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Thermodynamically and Physically Stable Dendrite-Free Li Interface with Layered Boron Nitride Separators

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Abstract

The development of promising separator candidates, offering both great physical rigidity and high thermal resistance, is still a global challenge to guarantee high-performance electrochemical cells. Freestanding hexagonal boron nitride (*h*-BN) separators are developed using the strategically combined synthetic steps with liquid and thermal-expansion exfoliation methods, which can simply fabricate the largely yielded *h*-BN nanosheets. The as-prepared freestanding *h*-BN nanosheet separator presents the better ionic conductivity than commercial polymer separators, and furtherly provides the excellent stability of Li-ion cells by suppressing the protruding dendritic Li growth during cycles on the electrodes as well as the high thermal and electrochemical stability of *h*-BN nanosheet separators even after aging process at high temperature 80~120 °C. Furthermore, the lithium titanate (LTO) batteries with the freestanding *h*-BN separators maintain an outstanding reversible coulombic efficiency of ~ 99 % after 600 cycles as well as high cycling retention, indicating the significant improvement on battery performance compared to the LTO batteries with commercial polymer separators. Thus, the freestanding *h*-BN separators may provide a new strategic way for the efficient storage of charge in next-generation rechargeable batteries and high-safety energy storage applications.

Keywords: hexagonal boron nitride, two-dimensional atomic crystal, layered separator, thermal stability, lithium metal anode

1 Introduction

2 Currently, to meet the ever-increasing demands of electrical energy delivery,
3 electrochemical lithium ion batteries (LIBs) are a primary class of energy storage and delivery
4 sources in portable electronic applications [1-4]. Particularly, in order to optimize and further
5 improve the overall energy-storing performance of LIBs, the rational selection and design of
6 each conventional battery component (such as electrodes, electrolyte, and separators) are very
7 essential [5-7]. In fact, many studies on LIBs have intensively focused on the improvement of
8 energy storage capacitance and cyclability by the optimization of electrode and electrolyte
9 materials [8-9]. However, there are more crucial factors which may limit the overall
10 performance of LIBs such as the growth of protruding Li dendrites during cycling and the
11 physical and chemical damage through internal and external heat environments [10-11]. Such
12 factors can induce the serious deformation of separator membranes and cause drastic
13 performance failure as well as severe problems on battery safety [12]. Commercial separators
14 are largely consisted of polymer-based compounds, which can be certainly damaged by the
15 aforementioned factors during cycling. Therefore, the development of novel separators with
16 high electrochemical, physical, and thermal stability is important not only for viable long-term
17 and high-performance LIB applications but also for the use of Li metal as the anode, possessing
18 a high theoretical specific capacity of 3860 mAh g^{-1} and a low redox potential (-3.04 V vs.
19 standard hydrogen electrode) [13-17]. Recently, to suppress Li dendrite growth, the deposition
20 of some nanomaterials (such as silicone nanofilaments, dopamine and mesoporous silica thin
21 films) on commercial separators has been reported and presented the high electrochemical
22 stability [15-16]. Moreover, the coated Prussian blue and reduced graphite oxides on
23 commercial separator also show the ultra-long-term stability of Li plating/stripping [17].
24 However, instead of focusing on new coating materials on commercial separators, the
25 development of freestanding materials candidates for separators may be a crucial way to solve
26 Li dendrite problems which are usually originated from polymer-based commercial separators.

Hexagonal boron nitride (h-BN), as a typical two-dimensional (2D) insulating material with a wide band gap (5-6 eV), has been considered to be a promising candidate for separator membranes in LIBs because of its outstanding ion permeability as well as superior thermal, chemical and physical stability [18-20]. h-BN films have recently revealed great success as stable coating materials on electrodes and polymer membranes, which can lead to the great strides in increasing the performance and stability of LIBs. For example, Yan et al. and Xie et al. reported that the 2D h-BN layers were directly synthesized on a Cu foil using a chemical vapor deposition (CVD) method [21-22]. The 2D h-BN layers can effectively suppress the growth of Li dendrites and hence, improve the cycling stability. Furthermore, Kim et al. and Luo et al. reported that the commercial polymer separators coated with the h-BN materials can improve the coulombic efficiency and ensure the reliability of Li metal anodes [23-24]. Nevertheless, the CVD-grown h-BN films are not practical to utilize as commercial and practical products due to its low reproducibility and limitation of large-scale fabrication. Indeed, the h-BN coated commercial polymer separators can be also damaged by any internal or external heat sources, resulting in the issues of dramatic performance degradation or safety failure. Therefore, it is highly desirable to fabricate freestanding h-BN separators, which are made of h-BN itself from new practical and scalable synthetic routes.

In this work, freestanding h-BN nanosheet (BNN) separators represent the outstanding thermal endurance under high-temperature conditions and possess the excellent physical rigidity against the dendritic Li growth after long cycling test as well as provide the favorable Li ion permeability, which is also theoretically calculated by a density function theory (DFT) model. The high quality and nanosized h-BN sheets were fabricated by employing both the simple liquid and thermal expansion exfoliation method, which might give the great scalability and processability. The freestanding BNN separators (BNN-Ss) can successfully suppress the formation of protruding Li dendrites and possess the high thermal stability even at the high thermal conditions at ~120 °C, where the commercial polymer separators cannot handle those

1 thermal conditions. In addition, the spinel lithium titanate (LTO) batteries with the freestanding
2 BNN-Ss maintain a reversible coulombic efficiency of $\sim 99\%$ after the 600 cycles. These
3 electrochemical and physical results imply the great potential of freestanding BNN-Ss as a key
4 separator component for the future battery technology in stability and cyclability against
5 internal and external surrounding environments.

