Red mud particle adsorbent and its adsorption mechanism for phosphate ion

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ABSTRACT: There is a large amount of phosphorus in the flotation tailwater of phosphate (PW), which will cause eutrophication of water body if discharged directly, effective removal of phosphorus is of great importance in prevention of eutrophication. In this article, we report a Non-thermally activated red mud particle adsorbent (ARMPA) for removal of phosphate, the main mineral components were hematite, calcite, oolitic chlorite, katenite and calcium nepheline and the adsorbent was found to have a pH 9.25, a specific surface area of 40.54 m\textsuperscript{2}/g, a pore volume of 2.10 cm\textsuperscript{3}/g, a compressive strength of 1.31 KPa, and an immersion pulverization rate of 3.72\% at 24 h. This adsorbent can be used to adsorb phosphate ions (P) in PW. When the initial total phosphorus (TP) concentration was 156.7 mg/L, the amount of ARMPA was 4 g/L, and the adsorption time was 10 h, the adsorption capacity and removal efficiency of the TP were 38.46 mg/g and 98.17\%, respectively. The zeta potential, XRF, SEM, FT-IR, and XPS analysis showed that when pH 8~9, P mainly existed in the form of HPO_{4}^{2-} and PO_{4}^{3-}. These ions reacted with Ca\textsuperscript{2+}/Na\textsuperscript{+}/Al\textsuperscript{3+}/Mg\textsuperscript{2+}, etc., and formed strong chemical bonds through surface deposition and ion exchange, which then distributed on the inner surface of the ARMPA channels. The adsorption of TP by ARMPA accords with the pseudo-first-order dynamics model, and the Langmuir model can better describe the adsorption process. Phosphorus, fluorine and other toxicity indicators in the tail water after adsorption have reached the standard requirements. After adsorption of phosphate material, it is equivalent to using phosphoric acid tailwater to comprehensively modify alkaline red mud, so as to achieve the purpose of treating waste with waste.

Keywords: Phosphate ion, Phosphate flotation, Adsorption mechanism, Non-thermally activated red mud particle adsorbent.

1 INTRODUCTION

The Percolating Water from the phosphate ore flotation tailings reservoir is that the phosphate ore flotation wastewater is leached from the bottom of the tailings dam by dilution with rainwater and percolation of phosphate rock tailings or soil. The diversity of phosphate rock properties determines the complexity of phosphate ore beneficiation wastewater. In general, pH, chemical oxygen demand (COD), biological oxygen demand (BOD), suspended solid(SS), TP, Sulfides (S), fluoride (F) and other indicators in phosphate rock flotation wastewater exceed the National Discharge Standard Rank 2 for industrial wastewater.
Discharging this kind of wastewater directly into the water body will change the pH value of the water body, inhibit the growth of bacteria and microorganisms, hinder the self-purification of the water body, and destroy the normal ecological cycle (Suman Mor et al.2016). Specifically, the TP content of this percolating water is as high as 50~150mg/L. Therefore, it must meet national standards before it can be discharged. Otherwise, it will lead to eutrophication of water bodies (State Environmental Protection Administration, 2002).

At present, wastewater treatment methods mainly include solidification, froth flotation, filtration, ion exchange, aerobic and anaerobic treatment, advanced oxidation treatment, solvent extraction, adsorption (Ying Wang et al.2016), electrolysis, microbial reduction and activated sludge (Ru Cai et al.2017), etc. However, these technologies need to be further improved, mainly due to insufficient removal of pollutants, high cost, and high requirements for reagents or energy consumption, and in addition, some may produce toxic residues or other wastes that require further safe disposal (Amit Bhatnagar et al.2011). Removing contaminants from water through adsorption is considered to be the most versatile method. Activated carbon is a very promising adsorbent commonly used to remove different types of contaminants from water and wastewater. However, the widespread use of activated carbon in water treatment is limited by its high cost. Therefore, the development of low-cost materials of adsorbents has become the direction of research in this field (Yi Zhi Wang et al.2016; Patricia B. et al.2018).

Red mud (RM) is the red muddy solid waste remaining after extracting A12O3 component from bauxite in the aluminum industry production process. Because the surface of calcium, sodium, aluminum and magnesium oxides in red mud has strong reactivity (Vaibhav R. et al.2018; Tengfei Guo et al.2018), red mud shows good adsorption to heavy metal ions (Wu Ming Xie et al.2018) such as Cu2+, Pb2+, Zn2+, Ni2+, Cr2+ and Cd2+ in water (A. Naga Babu, et al.2017; S. Lakshmi Narayanan G et al.2018). Red mud contains a large amount of metal oxides and silicates, which can form a structure with a certain functional group on the surface, and remove specific anions through ligand exchange and surface precipitation (Thiquynhxuan Le et al.2018). Many scholars have found that the original or modified red mud can exhibit certain removal characteristics for anionic pollutants (AsO43-, AsO3-, F-, PO43-) in water (Zhong Pan Hu et al.2018). Red mud can effectively remove phosphate ions from water (Pan Hu et al.2017). Because, specifically, CaO, A12O3 and Fe2O3 can form insoluble precipitates with phosphate, while A12O3 can also undergo hydroxylation at a certain pH value, and can adsorb and coagulate the phosphorus in water (Ratnamala Gadigayya Mavinkattimath et al.2017). The iron, aluminum and silicon oxides on the red mud surface can be hydrolyzed in water, and thus, the adsorption of phosphorus can be explained by the mechanism of ligand exchange reaction.

To improve the adsorption capacity of red mud to phosphate, many scholars have carried out a great amount of research work. In summary, it can be divided into shape regulation (powder, columnar, granular, flake), acid activation (Hydrochloric acid, sulfuric acid, nitric acid, carbon dioxide) (G. Venkatesan et al.2018), calcination activation (control sintering temperature) (Nayara T. do Prado et al.2017), combined activation technology, and static, dynamic adsorption and desorption control techniques, etc., as well as related mechanism research work. These works laid a good foundation for the application of phosphate ions removal from phosphorus-containing wastewater by red mud.

