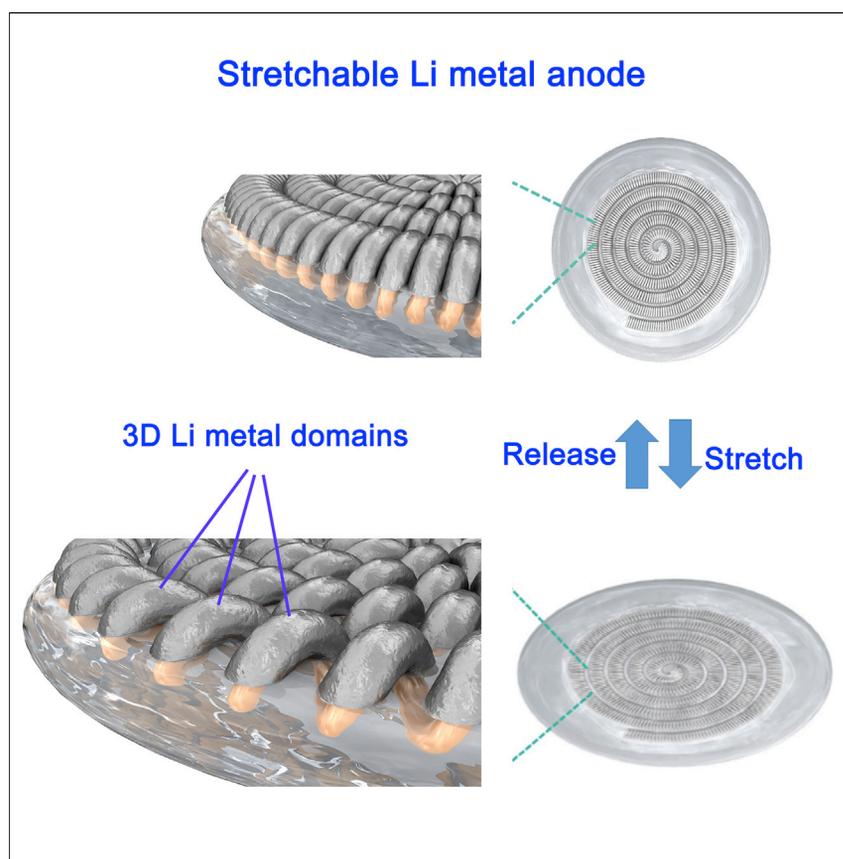


## Article

# Stretchable Lithium Metal Anode with Improved Mechanical and Electrochemical Cycling Stability



Lithium (Li) metal is the ideal anode material for stretchable batteries. However, Li metal is ductile with unstable electrochemical performance. We have fabricated a stretchable Li metal anode with stable mechanical and electrochemical performance. It consists of one-entity 3D-patterned Li metal microdomains connected by highly elastic polymer rubbers. This is a key step in the development of novel stretchable “lithium batteries” rather than traditional stretchable “lithium-ion batteries,” which could advance the development of soft electronics.

Kai Liu, Biao Kong, Wei Liu, ...,  
Dingchang Lin, Allen Pei, Yi Cui

yicui@stanford.edu

**HIGHLIGHTS**

We have made Li metal anode stretchable

It exhibits stable mechanical and electrochemical performance

It is a key step in the development of stretchable “lithium batteries”

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Article

# Stretchable Lithium Metal Anode with Improved Mechanical and Electrochemical Cycling Stability

Kai Liu,<sup>1</sup> Biao Kong,<sup>1</sup> Wei Liu,<sup>1</sup> Yongming Sun,<sup>1</sup> Min-Sang Song,<sup>2</sup> Jun Chen,<sup>1</sup> Yayuan Liu,<sup>1</sup> Dingchang Lin,<sup>1</sup> Allen Pei,<sup>1</sup> and Yi Cui<sup>1,3,4,\*</sup>

