**Optimize the Preparation of Novel Pyrite Tailings Based Non-Sintered Ceramsite (PTNC) by Plackett-Burman Design Combined with Response Surface Method For Phosphorus Removal**

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**Text S1** Adsorption kinetic models

Nonlinear form pseudo fist-order:

 (S1)

where qt is the adsorption amount at time t (mg/g), qe is the equilibrium adsorption amount (mg/g), and k1 is the equilibrium rate constant of the model (min-1).

Nonlinear formula of pseudo second-order model:

 (S2)

where k2 is the pseudo second-order rate constant (mg/g/min).

**Text S2** Adsorption isotherm models

Langmuir empirical model:

 (S3)

where Ce (mg/L) is the equilibrium adsorbate concentration in liquid, qm and kL are the Langmuir maximum adsorption capacity and the energy related constant respectively.

The Freundlich isotherm is described as the following equation:

 (S4)

where kF is the Freundlich constant and n is the heterogeneity factor (Freundlich coefficient).

**Text S3** Thermodynamic parameters calculation method

The free energy of adsorption (∆G), enthalpy of adsorption (∆H), and entropy of adsorption (∆S) during the adsorption process by PTNC was be calculated as with following equations:

 (S5)

 (S6)

Where R is the universal gas constant, 8.314 (J/mol/K); T (K) is the absolute solution temperature; K is the dimensionless thermodynamic equilibrium constant. The ΔH and ΔS values are calculated from the slope and intercept of the plot with the plot of lnK versus 1/T. Before the calculation process, the K gained from the best fitted model (Langmuir isotherm) need to be recalculated by multiplying it by 106 to become dimensionless, due to its unit was L/mg.

**Table S1** Chemical composition of original pyrite tailings

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Composition | S | Fe | Si | P | C | Mn |
| Percent (%) | 45.32 | 50 | 2.2 | 0.23 | 0.26 | 0.4 |

**Table S2** Chemical composition of dehydrated sludge

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Composition | SiO2 | Al2O3 | Fe2O3 | CaO | MgO | Na2O | K2O |
| Percent (%) | 63.51 | 17.21 | 6.53 | 0.96 | 3.65 | 1.71 | 2.89 |

**Table S3** List of levels and factors of Plackett-Burman experiment

|  |  |  |
| --- | --- | --- |
| Factors | Code | Levels |
| -1 | +1 |
| Cement  | A | 10 | 30 |
| Calcium lime | B | 1 | 3 |
| Anhydrous gypsum | C | 1 | 3 |
| Dehydrated sludge | D | 2 | 6 |
| Sodium bicarbonate | E | 0.5 | 1.5 |

**Table S4** Variables and levels in Box-Behnken design response surface method

|  |  |
| --- | --- |
| Levels | Factors |
| X1Dehydrated sludge (g) | X2Sodium bicarbonate (g) | X3Cement (g) |
| -1 | 2 | 0.5 | 10 |
| 0 | 4 | 1 | 20 |
| +1 | 6 | 1.5 | 30 |

**Table S5** Analysis results of specific surface area and pore size with BET method

|  |  |  |  |
| --- | --- | --- | --- |
| Parameters | Units | Original PTNC | PTNC after adsorption |
| Specific surface area | m²·g-1 | 7.21 | 11.39 |
| Average pore volume | cm³·g-1 | 0.024 | 0.029 |
| Average pore aperture | nm | 13.42 | 10.08 |

**Table S6** Physical characteristics of PTNC

|  |  |  |
| --- | --- | --- |
| Indicators | Standard value | PTNC performance |
| Water adsorption (%) | 22 | 25.08 |
| Cylinder compressive strength (MPa) | 1.0 | 1.80 |
| Porosity (%) | ≥40 | 53.90 |
| Solubility in hydrochloric acid (%) | ≤2 | 5.36 |
| Specific surface area (m2/g) | 0.5 | 7.21 |
| Wear rate (%) | ≤6 | 2.08 |
| Silt content (%) | ≤1 | 0.54 |

**Table S7** The concentrations of heavy metals in the PTNC leachate

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  | Cu | Pb | Cr | Zn | Ni | Cd |
| Detected value (mg/L) | 0.096 | 0.003 | 0.007 | 0.258 | 0.002 | 0.012 |
| Standard limitation （mg/L） | 100 | 5 | 5 | 100 | 5 | 1 |

**Table S8** The fitted parameters of two adsorption dynamic models

|  |  |
| --- | --- |
| Pseudo first order | Pseudo second order |
| qe1 (mg/g) | k1 (1/h) | R2 | qe2 (mg/g) | k2 (g/mg/h) | R2 |
| 0.2995 | 0.1661 | 0.9155 | 0.3527 | 0.4691 | 0.9100 |

**Table S9** The isothermal adsorption model parameters of TP adsorption

|  |  |  |
| --- | --- | --- |
| T (K) | Langmuir | Freundlich |
| qm (mg/g) | KL (L/mg) | R2 | KF (L/mg) | n (L/g) | R2 |
| 293 | 6.9603 | 0.8543 | 0.9421 | 3.0483 | 0.2487 | 0.8348 |
| 303 | 6.9824 | 0.8678 | 0.9574 | 3.0880 | 0.2495 | 0.8382 |
| 313 | 6.9978 | 1.1648 | 0.8530 | 3.2436 | 0.2353 | 0.7875 |

**Table S10** Thermodynamic parameters for TP adsorption by PTNC

| T (K) | ∆G (kJ/mol) | ∆H (kJ/mol) | ∆S (J/mol/K) |
| --- | --- | --- | --- |
| 293 | -33.27 | -11.69 | 153.09 |
| 303 | -34.45 |  |  |
| 313 | -36.35 |  |  |



  **Fig. S1** XRD pattern of pyrite tailings



**Fig. S2** Experimental device for dynamic adsorption experiment



**Fig.S3** Comparison analysis of experimental and predicted TP removal rates



**Fig. S4** Comparison XRD analysis of pyrite tailings, PTNC before and after adsorption of TP.



**Fig. S5** SEM image of PTNC after adsorption