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Ultra-effective removal of methylene blue dye and lead heavy metal from aqueous solution using novel Co and Fe co-doped magnetic activated carbon nanohybrid material from *Nigella sativa* (*Nigella sativa* L.) industrial processing wastes

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Abstract

In this study, a novel magnetic carbonaceous nanohybrid material (CoF@NAC) was synthesized by microwave-assisted simple chemical co-precipitation method in the basic medium using iron and cobalt nitrate salts in the presence of *Nigella sativa* (*Nigella sativa* L.) industrial processing waste-based activated carbon (NAC) and then characterized by SEM, EDX, Mapping, BET, XRD, VSM, FTIR, Boehm titration, and pH_{ZPC} techniques. Its BET surface area, total pore volume, mesoporosity, average pore diameter, and saturation magnetization values were 729 m²/g, 0.426 cm³/g, 61.5%, 2.4 nm, and 23.3 emu/g, respectively. To test its adsorptive performance, methylene blue (MB) dye and lead (Pb(II)) heavy metal water pollutants were selected as model adsorbates. Optimum adsorption conditions were determined according to the maximum effects of key experimental parameters. The kinetic and isotherm findings of each adsorption system fit best the pseudo-second-order kinetic and Langmuir isotherm models, respectively. The maximum removed MB and Pb(II) quantities of

MB and Pb(II) were 667 and 455 mg/g, respectively, at their natural pH levels (pH 6 for MB and pH 5 for Pb(II)) in water. Thermodynamic parameters calculated showed that their processes were spontaneous, endothermic for MB, and exothermic for Pb(II). Moreover, it showed high recyclability stability over four cycles for the studied adsorbates.

Keywords: Nigella sativa oil waste; Cobalt ferrite; Activated carbon; Water treatment; Adsorption; Reusability

Introduction

Water is one of the most important and valuable resources that maintain the world's ecosystems and diversity, and is consumed by almost all life forms, of which approximately 70% is used for agriculture and other commercial purposes (Sharma et al. 2022; Chong et al. 2023). The rapid pace of uncontrolled modernization, urbanization, and industrialization in the face of increasing demands due to increasing population growth has greatly affected the quality and quantity of water reservoirs. Water resources are at risk of contamination in developing countries where sanitation and hygiene facilities are scarce. Polluted water significantly impacts the health of communities, aquatic ecosystems, the sustainability of the natural environment, and the economic and social well-being of society (Kefeni et al., 2017; Sharma et al. 2022; Chong et al. 2023). Water pollution usually occurs due to the discharge of large amounts of untreated wastewater from industries such as food, textile, pharmaceutical, and metallurgy into surface and groundwater resources (Wu et al. 2021; Kumar et al. 2022). According to the United Nations World Water Development Report, in 2020, approximately 4 billion people faced serious physical water scarcity for at least one month a year (Tee et al. 2022). Therefore, to this day, it remains a global problem and a source of concern all over the world. It is essential to purify industrial wastewater containing organic and inorganic-based pollutants, which

damagingly affect living organisms in different ecosystems before they mix into clean water resources and soil.

Various physical, chemical, and biological techniques such as ion exchange, adsorption, reverse osmosis, electrodialysis, coagulation, flotation, ultrafiltration, membrane separation, ozonation, and others are applied for wastewater treatment, which is disadvantageous in terms of environmental damage and high cost (Siddiqui et al. 2018; Amusat et al. 2021; Yan et al. 2022; Abdul Kader 2022). However, these treatment methods, except adsorption, are insufficient to provide the desired amount of water with the highest quality, and they are environmentally damaging and costly, so they have been used less recently. Adsorption is a more preferred technique than other techniques due to its low cost, high efficiency, easy use, and simplicity of design. Recently, the increasing interest in the production of low-cost, effective, reusable, and eco-friendly sustainable adsorbents further increases the usability of this method. To this end, interest in the evaluation of industrial and agro-industrial solid wastes as a sustainable precursor in the production of activated carbon (AC), which exhibits much greater adsorption capacity than other adsorbents, is increasing day by day (Danish and Ahmad 2018; Xiao et al. 2020; Wu et al. 2021). ACs are a carbon-rich adsorbent group with a high surface area and large pore structure. However, it limits the practical use of powdered AC due to the disadvantages such as clogging of filters in the filtration method used to separate it and causing secondary pollution by being thrown away with the process sludge after being used in water and wastewater treatment (Zhang et al. 2007; Shao et al. 2012). To eliminate these disadvantages, studies on synthesizing easily removable activated carbon-based magnetic nanohybrids with magnetic separation techniques loaded with magnetic nanoparticles to solve environmental problems have recently gained importance (Xiao et al. 2020; Sirajudheen et al. 2021).

Magnetic nanoparticles have received great attention due to their special superparamagnetic properties and high adsorption capacity due to their high surface area and large pore volume. Mainly, transition metal oxides with spinel structure, commonly called ferrite, are among the most important magnetic nanoparticles (Kefeni et al., 2017). Ferrites are classified as spinel (MFe_2O_4 , where M = divalent metal cation as Mn, Fe, Co, Ni, Co, Zn, etc.), garnet ($M_3Fe_5O_{12}$, where M = rare earth cations), hexaferrite ($SrFe_{12}O_{19}$ and $BaFe_{12}O_{19}$) and orthoferrite ($MFeO_3$, M = rare earth cations) according to their crystal structures and magnetic properties (Litsardakis et al. 2007; Reddy and Yun 2016; Yunang Kefeni et al., 2017). Compared to others, cobalt ferrite ($CoFe_2O_4$; CoF) nanoparticles are more preferred in adsorbent surface modification due to their strong anisotropy, high coercivity at room temperature, high saturation magnetization, good mechanical and chemical stability (Liang et al. 2019; Mmelesi et al. 2021).

According to our detailed literature review, there are very few studies on plant waste-based magnetic-AC nanohybrid production loaded with CoF nanoparticles and their use as adsorbents in treating water containing organic and inorganic pollutants. This study aims to (1) synthesize a novel magnetically separable nanohybrid material (CoF@NAC) with microwave-assisted chemical co-precipitation method of nigella sativa (*Nigella sativa* L.) industrial processing solid waste (NW)-derived AC (NAC) loaded with CoF nanoparticles, (2) characterize with different physicochemical techniques, (3) investigate its performance in removing water pollutants such as methylene blue (MB) dye and lead (Pb) heavy metal, which frequently detected in industrial wastewater, and (4) test its reusability stability as an adsorbent.

Materials and methods

Materials and reagents

NAC was produced under conditions of optimum impregnation ratio (1:1, $w_{K_2CO_3}/w_{NW}$), carbonization temperature (900 °C), and carbonization/activation time (1 h) determined by

potassium carbonate (K_2CO_3 , purity 99%) chemical activation of wastes (NW) sourced from A local *Nigella sativa* oil production facility in Diyarbakir, Turkey. The determination of its optimal preparation conditions according to the effect of process variables and some physicochemical characterizations were examined and interpreted in detail in our previous study (Teymur et al. 2023). Chemicals such as cobalt(II) nitrate hexahydrate ($Co(NO_3)_2 \cdot 6H_2O$, $\geq 99\%$), iron(III) nitrate nonahydrate ($Fe(NO_3)_3 \cdot 9H_2O$, 98%), sodium hydroxide (NaOH, 99%) to synthesize CoF nanoparticles and CoF@NAC, and methylene blue (MB) (Chemical formula: $C_{16}H_{18}ClN_5S$; Molecular weight: 319.9 g/mol; Chemical class: cationic dye; Ionization: basic; Maximum wavelength: 665 nm), lead(II) nitrate (Pb(II)) (Molecular formula: $Pb(NO_3)_2$; Molecular weight: 331.2 g/mol; Chemical class: heavy metal; Ionization: cationic; λ_{max} : 283 nm) selected as adsorbates for adsorption experiments were obtained from Sigma-Aldrich Co. (Ankara, Turkey), and were used in AR grade. Fresh ultra-distilled water was used in all solution preparation and washing processes.

Synthesis of CoF@NAC nanohybrid

The CoF@NAC nanohybrid material was synthesized, which is shown in the schematic representation in **Fig. 1**, using microwave-assisted chemical co-precipitation as reported in a previous study (Prabhakaran et al. 2018). First, a 40 ml solution with a 1:2 stoichiometric molar ratio of Fe (III) and Co (II) ions was added into a 100 ml Teflon beaker containing 0.39 g of NAC. This mixture was refluxed at 80 °C for 30 min by adding 20 mL of 2.5 M NaOH solution dropwise on a magnetic stirrer. Then, the formed CoF@NAC was magnetically separated from the medium using an external magnet, and exposed to 600 W microwave irradiation for 3 min. Finally, to remove unreacted chemicals in the synthesized material structure, it was washed repeatedly with ultra-distilled water until the ambient pH was approximately 7. It was then dried in an oven at 110 °C overnight and stored in a dark glass container to be used as an adsorbent

material in characterization and subsequent adsorption experiments. Pure CoFe_2O_4 nanoparticles were synthesized using similar steps without the addition of NAC.

