

Supplementary Information

Cross-linker Mediated Formation of Sulfur-functionalized V₂O₅/Graphene Aerogels and Their Enhanced Pseudocapacitive Performance



Figure S1. Mass of the VOGS aerogel in 2 cm diameter x 2 cm height.

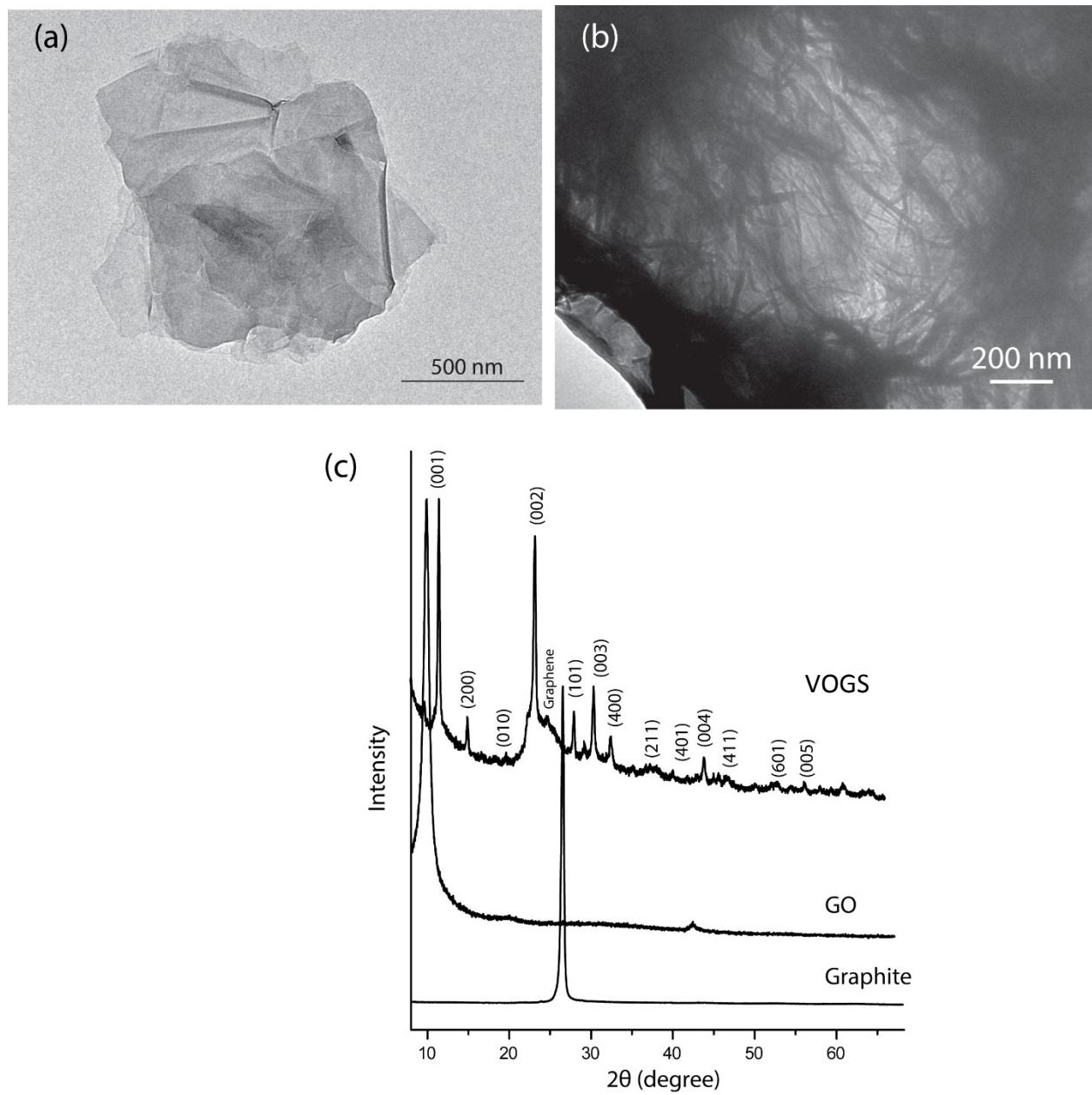


Figure S2. TEM image of the (a) graphene oxide and (b) VOGS aerogel. (c) XRD patterns for graphite, GO and VOGS.

