

Article

Preparation, Characterization, and Application of P(aluminum chloride-co-diallyldimethylammonium chloride) Hybrid Flocculant

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Abstract: The hybrid flocculant P(aluminum chloride-co-diallyldimethylammonium chloride) was synthesized in this study. Diallyldimethylammonium chloride monomers were used and ammonium persulfate served as the initiator. The structure of P(aluminum chloride-co-diallyldimethylammonium chloride) was characterized using Fourier-transform infrared spectroscopy, scanning electron microscopy, an electrical conductivity test, and thermogravimetric analysis. Single-factor experiments were conducted to optimize the synthetic conditions of the hybrid flocculant. An optimized product with an intrinsic viscosity of 926.36 mL/g and a flocculation decolorization rate of 99% was obtained under the following reaction conditions: the total monomer concentration was 30%, the initiator concentration was 0.7%, the reaction temperature was 60 °C, and the reaction time was 3 h. The results demonstrated that the PAC-PDMAAC hybrid flocculant exhibited covalent bonding between its organic–inorganic components and displayed enhanced stability properties due to its high intrinsic viscosity and spatial structure. Moreover, this hybrid flocculant showed superior decolorization performance in disperse-violet-H-FRL-dye wastewater.

Keywords: copolymers; preparation; characterization; flocculant; infrared spectrogram



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1. Introduction

Municipal and industrial wastewaters, such as from printing and dyeing, food processing, or chemical production industries, contain large amounts of organic pollutants, are of poor quality, and pose a severe threat to underground and surface water resources. Flocculation technology is of vital importance, with a wide range of applications in water and wastewater treatment plants. The performance of flocculation is largely affected by the presence of flocculants that enhance the aggregation of particles to form large, rapid-setting flocs either through charge neutralization or chain-bridging mechanisms [1]. Chloride is a mainstream flocculating agent in the water and wastewater treatment industry. It has characteristics such as a low required dosage, a high electrical-neutralization capability, fast floc growth, and decreased sludge production [2,3]. But its flocculation stability is poor, especially for colloid adsorption that bridges weaker-than-organic polymer flocculants. Poly-diallyldimethylammonium chloride is a stable, organic polymer flocculant. It has an excellent flocculation effect but with a high cost [4–6]. Through the study of organic–inorganic composite flocculants, we found that the synergistic effect of polydimethyl diallyl ammonium chloride and polyaluminum chloride can significantly improve the flocculation effect [7,8]. However, due to the weak hydrogen bond between the inorganic and organic phase, the anti-shear ability of the floc is poor. Therefore, the preparation of a new organic–inorganic hybrid flocculant by the hybrid method has been proposed. The formation of a strong bond between polydimethyl diallyl ammonium chloride and polyaluminum chloride polymer can not only electrically neutralize inorganic particles but also bridge the polymerization of organic polymers. Moreover, P(aluminum chloride-co-diallyldimethylammonium chloride) is cheaper than other cationic polymers. Therefore,

P(aluminum chloride-co-diallyldimethylammonium chloride) is expected to be used in water treatment, printing and dyeing wastewater, sewage treatment, etc.

To produce P(aluminum chloride-co-diallyldimethylammonium chloride) hybrid flocculant, the high charge density of an inorganic flocculant and the high bridging ability of an organic polymer flocculant were combined [7,8]. This improved the surface morphology structure, the charge distribution of the inorganic particles, and its adsorption bridging and charge neutralization capabilities. The organic and inorganic particles can be linked by hydrogen or covalent bonding to keep their performance stable. A hybrid flocculant can maximize the synergies between inorganic and organic components, with a better flocculating efficiency than single-component flocculants or composite flocculants. Usually, there are three ways to form covalent bonds in an organic–inorganic phase. First, the active groups on the polymer chain, such as a carboxyl group, a halogen, or a sulfonic acid group, or other chemical reactions with an inorganic particle's active surface groups form covalent bonds [9–11]. Second, the hydrolysis and condensation process of certain groups on the polymer chain forms covalent bonds, such as $-\text{Si}(\text{OR})$ [12,13]. Last, there is the grafting method on an inorganic particle's surface [14–16]. Template copolymerization is the combination of a preformed macromolecule through hydrogen or ionic bonds and another monomer that is not, and the monomers adsorb on the template to form a block structure. In this study, the basic idea of synthesizing covalently bound hybrid flocculants by template copolymerization was as follows: Firstly, a “molecular bridge” was established between inorganic hydroxyl polyaluminum and organic dimethyldiallyl ammonium chloride by using a silane coupling agent, which has groups that react with both inorganic substances and organic substances. Dimethyldiallyl ammonium chloride polymerization was initiated by using ammonium persulfate, and a covalent PAC-PDMAAC hybrid flocculant was obtained. The optimum preparation conditions, the structural characterization, and the decolorization properties of this new hybrid flocculant were studied.

