Supplementary Information

Optimization of the Preparation Conditions of Aluminum-Impregnated Food Waste Biochar Using RSM with an MLP and Its Application in Phosphate Removal

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2. Material and methods

2.2. Response Surface Methodology (RSM) application for optimization of Al-FWB preparation condition for phosphate removal

$$X_i = \frac{x_i - \min(x_i)}{\max(x_i) - \min(x_i)} \quad \text{(S1)}$$

where X_i is the normalized input variable (−). x_i is the ith variable, min(x_i) is the minimum value of variable x_i, and max(x_i) is the maximum value of variable x_i.

2.5. Data analysis

For all experiments, the phosphate adsorption capacity was calculated using the following equation:

$$q_e = \frac{(C_i - C_e) \times V}{m} \quad \text{(S2)}$$

where C_i is the initial phosphate concentration, C_e is the residual phosphate concentration after adsorption, V is the volume of the solution (L), and m is the amount of the adsorbent (g).

The adsorption mechanisms were explored using kinetics-based pseudo-first- and pseudo-second-order models. The following equations describe pseudo-first- and second-order kinetic models [81,82]:

$$q_t = q_e(1 - e^{-k_1 t}) \quad \text{(S3)}$$

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \quad \text{(S4)}$$

where k_1 (1/h) and k_2 (g/mg/h) are the pseudo-first-order and pseudo-second-order rate constants, respectively.

The Langmuir and Freundlich models were used to analyze the adsorption equilibrium data, and the isotherm equations were as follows [83,84]:

$$q_e = \frac{Q_m K_L C_e}{1 + K_L C_e} \quad \text{(S5)}$$

$$q_e = K_F C_e^{\frac{1}{n}} \quad \text{(S6)}$$

where Q_m (mg/g) is the maximum phosphate adsorption capacity of the adsorbent, K_L is the Langmuir constant related to adsorption energy, K_F (mg g^{-1}(mg L^{-1})$^{-1/n}$) is the Freundlich constant, and n (−) is the adsorption intensity.

The thermodynamic parameters were calculated using the following equation [85,86]:

$$\Delta G^0 = -RT \ln (K_e) \quad \text{(S7)}$$

$$\ln(K_e) = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \quad \text{(S8)}$$

$$K_e = \frac{aq_e}{C_e} \quad \text{(S9)}$$

where ΔG^0 is the standard Gibbs free energy, ΔS^0 is the entropy change, ΔH^0 is the enthalpy change, a is the adsorbent dosage (g/L), q_e is the maximum amount adsorbed (mg/g), T is the absolute temperature (K), and R is the universal gas constant (8.314 J/mol·K)). ΔH^0 and ΔS^0 were calculated using the slope and intercept of the va n't Hoff plot of $1/T$ vs. ln(aq_e/C_e) [87].

Table S1. Ranges of hyperparameters of multi-layer perceptron for grid-search and selected values of hyperparameters (using sklearn library, by 5 folds cross validation).

Hyperparameters	Range	Selected value
hidden_layer_sizes	10, 20, 30	**30**
activation	relu, logistic, tanh	**tanh**
learning_rate	constant, adaptive	**constant**
solver	adam, lbfgs, sgd	**lbfgs**
Alpha	0.0001, 0.001, 0.01	**0.01**
max_iter	500, 1000	**500**

Table S2. Analysis of variance and equation of the all term included quadratic equation for predicting the phosphate removal rate of Al-FWB prepared using Box-Behnken designed conditions.

Source	Sum of squares	Degree of freedom	Mean square	F-value	p-value prob > F
Model	0.041	9	4.5×10^{-3}	10.23	0.0029
X_1	0.019	1	0.019	43.46	0.0003
X_2	2.8×10^{-4}	1	2.8×10^{-4}	0.63	0.4527
X_3	7.0×10^{-3}	1	7.0×10^{-3}	15.89	0.0053
X_1X_2	5.6×10^{-5}	1	5.6×10^{-5}	0.13	0.7316
X_1X_3	3.9×10^{-3}	1	3.9×10^{-3}	8.75	0.0211
X_2X_3	4.7×10^{-4}	1	4.7×10^{-4}	1.06	0.3373
X_1^2	9.2×10^{-3}	1	9.2×10^{-3}	20.85	0.0026
X_2^2	2.4×10^{-4}	1	2.4×10^{-4}	0.55	0.4816
X_3^2	3.7×10^{-5}	1	3.7×10^{-5}	0.083	0.7814
Residual	3.1×10^{-3}	7	4.4×10^{-4}		
Lack of fit	3.0×10^{-3}	3	1.0×10^{-3}	41.60	0.0018
Pure Error	9.6×10^{-5}	4	2.4×10^{-5}		
Total	0.044	16			
Equation: $Y = 0.71 - 0.049X_1 - 0.0059X_2 + 0.030X_3 - 0.0038X_1X_2 - 0.031X_1X_3 - 0.011X_2X_3 + 0.047{X_1}^2 + 0.0076{X_2}^2 + 0.030{X_3}^2$					

Table S3. Modeling statistics from the analysis of variance of the optimized quadratic equation used in predicting the phosphate removal rate of Al-FWB prepared using Box-Behnken designed conditions.