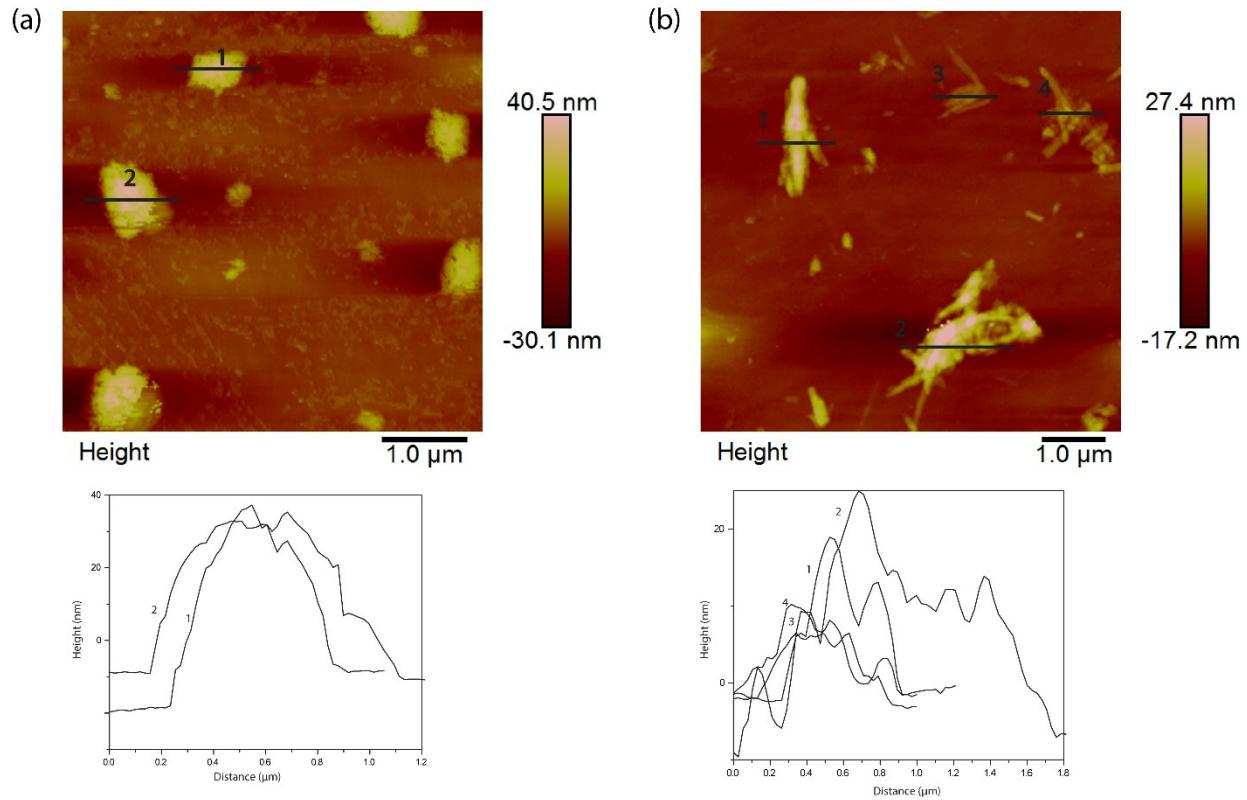


Figure S3. AFM images of the (a) VOGS and (b) VOG.

