Supplementary Information for

"A Hybrid Aerogels of Co-Al Layered Double

Hydroxides/Graphene with Three-dimensional Porous Structure

as a Novel Electrode Material of Supercapacitors" by

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Fig. S1 SEM images of (a) and (b) Co-Al LDHs/GHAs-15.2% and (c) and (d) Co-Al LDHs/GHAs-10.6%.

With the purpose of confirming the effects of GAs content on the morphologies of Co-Al LDHs/GHAs composites, SEM of Co-Al LDHs/GHAs-10.6%, and Co-Al LDHs/GHAs-15.2% are also inspected and the corresponding images are exhibited in Fig. S1. It can be clearly observed in Fig. S1 a and b that because of the high GA content in Co-Al LDHs/GHAs-15.2%, 3D structure has been well maintained but the existence of Co-Al LDHs in this composite is not enough. As shown in Fig. S1 c and d, for Co-Al LDHs/GHAs-10.6%, because of the lower content of GA and higher content of Co-Al LDHs, 3D structure cannot well maintained and obvious aggregation appeared obviously.



Fig. S2 The charge-discharge curve of (a) Co-Al LDHs/GHAs-15.2% and (b) Co-Al LDHs/GHAs-10.6%.

In order to ensure the effects of GAs content on the electrochemical performance of Co-Al LDHs/GHAs composites, the charge-discharge curve of Co-Al LDHs/GHAs-15.2% and Co-Al LDHs/GHAs-10.6% at various current densities are also detected and exhibited in Fig. S2. It can be clearly observed in Fig. S2 that the specific capacitance of Co-Al LDHs/GHAs-15.2% and Co-Al LDHs/GHAs-10.6% at 1 A g⁻¹ is 294 F g⁻¹ and 425 F g⁻¹, respectively. At 2A g⁻¹, their specific capacitance decay to 160 F g^{-1} and 236 F g^{-1} , respectively. When compared with the target 3D Co-Al LDHs/GHAs-13.7%, further increase or decrease of GAs loading both result in a decrease of specific capacitance. This can be attributes to many factors: when the loading content of GAs in Co-Al LDHs/GHAs too much (Co-Al LDHs/GHAs-15.2%), the pseudocapacitance, which produces from Co-Al LDHs, of the composite will obviously decreases; as a result, the Co-Al LDHs/GHAs shows a low specific capacitance. When the loading content of GAs in Co-Al LDHs/GHAs too little (Co-Al LDHs/GHAs-10.6%), it does not favor the formation of effectively dispersed Co-Al-LDH layers on GAs and the 3D structure becomes unobvious, which results in the low utilization of Co-Al-LDH. That is to say, if the loading content of GAs in Co-Al LDHs/GHAs is suitable, the structural advantage of the composite can be reflect apparently, as a result, the electrochemical properties of the composite will obviously improved.



Fig. S3. Large-multiple SEM images of 3D Co-Al LDH/GHAs in some selected regions.

In order to further confirm the composite state of Co-Al LDH and grapheme areogels in3D Co-Al LDH/GHAs compounds, large-multiple SEM image of 3D Co-Al LDH/GHAs in some selected regions has been supplemented in Fig. S3, the close observation of the composite is shown in Fig. S3 and shows that Co-Al LDHs are uniformly dispersed on every area of graphene areogels nanosheets, indicating efficient assembly between Co-Al LDHs and graphene layers.