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Supporting Information for

One-pot preparation of MnO_2/graphene-carbon nanotube hybrid material for the removal of methyl orange from aqueous solutions

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Isotherm Model

The Langmuir model assumes a monolayer adsorption onto a homogeneous surface with a finite number of identical sites. Its linearized form is represented by the following equation [1]:

$$\frac{C_e}{q_e} = \frac{1}{bq_m} + \frac{C_e}{q_m} \quad (1)$$

where C_e (mg L^{-1}) is the solute equilibrium concentration, q_e (mg g^{-1}) is the amount of dye adsorbed per unit mass of the adsorbent at the equilibrium point, and q_m (mg g^{-1}) and b (L mg^{-1}) are the Langmuir constants related to the maximum adsorbate amount and adsorption affinity of the binding sites. When C_e/q_e is plotted versus C_e, the q_m and b constants can be determined from the slope and the intercept. Another parameter, R_L, called separation factor or equilibrium parameter, which can be used to express the efficiency of the adsorption, is defined as [2]

$$R_L = \frac{1}{1 + bC_0} \quad (2)$$

where b (L mg^{-1}) is the Langmuir constant, and C_0 (mg L^{-1}) is the initial MO concentration. The values of R_L express the isotherm shapes which can be unfavorable (R_L □ 1)， linear (R_L = 1), favorable (0 □ R_L □ 1) or irreversible (R_L = 0).

The Freundlich model can be considered to describe heterogeneous surfaces and multilayer adsorption systems, which can be given as follows:[3]

$$ln\ q_e = ln\ K_F + \frac{1}{n}\ ln\ C_e \quad (3)$$

where q_e and C_e have the same definitions as those in the Langmuir equation cited above, and K_F and n are the Freundlich constants that indicate the adsorption capacity and the adsorption intensity, respectively. They can be obtained from the intercept and slope of the linear plot of *ln* q_e *versus ln* C_e.

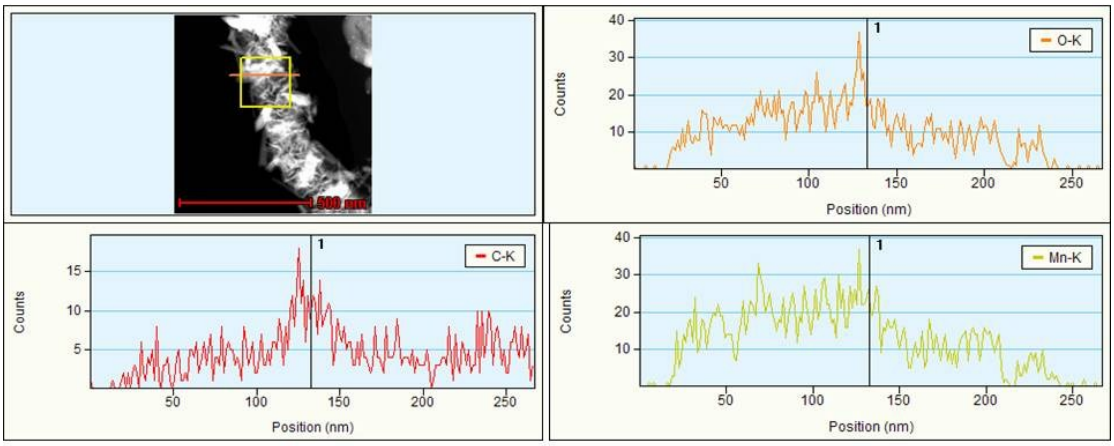

Fig. S1 The line scan results for C, Mn and O elements in the hybrid.

From Fig. S1, we can see that the edge of the curved petal-like sheets (in ~250 nm) is composed of only carbon without Mn and O, implying that the nanosheets are exactly graphene sheets.