6

Results and Discussion

The exfoliated *h*-BN nanosheets (BNNs) were prepared by using the combination of liquid exfoliation and thermal expansion techniques. The synthetic methods to yield the BNNs are schematically illustrated in **Fig. 1a** and **S1**. First, the bath and ultrasound probe sonification (liquid exfoliations) methods for *h*-BN bulk powders with a concentrated alkaline solution were carried out. These physical exfoliation methods can convert the bulk *h*-BN powders to the hydroxyl functionalized and small-sized thin *h*-BN layers. The hydrodynamic forces during the bath and ultrasound sonification can lead to the dramatic breakdown of the bulk BN powders into the thin BN layers. Simultaneously, the edge sites of the BN layers are simultaneously attacked by hydroxyl ions, and the hydroxyl ions are attached to the edge sites of thin *h*-BN as a functional group. Moreover, as shown in **Fig. S1**, during the gravimetric filtration process, the *h*-BN nanosheets can be stacked and laminated together with multiple layered structure. Moreover, during the lamination, the sheet-to-sheet van der Waals interactions become more effective. As shown in **Fig. S2**, X-ray photoelectron spectroscopy (XPS) spectra of both pristine BN powder and exfoliated BN have no difference. Especially, there are no split peaks and peak shift on the B1s and N1s of the both pristine BN powders and exfoliated BN layers, demonstrating the clear crystallinity of the exfoliated BN layers. Next, the secondary hydrogen (H₂)-assisted thermal expansion treatment was applied on the functionalized thin *h*-BN layers so as to further cleave the interlayers of the thin *h*-BN layers and to yield the nanosized BNNs. The finally obtained optimized BNNs were dispersed in an isopropyl alcohol (IPA) solution for the filtration process (**Fig. S3**). The freestanding BNN-Ss were further filtered by the vacuum filtration of the prepared exfoliated BNNs in the IPA solution. The vacuum filtration method enables the scalable fabrication of freestanding *h*-BN nanosheet-separators (BNN-Ss). Due to the fixed size of 2032-coin cells, the diameter and thickness of the freestanding BNN-Ss were carefully controlled by adjusting the amount and concentration of the BNN solution during

1 filtration. The thickness of BNNs was about 20 ~ 30 μm , which is close to that of the
2 commercial polymer separators.

3 It should be noted that the ion (H^+ and Li^+) permeability through the in- and out-plane of
4 thin layer *h*-BN at the level of mono- and few-layers has been already demonstrated by other
5 research groups [19,25,26]. The BNNs are successfully exfoliated into the mono- to few-layers
6 of *h*-BN. Therefore, when the BNNs are further applied to the separator application, their thin
7 in-plane structure can successfully provide the Li ion diffusion paths. **Fig. 1b** presents the
8 morphologies and height profiles of the exfoliated BNN samples examined by atomic force
9 microscopy (AFM). The thicknesses of representative BNNs were determined to be the mono-
10 and bilayers, which are about 0.48 nm, and 0.90 nm, respectively [27]. **Fig. S4a** presents the
11 Raman peak of bulk BN powders centered at near 1366 cm^{-1} (the $\text{E}_{2\text{g}}$ mode (G band), B-N
12 vertical vibrational mode within layers). Distinctly, the Raman peak of BNNs exhibits the
13 relatively slight blue shift up to 4 cm^{-1} , which is a clear signature of the thin thickness of BNNs
14 [28]. Note that the shift of the G band of *h*-BN is strongly correlated to its thickness: the thin
15 thickness of *h*-BN presents an upshifted G band, while the thicker *h*-BN shows the downshift
16 of the Raman peak position. **Fig. S4b** presents the Small-angle X-ray Scattering (SAXS) results
17 of BNNs. The slope of the SAXS scattering intensity profile at very low q values is found to be
18 2.14, indicating the perfect dispersion of BNNs before the filtration process [29-30]. High-angle
19 annular dark-field scanning transmission electron microscopy (HAADF-STEM) images of
20 BNNs were also performed as shown in **Fig. 1c**. The inset TEM image of **Fig. 1c** represents the
21 thin layers of BNNs. Moreover, the crystal images of BNNs include the well-organized
22 honeycomb structure. The magnified HAADF-STEM image also clearly depicts the atomic
23 structural characteristic of BNNs along with the distribution of B and N atoms. The hexagonal
24 symmetric selective area electron diffraction (SAED) patterns in **Fig. 1d** imply that the BNNs
25 are well matched with theoretical *h*-BN crystal structure [31]. Likewise, the STEM height line
26 profiling of BNNs further indicates the existence of the N atoms and B atoms along with the

in-plane direction of the BNN crystals. **Fig. S5** shows the TEM images of the exfoliated BNNs with few layered thicknesses. The most exfoliated BNNs have the diameters from few hundred nanometers to micrometers. These results indicate that the separator with the thin layers and small sizes of *h*-BN can guarantee the favorable ion diffusion paths through the in- or out-plane of BNNs. **Fig. 1e** depicts the freestanding BNN-Ss fabricated by the vacuum filtration method. The BNN-Ss can maintain its original structure even when it is strongly subjected to the different bending forces.

