ABSTRACT: The silicon–aluminum–iron flocculant (PAFSi) combines the most abundant resources of waste incineration bottom ash and unpurified water, being regarded as one of the most promising approaches toward water purification. Herein, in this research, waste incineration bottom ash was employed to produce a cost-effective and highly efficient flocculant. PAFSi with a particle size of 214 nm and a zeta potential of 8.63 mV reached the optimum performance using a dosage of 2 mL/50 mL at pH from 8 to 11. The results with the copolymer exhibited the following: (1) a good flocculation efficiency over a wide pH range, (2) superior flocculation performance compared to those of polyaluminum chloride and polyferric sulfate, (3) three-dimensional branching structure of PAFSi micelles with a high aggregation degree, (4) charge neutralization and bridging as the main flocculation mechanism, and (5) recycling the floc. Thus, this work provides an attractive solution to the pressing global clean water shortage problem.

INTRODUCTION

Global waste disposal and clean water shortage are two of the most pressing challenges of developed or developing cities that have driven scientists to pursue new and innovative techniques, such as flocculation, biodegradation, photocatalytic degradation, ozonation, and adsorption, for high-efficiency waste conversion and water purification. Among these methods, flocculation is industrially applicable and financially feasible owing to its relatively simple operation. This method is also regarded as one of the most promising techniques for water purification because of its rapid processing capacities. Highly efficient flocculation requires three general features: cheap raw materials for preparation, high-efficiency flocculants for purification, and recycling. Currently, various raw materials including aluminum sulfate, sodium silicate, magnesium chloride, zirconium sulfate tetrahydrate, etc., have been employed for the preparation of flocculants. Furthermore, chitosan, proteins, and lignins were constructed for efficient waste purification, showing great potential as high-performance and green flocculants. However, despite these successfully reported methodologies, challenges still remain in generating an ideal approach. The majority of current methods for removing impurities in wastewater are difficult to industrialize because of high cost, technology restriction, etc., which inhibit their practical applications. The development of a low-cost flocculant that simultaneously optimizes flocculation performance for water purification is highly desirable.

Waste incineration bottom ash, containing a variety of metal oxides (such as Al, Cu, Zn, Fe, Sn, and so forth) and soluble salts, is the residual solid waste after burning. However, because of the complexity of the extraction process and high cost, no effective and suitable technology has been applied to reclaim the aforementioned value metals. A polysilicate metal salt flocculant can implement two abundant resources: solid waste as a raw material and unpurified water; thus, it is possible to treat waste with actual waste by harnessing specific resources. Herein, we report that waste incineration bottom ash can be employed in the preparation of flocculants for water purification. The structure, performance, and flocculation mechanism of PAFSi were systematically characterized by Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), powder X-ray diffraction (XRD), and thermogravimetric analysis (TGA). It contains a variety of metal oxides, bestowing the flocculant with a three-dimensional branched structure, where PAFSi displays 98% flocculation efficiency (transmittance). Moreover, using this raw material, a low-cost flocculant and sufficient water purification can be realized.

RESULTS AND DISCUSSION

Figure 1 shows the design concept for the preparation of flocculants via waste incineration bottom ash for wastewater treatment. Currently, worldwide, approximately 100 billion tons of bottom ash is produced, of which only a part is utilized (Figure 1a). Another challenge is the presence of a wide...
range of toxic derivatives, in particular, heavy metals, suspended particles, and aromatic molecules in domestic and industrial wastewater, which require efficient removal for purification and possible reuse [Figure 1c(i)].²³,²⁴ Efficient bottom ash and wastewater disposal are of international concern, affecting both the environment and human health.²⁵−²⁷ Hence, research into resolving such issues is of considerable interest to our group and many others. Based on its unique characteristics and structure, the flocculant, an inorganic metal polymer, has proved its advantages in the separation wastewater,¹⁰ thus combining metal ions (bottom ash) and impurities (waste water) for purification.²⁸−³⁰ The above design was systematically optimized, and the preparation of the flocculant using bottom ash (Figure 2) as well as the flocculation performance and mechanism was explored. Therefore, we report that our flocculant, stemming from bottom ash, has a high purification efficiency in the preparation of clean water (Figure S1, Supporting Information) and can be recycled (Figure 1b). The proposed technology possesses two key advantages: (1) resolving the issues of waste resource and waste incineration bottom ash and (2) preparation of a new highly efficient and cheap flocculant and recycling flocs.