## SUMMARY

Stretchable batteries are key components of stretchable/flexible electronic devices. However, they typically exhibit low energy density due to the low lithium storage capability. Lithium (Li) metal is the ideal anode material, but it is ductile and the unstable electrochemical performance further hinders its practical applications. Herein, for the first time, a stretchable Li metal anode with stable mechanical and electrochemical performance is fabricated. It consists of one-entirety 3D-patterned Li metal microdomains connected by highly elastic polymer rubbers. Upon stretching, the rubber absorbs the mechanical energy while the electroactive Li domains are not mechanically strained. Moreover, the whole electrode is fabricated by simply winding one single Cu wire, which is facile and cost-effective. The stretchable Li metal anode is a key step in the development of novel stretchable “lithium batteries” rather than traditional stretchable “lithium-ion batteries” to boost the energy density of stretchable energy-storage devices.

## INTRODUCTION

Stretchable electronic devices, such as wearable electronics, smart clothes, electronic skin, displays, bendable/stretchable smart phones, and implantable medical devices, have attracted much attention in recent years.<sup>1–5</sup> To power these devices, stretchable energy-storage systems,<sup>6–9</sup> e.g., lithium-ion batteries (LIBs),<sup>10–12</sup> with comparable stretchability that can adapt to the dynamic motions of the electronic devices while maintaining good electrochemical performance, are highly desirable. However, current LIBs are intrinsically not stretchable because of the low elastic limits of its components, such as current collectors and cathode/anode materials. In recent years, great efforts have been devoted to make LIBs stretchable, either by enabling “device-scale”<sup>13–16</sup> or “electrode-scale”<sup>17–20</sup> stretchability. However, the energy density of these batteries has remained low. The former method is limited by the need for excessive electrochemically inactive components such as stretchable interconnects, while the latter strategy is hindered by extremely low active material mass loadings required to prevent materials detachment from the current collector upon stretching/releasing cycles. Furthermore, the active electrode materials used so far have low Li storage capabilities. For example,  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  (LTO) is the most popularly employed anode material for stretchable LIBs because of its stable electrochemical cycling performance.<sup>17,18,21</sup> However, it exhibits a low theoretical specific capacity ( $175 \text{ mAh g}^{-1}$ ) and high anode potential ( $\approx 1.5 \text{ V}$  versus  $\text{Li}/\text{Li}^+$ ), which greatly lower the battery energy density. Thus, new materials with high Li storage capacity, low working potential, and stable electrochemical performance are

## Context & Scale

Lithium (Li) metal is the ideal anode material for stretchable batteries. However, Li metal is ductile and cannot withstand mechanical deformation, and its unstable electrochemical performance further hinders its practical applications. Herein, for the first time, a stretchable Li metal anode with stable mechanical and electrochemical performance is fabricated. It consists of one-entirety 3D-patterned Li metal microdomains connected by highly elastic polymer rubbers. Moreover, the fabrication process is facile and cost-effective. The stretchable Li metal anode is a key step in the development of novel stretchable “lithium batteries” rather than traditional stretchable “lithium-ion batteries,” which could advance the further application of soft electronics.

required for the further development of next-generation stretchable/flexible energy-storage materials.

Different from traditional LIBs, “lithium batteries” directly use pure lithium metal as the anode material and can thus further boost the energy densities of the energy-storage devices. Li metal is the most ideal anode material due to its high theoretical capacity ( $3,860 \text{ mAh g}^{-1}$ , 10 times that of graphite) as well as its low electrochemical potential ( $-3.040 \text{ V}$  versus standard hydrogen electrode), both of which improve battery energy density.<sup>22–27</sup> Despite its appealing properties, Li metal is not able to accommodate significant elastic deformation. At above 25% irreversible plastic elongation the necking of Li metal occurs, followed by ductile fracture.<sup>28</sup> Moreover, Li metal anodes generally suffer from severe performance decay during repeated electrochemical cycles, further hindering the application of Li metal anodes in stretchable/flexible energy-storage materials.