In order to evaluate and compare the heat-resistant properties of BNN-Ss and commercial polymer separators, thermal shrinkage tests were carried out at a temperature range from room temperature to 550 °C. The visual images of BNN-Ss and commercial polymer separators after heat treatment at various temperatures are presented in the bottom and top of **Fig. 2a**, respectively. Shrinkage (%) is defined as the ratio of undamaged area to the area of an original separator and is also plotted as a function of increased temperature. The photo images clearly indicate that the commercial polymer separators started its volumetric degradation after the heat exposure about 80 °C and lost over 70% of their original area upon the high temperature treatment (200 °C), which is in good agreement with previously reported literatures [32-34]. On the other hand, the thermal shrinkage of BNN-Ss is negligibly small or not identified even up to 200 °C. The more detailed shrinkage images of the commercial polymer separators and BNN-Ss are shown in **Fig. S6** and **S7**, respectively. Even at 550 °C, the BNN-Ss well maintain its original round shape without any surface and volume degradation. These results indicate that the strong thermal durability of BNN-Ss can be strongly attributed to the superior intrinsic physical and chemical properties of *h*-BN. In addition, the wettability tests with an electrolyte solution and ionic conductivity tests were performed. Compared to the commercial polymer separators, the BNN-Ss exhibits the better wettability and electrolyte uptake, as shown in **Fig. 2b, 2c** and **S8**. The variation of contact angles using droplets of the electrolyte solution was measured between the commercial polymer separators and the BNN-Ss. Specifically, the

electrolyte uptake of BNN-Ss is found to be 256%, which is approximately 2 times higher than that of the commercial polymer separators. In addition, the ionic conductivity of BNN-Ss, evaluated by bulk resistance from electrochemical impedance spectroscopy (EIS) results (**Fig. S9**), exhibits the reasonable value of 0.496 mS cm^{-1} , which is similar to that of the commercial polymer separators (0.461 mS cm^{-1}).

The distinct structural, thermal and ionic characteristics of *h*-BN properties and the as-prepared BN separators can be beneficial for the overall stability of Li ion cells during cycling. Especially, its extremely thermal stability can overcome any external thermal resources and the following malfunctions such as the deformation and decreased ionic conductivity of polymer separators. Also, the high rigidity of *h*-BN can handle all negative outcomes in cells by the natural growth of Li dendrites and the corresponding physical damage on polymer separators during cycling. To investigate the both ionic advantages of BNN-Ss from its physical and chemical properties, the BNN-Ss and commercial polymer separators were employed in the Li ion cells for the Li plating/stripping electrochemical experiment. It can be seen from **Fig. 3a** that a Cu plate (substrate), a Li foil and a 1.0 M LiPF_6 electrolyte were used as the working electrode, counter/reference electrode and electrolyte, respectively. The total Li plating capacity was set to be 1.0 mAh cm^{-2} on the Cu working electrode at a rate of 0.5 mA cm^{-2} . The Li stripping was then cycled to a cutoff potential of 2.0 V at the same rate of 0.5 mA cm^{-2} . Specifically, after 10 cycles, the electrochemical tests of the Li cells with the BNN-Ss and commercial polymer separators were paused and resumed after the 1hr thermal aging process in an oven to identify the experimental thermal stability of those separators. The coulombic efficiency was defined as the ratios between total Li plating and stripping capacity. As shown in **Fig. 3b**, before the aging process, the coulombic efficiencies (CE) on the Cu substrate during the Li plating/stripping are similar between the cells with the both separators. All cells can well maintain its initial CE values until first 10 cycles. After the aging process (the heat treatment while Li plating/stripping is stopped), the CE of the cells with the commercial polymer

separators dropped to about 50% of its initial CE after 20 cycles. After aging at 80 °C, the CE for commercial polymer separators is lowered due to the physical damage and shrinkage on the commercial separator. In contrast, the Li plating/stripping cells with the BNN-Ss can continuously guarantee its initial electrochemical properties, which are similar to the initial CE before the aging process. The dramatic CE differences right after the aging process can strongly represent that the high thermal stability of BNN-Ss might be essential to protect the stability of Li ion cells. Even in the severe conditions (after the aging process up to the 125 °C, **Fig. S10**), the CE of the cells with BNN-Ss is close to ~ 80 % after 20 cycles. After aging at 125 °C, the CE for the h-BN separators is well maintained, but slightly lowered. However, the CE for the commercial polymer separators is dramatically lowered due to the large physical damage and shrinkage on the commercial separator. It has been reported that the LiPF₆ electrolyte solution in organic carbonates is thermally stable up to 140 °C with non-electrochemical charge/discharge process [35]. Thus, these results are strongly attributed to the high thermal stability of *h*-BN while the degradation of commercial polymer separators occurs after thermal aging process.

