LaLiO\textsubscript{2}-Based Multi-Functional Interlayer for Enhanced Performance of Li-S Batteries

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The “shuttle effect” of polysulfides and sluggish kinetics of sulfur redox are main problems in Lithium-Sulfur batteries. As a result, the Lithium-Sulfur battery systems are still far from commercialization for practical utilities. In this work, we design and prepare a Lanthanum lithium oxide - based multi-functional interlayer with good electronic and lithium ion conduction as well as efficient for polysulfide blocking. We obtain the multi-functional interlayer combined cathode by coating a composite of Lanthanum lithium oxide on as-prepared sulfur electrode. The modified sulfur cathode with such interlayer configuration gives enhanced electrochemical performance (1458.00 mA h g\textsuperscript{−1} capacity at initial cycle and 1004.40 mA h g\textsuperscript{−1} during the first cycle with 1C rate) with outstanding cycling stability up to 250\textsuperscript{th} cycle delivering the corresponding discharge capacity of 872.79 mA h g\textsuperscript{−1} in Li-S batteries. And, the rate performance has been improved by the facile strategy.

The dependencies of the society on various batteries is reaching outburst level in the modern days. Lithium–sulfur (Li–S) battery has been regarded as one of the best devices for next generation high-energy storage systems with promising high theoretical energy density of 2600 Wh kg\textsuperscript{−1}. However, it suffers from various problems associated with sulfur and its soluble and insoluble conversion products (Lithium polysulfides (Li\textsubscript{2}S\textsubscript{x}, 2 ≤ x ≤ 5)). Among them, the so-called shuttle effects derived from the polysulfides are very stubborn, which lead to a terrible cycling stability, poor rate capability and low utilization of sulfur. Striving to alleviate such problem in Li-S batteries, the porous carbons on the one hand, have been widely applied as a potential remedial material to host sulfur either alone or mixed with other metal compounds. The porous structures of carbon have been found to be efficient in encapsulating the active elemental sulfur in its pore. On the other aspects, incorporating interlayer\textsuperscript{9} between the separator and the sulfur cathode is used to mitigate the polysulfide shuttling issue in Li-S batteries.\textsuperscript{10–13} The effect of the interlayers is to block the polysulfide from diffusing in between the two electrodes either by physically trapping on its surface (physio sorption) or by electrochemical interactions (chemisorption and electrocatalytic conversion). However, most of the interlayers, which are usually employed to coat on a separator, are just acted as a simple blocking wall, will decrease the content of sulfur and make a barrier for the Li ion diffusion. In addition, the complex preparation processes limit their large-scale application. In this work, we design and prepare a simple LaLiO\textsubscript{2}-based multi-functional interlayer (LMFI) in Li-S batteries (Fig. 1), which is not only a good lithium ion conductor but also prevent from penetrating of polysulfides due to its unique properties and structure. At the same time, the LMFI can promote the sulfur conversion reactions as well. By the strategy, a Li-S battery with improved electrochemical performance has been achieved.

**Experimental**

**Synthesis of LaLiO\textsubscript{2} nanoparticles.**—The non-aqueous synthesis route was used for the preparation of binary metal oxides. Typically, the binary metal oxide, Lanthanum lithium oxide (LaLi\textsubscript{2}O\textsubscript{3}) nanoparticles was synthesized by mixing 1g Lanthanum acetylacetonate monohydrate (La(C\textsubscript{5}H\textsubscript{7}O\textsubscript{2})\textsubscript{3}\textcdot H\text{2}O), 0.5g Lithium formate monohydrate (HCO\textsubscript{2}Li.H\text{2}O), 0.05g hexadecyl trimethyl ammonium bromide (HTAB) in 30ml benzylamine. The solution was stirred vigorously for 1 hour and transferred to autoclave followed by solvothermal treatment at 200°C for 24 hours in an oven. When cooled, the solution was centrifuged and washed five times with ethanol and the precipitate was allowed to dry at 60°C. Finally, the dried solid was crushed properly and calcinated at 400°C in furnace for 1 hour.