Here, for the first time, we have proposed and fabricated a novel Li metal anode that is mechanically stretchable with improved electrochemical cycling performance. The stretchable Li metal anode is composed of three-dimensional (3D) patterned Li metal microdomains connected by highly elastic polymer rubber, poly(styrene-ethylene-butylene-styrene) (SEBS), which is electrochemically stable in the highly reductive environment of Li metal (Figure S1). The stretchable electrode structure consists of a doubly coiled Cu wire partially embedded within a rubber substrate; the exposed Cu coil surface serves as the electroactive current collector for Li metal deposition, while the rubber-encased Cu sections enable the metal coil to stretch uniformly and cohesively in parallel with the rubber. There are five aspects that make this design novel and efficient. (1) By winding a linear Cu coil into an Archimedean spiral pattern, the standard 1D coil is transformed into a novel hierarchical, 2D isotropically stretchable coil, which is more desirable for practical applications. (2) As Li is electro-deposited on the Cu coil, the Li metal is segmented into small microdomains connected by highly elastic SEBS rubber. Thus, upon stretching, only the rubber absorbs the mechanical energy; the electroactive Li domains remain attached to the freely moving Cu segments and are not affected. The integration of the Cu coil and rubber greatly alleviates the drastic modulus differences between the polymer rubber and metal materials and allows the electrochemical performance to be nearly unaffected by the mechanical deformation. (3) All the Li metal microdomains are one contiguous entity joined together by the 2D hierarchical Cu coil; the conductivity of the electrode is thus unchanged no matter how it is stretched, which is crucial for stable electrochemical performance. (4) The entire 2D Cu coil consists of only a single Cu wire, which can be readily fabricated through facile winding. This “one-body” design allows for very stable electrochemical performance under repeated stretching/releasing deformations. Moreover, the relatively simple design is more cost-effective compared with previous efforts that greatly rely on expensive and time-consuming micromanufacturing technologies, such as lithographic processes for making stretchable domains. (5) The 3D patterned Li metal anode has increased surface area compared with conventional planar electrodes, which greatly reduces the local current density and homogenizes the Li-ion flux. Thus, this new configuration of Li metal anode has not only made the electrode highly stretchable, but has also significantly improved the cycling stability of the Li anode. This is in great contrast to previous reports in which there has always been a trade-off between the electrochemical performance and the mechanical stretchability of the battery. With this design, the stretchable Li metal anode exhibits great mechanical and electrochemical cycling stability: the Li capacity loss is only 0.1% per mechanical

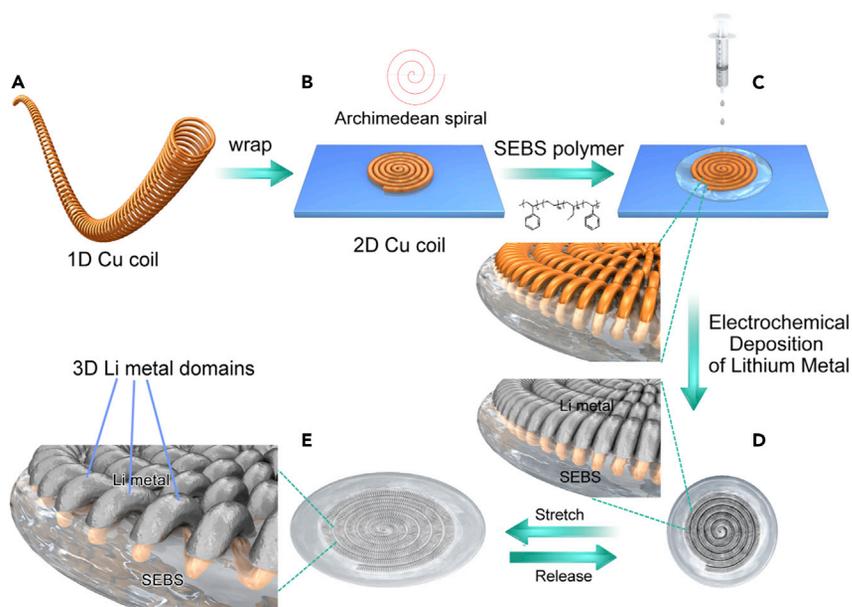
<sup>1</sup>Department of Materials Science and Engineering, Stanford University, Stanford, CA 94305, USA