The electrochemical behaviors of the Li plating/stripping cells with the BNN-Ss and commercial polymer separators were further characterized by the over-potential (voltage) versus capacity (time) results after 1st cycle and 20nd cycle (including the aging process) (**Fig. 3c and 3d**, respectively). For the first Li plating/stripping cycle, the Li metal is deposited on the Cu foil during the Li plating, but some of the Li metal is left over on the Cu foil while the stripping processes (C.E. is not 100%). After a few cycles, the initial nucleation barrier on the Li/Cu foil is lower than that of the pristine Cu foil. The high nucleation density leads to a comparatively low plating overpotential. The Li plating/stripping over-potential of the cells with the BNN-Ss is slightly lower than that of the cells with the commercial polymer separators at 20th cycle. The Li stripping over-potentials of the commercial polymer separator and BNN-S is 312 mV and 129 mV at the time of 0.75 h, respectively. Over-potential values can indicate

the status of Li plates on the Cu substrate which might result from the different structural and physical conditions of Li metal during cycling. The lower over-potentials of the cells with the BNN-Ss can representatively indicate the favorably deposited Li plates on the Cu substrate to continue the stable cycling performance of Li ion cells [22,24]. Moreover, there is a large voltage hysteresis (over-potential differences) at the cells with the commercial polymer separators, which is usually determined by the unfavorable interfacial charge-transfer properties. Those differences on the voltage versus capacity results might be strongly attributed to the morphologies of Li on the Cu substrate. The curve of the cells with the BNN-Ss after 20 cycles, after the aging process, indicates the longer capacitance (time) before reaching the cutoff potential of 2.0 V compared to the commercial polymer separators, subsequently demonstrating the CE with the BNN-Ss. The morphologies of the Li plates on the Cu substrate were visually investigated using scanning electron microscopy (SEM). The SEM images were taken after the heat treatment of 80 °C after 10 cycles and then the electrochemical test was continued for additional 10 cycles. As shown in **Fig. 3f**, the Li plates on the Cu substrate with the commercial polymer separators present the typical protruding and dendritic Li metal, including many one-dimensional Li wires. Such protruding structures can cause the failure of separator component in LIB. Using the BNN-Ss, however, the resulting Li granules show the smooth and uniform formation on the Cu substrate (**Fig. 3e**). The BNN-Ss show the high physical stability against the tensile strain from the Li dendrites after the electrochemical cycling. Due to the rigid and high physical stability of BNN-Ss, the cells with the BNN-Ss can physically inhibit protruding Li dendrite growth and successfully maintain its original electrochemical performance compared to the polymer-based separating materials as shown in schematically in **Fig. 3g** and **3h**. Collectively, the BNN-Ss can successfully provide the good thermal and physical stability of Li ion cells against the external and harsh heat environment as well as the physical deformation by the strain from dendritic Li growth. Therefore, in real LIB system, the BNN-Ss can decrease the risk of overall capacitance degradation and improve the cycling stability.

For the real application testing with the BNN-Ss, as shown in **Fig. 4a**, the electrochemical performance of BNN-Ss with lithium titanate (LTO) and Li metal electrodes was further tested. The spinel LTO ($\text{Li}_4\text{Ti}_5\text{O}_{12}$) electrode has been used as the electrode because LTO possesses high power processability, a unique zero-strain effect, stable charge and discharge voltage (~ 1.6 V vs. Li/Li^+), and excellent cycling stability. The LTO electrodes were used due to its high rate capability and long cycle life. **Fig. 4b** presents a cyclic voltammetry of the LTO electrode with the BNN-Ss [36]. The LTO electrode exhibits a pair of sharp peaks at about 1.50 V and 1.65 V, which are characteristic of the Li insertion/extraction on LTO. The charge–discharge curves of BNN-Ss at a current density of 100 mA g^{-1} are shown in **Fig. 4c**. A pair of distinct and flat voltage plateau at around 1.50 V and 1.65 V reflects the well-known two-phase Li insertion/extraction equilibria between $\text{Li}_4\text{Ti}_5\text{O}_{12}$ and $\text{Li}_7\text{Ti}_5\text{O}_{12}$ [37]. During the first cycle, the plateau at around 1.5 V (vs. Li/Li^+) would be related to the Li ion insertion/deintercalation of LTO and the small plateau at around 1.7 V (vs. Li/Li^+) is induced by Li ion insertion/deintercalation of TiO_2 phase. During the initial insertion, the unexpected side insertion of Li ion into the TiO_2 dominant phase can make the following additional reduction plateau [38–41]. However, after the 1st cycle, the observed small plateau is negligible. The LTO with the BNN-Ss delivers an initial discharge capacity of 171.2 mAh g^{-1} . The LTO electrode with the BNN-Ss exhibits better long-term cycling stability than that of the commercial polymer separators even in the room temperature (**Fig. S11**). As shown in **Fig. 4d**, the LTO electrode with the BNN-Ss present a high coulombic efficiency of $\sim 99 \%$ up to 600 cycles, whereas that of the commercial polymer separators drops to $\sim 78 \%$. **Fig. 4e** presents the rate-capability test of BNN-Ss and commercial polymer separators. At a current density of 1500 mA g^{-1} , the retention of the charge storage capacities of the LTO electrode with the BNN-Ss can reach a higher value of $\sim 60\%$ than that of the commercial polymer separators. These results might be attributed to the high wettability and ionic conductivity of BNN-S and further suggest that the BNN-Ss play a pivotal role for the enhanced energy storing kinetics and stable

