Supplementary Information

Rapid organic contaminants removal by Mg/Fe-based magnetic nanosheets synthesized using the molten salt method

Text S1 Preparation of materials

(a) Preparation of Mg/Fe-based magnetic nanosheet (MMN)

First, a $NaNO_3$ powder (5.0 g) was added into a crucible and transferred into a muffle furnace preheated to 350 °C. After 15 min, the $NaNO_3$ had become molten. Then, a mixture of $Mg(OAc)_2$ and $FeCl_3$ (Mg=140 mg, Fe=60 mg) was added to the molten salt, and the reaction was heated at 350 °C for an additional minute. After cooling to room temperature, the dark brown product was washed several times with a sufficient amount of deionized water to remove the unreacted reactants. Then, the product was filtered and dried overnight in an oven at 80 °C.

(b) Preparation of the MMN-coated sponge (MMN-S)

Pieces of MF sponge measuring 2×2×2 cm were ultrasonically cleaned with deionized (DI) water and ethanol to remove pollutants in the sponge and then dried overnight in an oven at 50 ℃ to use as a template for depositing adsorbents. As-prepared MMN (250 mg) was dispersed in distilled water (50 mL) for 15 min under ultrasonic agitation. The MMN dispersion and pieces of pre-cleaned MF sponge were transferred to a Teflon-lined autoclave bomb and incubated at 50 ℃ for 12 hours. After cooling to room temperature, the MMN-S was taken from the autoclave bomb and washed with DI water by shaking. Finally, the MMN-S was dried overnight in an oven at 50 ℃.

(c) Preparation of the Au nanoparticle coated MMN (Au-MMN)

The Au nanoparticle (AuNP)-coated MMN (Au-MMN) was prepared to evaluate its catalytic ability in the reduction of 4-nitrophenol (4-NPh) to 4-aminophenol (4-APh). MMNs (5 g) were dispersed in 5 mL of DI water using ultrasonic agitation for 5 minutes. An aqueous $HAuCl_4$ solution (2.5 mL, 1 mg mL^{-1}) was then added dropwise to the MMN dispersion while

stirring, followed by the addition of $NaBH_4$ (0.025 mL, 10 mM). After an additional 10 minutes of stirring, the Au-MMN was collected by magnetic separation and rinsed three times with DI water.

Text S2 Characterization

The morphologies of MMN was characterized by ultrahigh-resolution field emission-scanning electron microscopy (FE-SEM) using a Hitachi SU8600 instrument and by field emission-transmission electron microscopy (FE-TEM) using a JEOL JEM-2100F instrument. Powder X-ray diffraction (XRD) analysis was carried out using a PANalytical Empyrean multipurpose diffractometer (Cu Ka radiation, 298 K) in Daegu Center, KBSI. ζ-potential measurements were performed on a Malvern Nano-ZS Zetasizer at room temperature in ethanol as a solvent. X-ray photoelectron spectroscopy (XPS) studies were performed using a K-ALPHA+ (Thermo Scientific, UK) system with an aluminum anode (Al Kα, 1486.6 eV) at 12 kV and 72 W in Busan Center, KBSI. UV-vis absorption spectra were recorded on a UV-vis spectrophotometer (Thermo Fisher Scientific, GENESYS^{TM} 180). The magnetic properties of the samples were obtained using a vibrating sample magnetometer (MPMS@3 SQUID-VSM, Quantum Design, San Diego, CA, USA) at the KBSI, and magnetic performance over a range from –20 to +20 kOe was recorded at 300 K. The Brunauer-Emmett-Teller (BET) surface area and Barrett-Joyner-Halenda (BJH) pore size distribution were measured using an accelerated surface area and porosimetry system (Micromeritics ASAP2010, USA) at the KBSI.

Text S3 Adsorption kinetic study

The adsorption rate and adsorption mechanism of MMN adsorbents were investigated by an adsorption kinetic study employing pseudo-first-order (PFO) and pseudo-second-order (PSO) models. The two kinetic models can be described by the following nonlinear equations (1) and (2):

PFO: $ln(q_e - q_t) = ln\, q_e - k_1 \cdot t$ (1)

PSO: $\frac{t}{q_t} = \frac{1}{k_2 \cdot q_e^2} + \frac{1}{q_e} t$ (2)

where q_e (mg g^{-1}) and q_t (mg g^{-1}) are the quantities of organic dyes at equilibrium and at contact time t (min), respectively. k_1 (min^{-1}) and k_2 (g mg^{-1} min^{-1}) are the equilibrium rate constants calculated from the PFO and PSO models, respectively.

Text S4 Adsorption isotherm study

Adsorption isotherm experiments were performed to understand the adsorption behavior and determination of the maximum adsorption capacity of MMN adsorbents. The adsorption isotherm data were fitted using two equilibrium models, the Langmuir and Freundlich isothermal models, which are given by the following linear equations, respectively:

Langmuir: $\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{C_e}{q_m}$ (3)

Freundlich: $lnq_e = \frac{1}{n} lnC_e + lnK_F$ (4)

where C_e (mg L^{-1}) is the equilibrium concentration of dyes; q_e (mg g^{-1}) is the amount of dye adsorbed on the MMNs; q_m (mg g^{-1}) is the maximum adsorption capacity of the adsorbents and

K_L (L mg^{-1}) is the Langmuir constant, which relates to the enthalpy of sorption. K_F and n are the Freundlich constants related to adsorption capacity and affinity between the adsorbents and adsorbates, respectively.

