

Electronic Supplementary Information

Gel adsorbent material composed of a polymer hydrogelator and activated carbon

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1. Insolubilization of NaPPDT

Experiments were conducted to determine the extent of loss (dissolution) of **NaPPDT** in water in the presence of NaCl.

1.1 Experimental Methods

100 mg of B/A 100/0 was weighed and placed in a screw tube. 10 ml each of 1 M NaCl solution, 0.1 M NaCl solution, 0.01 M NaCl solution, and pure water were added, and the tube was kept in a thermostatic bath set at 25°C for 48 hours. The solution portion other than the gel was then collected and the remaining gel was observed. The collected solution was measured for absorbance using a spectrophotometer.

2. Figures

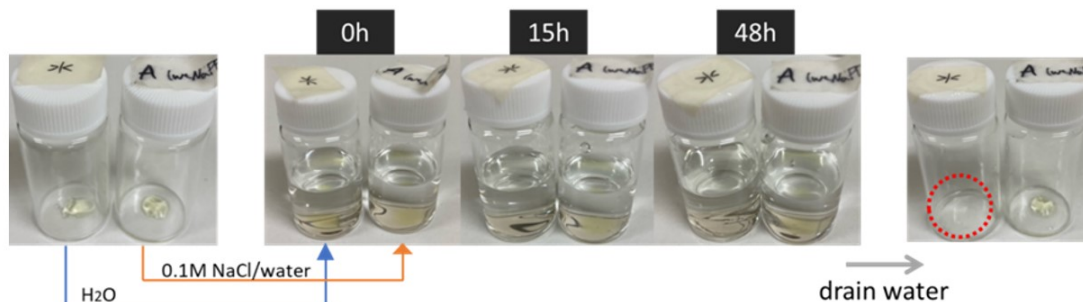


Fig. S1 Solubility test of the hydrogels using a lump of the 1 wt% **NaPPDT** hydrogel in NaCl aq. after 48 hours.

When the solution portion was collected, the gel remained at the bottom in 1 M NaCl solution and 0.1 M NaCl solution with a swollen appearance, and in 0.01 M NaCl solution, a slight thick precipitate was observed. Nothing remained in pure water.

The colour of the collected solution was clear, but absorbance measurements showed a clear difference. 0.1 M and 1 M NaCl solution with gel showed almost the same value as 1 M NaCl solution without gel, there was almost no change in the absorption band around 300 nm. On the other hand, in the pure water sample in which **NaPPDT** was completely dissolved, a large absorption band appeared between 300 and 400 nm. In the 0.01 M NaCl solution, bands were also observed in the same range as in the pure water sample. These absorption band values are consistent with those in previous studies, indicating that **NaPPDT** is not soluble in aqueous NaCl solutions above 0.1 M.¹