However, red mud as an alternative to activated carbon, does not have large-scale application of adsorbents for phosphorus-containing wastewater. There are still some limitations in current research (Kyung Won Jung et al.2016). First of all, the phosphorus-containing wastewater used in most studies is a simulated phosphorus-containing liquid, which has certain guiding significance for the mechanism discussion, but the actual industrial wastewater...
composition is more complicated, and it is necessary to evaluate the adsorption capacity of red mud to multi-component pollutants. Secondly, the adsorbent prepared by the calcination method has excellent adsorption performance, and the waterproof erosion meets the requirements, but the roasting cost is too high. However, powder adsorbents or particulate adsorbents prepared by non-calcination method do not take into account the relationship between porosity and particle compressive shear resistance, resulting in lower adsorption efficiency. Thirdly, the acid activation needs to be immersed in a large amount of acid solution and then dried. The acid decomposition in the filtration and drying process will cause secondary pollution to the water and air, and the cost is also high. Finally, the shape control of the pores of the adsorbent is neglected, and an efficient semi-open pore adsorption mode is not formed.

Therefore, in order to improve the anti-powdering performance, adsorption performance and strength of the red mud particle adsorbent, and to reduce production costs and environmental pollution, RM was used as raw material, and fly ash (FA) (pore-assisting agent), C2S-HPMC (binders), HCl (modified active agent) and H2O2-MnO2 (pore-forming active agents) were then added to prepare a kind of Non-thermally Activated Red Mud Particle Adsorbent (ARMPA) through a two-step method granulation technology. This product has the characteristics of environmental friendliness, high porosity, strong adsorption capacity, good mechanical properties and strong powder resistance. The phosphate ion (P) in the percolating water (PW) of the tailings dam of Guizhou Wengfu Phosphorus Ore Dressing Plant was adsorbed by the experimentally prepared ARMPA, and the adsorption mechanism of ARMPA to P was revealed, which laid a foundation for large-scale industrial application.

2 MATERIALS AND METHODS

2.1 Raw Materials and reagents

The sample of red mud was collected from Guizhou Huajin Aluminum Co., Ltd., with a water content of 30%. After drying at 50 °C for 12 hours, it was ground in a planetary ball mill through a 0.075 mm mesh sieve. The fly ash sample was collected from Qiantong Fly Ash Co., Ltd., Fuquan City, Guizhou Province. The original ash has a Loss on ignition of ≤ 5%, SiO2 content ≥ 30%, SO3 content ≤ 2%, it is a secondary fly ash. The fly ash was ground to less than 200 mesh before use. The water sample was taken from the leachate of tailings dam of WengFu phosphate ore dressing plant in guizhou province, China. The cement is 42.5 R high belite cement, purchased from Tangshan Polar Bear Building Materials Co., Ltd.; H2O2 (industrial grade, 30%) was purchased from Tianjin Hongshuoda Trading Co., Ltd.; MnO2 (industrial grade) from Chengdu Jinshan Chemical Reagent Co., Ltd.; HPMC (industrial grade, 2 million sticks) from Hebei Yanxing Chemical Co., Ltd.; HCl (concentration 36%~38%, analytical grade) was purchased from Hengyang Guangheng Hongda Chemical Co., Ltd.; 38% concentrated HCl (analytical grade) from Guangzhou Wan Cong Chemical Co., Ltd.; the standard solutions of Ca, Mg, Si, P, Al and Fe were developed by Beijing Research Institute of Nonferrous Metals; HNO3 (analytical grade, 68%) was purchased from Guangzhou Wancon Chemical Co., Ltd.; deionized H2O is self-prepared in the laboratory.

2.2 ARMPA particle adsorbent preparation

2.2.1 Preparation scheme

It has been found that when the particle size of the adsorbent particles is larger than 2 mm, the solution is difficult to penetrate into the center of the ball in a short time, thereby prolonging the adsorption time (Lan Ban et al.2019). In addition, if there is no supporting core, the strength of the adsorbent particles is low, and the dynamic adsorption loss rate would be relatively high. In the process of pore making, the pores are easy to become through-holes. Further if the pores are worn along the latitude and longitude lines, it is not good for adsorption (Lan Ban et al. 2019). Therefore, in this study, the granulation design was first carried out during the granulation process. The
ARMPA model is shown in Figure 1. The adsorbent particle has a two-layer structure, and the inner layer is a nucleating support body with particle size of less than 1 mm. It has relatively dense, and the powdering rate of which is less than 1% at 24 h, the strength should be greater than 2 KPa, and has good surface wettability and stickiness. The external one is the adsorption active coating layer, which has more adsorption active sites, a larger specific surface area, good wettability, certain strength and good anti-pulverization property. Blocked by the nucleation core, the pores are semi-via holes, which are favorable to physical or chemical adsorption than that of through holes. Since the particle adsorbent has a nucleating particle support, it is easy to prepare particles with high sphericity and good mechanical properties, and thus prepared particle adsorbent with good adsorption stability. When preparing the particle adsorbent, the particle size, weight, specific surface area, strength and powdering rate of each layer are first designed, then the nucleation support body is prepared, followed by the adsorption support body is prepared, followed by the adsorption active coating layer.

![Dense layer: nucleating support body](image1)

**Figure 1. ARMPA model (left) and laboratory prepared particulate adsorbent (right)**

### 2.2.2 Preparation of bonding active liquid

The main roles of the binder active solution are to change the pH and zeta potential of the complex system, to adjust the adsorption heat release rate of the adsorbent, to change the apparent morphology of the adsorbent, to adjust the specific surface area, pore volume and pore distribution, and to adjust particle strength and water immersion rate. For a test group, add 105 ml of deionized water, and then add HPMC as the binder with mass fractions of 0.5 , 1 , 1.5 , 2.0 , 2.5 %, respectively. After magnetic stirring for 5 minutes, the effect of HPMC on the performance of adsorbent particles were evaluated. On this basis, hydrochloric acid (2 mol/L) with mass fractions of 0.03, 0.06, 0.09, 0.12 and 0.15%, respectively, was added and followed by magnetic stirring for 5 minutes, the effect of the amount of HCl added to the performance of adsorbent particles was studied. After that, hydrochloric acid (2 mol/L) with mass fractions of 0.03, 0.06, 0.09, 0.12 and 0.15%, respectively, was added and followed by magnetic stirring for 5 minutes, the effect of the amount of HCl added to the performance of adsorbent particles was studied. After that, the pore modifier H2O2 with mass fractions of 0.2, 0.4, 0.6, 0.8, and 1.0% was continuously added, and the mixture was magnetically stirred for 5 minutes, and the effect of the amount of H2O2 was investigated.