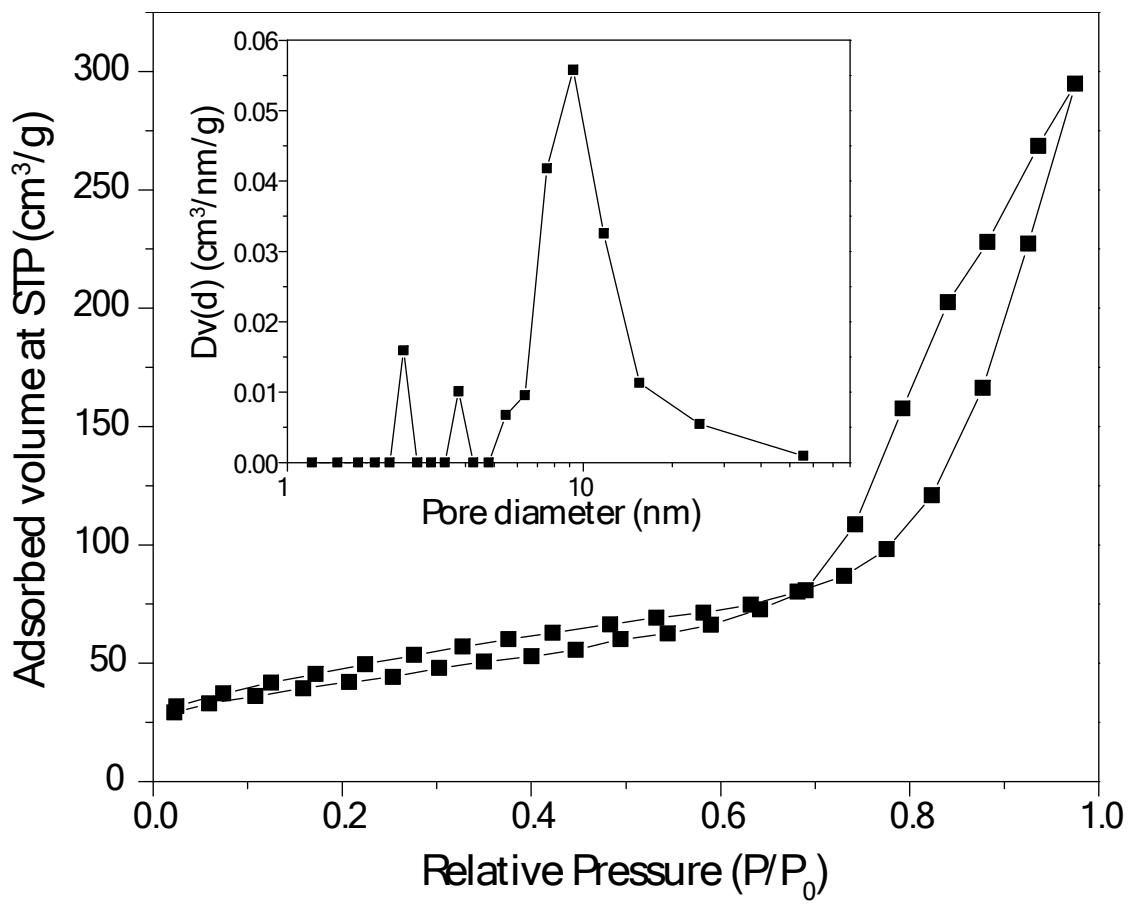


Figure S4. Nitrogen adsorption-desorption isotherms and BJH desorption pore size distribution of VOGS.

