Supplementary Material

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The ammoniation process of silica microspheres

The specific ammoniation process of silica microspheres was as follows. 0.6 g of silica microspheres previously prepared was added to the mixed solution of 20 mL of toluene and 0.3 mL of 3-aminopropyltriethoxysilane (APTES). After sonication 30 min, the mixed solution was transferred to oil bath and refluxed for 6 h at 105℃. Then, the obtained solution was washed three times with toluene and ethanol, respectively. Finally, the NH_2-SiO_2 microspheres were collected by drying in vacuum oven at 60˚C.

Adsorption kinetic model

Pseudo-first order kinetic model. Adsorbents that conform to this kinetic model are not selective for the adsorbed material and the adsorption process is faster. The pseudo-first-order equation is expressed as follows：

$$\ln(q_e - q_t) = \ln q_e - K_1 t \quad (1)$$

where q_e and q_t are the adsorption amounts (mg g^{-1}) at equilibrium and time t (min), respectively, K_1 is the first-order adsorption rate constant, and the unit is min^{-1}, determined by the slope of the $\ln(q_e-q_t)$~t linear graph, the specific values are shown in Table 1.

Pseudo-second order kinetic model. The model shows that the adsorption rate is controlled by the chemisorption mechanism, involving electron public or electron transfer between the adsorbent and the adsorbate, which belongs to chemical adsorption. The pseudo-second-order equation is expressed as follows:

$$\frac{\mathrm{T}}{q_{\mathrm{t}}} = \frac{1}{K_2 q_{\mathrm{e}}^{\ 2}} + \frac{1}{q_{\mathrm{e}}} t \qquad (2)$$

where K_2 is the rate constant of pseudo-second-order adsorption (g mg^{-1} min^{-1}), the specific values are shown in Table 1.

Weber and Morris kinetic model. The process of chemical reactions is complex and is usually completed in multiple steps. This model is used to analyze the control steps in the reaction to determine the internal diffusion rate constant of the adsorbent particles. If line passes through the origin indicates that the internal diffusion is controlled by a single rate. The equation is expressed as follows:

$$q_{\mathrm{t}} = \mathrm{K}_3 t^{1/2} + \mathrm{C} \qquad (3)$$

Where K_3 is the internal diffusion rate constant and C is a constant. q_t is the amount of dyes adsorbed at time t (mg g^{-1}) and $t^{1/2}$ is the square root of the time of contact ($min^{1/2}$). The fitting of the Weber and Morris kinetic model for MB and MG are shown in Fig.S6.

Adsorption isotherm model

Langmuir adsorption isotherm

The langmuir adsorption isotherm model assumes that the adsorption process is a single-layer surface adsorption, all adsorption sites are the same and the adsorbates are completely independent [35], which expression is as follows:

$$\frac{c\mathrm{e}}{q\mathrm{e}} = \frac{c\mathrm{e}}{q} + \frac{1}{q\,\mathrm{b}} \qquad (4)$$

where c_e(mg L^{-1}) is the equilibrium concentration of MG and MB solution, q_e(mg g^{-1}) is the saturated adsorption amount at equilibrium, q is the maximum adsorption capacity(mg g^{-1}) and b is the equilibrium adsorption constant (L mg^{-1}).

Freundlich adsorption isotherm

The Freundlich model is an empirical formula, and the adsorbent surface conforming to the model is heterogeneous [36,37]. The effect of pollutant concentration on adsorption is significant. As the concentration increases, the interaction between surface ions increases, so the number of layers that appear adsorbed increases, which expression is as follows:

$$\ln q_e = \ln K_F + \frac{1}{n}\ln c_e \quad (5)$$

where q_e (mg g^{-1}) is the saturated adsorption amount at equilibrium, c_e (mg L^{-1}) is the equilibrium concentration of MG and MB solution, K_F is the Freundlich constant (L mg^{-1}), indicating the adsorption capacity, 1/n indicating the adsorption intensity, the values of K_F and 1/n can be obtained from the intercept and slope of the linear plot of $\ln q_e$ versus $\ln c_e$, respectively. The value of 1/n is generally considered to be between 0 and 1, where if n = 1, the adsorption curve is linear; if n<1, adsorption is chemisorption; if n>1, it is a physical process.

The n values of the adsorbents for MB and MG were calculated separately, and the results showed that the adsorption for both was physical adsorption.