Text S5 Adsorption thermodynamic study

To confirm the interactions between dyes and MMN adsorbents, verify the nature of adsorption, and evaluate spontaneity, a comprehensive thermodynamic analysis was carried out. The thermodynamic parameters ΔG^0 (standard Gibbs free energy change, kJ mol^{-1}), ΔH^0 (standard enthalpy change, kJ mol^{-1}), and ΔS^0 (standard entropy change, kJ mol^{-1} K^{-1}) were calculated from the temperature-dependent adsorption isotherms using the following equations:

$$\Delta G^0 = -RTlnK_d \quad (5)$$

$$lnK_d = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \quad (6)$$

where R (8.314 J mol^{-1} K^{-1}) is the gas constant, T (K) is the absolute temperature and K_d is the thermodynamic equilibrium constant, calculated by plotting ln(q_e/C_e) versus q_e and extrapolating q_e to zero.

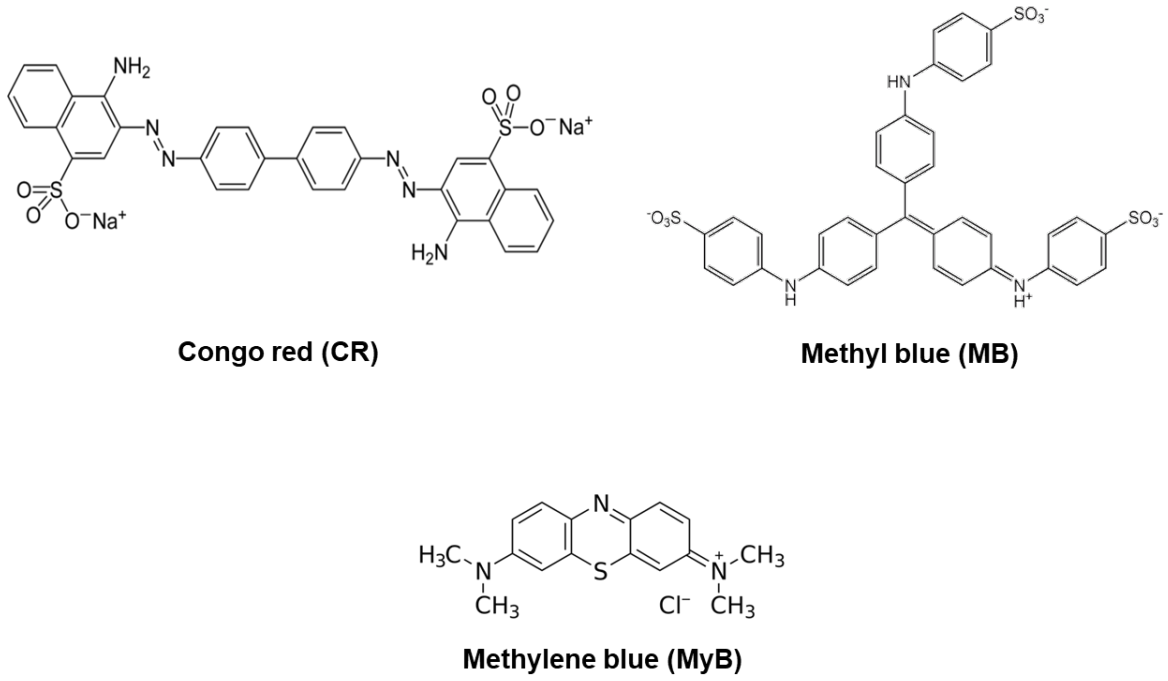


Fig. S1. Chemical structures of organic dyes Congo red, methyl blue and methylene blue.

