Supplementary Material for **"Effect of Oxide Content of Graphene Oxide Membrane on Remarkable Adsorption for Calcium Ions"**

PS1: Method

GO suspension with the concentration of ~4 mg/ml was prepared by the modified Hummers method. Ten-minutes' ultrasonic treatment with the frequency of 70 kHz has been performed to enhance the dispersion of GO sheets. After that, 1 ml GO suspension formed the GOMs via the drop-casting method followed by the drying process at 60℃ for 8 hours. The obtained GOMs are immersed in the 40-ml $CaCl_2$/NaCl solutions of various concentrations at room temperature. The extra $CaCl_2$ and NaCl solution adsorbed on the surface of the GOMs also had been removed by the absorbent paper. Then the obtained $CaCl_2$@GOMs were dried at 60℃ for 12 hours.

The metal ions in the GOMs were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES). The desorption amount of $CaCl_2$/NaCl during immersing $CaCl_2$@GOMs in DI water had been detected by the concentration variation in the solution by using conductance meter.

The prepared $CaCl_2$@GOMs samples were characterized by Siemens XRD and LEO 1530VP SEM operated at 10 kV.

PS2: Reusability of GO membranes

In industrial applications, reusability is used to evaluate the feasibility of an adsorbent. Their reusability was investigated by adsorption and desorption experiment repeated for several times. After adsorption, the GO membranes were desorbed DI

water, and then dried. The GOMs were then placed in a $CaCl_2$ solutions. The adsorption capacity for Ca^{2+} decreased slightly with increasing treatment cycles, but remain 0.425 g/g in the fourth cycle relative. After these cycles, the properties of the GOMs are still stable.

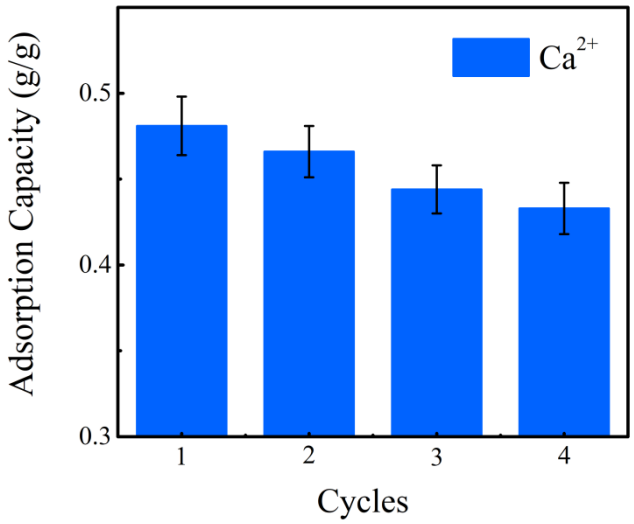

Fig S1: Recycling of GOMs for Ca^{2+} adsorption. Adsorption capacity with respect to the treatment cycles by GOMs. Error bars indicate the standard deviation.

PS3: Case of NaCl@GOMs

The GOMs contained NaCl (NaCl@GOMs) were also prepared via the same synthesis procedure as the case of $CaCl_2$@GOMs. Fig. S2a shows that the adsorption capacity of Na^+ in NaCl@GOMs can be as high as ~0.213 mg/mg after immersing GOMs heat-treated at 60 ℃ in the 6.0 M NaCl solution. It is also shown that the adsorption capacity of Na^+ increases from ~0.099 mg/mg to ~0.213 mg/mg with increases the concentration of NaCl solution from 3.0 to 6.0 M, respectively.

Desorption process of NaCl was also studied via immersing NaCl@GOMs into DI water. The NaCl@GOMs with the adsorption capacity of ~0.213 mg/mg are

chosen to use in our experiments. Similar to the case of $CaCl_2$@GOMs, we find that the desorption process of NaCl can also been divided into three stages as shown in Fig. S2b. The concentration of NaCl in the solution increases rapidly from 0 to 17 ppm in the first 1 min, then slowly to 28 ppm in the range of 1 - 5 min. Afterwards, the concentration of NaCl in the solution becomes stable.

Fig. S2c shows the XRD pattern of NaCl@GOMs and there are several Bragg peaks at different diffraction angles (2θ). The peak at ~11 degree for pristine GOMs and ~12 degree for NaCl@GOMs come from the GOMs. The weak peak at ~28 degree and strong peak at ~32 degree come from the (111) and (200) surface of the ordinary NaCl crystal in the NaCl@GOMs. SEM image shows extensive distribution of NaCl inside NaCl@GOMs, as shown in Fig. S2d. Fig. S2e shows elemental mappings of C, O, Na, Cl elements. We find that the signals of Na and Cl elements in the region covered with large amount of NaCl are shown very high intensities as the intensities of C and O elements, which further confirms the remarkable amount and wide distribution of NaCl in NaCl@GOMs.

