

Article

From Waste to Resource: A Circular Economy Approach for Landfill Leachate Treatment Using Coal Gangue-Based Coagulant and Beneficial Reuse of the Generated Sludge

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Abstract

This study explored the utilization of coal gangue with high iron content to synthesize polyferric-alum-sulfate (PAFS) and coal gangue leaching residue-PAFS (CG@PAFS) coagulants, which were subsequently applied to treat landfill leachate and its concentrate. The results demonstrated that the PAFS coagulant exhibited notable removal efficiency for key pollutants, including chemical oxygen demand (COD) and total phosphorus (TP), with removal rates of 23.8% and 77.5%, respectively. Its performance was comparable to polyferric sulfate (PFS) and superior to aluminum polysulfate (PAS). Mechanistic investigations revealed that Fe^{3+} in the PAFS coagulant reacted chemically with phosphate ions, forming insoluble precipitates and facilitating phosphate removal through flocculation. Furthermore, PAFS also exhibited moderate removal efficiency for COD and ammonia nitrogen ($\text{NH}_3\text{-N}$) via flocculation processes. From a sustainability perspective, this study advances a circular economy model by simultaneously addressing two pressing environmental challenges: the valorization of coal gangue (a massive industrial solid waste) and the treatment of landfill leachate (a hazardous wastewater). Importantly, the post-treatment sludge, rich in available phosphorus and silicon, demonstrates promising potential for beneficial reuse as a soil conditioner, thereby closing the material loop and reducing reliance on virgin chemical resources. This integrated waste-to-resource approach aligns with multiple Sustainable Development Goals (SDGs), particularly SDG 6 (Clean Water and Sanitation), SDG 11 (Sustainable Cities and Communities), and SDG 12 (Responsible Consumption and Production). Overall, this study provides valuable insights into the resource-efficient utilization of coal gangue and presents an effective, low-carbon, and sustainable approach for landfill leachate management.



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Keywords: PAFS; coal gangue; COD; landfill leachate; coagulant

1. Introduction

With the rapid development of the economy, the issue of industrial solid waste and municipal domestic waste treatment has become increasingly prominent. Coal gangue, as a major type of solid waste generated during coal mining and processing, has a huge annual output [1–3]. Globally, coal gangue accounts for approximately 10–15% of total

coal production. In China, the world's largest coal producer, coal gangue accumulation exceeds 7 billion tons, with an annual output surpassing 700 million tons [4–7]. Due to the limited treatment and utilization methods for coal gangue, a large amount of it is piled near mining areas, forming numerous gangue hills [8]. These coal gangue hills not only occupy a large amount of land but may also trigger geological disasters such as landslides and debris flows [9,10]. From a sustainability standpoint, the accumulation of coal gangue represents a significant waste of land resources and a potential source of environmental pollution, underscoring the urgent need for innovative and sustainable valorization strategies. Presently, there have been notable advancements in the research and application of coal gangue for resource recovery [2,11–18]. Nevertheless, these applications, while they do consume a significant amount of coal gangue, are not without their challenges, including lower utilization rates and products with minimal added value. Within the framework of sustainability and circular economy, an ideal utilization strategy should not only consume the waste but also convert it into high-value products capable of addressing other environmental problems, thereby creating a synergistic nexus of waste treatment, resource recovery, and environmental protection.

In recent years, the application of coagulants in the field of water and wastewater treatment has become increasingly widespread, as they can effectively remove suspended solids, colloids, and some dissolved pollutants from water [19]. However, coagulant preparations such as aluminum polysulfate (PAS) and polyferric sulfate (PFS) have issues like high costs and potential environmental impact [20]. Consequently, the development of a cost-effective and environmentally friendly coagulant has become a hot topic of current research. Compared to traditional inorganic coagulants, composite coagulants perform better, and compared to organic coagulants, they are more cost-effective, hence attracting increasing attention [21]. Aluminum-iron coagulants possess the advantages of both aluminum and iron polymers and overcome the defects of traditional coagulants [22]. Therefore, research has been conducted on aluminum-iron coagulants, including poly-alum-ferric-sulfate (PAFS) [23–25]. The raw materials for preparing PAFS coagulants include inorganic salts [25–27], ores [22], and waste materials [28,29]. PAFS coagulants were prepared using fly ash through the acid leaching method and have shown significant effects in the treatment of municipal wastewater [22].

Chen et al. [30] efficiently extracted aluminum from low-grade kaolin and prepared poly-aluminum-ferric sulfate (PAFS) coagulants. They used sulfuric acid to assist in the activation of low-grade kaolin and then conducted water leaching at room temperature. By optimizing the experimental parameters, they achieved high extraction rates of aluminum and iron (98.8% and 98.1%, respectively). During the acid leaching process of coal gangue, cations such as Al^{3+} and Fe^{3+} , as well as anions like Cl^- and SO_4^{2-} , can coexist in the leachate, indicating that coagulants can be prepared from coal gangue leachate. Gao et al. [22] prepared a composite hydrochloric acid-sulfuric acid leaching process to prepare poly-aluminum-ferric chlorosulfate (PAFCS) coagulants using fly ash, and applied it to the treatment of coking wastewater. Yang et al. [31] used acid pickling waste liquor from steel plants to prepare a composite inorganic coagulant PFASC and applied it to the treatment of paper mill wastewater. In their study, they added ferrous sulfate, aluminum sulfate, and sulfuric acid to the acid-washing waste solution, followed by the addition of sodium chlorate to induce an oxidation reaction, ultimately forming polyaluminum chloride sulfate (PFASC). These studies primarily focused on the impact of leaching temperature and acid concentration on leaching efficiency and the treatment effect of PFASC coagulant on printing and dyeing wastewater and coking wastewater. However, most of these studies employed mixed acids for leaching. From the perspective of mechanism exploration, research on the mechanism of single-acid leaching can more clearly reveal the interaction

mechanism between the acid and the target metals or minerals, which helps us understand the chemical reactions and physical changes during the leaching process. Moreover, these studies did not address the treatment scale, operational costs, and other information on PFAS coagulant in wastewater treatment applications, nor did they compare it with other coagulants to further demonstrate its economy and feasibility. Critically, the absence of comprehensive techno-economic and life-cycle assessments in previous studies represents a significant gap in evaluating the true sustainability of these proposed solutions—a gap that the present work aims to address.

Landfill leachate typically originates from landfill sites or waste piles, which are often located near residential areas or water sources [32]. If not treated promptly, the harmful substances in the leachate can quickly infiltrate groundwater, posing a direct threat to the health of nearby residents [33]. While printing and dyeing wastewater and coking wastewater are harmful, their impact is usually more localized [34–36]. Moreover, the composition of printing and dyeing wastewater and coking wastewater is relatively simple, and the treatment technology is relatively mature. The specificity of landfill leachate treatment is reflected in the growth potential of its market size, the complexity of technical requirements, and the profound impact of policies and society [37,38]. Therefore, focusing on the treatment of landfill leachate can help meet this broad market demand.

On the other hand, the sludge produced during the wastewater coagulation process, as a major byproduct, contains a large amount of organic matter and nutrients, and has certain value for resource utilization [33,36]. However, the treatment and disposal of sludge have always been a challenge for environmental protection [39]. Using sludge as a raw material for soil conditioners not only realizes the resource utilization of sludge but also improves soil structure and fertility, enhancing the ecological environmental quality of the soil.