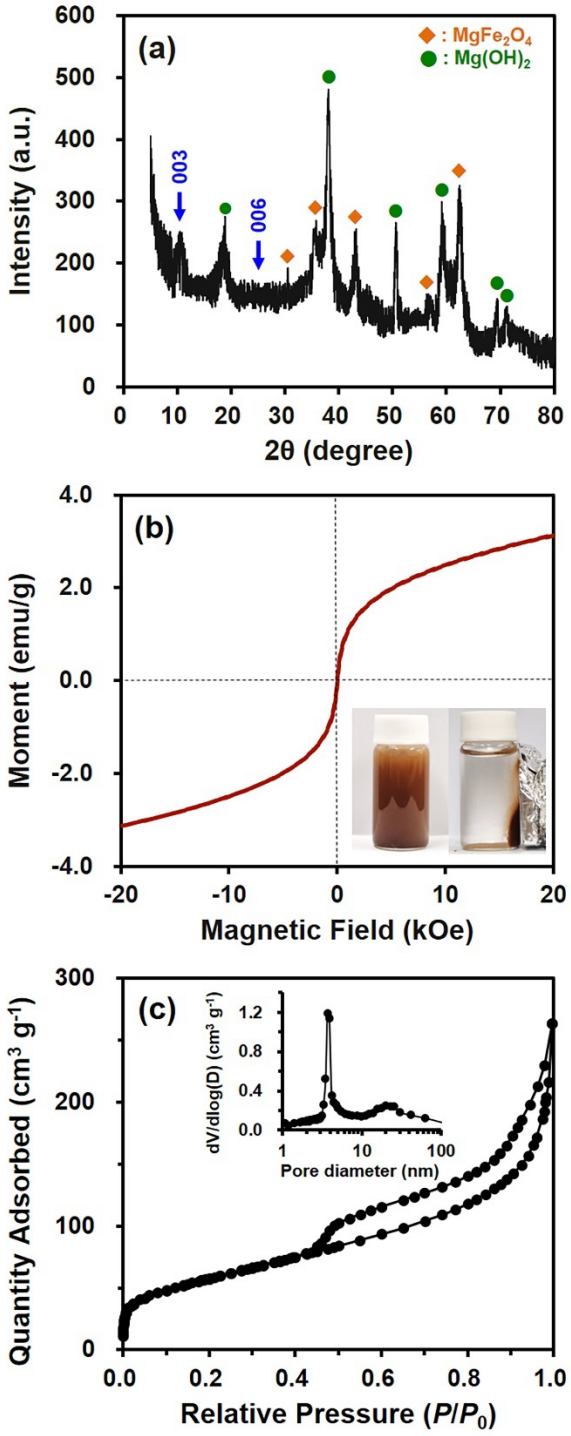


Fig. S2 (a) XRD pattern, (b) field-dependent magnetic curve and (c) N_2 adsorption-desorption isotherm and the corresponding pore size distribution (inset) of MMNs. Inset of (b): photographs of MMNs dispersed in water (left) and attracted to the side of the container after applying an external magnetic field (right).

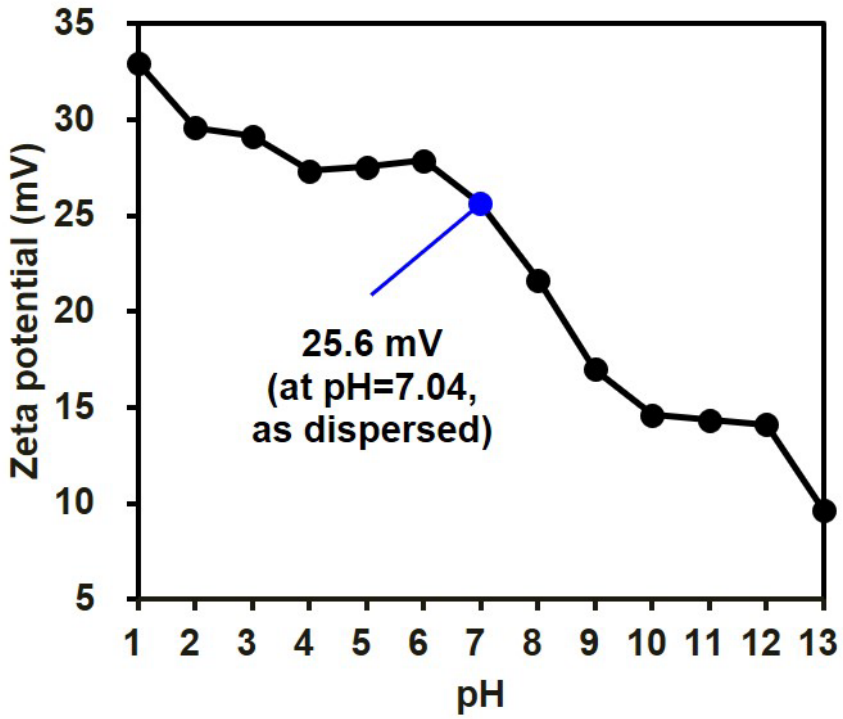


Fig. S3 (a) Zeta potential of MMNs in aqueous solution with various pHs. Adsorbent quantity = 0.5 mg mL^{-1} and T = 294 K.

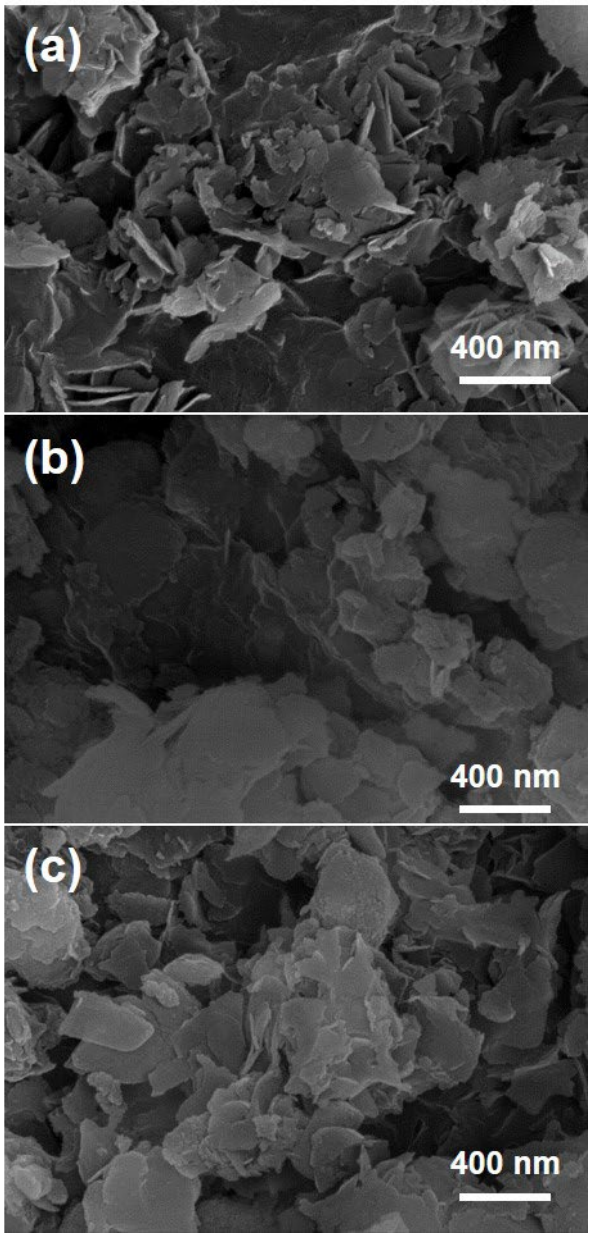


Fig. S4 SEM images of MMNs (a) before and after (b) MB and (C) CR adsorption.

