

Tailoring Mesoporous Y-Zeolite Molecular Sieve for Effective Removal of Micropollutants from Water

Wenxi Cen^{a, 1}, Bangguo Wang^{a, b, 1}, Yuwei Huang^a, Shengying Ye^a, Rui Xu^{c, d}, Liwei Cui^a, Yinghui Han^a, Yuanxun Zhang^{a, e}, Tao Lyu^{f, **, *}, Lijing Wang^{a, *}

^a College of Resources and Environment, University of Chinese Academy of Sciences, Beijing, 101408, China

^b Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen, 518055, China

^c Chinese Research Academy of Environmental Sciences, Beijing, 100012, China

^d National Joint Research Center for Yangtze River Conservation, Beijing, 100012, China

^e Yanshan Earth Critical Zone and Surface Fluxes Research Station, University of Chinese Academy of Sciences, Beijing, 101408, China

^f School of Water, Energy and Environment, Cranfield University, College Road, Cranfield, Bedfordshire MK43 0AL, United Kingdom

¹The authors contributed equally to this work. *Corresponding authors. wanglijing@ucas.ac.cn (L.W.); t.lyu@cranfield.ac.uk (T.L.)

Abstract

Overuse and misuse of antibiotics have led to the persistent antibiotic residues in water, posing significant challenges for remediation through conventional approaches. This study developed a tailored molecular sieve material, i.e. mesoporous Y-zeolite (M-Y zeolite), through hydrothermal synthesis to remove tetracycline (TC) as a model antibiotic from both simulated and real water matrices. The average pore size of M-Y zeolite was 3.16 nm, more than 1.7 times the second-widest dimension of the targeted TC molecule (0.81 nm), allowing effective TC adsorption. With a specific surface area of 516 m²·g⁻¹, M-Y zeolite achieved a maximum adsorption capacity of 88 mg·g⁻¹. The pseudo-second-order and Freundlich models indicated that the adsorption of TC on M-Y zeolite occurred on a multilayer heterogeneous surface

through chemisorptive interactions. The intra-particle diffusion model indicated that the adsorption process was governed by both liquid film diffusion and intraparticle diffusion stages. Mechanistic studies revealed that pore filling, complexation effects, and electrostatic interactions were the main adsorption mechanisms. After four regeneration cycles, M-Y zeolite retained 66% of its initial adsorption capacity. Tests in real water samples showed a slight decrease (4%-14%) in removal efficiency at 10 mg·L⁻¹ TC due to competing ions and organic matter, yet > 99% removal was achieved at 0.1 and 1 mg·L⁻¹ TC. These findings offer a novel, highly effective mesoporous material for the removal of antibiotics from water, marking a significant advancement in water treatment and environmental remediation technologies.

Keywords: Advanced material, Emerging contaminant, Y-zeolite, Mesoporous, Tetracycline

Synopsis

The tailored mesoporous Y-zeolite can effectively remove tetracycline from the aqueous system, providing a sustainable solution for advanced water treatment.

1. Introduction

Antibiotics are ubiquitous in aquatic ecosystem worldwide, emerging as significant pollutants due to their extensive use in controlling human and animal infections.¹ Global consumption of veterinary antibiotics is projected to reach 107k tons by 2030.² Among these antibiotics, tetracycline (TC) is the second most utilised in the aquaculture and livestock industries, favoured for its relatively low cost and effective antimicrobial properties.^{3, 4} However, TC is typically poorly absorbed by human and animals, with about 25%-75% of the ingested TC

excreted through urine and faeces as raw compounds or metabolites.⁵ The naphthol ring structure of TC makes it resistant to biological degradation by conventional wastewater treatment technologies.^{5, 6} Consequently, TC is frequently detected in effluents from sewage treatment plants.^{7, 8} Long-term exposure to trace levels of TC in aquatic ecosystem increases the risks of acute or chronic ecotoxicity to humans and promotes the transfer and spread of antibiotic-resistant genes among organisms.^{9, 10} Therefore, it is crucial to develop efficient and economical treatment technologies for the removal of TC residuals from waters.

Towards effective antibiotic removal from water, various chemical (e.g. photocatalytic degradation and Fenton oxidation) and biological (e.g. aerobic and anaerobic microbial digestion) approaches^{11, 12} have been employed to completely break down their structures. Nevertheless, the low residual levels of antibiotics in solution present a persistent challenge for these technologies in terms of sustainability and cost-effectiveness. Generally, chemical methods are energy-intensive, which require harsh reagents and generate secondary pollutants.¹³ Some biological methods are time-consuming and susceptible to operational variability.^{14, 15} In comparison, physical adsorption is a simple, high efficiency, and cost-effective approach,¹⁶ which can selectively separate precious metal ions for resource recovery and remove or pre-concentrate organic contaminants (e.g. antibiotics) to support subsequent treatment.^{17, 18} To enhance the active sites for antibiotic-adsorbent interactions, porous materials with high surface area and complex pore structures have been appraised to intensify treatment performance.¹⁹ Previous studies have investigated numerous adsorbent surface modification methods, such as elemental doping and metal ion loading, to improve antibiotic surface

adsorption. However, the importance of pore size design in relation to the dimensions of target organic pollutants has not been well studied. Random pore size distribution in adsorbents can result in ineffective antibiotic removal due to size exclusion effects (small pores) and wasted active sites (big pores).¹⁹ Thus, it is essential to design a porous adsorbent with an appropriate pore structure to achieve optimal treatment performance.

Zeolite is a porous material often used as a competitive adsorbent for removing organic compounds from water due to its stability, low cost, and settleability.^{20, 21} Specifically, Y-zeolite consists of silicon-oxygen tetrahedron and aluminium-oxygen octahedron connected through oxygen atoms, featuring a stable crystalline framework and a pore size of 0.74 nm.²² As a molecular sieve, Y-zeolite can trap chemicals in fluid environments, a function utilised in fields such as drug delivery and catalysis.^{23, 24} In environmental remediation, the trapped chemicals can be desorbed for resource recovery or further degraded through chemical and biological treatments. When used as a medium in biological reactors, the antibiotic adsorption can enhance biofilm-chemical interactions and benefit biodegradation. Notably, porous materials preferentially adsorb target chemicals when the molecular size is similar to or slightly smaller than the pore size of the adsorbent.¹⁹ Therefore, to adopt Y-zeolites for the removal of TC, with molecular dimensions of $1.41 \times 0.46 \times 0.81$ nm, introducing mesopores into the structure may be necessary to facilitate the pore filling effect.

Beyond the appropriate pore size that allows TC molecules to travel through, adsorbent surfaces loaded with metals such as Fe, Cu, and Zr have demonstrated superior TC adsorption performance.^{5, 25, 26} However, exogenously loaded metals may cause secondary pollution when

applied in water. The endogenous aluminium in Y-zeolite is expected to interact with TC through complexation, naturally enhancing adsorption. In addition, TC possesses tricarbonyl-amide, phenolic diketone, and dimethylamine groups that are pH-dependent, causing it to exist as cationic, zwitterionic, or anionic species under different conditions.^{7, 27} The ionisation behaviour could potentially enhance TC adsorption via electrostatic attraction. Furthermore, TC adsorption is also controlled by cation bridging, π - π electron coupling, Lewis acid-base interactions, and hydrophobic effects between the functional groups of TC and the corresponding charged/polar sites of adsorbents.^{25, 28-30} Therefore, designing an appropriate mesoporous structure for zeolite adsorbents is urgently needed to address the growing issue of antibiotic pollution. Meanwhile, evaluating adsorption mechanisms is crucial to improve adsorption performance and guide environmental applications.

This study aimed to synthesize a mesoporous Y-zeolite (M-Y zeolite) tailored for effective antibiotic removal and to elucidate associated adsorption mechanisms. The mesoporous structure, targeting the specific dimensions of the TC molecule, was confirmed by detailed characterisation analyses (SEM, BET, XRD, XPS, and FTIR). The adsorption performance of M-Y zeolite was systematically evaluated through analyses of adsorption kinetics, isotherms, and thermodynamics. Model fitting and physicochemical property comparisons before and after adsorption elucidated the underlying adsorption mechanisms. Furthermore, regeneration experiments and real water matrices (tap water, natural lake water, and domestic wastewater treatment effluent) validation were conducted to assess the reusability and sustainability of M-

Y zeolite. This study emphasizes the innovation of tailoring mesoporous Y-zeolite for targeted antibiotic removal, offering insights for practical applications in advanced water treatment.

2. Experimental Methods

2.1. Synthesis of mesoporous Y-zeolite

The mesoporous Y-zeolite was synthesized through a modified hydrothermal method.³¹ The procedure began by dissolving aluminum isopropoxide (10.86 g) in an aqueous solution of TEAOH (12.62 g) with stirring, followed by NaOH (0.20 g) addition. TEOS (52.08 g) was gradually introduced dropwise and stirred vigorously for 2 h to obtain solution 1 (S1). S1 was transferred into a Teflon-lined autoclave and first heated at 50 °C for 12 h, followed by 100 °C for 48 h. The final molar composition of S1 was 10SiO_2 : Al_2O_3 : 0.2NaOH : 1.2TEAOH : $183\text{H}_2\text{O}$.

For solution 2 (S2), NaAlO_2 (0.82 g) was dissolved in an aqueous solution containing NaOH (1.28 g), TBAOH solution (22.70 g) and deionized water (49.43 g) with stirring. TPOAC (3.26 g) was then added, and the mixture was stirred for 10 min. Finally, silica gel (10.62 g) was slowly added into the mixture under stirring to create a gel with the molar composition of 35SiO_2 : $1\text{Al}_2\text{O}_3$: 8.4NaOH : 7TBAOH : 0.525TPOAC : $700\text{H}_2\text{O}$.

To prepare the final gel, S1 was added dropwise to S2 under vigorous stirring for 2 h. The silica from solution 1 constituted 10 wt% of the total silica in the gel. The gel was transferred into a Teflon-lined autoclave and crystallized by rotating at 50 rpm at 120 °C for 8 d. The crystallized solid was collected by filtration, thoroughly washed with deionized water, and dried in air at

80 °C for 12 h. To remove any residual organic templates, the product was calcined in air at 550 °C for 7 h.

More details on materials, characterisation, UV-Visible spectroscopy are provided in Text S1-S3 (supporting information).

2.2. Adsorption experiments

In the adsorption kinetics experiments, 1 g·L⁻¹ M-Y zeolite were added to 100 mg·L⁻¹ TC solutions. The pH of the M-Y zeolite mixed solution was adjusted to 7 and 8 (± 0.05) to simulate practical sewage and natural water conditions, and the solution was shaken at 200 rpm at 298 K. Water samples were collected at different time intervals from 0 to 3600 min to measure the residual TC concentration using a UV-Visible spectrophotometer (Varian Cary 60, Agilent Technologies Co., Ltd. USA) at a wavelength of 355 nm. The results were fitted to the pseudo-first-order³², pseudo-second-order³³, and intra-particle diffusion models³⁴ (Text S4). To further investigate the effect of pH on TC adsorption, 0.1 M HCl and NaOH were used to adjust the initial pH of TC solutions to 3, 4, 5, 6, 7, 8, 9, and 10, respectively. The mixture were shaken at 200 rpm at 298 K for 3600 min.

In the adsorption isotherm experiments, 50 mL of TC with different initial concentrations (0, 20, 50, 80, 100, 150, 200 mg·L⁻¹) was mixed with 0.05 g of M-Y zeolite and shaken at 200 rpm at 288, 298, 308 and 318 K for 3600 min at pH 7. The Langmuir³⁵, Freundlich³⁶ and Temkin³⁷ isotherm models were adopted to fit the adsorption equilibrium data (Text S5). Thermodynamic parameters were calculated based on the method detailed in Text S6.

