

Ultrathin, Lightweight, and Wearable Li-O₂ Battery with High Robustness and Gravimetric/Volumetric Energy Density

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Flexible electronics have attracted worldwide attention due to their advantages such as deformability, lightweight, and portability, which would revolutionize many related industries including consumer electronics, automotive, and aerospace.^[1–7] To fulfill the alluring "flexible electronics" society, it is urgent to develop well-matched flexible, stretchable, and rechargeable energy storage devices. In response, several inspirational prototypes have been successfully developed, such as organic solar cells,^[8–11] lithium-ion batteries,^[12–16] and electrochemical supercapacitors.^[17–21] Nevertheless, the low theoretical energy density of these devices has restricted their applications in the next-generation flexible devices.

Fortunately, the rechargeable aprotic lithium-oxygen (Li-O₂) battery has emerged as a very promising candidate due to its ultrahigh theoretical energy density.^[22–33] Although four flexible Li-O₂ battery prototypes have been developed very recently,^[34–37] to achieve good wearability and robustness, especially in conjunction with high energy density, remains a daunting challenge due to the absence of effective components and favorable cell configurations: (1) lithium metal anode is problematic – the cracks generated after repetitive folding/bending would inevitably result in poor robustness and even short circuit and/or premature battery death; (2) assembly of a flexible Li-O₂ battery still heavily

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rely on soft packaging fixation (e.g., shrinkable tube and aluminum soft packaging materials) and thick air diffusion layer (which serves as an important component for cell assembly, such as steel mesh, nickel foam, etc.), which greatly decreases the energy density of the entire practical battery (currently, most reported Li-O₂ battery energy densities are based only on the weight of the cathode or even only on the weight of the catalyst); (3) the available soft-package and cable-type batteries are still "stack-type" Li-O₂ batteries, limiting their application in practical wearable electronic devices. Besides unsatisfying mechanical properties and low energy density, the current Li-O₂ batteries are still facing poor electrochemical performances, including low energy efficiency and poor cycling stability, due to the absence of highly efficient air cathode. Therefore, development of novel battery components and a sophisticated battery structure design to build an ultrathin, lightweight, and wearable Li-O₂ battery with high robustness and energy density is highly desirable, while still very challenging.

Herein, as a proof-of-concept experiment, we propose and demonstrate a "break up the whole into parts" strategy to build a novel segmented Li-O_2 battery, wherein the overall electrode consisting of an array of electrode disks and, especially, no air diffusion layer is employed. Unexpectedly, thanks to this unique configuration, this novel Li-O_2 prototype is ultrathin, lightweight, wearable, robust (no degradation even after 10 000 folding/stretching cycles), and holds superior electrochemical performances, including low overpotential, good rate capability, excellent cycle stability, and, especially, high gravimetric/volumetric energy density. This new strategy would promote widespread application of Li-O_2 batteries, leading to highly flexible energy storage devices in the future.

Figure 1 shows the novel design of the ultrathin, lightweight, and wearable Li-O₂ battery that consists of arrays of small-scale carbon cathode disks and lithium anode disks interconnected by carbon ropes and copper wires, respectively. Significantly, this strategy for the ultrathin, lightweight, and wearable Li-O₂ battery holds many advantages: (1) the anodes (composed of small lithium disks) were well confined in holes of the orderly punched polyethylene terephthalate (PET) film, serving as a skeleton of the battery (Figure S1, Supporting Information). Thanks to this unique structure, the whole segmented lithium metal anodes were almost kept unchanged even after folded in half and then stretched up

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Figure 1. a) Schematic illustration of the ultrathin, lightweight, and wearable Li-O₂ battery, in a state of bending. b) Exploded view layout of the layers in the battery structure. Photographs of the fabricated wearable Li-O₂ battery, c) woven into a glove and powered a commercial green light-emitting diode, d) held by a branch of the asparagus fern, e) tested by a screw micrometer.

to 1000 cycles. In sharp contrast, the whole piece of lithium metal sheets cracked after only 100 folding/stretching cycles (Figures S2 and S3, Supporting Information); (2) thin and lightweight PET film and polypropylene tape are employed as the skeleton or the ultrathin packing layers, respectively, endowing the obtained Li-O₂ battery with superior flexibility and ultralow density; (3) the novel cell configuration, free of air diffusion layer (nickel foam), further reduces the thickness and weight of the battery and thus significantly improve the practical energy density of Li-O₂ battery (based on the weight of whole battery). Thanks to all of these core features, the thus fabricated novel Li-O₂ battery is wearable, lightweight, and ultrathin (Figure 1c–e).