Henry adsorption isotherm

The Henry adsorption isotherm equation shows that the amount of adsorption is proportional to the concentration of pollutants during the adsorption process. The specific equation is as follows:

$$q_e = K c_e \quad (6)$$

where c_e is the equilibrium concentration of dye in aqueous solution (mg L^{-1}), q_e is the adsorption amount (mg g^{-1}) at equilibrium and K is the Henry constant. This formula is only applicable to the adsorption amount that accounts for 10% of the total adsorption of a single layer, which is a maximum of 10% of the adsorbent surface is covered by adsorbent molecules.

Temkin adsorption isotherm.

Temkin adsorption isotherm reveals the effect of temperature on the isotherm, and the form of the equation is similar to the Freundlich isotherm, which expression is as follows:

$$q_e = \frac{RT}{b_T}\ln c_e + \frac{RT}{b_T}\ln A_T \quad (7)$$

where q_e(mg g^{-1}) is the saturated adsorption amount at equilibrium, c_e (mg L^{-1}) is the equilibrium concentration of MG and MB solution, A_T (L mg^{-1}) is the Temkin constant and b_T (J mol^{-1}) is the Temkin isotherm adsorption heat. R (8.314 J mol^{-1} K^{-1}) is the gas constant, T is the temperature in Kelvin.

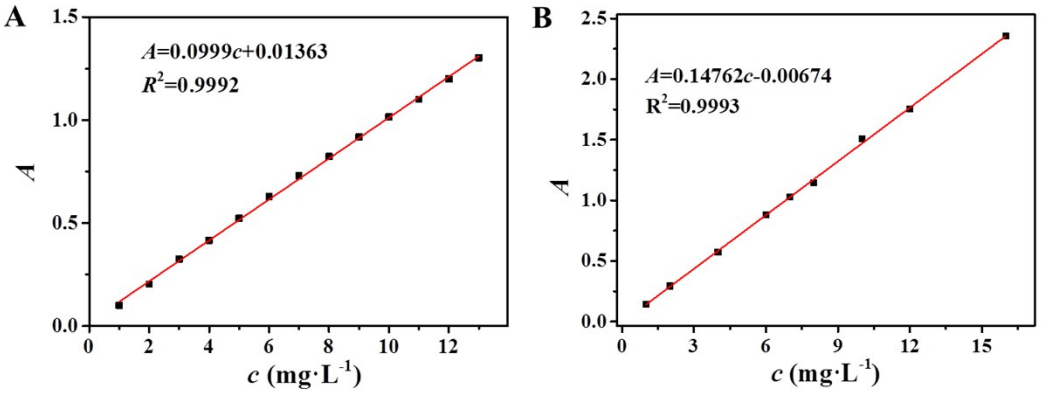

Fig. S1 The standard curve of (A) methylene blue at the 664 nm of wavelength and (B) malachite green at the 617 nm of wavelength