The purpose of this study is to demonstrate that the specific mineralogical structure of coal gangue allows for the extraction of Al and Fe to form a composite coagulant (PAFS), and the residual leaching residue can act synergistically with the flocs to enhance pollutant removal while simultaneously producing a sludge suitable for soil amelioration. The overarching goal is to demonstrate a closed-loop, sustainable process that addresses multiple environmental challenges in an integrated, cost-effective, and low-carbon manner. The novelty of this work lies in: (i) utilizing a single sulfuric acid leaching system to simplify the preparation mechanism compared to mixed-acid systems reported previously; (ii) proposing a dual-benefit strategy where the leachate is used for coagulation and the generated sludge is evaluated for beneficial reuse as a silicon-phosphorus fertilizer, thereby providing a holistic waste-to-resource solution that contributes directly to the United Nations Sustainable Development Goals (SDGs), particularly SDG 6 (Clean Water and Sanitation), SDG 9 (Industry, Innovation and Infrastructure), SDG 11 (Sustainable Cities and Communities), and SDG 12 (Responsible Consumption and Production).

2. Materials and Methods

2.1. Materials

The coal gangue used in this study was provided by Jixi, Heilongjiang province, China. The chemical (elemental) composition of the coal gangue was determined by X-ray fluorescence spectroscopy (XRF) (S8 TIGER, Bruker, Bremen, Germany), while its mineralogical composition and phase identification were characterized by X-ray diffraction (XRD) (S8 TIGER, Bruker, Germany). In the experiment, the landfill leachate and its concentrate, used as actual wastewater, were collected from the Fengshun Landfill in Shangrao, Jiangxi, China. The landfill has been in operation for approximately 8 years (middle-aged stage), primarily receiving municipal solid waste (MSW) with a small proportion of stabilized

industrial waste. The waste composition is typical of southern Chinese cities, dominated by kitchen waste, plastics, and paper. The following items were purchased from Xilong Scientific Co., Ltd. (Guangzhou, China): 98 wt% concentrated sulfuric acid, sodium silicate, sodium hydroxide, aluminum polysulfate (PAS), and polyferric sulfate (PFS). All chemical reagents were of analytical grade, and all solutions were prepared using deionized water.

2.2. Preparation of PAFS and CG@PAFS

The schematic diagram of the experiment, including the preparation of PAFS and CG@PAFS and their application in landfill leachate, is shown in Figure 1. The preparation of PAFS and CG@PAFS from coal gangue involved three steps: acid leaching with sulfuric acid solution under atmospheric pressure, filtration, and polymerization.

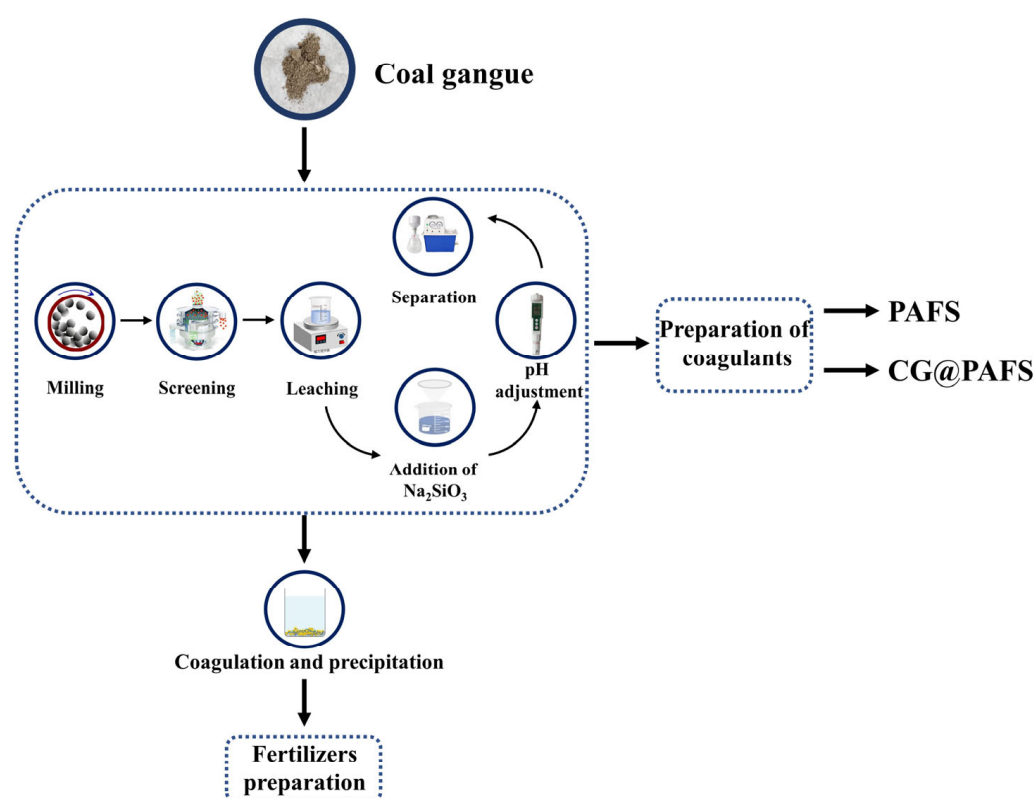


Figure 1. PAFS and CG@PAFS preparation process.

The acid leaching process was conducted in a beaker equipped with a magnetic stirrer. Add 100 mL of sulfuric acid solution and 25 g of coal gangue to the beaker, and then stir the mixed solution at 25 °C for 30 min. To characterize and analyze the leaching efficiency and metal content, the leachate was diluted (and mixed with the sulfuric acid solution). Sodium hydroxide and sodium silicate were added, and the leachate residue was collected after drying at 75 °C for the following experiments.

2.3. Coagulation Experiments

The treatment process of landfill leachate (Figure 2) was a multi-stage and multi-technology integrated process. The full-scale A/O + MBR process operates with an anaerobic HRT of 5 days and an aerobic HRT of 10 days, with a mixed liquor suspended solids (MLSS) concentration of approximately 8000~10,000 mg/L. In this stage, coagulants were added to the landfill leachate. The anaerobic-oxic process (A/O) consisted of anaerobic and aerobic stages [40]. In the anaerobic stage, organic matter in the wastewater was converted into biogas (mainly composed of methane and carbon dioxide) under the action

of anaerobic microorganisms. Subsequently, the wastewater entered the aerobic stage, where aerobic microorganisms further degraded the remaining organic matter. In the membrane bioreactor system (MBR), the activated sludge process or biofilm method was used for biodegradation, while the membrane module was used for solid–liquid separation, replacing the traditional sedimentation tank [41]. After the MBR, coagulation treatment was carried out again to further remove suspended solids and some dissolved organic matter. During the nanofiltration process (NF), the landfill leachate was passed through the nanofiltration membrane under pressure, and most divalent and multivalent ions, as well as some organic matter, were retained [42]. Under reverse osmosis (RO) and high-pressure reverse osmosis (HPRO), the landfill leachate was passed through a semi-permeable membrane, with water molecules passing through the membrane while most dissolved solids were retained, resulting in purer water [43,44]. The MVR system increased the pressure and temperature of the steam through a compressor and then reinjected the steam into the evaporator, improving thermal efficiency and reducing energy consumption [45]. In this study, the prepared coagulants were applied specifically to the raw leachate and the concentrated brine to evaluate their potential for pretreatment and deep treatment, respectively (Tables 1 and 2). Biological effluent was not a direct subject.