### 2.2.3 Preparation of nucleation particle support

Mix RM and C2S (mass ratio of 6:4) in a high-speed powder dispersion mixer, and then put it into a disc granulator for molding. During the molding process, the rotation speed of the disk is 90r/min, and a certain amount of deionized water is added by a sprayer until the molding work is completed. After molding, the product was sieved with a sieve having a mesh size of 1 mm, and then the undersize proportion was placed in a constant temperature electric drying oven (at 40 °C) to dry to a moisture content of <5%.

### 2.2.4 Preparation of particle adsorbent

Add RM, FA, MnO2 and other raw materials into a high-speed powder disperser for mixing. The total mass of the test materials is about 300g. Add FA with mass fractions of 1.7, 3.4, 5.1, 6.8 and 8.5%, respectively, and keep the mass ratio of MnO2 to H2O2 as 1 to 5. First,
add nucleating particle support with a certain proportion, then start the disc granulator, add the main ingredients and the prepared adhesive active liquid while rotating. During the molding process, control the addition speed and addition amount of the bonding active liquid until the adsorption active coating layer reaches a certain thickness, and then continue to rotate for 10 minutes. After that, the product is put into a standard cement concrete curing box with constant temperature and humidity for 3 days, keep the curing temperature at 22-25 °C and the curing humidity at 80%. After curing, dry it at 40 °C.

2.3 Test methods

2.3.1 Adsorption test

Take 1000 ml of phosphorus-containing wastewater into a 1500 ml beaker, then add 4 g of ARMPA adsorbent, place the beaker after slight hand shaking. Take 5 ml of supernatant from the beaker after 2nd, 4th, 6th, 8th and 10th hour, dilute each of the water sample to 15 ml with deionized water and then measured the content of the TP. Track the pH variation of the test solution in the beaker with a pH meter. Thus, phosphorus removal efficiency, adsorbent adsorption capacity, and immersion pulverization ratio were calculated by using formula 1-3 as following (Lv J H et al, 2014).

\[ \eta = \frac{C_0 - C_i}{C_0} \times 100\% \]  \hspace{1cm} (1)

\[ Q = \frac{V(C_0 - C_i)}{m} \times 100\% \]  \hspace{1cm} (2)

\[ \eta_m = \frac{m - m_i - m_s}{m} \times 100\% \]  \hspace{1cm} (3)

Where \( \eta \) means phosphorus removal efficiency in solution (\%), \( C_0 \) means phosphorus concentration in solution before adsorption (mg/L), \( C_i \) means phosphorus concentration in solution after adsorption (mg/L), \( \eta_m \) means the immersion pulverization ratio (%), \( m_i \) means the dry weight of adsorbent after adsorption(g), \( m_s \) means the adsorption weight (g).

2.3.2 Product characterization methods

The concentration of TP and other ions (Ca2+, Mg2+, Al3+, Fe2+, K+, Na+, F- and As2+) in the percolating water were determined by yttrium internal standard method using an inductively coupled plasma emission spectrometer (ICP-7400, Thermo Fisher Scientific, USA). The phase and crystal structure of the sample were characterized by an X-ray diffractometer (POWDER PRO, PANalytical, Netherlands) using a Cu target Kα source (\( \lambda = 0.154 \) nm), a tube voltage of 40 kV, and a tube current of 40 mA. The test uses a step scan with a step size of 0.05, a scan speed of 0.5o/s, and a scan range of 10o-70o. The specific surface area, pore volume and pore size of the particulate adsorbent were tested using an N2 adsorption/desorption physical adsorption instrument (BET) (ASAP2020, Micrometrics, USA), in which the specific surface area (SBET) is calculated by the Barrett-Emmer-Teller method, while the pore volume (VBJH) is calculated by the Barrett-Joyner-Halanda (BJH) model from the isothermal adsorption branch. Specifically, the pore volume is calculated by the adsorption capacity at the relative pressure \( P/P_0 = 0.975 \). Determination of particle strength is conducted by using APT-3 particle strength tester. The chemical composition of the samples was determined by X-ray fluorescence spectrometer (XRF) (AxiosmAX, 4kW, PANalytical B.V., Netherlands). The surface structure of the red mud original sample and the particulate adsorbent was observed by a S-3400 scanning electron microscope (SEM) manufactured by Japan Tianli Co., Ltd., and the test operating voltage is 20 kV. The functional groups contained in the samples before and after adsorption were characterized by a VERTEX 70 type Fourier infrared spectroscopy analyzer from BRUKER, Germany. A solid infrared method is applied as follow: a small amount of sample (about 0.6 mg) and a certain amount of potassium bromide (about 200 mg) were uniformly ground in an agate mortar, compressed, and the sample was subjected to FT-IR test with a wavenumber range of 4000-
400 cm⁻¹. The surface zeta potential of the sample and the adsorbent was measured using a Nano-particle size/Zeta potential analyzer (Delsa Nano C, Beckman Coulter, USA). The heat value of the adsorption process of the adsorbent was measured by a microcalorimeter (C80, Setaram, French). X-ray photoelectron spectroscopy (AXIS ULTRADLD, Kratos, Shimadzu Group, Monochromated Al Target, 150 W) was used to scan the particle samples before and after adsorption to analyze the composition changes of the surface materials of the adsorbents. The relative content and bonding mode of phosphorus (P 2p) in the particle sample after adsorption of phosphate were analyzed to reveal the mechanism of action of phosphorus on the surface of the adsorbent.

2.3.3 Adsorption kinetic analysis

The adsorption rate of phosphate ions is evaluated by primary and secondary kinetic models. The expressions of the pseudo-first-order dynamic equation and the pseudo-second-order dynamic equation of the Lagergren equation are shown in Equation 4 (Lagergren, 1898) and Equation 5 (Freundlich H. 1906), respectively.

\[ \log (Q_e - Q_t) = \log Q_e - \frac{k_1}{2.303} t \]  
\[ \frac{1}{Q_t} = \frac{1}{Q_e} + \frac{t}{k_2 Q_e} \]  

Where \( Q_e \) means the adsorption capacity of P at equilibrium (mg/g), \( Q_t \) means the amount of adsorption of TP at the time of \( t \) (min) (mg/g), \( K_1 \) means the rate constant for first-order kinetic adsorption, \( K_2 \) means the second-order kinetic adsorption constant, \( K_2 \) can be calculated from the slope and intercept of curve \( t/Q_t \), the adsorption rate constant can be determined from the slope in a linear graph of \( \log(Qe-Qt) \) versus \( t \).