2.3. Validation in real water matrices

To validate the adsorption potential of M-Y zeolite, experiments were further conducted using tap water from the lab, natural lake water from Yanqi Lake in Beijing, and secondary effluent from the Bishui Wastewater Treatment Plant in Beijing. Water quality tests (**Table S1**) revealed that secondary effluent contained significantly higher levels of phosphorus and nitrogen nutrients, with concentrations 2.7 to 12.7 times greater than those detected in tap water and lake water. While COD level was undetectable in tap water, the COD in secondary effluent was 8.2 times higher than that in lake water. TC was spiked into these water samples at concentrations of 0.1, 1, and 10 mg·L⁻¹. The experimental setup adhered to the protocols previously established for the adsorption kinetics experiments. All experiments were performed in triplicate to ensure reliable results.

More details on desorption and re-adsorption experiments are provided in Text S7.

3. Results

3.1. Properties of mesoporous Y-zeolite

The BET surface area of the synthesized M-Y zeolite was 516 m²·g⁻¹, along with a large total pore volume and mesoporous volume (**Table S2**). The average pore size was 3.16 nm, indicating a mesoporous structure (**Figure S1a**). The N₂ adsorption-desorption isotherms of M-Y zeolite exhibited a type IV pattern with a noticeable H3 hysteresis loop, further confirming its mesoporosity (**Figure S1b**). Additionally, EDS analysis identified four elements (O, Si, Al, and Na) on the M-Y zeolite surface before adsorption. The Si to Al molar ratio was found to be

14.9, with oxygen representing the largest mass proportion at 60% (**Table S3**). The large BET surface area and mesoporous volume of M-Y zeolite are expected to provide ideal accessible channels for TC transfer and adsorption.

3.2. Adsorption kinetics

The TC adsorption kinetics experiments showed that M-Y zeolite exhibited a slightly higher adsorption capacity at pH 8 ($38 \text{ mg} \cdot \text{g}^{-1}$) compared to pH 7 ($35 \text{ mg} \cdot \text{g}^{-1}$) (**Figure 1a**). The intra-particle diffusion model effectively described the adsorption kinetics at both pH levels ($R^2 > 0.9$), dividing the adsorption process into two stages: liquid film diffusion and intraparticle diffusion (**Figure 1b**, **Table 1**). In addition, the intra-particle diffusion constant at the first and second stage (k_1 and k_2) was higher at pH 8 (0.60 and 0.47) than at pH 7 (0.48 and 0.46). Although equilibrium was achieved after 3600 min, the adsorption kinetics revealed a rapid initial adsorption of TC, with nearly 30% of TC adsorbed in the first minute, compared to only 8% in the final 14 hours. Both the pseudo-first-order (**Figure 1c**) and the pseudo-second-order models (**Figure 1d**) were applied to fit the adsorption data at pH 7 and 8. The R^2 value (0.95; 0.93) of the pseudo-second-order model fitted better than that of the pseudo-first-order model (0.89; 0.85) (**Table 1**).

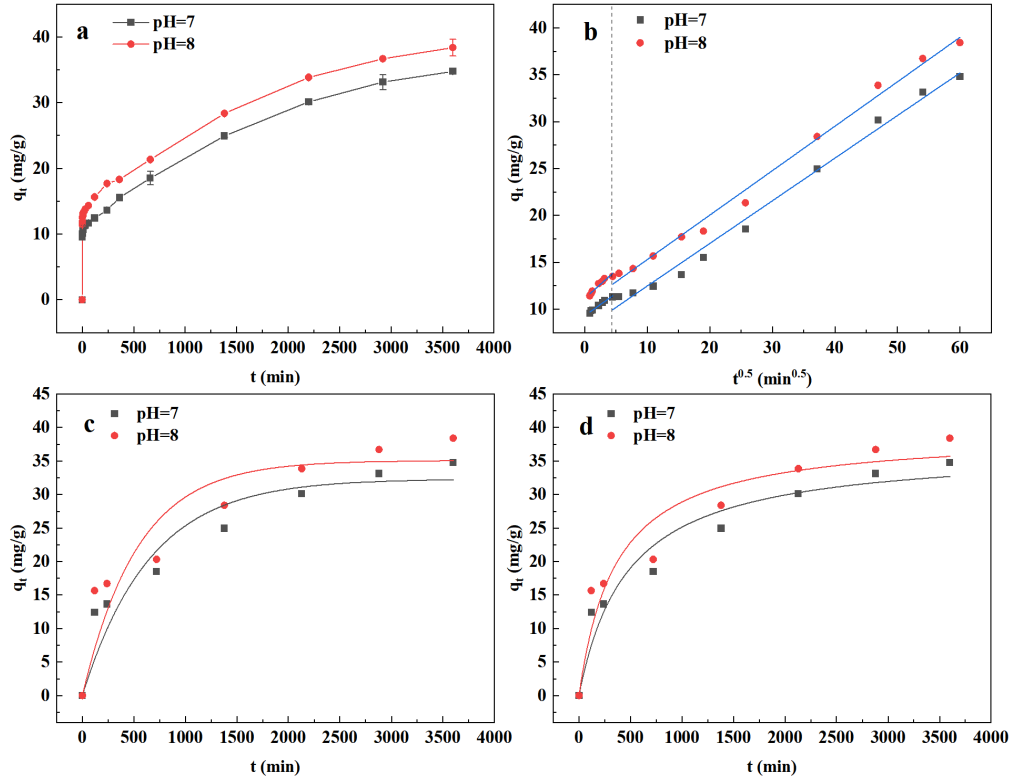


Figure 1. TC adsorption process (a), intra-particle diffusion model fitting (b), pseudo-first-order model fitting (c) and pseudo-second-order model fitting (d) for M-Y zeolite at pH 7 and 8.

Table 1

Fitting parameters of adsorption kinetics models

Model	Parameter	pH=7	pH=8
Intra-particle diffusion model	k_1 (mg·g ⁻¹ ·min ^{-1/2})	0.476	0.597
	C	9.30	11.14
	R ²	0.973	0.917
	k_2 (mg·g ⁻¹ ·min ^{-1/2})	0.455	0.474
	C	7.91	10.55
	R ²	0.989	0.992
Pseudo-first-order model	$k_1 \times 10^{-3}$ (min ⁻¹)	1.53	1.86
	q_e (mg·g ⁻¹)	32.34	35.09
	R ²	0.892	0.851
Pseudo-second-order model	$k_2 \times 10^{-5}$ (g·mg ⁻¹ ·min ⁻¹)	5.74	7.10
	q_e (mg·g ⁻¹)	36.98	39.26
	R ²	0.947	0.927

3.3. Adsorption isotherm

Isotherm analysis revealed the transfer of TC from the liquid phase to the solid phase of M-Y zeolite. The Langmuir, Freundlich and Temkin isotherm models were employed to fit the adsorption equilibrium data (**Figure 2a-c; Table 2**). The results indicated that the Freundlich model provided the best fit across 288-318 K, with R^2 ranging from 0.91 to 0.95, compared to the Langmuir model ($R^2 = 0.73-0.80$) and Temkin model ($R^2 = 0.85-0.91$). Furthermore, the magnitudes of the Freundlich parameter $1/n$ ranged from 0 to 0.5, and the K_F value increased as the temperature rose from 288 K to 318 K. The maximum adsorption capacity of M-Y zeolite reached $88 \text{ mg}\cdot\text{g}^{-1}$ at an initial TC concentration of $200 \text{ mg}\cdot\text{L}^{-1}$, at a temperature of 318 K and pH of 7.

In order to investigate the thermodynamic feasibility of the adsorption, various thermodynamic parameters were calculated, including Gibbs free energy (ΔG), enthalpy change (ΔH), and entropy change (ΔS). The ΔG values were negative, decreasing from $-3.8 \text{ kJ}\cdot\text{mol}^{-1}$ to $-6.4 \text{ kJ}\cdot\text{mol}^{-1}$ as the temperature increased from 288 K to 318 K (**Table 2**), indicating that the adsorption was spontaneous and became more feasible and favourable at higher temperatures.³⁸ This is consistent with the results of the batch adsorption experiments (**Figure 2a-c**). The positive ΔH and ΔS values suggested that the adsorption was an endothermic process, characterised by increased randomness at the solid-solution interface.³⁹

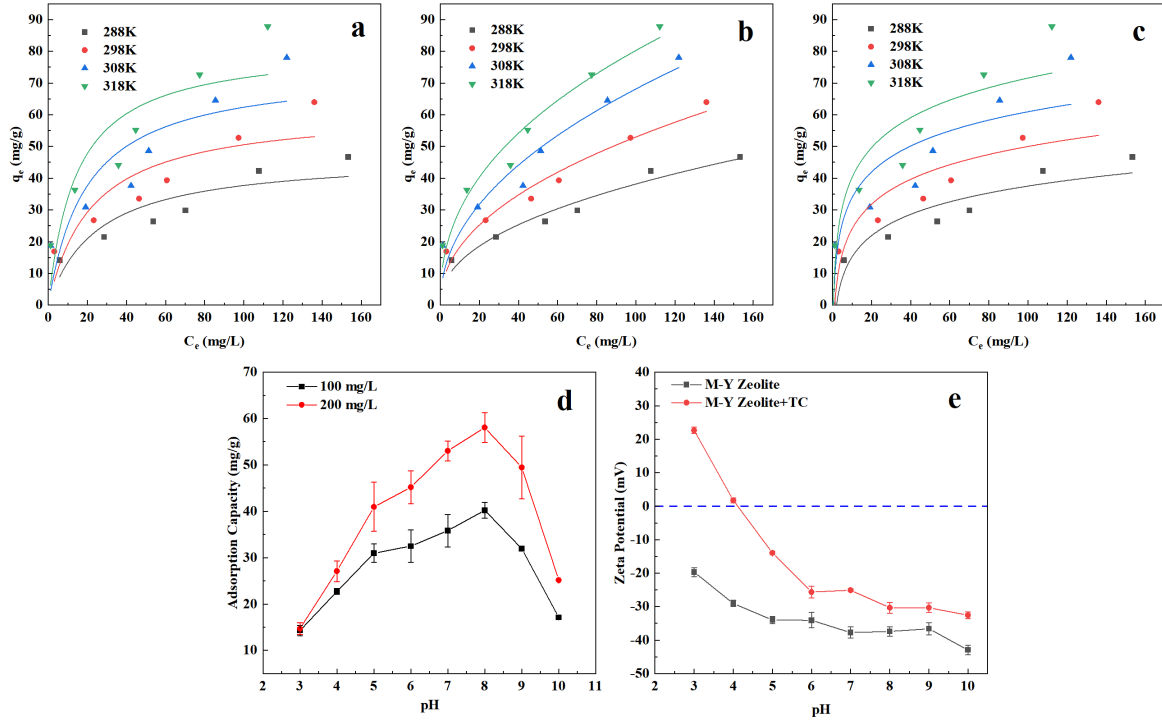


Figure 2. Adsorption isotherms of TC on M-Y zeolite fitted by Langmuir model (a), Freundlich model (b), and Temkin model (c), along with adsorption capacity of M-Y zeolite (d) and zeta potential of M-Y zeolite before and after adsorption (e) at different pH levels.

Table 2

Langmuir, Freundlich, Temkin isotherm, and thermodynamics parameters of TC adsorption on M-Y zeolite.