In this paper, we present the synthesis of a novel organic–inorganic hybrid polymer flocculant, PAC-PDMAAC, which leverages the strong bridging effect of organic polydimethyl diallyl ammonium chloride and the robust electric-neutralization properties of inorganic aluminum flocculants. This innovative approach facilitates the integration of organic and inorganic components at the molecular level, thereby achieving a successful synthesis of hybrid flocculants that combines both inorganic and organic polymers. This research opens new avenues for exploring hybrid flocculants and holds significant implications for advancing adaptive technologies in wastewater coagulation treatment.

2. Materials and Methods

2.1. Materials

The monomer DMAAC (60%, *w/w*) was obtained from the Aladdin Reagent Company (Shanghai, China). PAC was obtained from Lanjie Tap Water (Chongqing, China). The composite flocculant in the experiment was obtained by mixing PAC with PDMAAC. Disperse violet H-FRL (purity 100%) was obtained from Shanyu Dyestuff Chemical Company (Taizhou, China). The other reagents used in the experiments, including ammonium persulfate, silane coupling agent (KH570), sodium hydroxide, acetone and ethanol, were of analytical grade. All aqueous and standard solutions were prepared with deionized water. The purity of nitrogen gas was higher than 99.999%.

2.2. Preparation of the Template Polymer

Polyaluminum chloride (PAC) does not exist as Al^{3+} in aqueous solution, but it is dispersed in the form of hydroxyl polyaluminum. It is difficult for hydroxyl aluminum ions to polymerize directly with dimethyl diallyl ammonium chloride, so surface modification of hydroxyl aluminum must be carried out first. γ -methylacryloxy propyl trimethoxysilane (KH570) was used as a modifier. KH570 was hydrolyzed to produce silanol, which could be combined with hydroxyl aluminum by a dehydration reaction. Then, after the initiation of ammonium persulfate, hydroxyl aluminum/KH570 free radicals and dimethyldiallyl

ammonium chloride free radicals were produced, and the hybrid polymerization product could be obtained through the addition of double bonds.

The preparation of hybrid flocculant was performed as follows: first, the modified agent KH570 was hydrolyzed for 12 h with 5% hydrochloric acid to adjust the pH to 3~4. Polyaluminum chloride and KH570 were added to a reaction vessel made of silicate glass. The pH value of the aqueous solution was adjusted to alkaline, in favor of polymeric hydroxy aluminum surface modification. Then, a certain amount of dimethyl diallyl ammonium chloride (DMDAAC) solution was added, evacuated, and filled with nitrogen three times. Under nitrogen, a certain amount of ammonium persulfate initiator was slowly added. The reaction vessel was sealed and placed in a thermostatic oscillator for a specific period. The flocculant was produced, and the aqueous solution changed to a pale-yellow colloid. The polymer products were precipitated with ethanol and washed with acetone. The polymer was then ground into a powder after it had been dried for 48 h in a vacuum-drying oven (Shanghai, China) at 60 °C. The possible reaction scheme of P(aluminum chloride-co-diallyldimethylammonium chloride) is presented in Figure 1.