2.3.4 Adsorption equilibrium isotherm

The phosphorus-containing wastewater was filtered through a 45 μm filter membrane and diluted to a solution having a phosphorus content of 20, 25, 30, 40, 50, 65, 90, 120, 150 mg/L by stepwise dilution, and 0.1 mol/L NaOH adjusts the pH to around 8. To ensure that the adsorption reaches equilibrium, the reaction time is set at 10 h and the temperature is 25°C. The adsorption isotherms of RM and ARMPA were determined separately, and their adsorption isotherms were thus fitted respectively. The adsorption capacity of the adsorbent was determined by the Freundlich and Langmuir isothermal models, where the Freundlich isotherm model was expressed as Equation 6 (Freundlich H. 1906).

\[ Q = \frac{k_f C_e^{1/n}}{1 + \frac{C_e}{Q_m}} \]  

Where \( Q \) (mg/g) is adsorption capacity of P by unit mass of adsorbent, \( C_e \) (mg/g) is the concentration of P at equilibrium, \( K_f \) is the Freundlich constant related to adsorption capacity, \( n \) is the Freundlich constant indicating the adsorption intensity, The expression of the Langmuir isotherm model is shown in Equation 7 (Langmuir I. 1918).

\[ \frac{C_e}{Q_e} = \frac{1}{bQ_m} + \frac{C_e}{Q_m} \]  

Where \( Q_e \) (mg/g) is adsorption capacity of TP by unit mass of adsorbent, \( C_e \) (mg/L) is the single layer capacity required for unit weight adsorbent adsorption, \( b \) (L/mg) is Langmuir constant.

To further investigate the adsorption mechanism, the experimental data were fitted to the Dubinin-Rasdushkevich (DR) isotherm. The DR isotherm has the form of Equation 8 (Thomas W J et al. 1918).

\[ \ln Q = \ln Q_m - K_{DR} \varepsilon^2 \]  

Where \( K_{DR} \) is constant associated with adsorption energy, \( Q_m \) is adsorption capacity, \( \varepsilon \) (Polanic potential) is expressed as \( RT \ln (1+1/C_e) \), in which \( R \) is the gas constant and \( T \) is the temperature. Usually DR isotherms are used to distinguish between physical
adsorption and chemisorption. The average amount of adsorption (E) can be calculated according to the KDR value using the formula (Maiti A et al. 2007):

\[ E = -(2K_{DR})^{0.5} \]  

(9)

The type of adsorption can be distinguished by estimating the value of E. If the E value is between 8 and 16 kJ/mol, it is chemisorption. If E < 8 kJ/mol, it is physical adsorption.

3 RESULTS AND DISCUSSION

3.1 Red Mud XRD Analysis

The composition analysis results of PW is shown in Table 1. The TP content is 156.7 mg/L, which exceeds the GB3838-02 national industrial wastewater discharge standard Rank 2 (the emission standard of P is less than 0.5 mg.L-1), and pH also exceed the standard, so it cannot directly discharged without treatment.

Table 1. The composition of the percolating water from the phosphate ore flotation tailings reservoir

<table>
<thead>
<tr>
<th>Main ingredients</th>
<th>value</th>
<th>standard</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca²⁺/mg.L⁻¹</td>
<td>213.15</td>
<td>--</td>
</tr>
<tr>
<td>Mg²⁺/mg.L⁻¹</td>
<td>401.4</td>
<td>--</td>
</tr>
<tr>
<td>Al³⁺/mg.L⁻¹</td>
<td>0.253</td>
<td>--</td>
</tr>
<tr>
<td>Fe²⁺/mg.L⁻¹</td>
<td>0.022</td>
<td>--</td>
</tr>
<tr>
<td>K⁺/mg.L⁻¹</td>
<td>66.35</td>
<td>--</td>
</tr>
<tr>
<td>Na⁺/mg.L⁻¹</td>
<td>277.01</td>
<td>--</td>
</tr>
<tr>
<td>TP/mg.L⁻¹</td>
<td>156.7</td>
<td>&lt;0.5</td>
</tr>
<tr>
<td>F/mg.L⁻¹</td>
<td>33.2</td>
<td>&lt;6</td>
</tr>
<tr>
<td>As²⁺ /mg.L⁻¹</td>
<td>3.771</td>
<td>&lt;0.3</td>
</tr>
<tr>
<td>pH</td>
<td>5.22</td>
<td>6~9</td>
</tr>
</tbody>
</table>

The results of the phase identification and quantitative phase analysis of the red mud sample are given in Figure 2, showed that the outputs from the Rietveld analyses of the sample. Figure 2 output from rietveld analysis of the red mud sample where the blue line is the observed data, the red is the calculated pattern and the grey the difference. The vertical lines are the Bragg peak markers for (top to bottom) katoite(39.1%), calcite(7.4%), hematite(5.3%), cancrinite(26.1%), diaspore(1.3%), anatase(1.5%), kaolinite(3.6%), clinoclore(12.9%), and muscovite(3.0%).

3.2 ARMPA preparation and performance testing

3.2.1 Effect of FA on the performance of adsorbents

The granulation molding and adsorption tests were carried out according to the method described in 2.3. After the adsorption, the adsorption capacity, the removal efficiency, the particle compressive strength and the immersion pulverization ratio in 24 h were tested, respectively. It can be seen from Figure 3 that when the adsorption temperature is 298 K and the adsorption time is 10 h, the adsorption effect of RM on P is not satisfactory, and the removal efficiency is only 62.62%. In addition, it can be seen from Figure 4 that the particle adhesion is unsatisfactory, the strength is only 0.36 KPa, the particles placed in the PW are disintegrated and eroded, and the immersion pulverization ratio is as high as
87.4% in 24 hours. After the addition of FA, the removal efficiency of TP gradually increased. However, FA reduces the viscosity of the adsorbent and lowers the strength. For example, when the amount of FA added is 8.3%, the removal efficiency of TP reaches 76.6%, but at the same time the immersion pulverization ratio reaches 94.30%. The addition of FA increases the adsorption active substances such as Al2O3 and Fe2O3, thus increasing the specific surface and active sites of the adsorbent. However, the adhesion of FA is poor, resulting in a decrease in the true density of the particles and a loose structure. This lowered the particle strength and increased the immersion pulverization ratio.