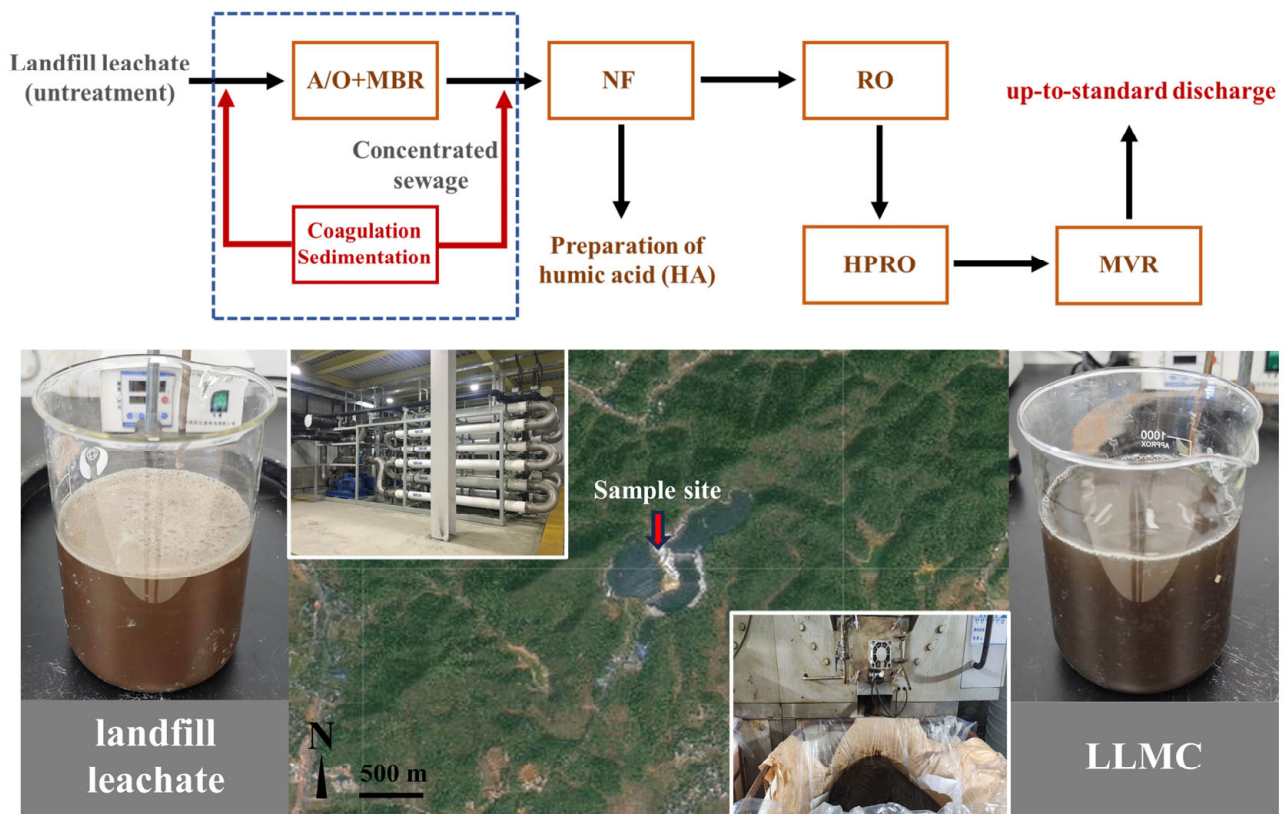


Figure 2. Source and sampling of landfill leachate and its concentrate.

Table 1. Indicators of pollutants in landfill leachate.

COD	NH ₃ -N	TN	TP	Colority	pH	ss	Ca
2393 ± 46	416 ± 12	695 ± 45	27.1 ± 2.4	3500 ± 200	6.4~7	37 ± 3	79 ± 6.3
Hg	Cr	Cd	As	Pb	Cu	Ba	Mg
<0.03	<0.004	<0.02	<0.004	1.26 ± 0.05	<0.006	0.07 ± 0.02	132 ± 12.7

Organic pollutants and heavy metals in mg/L.

Table 2. Indicators of pollutants in landfill leachate concentrate.

COD	NH ₃ -N	TN	TP	Colority	pH	ss	Ca
16,587 ± 352	2218 ± 87	4215 ± 263	75.3 ± 11	6800 ± 300	7.6~8.8	258 ± 42	425 ± 35
Hg	Cr	Cd	As	Pb	Cu	Ba	Mg
<0.03	<0.004	<0.02	<0.004	2.65 ± 0.1	<0.006	0.28 ± 0.05	584 ± 52

Organic pollutants and heavy metals in mg/L.

The coagulation experiment was also conducted using a magnetic stirrer. In total, 100 mL of landfill leachate and concentrated leachate were placed in a beaker, and different coagulants were added, followed by stirring. The stirring process was divided into the following three steps: (1) rapid stirring at 200 rpm for 2 min; (2) slow stirring at 80 rpm for 8 min; (3) stop stirring and allow for sedimentation for 10 min. Supernatant samples were extracted from the beaker at a depth of 2 cm below the water surface and were measured for turbidity, COD, SS, pH, ammonia nitrogen, total nitrogen, and total phosphorus using a turbidimeter (WGZ-200 Ratio Turbidimeter, Shanghai Instrument Physical Optics Instrument Co., Ltd. (Shanghai, China)) and an ultraviolet-visible spectrophotometer (Shimadzu UV1780, Shimadzu, Kyoto, Japan). The leachable COD, NH₃-N, and TN in the sludge generated from the treatment of landfill leachate were quantified using the horizontal oscillation method (HJ 557-2010) [46]. The available silicon and phosphorus were determined using buffered citric acid (DB63/T 1823-2020) [47] and sodium hydrogen carbonate solution (HJ 704-2014) [48]. Each experiment was repeated three times, and the results were the average of three parallel experiments.

3. Results and Discussion

3.1. Leaching of Coal Gangue with Sulfuric Acid

To illustrate the leaching behavior of metals from coal gangue during the sulfuric acid solution leaching process, the effect of sulfuric acid at a fixed concentration on the metal leaching efficiency was studied (120 min, liquid-to-solid ratio: 4 mL/g), as shown in Figure 3. The mineral phases and elemental content of the coal gangue were shown in Figure 3a,b. It can be observed that aluminum and iron had higher leaching concentrations (7.97 g/L and 11.8 g/L) and leaching rates (28.4% and 48.6%) (Figure 3c). In contrast, the leaching concentrations of silicon, magnesium, and calcium (0.52 g/L, 0.22 g/L, and 0.074 g/L) (Figure 3d) and their leaching rates were lower (0.8%, 12.4%, and 6.58%). This indicated that 10% sulfuric acid can significantly promote the release of iron and aluminum from the coal gangue, which may be due to the acceleration of molecular motion and the increase in activity of the acid leaching process. Silicon primarily existed in the form of silicate, and the silicic acid generated by its reaction with sulfuric acid had a low solubility in water, resulting in lower leaching concentrations and rates of silicon. Although magnesium and calcium can react with sulfuric acid to form soluble sulfates, their forms of existence in coal gangue (SiO₂ PDF#46-1045 and CaPO₃(OH)·2H₂O PDF#11-0293) may not be conducive to dissolution by sulfuric acid, thus reducing the leaching rates [49,50].