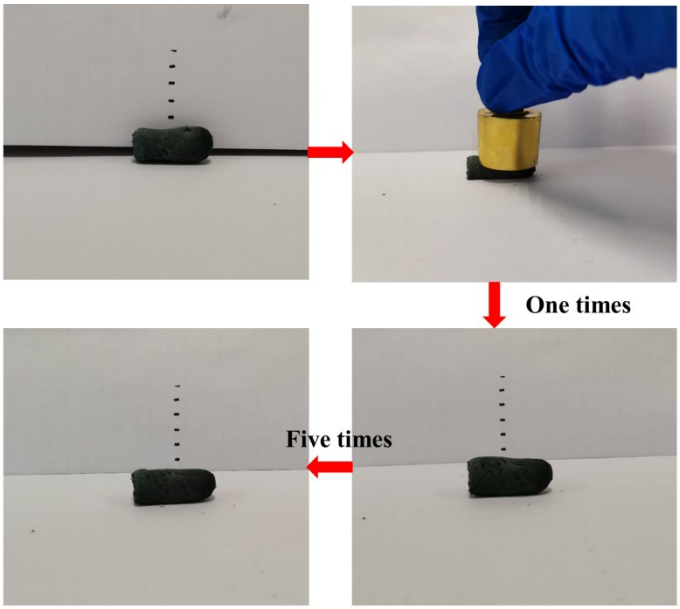

Fig.S2 Mechanical testing about the PANI/SiO_2/GA

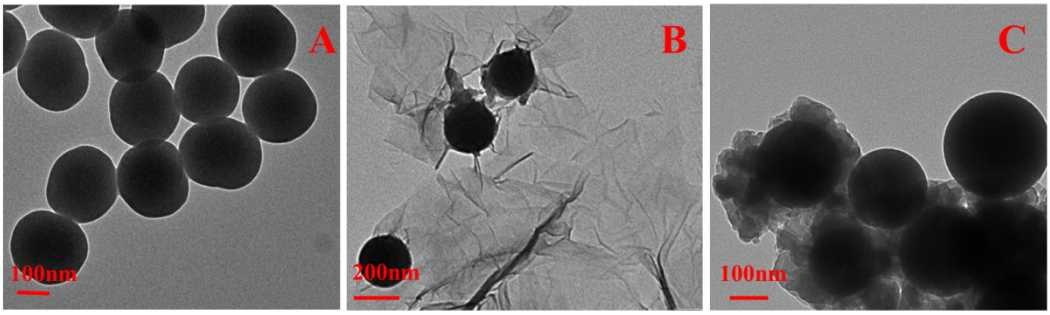

Fig.S3 The TEM image of (A) SiO_2, (B) PANI/SiO_2, and (C) PANI/SiO_2/GA

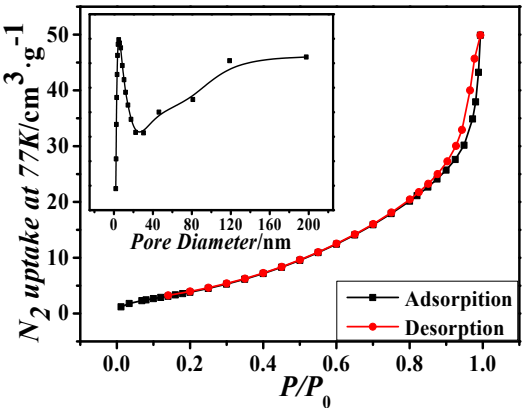

Fig.S4 Nitrogen adsorption-desorption isotherms for the pore size distribution curves (inset) of PANI/SiO_2/GA

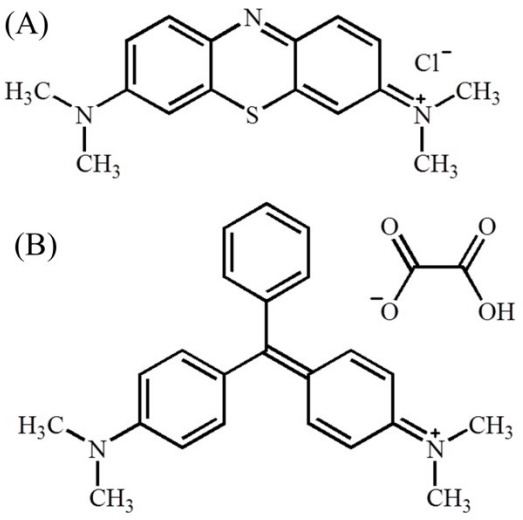

Fig.S5 The molecular structure of methylene blue (A) and malachite green (B)

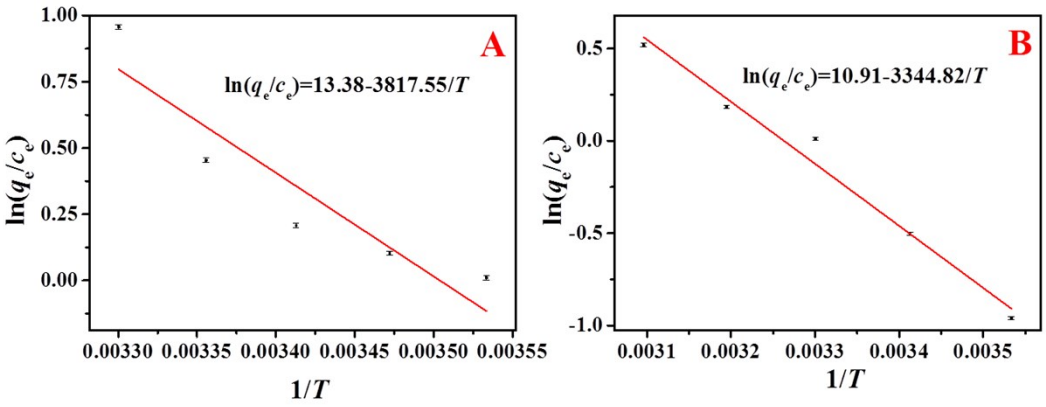

Fig. S6 Van der Hough equation curves of MB (A) and MG (B)