Figure 3. Effect of FA on the adsorption capacity of adsorbents

Figure 4. Effect of FA on the compressive strength and the immersion pulverization ratio of adsorbents

3.2.2 *Effect of HPMC on the performance of adsorbents*

In order to increase the strength and reduce the immersion pulverization ratio of the adsorbent, HPMC was added as a binder on the basis of adding FA. It can be seen from Figure 5 and Figure 6 that under the same conditions, the adsorption capacity of the adsorbent to TP decreases with the addition of HPMC. When the dosage of HPMC increased from 0.05% to 0.25%, the removal efficiency of TP by the adsorbent decreased from 79.69% to 68.95%, but the strength increased from 0.58 KPa to 0.81 KPa, and the immersion pulverization ratio of the particles decreased to 3.5% when the dosage of HPMC is 0.25%. HPMC and water form a sol during the granulation of the adsorbent, which can increase the consistency and viscosity of the binder. At the same time, HPMC and solid particles form a network-like structure after hydration, increasing the contact angle of the surface of the particles and partially filling it and reducing the adsorption capacity. This property of HPMC can increase the strength of the particulate adsorbent and the rate of soaking loss, and also improve the adsorption stability of the adsorbent.

Figure 5. Effect of HPMC on the adsorption capacity of adsorbent

Figure 6. Effect of HPMC on the compressive strength and the immersion pulverization ratio of adsorbents
3.2.3 Effect of C2S on the performance of adsorbents

The compressive strength and the immersion pulverization ratio of adsorbent added with HPMC still fail to meet the requirements. Adding C2S as a strength enhancer, the purpose is, by synergistic action with HPMC, to increase the compressive strength and reduce the immersion pulverization ratio of the adsorbent. It can be seen from Figure 7 and Figure 8 that when the amount of C2S is increased from 1% to 5%, the compressive strength of the adsorbent is increased from 0.93 KPa to 1.42 KPa, and the immersion pulverization ratio is reduced from 3% to 1.2%, but at the same time, the removal efficiency of TP reduced from 70.35% to 60.21%. C2S is a high-performance low-environment load cement containing more than 50% dicalcium silicate. It can be characterized by low hydration heat, high strength and fast hydration time. By adding C2S during the granulation process, the granules can be quickly hydrated and the processing time is shortened. However, the addition of C2S increases the compactness of the particles, and some of the pores are blocked, resulting in a decrease in adsorption capacity.

3.2.4 Effect of H2O2&MnO2 on the performance of adsorbent

While the particle strength increases but the pulverization rate decreases, the adsorption effect weakens again. It can be seen from Figure 9 and 10 that the addition of H2O2&MnO2 as a pore-forming agent can increase the adsorption capacity of the adsorbent from 76.91% to 93.71%. MnO2 is a catalyst for the decomposition of H2O2. Under alkaline conditions, H2O2+MnO2 can generate a large number of microbubbles, dredge the channels of adsorption sites, and expose larger adsorption surfaces. Both H2O2 and MnO2 have good oxidation performance, which lowers the metal valence state and thus increases the adsorption activity point during the adsorption process. However, the strength of the particle adsorbent will decrease sharply due to the generation of microbubbles, leading to a large mass loss during immersion. This has a certain adverse effect on the repeated regeneration of the adsorbent.
3.2.5 Effect of HCl on the performance of adsorbents

As can be seen from Figure 11 and Figure 12, the porosity was improved by HCl activation and addition of H2O2 and MnO2 foaming agent, the removal efficiency of phosphorus reached 98.12%, the strength is 1.03 KPa, and the 24 h immersion pulverization ratio was <5%. HCl is a monobasic strong acid that releases a large amount of H+ in the binder. With the increase of the amount of HCl, the adsorption capacity of the adsorbent increases first and then decreases, and the adsorption performance is best when the addition amount is 0.5%. HCl activates minerals, which changes the pore structure of the adsorbent, removes impurities in the pores, and facilitates the entry and adsorption of pollutant molecules. After that, as the HCl dosage increases, the adsorption capacity decreases. This is because during the granulation process, HCl dissolves K+, Ca2+, Na+, Fe2+ and Al3+ in the adsorbent, eliminating the original interlayer bond force and causing the cracking of layer crystal lattice. HCl activates minerals, which changes the pore structure of the adsorbent and removes impurities in the pores, and therefore, facilitates the entry and adsorption of pollutant molecules. When the amount of HCl is increased, the OH- and H+ in the system are neutralized, and through pores are thus partially formed which reduce the adsorption capacity. At this time, the strength of the adsorbent is decreased, and the immersion pulverization ratio is increased.

Figure 9. Effect of H2O2&MnO2 on the adsorption capacity of adsorbent

Figure 10. Effect of H2O2&MnO2 on the compressive strength and the immersion pulverization ratio of adsorbents

Figure 11. Effect of HCl on the adsorption capacity of adsorbent

Figure 12. Effect of HCl on the compressive strength and the immersion pulverization ratio of adsorbents

3.3 Performance Optimization and Testing

3.3.1 Performance Optimization

In order to achieve the best adsorption performance, we keep the amount of RM, FA
and water unchanged, and carry out a 4 factor-3 level orthogonal test with HPMC, C2S, H2O2+MnO2 and HCl as variables. The test results are shown in Table 2. The order of the range of the difference: (1) Influencing factors of the adsorption capacity is B>D>C>A, thus the optimal design should be B3D3A2C2; (2) Influencing factors of strength is A>C>D>B, thus the optimal design should be B2A2C3D2. Considering the cost and effect, B3A2C2D3 is the optimal design. Considering the effects of adsorption capacity, removal efficiency, particle strength and immersion pulverization ratio, we found the best ratio (mass ratio) of ARMPA particle adsorbent is RM:FA:water=19:1:7, and 0.15% HPMC, 0.17% HCl, 4% C2S, 1% H2O2 and 0.2% MnO2. The pH, the specific surface area and pore volume of the ARMPA particle adsorbent prepared under the above conditions were 9.25, 40.54 m2/g and 2.10 cm3/g, respectively. For the PW with initial total TP concentration of 156.7 mg/L, when keep the amount of ARMPA is 4 g/L and the adsorption time is 10 h, the adsorption capacity is 38.46 mg/g, the removal efficiency of P is 98.17%, the strength is 1.31 KPa, and the immersion pulverization ratio is 3.72%.