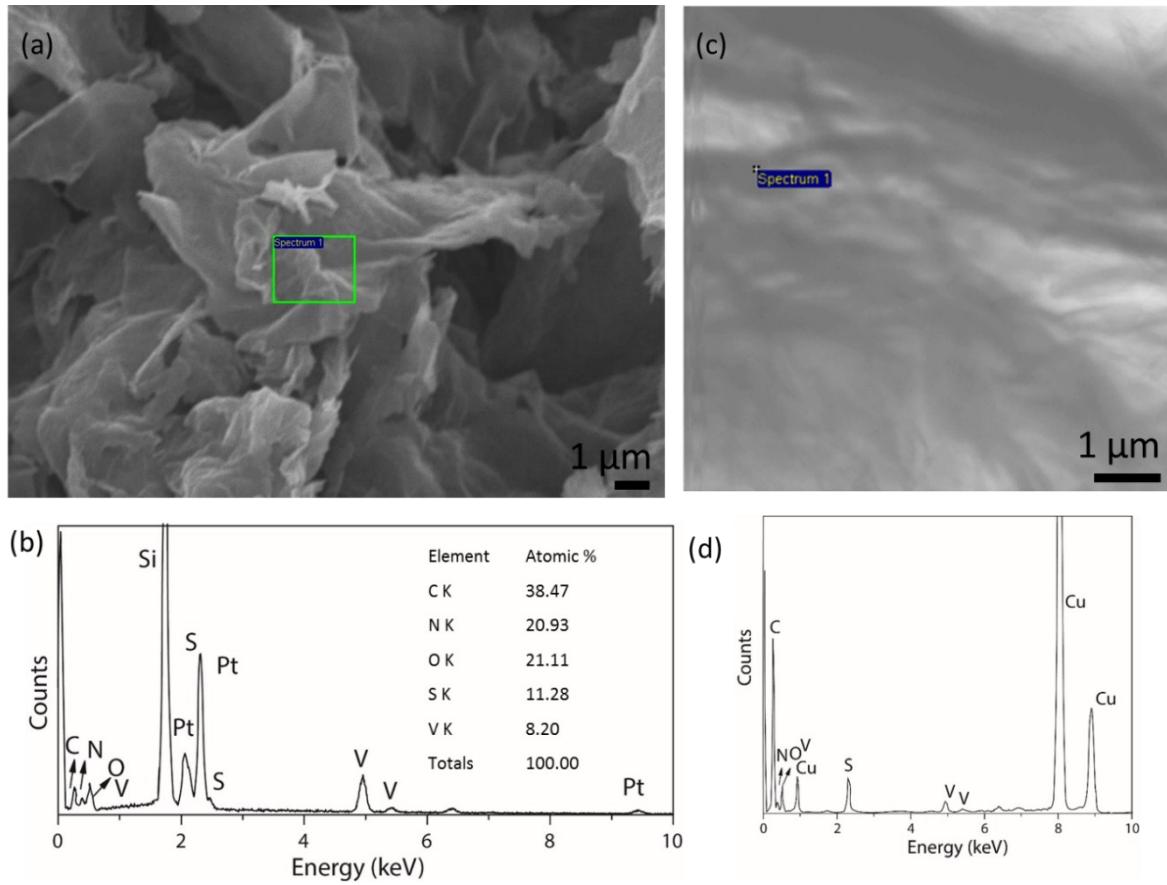


Figure S5. (a) SEM and (c) TEM-EDX images of the VOGS and the corresponding EDX spectra for (b) the square region in SEM image and (d) the point in TEM image.

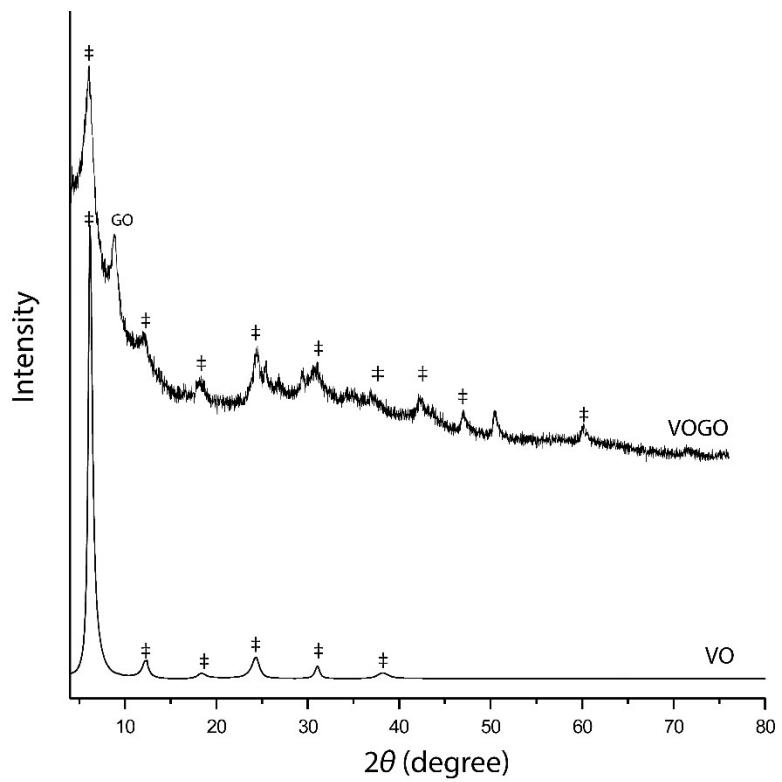


Figure S6. XRD patterns for VO and VOGO.