T (K)	Langmuir isotherm constants			Freundlich isotherm constants			Temkin isotherm constants			Thermodynamics parameters		
	q_m (mg·g ⁻¹)	K_L (L·m ³ ·g ⁻¹)	R^2	K_F (mg ^{1-1/n} ·L ^{1/n} ·g ⁻¹)	$1/n$	R^2	K (L·mg ⁻¹)	B (J·mol ⁻¹)	R^2	ΔG (kJ·mol ⁻¹)	ΔH (kJ·mol ⁻¹)	ΔS (J·mol ⁻¹ ·K ⁻¹)
288	47.12	0.040	0.801	4.86	0.447	0.944	0.47	9.72	0.908	-3.79	20.44	83.94
298	61.78	0.045	0.772	6.33	0.461	0.942	0.83	11.32	0.875	-4.57		
308	74.58	0.051	0.733	7.67	0.474	0.911	1.75	11.79	0.851	-5.22		
318	81.87	0.070	0.786	11.19	0.428	0.950	1.89	13.65	0.875	-6.38		

3.4. Effect of pH

The adsorption capacity of M-Y zeolite to TC initially increased and then declined within the pH range of 3 to 10 (**Figure 2d**). At pH 8, the maximum adsorption capacities of M-Y zeolite were 58 mg·g⁻¹ and 40 mg·g⁻¹ for initial TC concentrations of 200 mg·L⁻¹ and 100 mg·L⁻¹,

respectively (**Figure 2d**). Previous studies on natural zeolite and graphene oxide for TC adsorption reported similar trends.^{40, 41} The zeta potential of M-Y zeolite was pH dependent and exhibited higher stability under more alkaline conditions. It ranged from -20 mV to -43 mV within the pH range of 3-10, without showing an isoelectric point (pH_{pzc}). However, after adsorption, the zeta potential of M-Y zeolite significantly increased, fluctuating between 23 and -33 mV at pH 3-10, with an isoelectric point at 4 (**Figure 2e**).

3.5. Surface structure changes

Before adsorption, the M-Y zeolite exhibited an irregular, blocky crystalline shape with distinct surface depressions (**Figure 3a**). After adsorption, although the blocky stacked structure of M-Y zeolite remained largely unchanged, the surface appeared noticeably rougher (**Figure 3b**), likely due to the adsorption of TC. Additionally, nearly 2% N was newly detected after adsorption, marking the most notable change in the element content (**Table S3**).

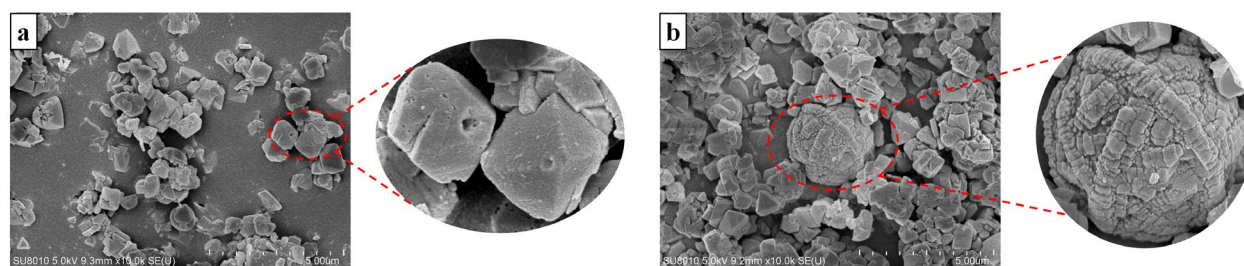


Figure 3. SEM images of M-Y zeolite before (a) and after (b) adsorption.

3.6. Spectroscopic analysis of mesoporous Y-zeolite

The XRD analysis showed that the major diffraction peaks of M-Y zeolite occurred at 6.16, 10.06, 11.81, 15.89, 18.96, 20.66, 24.00, 27.45, and 31.87°, corresponding to the characteristic peaks of Y-zeolite (**Figure 4a**). After adsorption, there were no significant changes in the positions of the diffraction peaks, indicating that the main structure of M-Y zeolite was

maintained during adsorption. However, the peak intensity at 6.16, 10.06, 11.81, and 15.89° decreased noticeably after adsorption (**Figure 4a**).

FTIR spectra were used to investigate the functional group changes of M-Y zeolite after adsorption (**Figure 4b**), and the absorption peaks in the 600-450 cm^{-1} range were attributed to Si-Al framework vibration, with no apparent shift after adsorption. The results illustrated that the M-Y zeolite structure did not change during adsorption.⁴² This was further confirmed by the XRD pattern of M-Y zeolite after adsorption (**Figure 4a**). The new characteristic absorption peaks at 1459 and 1085 cm^{-1} after adsorption were attributable to the C=C stretching vibration of the benzenoid framework and C-OH groups, corresponding to the functional groups in TC molecules.^{27, 40} The newly detected absorption peaks confirmed successful TC adsorption on M-Y zeolite. Moreover, the broad peak at 3436 cm^{-1} , attributed to the H-O-H stretching vibration of physically adsorbed water on the M-Y zeolite surface, shifted to 3440 cm^{-1} after adsorption, indicating interaction between TC and the physically adsorbed water on the M-Y zeolite surface.⁴³

In comparing the XPS N 1s spectra before and after adsorption (**Figure 4c and d**), a new peak appeared at a binding energy of 399.60 eV after adsorption, suggesting the adsorption of TC. This observation aligns with the EDS results (**Table S3**) and FTIR findings (**Figure 4b**). The peak deconvolution of the Al 2p spectra was performed and the result showed that the peak of Al 2p spectra had shifted 0.22 eV to lower binding energy from 74.66 eV (**Figure 4e**). Meanwhile, analysis of the O 1s spectra (**Figure 4f**) revealed three peaks at 531.53, 533.03, and 534.14 eV, corresponding to the Al-O bond, Si-O bond, and hydroxyl functional group in the

M-Y zeolite structure, respectively.⁴⁴ After adsorption, all three peaks also have shifted to lower binding energies.

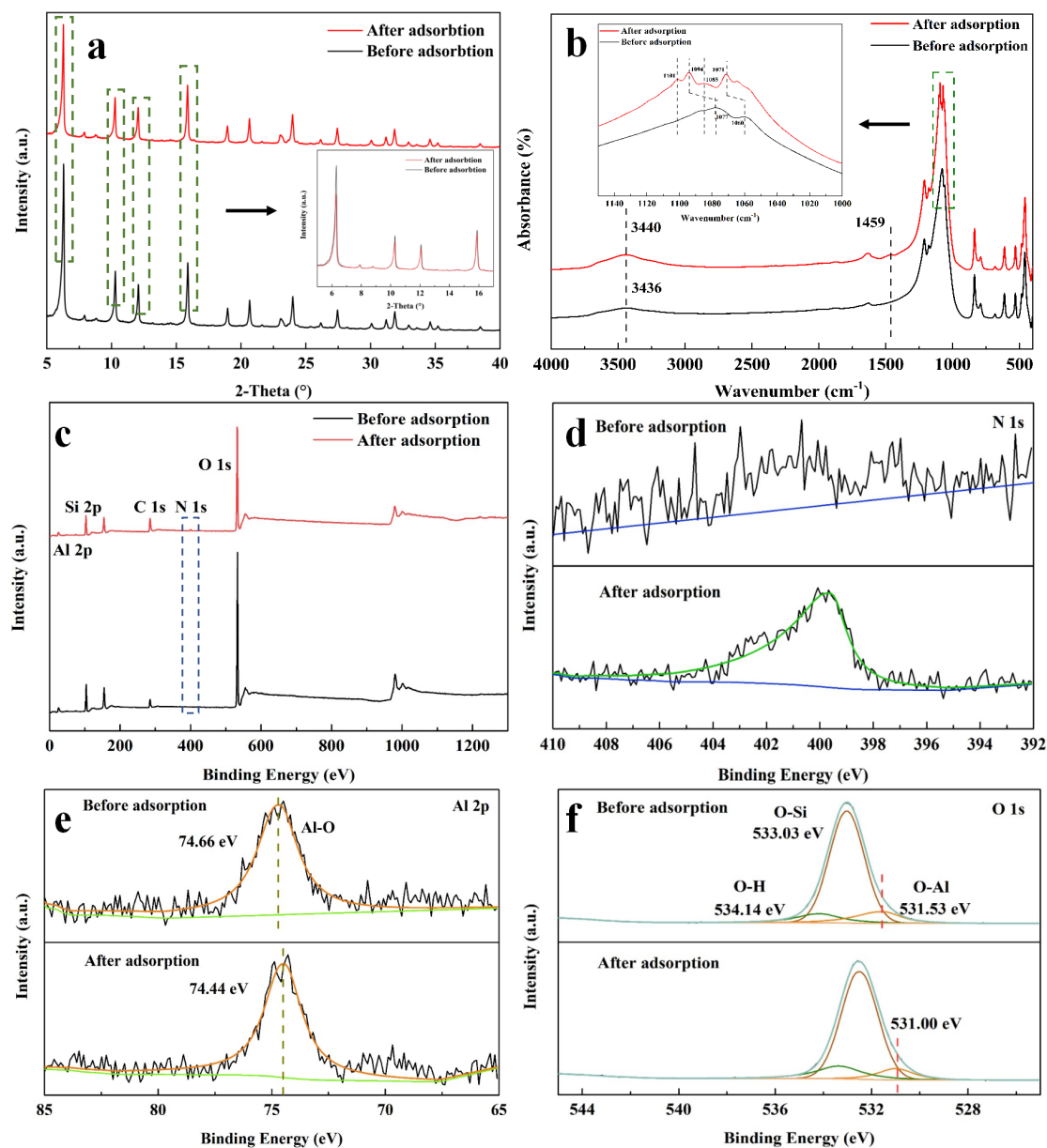


Figure 4. The XRD patterns (a), FTIR spectra (b), XPS survey spectra (c), XPS N 1s spectra (d), XPS Al 2p spectra (e), XPS O 1s spectra (f) of M-Y zeolite before and after adsorption of TC.

3.7. UV-Visible spectroscopy

In order to investigate the adsorption mechanisms, the UV-Visible spectra of TC solutions with various additions were collected in the wavelength between 200 and 600 nm (**Figure 5a**). The spectrum of TC exhibited two absorption peaks at 275 and 355 nm, corresponding to the π - π^*

transition of the amide and hydroxyl groups linked with a ring and the π - π^* transition of the other three-ring chromophores, respectively.⁴⁵ The spectrum of TC+M-Y zeolite solution after adsorption equilibrium could represent the adsorbed TC⁴⁶ and its intensity was significantly lower than that of the pure TC solution (**Figure 5a**). Meanwhile, the absorption peak at 355 nm disappeared while a new peak appeared at 375 nm in the adsorbed TC spectrum. The spectrum of TC in the presence of Si or Na⁺ was almost the same as the pure TC solution spectrum. However, a new absorption peak at 235 nm appeared in the spectrum of TC in the presence of Al³⁺, which was attributed to free Al³⁺ in the solution.⁴⁶ The peak at 375 nm was due to the shift of the primary characteristic peak of TC, which will be discussed in detail in the following section. Moreover, the spectrum of the filtrate matched well with the pure TC solution (**Figure 5a**).

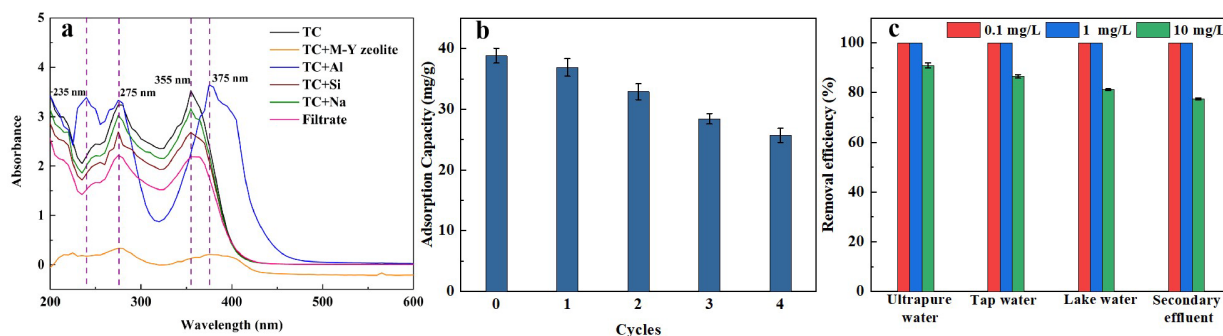


Figure 5. UV-Visible spectra of TC solution, M-Y zeolite+TC solution after adsorption equilibrium (adsorbed TC), mixture solution of TC and Al³⁺, Si, and Na⁺, and filtrate after the interaction of TC with M-Y zeolite (a), regenerative adsorption of TC on M-Y zeolite (b), adsorption capacity of M-Y zeolite in real water samples (c).

