

## Supporting Information

### **Three-dimensional nitrogen-doped graphene framework decorated with atomic layer deposited ultrathin V<sub>2</sub>O<sub>5</sub> layer for lithium sulfur battery with high sulfur loading**

Xiaowei Liu<sup>a</sup>, Zhaohuai Li<sup>a</sup>, Xiaobin Liao<sup>b</sup>, Xufeng Hong<sup>a</sup>, Yan Li<sup>b</sup>, Cheng Zhou<sup>a</sup>, Yan Zhao<sup>b</sup>, Xu Xu<sup>\*a</sup> and Liqiang Mai<sup>\*a</sup>

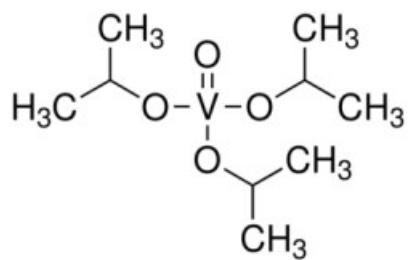
<sup>a</sup>State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, International School of Materials Science and Engineering, Wuhan University of Technology, Wuhan 430070, Hubei, China.

<sup>b</sup>State Key Laboratory of Silicate Materials for Architectures, International School of Materials Science and Engineering, Wuhan University of Technology, Wuhan 430070, Hubei, China.

\*E-mail: xuxu@whut.edu.cn(X. Xu); mlq518@whut.edu.cn(L. Mai).

**Precursor:**

Vanadium(V) triisopropoxy oxide (VTIP)