operation/cyclability of LIBs (Table S1). Due to the thickness difference, the ionic conductivity of BNN-Ss, calculated using R_b and thickness of separator, shows the value of 0.496 mS cm^{-1} , which is slightly higher than or comparable to that of the commercial polymer separators (0.461 mS cm^{-1}). Moreover, the contact angle and electrolyte uptake results obtaining from the droplets of the electrolyte solution on the separators samples represent the high interface interaction between the solution and BNN-S as shown in Figure 2b and 2c. As illustrated in **Fig. 4f** and **4g**, similar to previous results,[21,22,24] the commercial polymer separators are not proper candidates to block the piercing Li dendrites growth on the Li metal electrode. In contrast, the uniform and smooth deposition of the additional Li dendrites by the BNN-Ss with superior thermal and physical stabilities can induce the better cyclability than the commercial polymer separators. The possible diffusion mechanism of Li ions through BNN-Ss might be attributed to the mitigation through atomic defects which are formed during the exfoliation steps. The feasibility of those defects working as the potential ion diffusion paths is demonstrated by density-functional theory (DFT) calculations (**Fig. S12**). **Fig. S12d** also shows the auxiliary DFT calculations for input models obeying the exact charge neutrality. On the exfoliated *h*-BN sheets, there can be two possible atomic defects which are placed in the location where the nitrogen or borone atomic bonding is broken. Based on the activation barrier energy calculation, the diffusion paths thorough those two defects show the lower activation barrier energy of Li electrolyte ions compared to the pristine BN structure.

Conclusion

In summary, new synthetic methods of thin *h*-BN nanosheets are developed and have successfully resulted in the *h*-BN nanosheets as the freestanding separators for electrochemical energy storage applications. The resultant freestanding *h*-BN nanosheet separators exhibit the excellent physical rigidity during the cycling and thermal endurance against the high-temperature environments without the dramatic performance degradation as well as show the

1 good Li ion diffusion kinetics. The *h*-BN separators can handle ~ 120 °C external heat without
2 the structural collapse and also well maintain its initial coulombic efficiency during the Li
3 deposition on the Cu substrate. The free-standing *h*-BN separators can maintain a reversible
4 coulombic efficiency of ~ 99% after the 600 cycles with the LTO electrodes. The strong thermal
5 and physical durability of the freestanding *h*-BN nanosheet separators can be strongly correlated
6 to the superior intrinsic physical and chemical feasibility of *h*-BN. Moreover, the density
7 functional theory (DFT) calculation reveals that the small defect sites on the *h*-BN nanosheets
8 can induce the much favorable Li ion penetration along with the sheets and corresponding
9 separators. Thus, based upon these findings, the facile and efficient processability of the *h*-BN
10 nanosheets and the corresponding freestanding separators can further extend the applicability
11 of energy storage applications in near future.

12

ASSOCIATED CONTENT

- Supporting Information

Experimental methods, Schematic illustration of the exfoliation process, XPS spectra before and after exfoliation method, Real image of exfoliated h-BN nanosheets, Raman spectra of pristine h-BN powder and exfoliated h-BN, SAXS analysis of BNNs, TEM images, Thermal stability tests of commercial separators and BNN-S, Electrolyte wettability test, Impedance plots, Coulombic efficiency (CE) versus cycle plots after 125 °C thermal aging step, Capacity cycling retention of LTO//Li cells, DFT crystal models of h-BN defects, Parameters for ionic conductivity calculation.

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- Author Contributions

J.H and A.-R.J. equally contributed to this work. J.H, A.-R.J., Y.-W.L. and J.I.S. conceived the idea. J.H, A.-R.J., and Y.-W.L. synthesized the samples, analyzed the data, and wrote the manuscript; W.B.P. and K.-S.S. analyzed the DFT data. B.H. analyzed the TEM data. K.-S.S., J.-O. L. S.N.C., Y.-W.L, J.I.S. contributed in reviewing the manuscript and provided scientific input.