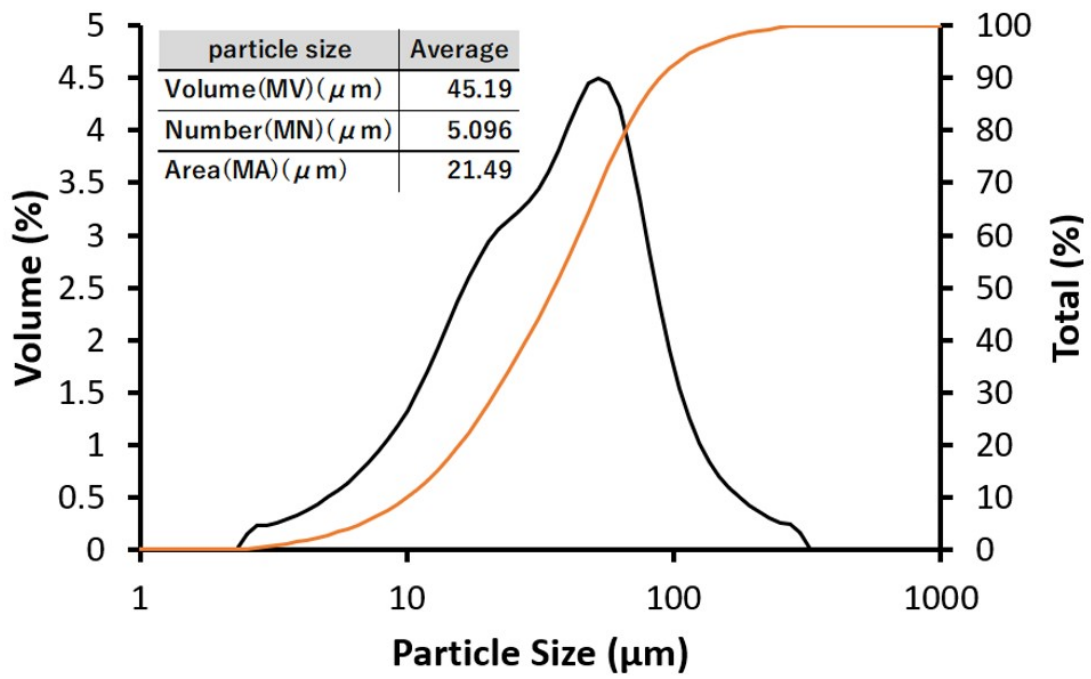
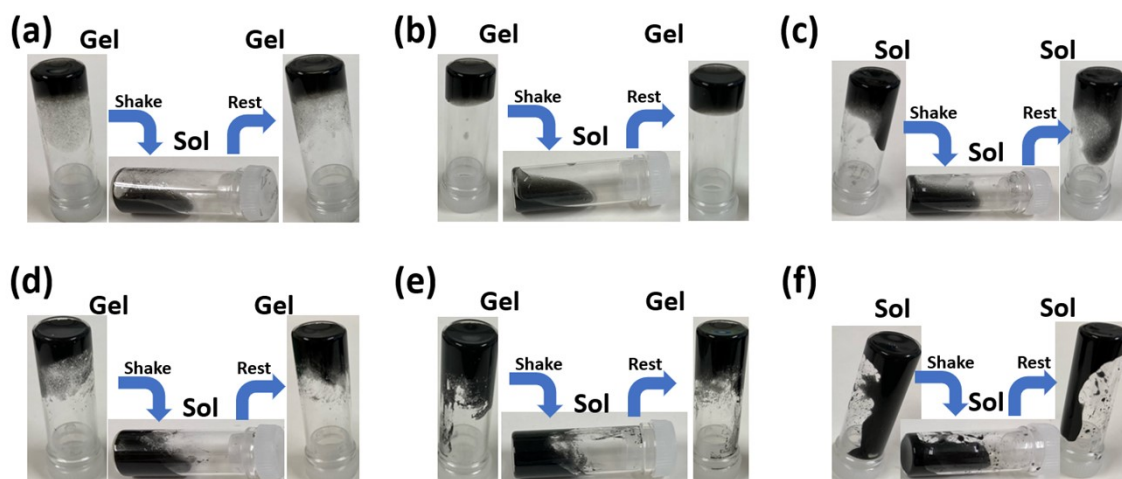


Fig. S2 Particle size distribution and particle size of activated carbon.

Fig. S3 Composite gels with activated carbon in water dispersion, inverted images of composite gels of 1 wt% **NaPPDT** and 1 wt% or 10 wt% activated carbon in water dispersion (a) N/Aw 2/1 (1w), (b) N/Aw 1/1 (1w), (c) N/Aw 1/2 (1w), (d) N/Aw 2/1 (10w), (e) N/Aw 1/1 (10w), (f) N/Aw 1/2 (10w), The resulting **NaPPDT**/activated carbon water dispersion (N/Aw) composite was labelled "N/A (1 wt% **NaPPDT** by weight)/(1 or 10 wt% activated carbon water dispersion by weight) (mixing ratio) (n wt% activated carbon water dispersion)."



