Response to Comment on *Sponge-Templated Preparation of High Surface Area Graphene with Ultrahigh Capacitive Deionization Performance*

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The comment from Porada et al. on our paper entitled "Sponge-Templated Preparation of High Surface Area Graphene with Ultrahigh Capacitive Deionization Performance" raises some concerns on the expression, structural advantage of sponge-templated graphene sheets (STGS), capacitive deionization performance, geometrical information of the CDI cell, and desalination testing details. We appreciate Porada et al. for their interest in our work, as well as for their useful suggestions regarding our paper. However, we do not concur with part of the raised criticism, even though some suggestions help us to improve the quality of the paper. Here are detailed responses to the issues raised by Porada et al.:

1. Porada et al. claimed the terminology "high surface area" is not accurate. The effective surface area of our sponge-templated graphene sheets (STGS) as measured by the N_2 absorption Brunauer-Emmett-Teller (BET) method is 305 $m^2\ g^{-1}$. As comparison, the sample of GN prepared without sponge template was calculated to be 178 $m^2\ g^{-1}$. Therefore, the effect of the sponge template on increasing the surface area has been clearly demonstrated. Although the theoretical value for the surface area of graphene sheets is estimated to be 2600 $m^2\ g^{-1}$,[1] we have to note that achieving significantly high specific surface area of graphene materials is generally difficult, especially for bulky GN samples. We have compared our results with several published works, and found that the specific surface area of STGS is obviously higher than some other reports.[2] Thus, we feel that it is appropriate to describe our STGS with the word of "high surface area", despite the fact that a 800–1000 $m^2\ g^{-1}$ value was previously reported by Cheng et al.
2. According to several cited references, Porada et al. conclude that pore size distribution is important for CDI test, and believe that materials with solely meso- and macroporous are not most favorable for CDI electrodes. We strongly agree with the viewpoint that an appropriate porous structure benefits the CDI performance of carbon materials. However, we have to note that the optimal structure of carbon materials for best CDI performance is still controverse. Previous studies have demonstrated that the adsorption capacity of the carbon aerogels is mainly affected by electrical double-layer capacity due to the electrostatic attractive force between the ions and the electrode.[3] It is known that the electrical double layers form inside the pores of the porous electrodes. Also, previous studies have demonstrated that the electrical double layer inside the pores should overlap when the pore size is of similar magnitude to the electrical double layer thickness, therefore resulting in a loss of electrical capacity and consequent limitation of the mass transfer.[3a,4] Such an overlapping effect exists in microporous (<2 nm) and part of mesoporous (2–50 nm) regions.[4] Based on these studies, it is suggested that the meso- and macroporous structure obtained in our STGS should be favorable for CDI application. Therefore, we feel that the meso- and macroporous structure of STGS should be favorable for CDI process.
3. Porada et al. think that the electrosorption capacity of our STGS is not enough high. It is known that the reported values for salt adsorption vary as a factor of applied cell voltage, initial salt concentration, setup design, and calculation method. Particularly, large values for salt adsorption can be easily obtained with a high initial salt concentration, as listed by Porada et al. Here, we note that the adoptive initial salt concentration in our work is of about 50 mg L^{-1}. Based on a similar initial salt concentration, we could compare our results with the previously reported data, and it is easy to find that the electrosorption capacity of our STGS is high enough. More importantly, we emphasize that, in this work, the STGS possess obviously higher electrosorption capacity than the GN prepared without template.
4. We agree that the metric of desorption time depends on electrode architecture and system setup. As a result, the desorption time is not a normalized parameter to estimate the electrode performance. We referred the "significant desorption rate of 25 min" to that of GN. Nevertheless, we note that the desorption time of 25 min for STGS is also shorter than that found in some previous work.[5]
5. For clear understanding the CDI data, the structure and geometrical information of the CDI cell and electrode are presented in **Figure 1**. The CDI cell consists of two plexiglass half-cell, tightly fixed with six rivets. The electrode thickness is about 200 μm and the separation distance between the electrodes is about 600 μm. The mass of active materials on one electrode is 20 mg and the size of the effective electrode is 50 mm × 50 mm.
6. We agree that the testing in 6 M KOH is less relevant for CDI. We note that the experimental data was kindly suggested and required by the reviewers. Meanwhile, we noticed that