<sup>2</sup>Energy Material Lab Material Research Center, Samsung Advanced Institute of Technology, Samsung Electronics, 130 Samsung-ro, Yeongtong-gu, Suwon-si, Gyeonggi-do 16678, Republic of Korea

<sup>3</sup>Stanford Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, USA

<sup>4</sup>Lead Contact

\*Correspondence: [yicui@stanford.edu](mailto:yicui@stanford.edu)  
<https://doi.org/10.1016/j.joule.2018.06.003>



**Figure 1. Schematic Showing the Fabrication Process of the Stretchable Electrode**

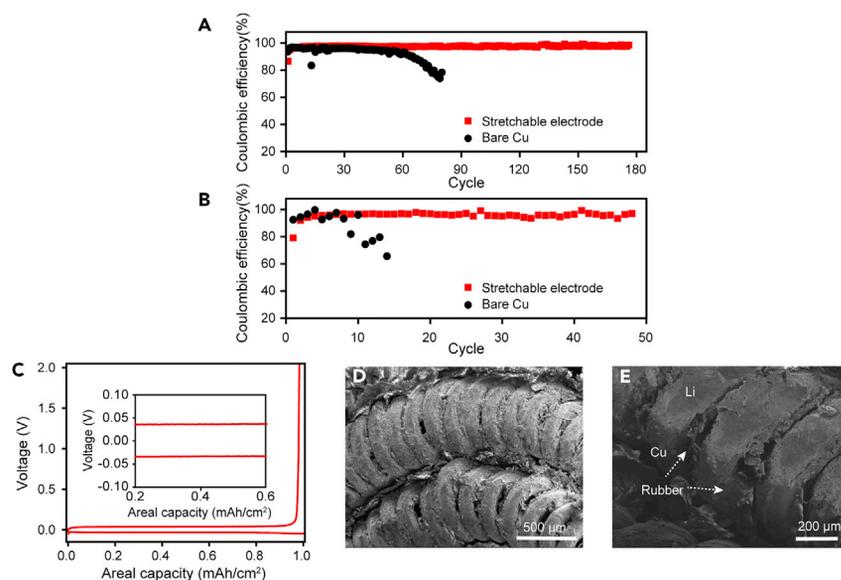
(A) Cu wire with a diameter of 150  $\mu\text{m}$  was wrapped into a coil with an inner diameter of  $\sim 500 \mu\text{m}$ . (B) The Cu coil was further wrapped in the pattern of an Archimedean spiral. (C) A solution of SEBS rubber was then cast on it. After drying the solvent out of the SEBS, the rubber on one side was polished away, exposing the Cu coil embedded in the SEBS rubber. (D) Li metal can be electrochemically deposited onto the polished side of the Cu coil, resulting in a stretchable Li metal anode. (E) Upon being stretched, the 2D Cu coil exhibits excellent extensibility due to the hierarchical spiral structures. As the Li metal is segmented into small domains separated by highly elastic SEBS rubber, the rubber absorbs the mechanical energy while the electroactive Li domains are not affected. Once the external stress is released, the extended SEBS rubber exerts a contraction force onto the Li metal, allowing the whole electrode to recover its original shape and dimension.

stretching/releasing cycle at a large strain of 60%, and the average Coulombic efficiency is  $\sim 97.5\%$  for the first 176 cycles at  $1 \text{ mA cm}^{-2}$  in ether-based electrolytes.