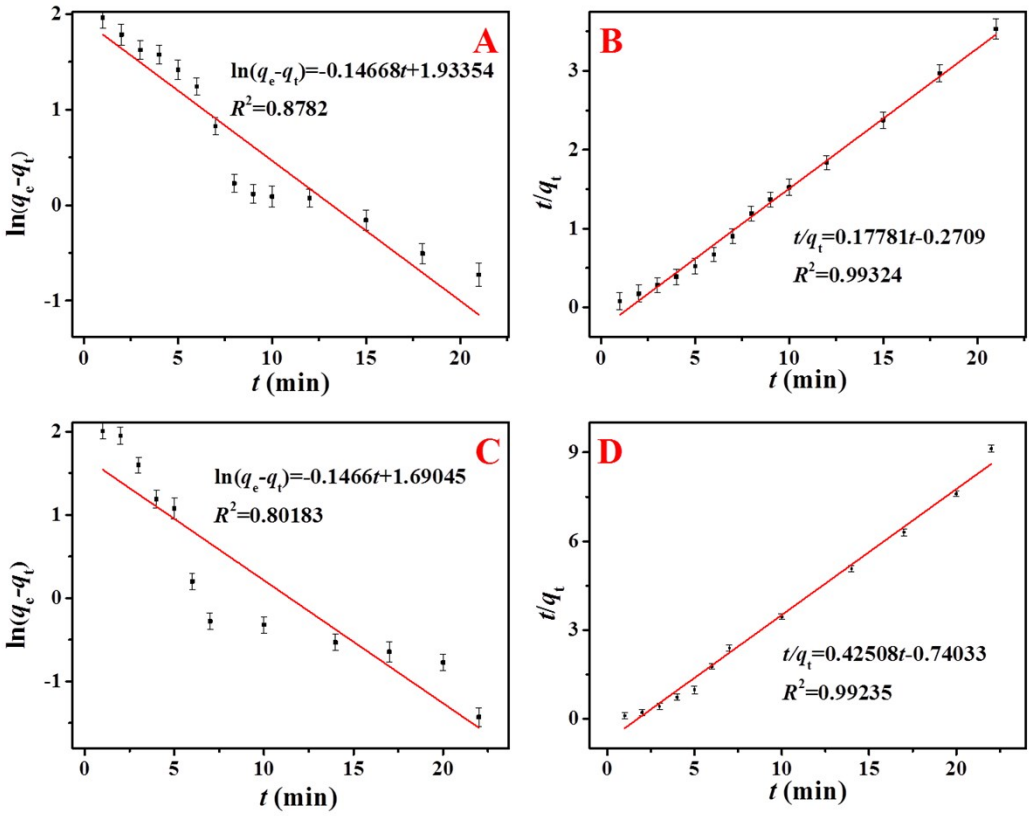

Fig.S7 The pseudo-first-order models of MB (A) and MG (C), pseudo-second-order models of MB (B) and MG (D)