Initially, as in previous studies¹⁻³, 1 wt% activated carbon aqueous dispersions were prepared and combined with 1 wt% **NaPPDT** in the ratios of 2/1, 1/1, and 1/2 (N/Aw (1w)). N/Aw 2/1 (1w) and N/Aw 1/1 (1w) were observed to be in the gel state, while N/Aw 1/2 (1w) was completely in the sol state (Fig. S3 (a)-(c)). However, 0.5 wt% **NaPPDT** alone was in a sol state, whereas N/Aw 1/1 combined with activated carbon was in a gel state. This indicates the possibility of strengthening the overall composite by composing activated carbon, a phenomenon that has been confirmed in previous studies.^{1,3}

Secondly, a composite with less water was then attempted. An activated carbon aqueous dispersion solution with a concentration of 10 wt% was prepared and composited with **NaPPDT** (N/Aw (10w)) using the same procedure and concentration as for the 1 wt% (N/Aw (10w)). N/A 2/1 (10w) clearly showed gel morphology, while N/A 1/1 (10w) showed an intermediate N/A 1/1 (10w) appeared to be a viscous liquid, somewhere between a gel and a sol (Fig. S3 (d), (e)). N/A 1/2 (10w) was in a clear sol state.

The reason for the relatively stable gel state in N/A 1/1 (1w) but unstable gel in N/A 1/1 (10w) may be due to the increased complex amount of activated carbon, combined with the decreased concentration of the gelator (**NaPPDT**), which resulted in a sparse gel network structure. However, reducing the amount of activated carbon, which does the work of adsorption, is expected to decrease adsorption performance. In addition, the high-water content may cause the composite gel to dissolve easily in seawater when put into seawater. Therefore, we decided to use a means to composite activated carbon with hydrogel in powder form without dispersing it in water.