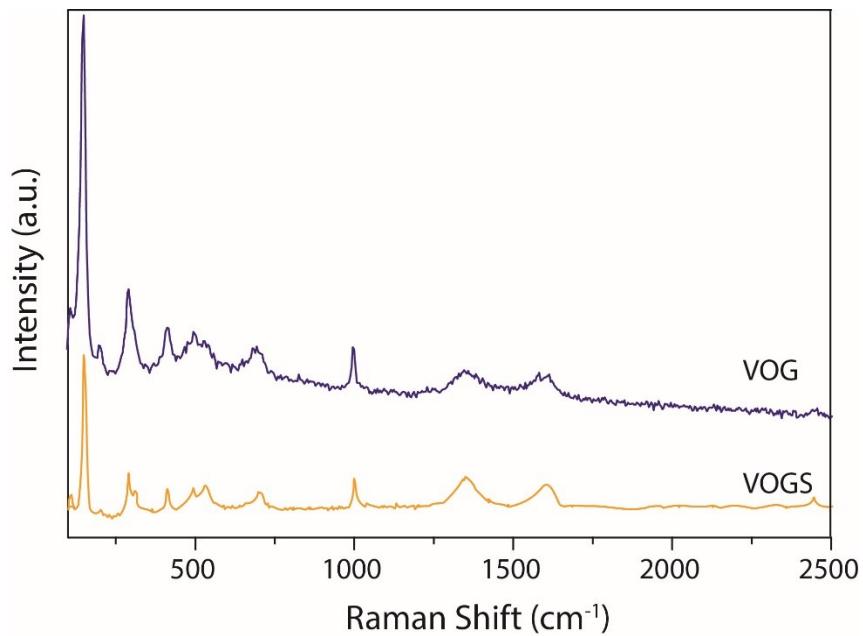


Figure S7. Raman scattering spectra of VOG and VOOGS aerogels.

Figure S7 shows the Raman spectra of VOG and VOOGS. Both VOG and VOOGS exhibit bands at 147, 200, 287, 410, 496, 690 and 1000 cm⁻¹, presenting the characteristic peaks for stretching and bending vibration modes of V₂O₅.^{1,2}