Fig. 1 Schematic representation of CoF@NAC synthesis.

Characterization

The surface morphology and elemental compositions were viewed using a Leo-Evo 40 model scanning electron microscope (SEM) outfitted with a Bruker-125 eV model energy-dispersive X-ray (EDX) detector. Pore characteristics such as BET (Brunauer-Emmett-Teller) surface area (m^2/g), total pore volume (cm^3/g), micropore volume (cm^3/g), and average pore diameter (nm) were obtained from nitrogen adsorption-desorption isotherm and pore size distribution analysis data at 77 K using a Quantachrome-Quadrasorb Evo 4 model surface area and porosity analyzer. The mesopore volume (cm^3) was determined by subtracting the micropore volume from the total pore volume while the micro-and mesoporesity (%) were based on the total pore volume. The crystal structure was elucidated from the X-ray diffraction (XRD) pattern obtained in the range of $10-80^\circ 2\theta$ with a scanning speed in $2^\circ/\text{min}$ increments using monochromatic copper ($\text{Cu K}\alpha$, $k = 1.541 \text{ \AA}$) radiation at 40 kV. PANalytical Empyre model powder XRD device. The structural defects were brightened by scanning the spectral range of $1000-2000 \text{ cm}^{-1}$ with a 1% laser power input and 50x optical magnification at 532 nm using a Renishaw InVia model Raman microscope equipped with an excitation laser. The magnetic property was measured with a magnetic field of $\pm 30 \text{ kOe}$ at room temperature using a Lakeshore 7404 model vibrating sample magnetometer (VSM). Qualitative identification of surface functional groups was detected between 4000 and 450 cm^{-1} at a resolution of 4 cm^{-1} and 20 scans using a Scientific-Nicolet iS20 model Fourier transform infrared (FT-IR) spectrophotometer. The basic and oxygenic acidic groups on the surface, which are lactonic, phenolic, and carboxylic acid groups,

were measured by Boehm's selective neutralization method mentioned in an earlier study (Kyzas et al. 2016). This method is based on the principle that the oxygen groups on the sample surface have different acidity and can be neutralized with bases with increased strength (i.e. sodium bicarbonate (NaHCO_3) < sodium carbonate (Na_2CO_3) < sodium hydroxide (NaOH)). The total acidity and basicity of the surface were determined from the amount of NaOH and HCl , respectively. For this purpose, approximately 0.05 g of the CoF@NAC was placed in Erlenmeyer flasks containing 50 mL of 0.1 M NaOH or Na_2CO_3 or NaHCO_3 aqueous solution. The mixtures were stirred in a water bath at room temperature at a constant speed of 120 rpm for 48 h. Then, the suspensions were filtered on membrane filters (0.45 μm). To determine the content of oxygenated acidic groups, 20 mL of the filtrates were back-titrated with a standard 0.1 M HCl solution. The pH at the point where the surface charge is zero (pH_{ZCP}), which reflects the general chemical character of the surface, was determined using the method described in a previous study (Jang and Kan 2019). Briefly, 50 mL of NaCl solution (0.01 M) adjusted from 3.0 to 10.0 using 0.1 M NaOH or HCl solution was placed in a 100 mL Erlenmeyer and 0.01 g of CoF@NAC was added. They were filtered after shaking in a thermostatted water bath at 20 $^{\circ}\text{C}$ and 150 rpm for 48 h, and the final pH of the supernatants was measured. The point of intersection of the curves in the plot (not shown) drawn ΔpH ($\text{pH}_{\text{initial}} - \text{pH}_{\text{final}}$) vs. $\text{pH}_{\text{initial}}$ was taken as the pH_{ZPC} of the CoF@NAC .

Batch adsorption experiments and evaluation

Batch adsorption experiments were performed to determine the optimum adsorption performance of CoF@NAC nanohybrid material for water pollutants such as MB and Pb(II) , which were selected as model adsorbates, and to investigate their adsorption mechanism. For this purpose, these experiments were done by agitating the mixture containing 0.05 g of adsorbent with 50 ml of known adsorbate concentration solution in 100 ml Erlenmeyer flasks

at 120 rpm in a thermostated water bath shaker. The effects of important experimental parameters such as adsorbent dose (0.05-0.1 g), adsorbate concentration (650-800 mg/L), contact time (0-360 min), and system temperature (293-313 K) at their natural pH levels (5.55 for MB; 5.06 for Pb(II)) in water on the adsorption of each adsorbate were investigated. After each experiment, the CoF@NAC in the suspension was isolated with an external magnet, then the solution was filtered through millipore membranes (0.45 µm). The residual concentrations of each adsorbate were measured using Shimadzu UV-1280 model UV-Vis and PerkinElmer AAnalyst, 100A model atomic absorption spectrophotometers at their maximum wavelengths (665 nm for MB and 274 nm for Pb(II)).

Kinetic experiments were performed by adding 50 mL of adsorbate solution with initial concentrations varying between 650-800 mg/L into 0.05 g of adsorbent in 100 mL Erlenmeyer flasks at a system temperature of 25 °C, a shaking rate of 120 rpm, contact time intervals ranging from 5 to 360 minutes, and the natural pH level (5.55 for MB and 5.06 for Pb(II)) in the water of examined adsorbates. The quantity (Q_t , mg/g) removed at any time t (min) was calculated from Eq. 1.

$$Q_t = \frac{(C_0 - C_t)V}{m} \quad (1)$$

where, C_0 (mg/L), C_t (mg/L), m (g), and V (L) represent the initial concentration of adsorbate, the adsorbate concentration at time t , the mass of the adsorbent, and the volume of the adsorbate solution, respectively. The kinetic findings of each adsorption system were modeled by evaluating the linear equations (Eqs. 2 and 3) of commonly used Lagergren's pseudo-first-order (PFO) (Lagergren 1898) and Ho-McKay's pseudo-second-order (PSO) (Ho and McKay 1998) kinetic models. In addition, these findings were analyzed given Eq. (4) of the intraparticle diffusion (IPD) model based on Weber-Morris's theory (Weber and Morris 1963) to determine the effective rate-controlling step.

$$\text{PFO:} \quad \ln(Q_e - Q_t) = \ln Q_e - k_1 t \quad (2)$$

200 PSO:
$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{1}{Q_e} t \quad (3)$$

201 IPD:
$$Q_t = k_{id} t^{0.5} + C \quad (4)$$

202 where k_1 (1/min), k_2 (g/mg/min), and k_{id} (mg/g min^{0.5}) are rate constants for PFO, PSO, and IPD
 203 models, respectively. C (mg/g) is a constant for the liquid-solid boundary layer thickness.

204 Equilibrium isotherm experiments were carried out at various temperatures (293, 303, and
 205 313 K) using solutions with concentrations of each adsorbate varying between 650-1000 mg/L
 206 at predetermined optimum removal conditions for each adsorption system. The quantity (Q_e ,
 207 mg/g) removed at equilibrium was calculated from Eq. (5).

208
$$Q_e = \frac{(C_0 - C_e)V}{m} \quad (5)$$

209 where C_e is the adsorbate concentration in the medium at equilibrium (mg/L). Equilibrium
 210 isotherm findings are modeled by evaluating the linear equations of commonly used Freundlich
 211 (Freundlich 1906) and Langmuir (Langmuir 1918) isotherm models given in Eqs. (6 and 9).

212 Freundlich:
$$\ln Q_e = \ln K_F + \frac{1}{n_F} \ln C_e \quad (6)$$

213 Langmuir:
$$\frac{C_e}{Q_e} = \frac{1}{Q_m K_L} + \frac{1}{Q_m} C_e \quad (7)$$

214 where K_F ((mg/g) (L/mg)^{1/n}) is the Freundlich constant. $1/n_F$ is the adsorption density, which
 215 indicates that the process is favorable and unfavorable, respectively, if this value is less than
 216 and greater than one for an adsorption system (Zhou et al. 2022). Q_m (mg/g) is the maximum
 217 adsorption capacity. K_L (L/mg) is the Langmuir constant.