Parameters	**Value**	**Parameters**	**Value**
Square root of the residual mean square	0.019	**R^2**	0.904
Overall average of all the response data	0.74	**Adjusted R^2**	0.872
Coefficient of variation	2.54	**Predicted R^2**	0.776
Predicted residual error sum of squares	9.83E-3	**Adequate precision**	18.4

Table S4. Values of parameters determined from fitting the adsorption data of phosphate removal using Opt-Al-FWB to kinetic models.

Models	**Pseudo-first order**			**Pseudo-second order**		
Parameters	q_e *(mg/g)*	k_1 *(1/h)*	$\mathbf{R^2}$	q_e *(mg/g)*	k_2 *(g/mg/h)*	$\mathbf{R^2}$
Values	126.5	4.05	0.983	130.0	0.078	0.993

Table S5. Values of parameters of equilibrium models for phosphate adsorption using Opt-Al-FWB.

Models	**Langmuir**			**Freundlich**		
Parameters	Q_m (mg^-/g)	K_L (L/mg)	$\mathbf{R^2}$	K_F (L/g)	$1/n$	$\mathbf{R^2}$
Values	197.8	0.039	0.982	30.22	0.369	0.995

Table S6. Values of thermodynamic parameters for phosphate removal using Opt-Al-FWB (initial concentration: 500 mg/L, adsorbent dosage: 3.33 g/L, reaction time: 24 h).

Temperature (°C)	ΔH^0 (kJ/mol)	ΔS^0 (J/mol·K)	ΔG^0 (kJ/mol)
15	19.6	83.4	-4.4
25			-5.2
35			-6.0

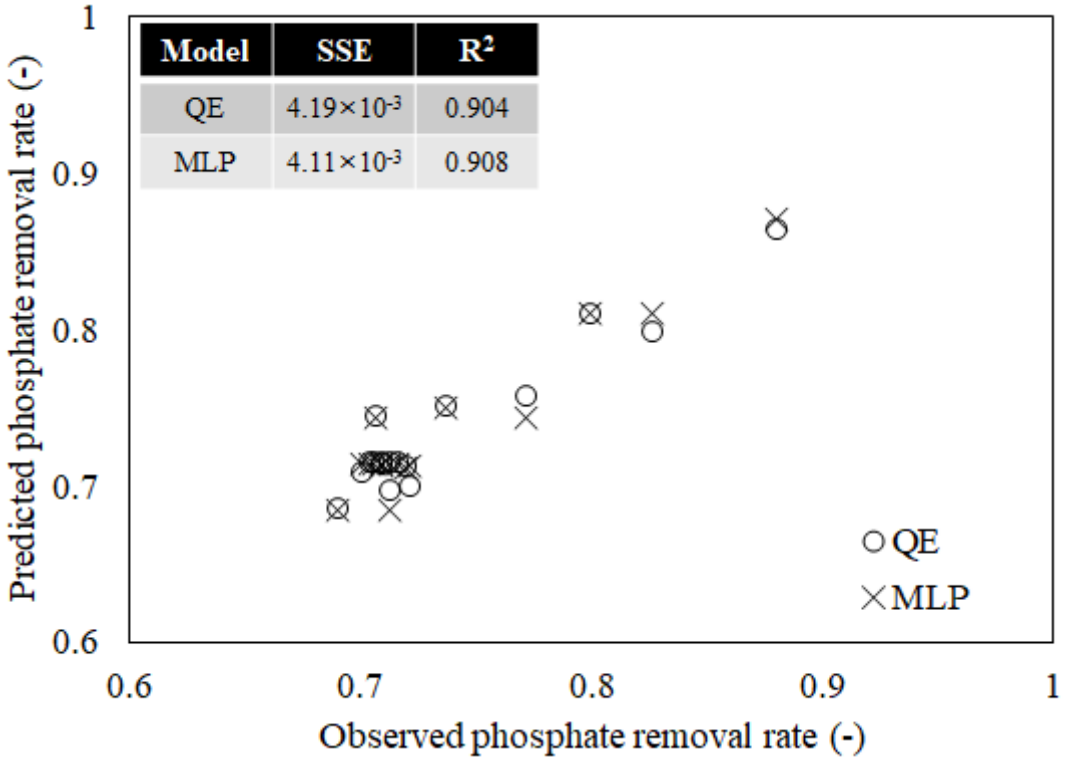

Figure S1. Pair plot, SSE, and R^2 for 17 datasets by quadratic equation (QE) and multi-layer perceptron (MLP).