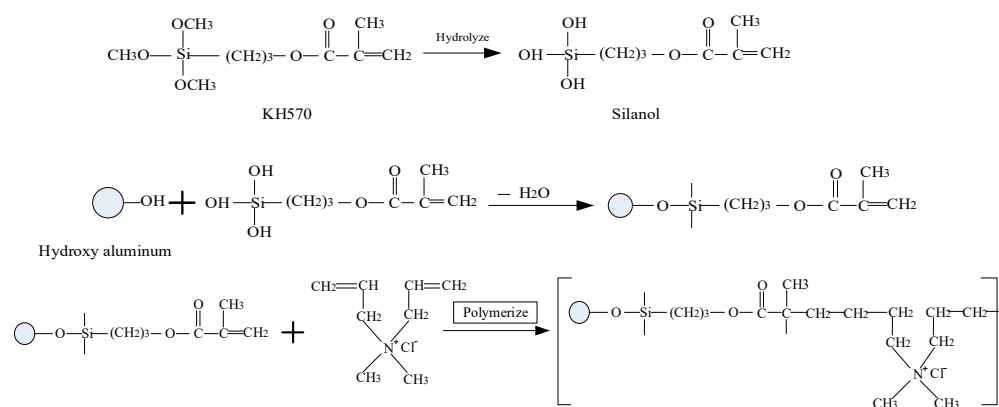


Figure 1. Scheme of the polymerization reaction.

2.3. Characterization of the Polymer

Intrinsic viscosity is an important index of a flocculant, because it directly affects the application performance of the flocculant. Research has shown that the higher the intrinsic viscosity, the larger the molecular weight of the flocculant; therefore, the easier it is to exert the adsorption-bridging capacity of the flocculant and the better the flocculation effect [17]. The intrinsic viscosity of the hybrid flocculant in deionized water was determined by using a 0.46 mm inner diameter, non-dilutive Uhlér viscometer at a constant temperature of 25 °C.

The FTIR of the polymer and the monomers was conducted using an AVATAR-370 infrared spectrometer (Nicolet, WI, USA). The interval of the measured wave numbers was from 400 to 4000 cm^{-1} .

SEM was used to characterize the polymer morphology on an S-3400N scanning electron microscopy instrument (Hitachi, Japan). After the sample was dried, it was affixed to the sample holder with double-sided tape, and the gold film was plated with an ion sputter.

The electrical conductivity test was used to test the ionization properties of the materials. The conductivity of the samples in deionized water was determined by a Sension5 conductivity meter (Hach, NM, USA) at a constant temperature of 25 °C.

TGA was used to determine the thermal degradation of the flocculant samples, which was carried out at a heating rate of 10 °C min^{-1} , a nitrogen flow rate of 50 mL min^{-1} , and a temperature range of 20–600 °C on a DTG-60H synchronal thermal analyzer (Shimadzu, Kyoto, Japan).

2.4. Decolorization Test

Using disperse violet dye, 100mg/L dye wastewater was formulated. Flocculation decolorization experiments were conducted in a jar test using a ZR4-6 program control coagulation experiment blender (Zhongrun Water Industry Technology and Development, Shenzhen, China). The disperse violet dye was poured into beakers and was stirred at a high speed of 500 rpm for 10 min, and then the flocculant solution was added immediately. This solution was mixed at a high speed of 200 rpm for 3 min and a lower rate of 50 rpm for 13 min. After 20 min settling, the water sample was removed from 3 cm below the liquid level and placed on a DR5000 desktop UV-VIS spectrophotometer (Hach, NM, USA) for continuous UV scanning. The absorbance was measured at the maximum absorption wavelength of the dye, and the decolorization rate was calculated.

3. Results and Discussion

3.1. Optimizing Synthesis Conditions

3.1.1. Effect of Modifier KH570 Concentration

KH570 played a role in the surface modification of hydroxyl aluminum. The hydroxyl group on the surface of the polymerized hydroxyl aluminum was used to produce a condensation reaction with KH570, so that the inorganic substance and the organic compound were connected by a chemical bond, thus forming polymer products.