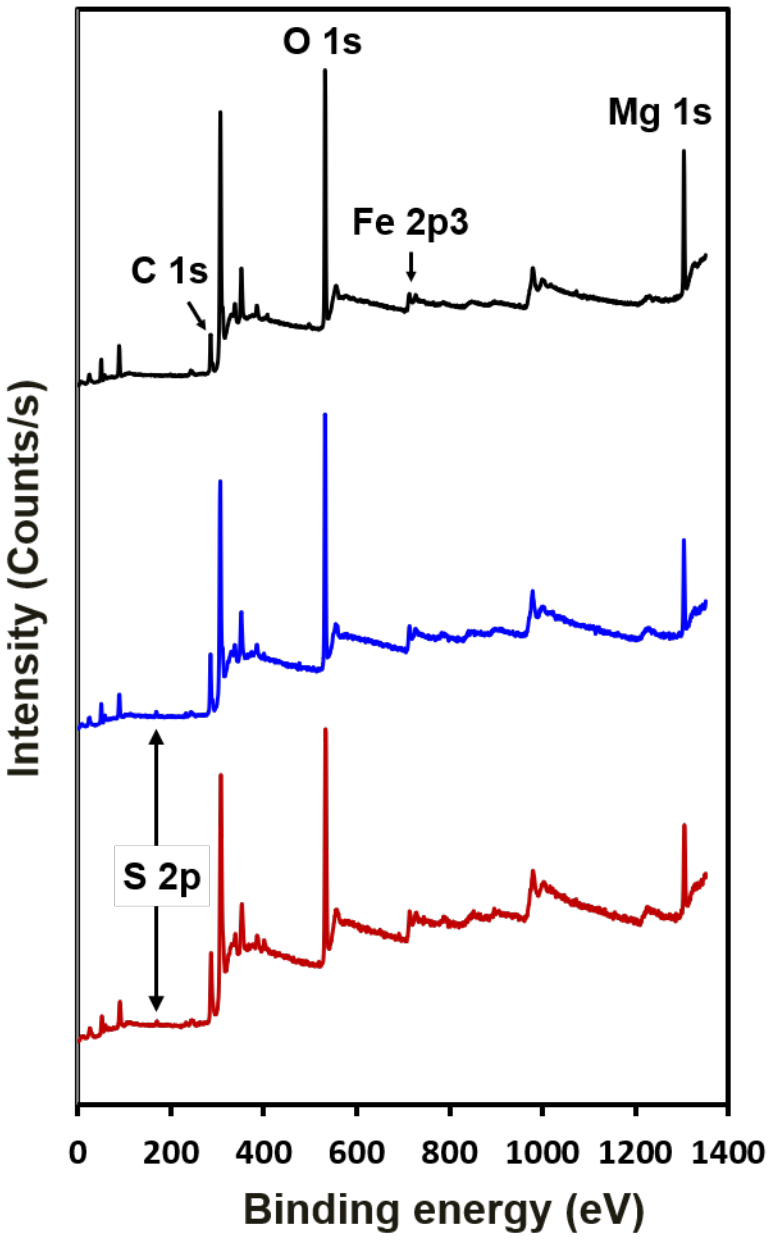


Fig. S5 XPS survey spectra of MMN before adsorption (black) and after adsorption of MB (blue) and CR (red).