- Notes

The authors declare no competing financial interest

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Figure Captions

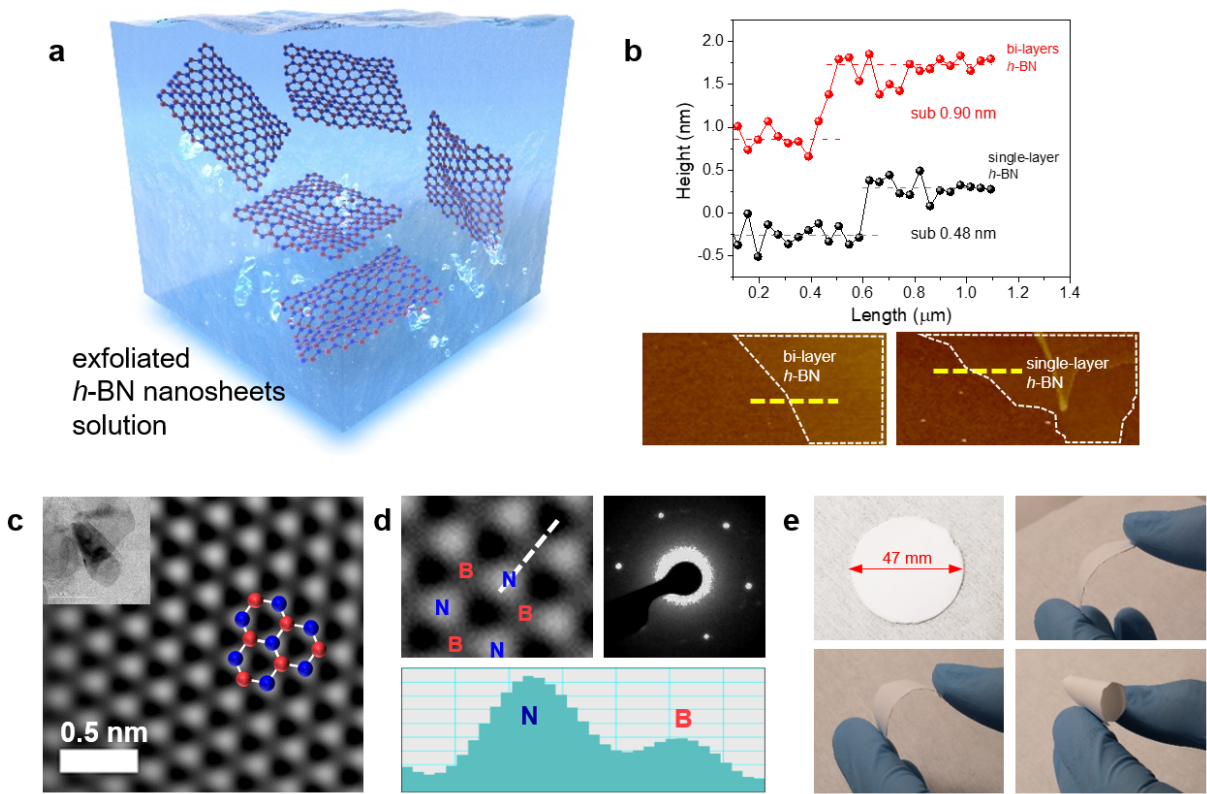
Figure 1. (a) A schematic image of exfoliated *h*-BN nanosheets. (b) AFM images of mono- and bi-layer BNNs & Linear AFM depth profiles of mono- and bi-layer BNNs. (c) A HAADF-STEM image of BNNs. The inset image shows a TEM image of BNNs. (d) A high resolution STEM image, SAED pattern and atomic line profile of BNNs. (e) Optical images of BNN-Ss.

Figure 2. (a) Thermal deformation tests of BNN-Ss and commercial polymer separators up to 300 °C (b) Contact angle (CA) of BNN-Ss and commercial separators with electrolyte solvents. (c) Electrolyte uptake (EU) calculation of BNN-Ss and commercial separators.

Figure 3. A schematic illustration of (a) Li plating/stripping experiment cells with BNN-Ss and commercial polymer separators. (b) Coulombic efficiency (CE) versus cycle plots with BNN-Ss and commercial polymer separators after 80 °C thermal aging process. Potential versus time plots during Li plating/stripping cycling with (c) BNN-Ss and (d) commercial polymer separators. SEM characterization of Li plating on the Cu substrate with (e) BNN-Ss and (f) commercial polymer separators. Schematic illustrations of deposited Li dendrites with (g) BNN-Ss (flat structures) and (h) commercial polymer (protruding structures).