In order to ensure the complete hydrolysis of the silane coupling agent to silanol, KH570 was hydrolyzed under acidic conditions for 12 h. The hydrolyzed solution of KH570 was tested by Fourier-transform infrared spectroscopy. The Fourier-transform infrared spectrum is shown in Figure 2. The wavenumber in the range of 1675~1500 cm^{-1} shows two absorption peaks of C=C double bond stretching vibration, while the absorption peak at 1000~650 cm^{-1} indicates C-H bending vibration. There is no Si-O-C siloxane stretching vibration located at 850 cm^{-1} , but instead, a very obvious stretching vibration absorption peak is observed in the range of 3200~3500 cm^{-1} . This suggests that KH570 has been completely hydrolyzed to silanol, meeting the experimental requirements. Subsequently, different ratios of hydrolyzed KH570 and PAC (such as 3/100, 5/100, 7/100, and 9/100) were mixed, and the solution was alkalized and reacted for 4 h to obtain the optimal modified product. The FTIR technique was used to test these modified products under different KH570 concentrations (Figure 3). According to the modification principle, the success of the modification test was based on the absence of, or the presence of very few, hydroxyl vibration absorption peaks. Upon the observation of Figure 3, it can be seen that, with an increase in KH570 content, the -OH absorption peak at 3200~3500 cm^{-1} gradually reduces until it reaches a KH570 and polyhydroxy aluminum quality ratio of 9%, where the alcoholic hydroxyl peak did not appear. Therefore, a quality ratio of KH570 and PAC at 9% was chosen as the best modification ratio based on these results.

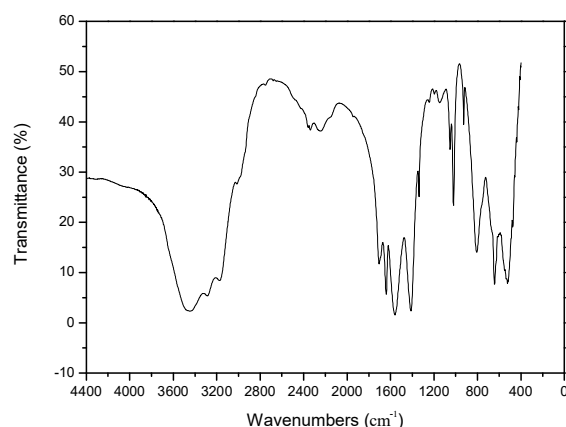


Figure 2. FTIR spectrum of KH570 hydrolysate.

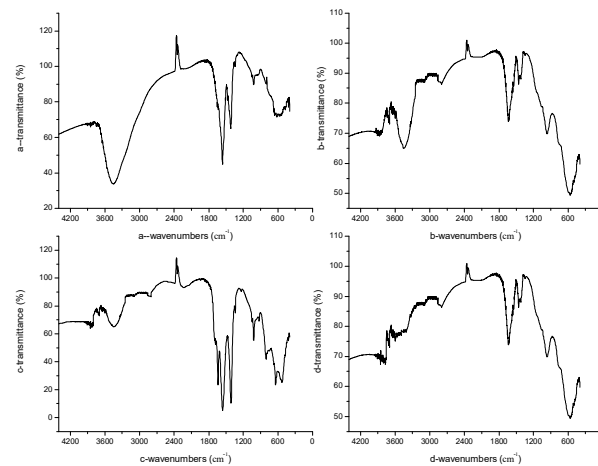


Figure 3. FTIR spectra of modified products when mass ratios of KH570 and polyhydroxy aluminum were 3/100 (a), 5/100 (b), 7/100 (c), 9/100 (d).

3.1.2. Effect of DMDAAC Concentration

The hybrid flocculant was prepared according to the Figure 1. Figure 4 shows the influence of the monomer mass fraction (mass ratio of DMDAAC monomer to PAC) on the intrinsic viscosity of the hybrid flocculant. According to Figure 4, as the dosage of DMDAAC increases, there is an initial increase in intrinsic viscosity followed by a decline after reaching a peak. The maximum intrinsic viscosity of the product was achieved when the DMDAAC and PAC quality ratio was 30%. This was attributed to the promotion of the copolymerization reaction with the increase in DMDAAC. Once a certain concentration of DMDAAC was reached in the system, it easily formed polydimethylene diallyl ammonium chloride. As a result, there was a decrease in monomers participating in copolymerization, leading to a reduced intrinsic viscosity. When the mass fraction of the monomer reached 30%, the intrinsic viscosity peaked at 416.51 mL/g.