Similar to the case of $CaCl_2$@GOMs, the remarkable amount of NaCl adsorbed inside the NaCl@GOMs can also be attributed to strong interactions between NaCl and GO sheets, including the ion-π interactions between Na^+/Cl^- and aromatic graphitic rings as well as the electrostatic interaction between Na and oxide groups. The desorption process of NaCl from NaCl@GOMs is similar to that of $CaCl_2$ from $CaCl_2$@GOMs.

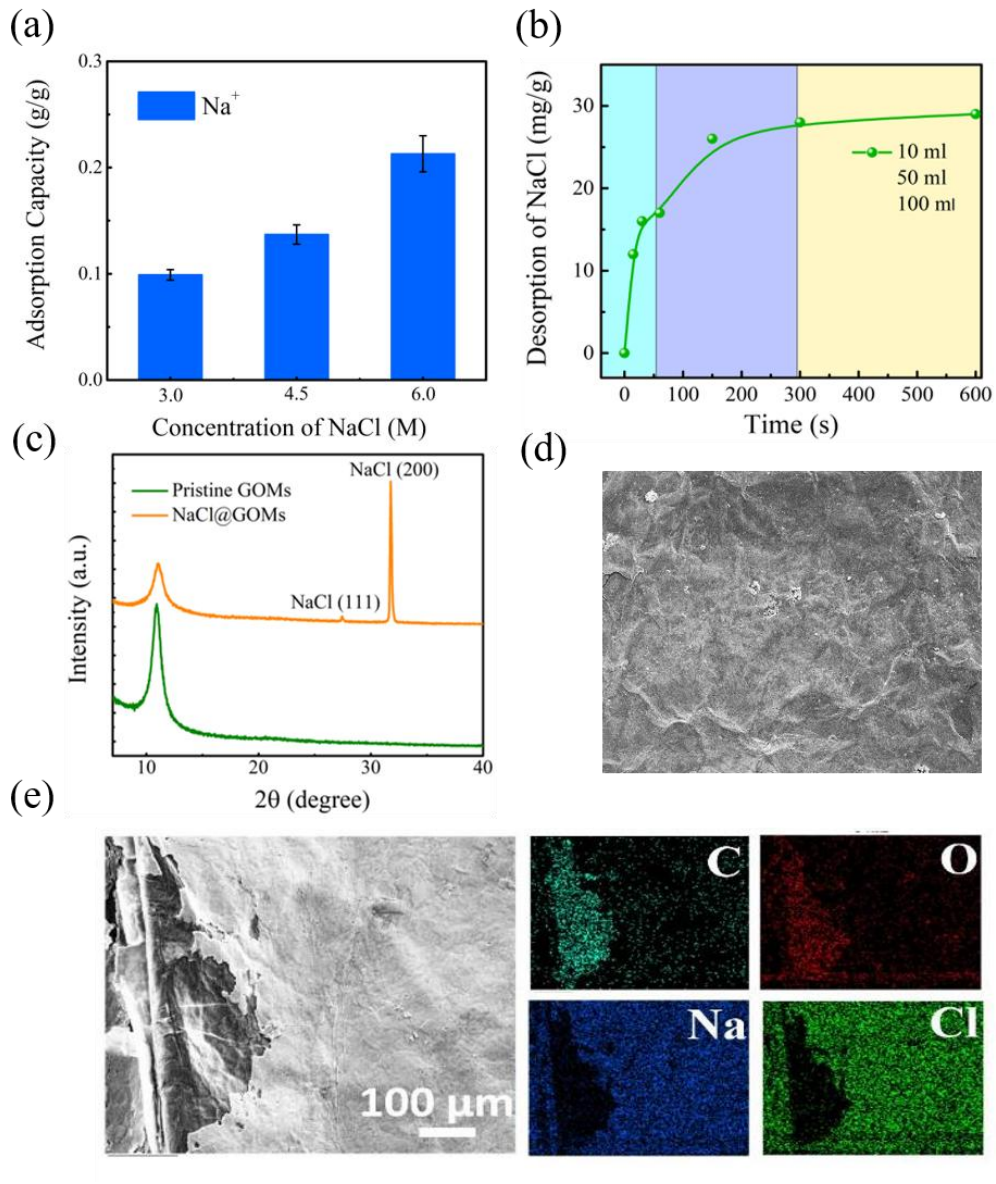

Fig. S2 Case of NaCl@GOMs. (a) Adsorption capacity of NaCl in NaCl@GOMs after immersing NaCl solution. (b) Desorption process of NaCl from NaCl@GOMs after immersing in DI water of 50 ml. The process can be divided into three stages: I. fast desorption stage (cyan-blue region), II. Slow desorption stage (purple region), III. stable stage (yellow region). (c) XRD patterns of NaCl@GOMs (orange line). (d) SEM image of NaCl@GOMs. (e) Elemental mappings of NaCl@GOMs. C, O, Na, Cl elements are displayed as the colors of cyan, red, blue and green, respectively. Error bars indicate the standard deviation.