## RESULTS AND DISCUSSION

The fabrication method of the stretchable Li metal anode is shown in Figure 1. Cu wire with a diameter of 150  $\mu\text{m}$  is wrapped into a coil with an inner diameter of  $\sim 500 \mu\text{m}$ . This 1D Cu coil is further self-wrapped in the pattern of an Archimedean spiral, creating the 2D Cu coil, which is then attached to tape to immobilize its overall shape. A solution of SEBS rubber is then cast onto the coil. After drying the solvent out of the SEBS, the tape is peeled off. The SEBS rubber facing the tape is then polished away, exposing part of the Cu coil. The exposed part of the Cu coil is thus segmented into small 3D microdomains with SEBS rubber filling the gaps. Li metal can be electrochemically deposited onto these 3D microdomains, resulting in a stretchable Li metal anode. Upon being stretched, the 2D Cu coil exhibits excellent extensibility due to the hierarchical spiral structures. Once the external stress is released, the extended SEBS rubber exerts a contraction force onto the small microdomains, allowing the whole electrode to recover its original shape and dimension.

We first tested the electrochemical performance of the stretchable electrode in the unstretched state. Coulombic efficiency is an important parameter for long cycle life and is defined as the ratio of the amount of Li stripped from the working electrode



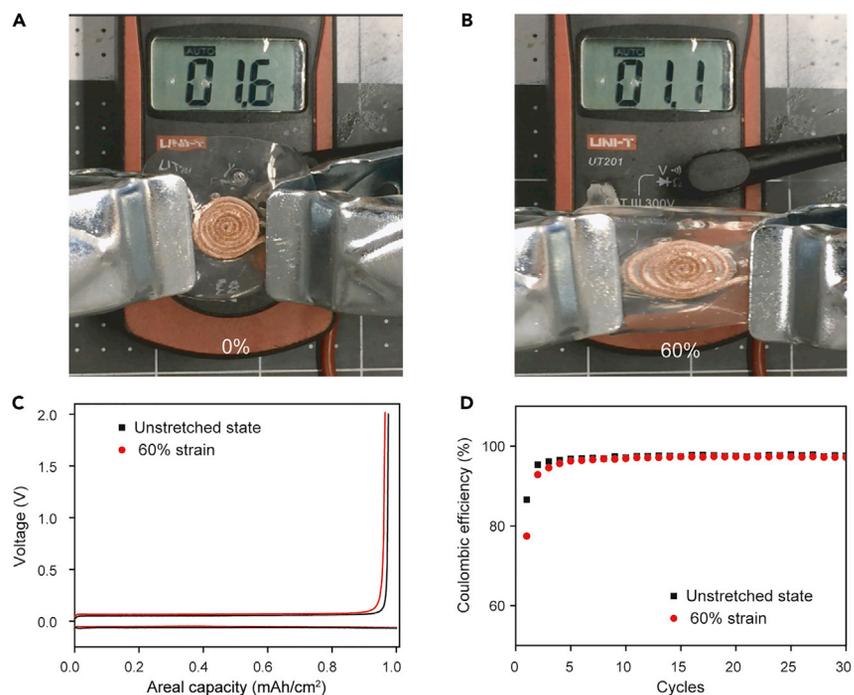
**Figure 2. Electrochemical Performance and Scanning EM Images of the Stretchable Li Metal Anode**

(A and B) Electrochemical cycling performance of the stretchable Li electrode (red squares) and flat Cu (black circles) at different current rates. The electrolyte is 1 M LiTFSI in 1,3-dioxolane and 1,2-dimethoxyethane (volume ratio 1:1) with 1 wt% LiNO<sub>3</sub>. (A) 1 mA cm<sup>-2</sup>, (B) 2 mA cm<sup>-2</sup>. The amount of Li deposited in each cycle is 1 mAh cm<sup>-2</sup>.

(C) Voltage profiles of the Li deposition/dissolution process with Li metal as the reference electrode at 1 mA cm<sup>-2</sup>.