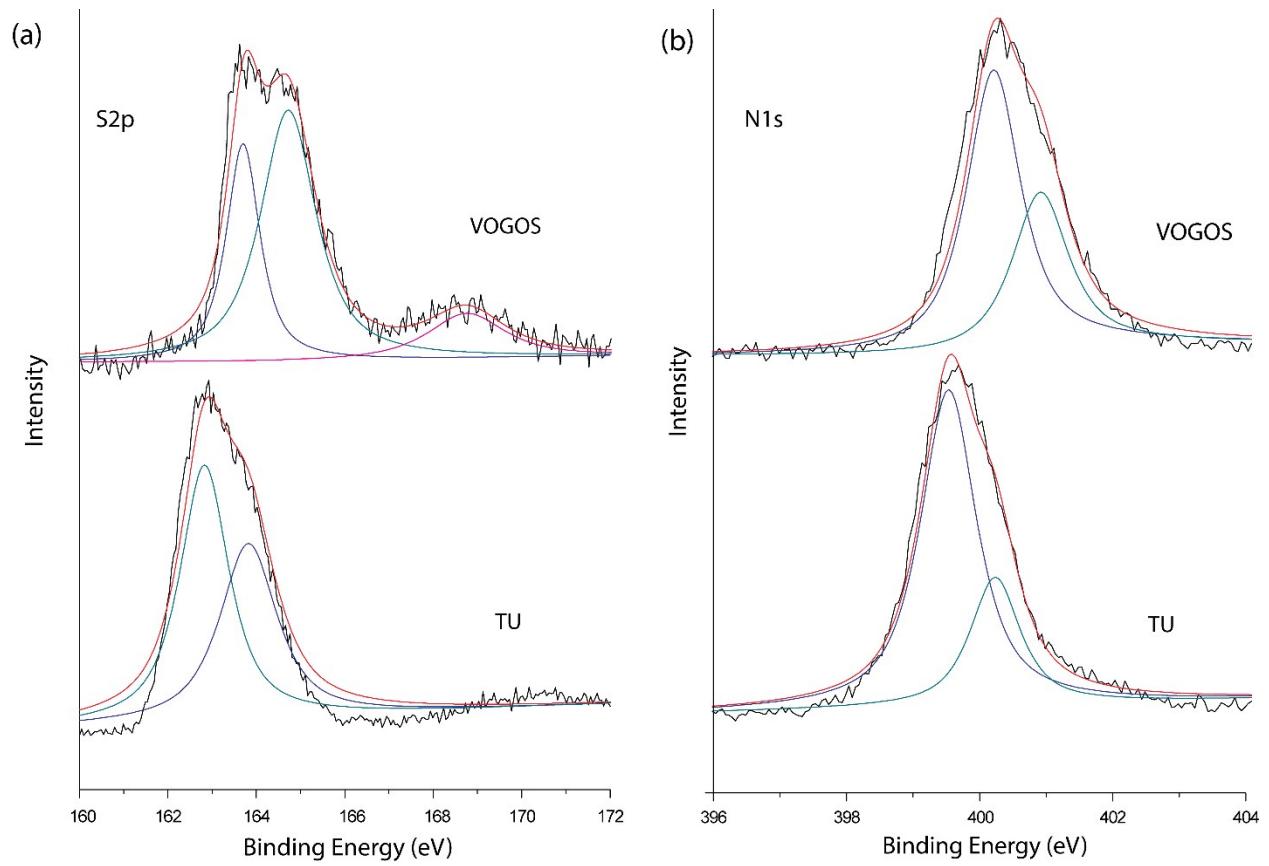


Figure S8. High-resolution (a) S2p and (b) N1s XPS spectra for TU complex and VOGOS aerogel.