218 The model that best describes the kinetic and isotherm findings was decided based on the
 219 largest and smallest statistical analysis data such as the regression coefficient (R^2) and the
 220 normalized standard deviation (Δq , %) determined utilizing the experimentally removed

221 quantity (Q_{exp} , mg/g) and calculated removed quantity (Q_{cal} , mg/g), and experimental data
 222 number (N) data in Eq. 8 and Eq. 9, respectively (Shi et al. 2023).

$$223 \quad R^2 = 1 - \left[\sum_{i=1}^n (Q_{e,\text{exp}} - Q_{e,\text{cal}})^2 / \sum_{i=1}^n (Q_{e,\text{exp}})^2 - \left((\sum_{i=1}^n Q_{e,\text{exp}})^2 / N \right) \right] \quad (8)$$

$$224 \quad \Delta q(\%) = 100 \sqrt{\sum_{i=1}^N [(Q_{e,\text{exp}} - Q_{e,\text{cal}}) / Q_{e,\text{exp}}]^2 / N - 1} \quad (9)$$

225 In addition, Langmuir's dimensionless separation factors (R_L), which indicate the nature of
 226 an adsorption process, were calculated from Eq. (10) to estimate whether the process was
 227 favorable for each adsorption system. If the R_L value is equal to zero, between zero and one,
 228 equal to one, and greater than one for an adsorption system, its nature is considered to be
 229 irreversible, favorable, linear, and unfavorable, respectively (Al-Musawi et al. 2022).

$$230 \quad R_L = \frac{1}{1 + K_L C_0} \quad (10)$$

231 Furthermore, thermodynamic parameters such as Gibbs free energy (ΔG^0 , kJ/mol), enthalpy
 232 (ΔH^0 , kJ/mol), entropy (ΔS^0 , kJ/(mol K)) changes were calculated for the analysis of
 233 thermodynamic applicability, spontaneity, and physical or chemical nature of the process in the
 234 studied adsorption processes. The determination of ΔG^0 values at the temperatures studied was
 235 calculated using the Van't Hoff equation (Eq. 11), which relates the changes in the corrected
 236 equilibrium constant for adsorption from solution to temperature by Lima et al (Lima et al.
 237 2019).

$$238 \quad \Delta G^0 = -RT \ln K_e^o \quad (11)$$

239 where R (8.314 J/K mol) is the universal constant of ideal gases. T (K) is the temperature. K_e^o
 240 is the dimensionless constant of thermodynamic equilibrium and is calculated from Eq.12.

$$241 \quad K_e^o = 1000(K_L M_w \gamma) \quad (12)$$

242 where 1000 is the conversion coefficient from L/mg to L/g. K_L (L/mg) is the equilibrium
 243 constant obtained from the intercept of the Langmuir isotherm linear plots at the temperatures

studied. M_w (g/mol) is the molar mass of the adsorbate. γ is the dimensionless activity coefficient of the adsorbate, which is assumed to be 1.0 since $\gamma \approx 1$ in the case of a very dilute solution. The average ΔH° and ΔS° values in the temperature range studied for each adsorption system were calculated from the slope and intercept values, respectively, of the linear graphs ($\ln K_e^\circ$ vs. $1/T$) plotted according to Eq. 14, which was derived by adapting Eq. 11 to Eq. 13, which reflects the third law of thermodynamics.

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (13)$$

$$\ln K_e^\circ = -\frac{\Delta H^\circ}{RT} + \frac{\Delta S^\circ}{R} \quad (14)$$

Desorption and regeneration experiments

To determine the reusability stability of the studied adsorbate-loaded CoF@NAC, batch adsorption-desorption cycle experiments were carried out using hydrochloric acid (HCl), nitric acid (HNO₃), methanol (CH₃OH) solutions with 0.5 M, and double-distilled water for effective desorbent agent selection. For this purpose, first, the adsorption experiments of the adsorbates examined on the CoF@NAC were carried out by agitating for 2 h at a stirring speed of 120 rpm and a temperature of 298 K the mixtures containing 0.05 g of CoF@NAC and 50 mL of adsorbate solution in 100 mL Erlenmeyer flasks. Then, desorption experiments were carried out with desorbent agents used to determine the chemical eluent that effectively removes each adsorbate from the CoF@NAC surface. To determine the reusability stability of the CoF@NAC for each adsorbate used, six-cycle adsorption-desorption experiments were carried out with the determined effective desorbent agent. The adsorbed and desorbed concentrations of each adsorbate were measured as mentioned above, and the adsorbed and desorbed quantities (Q_a and Q_d , mg/g) were calculated from them. Desorption and adsorption efficiencies were calculated from Eq. (15) and Eq. (16), respectively.

$$\text{Desorption efficiency (\%)} = \frac{Q_d}{Q_a} \cdot 100 \quad (15)$$

$$\text{Adsorption efficiency (\%)} = \frac{(C_i - C_e)}{C_i} \cdot 100 \quad (16)$$

270

271 **Results and discussion**

272 *Characterization of CoF@NAC*

273 *SEM-EDX/Mapping Analyses*

274 **Fig. 2** shows the SEM images, EDX spectra, and elemental composition maps of NAC and
 275 before and after MB and Pb(II) adsorption of CoF@NAC. It is seen from this figure that NAC
 276 has a honeycomb-like surface with open pores of various sizes, and the CoF@NAC surface
 277 covers most of the open pore gates by clustering the CoF nanoparticles on certain parts of the
 278 NAC surface. This supports the successful synthesis of the magnetic nanohybrid material.
 279 Furthermore, the existence of iron (18.48%) and cobalt (9.74%) elements along with carbon
 280 (52.58%) and oxygen (17.16%) elements, which are the main elements of the AC phase, in the
 281 EDX spectra and elemental mapping images confirms that the surface is covered with CoF
 282 nanoparticles.

283

284 **Fig. 2** SEM images/EDX spectra of NAC and before and after MB and Pb(II) adsorption of
 285 CoF@NAC

286

287 *Textural structure analysis*

288 Elucidating the textural structure of a solid is very important in applications, especially in
 289 adsorption studies. Preliminary information about the textural structures of solids is provided
 290 by comparing the similarity of nitrogen adsorption-desorption isotherms and pore size

distributions with the types recommended by IUPAC (International Union of Pure and Applied Chemistry). The nitrogen adsorption-desorption isotherms and hysteresis loops that solids exhibit according to their textural structures are classified by IUPAC as Type-I -Type-VI and H1-H4, respectively (Sing 1985). In addition, solids are defined by IUPAC in three groups according to their pore sizes: microporous (less than 2.0 nm), mesoporous (between 2.0 and 50.0 nm), and macroporous (larger than 50.0 nm) (Thommes et al. 2015) .

The nitrogen adsorption-desorption isotherm and pore size distribution (inset) at 77 K of CoF@NAC nanohybrid are presented in **Fig. 3a**, and the textural characteristics (BET surface area, total pore volume, micropore volume, and average pore diameter) determined evaluating in the BET and Barrett-Joyner-Halenda (BJH) methods of the data in this figure are listed in **Table 1**. It appears that the adsorption-desorption isotherms of CoF@NAC are a combination of Type I and Type IV with an H3 hysteresis loop indicating the presence of mesopores with cylindrical pore geometry in the materials, indicating the coexistence of micro- and mesopores, respectively, which is also supported by its textural characteristics given in **Table 1**. It is seen from this table that its BET surface area, total pore volume, micropore volume, mesopore volume, microporosity, mesoporosity, and average pore diameter values have pore characteristics of 729 m²/g, 0.426 cm³/g, 0.164 cm³/g, 0.262 cm³/g, 38.5%, 61.5%, and 2.4 nm, respectively. The comparison of textural properties of CoF@NAC with CoF-loaded ACs reported in the previous studies is given in **Table 2**. It is seen from this table that CoF@NAC has a higher BET surface area than the others. Accordingly, it indicates that CoF@NAC will be a more effective adsorbent, especially in treating water contaminated with toxic chemicals.

Fig. 3 N₂ adsorption-desorption isotherms and pore size distribution (inset) of CoF@NAC (a), XRD pattern of CoF and CoF@NAC (b), Raman spectra of CoF@NAC (c), Magnetic hysteresis loops of CoF and CoF@NAC, and photograph (inset) of adsorption system before and after

magnetic separation (d), FT-IR spectrum before and after MB and Pb(II) adsorption of CoF@NAC (e), pH_{PZC} determination of CoF@NAC (f)

Table 1 Textural, surface chemical, and magnetic characteristics of CoF@NAC

Table 2 Comparison of textural properties of CoF/NAC and some CoF-loaded carbonaceous materials reported in the literature

Crystal structure analysis

Fig. 3b shows the XRD patterns of CoF nanoparticles and CoF@NAC in the $2\theta = 10\text{-}80^\circ$ angle range. As seen from this figure, the XRD patterns of CoF and CoF@AC almost overlap with each other with the characteristic peaks at 18.9° , 30.4° , 35.3° , 43.7° , 54.1° , 57.3° , and 62.8° which corresponding to (111), (220), (311), (400), (422), (511) and (440) crystal planes defining the cubic spinel lattice of CoF nanoparticles (JCPDS card no. 22-1086), respectively, as stated in previous studies (Karthikeyan et al. 2020; Wu et al. 2021; Al-Hawary et al. 2023). Furthermore, two different diffraction angles of 23.1° and 43.5° belong to AC in the CoF@NAC structure, as observed in the CoF-loaded carbonaceous materials synthesized in previous studies (Xu et al. 2014; Karthikeyan et al. 2020; Wu et al. 2021).