Structural characterization of PAFSi is presented in Figure 3. The absorption peaks at 1054 and 552 cm⁻¹ correspond to the bending vibrations of Si–O–Si and the stretching vibrations of Fe–O bonds of waste incineration bottom ash, respectively [Figure 3a(i)].³¹,³² The peaks at 3412, 1103, 942, 610, and 474 cm⁻¹ (Figure 3a(ii)) relate to OH, Si–O–Al, Si–OH–Fe, Al–OH, and Fe–OH, respectively.³³ The peak at 1635 cm⁻¹ is the stretching vibration peak of H₂O.¹⁰ Figure 3b shows that the XRD pattern corresponding to mullite (b), CaO (a), and Fe₂O₃ (c) is in agreement with that recorded by Orooji,³⁴ Chen,³⁵ and Wang.³¹ In PAFSi [Figure 3b(ii)], metal compounds (including SiO₂, Al₂O₃, and Fe₂O₃) phases disappear, whereas NaCl (d) characteristic peaks are enhanced with the addition of Na₂CO₃, indicating that PAFSi is a complex rather than a simple mixture.

The microstructure for PAFSi was analyzed by transmission electron microscopy (TEM)—energy-dispersive system (EDS) and SEM (Figure 3c,d). Particles with different lengths of branches and different degrees of polymerization as well as PAFSi, being a chain-net structure with three dimensions [Figure 3c(ii)],³⁶,³⁷ are observed, and the amorphous polymer is copolymerized by Al, Fe, and Si elements [Figure 3c(ii)]. The morphology difference between SEM and TEM of

Figure 1. Schematic illustration of the silicon–aluminum–iron flocculant from waste incineration bottom ash. (a) Waste incineration bottom ash. (b) Recycling utilization for PAFSi. (i) Synthetic route for PAFSi. (ii) Flocculation for kaolin wastewater. (iii) Floc recycling. (c) Clean water after flocculation from kaolin wastewater, suggesting its low-cost potential application in water purification.

Figure 2. Chemical formula of PAFSi.
PAFSi is observed, and SEM images display larger and more irregular shapes than TEM images because of the drying treatment. The phenomenon homogeneous yellow colloid with Tyndall, which is consistent with FTIR, XRD, TEM−EDS, and SEM analysis. The TGA−differential scanning calorimetry (DSC) curves of PAFSi are shown in Figure 3f, which clearly show a significant weight loss stage (−43.34%) with an exothermic peak of 162.3 °C within the temperature range of 40−300 °C, which is related to the loss of physisorbed water. In the temperature range between 300 °C and approximately 800 °C, the structural hydroxyl groups condense and dehydrate, displaying an exothermic peak of 589.5 °C and 6.13% weight loss. Because of breaking of Si−O−Al, Si−O−Fe, and Si−O−Si chemical bonds, 10.01% weight loss occurs above 800 °C. FTIR, XRD, TEM−EDS, SEM, and TGA−DSC confirm that a new chemical species of PAFSi consisting of iron, aluminum, and silica is formed. Based on the structural characterization of PAFSi, the polymerization of waste incineration bottom ash promotes the formation of irregular polymers with Fe−Si, Al−Si, Fe−O−Si, or Al−O−Si bonds.