To enhance the electrochemical performance of the ultrathin, lightweight, and wearable Li-O2 battery, we first introduced ruthenium dioxide confined inside the channels of carbon nanotubes (CNTs) (RuO2@CNTs) for the air cathode. A pristine CNT cathode was also investigated as a control experiment. As shown in Figure 2a, pristine CNTs were opened up and shortened by treating in concentrated nitric acid, which were then mixed with ruthenium (III) chloride hydrate/diethylamine solution followed by solvothermal treatment. The high-resolution transmission electron microscopy (TEM) image shows the integration of ruthenium oxide (RuO₂) nanoparticles and the CNTs' channels (Figure 2b and Figure S4 (Supporting Information)), which might strengthen the surface electron density of CNTs and also avoids agglomeration/detachment of RuO2 nanoparticles during battery operation, leading to enhanced electrochemical performance of the fabricated wearable Li-O₂ battery. The powder X-ray diffraction (XRD) analysis of the RuO₂@CNT in Figure 2c reveals that only a very weak and broad diffraction peak of RuO₂ between 30° and 40° was observed, which might be due to the small size of RuO2 nanoparticles and/or shielding



effect of the CNTs. We then performed X-ray photoelectron spectroscopy (XPS) measurements to further confirm the presence of RuO₂ in the composite. In comparison to the XPS spectra of the CNTs in Figure S5 (Supporting Information), the two peaks centered at 281.0 (Ru_{3d5/2}) and 282.1 (Ru_{3d1/2}) eV can be attributed to Ru (IV) (Figure 2d), which is consistent with the XRD results. The specific surface area of the as-prepared RuO₂@CNTs is as high as 259 m² g⁻¹, which could provide sufficient catalytic sites for the necessarily required oxygen reduction/evolution reactions (ORR/OER) during discharging and charging processes of Li-O2 battery, respectively (Figure 2e). Furthermore, the pore-size distribution results show both micropores and mesopores exist in the RuO₂@CNTs, which is beneficial for the transportation of lithium ions and oxygen as well as storage of discharge product (Li_2O_2) , to improve the electrochemical performance of Li-O2 batteries (vide infra).

To investigate the flexibility, the obtained ultrathin, lightweight, and wearable Li-O₂ battery are intentionally deformed into different shapes with various stresses and strains, including planar, twisted, folded, and even curled. The red light-emitting diode remains constantly powered under all the testing conditions (Figure 3a), showing the superior flexibility and stability of the fabricated wearable Li-O₂ battery. To confirm whether the electrochemical performance of ultrathin, lightweight, and wearable Li-O2 battery degrades subjected to repetitive external forces, as shown in the inset of Figure 3b, they are first folded in half and then stretched to planar states, repeatedly for thousands of times and followed discharged/charged. Surprisingly, the discharge-charge curves of the wearable Li-O₂ battery keep almost unchanged even after 10 000 cycles of folding/stretching, showing excellent mechanical stability.

It should be noted that both the gravimetric energy density and the volumetric energy density are two important aspects for the practical application of flexible Li-O2 battery. Surprisingly, the weight of the ultrathin and wearable Li-O₂ battery is only 0.63698 g, which is more than four times lighter than that of conventional Li- O_2 coin battery (2.63444 g, Table S1, Supporting Information). As shown in Figure 3c, benefitting from the lightweight and high electrocatalytic activity of the RuO₂@CNTs, the gravimetric energy density of wearable Li-O₂ battery (even not optimized) reaches a very high value (up to 294.68 Wh kg⁻¹, calculated based on the weight of the whole device, not based only on the weight of the cathode or even only on the weight of the catalyst for most reported Li-O₂ batteries' energy densities) at a discharge current density of 100 mA g⁻¹, which far more exceed than those of conventional Li-O₂ coin battery (4.92 Wh kg⁻¹), cable-type flexible Li-O₂ battery (27.70 Wh kg⁻¹), and soft package flexible Li-O₂ battery (72.15 Wh kg⁻¹).^[34,37] Furthermore, due to the ultrathin