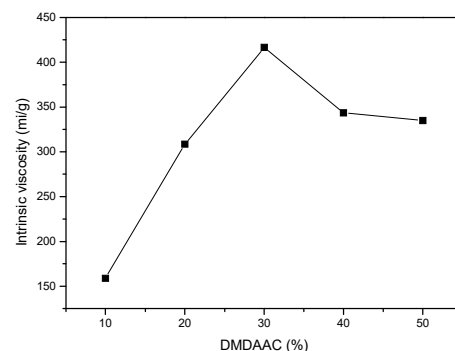


Figure 4. The influence of DMDAAC on intrinsic viscosity.

3.1.3. Effect of Initiator Concentration

Figure 5 shows the influence of the mass fraction of the initiator ammonium persulfate (mass ratio of ammonium persulfate to the DMDAAC monomer) on the intrinsic viscosity of the hybrid flocculant. As illustrated in Figure 5, the quantity of initiator exerted a significant influence on the intrinsic viscosity of the hybrid flocculant. With an increase in the initiator dosage, there was an initial rise in intrinsic viscosity followed by a gradual decrease. This phenomenon could be attributed to the initiation of monomer polymerization by the initiator. Upon reaching a certain threshold of concentration, an excess of free radicals may lead to an elevated risk of termination reactions between these free radicals, ultimately resulting in a reduction of intrinsic viscosity. This pattern closely resembles the typical behavior observed in free-radical polymerization processes. When the amount of

ammonium persulfate was 0.7% of the mass of dimethyl diallyl ammonium chloride, the intrinsic viscosity of the product reached the maximum, 719.59 mL/g.

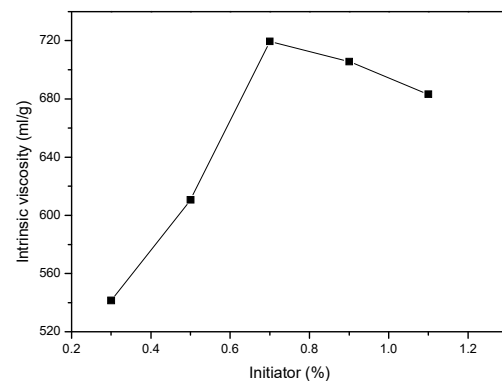


Figure 5. The influence of initiator on intrinsic viscosity.

3.1.4. Effect of Reaction Temperature

Figure 6 shows the effect of the reaction temperature on the intrinsic viscosity of the hybrid flocculants. In Figure 6, it can be observed that the intrinsic viscosity of the hybrid flocculant increases with an increase in the reaction temperature. The intrinsic viscosity reached a maximum at 60 °C and then slightly decreased, which was consistent with the general laws of free-radical polymerization. This was because, with the increase in the reaction temperature, the molecular motion speed accelerated, the collision probability increased, the polymerization speed accelerated, the polymerization speed of the monomers rapidly increased, and the relative molecular weight of the product also increased. However, when the temperature rose to a certain point, the initiator initiation speed accelerated, and the free radicals of the system reached their highest concentration in a short time. The probability of a collision between free radicals also increased rapidly, resulting in an increase in the speed of chain transfers and chain termination reactions; therefore, the characteristic viscosity of the product decreased, and the relative molecular mass also decreased.

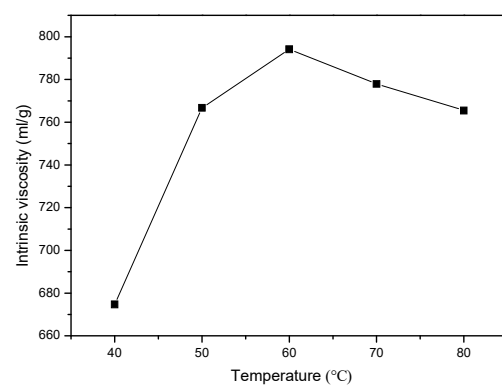


Figure 6. The influence of reaction temperature on intrinsic viscosity.

3.1.5. Effect of Reaction Time

Figure 7 shows the effect of the polymerization reaction time on the intrinsic viscosity of the hybrid flocculant. According to Figure 7, as the reaction time was prolonged, the intrinsic viscosity of the hybrid flocculant initially increased and then decreased. The maximum intrinsic viscosity was achieved at 3 h. This behavior can be attributed to an increase in free radicals within the system during a prolonged reaction time, facilitating polymerization reactions. However, after a certain point, the monomer concentration gradually decreased, leading to a slower reaction rate and a reduced degree of polymerization, resulting in a decrease in the product's intrinsic viscosity.

