magnetochemistry

MDPI

Review

Research Progress of Magnetic Flocculation in Water Treatment

Zhihao Hu, Kun Wu, Zihan Wang, Kinjal J. Shah and Yongjun Sun *

College of Urban Construction, Nanjing Tech University, Nanjing 211816, China
* Correspondence: sunyongjun@njtech.edu.cn

Abstract: As people's material quality of life continues to improve, water resources become subjected to varying degrees of contamination. As one of the most commonly utilised agents in water treatment, a flocculant exhibits a diverse range of forms and a vast scope of applications. However, the application of flocculants gives rise to a series of issues, including the use of large doses, the formation of sludge, the difficulty of recycling flocculants, and other concerns. The development of new flocculation technology has become a crucial step in enhancing the purification of wastewater and reducing environmental pollution. Magnetic flocculation can be classified into two main categories: magnetic seeds flocculation and magnetic flocculation. This paper presents an overview of the factors influencing magnetic flocculation, including the type of magnetic seeds, magnetic seeds particle size, and other pertinent considerations. Furthermore, the classification of magnetic flocculants in the process of magnetic flocculation is discussed. This includes the types of magnetic flocculant, namely, inorganic composite magnetic flocculants, organic composite magnetic flocculants, and biological composite magnetic flocculants. Inorganic composite magnetic flocculants are inexpensive and simple to produce; however, their dosage is considerable, and the resulting floc is not tightly formed, which impairs the efficacy of flocculation. The use of organic composite magnetic flocculants requires a smaller dosage and exhibits a strong flocculating ability; however, it may possess toxic properties and potentially cause harm to the water body. The biological composite magnetic flocculant exhibits high efficiency and no pollution, yet it is subject to stringent environmental conditions, displays poor stability, and is applicable to a relatively limited range of treatment scenarios. Furthermore, the integration of magnetic flocculation technology with other techniques is classified and summarised in diverse contexts, and the prospective research focus and direction of magnetic flocculants are proposed.

Keywords: magnetic flocculation; water treatment; magnetic seed flocculation; magnetic flocculant; combination

check for updates

Citation: Hu, Z.; Wu, K.; Wang, Z.; Shah, K.J.; Sun, Y. Research Progress of Magnetic Flocculation in Water Treatment. *Magnetochemistry* **2024**, *10*, 56. https://doi.org/10.3390/magnetochemistry10080056

Academic Editor: Carlos J. Gómez García

Received: 2 July 2024
Revised: 28 July 2024
Accepted: 5 August 2024
Published: 7 August 2024

Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/).

1. Introduction

With the rapid development of society and the continuous growth of the world population, global water resources have become particularly valuable in recent years. The Earth is 70% covered by water, but only 2.5% is freshwater, and the freshwater resources available are extremely limited. It is estimated that almost four billion people are currently experiencing water scarcity globally, with this figure potentially exceeding two billion in developing countries by 2050 [1]. The discharge of untreated wastewater from human industrial activities into natural water bodies has a detrimental impact on the environment, affecting the availability of water for plants, wildlife, and humans alike [2,3]. The conventional techniques employed for the purification of water can be broadly categorised into three principal groups: physical, chemical, and biological. Physical methods, including filtration, air flotation [4], and sand sedimentation, can be employed to target insoluble or suspended substances in water. Chemical methods, including coagulation and flocculation [5], oxidation [6], and ion exchange [7], have the potential to transform dissolved or insoluble hazardous substances into non-toxic or low-toxicity substances. The biological methods that are currently in use can be divided into two main categories: the activated

sludge method and the biofilm method. In these methods, green microorganisms are used to convert large organic molecules into smaller molecules, which can then be treated further to achieve the desired effect of water purification. While these methods can remove pollutants from water to a certain extent, they often suffer from a number of issues, including low treatment efficiency, high energy consumption, and high treatment costs.

Coagulation and flocculation represent one of the most critical and widely used surface water treatment processes for the removal of colloidal particles, phosphorus, and organic matter. The principal mechanisms of flocculation include compression of the bilayer [8], electro-neutralisation, adsorption and bridging, and netting and sweeping [9]. Flocculants act on suspended and dissolved substances in wastewater, destabilising the original colloidal particles to form flocs. These are then transformed into larger flocs through net sweeping, adsorption bridging, and so forth. Finally, the flocs are formed into sludge through sedimentation, which effectively reduces suspended solids, chromaticity, turbidity, and other indexes in the water. In order to optimise the efficiency of the treatment process, scientists have developed a range of different flocculants, including inorganic flocculants (such as aluminium salts, iron salts [10], and silicates [11]), organic flocculants (such as cationic polyacrylamide (PAM) and anionic PAM), and natural flocculants (such as humic acid and starch). Furthermore, a variety of flocculants have been combined in order to address the limitations of a single flocculant and to enhance the overall effectiveness of the treatment process. Nevertheless, it is unable to fulfil the requisite treatment requirements due to its inadequate treatment effect.