<table>
<thead>
<tr>
<th>No.</th>
<th>A (HPMC/%)</th>
<th>B (H2O2&amp;MnO2/%)</th>
<th>C (C2S/%)</th>
<th>D (HCl/%)</th>
<th>j (Adsorption capacity/mg.g⁻¹)</th>
<th>i (Intensity/KPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.1</td>
<td>0.27</td>
<td>3</td>
<td>0.09</td>
<td>35.82</td>
<td>0.89</td>
</tr>
<tr>
<td>2</td>
<td>0.1</td>
<td>0.80</td>
<td>4</td>
<td>0.17</td>
<td>37.65</td>
<td>0.97</td>
</tr>
<tr>
<td>3</td>
<td>0.1</td>
<td>1.00</td>
<td>5</td>
<td>0.26</td>
<td>38.04</td>
<td>0.96</td>
</tr>
<tr>
<td>4</td>
<td>0.15</td>
<td>0.27</td>
<td>4</td>
<td>0.26</td>
<td>36.72</td>
<td>1.27</td>
</tr>
<tr>
<td>5</td>
<td>0.15</td>
<td>0.80</td>
<td>5</td>
<td>0.09</td>
<td>37.05</td>
<td>1.31</td>
</tr>
<tr>
<td>6</td>
<td>0.15</td>
<td>1.00</td>
<td>3</td>
<td>0.17</td>
<td>38.46</td>
<td>1.23</td>
</tr>
<tr>
<td>7</td>
<td>0.2</td>
<td>0.27</td>
<td>5</td>
<td>0.17</td>
<td>36.39</td>
<td>1.11</td>
</tr>
<tr>
<td>8</td>
<td>0.2</td>
<td>0.80</td>
<td>3</td>
<td>0.26</td>
<td>38.43</td>
<td>1.13</td>
</tr>
<tr>
<td>9</td>
<td>0.2</td>
<td>1.00</td>
<td>4</td>
<td>0.09</td>
<td>37.41</td>
<td>1.12</td>
</tr>
</tbody>
</table>

Comparing the results of similar studies, as shown in table 3, Many studies have shown that its adsorbents are phosphorous preparations. In this paper, phosphorus separation tail water in actual phosphate ore dressing plant is used. ARMPA adsorbed total phosphorus up to 38.46 mg/g, which has a good absorption efficacy.
Table 3. Comparison of research results

<table>
<thead>
<tr>
<th>Adsorbent</th>
<th>Adsorbate</th>
<th>Adsorption Quantity/mg g⁻¹</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chitosan/Ca-organically modified montmorillonite</td>
<td>H₃PO₄</td>
<td>76.15</td>
<td>Jiseon Jang et al. 2019.</td>
</tr>
<tr>
<td>Oxygen furnace slag</td>
<td>KH₂PO₄</td>
<td>21-30</td>
<td>Chong Han et al. 2018.</td>
</tr>
<tr>
<td>PVA-CSH</td>
<td>KH₂PO₄</td>
<td>31.06</td>
<td>Shilin Ding et al. 2018.</td>
</tr>
<tr>
<td>Fe-loaded ceramic</td>
<td>NaH₂PO₄</td>
<td>45.88</td>
<td>Di Wang et al. 2016.</td>
</tr>
<tr>
<td>ARMPA</td>
<td></td>
<td>38.46</td>
<td>This paper</td>
</tr>
</tbody>
</table>

3.3.2 Adsorption time and adsorbent dosage

It can be seen from Figure 13 that when the adsorption time is longer than 10 h, the adsorption rate is lowered, so the optimum adsorption time is determined to be 10 h. Add 1, 2, 3, 4, 5 and 6 g ARMPA to a set of 1 L PW to investigate the effect of adsorbent dosage on adsorption. The result (Figure 14) showed that when the amount of the adsorbent is 4g/L, the adsorption efficiency is the best, and the removal efficiency of TP is 98.17%.

![Figure 13. Effect of adsorption time on TP adsorption from PW](image1)

![Figure 14. Effect of ARMPA dosage on TP removal from PW](image2)

3.3.3 pH variation of the adsorption system

The pH of RM and particulate adsorbent ARMPA were 10.35 and 9.35, respectively. As the adsorption time increased, the pH of ARMPA decreased to about 8.45 (see a in Figure 15). The original pH of PW is 6.80, and this figure increases to about 8.50 after 10 hours adsorption (see b in Figure 15). Therefore, as time increases, the pH of the entire adsorption system tends to be around between 8.45 and 8.52. This is because a large amount of alkaline substances in the red mud (such as sodium hydroxide, sodium bicarbonate, sodium carbonate, sodium aluminate, calcite, sodalite, hydrated garnet, etc.) are encapsulated by HPMC and C2S after being made into a particulate adsorbent, exhibiting a relatively
lower initial pH. However, when the adsorbent is immersed in PW, alkaline substances are hydrolyzed to produce OH- ions, resulting in an increase in pH. The OH- released by the adsorbent combines with H+ in the PW, and finally the pH of the system tends to be stable.

Figure 15. pH of ARMPA and PW with variation of adsorption time

3.4 Analysis of the adsorption mechanism of ARMPA

3.4.1 Chemical composition during the adsorption process

It can be seen from Table 4 that the content of components in the ARMPA which are related to adsorption performance (such as Fe2O3, Al2O3) are obviously increased. After adsorption, the P2O5 content in ARMPA increased, indicating that TP was successfully adsorbed. However, the content of CaO, Al2O3, Fe2O3 and K2O in ARMPA decreased, indicating that the alkaline matter decreased after adsorption and formed (Al2O3)10(MgO)10(Na2O)10(P2O5)70, CaHPO4, Na2HPO4 and other substances. The pH was also observed to have a downward trend, which is consistent with the pH test results in Figure 15.