**Preparation of cathode.**—The sulfur cathode composite was prepared by following well established melt-diffusion strategy. Typically, cautiously weighed sublimed 73wt% sulfur and 27wt% electrically conductive carbon (Super-P) were mixed to homogenize and closed under vacuum followed by heating for 6 hours at 155°C to allow complete diffusion of melted sulfur into the porous structure of the carbon. Then 70wt% of the C/S composite, 20wt% of conductive Super-P carbon and 10wt% of n-lauryl acrylate polymer binder (LA, Chengdu, China) were mixed and ball-milled for 6 hours to produce homogeneous thick slurry and casted on circular aluminum foil current collector (diameter = 16.3mm) with simple roll-casting strategy and dried overnight in a vacuum oven at 60°C and 0.05bar. As-prepared cathode discs obtained in this way achieved the areal

![Figure 1. Schematic of typical Li-S battery with LMFI composite coated on sulfur/carbon cathode (LMFI-battery) undergoing discharging/charging process. The LMFI coating trap the polysulfide on the cathode side and avoids the shuttling effect, enhance the kinetics of interconversion reactions of (S, Li\textsubscript{2}S\textsubscript{2} and Li\textsubscript{2}S\textsubscript{3}) and also allowing the Li\textsuperscript{+} ion transfer.](image-url)
sulfur loading of around 2.3 mg cm$^{-2}$. Therefore, the corresponding active sulfur content in every electrode was used to calculate the specific capacities reported here in this work. All the experiments were tested at room temperature.

**LMFI fabrication.**—The LMFI wall between the as-prepared cathode and the polypropylene separator was fabricated by simple slurry coating strategy on the as-prepared cathode disc. Typically, the coating slurry was prepared from the lanthanum lithium oxide nanoparticles, Super-P carbon and n-lauryl acrylate polymer (LA) binder with weight ratio of (4.5:4.5:1) respectively were mixed well for 6 hours at room temperature using planetary ball milling. Then the slurry was casted on as-prepared cathode and dried in a vacuum oven for 24 hours at 60 °C and 0.05 bar. The average mass loading of LaLiO$_2$ on LMFI coated cathode is 0.2 mg. The obtained hybrid interlayer coated cathode is denoted as LMFI cathode here after. On the other hand, the Super-P interlayer coating on as-prepared cathode was also fabricated following the same procedure but without lanthanum lithium oxide nano particles. In this case we assigned the resulting interlayer coated cathode as control cathode throughout this work.

**Cell assembly.**—The cell assembling was all carried out in Argon filled dry glove box in which the water content and the oxygen level were kept less than 1 ppm. The LMFI cathode, polypropylene separator and the Li-metal foil were arranged properly to fit with coin type cell (CR2032). The separator was impregnated with electrolyte 0.5 M lithium trifluoromethanesulfonate (LiCF$_3$SO$_3$), 0.5 M lithium nitrate (LiNO$_3$) in (1:1 v/v DOL/DME) (DoDoChem, China). Then the components were assembled followed by crimping the cell at the pressure of 55 bar. Other coin cells with control cathode were also assembled in similar way for comparison. The assembled cells (LMFI battery and control battery) were given 30 minutes of resting time before electrochemical measurements.

** Ionic conductivity test.**—The pellet of LaLiO$_2$ was prepared by coating the slurry on the nickel disc. Typically, the slurry was prepared by mixing 9:1 ratio of LaLiO$_2$ and LA binder followed by ball milling for 10 hours. The thickness of the as-prepared pellet was measured to be 1 mm. For ionic conductivity test, the LaLiO$_2$ pellet was assembled in between two nickel discs as blocking electrodes in Argon filled glove box. Then the measurements were performed using electrochemical impedance instrument (Zahner Zennium Electrochemical Workstation, Germany) within the frequency range of 100 mHz - 100 kHz with applied voltage signal of 5 mV.

**Material characterizations.**—The structural morphologies of as synthesized samples of LaLiO$_2$ were examined using scanning electron microscope (SEM S-4800). The weight percent composition of the sulfur in the cathode materials was determined using thermogravimetric analysis (TGA) technique employing (TA Instrument Q600 Analyzer) in N$_2$ atmosphere followed by air.

The (Micrometrics TriStar II 3020 V1.01, surface area and porosity analyzer) instrument was used to collect the nitrogen adsorption/desorption isotherms at 77 K. The Brunauer–Emmett–Teller (BET) technique was employed to calculate the specific surface area of the LaLiO$_2$ particles in accordance with multi-point analysis and the total volume of pores was determined with single point analysis at relative pressure (P/P$_0$ = 0.990). The Barrett–Joyner–Halenda (BJH) model was used to determine the pore size distribution of the particles from N$_2$ - desorption isotherm.