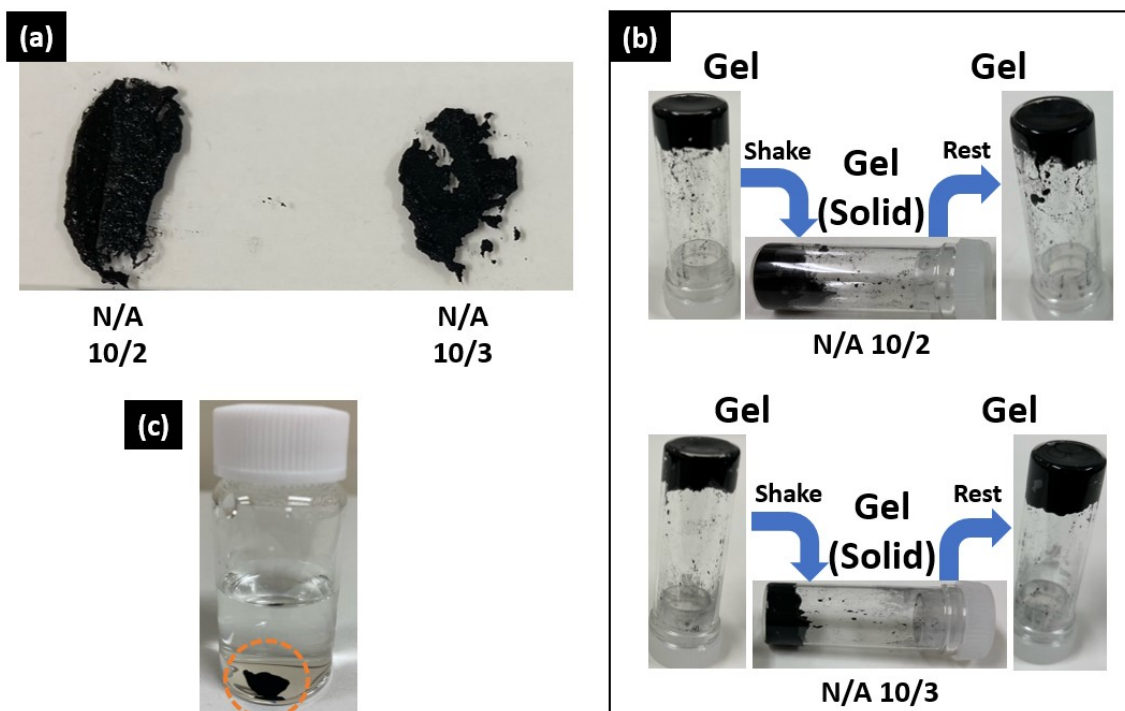


Fig. S4 Composite gel of 1 wt% NaPPDT and activated carbon powder when combined with high concentration of activated carbon: (a) The gelation vial inversion method of N/A 10/2, (b) N/A 10/2 applied to glass plates, (c) N/A 10/2 soaked in 1 M brine for 48 hours.

From the results of Fig. S3, it was confirmed that a better gel state is obtained by combining hydrogel and activated carbon in a way that does not disperse them in water. It is also suggested that the ratio of activated carbon to hydrogel may be very important as a condition for the gel state. As described below, the higher the amount of activated carbon in the composite gel, the more methylene blue can be adsorbed, so increasing the amount of adsorbent in the composite gel as

much as possible has a significant impact on improving adsorption performance. Therefore, we composited activated carbon at a higher concentration than N/A 10/1 and observed the physical properties of the composite gel.

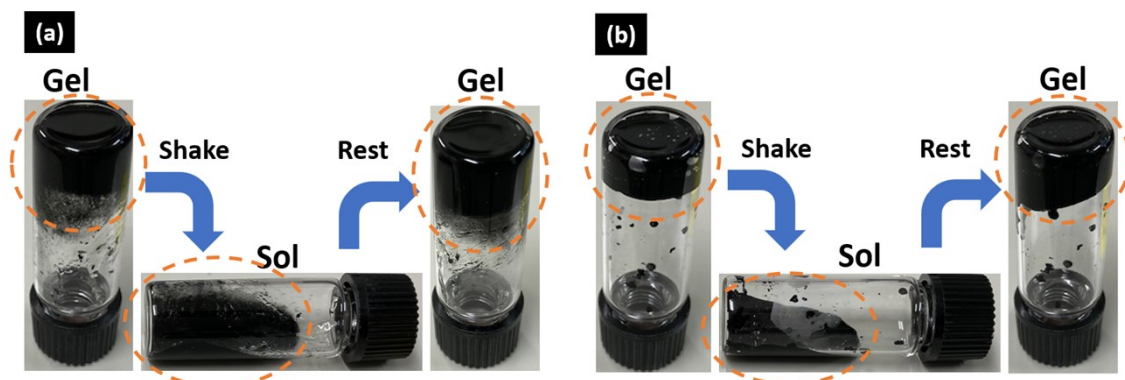
This composite gel was compounded in such a way that the concentrations of N/A 100/1, 100/3, and 10/1 were approximately tripled. N/A 10/3 sample contained so little moisture that it could not be mixed with a vortex mixer alone. When spread out, the paste-like sample was observed to break apart without the smoothness characteristic of gels (Fig. S5a). When the inversion method was applied, no solidification (i.e., a deformation that causes the sample to drip like a liquid) was observed even when an external force was applied (Fig. S5b). This result indicates that N/A10/3 is stable as a solid but has lost its thixotropic property.

N/A 10/2 was then prepared by slightly decreasing the concentration of activated carbon, which caused some tearing when spread, but could barely be spread (Fig. S5a).

N/A 10/2 showed a solid aspect and did not show a liquid state after shaking when the inversion method was performed (Fig. S5b). In other words, N/A 10/2 is a very hard gel. In addition, the composite gels of N/A 100/1, 100/3, and 10/1 could be aspirated with a dropper, but N/A 10/2 could not be aspirated.