3.2. Preparation of Flocculants

The polymerization mechanism of the leachate solution was investigated (Figure 4), mainly including the hydrolysis and polymerization of Al³⁺ and Fe³⁺. The results indicated that the addition of SiO₃^{2−} had almost no effect on the production of PAFS when the concentration exceeded 0.5 g/L (Figure 4a). Therefore, the amount of Na₂SiO₃ was set to a fixed value. Changes in pH may affect the polymerization reaction and the structure of the flocculant. To study the influence of pH on the production and performance of PAFS

floculants, PAFS floculants were synthesized at different pH levels from the sulfuric acid leachate solution. Figure 4b shows the impact of different pH levels on the production of PAFS floculants. The initial pH of the system was 0.12 (without added sodium hydroxide). When 2 g of sodium hydroxide was added, the pH reached 1.76. The production of PAFS floculant was 6.8 g, with a yield of 27.2%. At the same time, the concentrations of aluminum and iron in the solution decreased by 50.6% and 88.75%, respectively (Figure 4c). When the addition of sodium hydroxide reached 2 g, the pH was 1.76. When the addition of sodium hydroxide reached 3.5 g, the pH was 2.97. The production of PAFS floculant was 10.6 g, with a yield of 42.4%. The concentrations of aluminum and iron in the solution decreased by 83.9% and 100%, respectively. As the pH increased to 2.97, the concentration of H^+ in the solution decreased, and hydroxyl compounds were generated through hydrolysis reactions, which further polymerized to form macromolecular floculants. Therefore, at a pH of 2.97, the yield of PAFS floculant increased to 42.4%, indicating a significant improvement in the synthesis efficiency of the floculant. However, floculants synthesized at different pH levels may exhibit different flocculation effects (Figure 4d,e). To confirm this, PAFS prepared at different pH levels was used to treat concentrated landfill leachate, and the flocculation effects were shown. These results showed that the pH during the preparation process did not significantly affect the actual performance of PAFS. This indicated that the structure of the PAFS floculant was relatively stable and not easily influenced by changes in pH. Even at a broad range of pH levels, its basic structure remained intact, thus maintaining its flocculation performance. The yield of PAFS floculant at pH 2.97 (42.4%) was significantly higher than at pH 1.15 (16.8%) and pH 0.12 (15.2%). Meanwhile, at this pH, most of the iron and aluminum were converted into precipitates. Therefore, pH 2.97 was more favorable for the preparation of PAFS.

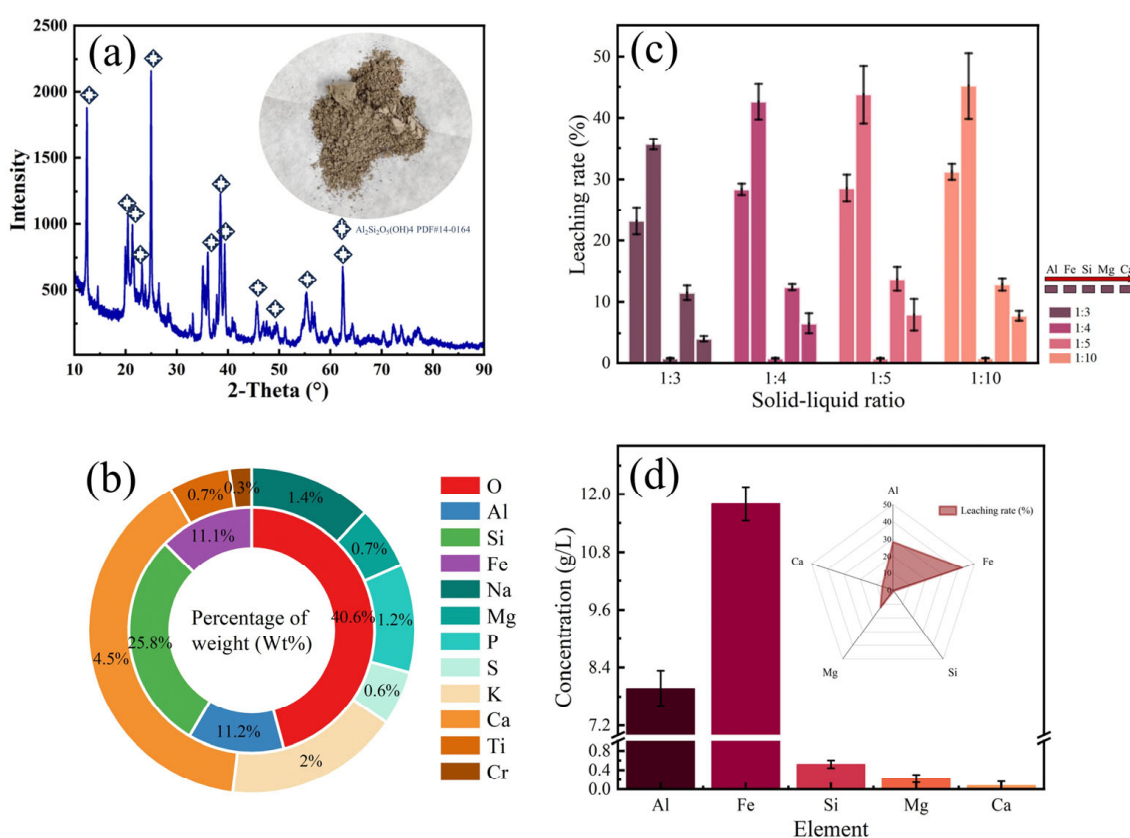


Figure 3. (a) XRD results of untreated gangue; (b) elements and contents of untreated gangue; (c) sulfuric acid leaching rate under different solid-liquid ratios; (d) metal leaching concentration and leaching rate when the solid-liquid ratio is 1:4.