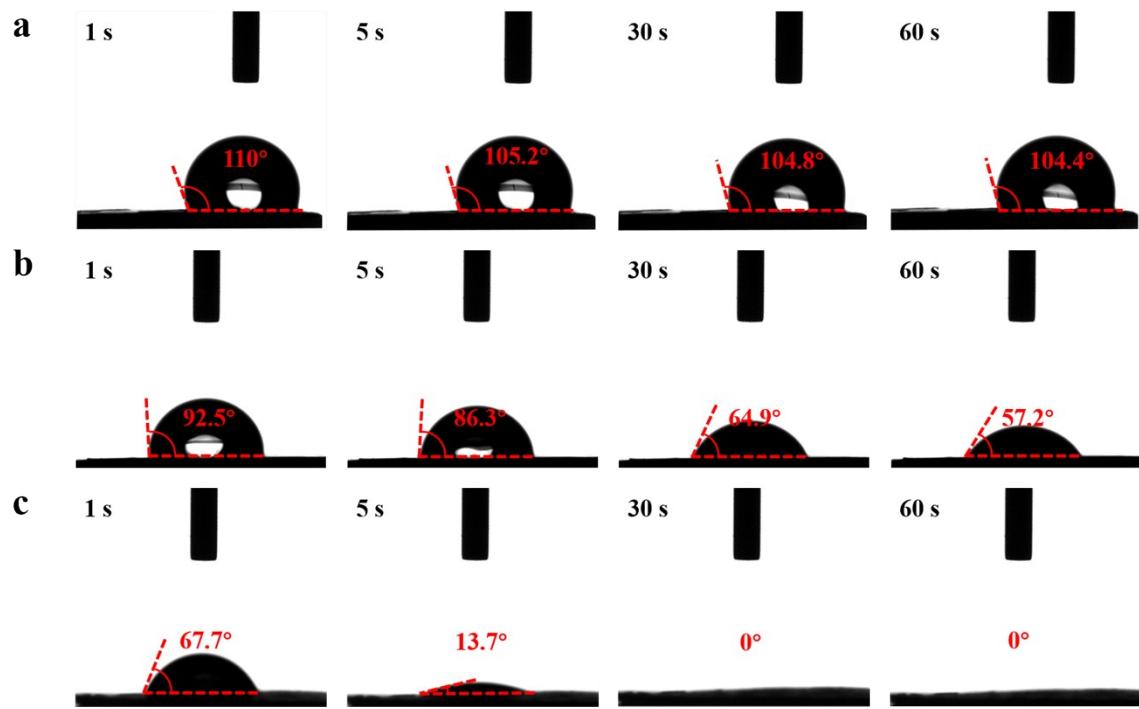


CAS Number: 5588-84-1

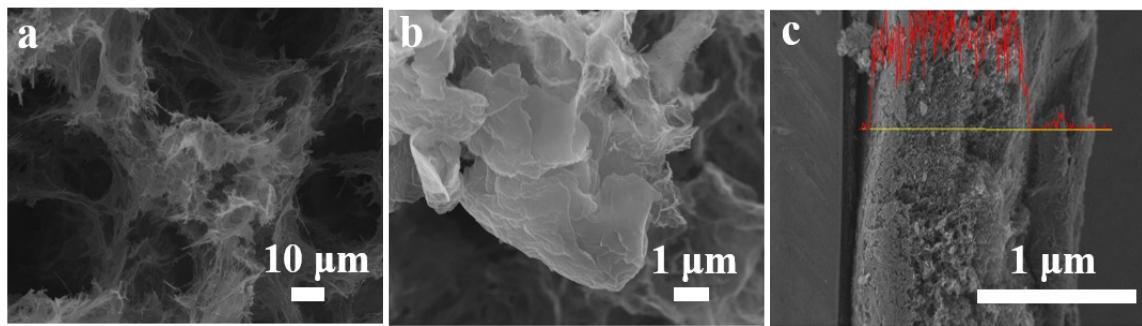
Heated to 70 °C

	Instruction	#	Value
0	Heater	9	150
1	Heater	8	150
2	Stabilize	9	
3	Stabilize	8	
4	Flow		20
5	Wait		1800
6	Flow		5
7	Wait		2
8	Stopvalve		0
9	Pulse	1	0.15
10	Wait		5
11	Stopvalve		1
12	Flow		20
13	Wait		20
14	Flow		5
15	Wait		2
16	Stopvalve		0
17	Pulse	0	0.03
18	Wait		5
19	Stopvalve		0
20	Flow		20
21	Wait		20
22	goto	6	100
23	Flow		5

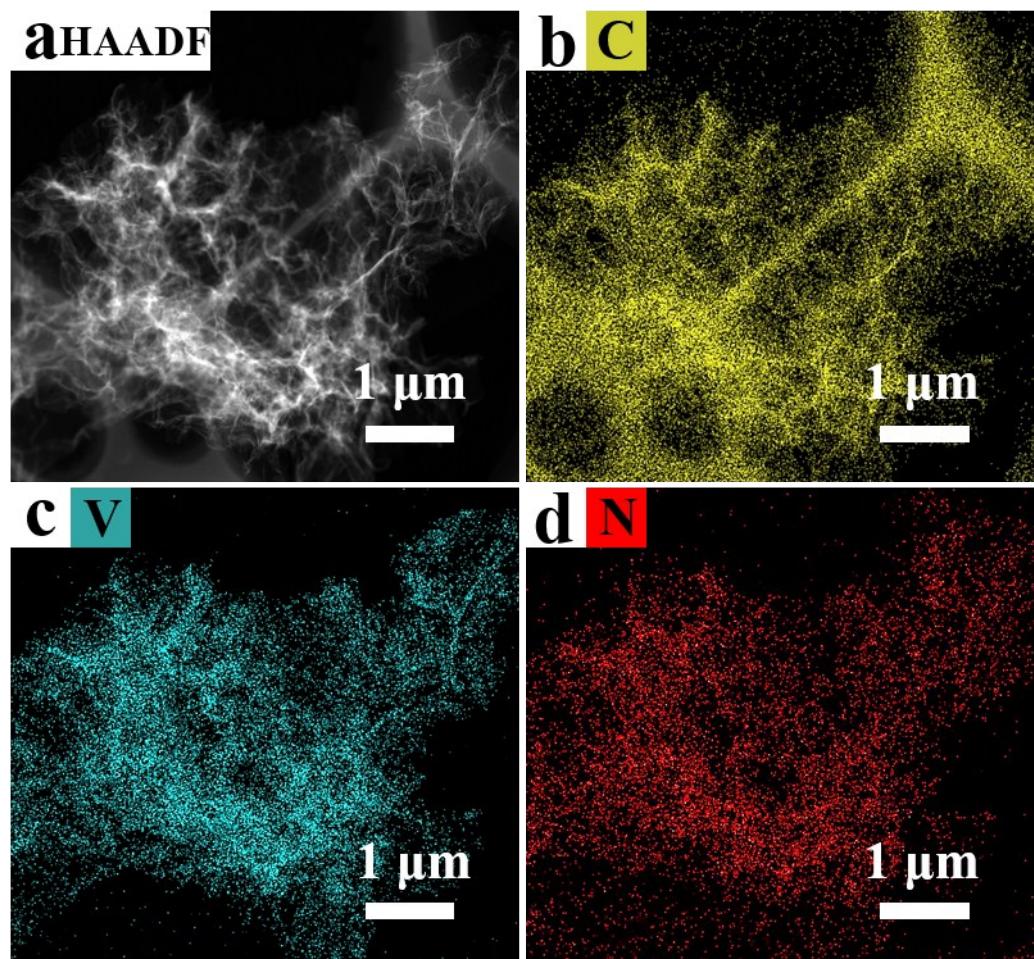
**Fig. S1.** Details about precursor and operation of ALD process.