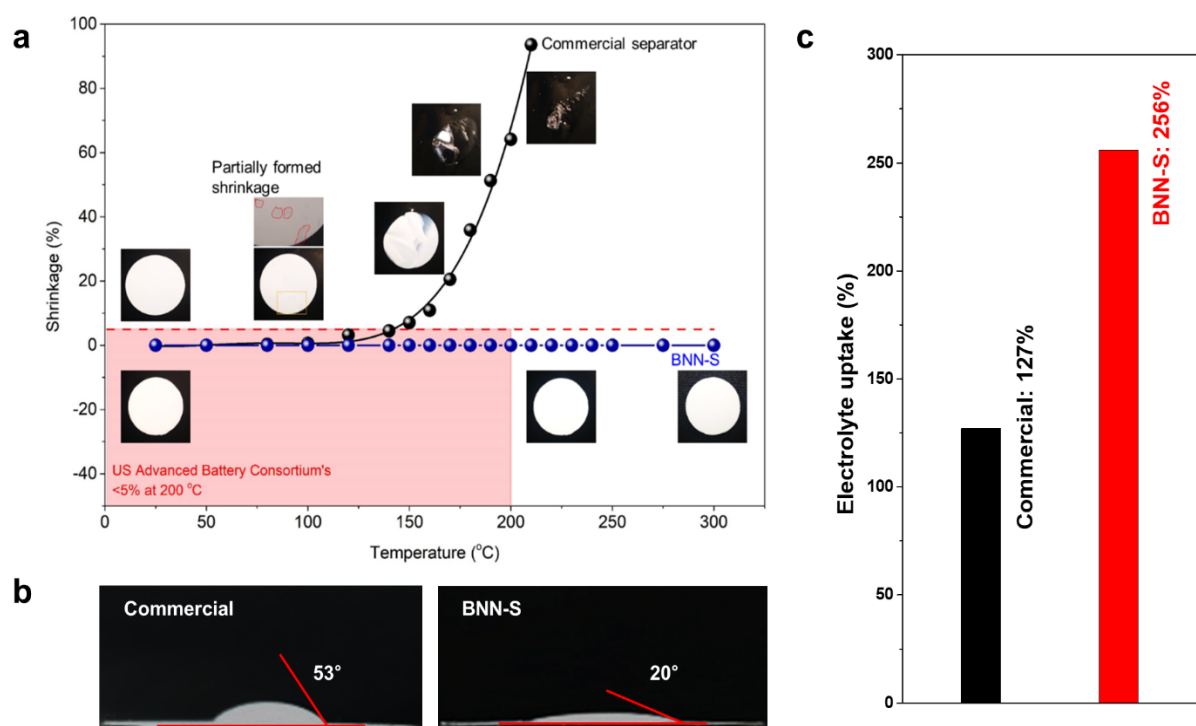
Figure 4. (a) A schematic ion diffusion illustration of LTO//Li cells with BNN-Ss. (b) Cyclic voltammetry of LTO//Li cells. (c) Charge-discharge curves of LTO//Li cells with BNN-Ss. (d) Coulombic efficiency of LTO//Li cells with BNN-Ss and commercial polymer separators. (e) Capacity rate retention of LTO//Li cells with BNN-Ss and commercial polymer separators. Schematic characterization of cycled Li metal electrodes in LTO//Li cells with (f) BNN-Ss and (g) commercial polymer separators.

Fig. 1. J. Hong et al.



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Fig. 2. J. Hong et al.



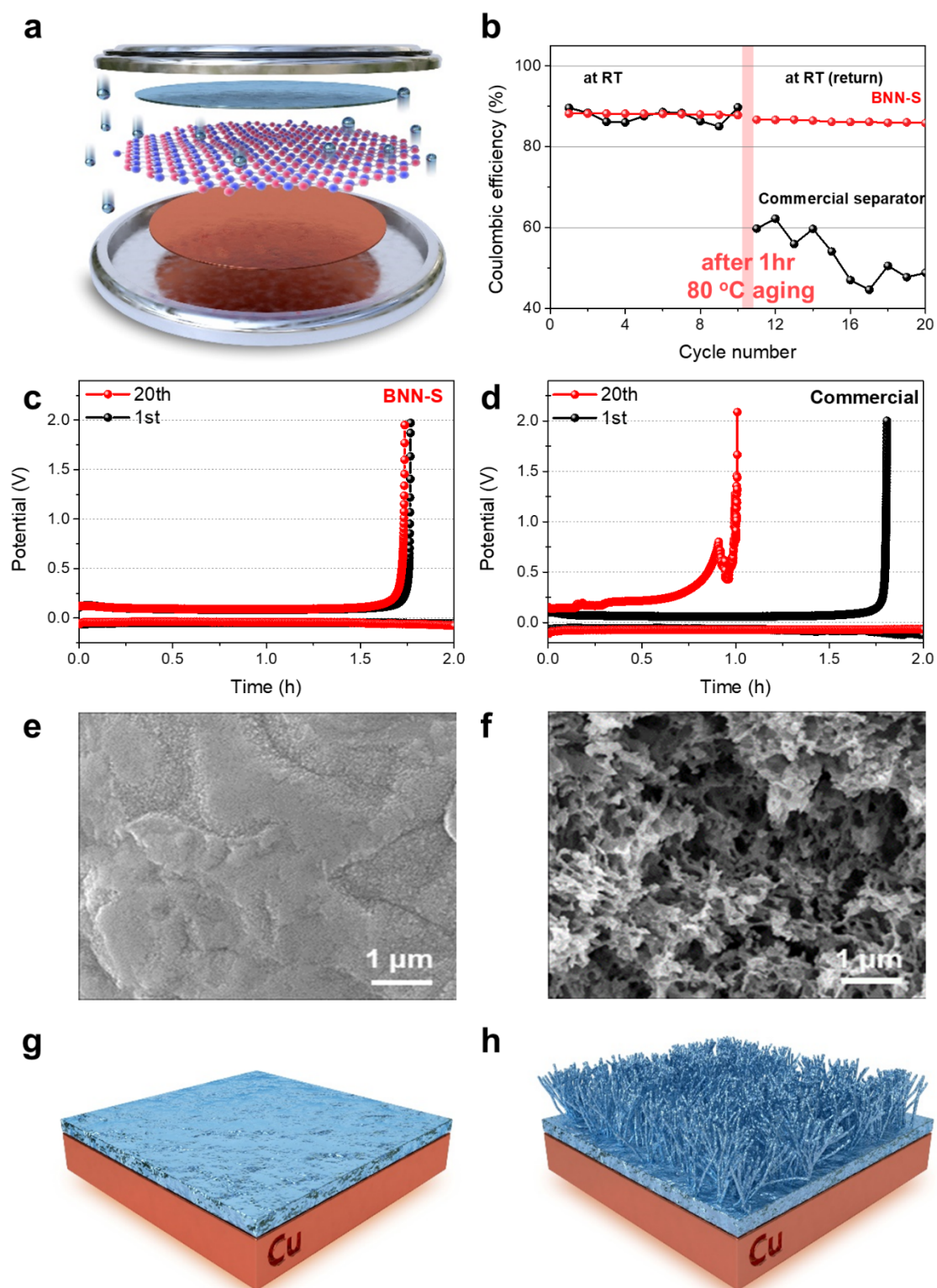
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Fig. 3. J. Hong et al.