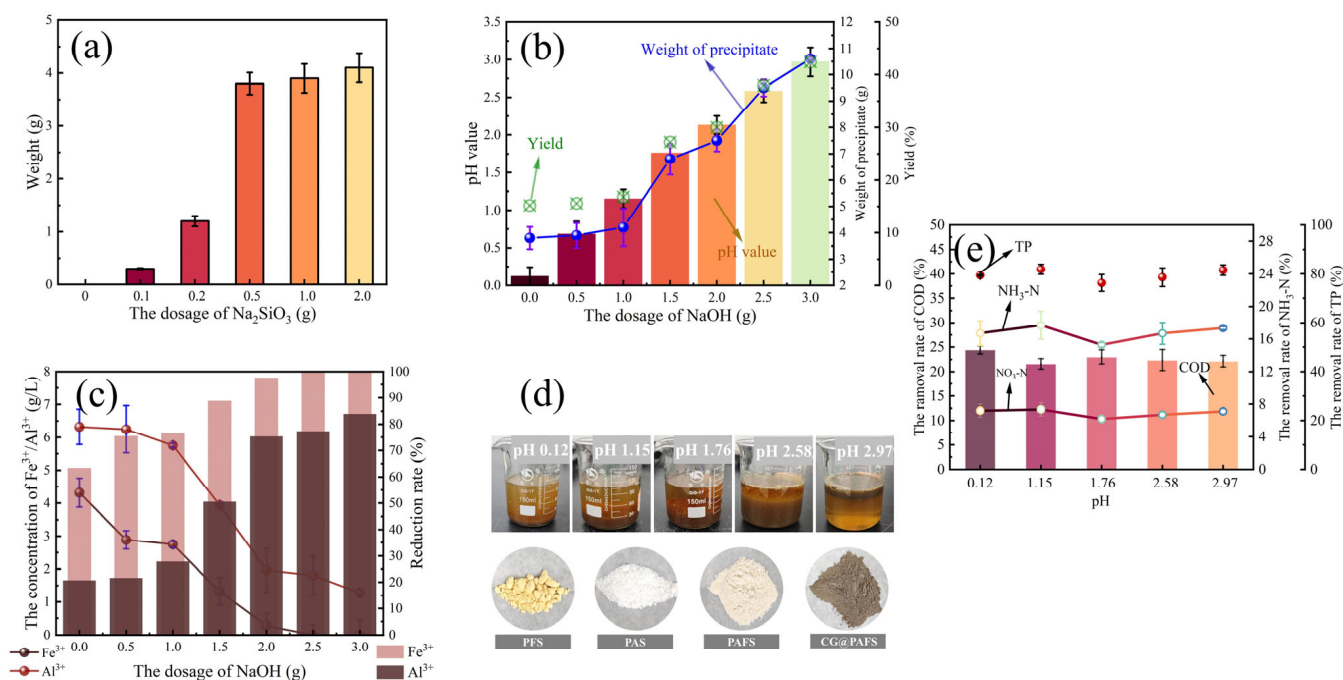


Figure 4. Yield of PAFS under different preparation conditions: (a) the effect of sodium silicate addition on the yield of PAFS; (b) the effect of hydroxide addition on the yield of PAFS; (c) the effect of sodium hydroxide addition on the concentrations of iron and aluminum; (d) images of landfill leachate treatment with PAFS obtained under different sodium hydroxide addition (pH changes during preparation) and comparison of PAFS with other coagulants (pH during preparation 2.97); (e) the treatment effect of PAFS obtained under different sodium hydroxide addition (pH during preparation) on landfill leachate.

3.3. Characterization of Materials

To analyze the morphological and compositional changes during the sulfuric acid leaching and PAFS preparation process, Figure 5 provides the SEM-EDS (Figure 5a,d), XRD (Figure 5b,c), and FTIR (Figure 5e) of the leached coal gangue and the prepared PAFS. After the sulfuric acid leaching process, the diffraction peaks of kaolinite essentially disappeared, indicating that the structure of coal gangue can be destroyed. Following polymerization, amorphous ferrisilicate protrusions were observed, suggesting the formation of new iron oxides. Meanwhile, the leachate residue contained only quartz, indicating that most of the silicon in the coal gangue was retained.

After leaching with 10% sulfuric acid, diffraction peaks of kaolinite disappeared, and new diffraction peaks appeared at 10.6° , 25.7° , 29° , 32.4° , 25.1° , 32.9° , etc. These peaks corresponded well to the (110), (131), (310), (330) of Grunerite and the (111), (-201) of Cronstedtite. The diffraction peaks were sharp and intense, which was consistent with their highly crystalline. After polymerization of the leachate, the diffraction pattern of the residue showed amorphous characteristics. PAFS exhibited broad and scattered peaks, indicating that Al^{3+} , Fe^{3+} , and SO_4^{2-} had polymerized into new compounds, resulting in the absence of crystal diffraction signals for $\text{Fe}_2(\text{SO}_4)_3$, $\text{Al}_2(\text{SO}_4)_3$, $\text{Al}(\text{OH})_3$, and $\text{Fe}(\text{OH})_3$. EDS analysis indicated that the prepared PAFS was mainly composed of iron, aluminum, and sulfur. There were two broad peaks near 697 cm^{-1} and 1381 cm^{-1} , corresponding to the stretching vibration of Fe-O [31,51]. A broad and intense peak was displayed at 1206 cm^{-1} , attributed to the stretching vibration of Fe-O-Fe or Al-O-Al [52]. The peak at 778 cm^{-1} corresponded to the asymmetric stretching vibration of Si-O [51]; the peak at 856 cm^{-1} was attributed to the bending vibration of Si-O-Si [53]. Overall, all the results indicated the formation of a new chemical substance PAFS, composed of aluminum and iron.

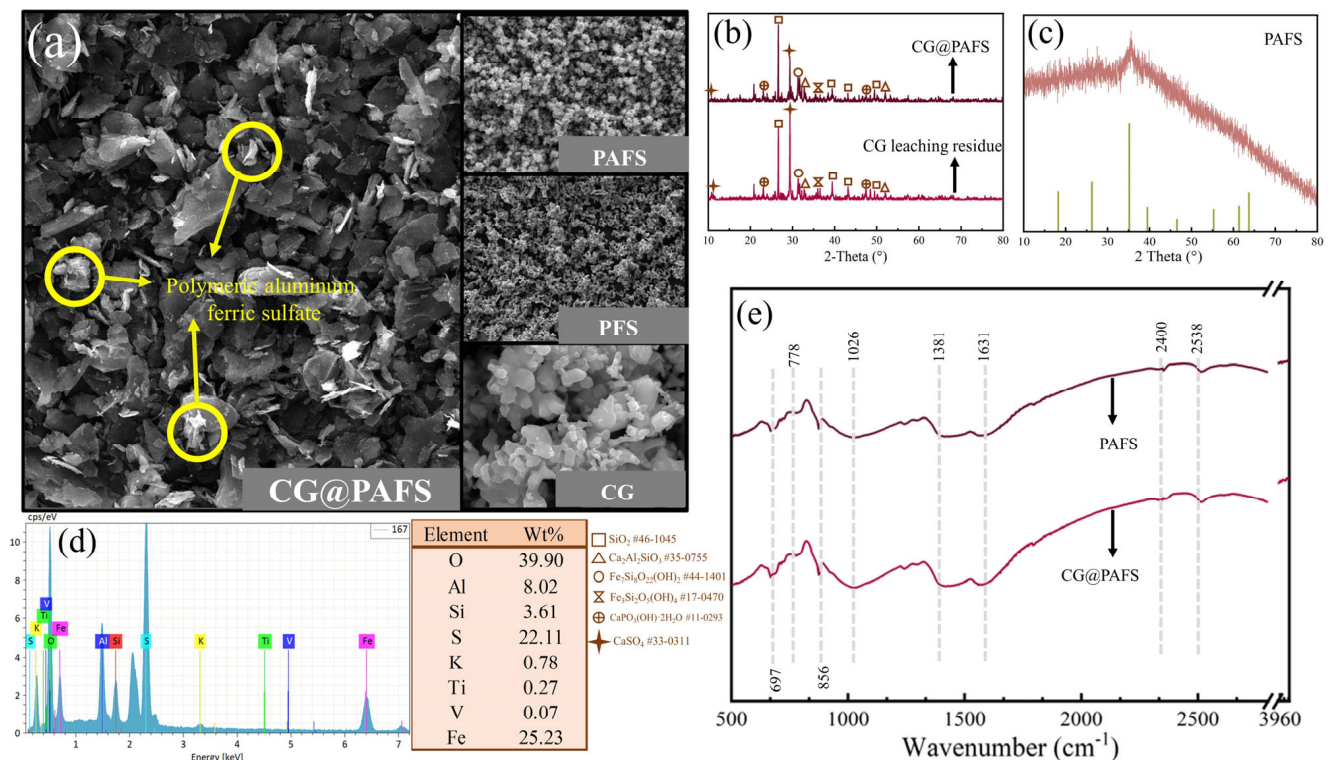


Figure 5. (a) SEM images of CG@PAFS, PAFS, PFS, and CG (Accelerating voltage: 15 kV, working distance: ~10 mm, vacuum mode); (b) XRD patterns of leached CG and CG@PAFS; (c) XRD patterns of PAFS; (d) EDS results of PAFS; and (e) IR spectral results of CG@PAFS and PAFS.