In recent years, with the continuous progress of science and technology, new coagulation and flocculation water treatment methods have emerged, among which magnetic flocculation technology has received widespread attention as a highly efficient and environmentally friendly means of water treatment. Based on magnetic seeds, magnetic flocculation enhances particle formation and accelerates the sedimentation process. Compared with conventional coagulation (CF), magnetic flocculation has the advantages of a short process, energy saving, and low cost. Magnetic flocculation can be divided into two main categories based on the method used: magnetic seed flocculation and magnetic flocculation. Figure 1a shows the magnetic seeds flocculation, which indicates that the magnetic seed and flocculant are added into the water body respectively, and the pollutants are magnetically inoculated by adsorbing the magnetic seed itself, and then the suspended particles in the water body are settled by using the effect of the applied magnetic field as well as the flocculant, so that the water body is purified. Magnetic nanoparticles can be introduced into the water body as magnetic seeds. Fe_2O_3 and Fe_3O_4 are typical magnetic nanomaterials with low toxicity, high magnetic properties, etc., and their porous structure can determine the ability to adsorb pollutants. The magnetic flocculation in Figure 1b focuses on the development of a composite magnetic flocculant with a synergistic flocculation effect, in which magnetic species are added in the preparation of the flocculant, which combine to form a modified magnetic flocculant, thereby enhancing the adsorption and settling of contaminants and separating contaminants for the purpose of purification. Magnetic particles can act as nuclei to promote the formation of settleable flocs of greater density, size, and strength [12]. The formation of larger magnetic particles—Al flocs—can be effectively removed under an applied magnetic field. The preparation of magnetic flocculants is similar to that of ordinary flocculants, using inorganic substances such as aluminium and iron salts and organic substances such as chitosan and moringa seeds. Various polymers such as polyacrylamide (PAM) [13], chitosan [14], starch [15], and alginate [16] have also been successfully grafted onto MPs and have shown high pollutant removal rates. This paper classifies magnetic flocculants into inorganic composite magnetic flocculants, organic composite magnetic flocculants, biocomposite magnetic flocculants, and other composite magnetic flocculants, and it compares their advantages and disadvantages, after first introducing the factors of external force influence on magnetic species in magnetic flocculation: force, particle size, type, and other factors. On this basis, the scenarios of magnetic flocculation in combination with other technologies are presented, and future

research priorities and directions for magnetic flocculants are suggested to provide new ideas for urban wastewater treatment. This study provides researchers with important information on the latest developments in their field of study.

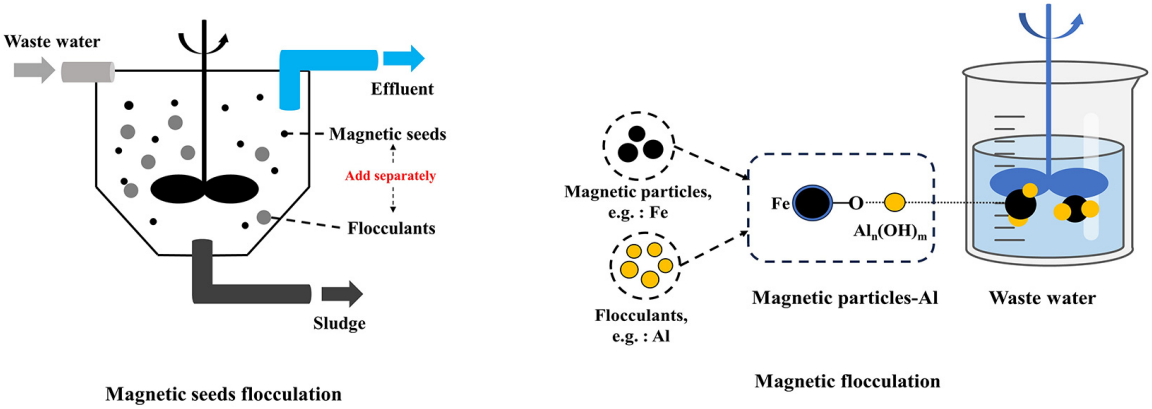


Figure 1. (**a**) Magnetic seeds flocculation; (**b**) magnetic flocculation.