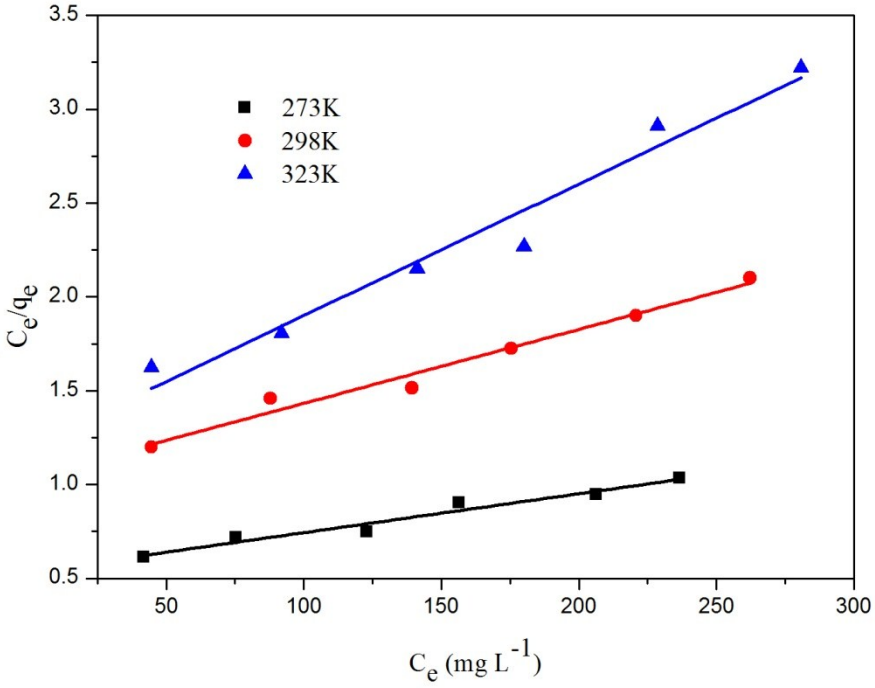

Fig. S2 Langmuir adsorption isotherm plots for MO adsorption onto MnO_2/G-CNT hybrid material.

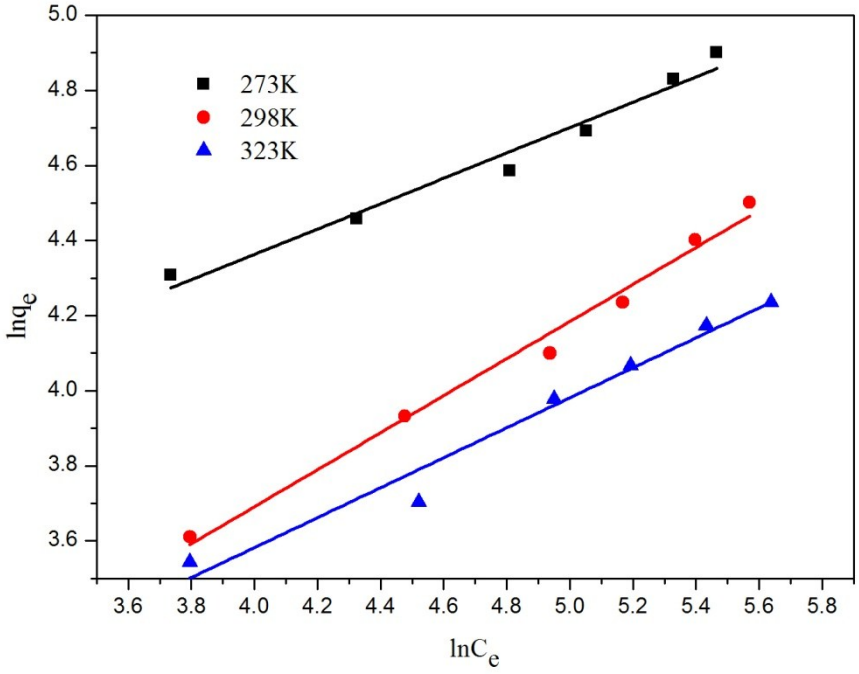

Fig. S3 Freundlich adsorption isotherm plots for MO adsorption onto MnO_2/G-CNT hybrid material.

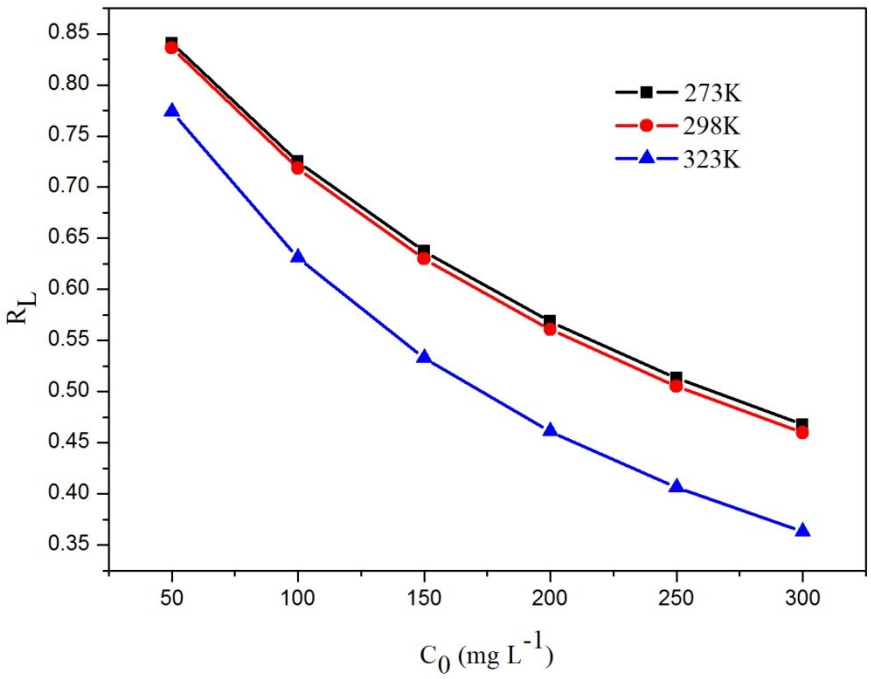

Fig. S4 Separation factor of MO adsorbed onto MnO_2/G-CNT hybrid material at different temperatures.