The results showed the SEM of the prepared PAFS, CG@PAFS, and the leachate residue. The SEM of PFS and PAS at a high magnification was shown. After sulfuric acid leaching, the coal gangue was mainly composed of large chunks and small fragments; the surface of the large chunks was smooth and pore-free, with many small fragments adhering to the surface. PAFS had a smooth but uneven surface, with some small fragments tightly adhered together, resembling many particles being encapsulated. After polymerization but without filtration, CG@PAFS was mainly composed of aggregates of small fragments and fine particles, with a large number of cracks about 10 nm in size. This indicated that the lamellar structure of the coal gangue was destroyed during the acid leaching process, with iron-containing polymers encapsulated on the surface [54,55].

3.4. Effect of PAFS and CG@PAFS on the Treatment of Landfill Leachate and Concentrate

3.4.1. Effect of Treatment of Landfill Leachate

We tested the treatment effects of PAFS and CG@PAFS on landfill leachate (Figure 6a,b) and concentrated leachate (Figure 6c,d), and compared them with PAS and PFS. We used CG, PAS, PFS, PAFS, and CG@PAFS as coagulants, with an addition of 2%. The coagulation effects were shown in Figure 6. PAFS had a COD removal rate of 23.8%, which was close to the COD removal rates of PAS and PFS (17.5% and 20.3%). The CG@PAFS prepared without filtration had a COD removal rate of 12.9%. This indicated that the prepared PAFS and CG@PAFS had a certain removal effect on COD in landfill leachate. For NH₃-N and NO₃-N, PAFS had a removal rate of 16.1% for NH₃-N and 5.6% for NO₃⁻-N. CG@PAFS had a removal rate of 5.9% for NH₃-N and 3.4% for NO₃⁻-N. For total phosphorus removal, PAFS had a removal rate of 77.5% for total phosphorus, while CG@PAFS had a removal rate of 60%. Overall, PAFS had a similar effect to PFS and was better than PAS on landfill leachate. However, the effect of CG@PAFS was significantly lower than PAS, PFS, and PAFS. This also confirmed that untreated coal gangue (CG) had no significant coagulation

effect on landfill leachate. According to the research of Gao et al. [22], the polymerization rate was different due to the difference in the radius of Al^{3+} and Fe^{3+} and their ability to bind with $-\text{OH}$ hydroxyl groups. Therefore, an appropriate $\text{Al}^{3+}/\text{Fe}^{3+}$ molar ratio may significantly improve the flocculation performance of the flocculant. The appropriate Al/Fe (1:2) ratio made it have a three-dimensional chain network structure, with higher strength and recovery factors of PAFCS flocs, indicating a more compact floc structure, stronger shear resistance, and easier re-aggregation after fragmentation. Without the addition of Al or Fe from external sources, the Al/Fe ratio in PAFS was close. This also explained why PAFS had a better effect than PAS on the removal of COD and TP. Moreover, the Fe^{3+} in the polymeric ferric sulfate reacts chemically with phosphate ions to form insoluble phosphate iron (FePO_4) precipitates. The Fe^{3+} further hydrolyze in water, forming $\text{Fe}(\text{OH})_3$ flocs with a higher positive charge, which can adsorb phosphate ions in the water and remove them through flocculation [56].

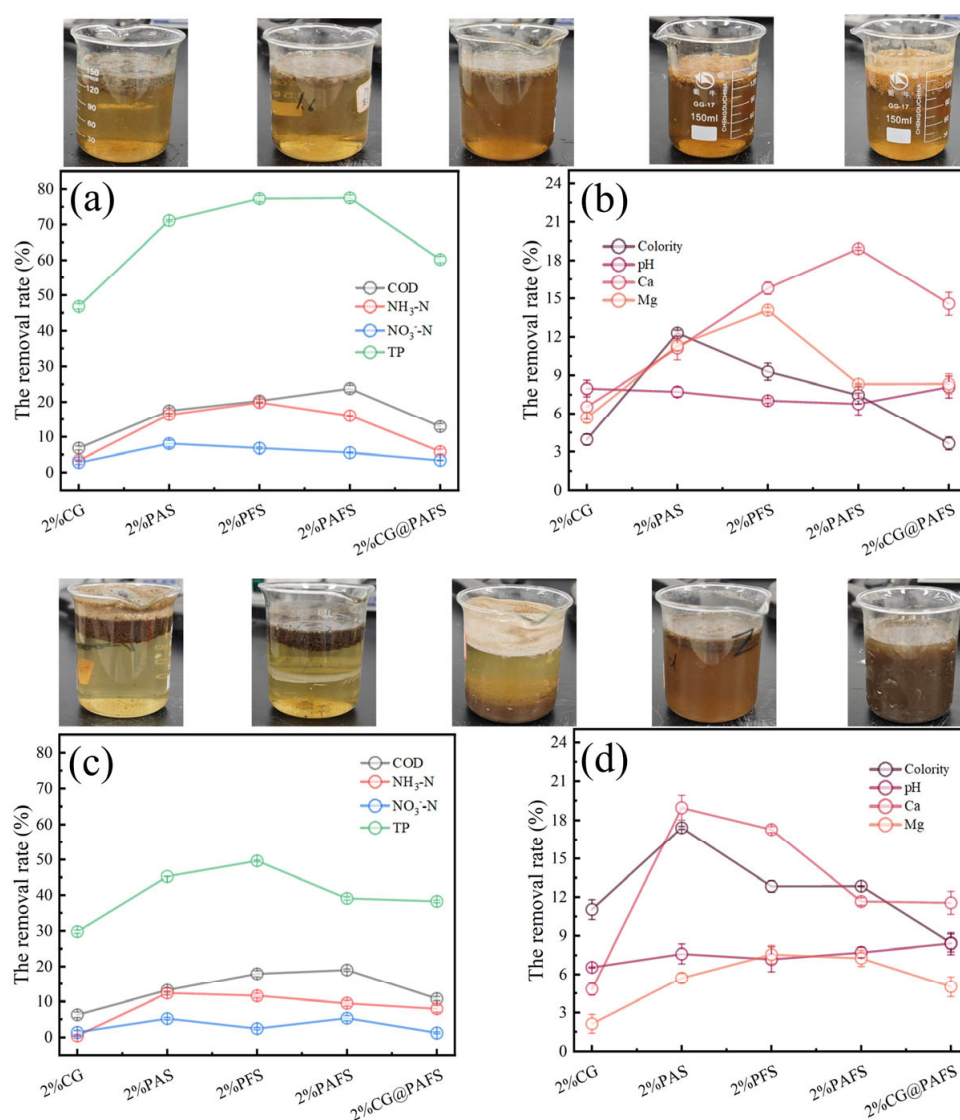


Figure 6. (a) Removal rates of COD, $\text{NH}_3\text{-N}$, $\text{NO}_3\text{-N}$, and TP after treatment of landfill leachate with CG, PAS, PFS, PAFS, and CG@PAFS; (b) Decrease rates of pH, chroma, and removal rates of Ca and Mg after treatment of landfill leachate with CG, PAS, PFS, PAFS, and CG@PAFS; (c) Removal rates of COD, $\text{NH}_3\text{-N}$, $\text{NO}_3\text{-N}$, and TP after treatment of landfill concentrate with CG, PAS, PFS, PAFS, and CG@PAFS; (d) Decrease rates of pH, chroma, and removal rates of Ca and Mg after treatment of landfill concentrate with CG, PAS, PFS, PAFS, and CG@PAFS.