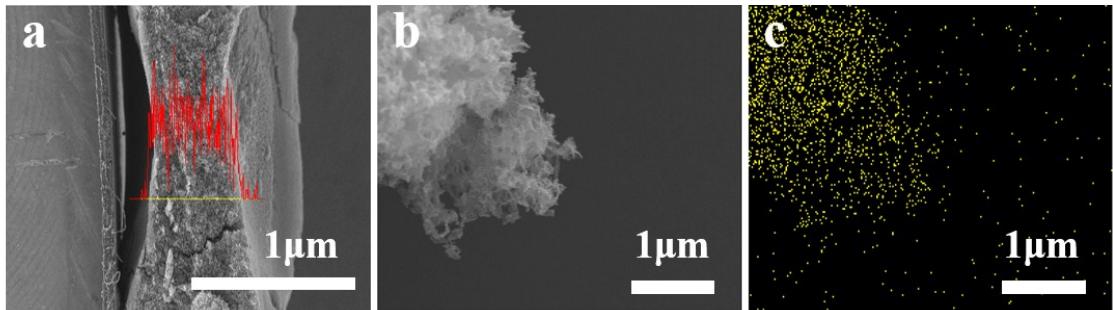
**Fig. S2.** Contact angle tests of (a) 3DG, (b) 3DNG, (c) 3DNG with plasma enhanced.



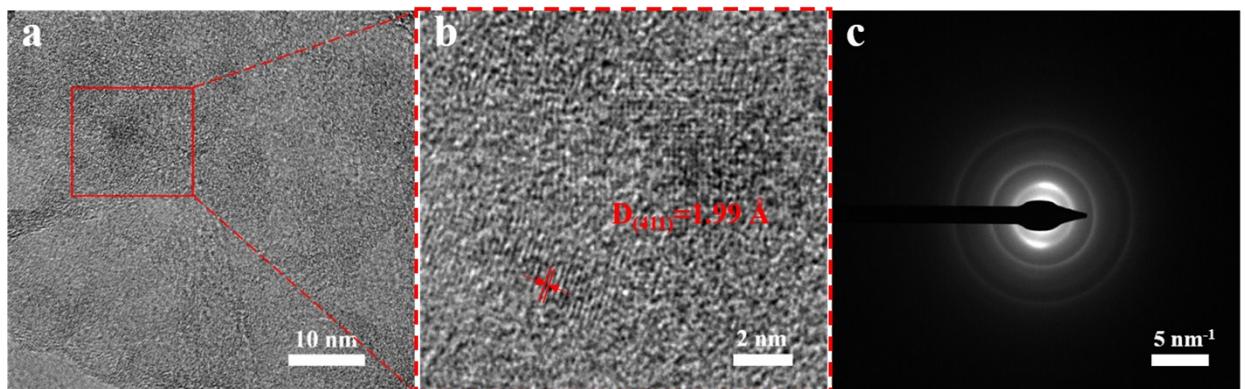
**Fig. S3.** (a, b) SEM images of TVO@3DNG, (c) element V linear analysis of TVO@3DNG.



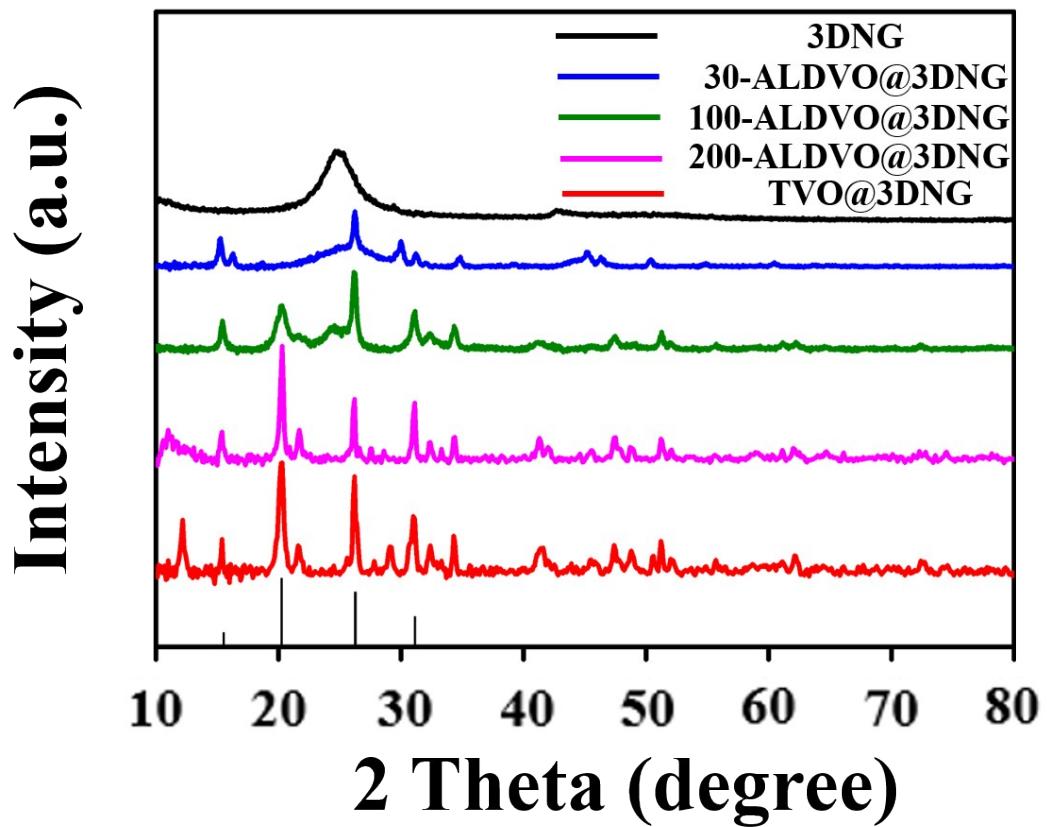
**Fig. S4.** TEM mapping analysis of the remaining elements of 100-ALDVO@3DNG cathode.