The X-ray diffraction (XRD) patterns of the samples were collected using Rigaku Ultima IV diffractometer, with an X-ray source operating condition 40 kV/30 mA, using Cu K$_\alpha$ radiation ($\lambda = 1.5406$ Å). The absorption of ultraviolet-visible light by the LMFI composite and Super-P particles in Li$_2$S$_6$ solution was measured by the UV-Vis spectrophotometer (UV 2550) to investigate the adsorption of Li$_2$S$_6$ on the particles.

**Electrochemical measurements.**—The assembled coin cells after 30 minutes of resting time were tested for electrochemical performance. The cyclic voltammograms were acquired from electrochemical work station (VMP2 Multichannel potentiostat, Princeton Applied Research) with scan rate of 0.1 mV s$^{-1}$ within the potential range of 1.7–2.8 V where the sulfur undergoes various electrochemical interconversion reactions with lithium metal. The electrochemical impedance measurements of cells were also performed before and after discharge/charge cycling test using impedance analyzer (Zahner Zennium Electrochemical Workstation, Germany) within the frequency range of 100 mHz - 100 kHz with applied voltage signal of 5mV.

The electrochemical cycle stability as well as the rate capabilities of the coin cells were investigated by performing Galvanostatic discharge/charge cycles at room temperature using NEWARE battery tester within the cutoff potential range of 1.7–2.8 V. The current density applied to every Galvanostatic cycling tests was based on the active mass of sulfur loading where (1C = 1675 mA g$^{-1}$).

**Results and Discussion**

Fig. 2a shows the SEM image of the LaLiO$_2$ nano-particles clustered together. The respective X-ray powder diffraction pattern collected Fig. 2b, showed a very good peak fit with the standard reference pattern of (JCPDS card no. 37-1023) belonging to the monoclinic phase of lanthanum lithium oxide with the space group P21/c.

The surface area measurement of the LaLiO$_2$ nano particles showed a Brunauer–Emmett–Teller (BET) specific surface area of 64.04 m$^2$ g$^{-1}$ and a total pore volume of 0.29-49 cm$^3$ g$^{-1}$ Fig. 2c, with an average pore size of 15.88 nm. This characteristic porosity and surface area of LaLiO$_2$ together with the ionically conductive nature can contribute to the LMFI layer to hold polysulfides within its pores and allow lithium ions to pass quickly.

To demonstrate the interaction between the polysulfides and the LMFI composite and its effect on the binding of polysulfide, 20mM of Li$_2$S$_6$ was prepared in DOL/DME (1:1, v/v) as representative for higher order soluble polysulfide. Hence, the adsorption of the Li$_2$S$_6$ on Super-P (C) and LMFI composite was compared. The digital image in Fig. 3a indicates the decoloration of the polysulfide as the result of adsorption. The apparent fading from the yellowish color to light appearance is indicative of the stronger adsorptive effect of the LMFI composite than the C-counterpart. This effect is confirmed by the UV-Vis spectrophotometric measurement of the samples. As a result, Fig. 3b shows the UV-Vis spectra of three samples of Li$_2$S$_6$ (blank), with C and LMFI composite. As clearly seen on the spectra, the intensity of Li$_2$S$_6$ absorbance band decreased when C as well as the LMFI composite are added. The more prominent decline in the intensity is noticeable in the sample containing the composite. This result ascribes the effect of LaLiO$_2$ nano-structures on strong entrapment of Li$_2$S$_6$.