2. Factors Affecting Magnetic Flocculation

2.1. External Forces on Magnetic Seed Flocculation

The influencing factors of magnetic flocculation are shown in Figure 2. The force distribution of magnetic seeds during the magnetic flocculation process is one of the key factors affecting their performance. In a magnetic field, the forces acting on magnetic particles mainly include magnetic force, gravity, hydrodynamic drag, and the interaction forces between particles [17]. Magnetic force is the force exerted by an external magnetic field on a magnetic seed, and its magnitude depends on the magnetization intensity and shape of the magnetic seed; it is usually proportional to the particle size of the magnetic seed. The influence of gravity on the volume of magnetic particles is particularly important for larger particle sizes. The hydrodynamic resistance is related to the motion state of the magnetic seed in the fluid and the flow velocity of the fluid, which affects the settling speed and efficiency of the magnetic seed. In magnetic flocculation, the magnetization of the magnetic nuclei is the basis for their functionality. The magnetization state of a magnetic seed determines its orientation and ability to migrate in the magnetic field and, thereby, influences its binding efficiency towards pollutants. The balance of forces of magnetic seeds in the magnetic field determines their trajectory in the water treatment plant, which, in turn, influences the formation and settlement of flocks. For example, when the magnetic force is greater than the hydrodynamic resistance, the magnetic seed moves quickly along the magnetic field line, which contributes to rapid flocks formation. On the contrary, if the dynamic resistance of the liquid predominates, the speed of movement of the magnetic seed slows down, which can lead to a reduction in the flocculation efficiency. In addition, the interaction forces between magnetic species, such as van der Waals forces and electrostatic forces, also play an important role in the flocculation process. These forces promote or inhibit the aggregation of magnetic species and, thereby, affect the stability and settling performance of the flocks. In practical applications, by adjusting the magnetic field strength and optimising the physical and chemical properties of magnetic seeds, the effect of these forces can be effectively controlled to achieve the best flocculation effect.

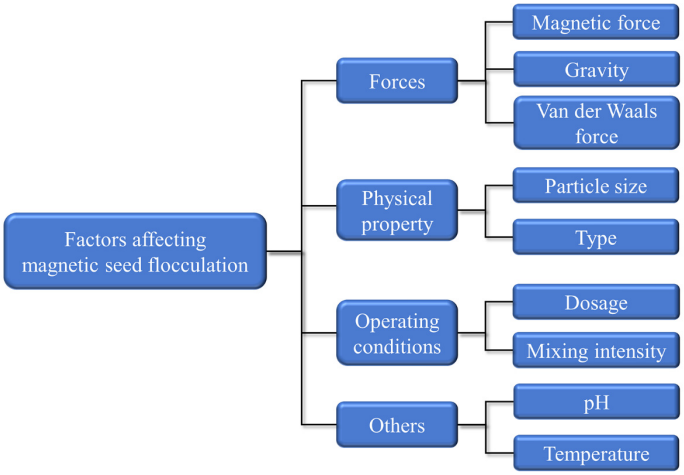


Figure 2. Factors affecting magnetic flocculation.