N/A 10/2 was placed in 1 M NaCl solution and stirred with a stirrer tip for 48 h. N/A 100/1, 100/3, and 10/1 were formed with a syringe to increase the surface area exposed to aqueous solution during adsorption. However, N/A 10/2 was too hard to be formed by syringe, so it was scooped out with a spoon. As a result, the gel surface did not collapse and the activated carbon did not desorb, as we had feared (Fig. S5c). However, the difficulty of spreading the gel and forming it with a syringe made it different from the composite gel with thixotropic and ductile properties that we were aiming for.

Fig. S5 Photographs of the gelation test (vial inversion method); (a) N/A 100/3, (b) N/A 10/1. (left) Before shaking (gel state), (centre) immediately after shaking (sol state), and (right) after resting for 1 min. The resulting **NaPPDT**/activated carbon (N/A) composite was labelled "N/A (1 wt% **NaPPDT** by weight)/(activated carbon powder by weight) (mixing ratio)."



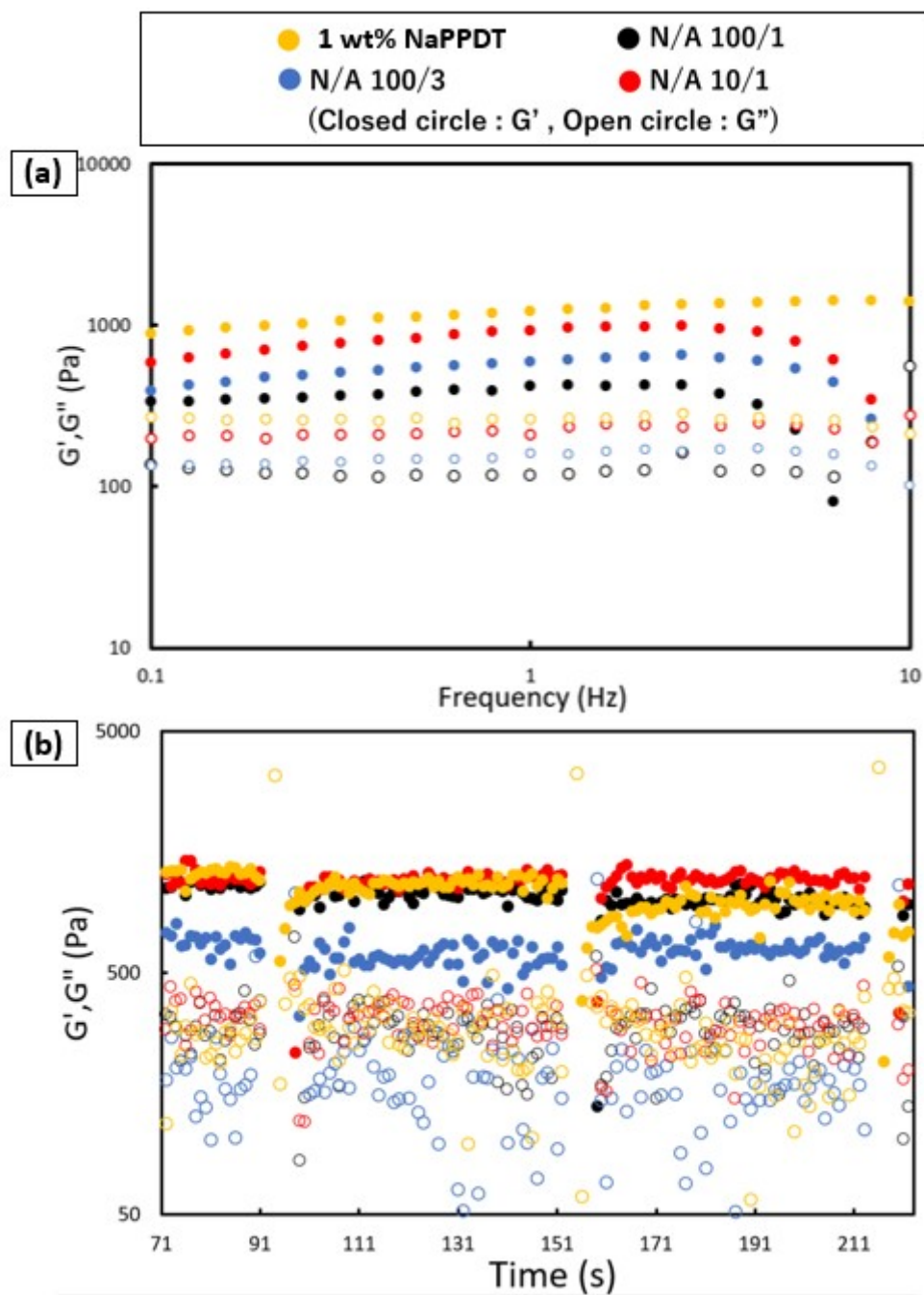


Fig. S6 Rheometric results of composite gels: (a) Frequency sweep, (b) Periodic step-shear test. Part of the result. The resulting NaPPDT/activated carbon (N/A) composite was labelled "N/A (1 wt% NaPPDT by weight)/(activated carbon powder by weight) (mixing ratio)."

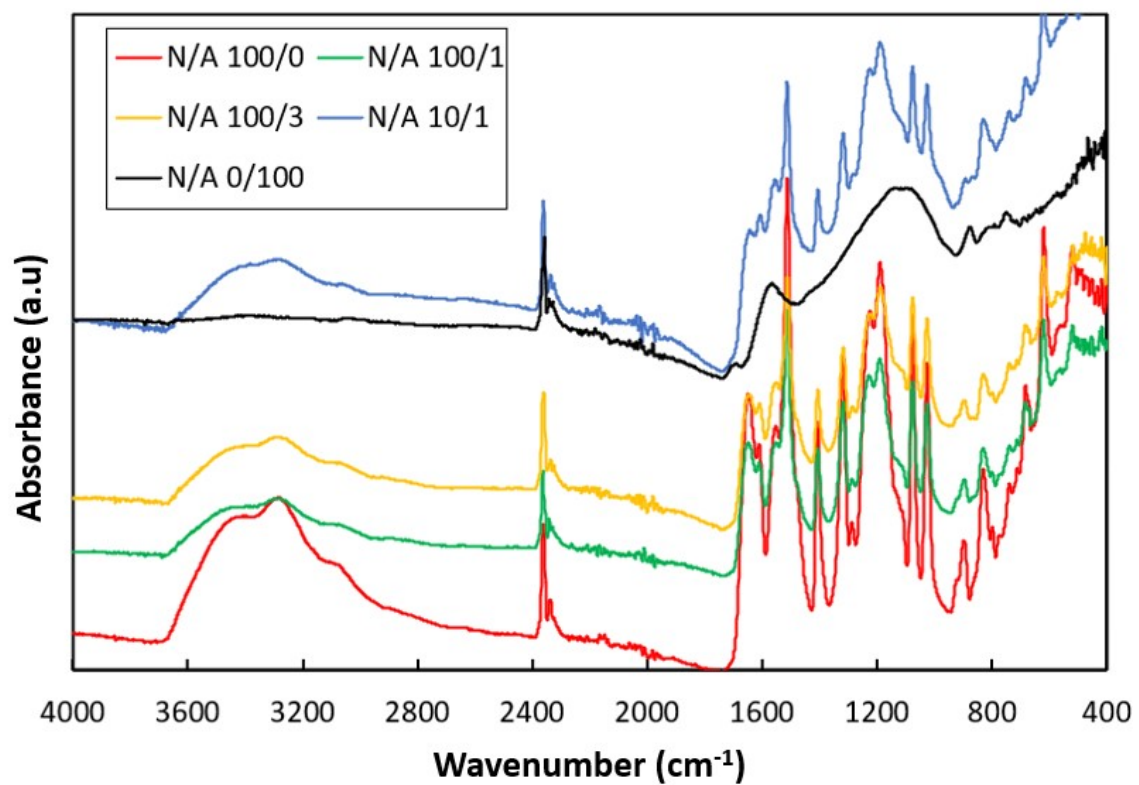


Fig. S7 ATR-FTIR measurement. The resulting NaPPDT/activated carbon (N/A) composite was labelled "N/A (1 wt% NaPPDT by weight)/(activated carbon powder by weight) (mixing ratio)."