3.8. Desorption and re-adsorption of M-Y zeolite

Regeneration experiments were conducted to evaluate the practical reusability of the M-Y zeolite. Efficient regeneration using a low concentration of NaOH (0.01 M) was tested (**Figure**

5b). The initial adsorption capacity of TC on fresh M-Y zeolite was $39 \text{ mg}\cdot\text{g}^{-1}$; after four cycles, the adsorption capacity of TC on M-Y zeolite still remained above $25 \text{ mg}\cdot\text{g}^{-1}$.

3.9. Validation in real water matrices

To assess the performance of M-Y zeolite in real water samples compared with the ultrapure water, TC removal efficiency was tested in tap water, lake water, and secondary effluent (**Figure 5c**). A reduction of approximately 4%-14% in TC removal efficiency was observed in the three real water samples compared to the performance in the ultrapure water matrix (91%) at a TC concentration of $10 \text{ mg}\cdot\text{L}^{-1}$. Nevertheless, at lower TC concentrations (0.1 and $1 \text{ mg}\cdot\text{L}^{-1}$), complete removal was achieved in both ultrapure and real water matrices.

4. Discussions

4.1. Pore and surface characteristics of M-Y zeolite

The pore size of the porous adsorbent significantly affects the adsorption capacity for the aqueous organic pollutants with different sizes and geometry.^{19, 47} The hydrothermal synthesis process used in this study yielded Y-zeolite with a mesoporous structure (**Table S2**). As a result, TC could effectively transfer into the pores of M-Y zeolite without being hindered by size exclusion, as the pore size of M-Y zeolite (3.16 nm) exceeded 1.7 times the second-widest dimension of TC (0.81 nm). This result is identical to the study investigating the effects of the pore size distribution of magnetic hyper crosslinked resins on TC adsorption.¹⁹

The Freundlich parameters ($1/n$ and K_F) indicated that the adsorption was favorable and occurred on a multilayer heterogeneous surface, with stronger adsorption capacity observed at higher temperatures (**Table 2**).⁴⁸⁻⁵⁰ In addition, the pseudo-second-order model fitted better,

indicating that the adsorption rate was primarily governed by the availability of active sites on M-Y zeolite surface. The rate-limiting step was strongly influenced by chemical interactions, rather than the physical interactions characterised by the pseudo-first-order model.^{38, 51} The decrease in the adsorption rate at the later stage might be attributed to the reduction of active adsorption sites.⁴⁰ The intra-particle diffusion model also divided the adsorption into two stages: liquid film diffusion that the transfer of TC molecules from the solution to the surface of M-Y zeolite; intraparticle diffusion that the TC diffused into the internal mesoscale pores of M-Y zeolite until the adsorption equilibrium was attained (**Figure 1b**). The fitted lines did not pass through the origin ($C \neq 0$), indicating that the rate-limiting steps were controlled not only by intraparticle diffusion but also by liquid film diffusion.^{52, 53} The intraparticle diffusion stage was much longer than the film diffusion stage, as the initial occupation of channels by adsorbed TC slowed surface mass transfer.³⁸ The higher adsorption capacity observed at pH 8 (**Figure 2d**) correlated with the increased diffusion constants (k_1 and k_2), suggesting that the adsorption process was more favorable at this pH.¹⁹

The surface characteristics of M-Y zeolite also indirectly verified the indispensable role of pore filling effect in TC adsorption process. The newly detected N element in M-Y zeolite after adsorption (**Figure S2, Table S3**) confirmed the adsorption of TC, as N is a characteristic element of TC. Moreover, the noticeable decrease in the peak intensity (6.16, 10.06, 11.81, and 15.89°) of the XRD spectrum after adsorption (**Figure 4a**) indicated changes in the surface composition of M-Y zeolite due to the adsorbed TC.⁴³ The adsorption of TC would clog the mesopores of M-Y zeolite, causing changes in the adsorbent's grain surface, which may be

reflected in the decreased peak intensity of XRD spectrum.⁴³ This was also supported by SEM images showing changes in morphology of M-Y zeolite after the reaction (**Figure 3**). Therefore, based on the characterisation and model fitting, M-Y zeolite demonstrated its effectiveness as an adsorbent by providing ideal mesoscale channels that facilitated the diffusion and adsorption of TC molecules.

4.2. Complexation effect

The UV-Visible, FTIR, and XPS spectra analysis suggested that the formation of an Al-TC complex might enhance the adsorption. The UV-Visible spectra of individual TC, filtrate, and TC solutions with Si or Na⁺ exhibited identical patterns, indicating that Si or Na⁺ did not participate in the adsorption process (**Figure 5a**), and the aluminum element in the skeleton of M-Y zeolite did not dissolve in the TC solution during the adsorption process. In contrast, obvious shifts of original peaks at 355 nm to 375 nm were observed in the TC+M-Y zeolite and TC+Al³⁺ solutions (**Figure 5a**), which suggested that similar interactions might be involved in those two conditions.⁴⁶ Previous studies have shown that a complex could form between the tricarbonyl-amide group on the TC and aluminum ions.^{45, 46} The nitrogen of the tricarbonyl-amide group in the TC structure can act as an electron donor, giving electrons to Al to form complexation with M-Y zeolite. This resulted in the lower binding energy of Al after receiving electrons shown in the Al 2p spectra (**Figure 4e**). In addition, the electronegativity of O in the Al-O bond was higher than that of Al, leading to electron transfer towards O and a consequent decrease in O binding energy (**Figure 4f**). The generated Al-TC complex has strong stability due to a relatively high formation constant (log K=12.5).⁵⁴ Therefore, the peak at 375 nm in the

UV-Visible spectra (**Figure 5a**) might be related to the formation of stable complexes between the functional groups of TC molecules and Al on the skeleton of M-Y zeolite.

Furthermore, the FTIR spectrum analysis revealed absorption peaks of M-Y zeolite at 1077 and 1060 cm^{-1} , which was assigned to the asymmetric stretching vibration of O-T-O and T-O-T bonds (T= Si or Al) (**Figure 4b**).^{55, 56} After the reaction with TC, these peaks shifted to 1094 and 1071 cm^{-1} , respectively (**Figure 4b**), indicating that the Si-O or Al-O groups were involved in the TC adsorption.⁵⁷ However, the UV-Visible spectra in **Figure 5a** have excluded the reaction between TC and Si. In addition, a new peak at 1101 cm^{-1} was attributable to the strong binding of amide groups on TC with Al^{38, 58}, which further demonstrated that the chelation would occur on the amide groups during the adsorption process. Thus, with evidences from UV-Visible spectra, XPS spectra, and FTIR spectra, it could be concluded that the stable complexation between the amide groups on TC molecules and aluminium on the skeleton of M-Y zeolite was an essential mechanism for the adsorption.

4.3. Electrostatic interaction

The solution pH significantly affected the adsorption capacity of M-Y zeolite for TC (**Figure 2d**), which may be explained by the interactions between pH-dependent speciation of TC and surface charge properties of M-Y zeolite. Due to the multiple ionizable functional groups (tricarboxyl-amide, phenolic diketone, and dimethylamine) on TC compound, it exists in cation form (TCH_3^+) at $\text{pH} < 3.3$, zwitterion form ($\text{TCH}_2^\pm$) at $3.3 < \text{pH} < 7.7$, anionic form (TCH^-) at $7.7 < \text{pH} < 9.7$, and as TC^{2-} when $\text{pH} > 9.7$.^{26, 38} The increase by 6 mV to 42 mV in the zeta potential of the M-Y zeolite in the pH range of 3-10 after adsorption (**Figure 2e**) suggested that

electrical neutralization may be involved during the adsorption process. This result is similar to the previous study of TC adsorption on natural zeolite.⁴⁰ While electrostatic attraction is expected to be stronger under highly acidic conditions ($\text{pH} < 3.3$), the observed decrease in adsorption capacity at low pH can be attributed to a reduction in complexation between TC and M-Y zeolite. This reduction is likely due to the protonation of the tricarbonyl-amide group in TC, which diminished the electron-donating ability of the amide nitrogen, thus weakening its ability to complex with M-Y zeolite. At pH 3.3-7.7, TC exists predominantly as a zwitterion ($\text{TCH}_2^\pm$), optimizing its ability to complex with M-Y zeolite.⁵⁹ In this range, adsorption was primarily driven by pore filling and complexation, as electrostatic interactions were not significant.²⁶ Beyond pH 7.7, TC transitions to anionic forms (TCH^- and TC^{2-}), causing strong electrostatic repulsion with the negatively charged M-Y zeolite. This repulsion hindered TC's access to the zeolite pores⁶⁰, leading to a sharp decline in adsorption capacity and making complexation the dominant mechanism when $\text{pH} > 8$.

4.4. Effects of real water matrices and engineering implications

The removal efficiency of M-Y zeolite in real water samples showed variations at $10 \text{ mg} \cdot \text{L}^{-1}$ TC. In tap water, the slight decrease (4%) in removal efficiency (**Figure 5c**) likely resulted from the electrostatic attraction between cations (Ca^{2+} and Mg^{2+}) and the negatively charged M-Y zeolite. These cations may occupy active sites on the M-Y zeolite surface, leading to competitive adsorption against TC and weakening electrostatic interactions between M-Y zeolite and TC.⁶¹ The decreases were more pronounced ($< 14\%$) in lake water and secondary effluent (**Figure 5c**), involving not only competitive ions but also organic matter, as indicated

by the higher COD levels compared to tap water (**Table S1**). These contaminants may occupy active sites on the surface and within the mesopores of M-Y zeolite, hindering pore accessibility for TC molecules. Consequently, competitive adsorption intensified, further reducing removal efficiency to 14%.⁶² Despite the complex and variable conditions in real water samples, M-Y zeolite provided sufficient adsorption sites to achieve complete TC removal at low concentrations (0.1 and 1 mg·L⁻¹), confirming its potential for effectively removing residual antibiotics in real water bodies. Furthermore, after four regeneration cycles with NaOH, M-Y zeolite retained 66% of its initial TC adsorption capacity (**Figure 5b**), demonstrating strong reusability and sustainable adsorption performance.

Adsorption is a promising treatment for removing antibiotics from water. Adsorbents that are highly efficient and eco-friendly are highly sought after for antibiotic removal. Therefore, in this study, mesoporous Y-zeolite was synthesized through hydrothermal process to meet this need. With its mesoporous structure, M-Y zeolite achieved better antibiotic adsorption capacity than other adsorbents (**Table S4**), including biochar⁶³, goethite⁶⁴, and zerovalent iron⁶⁵. Notably, except for the exogenous input from wastewater, around 20% of residual antibiotics in the aquatic environment originate from sediment release.⁶⁶ The settlement characteristics of M-Y zeolite allow it not only remove antibiotics from the aqueous phase but also potentially inhibit their release from sediments through physical isolation and chemisorption at the water-sediment interface. Moreover, the application of M-Y zeolite in water and wastewater treatment as a molecular sieve enables the concentration of adsorbed pollutants from contaminated water. The organic chemicals can subsequently be recovered through desorption or further treated through

biological/chemical processes at higher concentrations. Nevertheless, the complexity of pollutant constituents in real water bodies, including hydrocarbons and heavy metal ions, introduces additional challenges. While M-Y zeolite shows promise in removing these pollutants through complexation, pore filling, and electrostatic interaction, further research is needed to fully elucidate the mechanisms and optimize the material's performance. At last, to enhance the adsorption performance of M-Y zeolite and extend its application (e.g. blocking and inhibiting effects), other convenient and cost-effective synthesis methods could be developed in future study.