The galvanostatic charge/discharge test of the Li-S batteries both with LMFI and the control group (Fig. 4a) show two voltage plateau at different potential regions, the first at high voltage region corresponding to the electrochemical reduction of sulfur to high order polysulfides, the second plateau at the lower potential region (mostly the working voltage region) where the high order lithium polysulfide undergoes further reduction to give insoluble lithium sulfides (Li$_2$S$_x$ / Li$_2$S). These features are in harmony with the CV result recorded Fig. 5a. Furthermore, the discharge/charge profiles of the Li-S battery displayed a very high initial capacity of 1458.00 mA h g$^{-1}$ at initial cycle Fig. 5b. While, when the battery was cycled at 1C rate, it imparted a capacity of 1004.40 mA h g$^{-1}$ at the first cycle and showed outstanding cycle stability up to 250th cycle with the corresponding discharge capacity of 872.79 mA h g$^{-1}$. However, the control battery just offered an initial capacity of 1084.00 mA h g$^{-1}$ at initial cycle Fig. 5b. When the cell was cycled at 1C rate for many cycles for
Figure 2. The SEM image and XRD pattern of Lanthanum lithium oxide (LaLiO₂) (a, b); The N₂ adsorption-desorption isotherm at 77K and the inset pore size distribution by the Barret–Joyner–Halenda (BJH) method of the LaLiO₂ (c); The TGA curve of S/C composite (d).

Figure 3. (a) The optical image of sealed bottles of 20mM Li₂S₆ (blank, with Super-P carbon and containing LMFI composite) displaying the decoloration of the Li₂S₆ solution. (b) The UV-Vis absorption spectra of the solutions.
Figure 4. (a) The discharge/charge profiles of LMFI-battery and Control battery. (b) The specific discharge capacity versus cycle number comparison of Li-S batteries with LMFI and without LMFI at 1C rate.

Figure 5. (a) The cyclic voltammogram of Li-S battery with LMFI scanned at 0.1mV/s within the voltage range of 1.7–2.8 V. (b) The initial cycle discharge/charge profiles of batteries with LMFI and without LMFI displaying the overpotential comparison. (c) Typical Nyquist plot of the EIS spectra of cycled Li-S batteries with LMFI and control (without LMFI) fitted with the inset equivalent circuit. (d) The linear plots of $Z'$ versus $w^{-1/2}$ of batteries with LMFI and without LMFI obtained from fresh and cycled cells at low frequency region.
comparison, it delivered the capacities of 710.40 and 560.36 mAh g$^{-1}$ respectively for 1$^{st}$ and 250$^{th}$ cycles (Fig. 4b). The capacity obtained after 250$^{th}$ cycle for LMFI-battery still exceeds by 36% that of the control one for the corresponding cycle implying that more stable cycle performance attributed to LMFI coating.

The comparison of the electrochemical cycle performance of Li-S battery obtained in this work with recently published literatures is given in Table I. Accordingly, the respective initial discharge capacitances with the corresponding employed C-rates have been listed. From those selected works Table I reported in the literature, the LMFI-battery has shown enhanced electrochemical cycle performance with greater residual specific capacity after 250 cycles at a current rate of 1C.

Table I. Comparison of the electrochemical performances of Li-S batteries employing various interlayers in the literatures with the present work.

| Interlayer                              | Initial capacity (mAh g$^{-1}$)/C-rate | Potential range (V) | Cycle performance Final capacity (mAh g$^{-1}$)/C-rate | Cycle number | Ref.  |
|-----------------------------------------|---------------------------------------|---------------------|-------------------------------------------------------|--------------|-------|
| LMFI                                    | 1458/0.1C                             | 1.7–2.8             | 872.79/1C                                             | 250          | This work |
| Fe/C carbon nanotubber web               | 1177/0.2C                             | 1.5–2.8             | 893/0.2C                                              | 100          | 29    |
| Self-assembled MWCNT                    | 1112/0.1C                             | 1.9–2.8             | 851/0.5C                                              | 100          | 30    |
| Reduced graphene oxide film             | 1260/0.1C                             | 1.0–3.0             | 895/1C                                                | 100          | 31    |
| TiO$_2$/carbon nanotube paper           | 1085/0.5C                             | 1.7–3.0             | 5760/5C                                               | 250          | 32    |
| TiO$_2$/carbon nanotube paper           | 875/0.2C                              | 1.7–2.8             | 805/0.1C                                              | 100          | 33    |
| Porous CoS$_2$/carbon paper             | 1239/0.2C                             | 1.7–2.8             | 818/0.2C                                              | 200          | 34    |
| 3D CCF                                  | 1085/0.5C                             | 1.0–3.0             | 870/0.5C                                              | 75           | 35    |
| Graphene wrapped sulfur particles       | 750/0.2C                              | 1.7–2.8             | 600/0.2C                                              | 100          | 36    |
| MWCNT                                   | 1446/0.2C                             | 1.5–2.8             | 804/1C                                                | 100          | 9     |
| Carbon monolith                         | 1120/0.5C                             | 1.5–3.0             | 650/0.5C                                              | 100          | 37    |
| Nickel Foam foam                        | 8190/0.2C                             | 1.5–3.0             | 604/0.2C                                              | 80           | 5     |