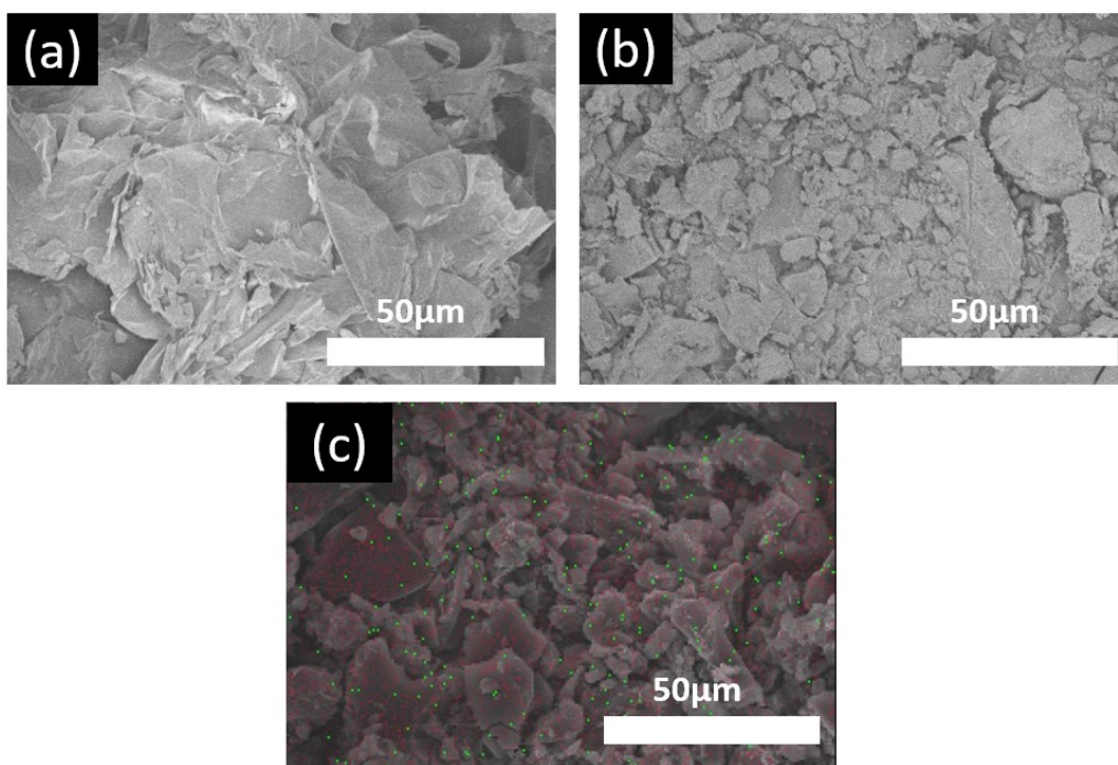


Fig. S8 SEM images: (a) N/A 100/1, (b) N/A 100/3, (c) Elemental analysis (EDS) image of N/A 10/1. (Red dots: C, Green dots: Na). The resulting **NaPPDT/activated carbon (N/A)** composite was labelled "N/A (1 wt% **NaPPDT** by weight)/(activated carbon powder by weight) (mixing ratio)."