Table 4. Chemical components of raw and activated red mud and fly ash /%

<table>
<thead>
<tr>
<th>Species</th>
<th>P2O5</th>
<th>SiO2</th>
<th>Fe2O3</th>
<th>Al2O3</th>
<th>CaO</th>
<th>MgO</th>
<th>MnO</th>
<th>TiO2</th>
<th>K2O</th>
<th>Na2O</th>
</tr>
</thead>
<tbody>
<tr>
<td>RM</td>
<td>0.33</td>
<td>18.93</td>
<td>6.92</td>
<td>25.56</td>
<td>13.74</td>
<td>0.74</td>
<td>0.024</td>
<td>4.35</td>
<td>1.33</td>
<td>3.35</td>
</tr>
<tr>
<td>FA</td>
<td>0.35</td>
<td>39.74</td>
<td>11.88</td>
<td>13.38</td>
<td>1.70</td>
<td>0.74</td>
<td>0.071</td>
<td>1.47</td>
<td>1.52</td>
<td>0.24</td>
</tr>
<tr>
<td>HCl modified</td>
<td>0.57</td>
<td>13.89</td>
<td>16.54</td>
<td>33.34</td>
<td>7.42</td>
<td>0.81</td>
<td>0.05</td>
<td>15.859</td>
<td>1.10</td>
<td>0.52</td>
</tr>
<tr>
<td>ARMPA</td>
<td>0.33</td>
<td>18.27</td>
<td>15.24</td>
<td>27.25</td>
<td>10.00</td>
<td>0.97</td>
<td>0.06</td>
<td>11.30</td>
<td>0.90</td>
<td>0.50</td>
</tr>
<tr>
<td>After adsorption</td>
<td>1.45</td>
<td>18.62</td>
<td>13.54</td>
<td>25.43</td>
<td>7.21</td>
<td>1.42</td>
<td>0.065</td>
<td>10.05</td>
<td>0.76</td>
<td>0.53</td>
</tr>
</tbody>
</table>

3.4.2 Surface topography analysis

As shown in Figure 16, there are small pores on the surface of the RM, and it is observed that powdery substance is filled around the pores, and the specific surface area is relatively small (Figure 16(a)). While for ARMPA, the surface is rough, the pores are evenly distributed, and the specific surface area is relatively large (Figure 16(b)). It can be seen from the Figure 16(c) and (d), that before adsorption ARMPA has an irregular surface with loose structure and large inter-particle gaps. and it consists of the gap between the aggregate, the agglomerate and the aggregate. The surface material mainly consists of voids between aggregates, agglomerates and aggregates. During the comprehensive modification of ARMPA particle adsorbent, the surface is eroded, which makes ARMPA display a rougher surface and richer pores. In addition, the loose porosity of fly ash makes the surface pores of the particles more developed, improving the adsorption capacity of the ARMPA. After adsorption, there are fewer pores on the surface of ARMPA, and the bulk and flocculent substances increase.
This is because ARMPA is soaked and eroded by PW, and hydrolysis occurs, causing the material around the pore to collapse and deposit and form a lump. In addition, Ca2+/Na+/Al3+/Mg2+ adsorbs P and phosphate and adheres to the surface of the pores, causing the surface of the adsorbent to be massive and flocculated.

Figure 16. Surface morphology of ARMPA and RM under SEM (a) Particle surface of RM; (b) Particle surface of ARMPA; (c) ARMPA before adsorption; (d) ARMPA after adsorption.

3.4.3 FT-IR analysis

The FT-IR spectrum of ARMPA before and after adsorption is shown in Figure 17. The peak at a frequency of 1114 cm\(^{-1}\) is a Si-O-Si group, the peak at 1456 cm\(^{-1}\) is a carboxylate COO\(-\) group and a CO3\(^2-\) group, and the peak at 995 cm\(^{-1}\) is the peak spectrum of the POP group. The peak at a frequency of 3600-3000 cm\(^{-1}\) is crystal water, and most of the substances do not change before and after adsorption. The strong peak at 1156 cm\(^{-1}\) after adsorption is PO4\(^3-\) group, which indicates that the adsorbent has loaded P in PW.

Figure 17. Infrared spectrum before and after ARMPA adsorption

3.4.4 XPS Analysis

According to the results of XRF, EDS, FT-IR, etc., ARMPA has successfully adsorbed P from PW. In order to determine the
composition of the adsorbed material and the form of the adsorbed P, the sample was subjected to XPS analysis. The XPS spectrum of the sample after adsorption is shown in Figure 18. As can be seen from the figure, the surface of the sample mainly includes Fe 2p, Al 2p, Ca 2p, Ti 2p, Si 2p, K 2p, Na 1s, Mg 1s and the like. Notably, there is no P2p spectrum on the surface of the ARMPA sample before adsorption (Figure 18(a)), but the surface of the ARMPA sample after adsorption has a P2p spectrum at 133.7 eV (Figure 18(b)), indicating that the phosphorus has been successfully adsorbed on the surface of the ARMPA, which were consistent with the results of XRF.

The XPS analysis of the ARMPA particle adsorbent after adsorption was carried out, and the results are shown in Figure 19, and the peaking results are shown in Table 5. Before adsorption (Figure 19(c)), the characteristic spectrum of P 2p on the surface of ARMPA is not obvious. Combined with XRD data, the binding energy has a little amount of P; while after adsorption (Figure 19(d)), the binding energy of P 2p exist in three forms. The binding energy has obvious characteristic peaks at 133.2eV, and its existence form is (PO4)2.3H2O, Na2HPO4, the corresponding ratio is 38.51%; the binding energy has obvious characteristic peak at 134.2eV, and its existence form is (Al2O3)10(MgO)10(Na2O)10(P2O5)70, the corresponding ratio is 48.34%; the binding energy has a weak characteristic peak at 133.2 mV, and its existence form is CaHPO4, and the corresponding ratio is 13.15%. It is indicated that in the adsorption system, when the pH is 8-9, P mainly exists in the form of HPO42- and PO43-, which is consistent with the existence state of phosphate in the aqueous solution reported in the literature. These ions and positively charged Ca2+/Na+/Al3+/Mg2+ form strong chemical bonds by precipitation, surface deposition and ion exchange coordination, and are distributed on the inner surface of the ARMPA tunnel.