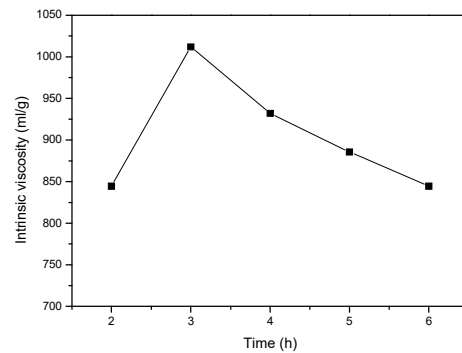


Figure 7. The influence of reaction time on intrinsic viscosity.

According to the test results, the optimum conditions for preparation of the PAC-PDMAAC hybrid flocculant were as follows: DMDAAC mass fraction of 30%; initiator mass fraction of 0.7%; reaction temperature of 60 °C; reaction time of 3 h. Three sets of parallel verification experiments were conducted under these experimental conditions, and the measured eigenviscosity values of the products were 912.85 mL/g, 937.35 mL/g, and 928.88 mL/g, respectively, with an average value of 926.36 mL/g.

3.2. Characterization

3.2.1. FTIR

Figure 8 displays the FTIR spectrum of the hybrid flocculant, revealing characteristic absorption peaks at wavenumbers 3020 cm^{-1} (C-H bond vibration), 1704 cm^{-1} (C=O bond vibration), 1471 cm^{-1} (C-H bond vibration), and 1408 cm^{-1} (C-N bond vibration). For the hybrid flocculant, there was no absorption peak in the range of 3500–3200 cm^{-1} , indicating that there was no hydroxyl group in the substance. Notably absent was any characteristic absorption peak corresponding to C=C double bonds within the spectrum's range from 1675 to 1500 cm^{-1} , indicating that the reactants undergo free-radical-initiated polymerization resulting in covalent bonding between the organic and inorganic components.

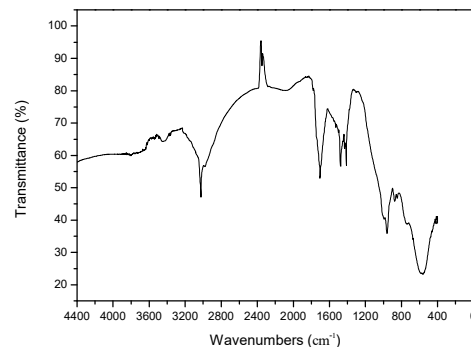


Figure 8. FTIR spectrum of hybrid flocculant.

3.2.2. SEM

An organic–inorganic composite flocculant is a simple superposition of organic matter and inorganic matter. In this study, a composite flocculant derived from mixing PAC and PDMAAC was compared to the PAC-PDMAAC hybrid flocculant prepared. The composite flocculant and the prepared hybrid flocculant were observed by SEM.

The SEM images of the PAC-PDMAAC composite flocculant (a) and the PAC-PDMAAC hybrid flocculant (b) are presented in Figure 9. It can be observed that the composite flocculant exhibited a relatively smooth surface with a blocky shape, featuring a few bulges that facilitate adsorption. On the other hand, the hybrid flocculant possessed a larger particle size and an irregular surface morphology with more convex folds, presenting an obvious spatial network structure. The increased surface area of the molecular particles

in the hybrid flocculant contributed to its enhanced adsorption capacity, while its spatial network structure made it have a superior bridging effect.

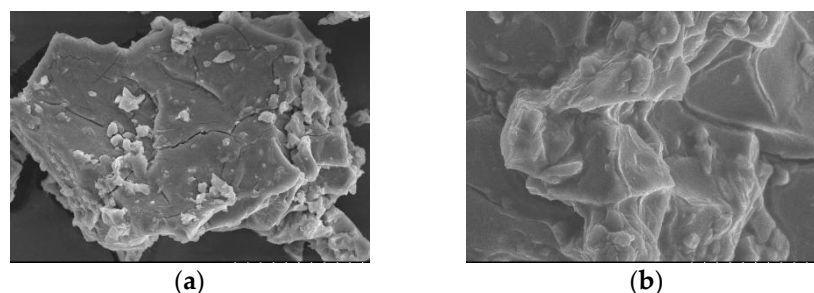


Figure 9. SEM pictures at 1.5k of PAC-PDMDAAC composite flocculant (a) and PAC-PDMDAAC hybrid flocculant (b).