3.4.2. Effect of Treatment of Landfill Concentrate

When using membrane separation technologies (such as reverse osmosis RO, nanofiltration NF, etc.) to treat landfill leachate, most of the pollutants in the leachate were retained on one side of the membrane, while water and some small molecular substances passed through the membrane. The concentration of the retained substances gradually increased, forming a liquid with a higher concentration of pollutants, known as landfill concentrate. Landfill concentrate also required coagulation treatment, and we tested the treatment effects of PAFS and CG@PAFS. Compared to PAS and PFS, PAFS had a higher removal rate for COD and total phosphorus, but a lower removal rate for $\text{NH}_3\text{-N}$ and $\text{NO}_3^- \text{-N}$, which was similar to the coagulation effect of landfill leachate. These results indicated that PAFS prepared from coal gangue has a coagulation effect on landfill leachate and landfill concentrate that was comparable to PFS. However, the unfiltered CG@PAFS also had a certain coagulation effect, but it was not as effective as PFS and PAFS.

3.5. Prospects for Application as a Stabilized Soil Amendment

The sludge (coagulation and sedimentation) produced after the treatment of landfill leachate (Figure 7a,b) and concentrate (Figure 7c,d) contains a large amount of organic matter and nutrients such as nitrogen, phosphorus and silicon. Application of these sludges to the soil can improve the quality of the soil, thus favoring plant growth. However, if the sludge has a high content of organic pollutants, it may cause secondary pollution to soil and groundwater. In this study, the contents of available phosphorus and available silicon of sludge generated from the treatment of landfill leachate and concentrate by CG, PAS, PFS, PAFS and CG@PAFS were tested, and the contents of leachable organic pollutants (COD), $\text{NH}_3\text{-N}$ and TN were also determined. The results are shown in Figure 7. For the treatment of landfill leachate, the available phosphorus in the PFS and PAFS precipitates reached 1.97 mg/L and 1.22 mg/L, respectively. The available phosphorus content in the PAS and CG@PAFS precipitates was 0.97 mg/L and 0.13 mg/L, respectively. However, the available silicon in the PFS and PAFS precipitates was very low. The available silicon content in the CG@PAFS precipitate reached 0.14 mg/L. For the treatment of landfill concentrate, the available phosphorus in the PFS and PAFS precipitates reached 1.97 mg/L and 1.02 mg/L, respectively. The available phosphorus and silicon in the CG@PAFS precipitate reached 0.5 mg/L and 0.17 mg/L, respectively. Therefore, as a soil conditioner, the potential of PAFS precipitate as a phosphorus fertilizer was greater than that of CG@PAFS. After the leaching of iron and aluminum from coal gangue, the leachate residue was mainly composed of silicon compounds, with a higher available silicon content. In the unfiltered case, the flocs formed by PAFS accumulated on the surface of the leachate residue to form a passivation layer, which hindered the dissolution of silicon. This indicated that CG@PAFS can be used as a silicon-releasing material. From the perspective of environmental safety, the available phosphorus content in the PFS and PAFS precipitates exceeded the limit specified in the comprehensive discharge standard for sewage (0.5 mg/L GB 8978-1996) [57]. Therefore, CG@PAFS has more advantages in the prospect of soil conditioner preparation.

3.6. The Removal Mechanisms of PAFS and CG@PAFS on Landfill Leachate

From the experimental section above, it was evident that the aluminum and iron present in coal gangue were leached out under 10% sulfuric acid, resulting in the formation of aluminum ions (Al^{3+}) and iron ions (Fe^{3+}). At an optimal pH, Al^{3+} and Fe^{3+} underwent hydrolysis reactions to produce hydroxyl compounds, which subsequently polymerize to form the flocculant PAFS. Owing to its three-dimensional chain network structure, PAFS was capable of effectively adsorbing suspended solids, colloids, and

some dissolved pollutants in water, facilitating their removal through flocculation. Similarly, Al^{3+} and Fe^{3+} generated hydroxyl compounds via hydrolysis, which then polymerized to form PAFS. The flocs generated by PAFS accumulated on the surface of the coal gangue leachate residue, forming a passivation film that adsorbed certain water pollutants through flocculation. Although the flocculation efficiency of CG@PAFS was slightly inferior to that of PAFS, it can effectively adsorb some pollutants in water and possesses a higher concentration of effective silicon, qualifying it as a silicon slow-release material. The preparation and mechanisms of PAFS and CG@PAFS were depicted in Figure 8.