![Figure 18. XPS photoelectron spectroscopy before and after adsorption](image-url)
Figure 19. XPS photoelectron spectroscopy of P 2p before and after adsorption

Table 5. XPS of P2p peak results after adsorption

<table>
<thead>
<tr>
<th>Name</th>
<th>BE/eV</th>
<th>FWHM/eV</th>
<th>Area/CPS.eV</th>
<th>Combined state</th>
<th>ratio/%</th>
</tr>
</thead>
<tbody>
<tr>
<td>P 2p</td>
<td>133.2</td>
<td>1.02</td>
<td>852.89</td>
<td>(PO4)2·3H2O/Na2HPO4</td>
<td>38.51</td>
</tr>
<tr>
<td>P 2p</td>
<td>133.6</td>
<td>0.60</td>
<td>291.20</td>
<td>CaHPO4</td>
<td>13.15</td>
</tr>
<tr>
<td>P 2p</td>
<td>134.2</td>
<td>1.02</td>
<td>1070.49</td>
<td>(Al2O3)10(MgO)10(Na2O)10(P2O5)70</td>
<td>48.34</td>
</tr>
</tbody>
</table>

3.4.5 Adsorption kinetic analysis and thermodynamic analysis

It can be seen from the comparison of R2 value in Table 6 and Table 7 that the results of data fitting using pseudo first-order dynamic model are better than pseudo second-order dynamic model, so pseudo first-order dynamic model can better describe the adsorption process for phosphorus by RM and ARMPA.

Table 6. Pseudo-first and pseudo second-order kinetic parameters of P adsorption by RM and ARMPA

<table>
<thead>
<tr>
<th>Samples</th>
<th>Pseudo-first</th>
<th>Pseudo second-order</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>k1/ min⁻¹</td>
<td>R²</td>
</tr>
<tr>
<td>RM</td>
<td>0.25</td>
<td>0.96</td>
</tr>
<tr>
<td>ARMPA</td>
<td>0.31</td>
<td>0.99</td>
</tr>
</tbody>
</table>

Table 7. Comparison of experimentally determined Qe with fitting Qe values through pseudo first-order, pseudo second-order kinetic models

<table>
<thead>
<tr>
<th>Samples</th>
<th>Qe/mg·g⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Experiment</td>
</tr>
<tr>
<td></td>
<td>Pseudo-first-order</td>
</tr>
<tr>
<td>RM</td>
<td>7.65</td>
</tr>
<tr>
<td>ARMPA</td>
<td>13.83</td>
</tr>
</tbody>
</table>
### Table 8. Freundlich, Langmuir and DR adsorption parameters of RM and ARMPA

<table>
<thead>
<tr>
<th>Samples</th>
<th>Freundlich</th>
<th>Langmuir</th>
<th>DR</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$R^2$</td>
<td>$1/n$</td>
<td>$K_f$</td>
</tr>
<tr>
<td>RM</td>
<td>0.87</td>
<td>0.23</td>
<td>3.09</td>
</tr>
<tr>
<td>ARMPA</td>
<td>0.92</td>
<td>0.59</td>
<td>3.04</td>
</tr>
</tbody>
</table>

Comparing the $R^2$ values in Table 8, it is obvious to see that the adsorption process of P by RM and ARMPA is more consistent with the Langmuir model, indicating that the main adsorption forms of the first two adsorbents in the adsorption process are single-layer adsorption.

The results of adsorption microcoria test by RM and ARMPA was shown in Figure 20. RM and ARMPA adsorptions are both exothermic reactions, and the ARMPA micro-heat value is -5.022 J/g, which is greater than RM, indicating the adsorption effect of the adsorbent after comprehensive modification was better than before modification.

#### 3.4.6 Safety analysis of adsorbents for water treatment

In order to explore the safety of the use of solid adsorbent particles, the toxicity analysis was carried out according to the Chinese National Standard HJ 557-2010 solid waste leaching toxicity method horizontal oscillation method, and the measurement results are shown in Table 9. Compared with the heavy metal leaching results of RM, the leaching concentrations of Pb, Cr and Zn of RMFA and ARMPA are significantly reduced, indicating that C2S has a certain curing ability for the above-mentioned pollutants in the raw materials during the preparation of the adsorbent. At the same time, the heavy metal leaching rate of RMFA and ARMPA meets the Chinese National Standard GB5085.3-2007 "Dangerous waste identification standard leaching toxicity identification" for the limits of Pb, Cd, Cr, Hg, Zn, and the safety as a water treatment material basically meet the requirement.

### Table 9. Leaching concentration of heavy metal by RM, RMFA and ARMPA (mg·L$^{-1}$)

<table>
<thead>
<tr>
<th>Heavy metal</th>
<th>RM</th>
<th>ARMPA</th>
<th>National standard Toxicity limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>0.560</td>
<td>0.293</td>
<td>5</td>
</tr>
<tr>
<td>Cd</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>Cr</td>
<td>1.251</td>
<td>0.701</td>
<td>15</td>
</tr>
<tr>
<td>Hg</td>
<td>0</td>
<td>0</td>
<td>0.1</td>
</tr>
<tr>
<td>Zn</td>
<td>0.146</td>
<td>0.004</td>
<td>100</td>
</tr>
</tbody>
</table>
4 CONCLUSION

When RM: FA: Water = 19:1:7, and with addition of 0.15% HPMC, 4% C2S, 1% H2O2 and 0.2% MnO2, the non-thermally activated red mud particle adsorbent ARMPA is characterized by a pH of 9.25, a specific surface area of 40.54 m2/g, a hole volume of 2.10 cm3/g, a compressive strength of 1.31 KPA, and a soaking powdering rate of 3.72% at 24h. When the initial total P concentration was 156.7 mg/L, the amount of ARMPA was 4 g/L, and the adsorption time was 10 h, the TP adsorption capacity could reach 38.46 mg/g, and the removal efficiency of TP reached can be as high as 98.17%. The preparation process of the particle adsorbent by the non-thermally method has the advantages of not only environmental-friendly, but also of low cost due to the low energy consumption. Furthermore, the double-layer structure has significant beneficial effects on the enhancement of adsorption capacity and the strength.

The pseudo first-order kinetic model can better describe the adsorption process of phosphorus by RM and ARMPA, and the adsorption process of RM and ARMPA is more consistent with the Langmuir model, which is monolayer adsorption.

In the adsorption system, when the pH is 8-9, P mainly exists in the form of HPO42- and PO43-, which is consistent with the existence state of phosphate in the aqueous solution reported in the literature. These ions and positively charged Ca2+/Na+/Al3+/Mg2+ form strong chemical bonds by precipitation, surface deposition and ion exchange coordination, and are distributed on the inner surface of the ARMPA tunnel.

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REFERENCES