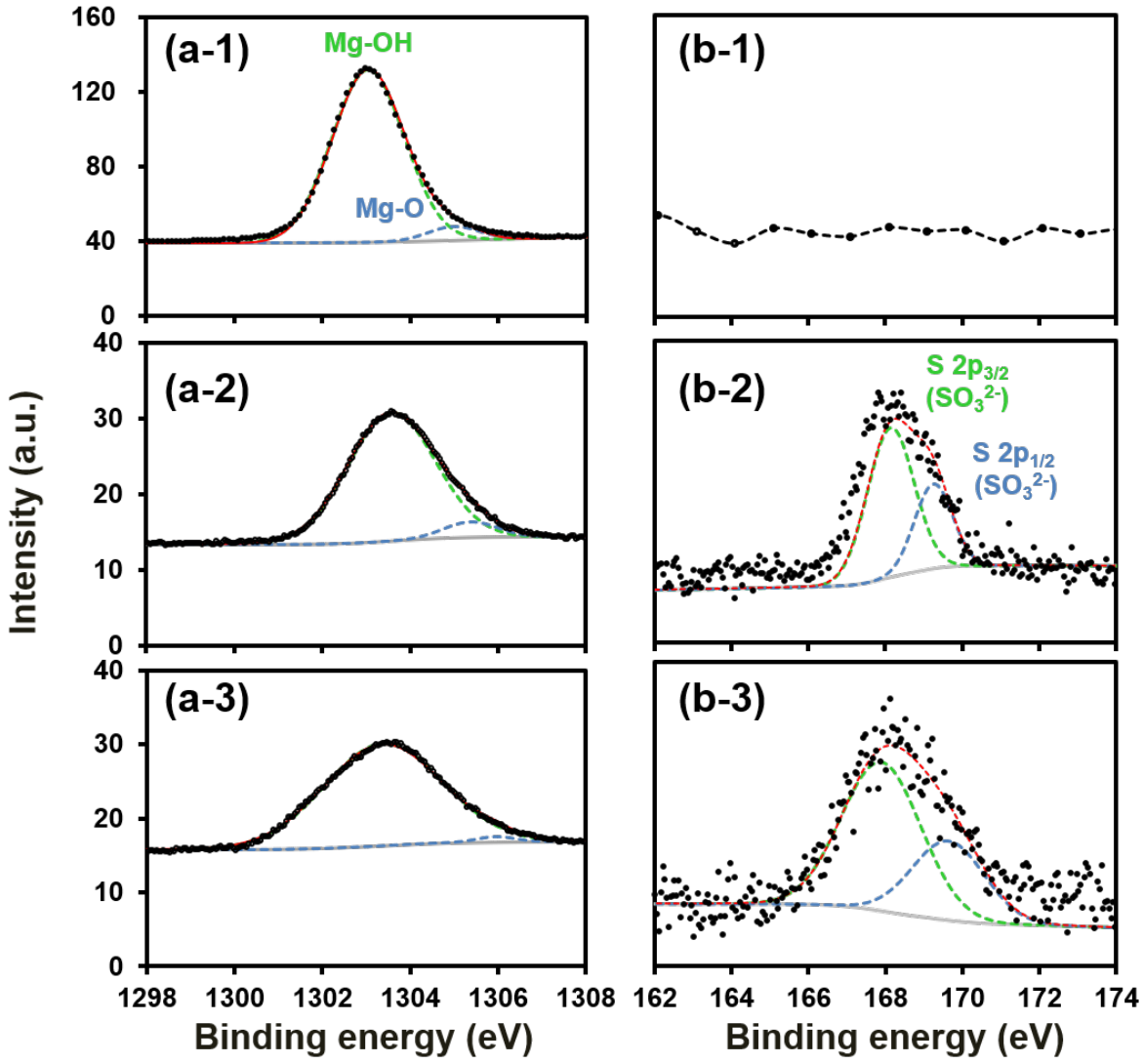


Fig. S6 High-resolution XPS (a) Mg 1s and (b) S 2p spectra of MMN (1) before adsorption and after adsorption of (2) MB and (3) CR.

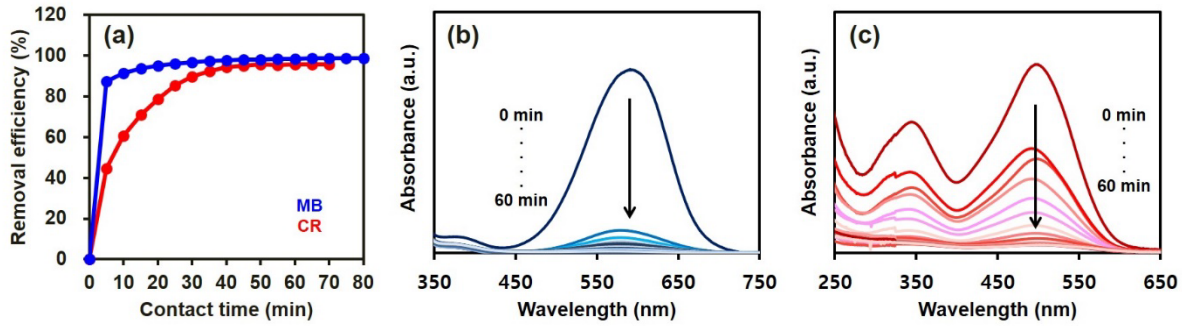


Fig. S7 (a) Removal efficiencies of MB (blue line) and CR (red line) by the MMN as a function of contact time, and time-dependent UV–vis absorption spectral changes in (b) MB and (c) CR adsorption by the MMN. Adsorbent quantity = 0.5 mg mL^{-1}, C_0 (dyes) = 100 ppm, reaction time = 0~90 min, and T = 294 K.

Fig. S8 Photographs showing the process for the dye absorption by the MMNs: left is before and center is after adding the MMNs, and right is after isolating the MMNs with an external magnetic field.

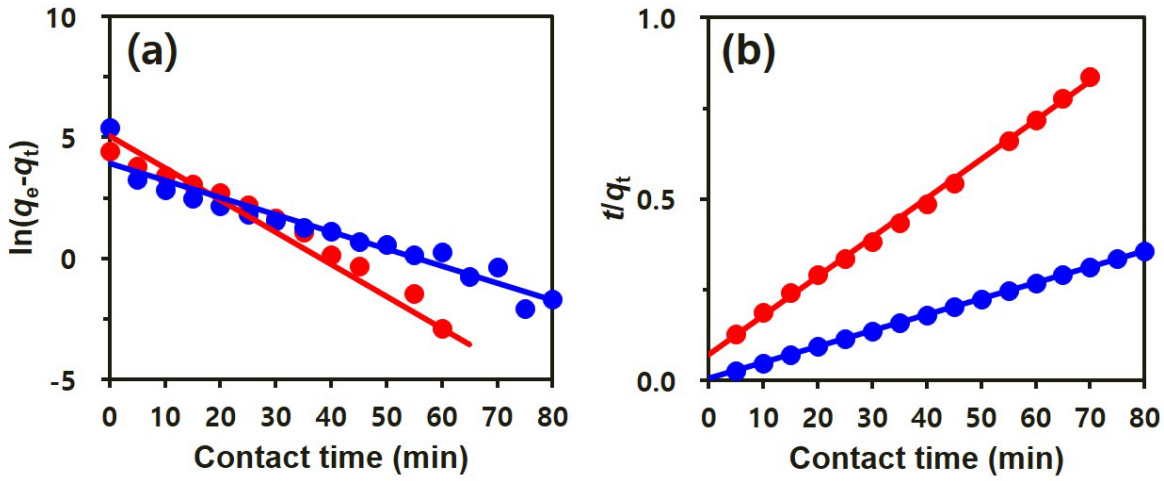


Fig. S9 Linear kinetic plots using the (a) pseudo-first-order (PFO) and (b) pseudo-second-order (PSO) models for MB (blue lines) and CR (red lines) adsorption on MMNs. Adsorbent quantity = 0.5 mg mL^{-1}, C_0(dyes) = 100 ppm, reaction time = 0~80 min, and T = 294 K.