Graphitization degree analysis

Fig. 3c illustrates the Raman spectrum of CoF@NAC. The broad peaks at 1352 and 1589 cm^{-1} in this figure relate to the CoF@NAC's D- and G- bands, respectively. The D-band is due to sp^3 structural defects for scattering due to disorder caused by the loss of hexagonal symmetry (Asadi et al. 2022). The G-band describes the stretching vibration of the carbon atoms on the sp^2 hybrid surface (Liao et al. 2021). In addition, the intensity ratio ($I_{\text{D}}/I_{\text{G}}$), which indicates the

degree of structural disorder, was calculated as 0.85 for CoF@NAC. In our previous study, this ratio was reported to be 0.80 for NAC (Teymur et al. 2023). The increase in this ratio for CoF@NAC compared to AC indicates that it has a more disordered and defective structure. This is most likely due to the presence of CoF nanoparticles in the CoF@NAC structure.

VSM analysis

Fig. 3d shows the magnetic hysteresis loops., indicating that they will exhibit superparamagnetic behavior, depicting the magnetization measurements of the synthesized CoF and CoF@NAC. From these analysis results, the saturation magnetization (M_s , emu/g), remnant magnetization (M_r , emu/g), and intrinsic coercivity (H_c , Oe) values of CoF and CoF@NAC are 44.68 and 23.29 emu/g, 11.74 and 5.49 emu/g, and 740.0 and 693.3 Oe, respectively. The decrease in the values of CoF@NAC compared to bare CoF nanoparticles can be attributed to the presence of non-magnetic NAC particles in the structure of the CoF@NAC, as reported in previous studies (Liang et al. 2019; Al-Musawi et al. 2022; Askari et al. 2023), confirming that the synthesis was successful. In addition, to understand the magnetic behavior of the CoF@NAC, a magnet was placed on the outer surface of the glass container where the CoF@NAC and adsorbate solution were located after adsorption, as seen in the additional photograph in the same figure. As a result of this test, it was observed that the CoF@NAC was completely collected in a short time at the level of seconds without leaving any secondary pollution in the medium, which supports that it has high magnetic sensitivity.

Surface chemical analysis

The FTIR spectrum of CoF@NAC before and after adsorption is shown in **Fig. 3e**. The peak at 3437.1 cm^{-1} belongs to the -OH stretching vibration. The peak at 1559.05 cm^{-1} is attributed to the stretching vibration of the amide group (-NH). Furthermore, the peak at 1413.7 cm^{-1}

confirms the presence of carboxylic group. Peak values show that amides and carboxylic acid groups are available for binding (Siddiqui et al. 2018). The sharp peak at 585.89 cm^{-1} belongs to the Fe/Co-O tetrahedral stretching region in spinel oxide (Mahmoud et al. 2022). This confirms that CoF@NAC was successfully synthesized. Furthermore, the Boehm titration findings and pH_{ZPC} value of the composite are listed in **Table 1**. The total surface acidity, which is the sum of the quantities of functional groups such as carboxylic (0.5401 mEq/g), phenolic (0.0185 mEq/g), and lactonic (0.5023 mEq/g), is 1.0609 mEq/g, and the total surface basicity is 1.0717 mEq/g. In addition, the pH_{ZPC} value was determined as 8.2 from **Fig. 3f**. These results reveal that the surface has a weak basic character, which is confirmed by its FT-IR spectra.

Cost analysis of CoF@NAC synthesis

The sustainable applicability of a technique for the adsorptive removal of pollutants from water in the commercial field depends greatly on the cost of adsorbent production together with its efficiency. The cost of synthesis of magnetic activated carbon nanohybrid material from biomass is generally determined by consumptions such as precursor supply and transportation fees, chemicals for activation, magnetization, precipitation, and neutralization, energy for carbonization, and water for washing.

The approximate cost per kilogram of CoF@NAC was determined by averaging the costs of three separate in vitro syntheses. The approximate cost per kilogram of NAC was determined to be \$2.830 in our previous study. In addition to this cost, the approximate net cost per kilogram of CoF@NAC is calculated as \$3.615, with costs of \$0.435 for $\text{Co}(\text{NO}_3)_2$ and FeCl_3 consumption for magnetization, \$0.019 for NaOH consumption in chemical precipitation, \$0.19 for water consumption and \$0.141 for energy consumption in microwave heating, distilled water production, and drying. The approximate total cost of its commercial production is \$3.977 adding a 10% additional charge to its net cost.

Effect of experimental parameters and optimization

Optimum removal conditions of MB and Pb(II) by CoF@NAC were estimated by examining their effects on their adsorption by systematically changing the experimental parameters such as CoF@NAC dose, contact time, adsorbate concentration, and system temperature at natural pH in the water.

Fig. 4a demonstrates the effect of CoF@NAC dose on the adsorptions of adsorbates used by CoF@NAC. As can be seen from this figure, the quantities of MB and Pb(II) removed decreased from 224.2 to 163.5 mg/g and to 163.4 mg/g from 106.3, respectively, by increasing the CoF@NAC dose from 0.05 to 0.10 g/50 mL. This decrease is probably due to reduced effective adsorption sites due to surface overlap/aggregation with increasing CoF@NAC dose, as reported in previously published studies (Sun et al. 2021; Gohr et al. 2022; Farooq et al. 2022).

Fig. 4b shows the effect of their concentrations/contact time on the adsorptions from the CoF@NAC surface of the adsorbates used. It can be seen from this figure that the quantity removed for adsorbates used increases rapidly from the beginning up to 120 min, and then slowly up to 240 min and then reaches a constant value. The fact that the adsorption is initially fast and then slow may be probably due to the large number of empty active adsorption sites on the CoF@NAC surface at the beginning and then gradually decreasing them, respectively. Also, it is seen from the same figure that by increasing the concentration of MB and Pb(II) ions from 650 to 800 mg/L, their removed quantities increase from 283.4 to 331.7 mg/g and 218.8 to 238.1 mg/g, respectively. This increase is due to the increase of adsorbate ions to be adsorbed at the active adsorbed sites on the adsorbent surface in the solution phase at increasing adsorbate concentrations. Similar trends have been noted in previously published studies by other researchers (Maneerung et al. 2016; Farooq et al. 2022). **Fig. 4c** displays the effect of system

temperature on the adsorption of the CoF@NAC surface of the adsorbates used. The removed quantities of MB increased from 339.6 to 402.7 mg/g. That of Pb(II) decreases from 308.7 to 258.5 mg/g by increasing the system temperature from 293 K to 313 K. This states that the MB and Pb(II) adsorption processes are endothermic and exothermic, respectively. The increase in MB adsorption with increasing system temperature may be due to the increase in interactions between CoF@NAC and adsorbate ions, as well as the decrease in solution viscosity, which facilitates the diffusion of adsorbate ions into the pores through the outer boundary layer between the solution phase and the CoF@NAC. The decrease in the adsorption of Pb(II) ions with increasing temperature is most likely due to the mobility increase of physically adsorbed Pb(II) ions, resulting in an increased tendency to migrate to the solution phase to leave the CoF@NAC surface. Similar observations have been reported in previous studies (Djilani et al. 2015; Tang et al. 2017).

The optimal adsorption conditions for equilibrium isotherm experiments of the adsorbates used from the above-mentioned experimental results were chosen as 0.05 g /50 mL of CoF@NAC dose, 240 min of contact time, 40 °C for MB, and 20 °C for Pb(II) of system temperature.

Fig. 4 Effects of CoF@NAC dose (a), contact time/initial adsorbate concentration (b), and system temperature (c) on the MB and Pb(II) adsorption capacity of CoF@NAC

Kinetic analysis

The investigation of adsorption kinetics is necessary to determine the rate of the adsorption process, which allows the adsorbent to be used practically in the water treatment system [15].

Fig. 4b shows nonlinear kinetic plots of CoF@NAC-MB/Pb(II) adsorption systems at different adsorption time intervals and adsorbate concentrations. The kinetic and statistical parameters

were calculated from the linear plots ($\ln(Q_t - Q_e)$ vs. t) and (t/Q_t vs. t) (**Fig. S1a, b** in Supplementary (S) material) drawn according to the linearized equations (Eqs. (2) and (3)) of the PFO and PSO kinetic models of the kinetic data in **Fig. 4b**, and are given in **Table 3**. Each adsorption system fits the PSO kinetic model with highest R^2 (>0.9992 for MB and >0.9991 for Pb(II)) and lower Δq (%) (<2.01 for MB, and <1.31 for Pb(II)) values calculated from Eqs. (8) and (9). In addition, the fact that the experimentally removed quantity ($Q_{e, \text{exp}}$) determined for each adsorption system are very close to those ($Q_{e, \text{cal}}$) calculated from the PSO kinetic model, confirms that they fit this model (**Table 3**). Furthermore, as seen in **Table 3**, PSO rate constant values decreased with increasing adsorbate concentration for each adsorption system. This decrease is probably due to the decrease in the affinity between adsorbate ions and the CoF@NAC surface as a result of the decrease of active adsorbing sites on the surface, or the inhibition of the orientation of adsorbate ions to the surface by increasing the boundary layer thickness between solution and adsorbent, with increasing adsorbate concentration (Ahmed et al. 2017; Fuat and Cumali 2021).