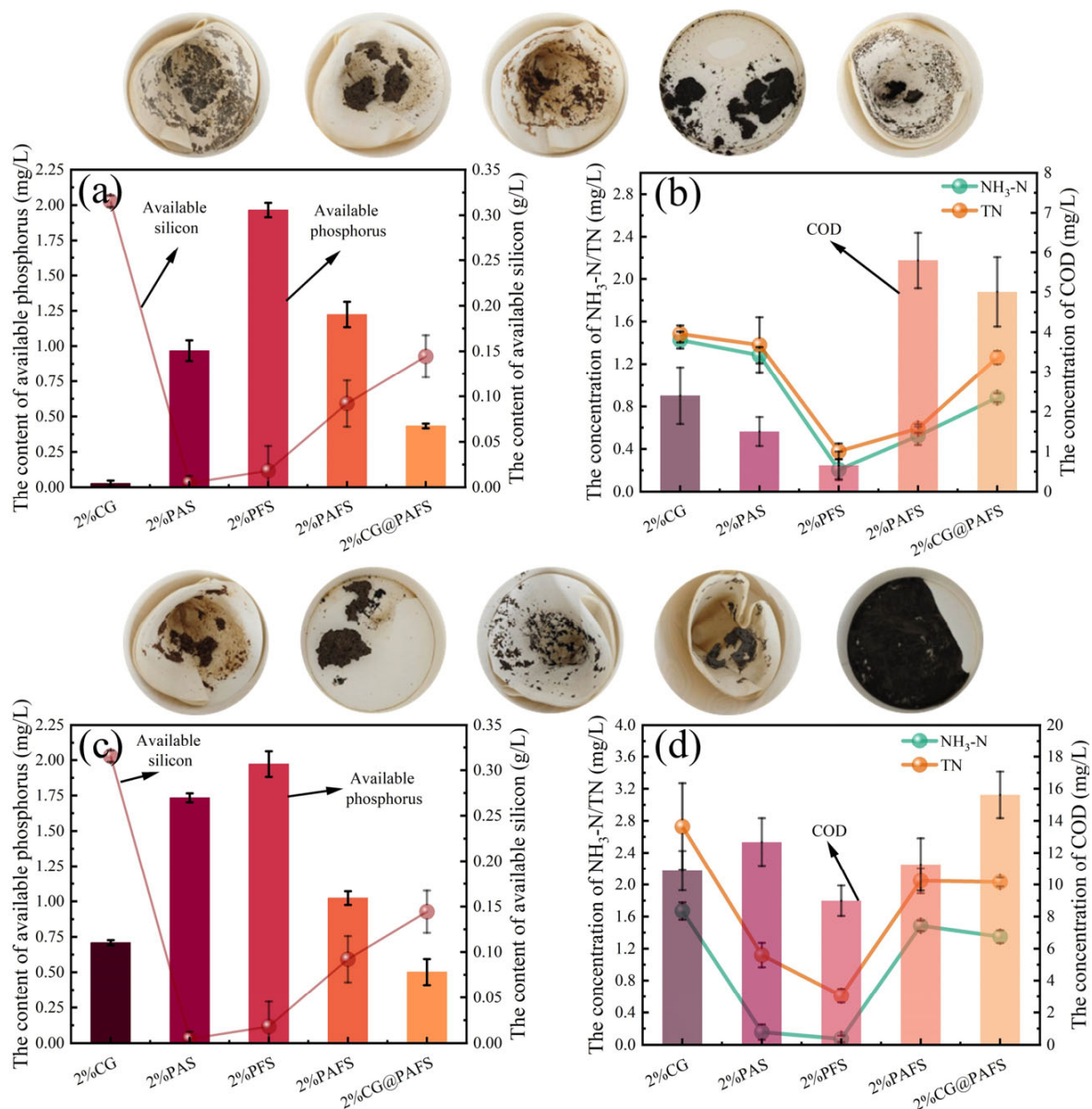


Figure 7. (a) Available phosphorus and available silicon content of sludge after CG, PAS, PFS, PAFS and CG@PAFS treatment of landfill leachate; (b) Leachable COD, $\text{NH}_3\text{-N}$ and TN concentration of sludge after CG, PAS, PFS, PAFS and CG@PAFS treatment of landfill concentrate; (c) Available phosphorus and available silicon content of sludge after CG, PAS, PFS, PAFS and CG@PAFS treatment of landfill concentrate; (d) Leachable COD, $\text{NH}_3\text{-N}$ and TN concentrations of landfill concentrate sludge after treatment by CG, PAS, PFS, PAFS and CG@PAFS.

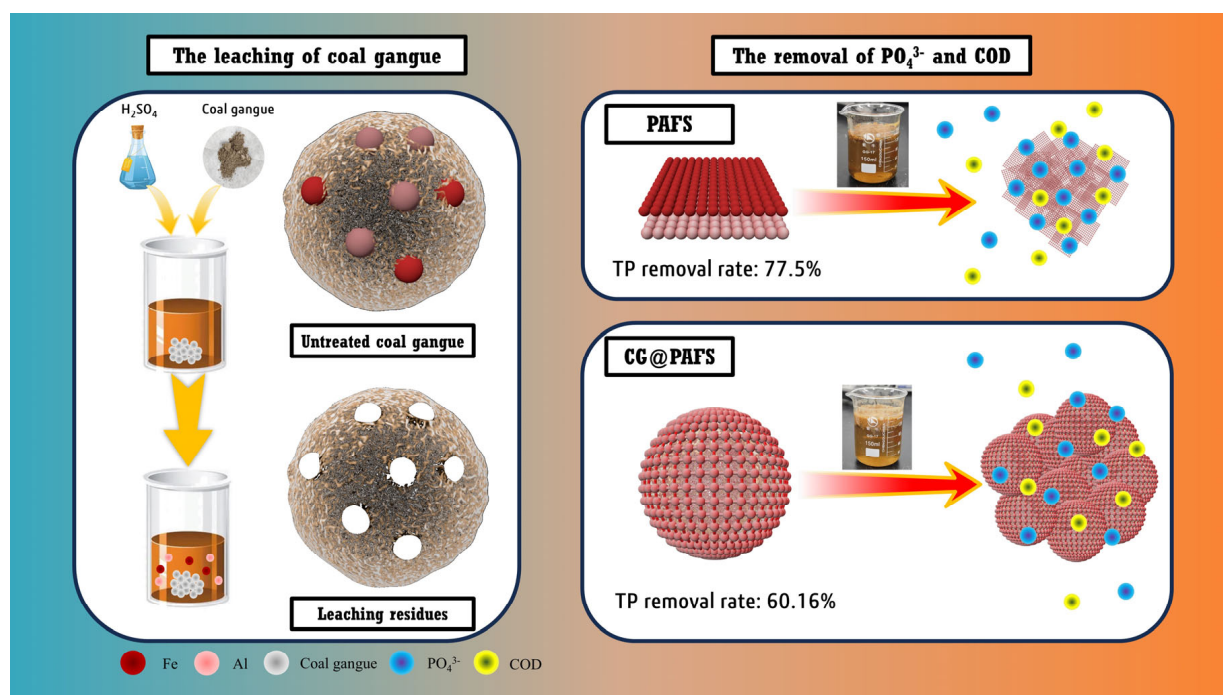


Figure 8. The preparation and mechanisms of PAFS and CG@PAFS.

4. Conclusions

In this study, aluminum and iron were extracted from coal gangue using sulfuric acid leaching, and the coagulants PAFS and CG@PAFS were prepared through polymerization reactions. PAFS exhibited favorable removal efficiencies for COD, total phosphorus, and NH_3-N in landfill leachate and concentrate, which were comparable to PFS and superior to PAS. The precipitates of PAFS and CG@PAFS contained a certain amount of available phosphorus, which could be utilized as phosphate fertilizer, while the CG@PAFS precipitate contained a higher concentration of available silicon, suitable for use as a silicon slow-release material. Overall, this study demonstrates a technically feasible and economically viable circular economy approach that effectively closes the material loop by converting two waste streams—coal gangue and leachate pollutants—into valuable products, namely a composite coagulant and a nutrient-rich soil conditioner. While the PAFS coagulant exhibits moderate removal efficiencies for bulk COD compared to advanced oxidation processes, its exceptional performance in TP removal and its low reagent cost (approx. 150–200 USD/ton) highlight its niche applicability for polishing treatment or pretreatment. From a sustainability standpoint, this cost-effectiveness, coupled with the valorization of waste-derived sludge, significantly reduces the overall environmental and economic burden of leachate treatment, offering a more sustainable and low-carbon alternative to conventional methods that rely heavily on virgin chemicals and generate problematic waste streams. The generation of silicon-phosphorus-rich sludge provides a pathway for resource recovery, provided that stringent environmental safeguards and stabilization protocols are implemented to mitigate pollutant risks. Future research should prioritize life-cycle assessment (LCA) and techno-economic analysis (TEA) to rigorously quantify the environmental and economic benefits of this integrated system across its entire life cycle. Additionally, long-term field trials are essential to validate the agronomic performance and environmental safety of the sludge-derived soil conditioner, ensuring that its application does not introduce secondary pollution. Therefore, this approach represents a cost-effective complementary technology for landfill leachate management rather than a replacement for conventional biological treatment. It constitutes a meaningful step-change towards a more

sustainable, resource-efficient, and circular waste management paradigm that aligns with the core mission of the sustainability agenda.

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