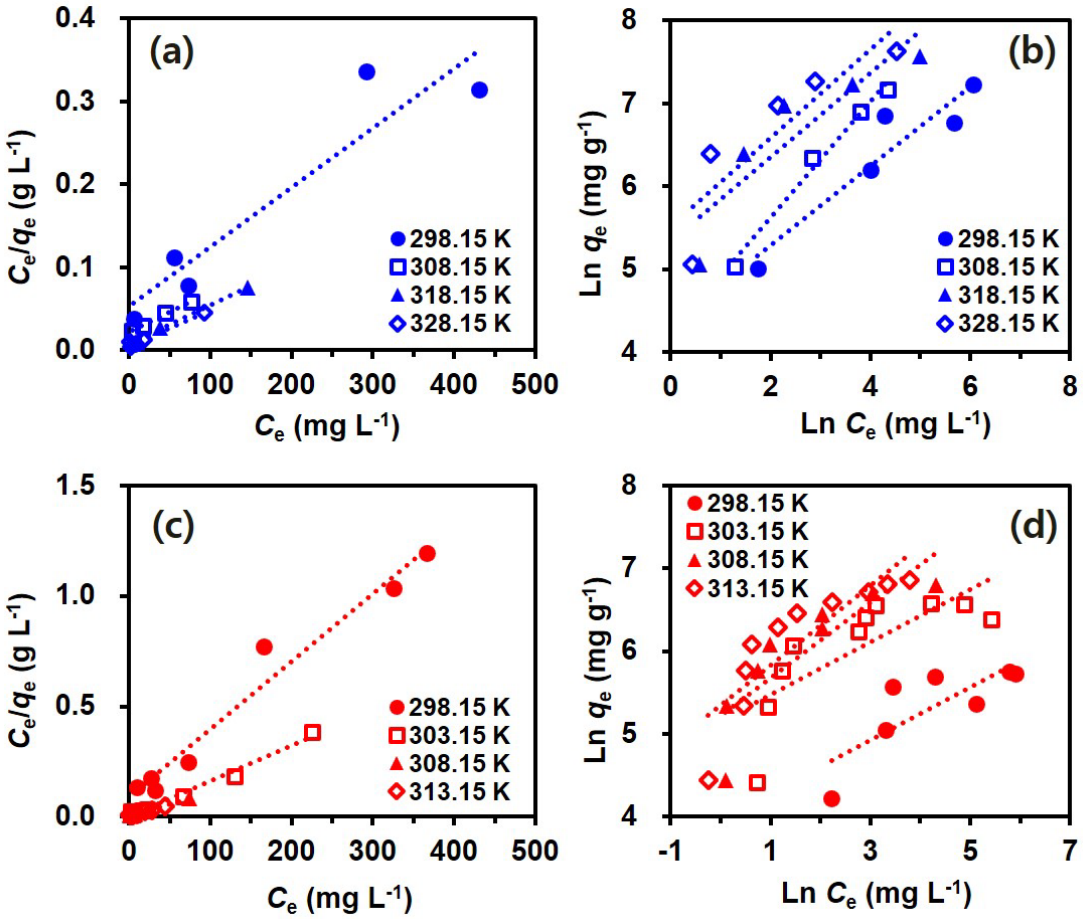
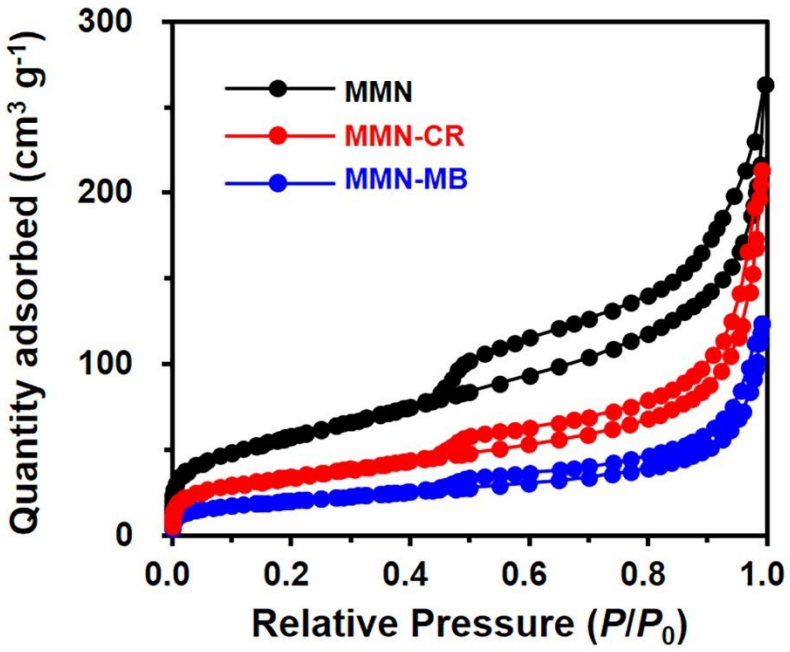


Fig. S10 Adsorption isotherms of (a and b) MB (blue lines) and (c and d) CR (red lines) on MMNs fitted by (a and c) the Langmuir model and (b and d) the Freundlich model. Adsorbent quantity = 0.5 mg mL^{-1}, C_0 (dyes) = 50~500 ppm, reaction time = 90 min.