As discerned in Fig. 5c, the overall impedance of the battery with LMFI is found to be lower than the control one. Table II shows the results of the EIS spectral fittings of typical Li-S batteries (with and without LMFI) of cycled cells (Fig. 5c) and fresh cells (supporting information, Fig. S3) with the inset equivalent circuit model in Fig. 5c.

The CV profile presented in Fig. 5a for LMFI-battery shows stable cycling with characteristic oxidation peaks at about (2.4 and 2.46V) and two reduction peaks at about (2.35 and 2.0 V). The slight divergence of the anodic and cathodic peak profiles of the first cycle from that of the subsequent cycles is due to the effect of the rearrangement of the anodic and cathodic currents in the repeated CV cycles implying that the presence of the LaLiO$_2$ coating contributed to the improved stability of the cathode composite. The two cathodic peaks at about 2.35 and 2.0 V confirms the reduction of cyclic octa sulfur (S$_8$) to soluble higher order polysulfides (Li$_x$S$_y$, 4 < x < 8) and consequently higher polysulfides to insoluble lower order sulfides (Li$_2$S$_2$ and Li$_2$S) respectively. Similarly, in anodic scan two peaks were discerned at about 2.4 and 2.46V which are ascribed to the oxidation of polysulfides and the insulating insoluble Li$_2$S products back to the octa sulfur.$^{22,23}$ These features imply that reasonable kinetics of electrochemical processes are occurring. From the CV of the blank cell (supporting information, Fig. S1), the LaLiO$_2$ capacity contributions can be ignored. The rate capabilities were tested at different C-rates of 0.1, 0.2, 0.3, 0.5, 1 and 2C. As shown in (supporting information, Fig. S2), the results revealed that the Li-S batteries with LMFI showed enhanced rate performance than the control counterpart. Moreover, the batteries were re-cycled with 0.1 and 0.2C rates after subsequent cycling at varied C-rates, and the results show that, the LMFI-battery could recover about 77% of its capacity at 0.1C rate and about 65% at 0.2C rate. As we can clearly see in Fig. S2 that, the profile of the battery with LMFI is stable throughout every rate tests carried out with higher rate capability than the control battery. In contrast, the rate profile of the battery without LMFI showed lower rate performance throughout the assigned C-rate tests with very low capacity output.

Also, the Li-S battery with LMFI has less over potential than the battery without LMFI, attributed to the synergistic effect of LMFI composite with good electronic and ionic conductivity, which formed an expanded sulfur cathode thus to enhance the charge transfer and boost the redox kinetics of sulfur (Fig. 5b).

Further electrochemical tests of Li-S cells were conducted using Electrochemical Impedance spectroscopic techniques. The EIS spectra of the Li-S battery in this study have been fitted with the equivalent circuit shown in the inset of Fig. S2 using ZView software (Scribner Associates). The characteristic spectra of these typical Li-S batteries have shown two semi-circles at high and medium frequency regions. And an inclined straight line at about 45° with the real impedance axis at low frequency region. The physical implications of these features of the EIS spectra are related to the equivalent circuit and useful information were acquired from the Li-S batteries under study. Accordingly, the Rs- represents the resistances associated with the electrolyte solution, separator and electrodes. R1 and CPE1 are in parallel configuration with each other and in series with Rs, resulting the first semi-circle at high frequency region of the spectra representing the resistance and capacitance of the solid film formed on the electrodes surfaces. R2 and CPE2 are the other category of the circuit in parallel arrangement with each other and series with the previous block of the circuit elements, giving the second semi-circle at medium frequency region stands for the charge transfer resistance and double layer capacitance of the electrolyte/electrode interface. The CPE3 in series configuration with previous RC blocks of the circuit, stands for low frequency region infinite length Warburg element associated with the Li$^+$ ion diffusion through active electrode particles.$^{22}$ The non-idealities of electrodes (roughness and or surface porosity) were compensated by using constant phase element (CPE) instead of pure capacitors in modeling of the electrochemical cells with physical equivalent circuit.$^{21,24}$ Similarly, the CPE was used instead of Warburg element in order to make suitable for spectral fitting at low frequency region.$^{21}$