3.2.3. Electrical Conductivity Test

Figure 10 illustrates the conductivity curve of the PAC-PDMDAAC hybrid flocculant and the composite flocculant as a function of concentration. The figure indicates that, within the experimental concentration range, the conductivity of the hybrid flocculant solution exhibited a strong linear correlation with the concentration. In contrast, the conductivity of the composite flocculant solution experienced a gradual decline following an abrupt increase during dilution, deviating from linearity. This behavior could be attributed to the fact that the composite flocculant was merely a physical mixture of two distinct flocculants without any chemical bonding interactions. In this solution, N^+-Cl^- ions in PDMDAAC become ionized. As dilution occurs, aluminum particles in polyaluminum chloride undergo multiple stages of hydrolysis, reaching maximum hydrolysis at a specific point in time, leading to a gradual decrease in electrical conductivity with decreasing concentration. Conversely, due to the covalent bond interactions that stabilize the modified PAC particles on PDMDAAC, no sudden increase was observed during dilution for the hybrid flocculant, thereby demonstrating consistent linear behavior.

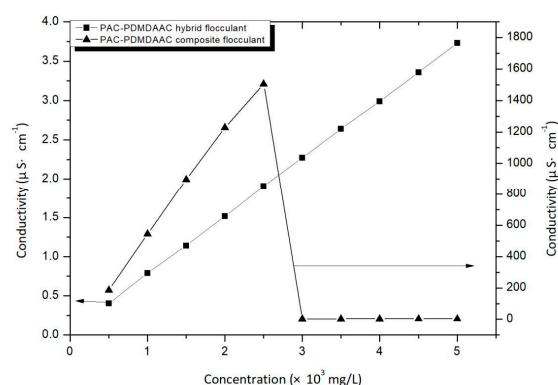


Figure 10. Variation in conductivity of PAC-PDMDAAC hybrid flocculant and PAC-PDMDAAC composite flocculant with concentration.

3.2.4. TGA

Figure 11 shows the TGA curves of both the PAC-PDMDAAC composite flocculant and PAC-PDMDAAC hybrid flocculant. Figure 9 provided the effect of temperature on product weight and the rate of weight change (dw/dt). Due to their inherent hygroscopicity, water molecules were evaporated during the heating process, as depicted in Figure 11, thus exhibiting a substantial weight loss prior to reaching temperatures above $150^\circ C$ due to water loss. As the temperature increased further, structural units within the flocculant molecules commenced decomposition, leading to bond breakage along molecular chains and a subsequent increase in weight loss until the complete decomposition

of organic components occurred. Notably, it can be observed in this figure that the maximum thermal decomposition rate for the hybrid flocculant was achieved at approximately 264.38 °C. However, the maximum thermal decomposition rate of the composite flocculant appeared at 256.36 °C. The TGA test report indicated an initial decomposition temperature of 230.29 °C for the PAC-PDMDAAC hybrid flocculant, which was significantly higher than that of 214.53 °C for the composite flocculant. The calculated half-life temperature for thermal decomposition of the hybrid flocculant was 671.93 °C, which was also higher than 621.29 °C for the composite flocculant. This may be because the organic and inorganic components of the hybrid flocculant were bonded in the form of covalent bonds, and the organic macromolecular chain end was fixed by polyaluminum chloride aluminum particles, which weakened the thermal motion of the macromolecular chain, thereby improving its heat resistance and increasing its thermal decomposition temperature.