(a)



(b)

Adsorbents	S_{BET} (m^2 g^{-1})	Zeta potential (mV)
MMN	200.9	+25.6
MMN-MB	72.7	-12.4
MMN-CR	123.8	-20.3

Fig. S11 (a) N_2 adsorption-desorption isotherms of MMNs after MB (blue line) and CR (red line) adsorption, and (b) a summary table of the BET surface area and zeta potential of MMNs before and after dye adsorption.

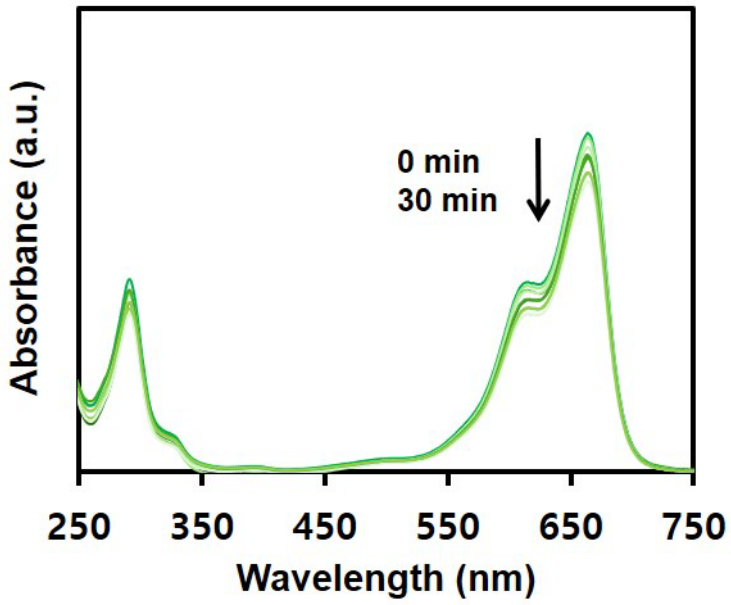


Fig. S12 Time-dependent UV–vis absorption spectral changes in methylene blue adsorption by the MMNs. Adsorbent quantity = 0.5 mg mL^{-1}, C_0 (dyes) = 100 ppm, reaction time = 0~30 min, and T = 294 K.

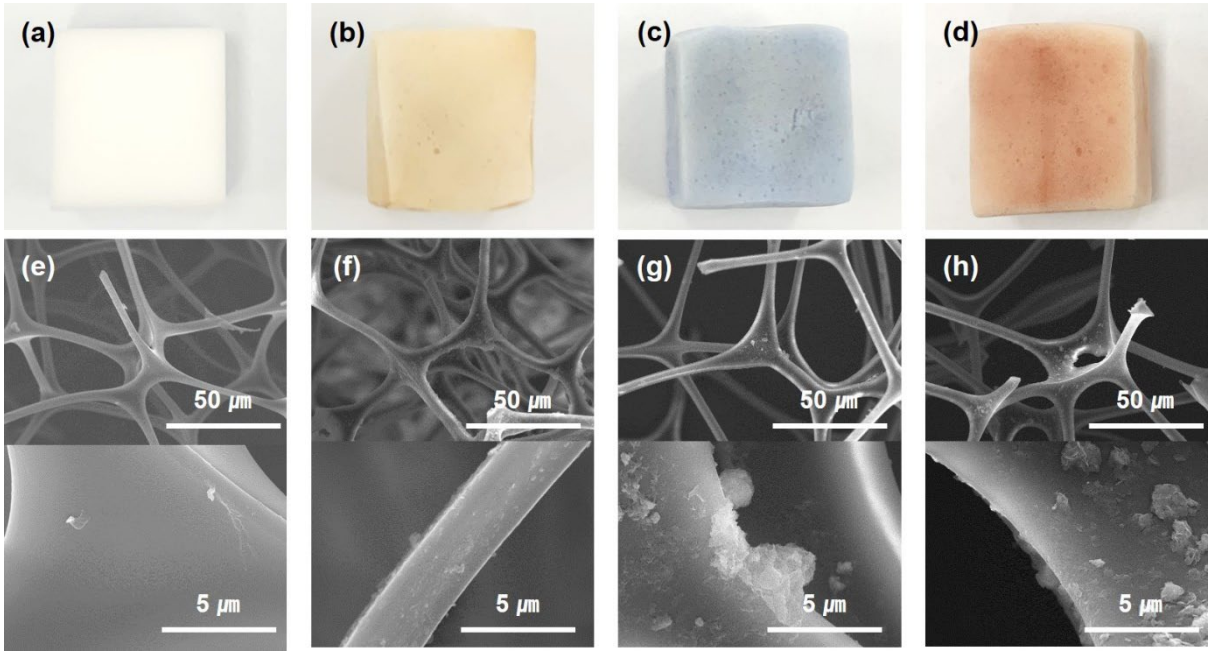


Fig. S13 (a-d) Photographs and (e-h) SEM images of MF sponges: (a and e) pristine MF sponge, (b and f) MMN-S, (c and g) MMN-S after MB adsorption, and (d and h) MMN-S after CR adsorption. C_0 (dyes) = 100 ppm, reaction times = 60 min (MB) and 240 min (CR), and T = 294 K.