Generally, insufficient dosage of the flocculant influences the flocculation performance; hence, it is crucial to evaluate this parameter on kaolin wastewater efficiency. As shown in Figure 4a(i), especially at a lower dosage (1 mL/50 mL), the flocculation performance of PAFSi is greater than that of polyaluminum chloride (PAC) and polyferric sulfate (PFS) because PAFSi possesses higher transmittance and removal rates of impurities at the same dosage. The optimal flocculation performance is observed for PAFSi that reaches 96.61% (transmittance) at a low dosage of 2 mL/50 mL. This demonstrates the treatment of high concentration wastewater at low dosage. Because of the PAFSi neutralization and adsorption bridging effect, the negatively charged colloidal particles in kaolin wastewater destabilize and aggregate, followed by flocculation, and cohere into large flocs, resulting in precipitation and separation of impurities. However, the transmittance decreases slightly upon overdosing of PAFSi, indicating that colloids stabilize again making it difficult to flocculate and settle. Owing to colloidal particles being completely covered by the absorbed PAFSi, regeneration of the electrostatic repulsion forces and a new well-established suspension are observed. Obviously, the volume of flocs formed by PAFSi is smaller than that of PAC but larger than that of PFS. This is due to PAFSi exhibiting strong aggregation ability to form more compact flocs [Figure 4a(ii)].

The flocculation performances (2 mL/50 mL dosage) under different pH conditions are displayed in Figure 4b(i). Under acidic conditions, the flocculation performance of PAFSi, PFS, and PAC is poor, with a transmittance below 90%. Because of excess of hydrogen ions, excluding the positive charges of aluminum and iron, the charge neutralization capacity is weakened between the positively charged flocs and negatively charged colloids.

Under alkaline conditions, PAFSi and PAC display a slight increase in transmittance with increasing dosage, whereas PFS decreases dramatically. In the pH range 8−11, the removal rates (higher transmittance) of PAFSi (above 95%) are higher.

Figure 3. Morphology and structural characterization of PAFSi. (a) FTIR spectra of PAFSi. (i) Waste incineration bottom ash. (ii) PAFSi. (b) XRD patterns of PAFSi. (i) Waste incineration bottom ash. (ii) PAFSi. (c) Microstructure of PAFSi. (i) TEM of PAFSi. (ii) EDS of PAFSi. (d) SEM of PAFSi. (e) Morphology of PAFSi. (i) Natural state of PAFSi. (ii) Tyndall phenomenon for PAFSi. (f) TGA−DSC for PAFSi.
than that of commercial flocculants (PFS and PAC), highlighting its enhanced flocculation performance and wider pH scope for application (Figure 4b(i)). The increase in OH$^-$ production within this pH range allows these ions to react with (Fe−Al)$_a$ or Fe$_a$ species generating more high polymers, thus improving the bridging flocculation. However, Fe and Al ions are unsuitable for the formation of hydroxide precipitates, which deteriorates the flocculation performance, suggesting that PAFSi may have a higher cation concentration in polymers that further strengthens the electrical neutralization. The volume of flocs formed by PAFSi is relatively small under acidic conditions because of poor flocculation effects [Figure 4b(ii)]. Therefore, PAFSi is more suitable for alkaline conditions.

In order to explore the flocculation mechanism, further characterization and experiments were carried out, as shown in Figure 5a. In the floc, FTIR displayed new characteristic peaks at 459 and 563 cm$^{-1}$ corresponding to Si−O−Si and Si−O−Al bending vibrations of kaolin, respectively. Intense broad bands observed at 3449 and 1636 cm$^{-1}$ are due to OH and H−OH stretching vibrations of flocs, respectively. This confirms that the structure of PAFSi remains unchanged (Figure 5a). Compared with kaolin, no new or other intense peaks appear in flocs, with only typical kaolin characteristic peaks, namely, mullite (Al$_6$Si$_2$O$_{13}$), being observed (Figure 5b). However, this may be due to kaolin covering the surface of PAFSi generating flocs. To further verify these results, SEM−EDS analysis of PAFSi and flocs was carried out. PAFSi displayed a stratified distribution of clusters with rough surfaces and covered by irregular branched long-chain structures, which is consistent with SEM results [Figure 5c(i)]. The branched structures of PAFSi, promoting charge neutralization as well as adsorption and bridging performance for purification, are more favored for the coagulation of colloidal particles and bridge-aggregation formation. Compared with PAFSi, flocs are composed of closely clustered and arranged fragments and particles of various sizes, showing irregular three-dimensional clusters [Figure 5d(i)]. The presence of larger and denser flocs, with relatively few elements (including O, Al, and Si), especially with a higher Al element content (21.51% in flocs, but 6.64% in PAFSi), proves that kaolin coats PAFSi (including C, O, Na, Al, Si, Cl, and Fe) [Figure 5c(ii),d(ii)].