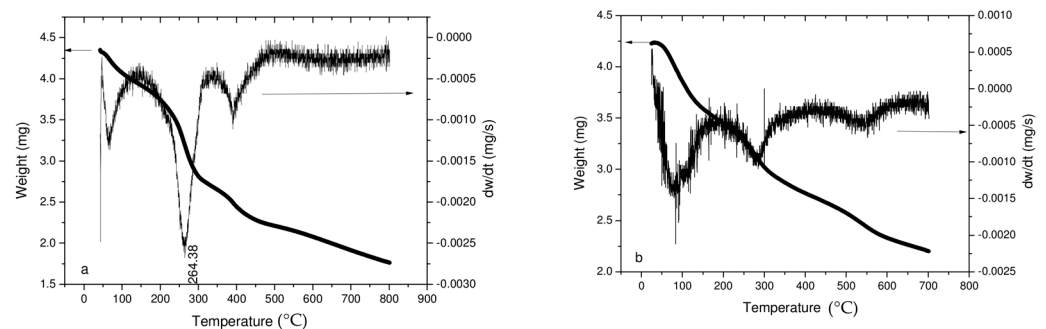


Figure 11. TGA curves of PAC-PDMDAAC hybrid flocculant (a) and PAC-PDMDAAC composite flocculant (b).

3.3. Flocculation Performance

The effect of any flocculant on the flocculation process is influenced by the dosages, with the flocculation efficiency decreasing at lower or higher dosages than the optimum dose. The flocculation decolorization rate can be used to measure the flocculation decolorization performance of polymer samples in dye wastewater. In this study, we determined the effect of the flocculant dosage on the decolorization rate using the hybrid flocculant and the composite flocculant, as shown in Figure 12.

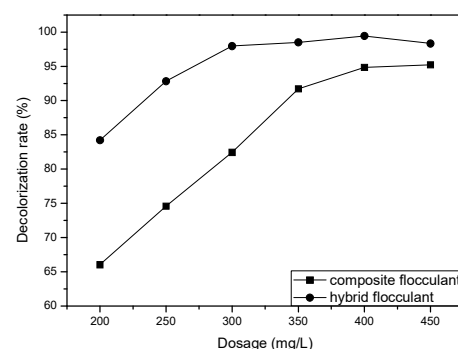


Figure 12. Effect of different flocculants' dosages in disperse-violet-dye wastewater on decolorization rate.

Figure 12 illustrates the decolorization rates of disperse-violet-dye wastewater with a concentration of 100 mg/L using both the hybrid flocculant and the composite flocculant. As depicted in the figure, an increase in dosage led to higher decolorization rates for both types of flocculants. However, a consistently superior performance was observed with the hybrid flocculant compared to the composite one. When dosed at 400 mg/L, a remarkable decolorization rate of 99% was achieved by the hybrid flocculant for disperse-violet-dye wastewater treatment. Disperse violet is a compound with the structure of anthraquinone,

which can only be evenly dispersed in water, so any treatment only needs to neutralize a small amount of negative charge on the surface of the dye, without the need to precipitate the dye from the water. Therefore, hybrid flocculants with long chain structures and high molecular weights are more effective in the treatment of disperse violet. This highlights that the synergistic effects between its inorganic and organic components were fully utilized by this hybrid flocculant, resulting in a better binding-bridging capacity than that exhibited by the composite counterpart.

4. Conclusions

Aluminum chloride-co-dienyldimethylammonium chloride complex flocculant (PAC-PDMAAC) was synthesized with dienyldimethylammonium chloride as a monomer, KH570 as a modifier, and ammonium persulfate as an initiator. The synthesis of the PAC-PDMAAC hybrid flocculant was conducted under the following preparation conditions: a mass fraction of 30% for monomer dimethyl diallyl ammonium chloride; a mass fraction of 0.7% for initiator; a reaction temperature of 60 °C; and a reaction time of 3 h. The results of FT-IR, the electrical conductivity test, and TGA demonstrated the covalent bonding and stability in properties between the organic and inorganic components of the PAC-PDMAAC hybrid product. SEM photographs revealed a loose spatial structure in the hybrid flocculant, facilitating its adsorption and bridging functions.

The decolorization rates observed in disperse-violet-dye wastewater indicated that the hybrid flocculant exhibited excellent decolorization effects by leveraging synergistic interactions between its organic and inorganic components.

Further work needs to be carried out to explore the chemical properties of the hybrid flocculant, encompassing its chemical composition, elemental composition, molecular structure, and other relevant aspects. In the realm of wastewater treatment, organic–inorganic hybrid flocculants are poised to emerge as a significant avenue for future development.

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