5. Conclusions

This study developed a Y-zeolite with a mesoscale pore size of 3.16 nm and a large surface area of $516 \text{ m}^2 \cdot \text{g}^{-1}$ through hydrothermal preparation, enabling effective TC adsorption. The kinetics and isotherm studies indicated that the TC adsorption involved multilayer chemisorptive interactions, with the process being controlled by liquid film diffusion and intraparticle diffusion. Regeneration experiments over four cycles confirmed the reusability of M-Y zeolite. While tests in real water samples at a concentration of $10 \text{ mg} \cdot \text{L}^{-1}$ showed a slight reduction in removal efficiency due to competing ions and organic matter, M-Y zeolite achieved > 99% TC removal at low concentrations (0.1 and $1 \text{ mg} \cdot \text{L}^{-1}$). The analysis of the adsorption process and alternations in the physicochemical properties of M-Y zeolite suggested that pore filling effect and electrostatic interaction were responsible for the adsorption. Moreover, based on the UV-Visible, XPS, and FTIR spectra, complexation between the amide groups on TC and aluminium on M-Y zeolite was another essential mechanism for the adsorption. In summary, this study

offers a solid theoretical foundation for the practical application of mesoporous molecular sieve material for antibiotic adsorption and supports subsequent resource recovery and remediation efforts.

Associated Contents

The supporting information is available free of charge via the internet at '<http://pubs.acs.org.xn-ivg/>'.

Materials, characterisation, equations, water quality parameters, BET analysis, EDS data and the comparison of adsorption capacity of different adsorbents

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Author contributions

Wenxi Cen: Investigation, Methodology, Software, Validation, Writing – original draft. **Bangguo Wang:** Investigation, Methodology, Formal analysis, Writing – original draft. **Yuwei Huang:** Data verification, Writing – review & editing. **Shengying Ye:** Investigation, Writing – review & editing. **Rui Xu:** Conceptualization, Data verification, Writing – review & editing. **Liwei Cui:** Experimental method guidance, Project administration. **Yinghui Han:** Experimental method guidance, Supervision. **Yuanxun Zhang:** Experimental method guidance, Data verification. **Tao Lyu:** Formal analysis, Writing – review & editing. **Lijing Wang:** Conceptualization, Investigation, Supervision, Writing – review & editing, Funding acquisition.

Acknowledgments

This work was supported by Fundamental Research Funds for the Central Universities (No. E3E40504X2), National Key R&D Program of China (No. 2022YFE0209500), Air Pollution Status Assessment and Refined Management Supporting Project of Qinghai Province (No. E341970201, E34050701).

Data availability

Data available within the article and its supporting information.

References

1. Tian, Y.; Zhu, J.; Ying, C.; Luo, H.; Zhang, S.; Zhang, L.; Li, R.; Li, J., Photoaging processes of polyvinyl chloride microplastics enhance the adsorption of tetracycline and facilitate the formation of antibiotic resistance. *Chemosphere* **2023**, 320, 137820.
2. Mulchandani, R.; Wang, Y.; Gilbert, M.; Van Boeckel, T. P., Global trends in antimicrobial use in food-producing animals: 2020 to 2030. *PLOS Global Public Health* **2023**, 3 (2), e0001305.
3. Sheng, D.; Ying, X.; Li, R.; Cheng, S.; Zhang, C.; Dong, W.; Pan, X., Polydopamine-mediated modification of ZIF-8 onto magnetic nanoparticles for enhanced tetracycline adsorption from wastewater. *Chemosphere* **2022**, 308, 136249.
4. Wei, M.; Marrakchi, F.; Yuan, C.; Cheng, X.; Jiang, D.; Zafar, F. F.; Fu, Y.; Wang, S., Adsorption modeling, thermodynamics, and DFT simulation of tetracycline onto mesoporous and high-surface-area NaOH-activated macroalgae carbon. *Journal of Hazardous Materials* **2022**, 425, 127887.
5. Zhang, Z.; Li, H.; Liu, H., Insight into the adsorption of tetracycline onto amino and amino-Fe³⁺ functionalized mesoporous silica: Effect of functionalized groups. *Journal of Environmental Sciences* **2018**, 65, 171-178.
6. Watkinson, A. J.; Murby, E. J.; Kolpin, D. W.; Costanzo, S. D., The occurrence of antibiotics in an urban watershed: From wastewater to drinking water. *Science of the Total Environment* **2009**, 407 (8), 2711-2723.
7. Rivera-Utrilla, J.; Gómez-Pacheco, C. V.; Sánchez-Polo, M.; López-Peñalver, J. J.; Ocampo-Pérez, R., Tetracycline removal from water by adsorption/bioadsorption on activated carbons and sludge-derived adsorbents. *Journal of Environmental Management* **2013**, 131, 16-24.
8. Sun, J.; Zeng, Q.; Tsang, D. C.; Zhu, L.; Li, X., Antibiotics in the agricultural soils from the Yangtze River Delta, China. *Chemosphere* **2017**, 189, 301-308.
9. Hu, Y.; Jiang, L.; Zhang, T.; Jin, L.; Han, Q.; Zhang, D.; Lin, K.; Cui, C., Occurrence and removal of sulfonamide antibiotics and antibiotic resistance genes in conventional and advanced drinking water treatment processes. *Journal of hazardous materials* **2018**, 360 (OCT.15), 364.

10. Povolo, V. R.; Ackermann, M., Disseminating antibiotic resistance during treatment. *Science* **2019**, *364* (6442), 737-738.
11. Liu, J.; Meng, C.; Zhang, X.; Wang, S.; Duan, K.; Li, X.; Hu, Y.; Cheng, H., Direct Z-scheme In₂O₃/AgI heterojunction with oxygen vacancies for efficient molecular oxygen activation and enhanced photocatalytic degradation of tetracycline. *Chemical Engineering Journal* **2023**, *466*, 143319.
12. Hu, P.; Shao, J.; Qian, G.; Adeleye, A. S.; Hao, T., Removal of tetracycline by aerobic granular sludge from marine aquaculture wastewater: A molecular dynamics investigation. *Bioresource Technology* **2022**, *355*, 127286.
13. de Souza Santos, L. V.; Meireles, A. M.; Lange, L. C., Degradation of antibiotics norfloxacin by Fenton, UV and UV/H₂O₂. *Journal of environmental management* **2015**, *154*, 8-12.
14. Terzic, S.; Udikovic-Kolic, N.; Jurina, T.; Krizman-Matasic, I.; Senta, I.; Mihaljevic, I.; Loncar, J.; Smital, T.; Ahel, M., Biotransformation of macrolide antibiotics using enriched activated sludge culture: kinetics, transformation routes and ecotoxicological evaluation. *Journal of hazardous materials* **2018**, *349*, 143-152.
15. Sravan, J. S.; Matsakas, L.; Sarkar, O., Advances in biological wastewater treatment processes: focus on low-carbon energy and resource recovery in biorefinery context. *Bioengineering* **2024**, *11* (3), 281.
16. Chaudhuri, H.; Dash, S.; Sarkar, A., SBA-15 functionalised with high loading of amino or carboxylate groups as selective adsorbent for enhanced removal of toxic dyes from aqueous solution. *New Journal of Chemistry* **2016**, *40* (4), 3622-3634.
17. Chaudhuri, H.; Lim, C.-R.; Yun, Y.-S., Single-step synthesis of prominently selective and easily regenerable POSS functionalized with high loadings of sulfur and carboxylic acids. *Journal of Materials Chemistry A* **2023**, *11* (43), 23463-23478.
18. Chaudhuri, H.; Lim, C.-R.; Yun, Y.-S., Amino-and sulfur-containing POSS for highly efficient and selective extraction of platinum group elements from acidic solution. *Journal of Cleaner Production* **2024**, *434*, 139912.
19. Zhang, M.; Li, A.; Zhou, Q.; Shuang, C.; Zhou, W.; Wang, M., Effect of pore size distribution on tetracycline adsorption using magnetic hypercrosslinked resins. *Microporous and mesoporous materials* **2014**, *184*, 105-111.
20. de Sousa, D. N. R.; Insa, S.; Mozeto, A. A.; Petrovic, M.; Chaves, T. F.; Fadini, P. S., Equilibrium and kinetic studies of the adsorption of antibiotics from aqueous solutions onto powdered zeolites. *Chemosphere* **2018**, *205*, 137-146.

21. Kiwaan, H.; Mohamed, F. S.; El-Ghamaz, N.; Beshry, N.; El-Bindary, A., Experimental and electrical studies of Na-X zeolite for the adsorption of different dyes. *Journal of Molecular Liquids* **2021**, 332, 115877.
22. Ali, M. M.; Ahmed, M.; Hameed, B., NaY zeolite from wheat (*Triticum aestivum* L.) straw ash used for the adsorption of tetracycline. *Journal of Cleaner Production* **2018**, 172, 602-608.
23. Losch, P.; Huang, W.; Vozniuk, O.; Goodman, E. D.; Schmidt, W.; Cargnello, M., Modular Pd/zeolite composites demonstrating the key role of support hydrophobic/hydrophilic character in methane catalytic combustion. *ACS Catalysis* **2019**, 9 (6), 4742-4753.
24. Servatan, M.; Zarrintaj, P.; Mahmodi, G.; Kim, S.-J.; Ganjali, M. R.; Saeb, M. R.; Mozafari, M., Zeolites in drug delivery: Progress, challenges and opportunities. *Drug Discovery Today* **2020**, 25 (4), 642-656.
25. Yu, Y.; Zhang, Y.; Yang, W.; Yang, Y.; Deng, Q.; He, X.; Peng, C.; Xu, Y.; Hu, T.; Jiang, L., Cu-loaded reduced graphene oxide with ultrahigh adsorption performance for tetracycline from aqueous solution. *Journal of the Taiwan Institute of Chemical Engineers* **2023**, 147, 104899.
26. Zhong, J.; Yuan, X.; Xiong, J.; Wu, X.; Lou, W., Solvent-dependent strategy to construct mesoporous Zr-based metal-organic frameworks for high-efficient adsorption of tetracycline. *Environmental Research* **2023**, 226, 115633.
27. Guo, Y.; Huang, W.; Chen, B.; Zhao, Y.; Liu, D.; Sun, Y.; Gong, B., Removal of tetracycline from aqueous solution by MCM-41-zeolite A loaded nano zero valent iron: synthesis, characteristic, adsorption performance and mechanism. *Journal of hazardous materials* **2017**, 339, 22-32.
28. Chen, C.; Wang, C. K.; Li, X. B.; Wang, J.; Zhang, X. J.; Liao, X. B., Application of conventional and O₃-BAC processes to treat organic matter and antibiotic pollutants in a lake in East China. *Water Supply* **2013**, 13 (6), 1470-1477.
29. Ji, L.; Chen, W.; Duan, L.; Zhu, D., Mechanisms for strong adsorption of tetracycline to carbon nanotubes: a comparative study using activated carbon and graphite as adsorbents. *Environmental science & technology* **2009**, 43 (7), 2322-2327.
30. Zhang, Y.; Li, Y.; Xu, W.; Cui, M.; Wang, M.; Chen, B.; Sun, Y.; Chen, K.; Li, L.; Du, Q., Filtration and adsorption of tetracycline in aqueous solution by copper alginate-carbon nanotubes membrane which has the muscle-skeleton structure. *Chemical Engineering Research and Design* **2022**, 183, 424-438.
31. Cui, W.; Zhu, D.; Tan, J.; Chen, N.; Fan, D.; Wang, J.; Han, J.; Wang, L.; Tian, P.; Liu, Z.,

Synthesis of mesoporous high-silica zeolite Y and their catalytic cracking performance. *Chinese Journal of Catalysis* **2022**, 43 (7), 1945-1954.

32. Lagergren, S., About the theory of so-called adsorption of soluble substances. **1898**.

33. Ho, Y.-S.; McKay, G., Pseudo-second order model for sorption processes. *Process biochemistry* **1999**, 34 (5), 451-465.