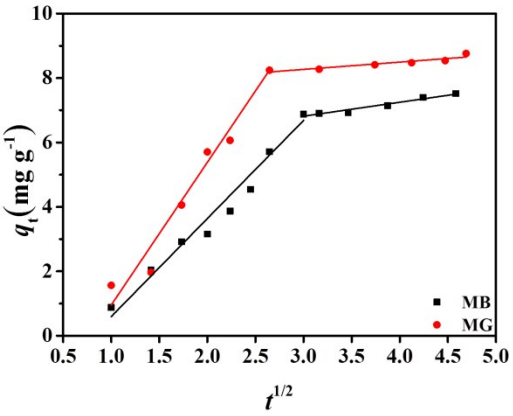

Fig.S8 Fitting of the Weber and Morris kinetic model for MB and MG

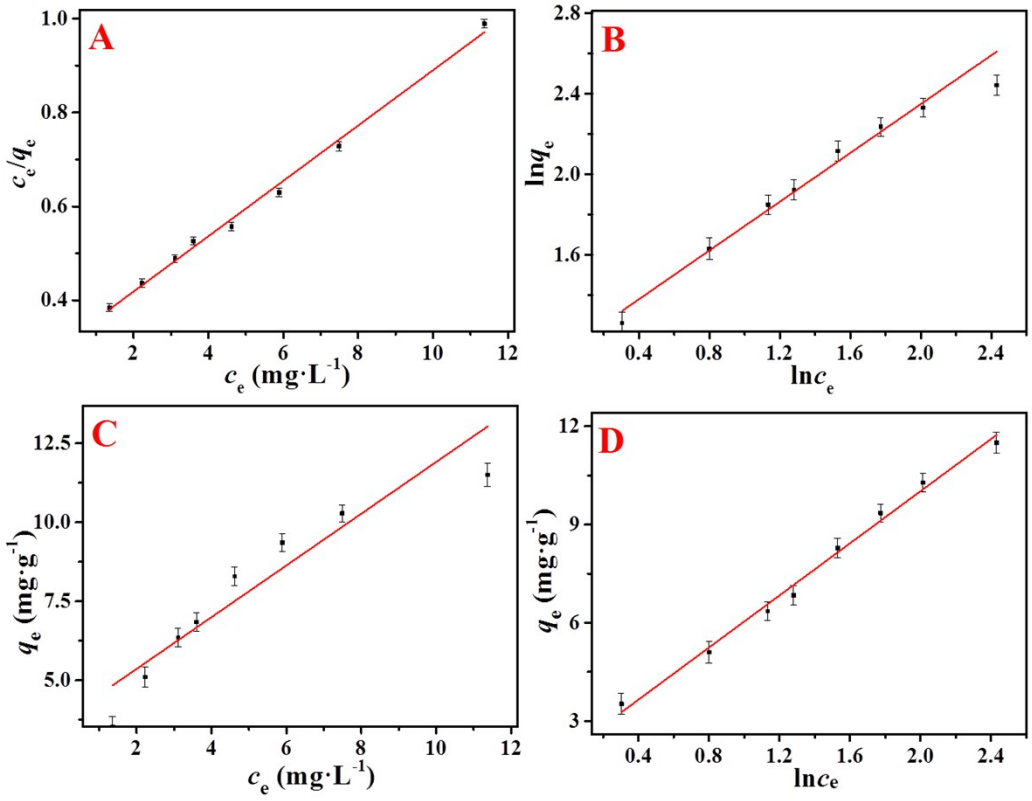

Fig.S9 The adsorption isotherm models of MB, (A) Langmuir, (B) Freundlich, (C) Henry, (D) Temkin