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Figure 2. a) Scheme for integration of RuO₂ nanoparticles with CNTs. b) The TEM image of the as-prepared integrated RuO₂ nanoparticles with CNTs, and its c) XRD patterns, d) C1s and Ru3d XPS spectra, e) nitrogen adsorption–desorption isotherms and pore-size distribution (inset).

feature (only 0.474 mm) and the unique battery configuration, the volumetric energy density of the fabricated wearable Li-O₂ battery reaches up to 274.06 Wh L⁻¹, which is far more higher than those of conventional Li-O₂ coin battery (13.97 Wh L⁻¹), cable-type flexible Li-O₂ battery (29.08 Wh L⁻¹), and soft package flexible Li-O₂ battery (32.18 Wh L⁻¹). As the research of flexible Li-O₂ batteries is still at its infant stage, there is plenty of room for further improvement of energy density of this novel kind of Li-O₂ battery.

From the viewpoint of practical application to make this novel battery more powerful, a foldable Li-O₂ battery pack,

consisting of several single ultrathin, lightweight, and wearable Li-O₂ batteries and a scrolled Li-O₂ battery, is first fabricated (Figure S9, Supporting Information). Obviously, the thus fabricated ultrathin, lightweight, and wearable Li-O₂ battery might be applied to next generation flexible pad/ mobile phones/e-readers due to its high specific energy density and flexibility.

Encouraged by the above obtained superior flexibility and mechanical stability, we then further studied the electrochemical performances of the obtained ultrathin, lightweight, robust, and wearable Li-O₂ battery. As shown in **Figure 4**a,



Figure 3. a) An ultrathin, lightweight, and wearable Li-O_2 battery powered a commercial red light-emitting diode at various bended and twisted conditions. b) Charge–discharge curves after bended thousands of times, inset is the battery at planar and folded in half states. c) The comparison of gravimetric energy density (left axis) and volumetric energy density (right axis) with four kinds of Li-O_2 batteries at a current density of 100 mA g⁻¹, insets are the photographs of the assembled Li-O_2 batteries.

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Figure 4. Electrochemical performances of the ultrathin, lightweight, and wearable Li-O_2 battery: a) first discharge curves at a current density of 200 mA g⁻¹ with a limited specific capacity of 1000 mA h g⁻¹, b) rate capacity curves at different current densities, c) first discharge curves at a current density of 200 mA g⁻¹, d) cycling performance at a current density of 200 mA g⁻¹ with a limited specific capacity of 500 mA h g⁻¹.

the overpotential is found to be only 0.86 V, which is much lower than that with pristine CNTs (1.46 V), indicating that the RuO₂@CNT cathode exhibits superior ORR/OER catalytic activities over CNTs, even for ultrathin, lightweight, robust, and wearable Li-O₂ battery, which is further supported by the cyclic voltammetry, wherein the first cycle curve reveals that the RuO₂@CNT cathode exhibits a higher current density compared with the CNT cathode (Figure S10, Supporting Information). The discharge rate performance investigations show that the discharge voltage plateau of

the RuO₂@CNT cathode is higher than that of CNTs at a wide variety of current densities (Figure 4b). Interestingly, the discharge capacity of the Li-O₂ battery can be significantly improved with the help of RuO₂@CNT cathode (Figure 4c, 11 400 mA h g⁻¹), compared to that of CNT cathode (3880 mA h g⁻¹) at a current density of 200 mA g⁻¹. Obviously, the background discharge capacity of the Ar-filled wearable batteries is negligible (Figure S11, Supporting Information), demonstrating that the above obtained discharge capacities of the Li-O2 cells are derived from ORR rather than intercalation reactions with RuO2@CNTs or CNTs.^[35,38] Furthermore, cycling performances of the ultrathin, lightweight, robust, and wearable Li-O₂ battery were measured, and the results are shown in Figure 4d. It was observed that the battery with RuO₂@CNT cathode can discharge/ charge for 288 cycles with a discharge terminal voltage >2.0 V, which is ca. 8.47 times