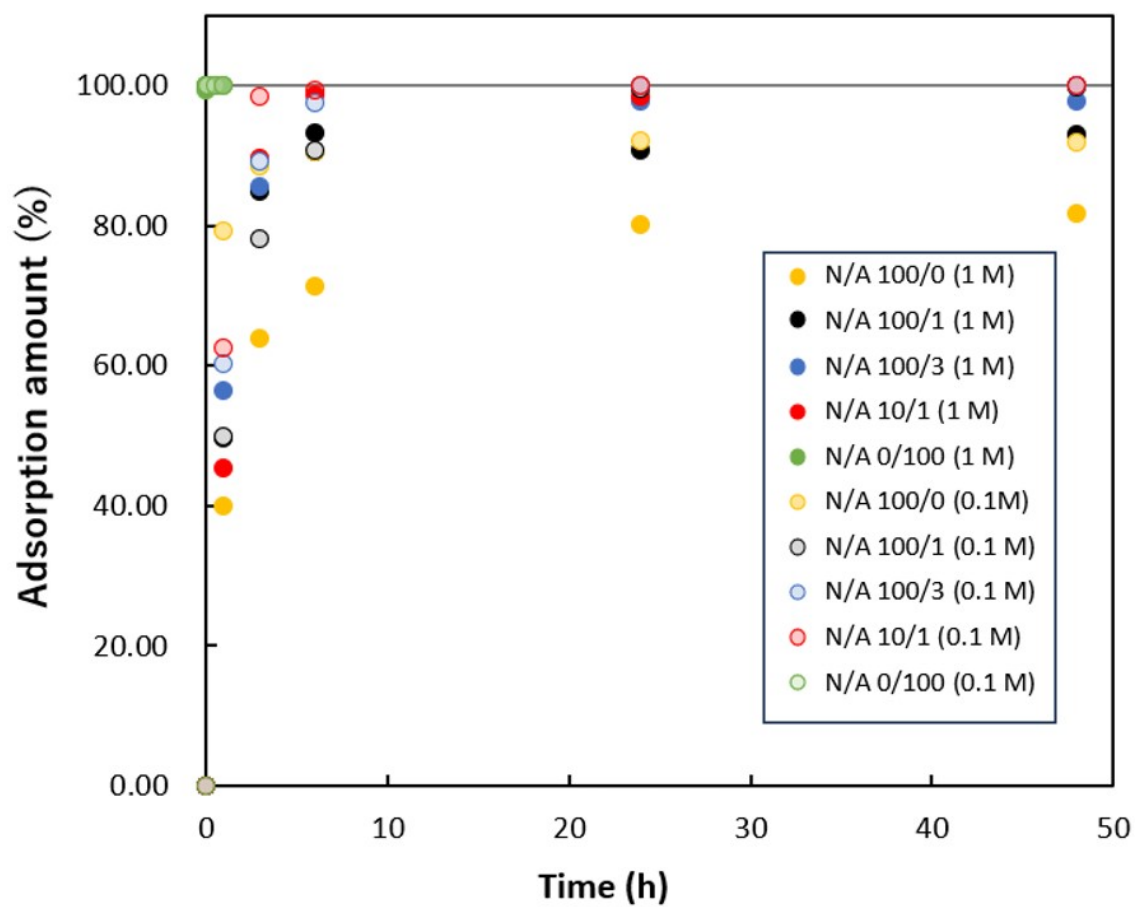


Fig. S9 Amount of each adsorbent adsorbed to methylene blue/1 M and 0.1 M NaCl solution for 48 hours. The resulting **NaPPDT**/activated carbon (N/A) composite was labelled "N/A (1 wt% **NaPPDT** by weight)/(activated carbon powder by weight) (mixing ratio)."

3. References

1. Y. Ohsedo and M. Sasaki, *Gels*, 2022, **8**, 469.
2. Y. Ohsedo, K. Saruhashi, H. Watanabe and N. Miyamoto, *New J. Chem.*, 2017, **41**, 9602–9606.
3. C. Nitta and Y. Ohsedo, *Chem. Eur. J.*, 2024, e202401469.