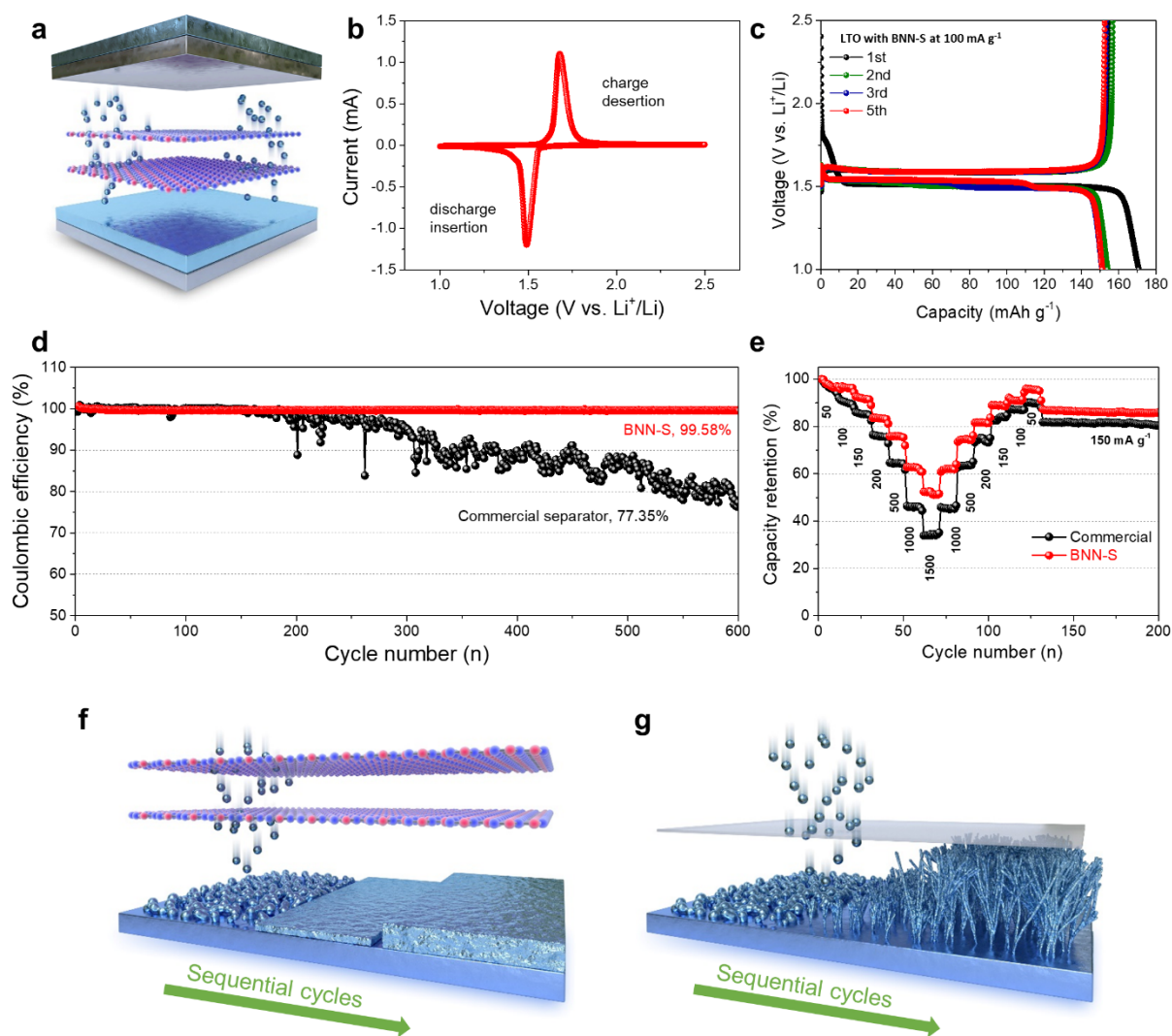
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Fig. 4. J. Hong et al.

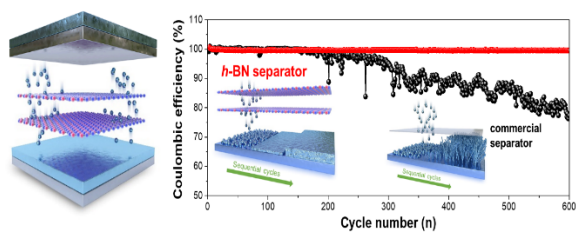


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5 Freestanding *h*-BN nanosheet separator show the high electrochemical performance in Li ion
6 battery including high physical and thermal stability.

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