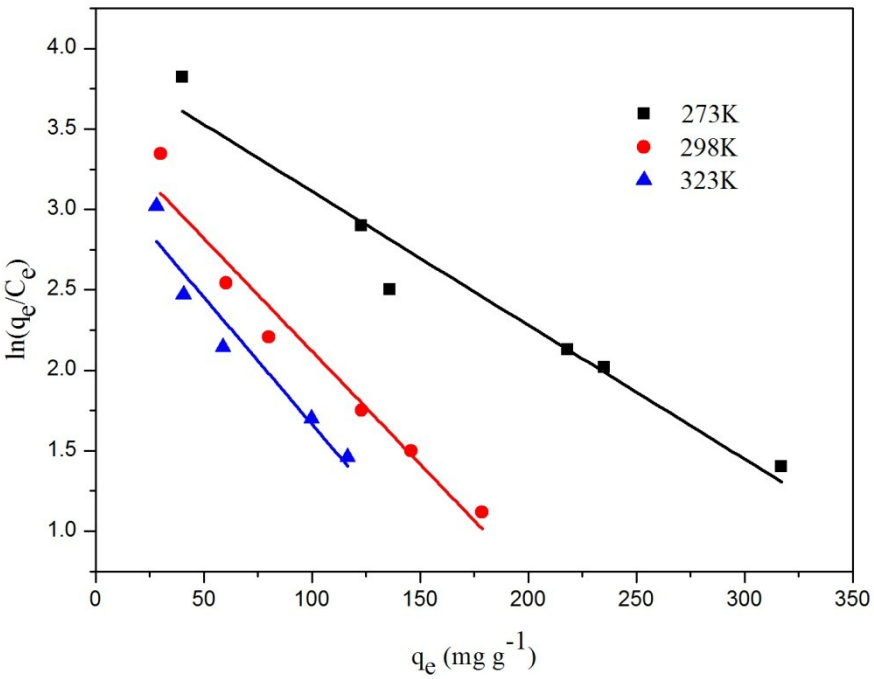

Fig. S5 Plots of ln q_e/C_e *vs.* q_e for calculation of thermodynamic parameters.

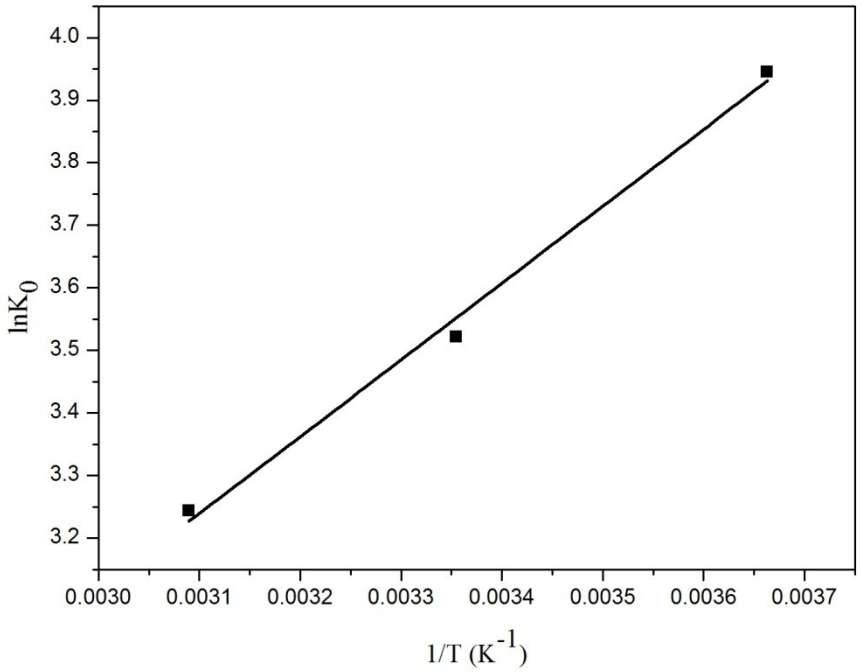

Fig. S6 Van't Hoff plot for the adsorption of MO onto MnO_2/G-CNT hybrid material.

References

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2. Kenneth R. Hall, Lee C. Eagleton, Andreas Acrivos and T. Vermeulen, *Ind. Eng. Chem. Fundam.*, 1966, **5**, 212-223.
3. M. Ahmaruzzaman and S. Laxmi Gayatri, *Chem. Eng. J.*, 2010, **158**, 173-180.