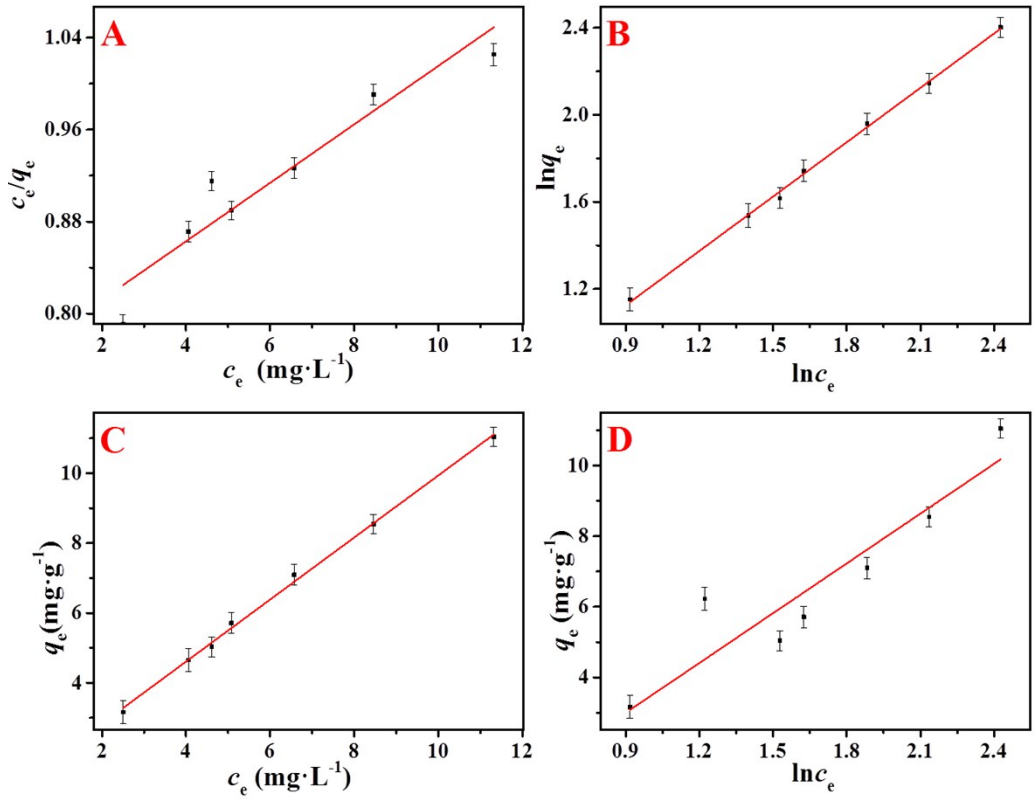

Fig.S10 The adsorption isotherm models of MG, (A) Langmuir, (B) Freundlich, (C) Henry, (D) Temkin

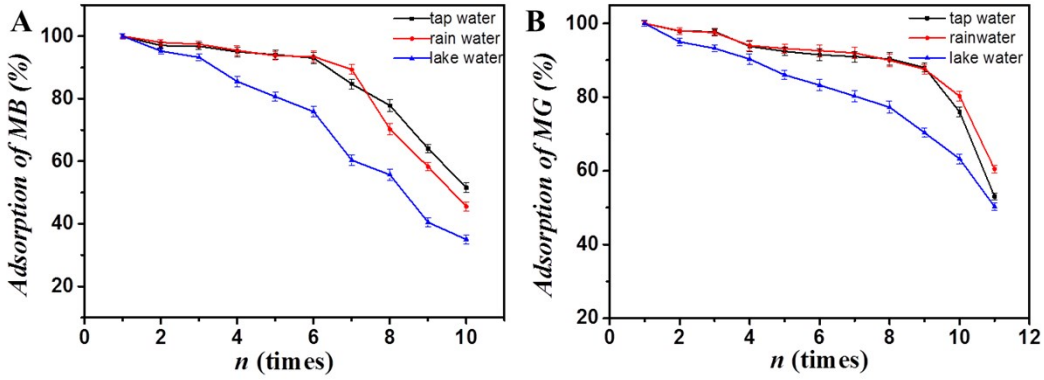

Fig.S11 PANI/SiO_2/GA eluted with 0.1M NaOH in different water matrices

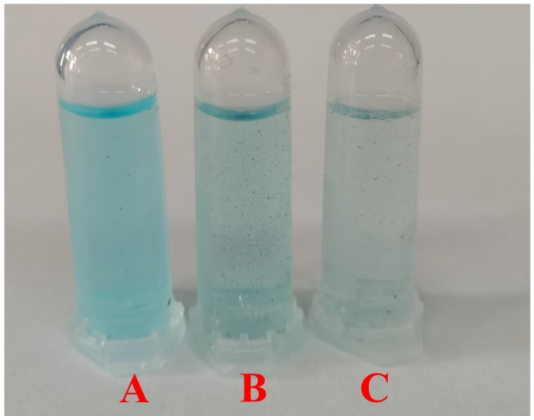

Fig.S12 The photoes of MB, (A) 10mg L^{-1} MB solution, (B) the MB solution after 5 minutes of GA adsorption, (C) the MB solution after 5 minutes of PANI/SiO_2/GA adsorption

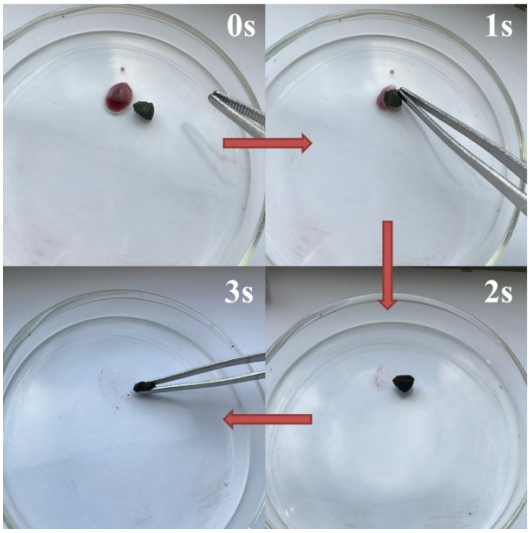

Fig.S13 Absorption of 50 μL of aniline dyed by Sudan II