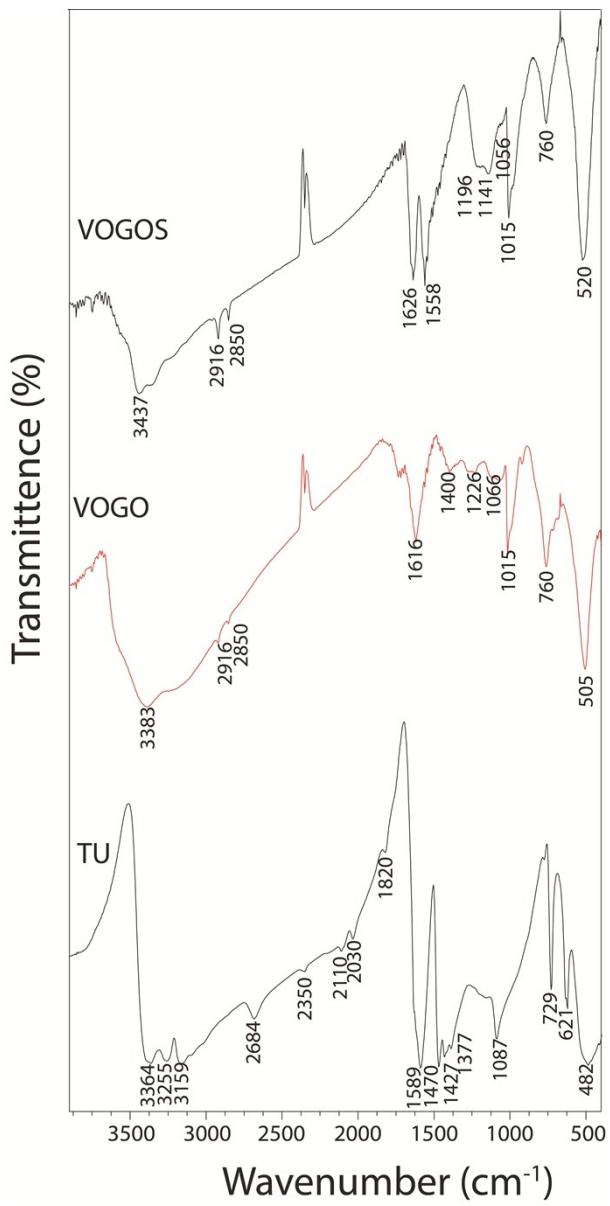


Figure S9. FTIR spectra of TU complex, and VOGO and VOGOS aerogels.

Figure S9 shows the FTIR spectroscopy of TU, VOGO and VOGOS. In the TU spectrum, the strong characteristic peaks belonging to C=S stretching bands at 729 and 1427 cm⁻¹ are weakened and/or disappeared in VOGOS spectrum. The disappearance/weakening can be explained by the reduced double bond character of the C=S bonds in thiourea-based radical

groups during redox reactions.³ Moreover, the sharp NH bending vibration at 1589 cm⁻¹ and N-C-N stretching frequency at 1470 cm⁻¹ in TU and are weakened and shifted to 1626 cm⁻¹ and 1558 cm⁻¹, respectively.⁴ The red shift can indicate the formation of C=N bond.⁵ Similarly, the appearance of N-H stretching absorption bands (~3000 cm⁻¹) at higher frequencies in the VOGOS spectrum compared with TU is another indication of the C=N double bond formation. These findings demonstrates the weakened and dominated double bond characteristics of C=S and C-N, respectively.^{5, 6} For V-O binding, the VOGO has the peaks at 505 cm⁻¹ and 760 cm⁻¹ assigned to V-O-V stretching modes and the peak at 1015 cm⁻¹ corresponds to the V=O stretching. A difference in the spectrum of VOGO and VOGOS appears in the peak around 500 cm⁻¹, which further suggests the higher absorption frequencies for TU cross-linked aerogel due to highly polar nature of nitrogen to carbon bond. Another interesting finding is the appearance of a new band at 1616 cm⁻¹ associated with the H-O-H bending since VOGO structure consist of a large number of interlaying water molecules similar to V₂O₅ gel (Figure S6). The graphene oxide peaks in VOGOS and VOGO mainly appeared at 1400 cm⁻¹ (O-H deformation vibration), 1226 cm⁻¹ (C-OH stretching vibration) and 1050-1060 cm⁻¹ (C-O stretching vibrations).⁷