(D and E) Scanning EM images of the stretchable Li metal anode. (D) Lower magnification; scale bar, 500 μm. (E) Higher magnification; scale bar, 200 μm.

versus the amount that is plated during each cycle. Half cells using Li metal as both the counter and reference electrode were fabricated. To standardize the electrochemical performance, we used ~60 μL of electrolyte in each coin cell test. In the half-cell configuration, Li was electrochemically deposited (at 1 mA cm<sup>-2</sup> to a capacity of 1 mAh cm<sup>-2</sup>) from the Li metal counter electrode onto the stretchable working electrode and then stripped away. Here, the Coulombic efficiency reflects the loss on the working electrode because the Li metal counter electrode has excess Li. In cycle life testing, these batteries fail due to the depletion of electrolyte as a result of reaction with the deposited Li metal. The electrochemical cycling performance of the stretchable electrode is stable, as indicated by Figures 2A and 2B. The average Coulombic efficiency is ~97.5% for the first 176 cycles at 1 mA cm<sup>-2</sup> in ether-based electrolyte (1 M lithium bis(trifluoromethanesulfonyl)imide [LiTFSI] in 1,3-dioxolane and 1,2-dimethoxyethane [volume ratio 1:1] with 1 wt% lithium nitrate [LiNO<sub>3</sub>]). After 176 cycles, the electrochemical performance started to decay. Even when tested at a high current density of 2 mA cm<sup>-2</sup>, the average Coulombic efficiency of the stretchable Li metal electrode is ~96% for the first 48 cycles. The voltage profiles upon discharging/charging show the typical electrochemical curves for Li deposition/stripping (Figure 2C). As indicated by the scanning electron microscopy (EM) images shown in Figures 2D and 2E, Li metal is firmly deposited on the self-wrapped Cu spiral. SEBS rubber fills in the gaps among the neighboring pitches of the Cu spirals (Figure S2), which returns the coil back to its original shape after stretching while the Li metal deposited on the Cu wire is not affected upon stretching. As such, the electrochemical performance of the stretchable Li metal anode is significantly improved compared with that of the flat Cu foil electrode. As shown in Figure 2A, batteries using Cu foil show a gradual decrease in Coulombic efficiency, which eventually drops



**Figure 3. Stretchability and Electrochemical Performance of the Novel Li Metal Anode at Its Stretched State**

(A and B) Photographs showing the stretchability of the stretchable electrode. (A) Original state; (B) 60% strain.

(C) Voltage profiles of the stretchable Li metal anode at unstretched state (black) and stretched state (60% strain, red).

(D) Cycling performance of the stretchable Li metal anode at unstretched state (black squares) and stretched state (60% strain, red circles).

to less than 95% after 45 cycles at  $1 \text{ mA cm}^{-2}$ . At a high current density of  $2 \text{ mA cm}^{-2}$ , the Coulombic efficiency on the planar Cu quickly decays to less than 90% after only 8 cycles, and the average Coulombic efficiency is  $\sim 88\%$  for the first 14 cycles. In addition, the stretchable electrode also showed improved electrochemical performance in carbonate-based electrolyte (Figure S3). This improved electrochemical performance of the stretchable Li metal anode should be attributed to the increased surface area of the Cu coil as compared with that of the flat Cu foil, which decreases the local current density (see Note S1). As shown by the scanning EM image in Figure S4A, the morphology of the Li metal on the stretchable anode is smooth without dendrites, which should be the reason for the improved electrochemical cycling stability. In contrast, Li dendrites were clearly observed on the conventional bare Cu electrodes (Figure S4B). Thus, the above results demonstrate that the stretchable Li electrodes show good electrochemical performance, which is a prerequisite for its further application as the anode in LIBs.

Moreover, it is equally important that the stretchable electrode can still function well in the stretched state. As shown in Figures 3A and 3B, the stretchable electrode shows good stretchability and stable electrical resistance upon being stretched. The electrode can be stretched by up to 60% and can fully recover its original state after the mechanical stress is released. Moreover, the original resistance across the electrode is measured to be  $1.6 \Omega$ . Upon gradually being uniaxially stretched to a strain of  $\sim 60\%$ , its electronic resistance is stable during the stretching process. The low electronic resistance originates from the one-body integration of the Cu