Furthermore, since PFO and PSO models cannot determine the effective rate step in an adsorption process, kinetic data for this are analyzed in the IPD model based on the Weber-Morris theory. In general, the rate of an adsorption process is controlled by film diffusion (or boundary layer diffusion) or intra-particle diffusion (or pore diffusion) steps (Lee et al. 2022). To describe the adsorption diffusion mechanism in detail for each adsorption system and to determine the rate-controlling step, the plots (Q_t vs. $t^{0.5}$) drawn by assessing kinetic data of each adsorbate in **Fig. 4b** in the equation (**Eq. 4**) of Weber-Morris's IPD model are presented in **Fig. S1c**. If the graph drawn according to this model gives a straight line at the origin, it indicates that the rate-controlling step is only the IPD step, otherwise, it indicates that the mechanism consists of a complex mechanism that is effective in other steps (Gao et al. 2013). The IPD graphs for the investigated adsorption systems (**Figure S1c**) consist of lines that do not pass

through multiple-step origins, indicating that the rate-controlling step in adsorption mechanisms is not only the IPD step. The k_{id} and C constants of the IPD model for each adsorption system were determined from the slope and intercept values of the second lines in **Fig. S1c** and are given in **Table 3**. This table shows that the C values, which measure the boundary layer thickness between the solution adsorbate, increase as the concentration of each adsorbate increases. The fact that the C value is close to zero for any adsorption process indicates that the IPD step is the more effective rate-controlling step in the mechanism, otherwise, other steps are also effective (Ahmed et al. 2017). This confirms that more than one rate-controlling step is effective in the mechanism of each adsorption system studied.

Table 3 Kinetic parameters for adsorption at different concentrations of MB and Pb(II) onto CoF@NAC

Isotherm and thermodynamic analysis

Equilibrium adsorption isotherm is an essential adsorption system design tool in describing the interaction between adsorbents and adsorbents and determining the maximum adsorption capacity. In this context, Giles et al. classified the adsorption isotherm shapes as S, L, H, and C-types to explain the relationship between adsorbent and adsorbate in adsorption from solution (Giles et al. 1974). **Fig. 4c** shows nonlinear isotherm plots of CoF@NAC-MB/Pb(II) adsorption systems at 293, 303, and 313 K. The isotherms of the investigated adsorption systems resemble the L-type in Giles' isotherm classification, which indicates the high affinity between the adsorbent and the examined adsorbates. The equilibrium isotherm data in this figure were evaluated in the linearized equation (**Eqs. (6 and 7)**) of Freundlich and Langmuir isotherm models. The isotherm parameters, Langmuir separation factors, R_L , R^2 , and $\Delta q\%$ statistical parameters of the linear graphs ($\ln C_e$ vs. $\ln Q_e$) and (C_e/Q_e vs. C_e) (**Fig. S2a, b**) drawn

according to these models were calculated and listed in **Table 4**. This table shows that all investigated adsorption systems fit the Langmuir isotherm model with the highest R^2 (>0.9506 for MB and >0.9501 for Pb(II)) and lower Δq (%) (<1.64 for MB, and <1.56 for Pb(II)) values calculated from Eqs. (8) and (9). Maximum adsorption capacities from the Langmuir linear graphs drawn at 293, 303, and 313 K temperatures of MB and Pb(II) were determined as 588, 625, and 667 mg/g and 455, 400, and 370 mg/g, respectively. The increase in the maximum adsorption capacity of MB and the decrease of Pb(II) with the increase in system temperature indicate that the adsorption processes are endothermic and exothermic, respectively. A comparison of the MB and Pb(II) adsorption performances of CoF@NAC and some other adsorbents is listed in **Table 5**. In addition, the fact that the Freundlich, $1/n$, and R_L values determined for each adsorption system, are less than one indicates that the adsorption processes are favorable. Moreover, the decrease for MB and the increase for Pb(II) of these values with increasing system temperature confirm that their adsorption is more favorable at high and low temperatures, respectively.

Determining the thermodynamic parameters (ΔG° , ΔH° , and ΔS°) for an adsorption process provides essential information about whether it is spontaneous, endothermic, or exothermic, depending on the process temperature. These parameters for CoF@NAC-MB/Pb(II) adsorption systems were calculated van't Hoff plots ($\ln K_c^\circ$ vs. $1/T$) (**Fig. S3**) drawn by assessing in **Eqs. (11) and (14)** adapted to adsorption from the liquid phase of K_L constants in **Table 4** determined from the Langmuir isotherm model at different temperatures and listed in the same table. The ΔG° values at 20, 30, and 40 °C for MB and Pb(II) are -15.9, -16.7, and -17.5 kJ/mol and -19.4, -17.1, and -16.7 kJ/mol, respectively. Negative ΔG° values of the investigated adsorption processes indicate that they occur spontaneously. Additionally, the absolute values of ΔG° increase with increasing temperature for MB and decrease for Pb(II), revealing that their adsorption occurs more spontaneously and not, respectively, as the temperature increases. This

is consistent with the fact that their Q_m values in **Table 4** are higher for MB and lower for Pb(II) at higher temperatures. Furthermore, it has been stated that the process is physical when the ΔG° values are less than -20 kJ/mol in an adsorption process (Karthikeyan et al. 2020). Accordingly, it indicates that the processes in the adsorption systems examined occur via the physical adsorption mechanism. The average ΔH° and ΔS° values for the studied temperature range for the adsorption systems studied (**Table 4**) were determined from the slope and intercept values of the van't Hoff plots (Fig. S3), respectively, drawn according to Eq. 14. Average ΔH° values that provide information about the effective forces during the adsorption process are positive for MB (7.9 kJ/mol) and negative for Pb(II) (-42.4 kJ/mol), indicating that the adsorption processes are endothermic and exothermic, respectively. These values also confirm that the Q_m values in **Table 4** increase for MB and decrease for Pb(II), respectively, with increasing temperature. Furthermore, it has been stated that in an adsorption process, different physical forces such as Van der Waals forces, hydrophobic bond forces, hydrogen bond forces, coordination exchange and dipole bond forces, and chemical forces are effective when the ΔH° value is 4-10, 5, 2-40, 40, 2-29 and >60 kJ/mol, respectively (Mattson and Mark 1971). Accordingly, it supports that the examined adsorbates occur through a mechanism in which they are adsorbed on the CoF@NAC surface with physical forces. The average ΔS° values for MB and Pb(II) in the studied temperature range are 0.081 and -0.21 kJ/mol K, respectively. This indicates that the disorder in the adsorption systems increases and decreases, respectively. These values are slightly positive for MB and negative for Pb(II), indicating a slight increase or decrease in the degree of freedom of the adsorbate molecules at the adsorbent/solution interface due to weak interactions, respectively, as reported in a previous study (Wu et al. 2020).

Table 4 Isotherm and thermodynamics parameters for MB and Pb(II) adsorptions onto CoF@NAC at different temperatures

Table 5 Comparison of maximum adsorption capacity of MB and Pb(II) of CoF@NAC with other magnetic composite adsorbents reported in the literature.

Elucidating the adsorption mechanism

The adsorption mechanism of MB and Pb(II) by CoF@NAC was elucidated by comparing the SEM-EDX/mapping images and FT-IR spectra before and after adsorption and also by the relationship between the pH of the medium in which their adsorption was carried out and the surface pH_{ZPC} value of CoF@NAC.

Fig. 2 shows the SEM-EDX/mapping images of CoF@NAC before and after MB and Pb adsorption. When the SEM images are compared, the disappearance of various-sized pores and pits on the surface of CoF@NAC after their adsorption and the changes such as the surface image turning from dark to light support the fact that their adsorption has taken place. Moreover, when the EDX spectra and mapped images of CoF@NAC in the same figure are compared, in addition to the C, O, Fe, and Co elements present in its structure after their adsorption, the presence of the spectra of S, Cl, and Pb elements confirms that their adsorption has occurred.