To further verify the flocculation process, the particle size distribution of PAFSi, kaolin wastewater, and flocs was tested. Figure 5e shows that the particle sizes of PAFSi and kaolin wastewater are 214 and 2622 nm, respectively, whereas flocs show an enlarged size of 10419 nm, implying that they are formed via close aggregation of numerous small molecular particles. These results are consistent with SEM data [Figure 5d(i),e].

As shown in Figure 5f(i), by estimating the neutralization ability of PAFSi, the zeta potential results of initial kaolin wastewater and PAFSi are −11.08 and 8.63 mV, respectively, whereas the zeta potential of wastewater after flocculation is 7.86 mV. Figure 5f(i) shows that PAFSi contains positively charged iron and aluminum that are more easily reacted with negatively charged kaolin particles through charge neutralization; thus, the small flocs and kaolin can be assembled and adsorbed onto PAFSi. The kaolin particles and smaller flocs attach to PAFSi via strong bridging effects producing larger-sized flocs (Figure S2, Supporting Information). As a result, bridge-aggregation flocculation gathered smaller flocs, generating larger ones with a compact floc structure. Consequently, a possible flocculation mechanism schematic diagram is deduced and displayed in Figure 6i,ii.
The zeta potential of kaolin wastewater by PAFSi (8.26 mV) is much higher than that by PFS (5.16 mV) and PAC (7.93 mV) after processing, indicating that PAFSi has a greater ability for charge neutralization, which is in agreement with experimental results (Figure 4a,b). This confirms that PAFSi shows better performance than the commercial flocculants PFS and PAC [Figure 5(ii)]. Compared with commercial flocculants, the positive charges of Fe and Al in mesh chains can absorb a large amount of negatively charged particles, thereby causing flocculation into larger and denser flocs. In addition, PAFSi exhibits an enormous mesh structure and discontinuous surface with many channels or voids and branches, facilitating charge neutralization, adsorption, and bridging effects.

Based on the above analysis, the positive charges of Al and Fe in PAFSi bearing many channels or voids and branches have optimal charge neutralization capacity, where silicon provides strong chelation to the impurity of kaolin particles, thereby improving the degree of bridge aggregation and sweep flocculation. Recycling. Recycling is an important factor when considering the large quantities of dangerous flocs with residual metals. According to inductively coupled plasma–optical emission spectroscopy analysis, 36 and 14% of Si and Al content, respectively, are found in flocs. The reusability of flocs was evaluated using Na2CO3 by calcining at 900 °C to prepare a soluble salt. By comparing XRD patterns of NaAlSiO4 (JCPDS35-0424) characteristic peaks, formation of the target product is confirmed (Figure S3, Supporting Information). Hence, flocs can be recycled by a simple process as a green technology for resource utilization.

Obviously, when considering practical applications of PAFSi, in addition to flocculation efficiency, low cost is essential. Waste incineration bottom ash was used in the fabrication of PAFSi via a simple chemical technology. The total cost, including material and processing, is significantly low.
example, the cost of waste incineration bottom ash is free, while the manufacturing cost is low because of the simple chemical process. Additional benefits include easy recycling of flocs. Previous studies in this area are summarized in Table 1.