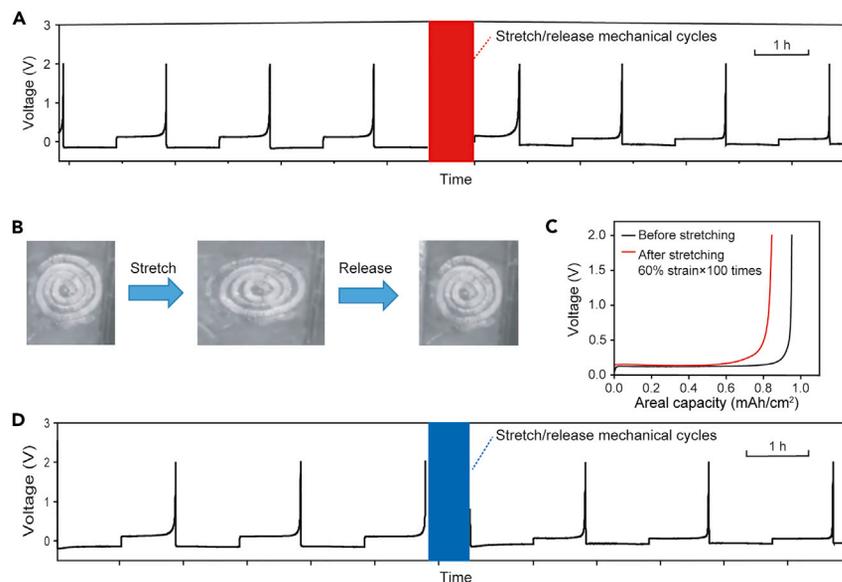
coil inside the rubber, meaning that the Cu coil remains as one entity no matter how it is stretched. Moreover, even after 100 cycles of repeated mechanical stretching/releasing, the electrode stays intact and the electronic resistance remains stable, indicating that the stretchable electrode is robust and durable in terms of mechanical deformation. Good stretchability and low and stable resistance are desirable for the stable electrochemical performance of stretchable electrodes during mechanical stretching/releasing cycles. Further tests of the electrochemical performance of the coiled Cu electrodes before and after mechanical cycles indicate their good durability (see [Figure S5](#)).

We then tested the electrochemical performance of the stretchable electrode at its stretched state (strain:  $\sim 60\%$ ). The stretched electrode was coupled with Li metal for electrochemical cycling. The discharge/charge voltage profiles are shown in [Figure 3C](#). For a direct comparison, the voltage profile of electrochemical cycling for the electrode in the unstretched state is also shown in [Figure 3C](#). The stretched electrode voltage profiles are similar to those of the unstretched state, both of which show typical Li deposition/stripping voltage profiles. In addition, the cycling performance of the stretchable electrode at 0% strain and 60% uniaxial strain is also shown in [Figure 4D](#), showing no obvious differences. The stable electrochemical performance of the electrode during stretching is attributed to the unchanged electrode structure and electrical resistance upon being stretched.

The stretchable electrode was also subjected to dynamic mechanical stretching/releasing cycles at the charged and discharged states. As shown in [Figure 4A](#), Li metal with a capacity of  $1 \text{ mAh cm}^{-2}$  was deposited onto the stretchable electrode at a current density of  $1 \text{ mA cm}^{-2}$  (charged state), after which all of the Li was completely stripped away to a voltage of 2 V (discharged state). After the battery electrochemical performance was stabilized after a few repeated Li deposition/stripping cycles, Li with a capacity of  $1 \text{ mAh cm}^{-2}$  was deposited onto the electrode, creating the charged state of the stretchable Li metal anode. The stretchable Li metal anode was then subjected to 100 mechanical stretching/releasing cycles to a strain of 60% each time ([Figure 4B](#)). Afterward, the electrode was discharged up to 2 V to check the capacity retention after the mechanical cycling. As can be seen in [Figures 4A and 4C](#), the Li can still be stripped away smoothly and the overpotential for stripping Li remains the same as that before the mechanical stretching/releasing cycles, indicating that the resistance of the electrode is not affected by the mechanical cycling. The Li capacity retention after these mechanical cycles reaches as high as 90%. Afterward, the electrode quickly recovers to the same stable electrochemical cycling as before stretching/releasing ([Figures 4A and S6](#)), indicating the good stretchability and stability of the stretchable Li metal anode. Additionally, the electrode was also subjected to mechanical stretching/releasing cycles at the discharged state. Li metal was fully stripped away from the electrode and 100 mechanical cycles were conducted with a strain of 60%, after which discharging-charging was continued. As can be seen from [Figure 4D](#), the Coulombic efficiency decreased by  $\sim 11\%$  compared with that of the unstretched electrode, which, however, quickly recovered within one cycle to the same value as that before the mechanical stretching/releasing cycles ([Figure S7](#)). Thus, the stretchable electrode shows good stretchability and stability at both the discharged and charged states, which are two representative states of anodes.