As discerned in Fig. 5c, the overall impedance of the battery with LMFI is found to be lower than the control one. Table II shows the results of the EIS spectral fittings of typical Li-S batteries (with and without LMFI) of cycled cells (Fig. 5c) and fresh cells (supporting information, Fig. S3) with the inset equivalent circuit model in Fig. 5c.

$$D_{Li+} = \frac{R^2T^2}{2A^2n^4 F^4 C^2 \sigma^2}$$

[1]

Table II. The EIS Data values extracted from fitting of experimental data of cycled cells in Fig. 5c and fresh cells (supporting information, Fig. S3) with the inset equivalent circuit model in Fig. 5c.

| Li-S cell | Rs (Ω) | R1 (Ω) | R2(Ω) | σ (Ω.s) | D$_{Li+}$(cm$^2$/s) |
|----------|--------|--------|--------|---------|---------------------|
| Fresh control battery | 10.87 | 12.05 | 10.69 | 4.39 | 1.05 × 10$^{-10}$ |
| Cycled control battery | 11.48 | 19.47 | 14.45 | 5.32 | 0.72 × 10$^{-10}$ |
| Fresh LMFI-battery | 9.98 | 8.08 | 2.32 | 1.92 | 5.46 × 10$^{-9}$ |
| Cycled LMFI-battery | 8.09 | 14.51 | 6.75 | 3.15 | 2.05 × 10$^{-9}$ |
Where, $D_{Li^+}$ is the diffusion coefficient of Li+, $R$ is the universal gas constant, $T$ is the absolute temperature, $A$ is surface area of the electrode, $n$ is the number of electrons transferred, $F$ is the Faraday constant, $C$ is the concentration of Li+ in electrolyte and $\sigma$ is the coefficient of Warburg line which is extracted from the slope of a plot of $Z'$ (or $Z''$) against the inverse square root of the lower angular frequency ($\omega^{-1/2}$) Fig. 5d, using the relation given in Equation 2.26.

$$Z' \propto \sigma \omega^{-1/2}$$

Where, $Z'$ and $Z''$ are real and imaginary impedances respectively. The results obtained for $D_{Li^+}$ and $\sigma$ are given in Table II. Evidently, the LMFI layer has larger Li+ ion diffusion coefficient comparing to the control one.

In addition, the solid state ionic conductivity of the LaLiO2 nanoparticles was tested by assembling the LaLiO2 pellet in between two nickel discs configured as (Ni/LaLiO2/Ni) at various temperatures ranging from 25 - 70 °C using EIS measurement. Ionic conductivities ($\rho$) were extracted from the EIS spectra and using the relation in Equation 3.27

$$\rho = l/R' A$$

Where, $l$ is the thickness of the LaLiO2 pellet, $R'$ is the resistance obtained from the intercept of impedance spectra with the real axis and $A$ is the area of the LaLiO2 pellet.

The results of the measurements were presented in (supporting information, Fig. S4). As it obviously seen in the figure that, with increasing the temperature, the impedance of the cells is getting lower and lower implying a linearly increase of the ionic conductivities. The activation energy ($E_a$) has also been determined from the experiment using Arrhenius relation in Equation 4.28 and its value is 1.35 eV.

$$\rho = \rho_0 \exp(-E_a/k_BT)$$

Where, $\rho_0$ is the pre-exponential factor, $k_B$ is the Boltzmann’s constant and $T$ is the temperature in Kelvin.

Four different cells were tested at 55 °C and the resulting spectra were used to determine the ionic conductivity. Accordingly, the ionic conductivity of the LaLiO2 was found to be 8.98 $\times$ 10$^{-7}$Scm$^{-1}$.

**Conclusions**

In this work we showed the enhanced electrochemical performance of Li-S battery with fabrication of LaLiO2-based multifunctional interlayer (LMFI) decorated on the as-prepared sulfur cathode. The LMFI was found to refrain the shuttling polysulfide and also enhance the kinetics of sulfur conversions in electrochemical processes.

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