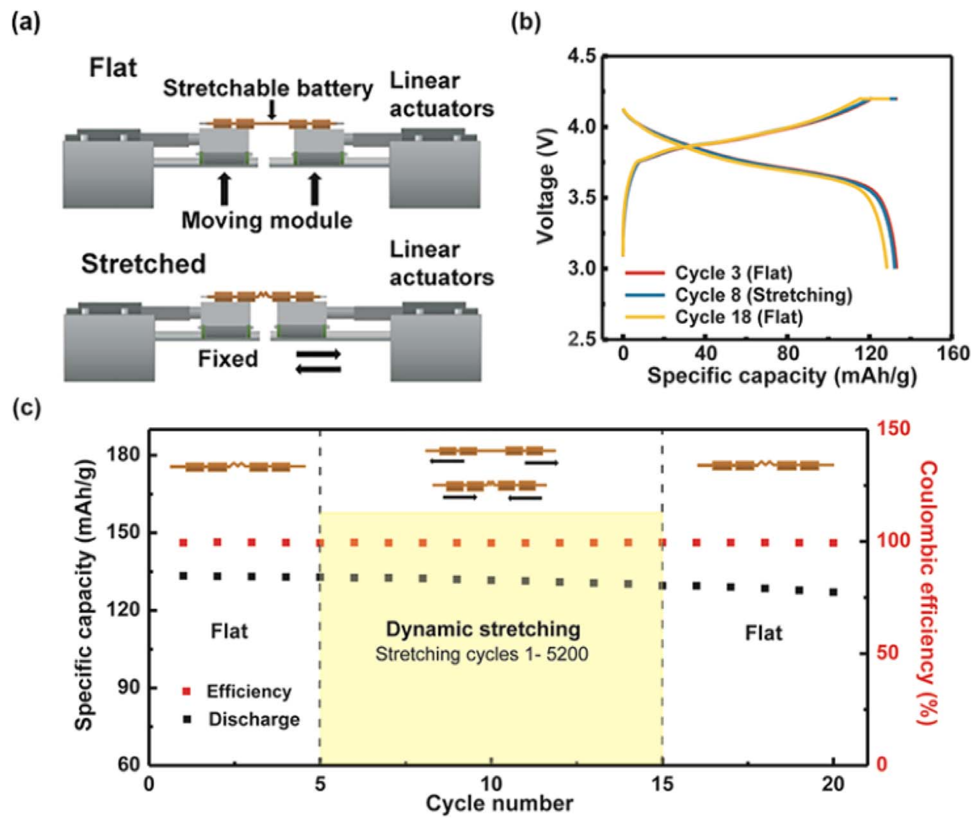

Fig. 4. The dynamic test of an accordion-like battery. a) The schematic of a stretchable battery on the testing device. b) The voltage profile of the stretchable battery in the 3rd, 8th, and 18th cycles. c) The dynamic cycling performance of the stretchable battery under different states.

can be achieved when electrode materials with high mass loading is applied. In addition, after stretching and bending at 90° and 180° for 10,000 times each, and 100 charge/discharge cycles, 95.6% of the cell capacity still remained. This is attributed to two factors: 1) Accordion-like design decouples mechanical stretching and energy storage, so that stretching does not apply stress onto electrode particles. 2) The protective tape provides mechanical support so that thin metal foils hardly break during stretching. Furthermore, the accordion-inspired cell presented excellent electrochemical stability during ten cycles of continuous and dynamic stretching. The potential for practical application and excellent thermal stability were also demonstrated in powering multiple LEDs and nailing test. Our design provides a new approach to fabricating stretchable battery in a scalable fashion.

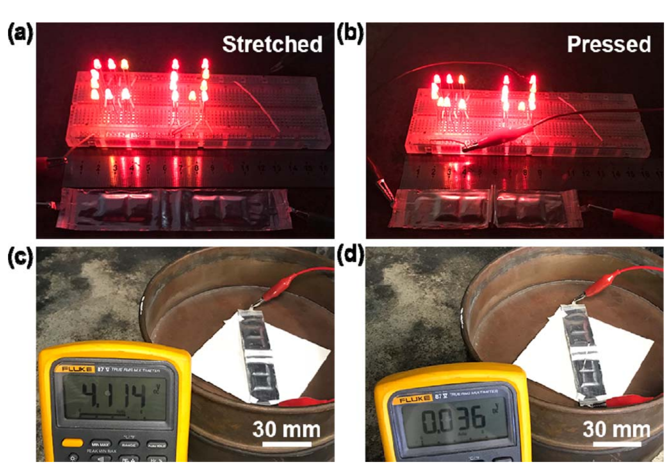

Fig. 5. Practical application and safety test of the stretchable battery. a) and b) The "CU" pattern lighted by a battery in a) stretched and b) pressed state. c) and d) The optical images of a stretchable cell c) before and d) after being nailed. The OCV before nailing is 4.11 V, and drop to 0 V after nailing.

5. Experimental section

5.1. Battery fabrication

Single-side commercial $LiCoO_2$ cathode and graphite anode are provided by Custom Electronics Inc. The typical mass loading is 6.6 mg cm^{-2} and 7.7 mg cm^{-2}, respectively. After electrodes are cut into predesigned shape, they are stacked with Celgard separators and wrapped according to Fig. 1. Then the electrode stack is packed in aluminized pouch bags (Sigma-Aldrich) and vacuum sealed to assemble pouch cells inside an argon-filled glovebox ($O_2 < 0.1$ ppm, $H_2O <$ 0.1 ppm). The electrolyte is 1 M $LiPF_6$ in ethylene carbonate/diethyl carbonate (1:1 vol/vol) (Gotion Inc.). The typical cell has a theoretical capacity of 34.6 mA h. The cell is rested for 6 h before electrochemical performance tests. For making stacking cells, the electrodes are first cut into the same width as the stretchable battery, and stacked together.

5.2. Electrochemical tests

The Landt battery analyzers and Bio-logic VMP3 potentiostat are used in electrochemical tests. The typical voltage range is 3.0 V to 4.2 V. In electrochemical impedance spectroscopy, the frequency range is 1 MHz to 0.1 Hz with an amplitude of 10 mV.

5.3. Mechanical simulations

The bending condition of electrode layers was simulated using 2D nonlinear finite element method, implemented in the commercial software ABAQUS. In all cases, four-node quadrilateral stress/displacement elements with reduced integration were used and the mesh density was verified by mesh convergence studies. The layers of tape/copper foil/tape/separator/tape/aluminum foil/tape/separator were stacked together without sliding. For simplicity, linear isotropic elasticity was adopted for the battery structure with effective modulus and Poisson ratio based on experimental parameters. The originally flat layers were laterally compressed into buckles structure with periodic boundary conditions at two lateral ends. The total length and thickness for simulations on unprotected structure are 1.0 cm and 142 µm, respectively and for tape-protected are 1.0 cm and 270 µm, respectively. The thicknesses of different layers are as follows. Tape: 32 µm, Cu foil: 18 µm, Al foil: 24 µm, Separator: 25 µm. A pressure of 1 atm is applied to both sides of layers to simulate the vacuum conditions inside the aluminized pouch bag.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ensm.2018.11.019.

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