Fig. 3e displays the FTIR spectra of CoF@NAC after adsorptions. When the FTIR spectra of the composite before and after adsorption were compared, it was observed that the -OH stretching peak of the composite at 3437.09 cm^{-1} shifted to 3322.46 , 3310.22 , and 3182.07 cm^{-1} after MB and Pb(II) adsorption, respectively. Additionally, the peak at 1413.7 cm^{-1} originating from the stretching of the carboxylic group (-OH) shifted to 1418.5 and 1402.8 cm^{-1} after MB and Pb(II) adsorption, respectively. The peak at 1216.42 cm^{-1} observed in the spectrum of CoF@NAC after MB adsorption is attributed to the S=O stretching vibration (Teymur and Güzel (2023)). This peak indicates that MB is adsorbed on the CoF/NAC surface. The shift of

the –OH stretching peak at 3437.09 cm^{-1} of CoF@NAC to 3182.07 cm^{-1} after Pb(II) adsorption may be due to the interaction between Pb(II) and O-H (Hu et al. 2016). The formation of new spectra and the shift of existing spectra indicate a good interaction between CoF@NAC and MB^+ and Pb(II) ions.

The possible adsorption mechanism of the investigated adsorbates can be elucidated by the change in surface chemical properties depending on the relationship between the pH_{ZPC} value of CoF@NAC, which was determined as 8.20 (**Table 1**), and the system pH values. In an adsorption mechanism, the chemical properties of the adsorbent and adsorbate in the medium play a very important role. In this context, it has been stated that the adsorbent surface is negatively and positively charged, respectively, depending on whether the pH of the medium in which adsorption takes place is greater or less than the pH_{ZPC} value of the adsorbent (Feng et al. 2022). Accordingly, the adsorption of the studied adsorbates is carried out at a pH lower than the pH_{pzc} value of CoF@NAC, so its surface is positively charged. MB and Pb are cationic in aqueous media and therefore they are in positively charged form. Accordingly, the positively charged investigated adsorbate ions are repelled by the protonated hydroxyl and/or carboxyl groups on the CoF@NAC surface, and their adsorption probably occurs via ion exchange between the protons on the surface, which is also supported by their ΔH° thermodynamic parameter and post-adsorption FTIR spectra.

The possible mechanism of the adsorption systems examined in line with the information mentioned above is depicted in **Fig. 5**.

Fig. 5 Possible MB and Pb(II) adsorption mechanism on the CoF@NAC

Regeneration and reuse of CoF@NAC

The economic feasibility of using any adsorbent to remove water pollutants from wastewater depends on its ability to be reused during multiple adsorption-desorption cycles (Farooq et al. 2022). **Fig. 6 (a) and (b)** show the drawings of the adsorption-desorption cycles related to the selection of the effective desorbent to remove the MB and Pb^{+2} adsorbed on the CoF@NAC, and its reusability stability with the determined effective desorbent, respectively. It is seen from **Fig. 6a** that desorption efficiencies calculated from Eq. (15) of MB and Pb(II) adsorbed on the CoF@NAC is 92.5, and 90.6 % with HCl, 62.1, and 71.5% with HNO_3 , 58.9, and 23.7% with CH_3OH and 21.6, 12.0 % with H_2O , respectively. From these findings, HCl was chosen as the effective desorbent for CoF@NAC-MB and Pb(II) adsorption systems. **Fig. 6b** shows that the CoF@NAC can be used with HCl desorption of MB and Pb(II) for up to four adsorption-desorption cycles with adsorption efficiencies of 59.2, and 64.0 %, respectively, calculated from Eq. (16). The gradual decrease in adsorption efficiency for the adsorbates studied at higher cycles is most likely due to the erosion of active adsorption sites on the CoF@NAC surface.

Fig. 6 Effect of different desorbents on desorption efficiencies of MB and Pb(II) (a) and reusability cycles for MB and Pb(II) adsorption efficiencies (b) onto CoF@NAC

Future research perspectives

From the findings in these experiments, we understood that utilizing *Nigella sativa* oil processing waste as a precursor in renewable and sustainable carbonaceous CoF-loaded magnetic nanohybrid adsorbent production is a very rational approach in terms of both environment and economy. Considering CoF@NAC's pore and structural properties and its ability to improve water containing MB and Pb(II), we aim to expand its usage areas by using it as an adsorbent in the treatment of water containing various water pollutants such as other antibiotics, dyes, and heavy metals in our future studies.

Conclusion

In the present study, magnetic CoF@NAC nanohybrid material synthesized by loading CoF nanoparticles from NAC was characterized, and its removal efficiency from aqueous solutions containing pollutants such as MB and Pb(II) in different chemical categories was tested. The results can be highlighted as follows:

- CoF@NAC has properties such as 729 m²/g BET surface area, 2.4 nm average pore diameter, 61.5% mesoporosity, and 14 emu/g saturated magnetization.
- The approximate cost per kilogram of CoF@NAC for commercial production is \$3.977.
- The best conditions to reach the highest adsorption capacity for the studied adsorbates were: CoF@NAC dose = 0.05 g, contact time = 180 min, system temperature = 313 K for MB, and 293 K for Pb(II).
- The adsorption kinetic data at various concentrations for each adsorbate fit the PSO kinetic model.
- The rate-controlling step in the adsorption of each adsorbate occurs through a complex mechanism in which not only the intra-particle diffusion step but also others are effective.
- The isotherm data obtained at different temperatures for each adsorbate was the best fit for the Langmuir isotherm model.
- The maximum removed quantities for MB and Pb(II) from the Langmuir isotherm model were determined as 667 and 455 mg/g, respectively.
- The values of Langmuir's separation factor, R_L , and Freundlich's isotherm constant, $1/n$ at the investigated temperatures for each adsorption system were less than one, proving that the process was spontaneous, and also that with increasing temperature, it gradually approached zero for MB and gradually approached one for Pb(II), proving that it was more favorable and unfavorable, respectively.

- The thermodynamic parameters calculated for the studied adsorption systems showed that the processes were spontaneous and that MB was endothermic, while Pb(II) was exothermic.
- The CoF@NAC was reused as an effective adsorbent up to four times, respectively, for MB and Pb(II) adsorption from adsorption-desorption cycle experiments with HCl.
- CoF@NAC was rapidly and completely separated from the medium by an external magnet after adsorption.

In conclusion, the findings clearly show that CoF@NAC can be used as an adsorbent that removes pollutants from water ultra-efficiently without leaving secondary pollution in the environment.

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879

880 **Ethics Declarations:**

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Figures



Figure 1

Schematic representation of CoF@NAC synthesis.