longer than the cell with the pristine CNTs (34 cycles). In addition, at the high current density of 500 mA g⁻¹, the charge voltage of the battery with RuO₂@CNT cathode is significantly lower than that with CNT cathode by ca. 560 mV (Figure S12, Supporting Information). As shown in Figure S13 (Supporting Information), with a discharge terminal voltage >2.0 V, the charge terminal voltage of RuO₂@CNT cathode is 4.30 V even after 75 cycles, which is still lower than that with the CNT cathode (4.51 V, 10 cycles). The low charge potential alleviated oxidative decomposition of the carbon-based



Figure 5. a-c) SEM images of the pristine, first discharged, and first recharged cathode of the ultrathin, lightweight, robust, and wearable Li-O₂ battery with high gravimetric/volumetric energy density. d) The XRD patterns of cathode with different discharge/charge states.

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ADVANCED SCIENCE NEWS www.advancedsciencenews.com electrode and possible decomposition of the electrolytes occurs in the presence of Li₂O₂, thus reducing the generation of byproducts.^[39-42] All of these improvements show that the RuO₂@CNT cathode exhibits superior ORR/OER activity, which may be attributed to the beneficial supervisitic effect

RuO₂@CNT cathode exhibits superior ORR/OER activity, which may be attributed to the beneficial synergistic effect of the high catalytic activity of RuO_2 toward both ORR and OER, the confinement of RuO_2 inside CNTs strengthening the surface electron density on CNT surfaces and avoiding agglomeration and detachment of RuO_2 nanoparticles. And the porous network structure of the RuO_2 @CNT cathode ensures accommodation of a large amount of discharge products and facilitates rapid mass (electrons, lithium ions, and oxygen) transportation. All these results show that, our obtained ultrathin, lightweight, robust, and wearable Li-O₂ battery also holds superior electrochemical performance, which is prerequisite for power devices for next-generation more powerful flexible electronics.

To further demonstrate the rechargeability of Li₂O₂ formation and decomposition within the ultrathin, lightweight, robust, and wearable Li-O₂ battery with a RuO₂@ CNT cathode, we tracked along the discharge and charge progresses by scanning electron microscope (SEM). As shown in Figure 5a, the pristine cathode composed of randomly dispersed RuO₂@CNTs holds a favorable porous structure. After the first discharge, the discharge product with toroidal structure appeared (Figure 5b). Interestingly, the generated discharge product is then almost fully decomposed during followed recharge progress (Figure 5c). The rechargeability is further supported by the XRD results, wherein the diffraction peaks centered at 32.9°, 35.0°, and 58.7° are observed after the first discharge, confirming that Li₂O₂ is the only crystalline product, and all of them disappeared after subsequent recharge progress (Figure 5d). The above results were also verified by Fourier transform infrared spectroscopy (Figure S14, Supporting Information). All these results demonstrate a favorable rechargeability of the novel ultrathin, lightweight, robust, and wearable Li-O₂ battery.

In summary, to endow Li-O2 battery with excellent flexibility, electrochemical performances, and robustness, and thus the great promise to power the next-generation versatile flexible electronics, as a proof-of-concept experiment, we have first proposed and demonstrated a facile, effective strategy inspired by great ideas from "break up the whole into parts" to fabricate an ultrathin, lightweight, and wearable Li-O2 battery. The superior flexibility and electrochemical stability of as-resulted wearable Li-O2 battery have been achieved even after 10 000 cycles of folding/stretching, benefiting from the special structure. Unexpectedly, the ultrathin, lightweight, robust, and wearable Li-O₂ battery also holds superior electrochemical performances, including low overpotential, high specific capacity, good rate capability, superior cycle stability, and, especially, high gravimetric/volumetric energy density $(294.68 \text{ Wh kg}^{-1}/274.06 \text{ Wh L}^{-1}; \text{ far more exceed the reported})$ flexible and even conventional Li-O₂ batteries), which are prerequisites for power devices for next-generation more powerful flexible electronics. The results reported here provide new designs of flexible Li-O2 battery, which would promote widespread application of Li-O2 battery for flexible

electronics and encourage further studies on flexible energy storage devices.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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