**CONCLUSIONS**

In summary, we report a silicon–aluminum–iron flocculant based on waste incineration bottom ash via chemical conversion for recovery and comprehensive utilization of waste resources. The structural characterization, performance, and flocculation mechanism of PAFSi were comprehensively characterized. The results show that PAFSi achieves water resource purification and waste resource utilization. Furthermore, for usual conditions of pH (from 6 to 8) in raw water, both PAC and PAFSi showed a comparative efficiency in the flocculation process. For other pH values PAFSi performed better owing to its excellent charge neutralization, adsorption, and bridging effects. The simultaneous optimizations of the PAFSi dosage and pH value on wastewater contributed to a higher water purification efficiency of over 95%. The floc after flocculation was recycled with zero emission, making it a type of green chemistry reaction for the flocculation system. Moreover, the utilization of waste incineration bottom ash enables high-efficiency, low-cost, and sustainable PAFSi for water purification in the real world, especially in countries where waste incineration is vast, but pure water is in great demand.

**EXPERIMENTAL SECTION**

**Materials.** Waste incineration bottom ash was supplied by a domestic waste incineration plant in the Yunnan province, China, and its main components are shown in Table S1 (Supporting Information). Kaolin, the PAC flocculant, and the PFS flocculant were supplied by Chengdu Aikeda Chemical Reagent Co., Ltd. All chemicals were used without further purification.

**Synthesis of PAFSi.** Waste incineration bottom ash (10.0 g) and sodium carbonate (4.0 g) were placed on a 50 mL crucible and heated at 900 °C for 1.5 h using a muffle furnace. After cooling, the calcined products were charged into a 250 mL beaker and mixed with 4 M HCl (100 mL) by titration within 0.5 h. The mixture was stirred for 1 h in a water bath at 80 °C and then at room temperature for 24 h. The target product was obtained by filtration. The synthetic pathway and schematic illustrations are shown in Figure S4 (Supporting Information).

**Flocculation Experiments.** First, kaolin wastewater (10 g/L) was prepared by combining kaolin (1.0 g) and deionized water (100 mL). The initial pH of kaolin wastewater was adjusted to the specified pH using 1 M HCl or 1 M NaOH. After standing for 30 min, PAFSi (2 mL, PFS or PAC) was added to wastewater (50 mL, 11.53% of light transmittance at 2 cm under the surface) and mixed at 200 rpm for 3 min, followed by quiescent settling for 30 min. Then, the wastewater 2 cm below the liquid surface was collected for transmittance measurement under 590 nm of the maximum absorption wavelength, and the transmittance was calculated using eq 1. The volume of flocs was recorded, and the flocs were dried under vacuum at 80 °C for further characterization.

\[ T = 10^{-A} \times 100\% \]

where \( T \) is the transmittance and \( A \) is the absorbance, L/(g·cm).

**Characterization and Measurements.** Infrared spectra of the samples were obtained via a Fourier-transform infrared spectrometer (Nicolet, USA) using the potassium bromide pellet method. XRD patterns of the samples were measured on a D/MAX-Ultima IV X-ray diffractometer (Rigaku, Japan) equipped with the diffraction angle scanned at a speed of 8° min⁻¹ over a 2θ range of 5°–80°. SEM images were obtained on a S4800 (Hitachi, Japan) at an acceleration voltage of 10 kV, and TEM images were obtained on a Jeol 2100F (JEOL, Japan). The zeta potential of the samples was obtained on a Malvern zetasizer Nano ZS90. The particle sizes of the samples were determined using a 90Plus PALS (Brookhaven, USA) operating at 25 °C. The thermal stability of the samples was obtained using a TGA2 (Mettler Toledo, Switzerland) ranging from 25 to 1000 °C at a rate of 10 °C min⁻¹. The element composition of the samples was obtained using X-ACT energy-dispersive X-ray spectroscopy (OXFORD, England).

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c01296.

Main components of waste incineration bottom ash; schematic diagram of experimental steps of PAFSi; flocculation picture of PAFSi; picture of flocs from an
optical microscope; and XRD patterns of recycling flocs (PDF)

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