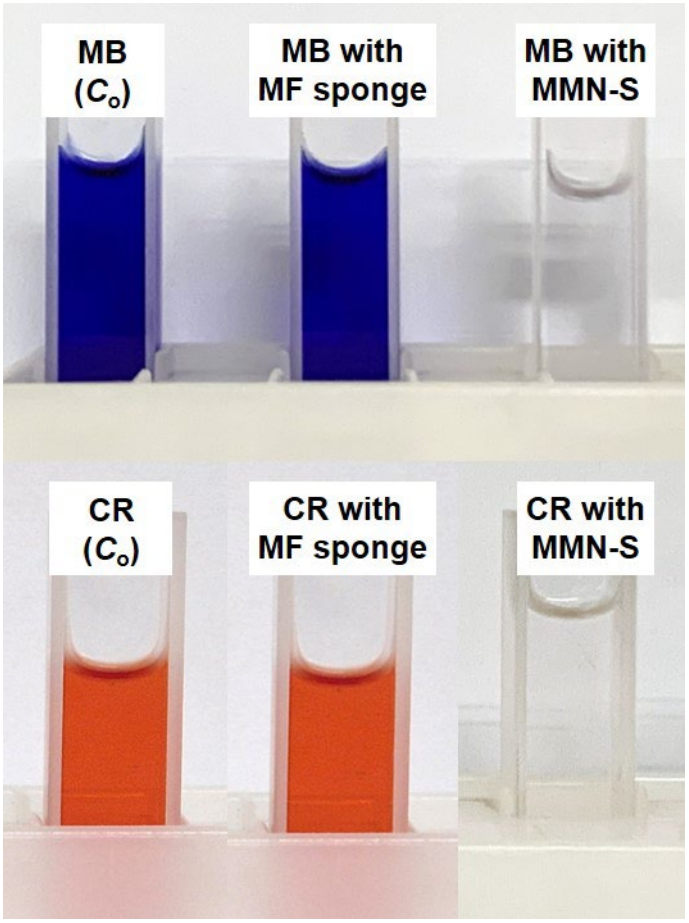


Fig. S14 Photographs of MB (blue color) and CR (red color) solutions before (left) and after adsorption with pristine MF sponge (center) and MMN-S (right). C_0(dyes) = 100 ppm, reaction time = 240 min, and T = 294 K.

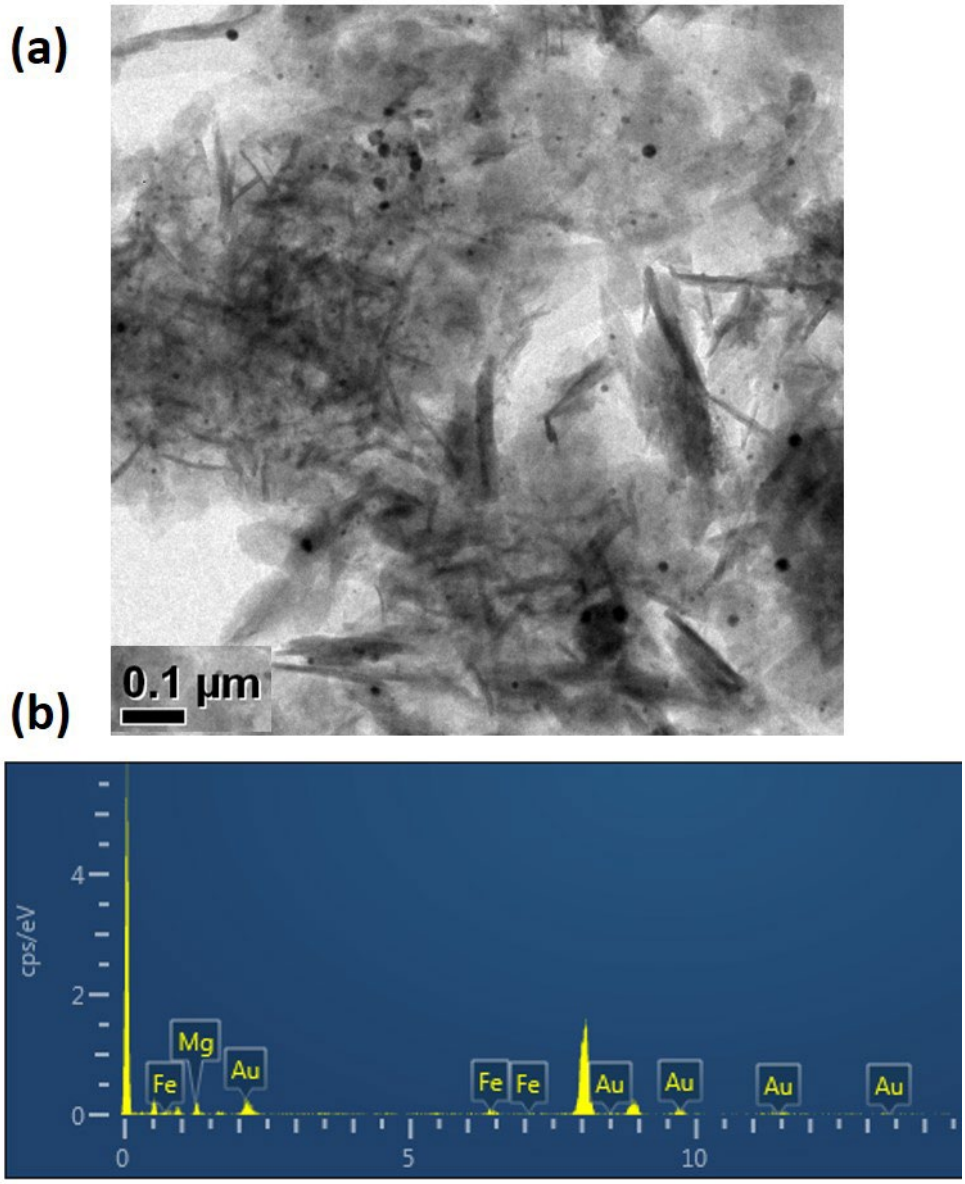


Fig. S15 (a) TEM image and (b) EDS spectrum of Au-MMN.