**Fig. S5.** (a) linear analysis in the cross section of 100-ALDVO@3DNG cathode. (b, c) V element EDS mapping analysis of 100-ALDVO@3DNG.



**Fig. S6.** (a) HRTEM image of 100-ALDVO@3DNG inner graphene surface, (b) enlarged image in red box of (a); (c) SEAD pattern of 100-ALDVO@3DNG.



**Fig. S7.** Comparison of XRD curves of different samples.

**Table S1.** Electrochemical performance comparison of various ALD process applied in Li-S batteries.

Materials	Rate (1 C = 1675 mA g <sup>-1</sup> )	Capacity (mAh g <sup>-1</sup> )	Ref.
<b>V<sub>2</sub>O<sub>5</sub>/3DNG</b>	<b>0.2 C</b>	<b>1341.8(100 cycles)</b>	<b>This Work</b>
	<b>0.5 C</b>	<b>1102(100 cycles)</b>	
	<b>2 C</b>	<b>542(350 cycles)</b>	
	<b>3.86 mA cm<sup>-2</sup></b>	<b>1204(Initial)</b>	
Al <sub>2</sub> O <sub>3</sub> /C-S	0.5 C	646(100 cycles)	33
	2 C	540(Initial)	
V <sub>2</sub> O <sub>5</sub> /CNTs	0.1 C	1209(Initial)	38
	0.2 C	800(Initial)	
Al <sub>2</sub> O <sub>3</sub> /C	0.2 C	1136(Initial)	34
NG/S-TiO <sub>2</sub>	0.1 C	1374(Initial)	37
	1 C	918.3 (500 cycles)	
Al <sub>2</sub> O <sub>3</sub> /C	0.2 C	423	32

**Table S2.** Comparison of areal capacity of the 100-ALDVO@3DNG cathode with that of recent publications in Li–S batteries which have high sulfur loadings more than  $4 \text{ mg cm}^{-2}$

Mass loading ( $\text{mg cm}^{-2}$ )	Rate ( $\text{mA cm}^{-2}$ )	Areal capacity ( $\text{mAh cm}^{-2}$ )	Ref.
<b>11.5</b>	<b>3.86</b>	<b>14.9(Initial) 11.2(100 cycles)</b>	<b>This work</b>
5	1.68	5.1(Initial) 4(50 cycles)	52
8.1	3.4	7.69(Initial) 6.8(50 cycles)	53
4.2	0.35 2	4.8(Initial) 2.44(60 cycles)	54
10.2	1	10.8(Initial) 6.0(50 cycles)	55
5.4	4.52	5.43(Initial) 4.3(200 cycles)	56
12	4.02	10.2(Initial) 5.04(100 cycles)	57
10.02	3.41	6.52(Initial) 5.14(500 cycles)	58
4	5.8	2.8(Initial) 2.08(500 cycles)	59
4	0.67	3.33(Initial) 1.68(100 cycles)	60
4.2	0.7	5.707(Initial) 4.09(160cycles)	61
6	1.01	6.648(Initial) 4.25(200 cycles)	62
12.1	4.05	12.6(Initial) 11.1(30 cycles)	63
5	0.83	4.87(Initial) 2.56(50 cycles)	64
8	2.68	9.6(Initial) 7.72(60 cycles)	65