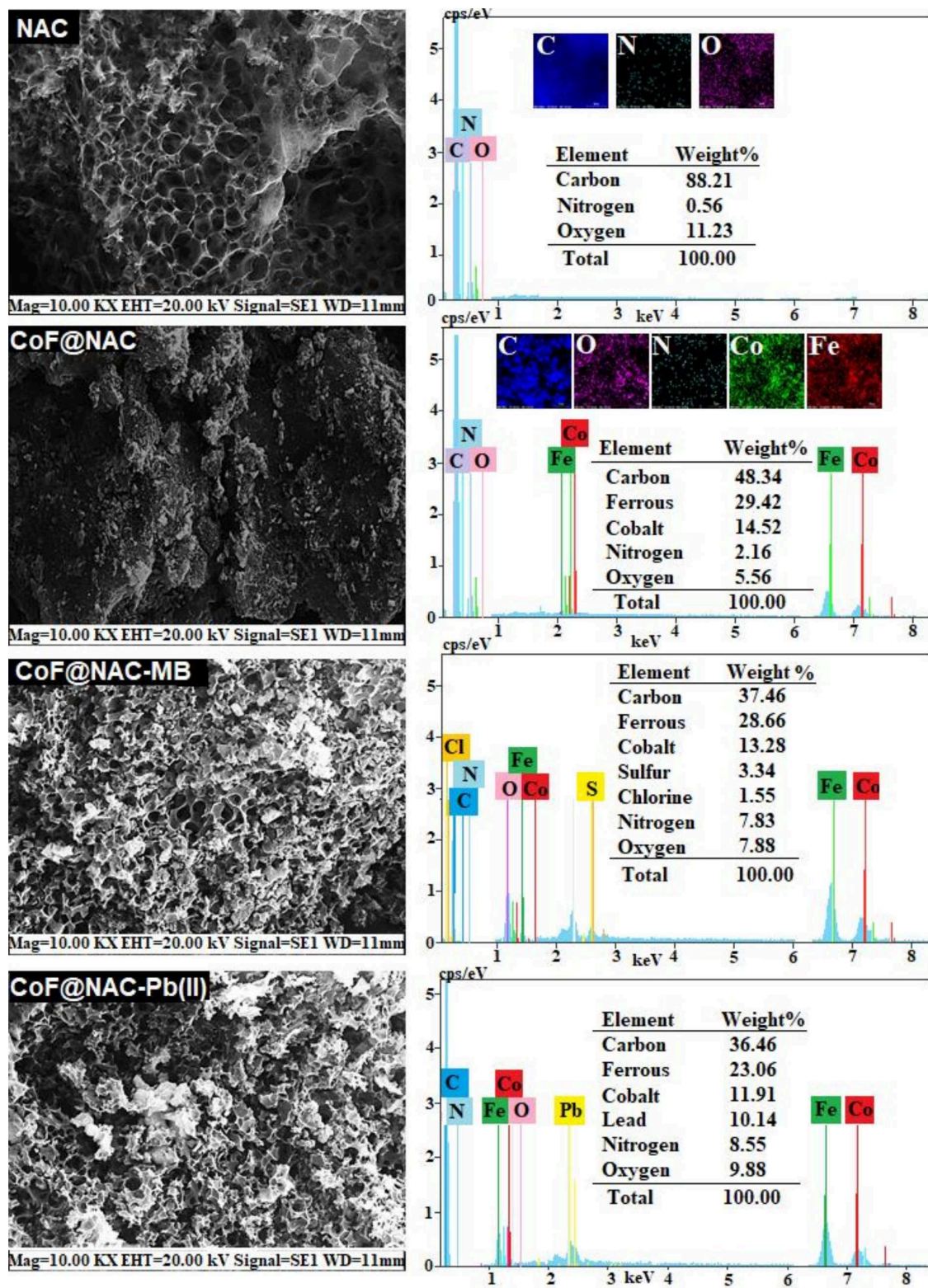


Figure 2

SEM images/EDX spectra of NAC and before and after MB and Pb(II) adsorption of CoF@NAC

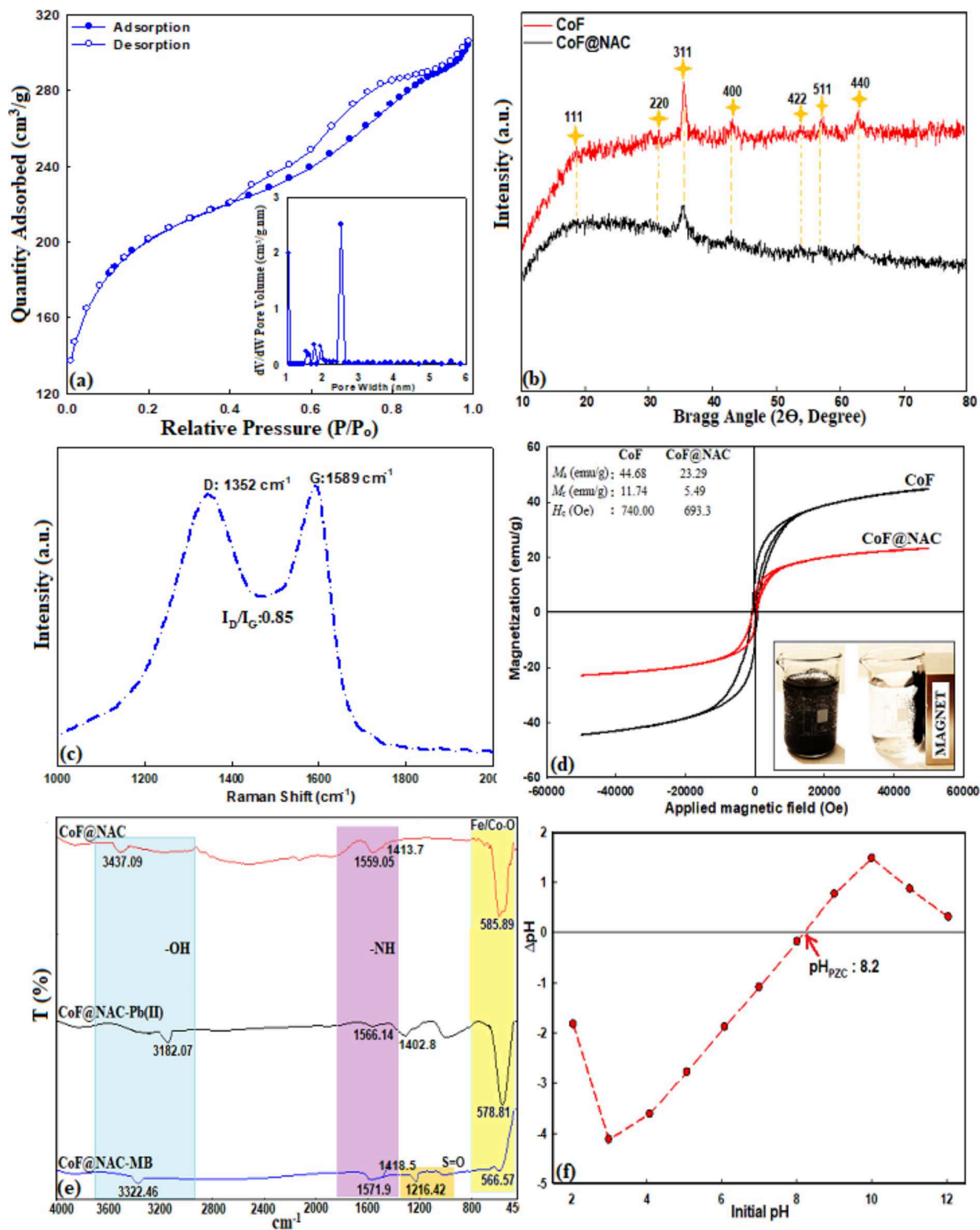


Figure 3

N₂ adsorption-desorption isotherms and pore size distribution (inset) of CoF@NAC (a), XRD pattern of CoF and CoF@NAC (b), Raman spectra of CoF@NAC (c), Magnetic hysteresis loops of CoF and CoF@NAC, and photograph (inset) of adsorption system before and after magnetic separation (d), FT-IR spectrum before and after MB and Pb(II) adsorption of CoF@NAC (e), pH_{pzc} determination of CoF@NAC (f)

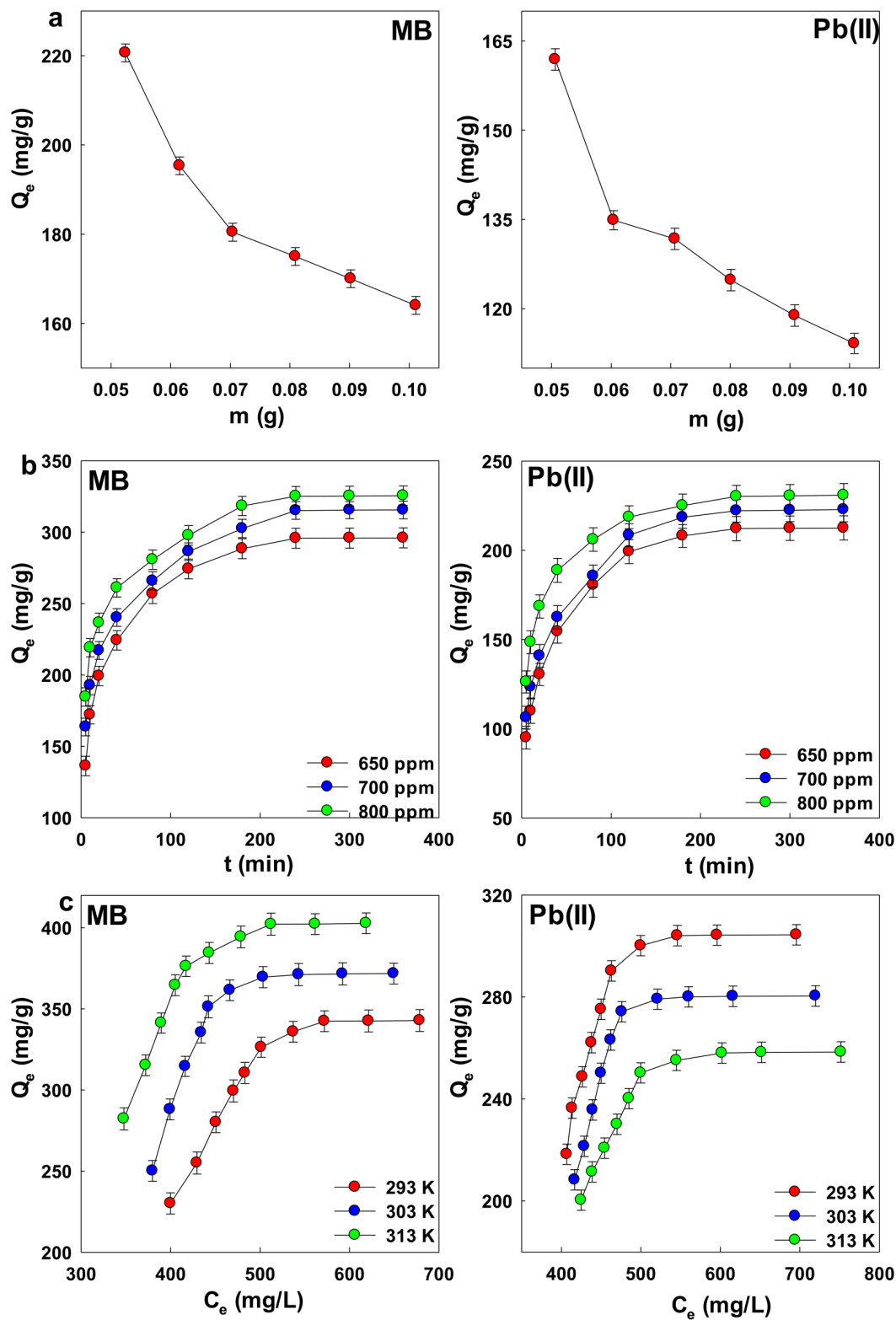


Figure 4

Effects of CoF@NAC dose (a), contact time/initial adsorbate concentration (b), and system temperature (c) on the MB and Pb(II) adsorption capacity of CoF@NAC