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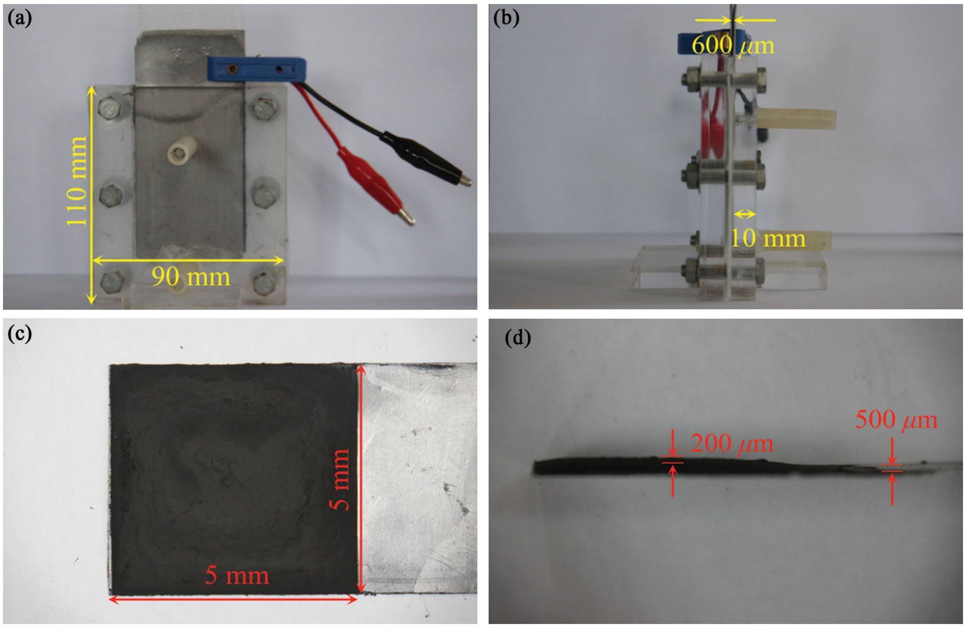

Figure 1. A,B) Front and side view of the CDI cell; C,D) The image of the front and side view of STGS electrode.

porous carbon electrodes and desalination cells behave very differently when operated at 1 M NaCl compared with 1 mM NaCl. However, the laboratorial yield of STGS in this work is quite low. Considering this factor, it is not possible to acquire a larger-sized STGS electrode. In our case, the size of the effective electrode is 50 mm × 50 mm, with a STGS loading of 20 mg. As a consequence, the little amount of STGS on the electrode can only afford low total adsorption capacity. Thus, when high initial salt concentration was adopted for our CDI system, the variation of conductivity was too small to be detected. Specifically, a 20 mL 1 M NaCl solution contains 1170 mg NaCl with a conductivity of 85.5 mS cm^{-1}. Assuming that the adsorption capacity is 4.95 mg g^{-1} for STGS, the maximum amount of 0.198 mg NaCl should be adsorbed during one cycle. Even with a very low flow rate, the variation of the conductivity was not detectable. **Figure 2** shows the recorded curves of the conductivity variation during the CDI process in 1 M NaCl with an applied potential of 1.5 V for STGS electrode. As seen, no detectable conductivity variation was observed. Considering the cell size and electrode capacity, in this work, we adopted 50 mg L^{-1} of NaCl solution with a conductivity of 106 µS cm^{-1} as our initial conditions.

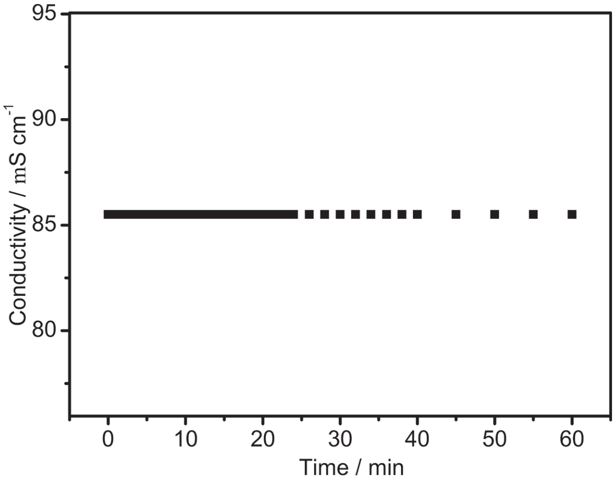

Figure 2. Variation of the conductivity during the CDI process in 1 M NaCl with an applied potential of 1.5 V for STGS electrode.

In conclusion, we thank Porada et al. for their special concern with this paper. We hope the supplied data and result will help the reader to better understand our work. Also, we refer the reader to the Supporting Information of the paper and some useful cited references to clarify our viewpoints.

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