## Conclusion

For the first time a novel stretchable Li metal anode with stable electrochemical cycling performance has been designed and fabricated. The stretchable anode is



**Figure 4. Electrochemical Cycling Stability under Stretching/Releasing Mechanical Cycles**

(A) Voltage versus time profile for the stretchable Li metal anode half cell during charge/discharge cycling. A Li metal disc is used both as the reference electrode and counter electrode. The current density is  $1 \text{ mA cm}^{-2}$  and the capacity is  $1 \text{ mAh cm}^{-2}$ . The stretchable Li metal anode was subjected to mechanical stretching/releasing cycles (100 times at 60% strain) at its charged state, when Li metal with a capacity of  $1 \text{ mAh cm}^{-2}$  was deposited onto the electrode. Thereafter, the anode was discharged to 2 V and the electrochemical performance quickly recovered.

(B) Photographs of the stretching/releasing of the stretchable Li metal anode.

(C) Comparison of the voltage profiles of the stretchable Li metal anode before and after mechanical stretching/releasing cycles.

(D) Voltage versus time profile for the stretchable Li metal anode half cell during charging/discharging cycling. The stretchable Li metal anode was subjected to mechanical stretching/releasing cycles (100 times at 60% strain) at its discharged state, when Li metal was fully stripped away. Afterward, the anode was cycled again with a capacity of  $1 \text{ mAh cm}^{-2}$  and the electrochemical performance quickly recovered.

based on a novel hierarchical 2D Cu coil that can be stretched isotropically, segmenting the Li metal into small microdomains separated by highly elastic SEBS rubber, leading to the 3D patterned Li metal anode. Here, the essence of our structural design is that the electrode structure can accommodate and sustain large elongation over many cycles, but the actual local strains experienced by the Li metal are lower than its failure strain. This facile “one-body” integration of the 2D Cu coil and rubber greatly alleviates the drastic modulus differences between the polymer and metal materials and allows the electrochemical performance to be nearly unaffected by the mechanical deformation. This new configuration of Li metal anode has not only made the electrode highly stretchable, but has also significantly improved the cycling stability of the Li anode. This is in great contrast to previous reports in which there has always been a trade-off between the electrochemical performance and the mechanical stretchability of the battery. It is anticipated that the facile and cost-effective fabrication of the stretchable Li metal anode will boost the development of stretchable lithium batteries and increase the energy density of stretchable/flexible batteries.

## EXPERIMENTAL PROCEDURES

Experimental Procedures can be found in the [Supplemental Information](#).

## SUPPLEMENTAL INFORMATION

Supplemental Information includes Supplemental Experimental Procedures, seven figures, and one note and can be found with this article online at <https://doi.org/10.1016/j.joule.2018.06.003>.

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## AUTHOR CONTRIBUTIONS

K.L. and Y.C. conceived this work and wrote the manuscript. B.K. drew the schematic **Figure 1**. All authors (K.L., B.K., W.L., Y.S., M.-S.S., J.C., Y.L., D.L., A.P., Y.C.) contributed to the discussion, analysis of the results, and discussion on the manuscript.

## DECLARATION OF INTERESTS

K.L., M.-S.S., and Y.C. are inventors on a US Provisional Patent Application related to this work (No. 62/575,079, filed October 20, 2017).

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