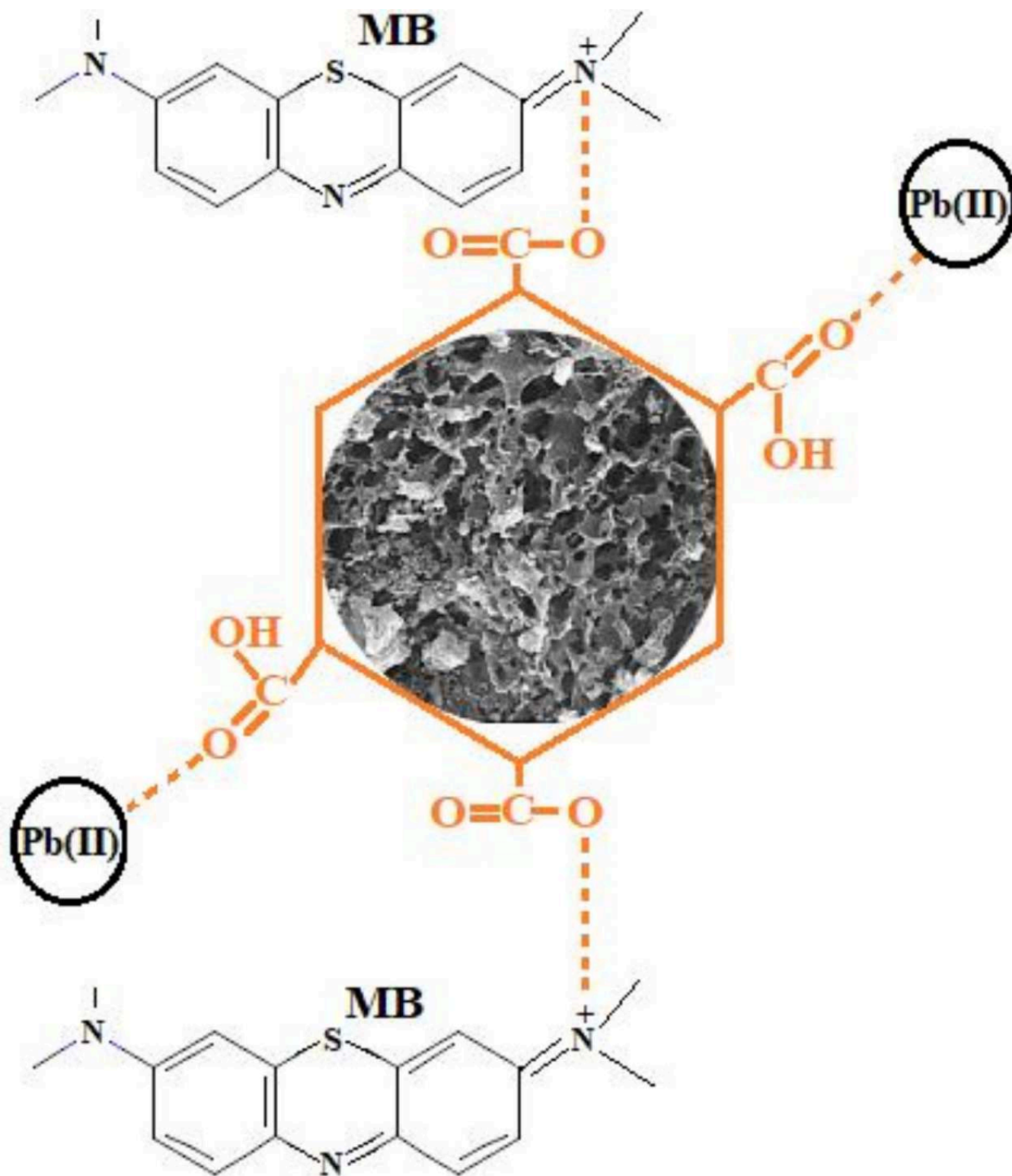


Figure 5

Possible MB and Pb(II) adsorption mechanism on the CoF@NAC

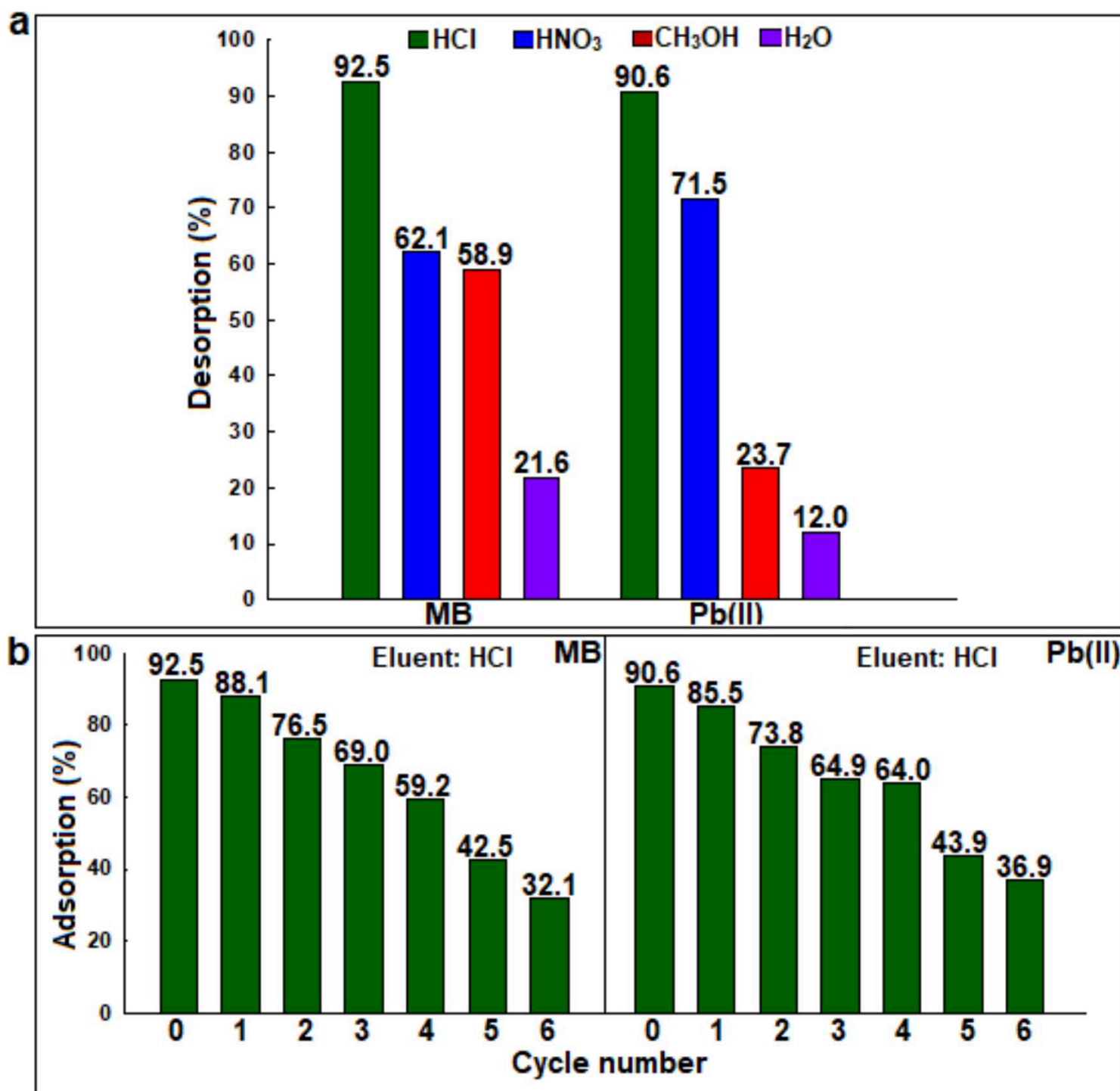


Figure 6

Effect of different desorbents on desorption efficiencies of MB and Pb(II) (a) and reusability cycles for MB and Pb(II) adsorption efficiencies (b) onto CoF@NAC

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