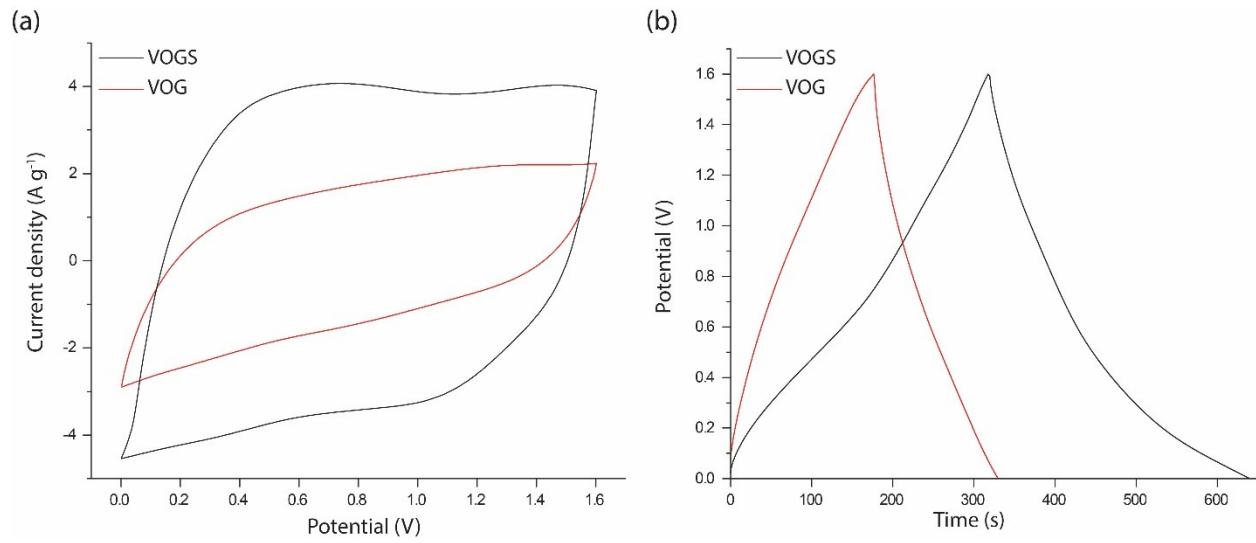


Figure S10. (a) CV curves at 30 mV s^{-1} and (b) CD curves at 0.6 A g^{-1} for VOGS and VOG.

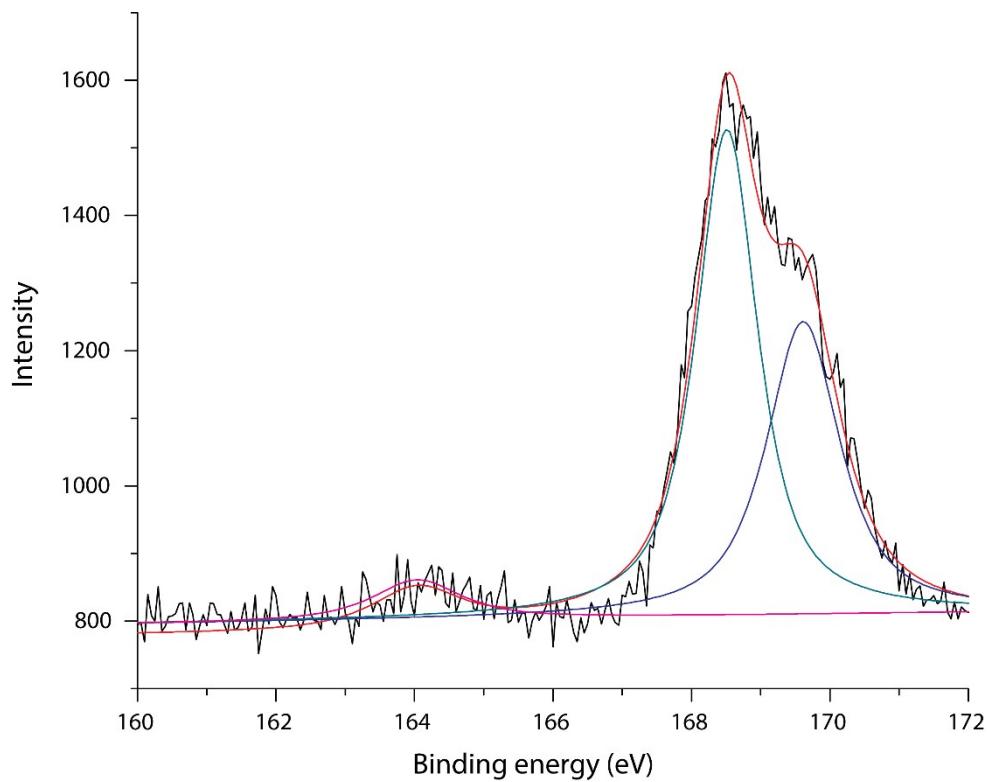


Figure S11. (a) S2p and XPS spectra for VOGS after annealing.

Table S1. Volumetric capacitance values for VOGS, calculated using the density of aerogel (20mg/cm³).

Current Density (A/cm ³)	Volumetric Capacitance (F/cm ³)
0.012	9.68
0.025	9.10
0.05	8.05
0.1	6.84
0.2	5.44

References

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