34. Weber Jr, W. J.; Morris, J. C., Kinetics of adsorption on carbon from solution. *Journal of the sanitary engineering division* **1963**, 89 (2), 31-59.

35. Langmuir, I., The constitution and fundamental properties of solids and liquids. Part I. Solids. *Journal of the American chemical society* **1916**, 38 (11), 2221-2295.

36. Freundlich, H., Über die adsorption in lösungen. *Zeitschrift für physikalische Chemie* **1907**, 57 (1), 385-470.

37. Temkin, M. J.; Pyzhev, V., Recent modifications to Langmuir isotherms. **1940**.

38. Liao, Q.; Rong, H.; Zhao, M.; Luo, H.; Chu, Z.; Wang, R., Strong adsorption properties and mechanism of action with regard to tetracycline adsorption of double-network polyvinyl alcohol-copper alginate gel beads. *Journal of Hazardous Materials* **2022**, 422, 126863.

39. Ebelegi, A. N.; Ayawei, N.; Wankasi, D., Interpretation of adsorption thermodynamics and kinetics. *Open Journal of Physical Chemistry* **2020**, 10 (3), 166-182.

40. Guo, X.; Wang, P.; Li, P.; Zhang, C., Effect of Cu(II) on adsorption of tetracycline by natural zeolite: performance and mechanism. *Water science and technology : a journal of the International Association on Water Pollution Research* **2019**, 80 (1), 164-172.

41. Tabrizian, P.; Ma, W.; Bakr, A.; Rahaman, M. S., pH-sensitive and magnetically separable Fe/Cu bimetallic nanoparticles supported by graphene oxide (GO) for high-efficiency removal of tetracyclines. *Journal of colloid and interface science* **2019**, 534, 549-562.

42. Chizallet, C.; Lazare, S.; Bazer-Bachi, D.; Bonnier, F.; Lecocq, V.; Soyer, E.; Quoineaud, A. A.; Bats, N., Catalysis of transesterification by a nonfunctionalized metal-organic framework: acido-basicity at the external surface of ZIF-8 probed by FTIR and ab initio calculations. *Journal of the American Chemical Society* **2010**, 132 (35), 12365-77.

43. Li, N.; Zhou, L.; Jin, X.; Owens, G.; Chen, Z., Simultaneous removal of tetracycline and oxytetracycline antibiotics from wastewater using a ZIF-8 metal organic-framework. *Journal of hazardous materials* **2019**, 366, 563-572.

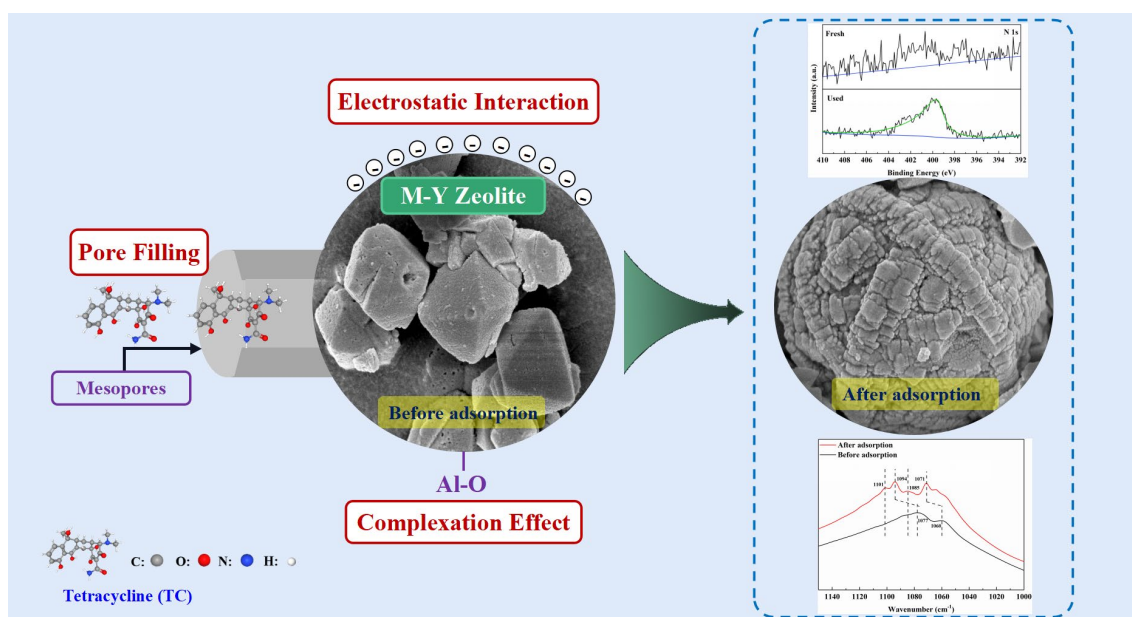
44. Medykowska, M.; Wiśniewska, M.; Szewczuk-Karpisz, K.; Panek, R., Interaction mechanism of heavy metal ions with the nanostructured zeolites surface–adsorption, electrokinetic and XPS studies. *Journal of Molecular Liquids* **2022**, 357, 119144.
45. Linares, C. F.; Brikgi, M., Interaction between antimicrobial drugs and antacid based on cancrinite-type zeolite. *Microporous and mesoporous materials* **2006**, 96 (1-3), 141-148.
46. Kang, J.; Liu, H.; Zheng, Y. M.; Qu, J.; Chen, J. P., Application of nuclear magnetic resonance spectroscopy, Fourier transform infrared spectroscopy, UV-Visible spectroscopy and kinetic modeling for elucidation of adsorption chemistry in uptake of tetracycline by zeolite beta. *J Colloid Interface Sci* **2011**, 354 (1), 261-7.
47. Güzel, F.; Saygılı, H.; Saygılı, G. A.; Koyuncu, F., New low-cost nanoporous carbonaceous adsorbent developed from carob (*Ceratonia siliqua*) processing industry waste for the adsorption of anionic textile dye: Characterization, equilibrium and kinetic modeling. *Journal of Molecular Liquids* **2015**, 206, 244-255.
48. da Rocha, M. S.; Iha, K.; Faleiros, A. C.; Corat, E. J.; Suárez-Iha, M. E. V., The influence of the temperature on the parameters of extended Freundlich's isotherm. *Journal of colloid and interface science* **1998**, 200 (1), 126-130.
49. Kalam, S.; Abu-Khamsin, S. A.; Kamal, M. S.; Patil, S., Surfactant adsorption isotherms: A review. *ACS omega* **2021**, 6 (48), 32342-32348.
50. Elsherif, K. M.; Alkherraz, A. M.; Edwards, H.; Mutawia, B. Y. A., Congo red dye adsorption on dry green pea husk: Effects of process parameters and modeling approaches. *Environmental Health Engineering And Management Journal* **2024**, 11 (3), 273-284.
51. Raganati, F.; Alfe, M.; Gargiulo, V.; Chirone, R.; Ammendola, P., Kinetic study and breakthrough analysis of the hybrid physical/chemical CO₂ adsorption/desorption behavior of a magnetite-based sorbent. *Chemical Engineering Journal* **2019**, 372, 526-535.
52. Yu, F.; Li, Y.; Huang, G.; Yang, C.; Chen, C.; Zhou, T.; Zhao, Y.; Ma, J., Adsorption behavior of the antibiotic levofloxacin on microplastics in the presence of different heavy metals in an aqueous solution. *Chemosphere* **2020**, 260, 127650.
53. Hasani, N.; Selimi, T.; Mele, A.; Thaçi, V.; Halili, J.; Berisha, A.; Sadiku, M., Theoretical, equilibrium, kinetics and thermodynamic investigations of methylene blue adsorption onto lignite coal. *Molecules* **2022**, 27 (6), 1856.

54. Ghandour, M. A.; Azab, H.; Hassan, A.; Ali, A., Potentiometric studies on the complexes of tetracycline (TC) and oxytetracycline (OTC) with some metal ions. *Monatshefte für Chemie/Chemical Monthly* **1992**, 123 (1), 51-58.
55. Duan, T. W.; Yan, B., Photophysical properties of metal ion functionalized NaY zeolite. *Photochemistry and Photobiology* **2014**, 90 (3), 503-510.
56. Liu, M.-C.; Hsieh, C.-C.; Lee, J.-F.; Chang, J.-R., Impact of Pt and V₂O₅ on ethanol removal from moist air using pellet silica-bound NaY. *Industrial & Engineering Chemistry Research* **2015**, 54 (35), 8678-8689.
57. Ahmed, M.; Islam, M. A.; Asif, M.; Hameed, B., Human hair-derived high surface area porous carbon material for the adsorption isotherm and kinetics of tetracycline antibiotics. *Bioresource Technology* **2017**, 243, 778-784.
58. Yang LiuQing, Y. L.; Li YanFeng, L. Y.; Jin XinLiang, J. X.; Ye ZhengFang, Y. Z.; Ma XiaoJie, M. X.; Wang LiYuan, W. L.; Liu YaPing, L. Y., Synthesis and characterization of a series of chelating resins containing amino/imino-carboxyl groups and their adsorption behavior for lead in aqueous phase. **2011**.
59. Wu, H.; Xie, H.; He, G.; Guan, Y.; Zhang, Y., Effects of the pH and anions on the adsorption of tetracycline on iron-montmorillonite. *Applied Clay Science* **2016**, 119, 161-169.
60. Galvan, Y.; Bauernfeind, J.; Wolf, P.; Zarraga, R.; Haumann, M.; Vogel, N., Materials with Hierarchical Porosity Enhance the Stability of Infused Ionic Liquid Films. *ACS omega* **2021**, 6 (32), 20956-20965.
61. Xiang, W.; Wan, Y.; Zhang, X.; Tan, Z.; Xia, T.; Zheng, Y.; Gao, B., Adsorption of tetracycline hydrochloride onto ball-milled biochar: Governing factors and mechanisms. *Chemosphere* **2020**, 255, 127057.
62. Cao, M.; Liu, X.; Wang, W.; Gao, M.; Yang, H., Bifunctional two-dimensional copper-aluminum modified filter paper composite for efficient tetracycline removal: Synergy of adsorption and reusability by degradation. *Chemosphere* **2022**, 287, 132031.
63. Zhang, X.; Li, Y.; Wu, M.; Pang, Y.; Hao, Z.; Hu, M.; Qiu, R.; Chen, Z., Enhanced adsorption of tetracycline by an iron and manganese oxides loaded biochar: Kinetics, mechanism and column adsorption. *Bioresource Technology* **2021**, 320, 124264.
64. Zhu, Y.; Yang, Q.; Lu, T.; Qi, W.; Zhang, H.; Wang, M.; Qi, Z.; Chen, W., Effect of phosphate on the adsorption of antibiotics onto iron oxide minerals: Comparison between tetracycline and ciprofloxacin. *Ecotoxicology and Environmental Safety* **2020**, 205, 111345.
65. Hanay, Ö.; Yıldız, B.; Aslan, S.; Hasar, H., Removal of tetracycline and oxytetracycline by microscale

zerovalent iron and formation of transformation products. *Environmental Science and Pollution Research* **2014**, *21*, 3774-3782.

66. 张芊芊, 中. 中国流域典型新型有机污染物排放量估算、多介质归趋模拟及生态风险评估. 2015.

Graphic for Table of Contents (TOC)/Abstract Art



Supporting Information

Tailoring Mesoporous Y-Zeolite Molecular Sieve for Effective Removal of Micropollutants from Water

Wenxi Cen^{a, 1}, Bangguo Wang^{a, b, 1}, Yuwei Huang^a, Shengying Ye^a, Rui Xu^{c, d}, Liwei Cui^a, Yinghui Han^a, Yuanxun Zhang^{a, e}, Tao Lyu^{f, **}, Lijing Wang^{a, *}

^a College of Resources and Environment, University of Chinese Academy of Sciences, Beijing, 101408, China

^b Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen, 518055, China

^c Chinese Research Academy of Environmental Sciences, Beijing, 100012, China

^d National Joint Research Center for Yangtze River Conservation, Beijing, 100012, China

^e Yanshan Earth Critical Zone and Surface Fluxes Research Station, University of Chinese Academy of Sciences, Beijing, 101408, China

^f School of Water, Energy and Environment, Cranfield University, College Road, Cranfield, Bedfordshire MK43 0AL, United Kingdom

*Corresponding authors. College of Resources and Environment, University of Chinese Academy of Sciences, Beijing, 101408, China.

**Corresponding authors. School of Water, Energy and Environment, Cranfield University, College Road, Cranfield, Bedfordshire MK43 0AL, United Kingdom.

E-mail address: wanglijing@ucas.ac.cn (L.W.); t.lyu@cranfield.ac.uk (T.L.)

¹ The authors contributed equally to this work.

Text S1. Materials

The chemical reagents used for the experiments included: aluminum isopropoxide (94 wt%, Maclin Biochemical Technology Co.), tetraethylammonium hydroxide (TEAOH, 35 wt% in H₂O, Sigma-Aldrich Co.), sodium hydroxide (99.9%, Maclin Biochemical Technology Co.), tetraethyl orthosilicate (TEOS, 99.99%, Maclin Biochemical Technology Co.), sodium aluminate (AR, Maclin Biochemical Technology Co.), tetrabutylammonium hydroxide (TBAOH, 40 wt% in H₂O, Sigma-Aldrich Co.), [3 - (trimethoxysilyl)propyl] octadecyldimethylammonium chloride (TPOAC, 40 wt% in methanol, Maclin Biochemical Technology Co.), silica gel (99 wt%, 200-300 mesh, Maclin Biochemical Technology Co.), tetracycline (TC, Shanghai Aladdin Industrial Co.), and hydrochloric acid (HCl, Sinopharm Chemical Reagent Co.).

Text S2. Characterisation

The specific surface areas, average pore sizes, and pore volumes of the M-Y zeolite were determined using a pore size analyser (ASAP2460, Micromeritics Co., Ltd, USA). The Brunauer-Emmett-Teller (BET) model was applied to determine the surface area at a relative pressure ranging from 0 to 1. The total volume (V_{total}) and mesopore volume (V_{meso}) were calculated using the t-plot model. The surface microstructure morphology and elemental composition were measured by a scanning electron microscope (SEM) (SU8010, Hitachi Co., Ltd, Japan) and Energy Dispersive Spectrometer (EDS), respectively. Zeta potential measurements were performed to determine the surface charge at pH 3-10 by a Zetasizer Nano ZS (90Plus PALS, Malvern Instrument Co., Ltd. UK). An X-ray powder diffractometer (XRD) (SmartLab, Rigaku Co., Ltd, Japan) was applied to detect the crystallographic structures with Cu-K α radiation ($\lambda=0.15406$ nm) at 45 kV and 200 mA in the 2θ range of 5° to 40° . The functional group composition before and after adsorption was identified by a Fourier Transform Infrared (FTIR) spectrometer (Nicolet iS50, Thermo Fisher Scientific Co., Ltd, USA) in the wavenumber range of 400 to 4000 cm^{-1} with a resolution of 4 cm^{-1} . Moreover, the surface chemical compositions and binding energies of each element before and after adsorption were analysed by X-ray photoelectron spectroscopy (XPS) (K-Alpha, Thermo Fisher Scientific Co., Ltd, USA), with a C 1s reference peak at 284.6 eV used for peak position calibration.

Text S3. UV-Visible spectroscopy

In addition to the previously mentioned materials characterisations before and after the adsorption tests, UV-Visible spectra of TC were also conducted at the wavelengths ranging from 200 to 600 nm to investigate the adsorption mechanisms of TC on M-Y zeolite. The tests were conducted under different conditions: (1) 100 mg·L⁻¹ TC solution; (2) 1 g·L⁻¹ M-Y zeolite mixed with TC solution until adsorption equilibrium was reached. To avoid interference from M-Y zeolite particles, the absorbance of a 1 g·L⁻¹ M-Y zeolite solution was used as the background correction; (3) 100 mg·L⁻¹ TC solutions with AlCl₃ (molar ratio of TC: Al³⁺ = 1:10); (4) 100 mg·L⁻¹ TC solutions with SiO₂ (molar ratio of TC: Si = 1:10); (5) 100 mg·L⁻¹ TC solutions with NaCl (molar ratio of TC: Na⁺ = 1:10); (6) the filtrate of 100 mg·L⁻¹ TC after adsorption by M-Y zeolite.

Text S4. Adsorption kinetics models

In this study, the pseudo-first-order¹ and pseudo-second-order² kinetic models were used to fit the data in the adsorption kinetics experiment. The correlation parameter (R^2) obtained from linear fitting was used to judge the fitting degree of kinetics and evaluate the quality of the kinetic models. The pseudo-first-order and pseudo-second-order kinetic models can be represented by the following equations (1) and (2), respectively.

$$q_t = q_e - q_e e^{-k_1 t} \quad (1)$$

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

where q_t ($\text{mg}\cdot\text{g}^{-1}$) and q_e ($\text{mg}\cdot\text{g}^{-1}$) represent the adsorption capacities of tetracycline (TC) at time t (min) and equilibrium time (min), respectively. k_1 (min^{-1}) and k_2 (min^{-1}) are the rate constants of the pseudo-first-order and pseudo-second-order, respectively.

In order to further investigate the diffusion process of TC during adsorption, it is necessary to identify the rate-controlling step in the adsorption process. Therefore, the Weber-Morris intra-particle diffusion model³ (equations (3)) was applied to fit the data. This model proposes that the adsorption process is controlled by the diffusion of adsorbate molecules into the pores of the adsorbent. According to the model, the amount of adsorbate uptake varies linearly with the square root of time.^{4, 5} The mathematical expression for the model is as follows:

$$q_t = k_i t^{1/2} + C \quad (3)$$

where k_i is the intra-particle diffusion constant ($\text{mg}\cdot\text{g}^{-1}\cdot\text{min}^{-1/2}$), and C is the intercept, representing the thickness of the boundary layer ($\text{mg}\cdot\text{g}^{-1}$).

Text S5. Adsorption isotherm models

In this study, the Langmuir⁶ (equations (4)), Freundlich⁷ (equations (5)) and Temkin⁸ (equations (6)) isotherm models were used to describe the adsorption process.

The Langmuir isotherm is based on the ideal monolayer adsorption theory, which assumes that adsorption occurs on a homogeneous surface with a fixed number of adsorption sites.⁹ The model can be mathematically expressed as:

$$q_e = q_m \frac{K_L C_e}{1 + K_L C_e} \quad (4)$$

where q_m ($\text{mg}\cdot\text{g}^{-1}$) is the saturated monolayer adsorption capacity; q_e ($\text{mg}\cdot\text{g}^{-1}$) is the adsorption capacity of M-Y zeolite for TC at equilibrium; C_e ($\text{mg}\cdot\text{L}^{-1}$) is the the TC concentration in solution at equilibrium; K_L is the Langmuir constant.

The Freundlich model mainly describes surface heterogeneity and the exponential distribution of active sites and their energies.¹⁰ The model can be mathematically expressed as:

$$q_e = K_F C_e^{1/n} \quad (5)$$

where q_e ($\text{mg}\cdot\text{g}^{-1}$) is the adsorption capacity of M-Y zeolite for TC at equilibrium; C_e ($\text{mg}\cdot\text{L}^{-1}$) is the the TC concentration in solution at equilibrium; K_F is the Freundlich adsorption capacity constant; $1/n$ (dimensionless) represents adsorption intensity. The K_F value provides insight into the adsorption capacity, where a higher K_F indicates stronger adsorption capacity.¹¹ The parameter $1/n$ reflects the energetic heterogeneity of the system.¹² When $0 < 1/n < 1$, the adsorption process is considered favorable.¹³

The Temkin isotherm model assumes that the heat of adsorption decreases linearly with increasing surface coverage.¹⁴ The model can be mathematically expressed as:

$$q_e = B \ln(K C_e) \quad (6)$$

where q_e ($\text{mg}\cdot\text{g}^{-1}$) is the adsorption capacity of M-Y zeolite for TC at equilibrium; C_e ($\text{mg}\cdot\text{L}^{-1}$) is the the TC concentration in solution at equilibrium; B ($\text{J}\cdot\text{mol}^{-1}$) is the Temkin constant related to heat of adsorption; and K ($\text{L}\cdot\text{mg}^{-1}$) is the Temkin equilibrium binding constant.

Text S6. Thermodynamic parameters calculation

The adsorption process involves energy changes caused by intermolecular interactions, and its characteristics can be explored through thermodynamic analysis. The energy changes during adsorption reflect the alternations of adsorption equilibrium at different temperatures, which can be expressed by thermodynamic parameters such as Gibbs free energy change (ΔG), enthalpy change (ΔH), and entropy change (ΔS). The ΔG indicates the spontaneity and feasibility of the adsorption process, which can imply the adsorption reaching equilibrium with or without external energy input.¹⁵ The ΔH is defined as the heat exchange during adsorption at constant pressure. Positive ΔH typically indicates endothermic processes, while negative ΔH suggests exothermic processes.^{15, 16} The ΔS reflects the change in randomness in the system during adsorption. A positive ΔS suggests increased randomness at the solid/liquid interface.¹⁷ These thermodynamic parameters can be calculated using the following formulas:

$$\Delta G = -RT \ln K_c \quad (7)$$

$$\ln K_c = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad (8)$$

where T (K) is the temperature; R ($8.314 \text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$) is the ideal gas constant; and K_c is the adsorption distribution coefficient.

Text S7. Desorption and re-adsorption experiments

The used adsorbent was recovered by ultrasonic treatment in 100 mL of 0.01 M NaOH solution for 30 min, followed by oscillation at 200 rpm at 298 K for 2 h. After desorption, the adsorbent was separated by centrifuging at 8000 rpm for 20 min and washed with ultrapure water. The adsorbent was then dried at 333 K for 12 h. For re-adsorption, the recovered adsorbent was introduced into 200 mL of a 100 mg·L⁻¹ TC solution at pH 7. This adsorption-desorption cycle was repeated four times to evaluate the reusability of M-Y zeolite.

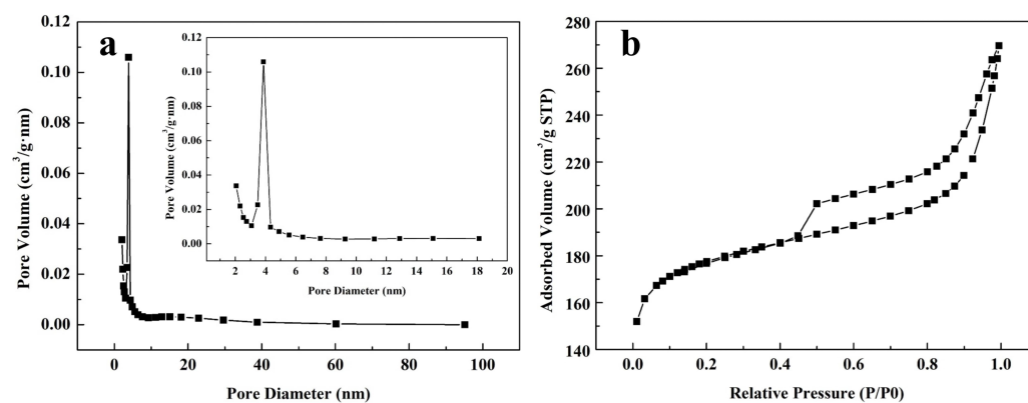


Figure. S1. The pore size distribution (a) and N_2 adsorption–desorption isotherm (b) of M-Y zeolite

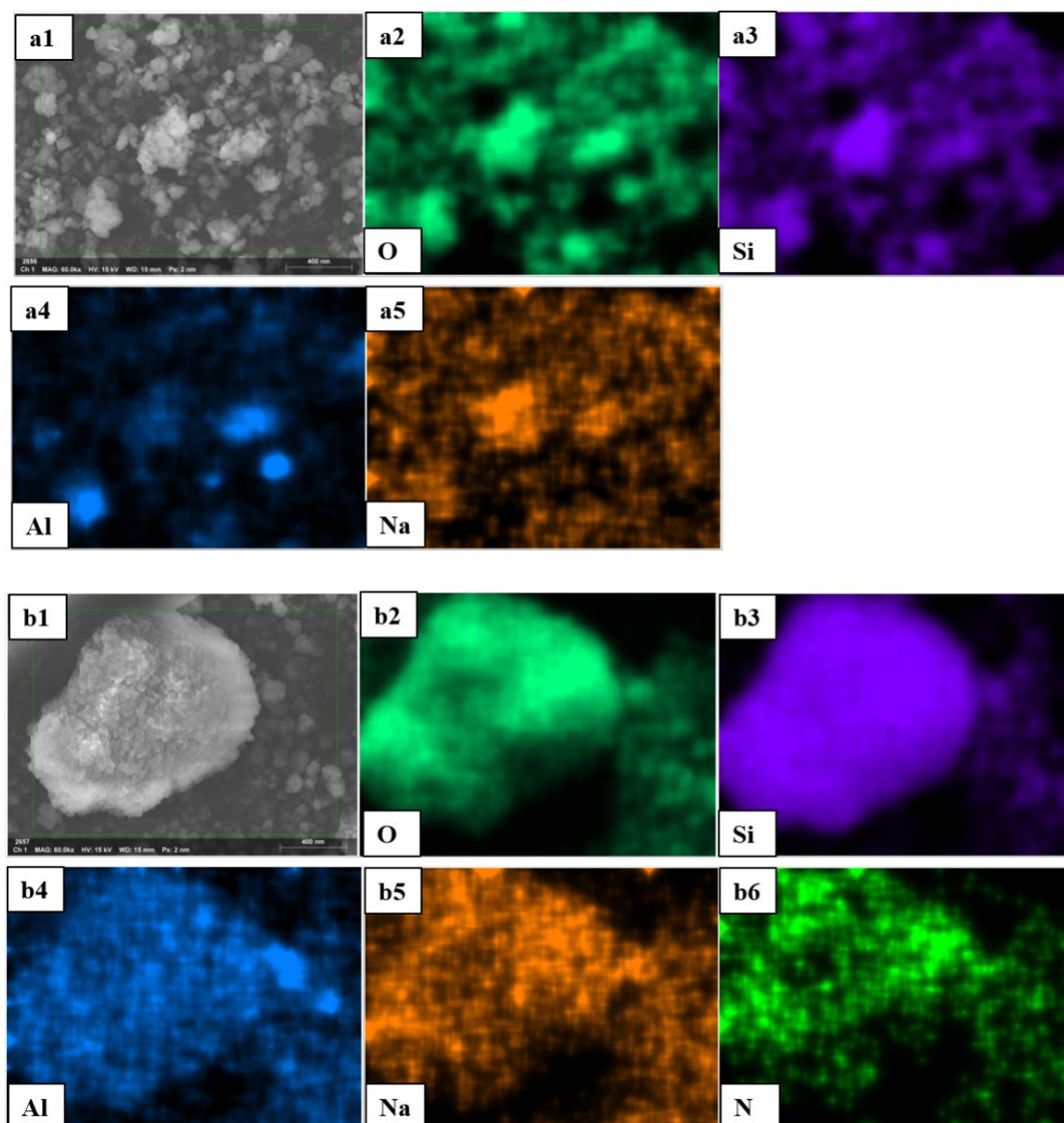


Figure. S2. EDS elemental mapping of M-Y zeolite before adsorption (a1-a5) and after adsorption (b1-b6).

Table S1

Selected water quality parameters for real water samples

Water quality parameters	Tap water	Lake water	Secondary effluent
Total phosphorus ($\text{mg}\cdot\text{L}^{-1}$)	0.03	0.14	0.38
Labile phosphorus ($\text{mg}\cdot\text{L}^{-1}$)	<DL	0.03	0.04
Total nitrogen ($\text{mg}\cdot\text{L}^{-1}$)	0.15	0.38	3.66
Ammonia nitrogen ($\text{mg}\cdot\text{L}^{-1}$)	0.04	0.21	0.46
DO ($\text{mg}\cdot\text{L}^{-1}$)	6.51	5.88	5.11
COD ($\text{mg}\cdot\text{L}^{-1}$)	<DL	1.8	14.7
Average particle diameter (μm)	<DL	15.71	48.98
Turbidity (NTU)	0.05	0.67	13.4
pH	7.25	8.01	7.84

DL: detection limit.

Table S2

Specific surface area and pore size distribution of M-Y zeolite

Material	S_{BET} (m²·g⁻¹)	V_{meso} (cm³·g⁻¹)	V_{total} (cm³·g⁻¹)	D_{average} (nm)
M-Y zeolite	515.97	0.20	0.41	3.16

Table S3

EDS data of M-Y zeolite before and after adsorption

Element	Weight (%)	
	Before adsorption	After adsorption
O	59.62	59.16
Si	37.70	35.32
Al	2.43	3.42
Na	0.25	0.23
N	-	1.87

-: not detected

Table S4

The comparison of adsorption capacity of different adsorbents for TC.

Adsorbent	C ₀ (mg·L ⁻¹)	Dosage (mg·L ⁻¹)	T (°C)	Time (h)	Q _e (mg·g ⁻¹)	Reference
Fe/Mn oxides loaded biochar	80	1	25	2	13	Zhang et al. ¹⁸ .
Magnetic molecularly imprinted nanoparticles	300	1	25	12	13	Dai et al. ¹⁹
Goethite	30	1	25	48	18	Zhu et al. ²⁰
HCl-modified zeolite	89	2	30	20	20	Zou et al. ²¹
Microscale zerovalent iron	60	0.4	60	8	24	Hanay et al. ²²
Mesoporous Y-zeolite	200	1	45	60	88	This study

References

1. Lagergren, S., About the theory of so-called adsorption of soluble substances. **1898**.
2. Ho, Y.-S.; McKay, G., Pseudo-second order model for sorption processes. *Process biochemistry* **1999**, *34* (5), 451-465.
3. Weber Jr, W. J.; Morris, J. C., Kinetics of adsorption on carbon from solution. *Journal of the sanitary engineering division* **1963**, *89* (2), 31-59.
4. Raganati, F.; Alfe, M.; Gargiulo, V.; Chirone, R.; Ammendola, P., Kinetic study and breakthrough analysis of the hybrid physical/chemical CO₂ adsorption/desorption behavior of a magnetite-based sorbent. *Chemical Engineering Journal* **2019**, *372*, 526-535.
5. Liao, Q.; Rong, H.; Zhao, M.; Luo, H.; Chu, Z.; Wang, R., Strong adsorption properties and mechanism of action with regard to tetracycline adsorption of double-network polyvinyl alcohol-copper alginate gel beads. *Journal of Hazardous Materials* **2022**, *422*, 126863.
6. Langmuir, I., The constitution and fundamental properties of solids and liquids. Part I. Solids. *Journal of the American chemical society* **1916**, *38* (11), 2221-2295.
7. Freundlich, H., Über die adsorption in lösungen. *Zeitschrift für physikalische Chemie* **1907**, *57* (1), 385-470.
8. Temkin, M. J.; Pyzhev, V., Recent modifications to Langmuir isotherms. **1940**.
9. Du, Y.; Liu, M.; Liu, Y.; Li, X.; Huang, Z.; Ding, D.; Yang, S.; Feng, J.; Chen, Y.; Chen, R., Modulating the pore and electronic structure for targeted recovery of platinum: Accelerated kinetic and reinforced coordination. *Journal of Hazardous Materials* **2024**, *469*, 133913.
10. Ayawei, N.; Ebelegi, A. N.; Wankasi, D., Modelling and interpretation of adsorption isotherms. *Journal of chemistry* **2017**, *2017* (1), 3039817.
11. Elsherif, K. M.; Alkheraz, A. M.; Edwards, H.; Mutawia, B. Y. A., Congo red dye adsorption on dry green pea husk: Effects of process parameters and modeling approaches. *Environmental Health Engineering And Management Journal* **2024**, *11* (3), 273-284.
12. da Rocha, M. S.; Iha, K.; Faleiros, A. C.; Corat, E. J.; Suárez-Iha, M. E. V., The

influence of the temperature on the parameters of extended Freundlich's isotherm. *Journal of colloid and interface science* **1998**, *200* (1), 126-130.

13. Kalam, S.; Abu-Khamsin, S. A.; Kamal, M. S.; Patil, S., Surfactant adsorption isotherms: A review. *ACS omega* **2021**, *6* (48), 32342-32348.

14. Guo, Y.; Yan, C.; Wang, P.; Rao, L.; Wang, C., Doping of carbon into boron nitride to get the increased adsorption ability for tetracycline from water by changing the pH of solution. *Chemical Engineering Journal* **2020**, *387*, 124136.

15. Ebelegi, A. N.; Ayawei, N.; Wankasi, D., Interpretation of adsorption thermodynamics and kinetics. *Open Journal of Physical Chemistry* **2020**, *10* (3), 166-182.

16. Ball, D. W.; Key, J. A., *Introductory Chemistry*. BCcampus: 2014.

17. Batool, F.; Akbar, J.; Iqbal, S.; Noreen, S.; Bukhari, S. N. A., Study of isothermal, kinetic, and thermodynamic parameters for adsorption of cadmium: an overview of linear and nonlinear approach and error analysis. *Bioinorganic chemistry and applications* **2018**, *2018* (1), 3463724.

18. Zhang, X.; Li, Y.; Wu, M.; Pang, Y.; Hao, Z.; Hu, M.; Qiu, R.; Chen, Z., Enhanced adsorption of tetracycline by an iron and manganese oxides loaded biochar: Kinetics, mechanism and column adsorption. *Bioresource Technology* **2021**, *320*, 124264.

19. Dai, J.; Pan, J.; Xu, L.; Li, X.; Zhou, Z.; Zhang, R.; Yan, Y., Preparation of molecularly imprinted nanoparticles with superparamagnetic susceptibility through atom transfer radical emulsion polymerization for the selective recognition of tetracycline from aqueous medium. *Journal of hazardous materials* **2012**, *205*, 179-188.

20. Zhu, Y.; Yang, Q.; Lu, T.; Qi, W.; Zhang, H.; Wang, M.; Qi, Z.; Chen, W., Effect of phosphate on the adsorption of antibiotics onto iron oxide minerals: Comparison between tetracycline and ciprofloxacin. *Ecotoxicology and Environmental Safety* **2020**, *205*, 111345.

21. Zou, Y. L.; Huang, H.; Chu, M.; Lin, J. W.; Yin, D. Q.; Li, Y. N., Adsorption research of tetracycline from water by HCl-modified zeolite. *Advanced Materials Research* **2012**, *573*, 43-47.

22. Hanay, Ö.; Yıldız, B.; Aslan, S.; Hasar, H., Removal of tetracycline and oxytetracycline by microscale zerovalent iron and formation of transformation products. *Environmental Science and Pollution Research* **2014**, *21*, 3774-3782.



DSPACE

<https://dspace.org/>

Tailoring mesoporous Y-Zeolite molecular sieve for effective removal of micropollutants from water

Cen, Wenxi; Wang, Bangguo; Huang, Yuwei; Ye, Shengying; Ma, Jian ...

2025-03-14

American Chemical Society

<https://doi.org/10.1021/acsestwater.4c01119>

Cen W, Wang B, Huang Y, et al., (2025) Tailoring mesoporous Y-Zeolite molecular sieve for effective removal of micropollutants from water. ACS ES&T Water, Volume 5, Issue 3, March 2025, pp. 1373-1383