2.2. Magnetic Seed Particle Size

The particle size of magnetic seeds has a significant impact on their performance in the magnetic flocculation process. The particle size determines the specific surface area, the magnetization, and the settling behaviour of the magnetic nucleus in the liquid. A smaller particle size means a larger specific surface area, which has a positive effect on the probability of contact between magnetic particles and pollutants in water, thereby improving the adsorption efficiency of pollutants. However, small particle sizes can lead to increased Brownian motion of the magnetic seed in the water, which can reduce the stability of the flocs and make it difficult for the magnetic seed to aggregate into larger flocks, affecting sedimentation efficiency. Although a larger particle size of the magnetic seed can reduce Brownian motion and improve the stability of flocs, an excessively large particle size can increase the gravity of the magnetic seed, resulting in rapid settling and shortening the contact time with pollutants, thereby reducing the removal efficiency. Additionally, magnetic particles with larger particle sizes can increase the risk of system blockage and affect the long-term operational stability of processing equipment. In practical applications, magnetic nuclei with a particle size in the range of 50–200 nanometers are usually chosen. Magnetic seeds in this range can maintain good magnetic responsiveness and provide sufficient specific surface area to achieve efficient pollutant adsorption. By optimising the particle size distribution of magnetic nuclei, the removal efficiency and settling performance of magnetic flocculation technology can be significantly improved. However, some studies have shown that larger particle sizes actually have a better impact on separation efficiency. In a pioneering study, Seo et al. [18] employed (3-aminopropyl) triethoxysilane (APTES) to impart a positive charge to the initially negatively charged $BaFe_{12}O_{19}$ magnetic particles. These particles were subsequently isolated from Chlorella (Chlorella sp.). It was observed that the recovery of the $BaFe_{12}O_{19}$ magnetic particles was significantly influenced by their particle size. The shedding efficiency increased from 12.5% to 85% when the particle size was increased from 108 nm to 1.17 µm. The greater specific surface area of smaller magnetic particles allows for a greater number of contact points with microalgae, resulting in the formation of stronger electrostatic bonds. This increases the difficulty of dislodging smaller particles. Conversely, as particle size increases, the spatial arrangement and interactions of larger particles within aggregates lead to an increase in dislodgement efficiency. The enhanced dislodgement efficiency enables the efficient harvesting of chlorella and the recovery and reuse of magnetic particles.

2.3. Magnetic Species

There are different types of magnetic species, including natural magnetic minerals and artificially synthesised magnetic materials. The most commonly used magnetic materials include Fe_3O_4 (magnetite) [19], Fe_2O_3 (hematite) [20], and various ferromagnetic

alloys. Different magnetic types have different magnetic properties, such as magnetization, coercivity, and stability, which directly affect the effectiveness of magnetic flocculation. For example, Fe_3O_4 has become a commonly used magnetic seed material in magnetic flocculation due to its high saturation magnetization and good chemical stability [21]. As shown in Figure 3, the mixed system is Fe_3O_4 and polymeric aluminium chloride (PAC). The magnetic seeds coated with PAC form Fe-O-Al bonds, which creates stronger structures and larger specific surface areas and strengthen the connection between the magnetic seeds and the hydrolysates of PAC [22]. In addition, Fe_2O_3 has good biocompatibility and a low cost, and it is also widely used in water treatment. Ferromagnetic alloys are used because of their special physical and chemical properties, such as high coercivity and high magnetic permeability, used in certain water treatment environments. In addition to traditional magnetic materials, researchers are also exploring new magnetic types such as magnetic nanoparticles, magnetic biochar [23], and magnetically modified natural minerals. These new magnetic species can improve their adsorption performance and selectivity and improve the removal efficiency of specific pollutants through surface modification or composite with other materials. When selecting magnetic seeds, their interaction with target pollutants, cost–benefit ratio, and environmental impact must be taken into account. For example, for waters contaminated with heavy metals, it may be necessary to choose magnetic seeds with a high adsorption capacity and good stability. To remove organic pollutants, it may be necessary to select magnetic adsorption materials with specific functional groups. By optimising the types and properties of magnetic seeds, the adaptability and treatment efficiency of magnetic flocculation technology can be improved.

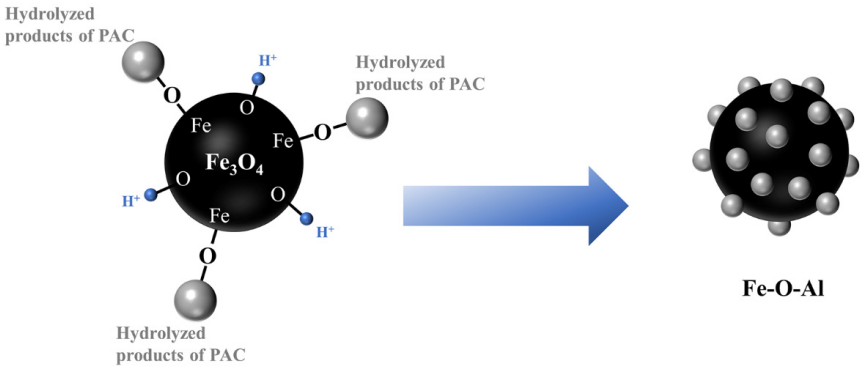


Figure 3. Mixed system of Fe_3O_4 and PAC.

2.4. Other Influencing Factors

In addition to the force, particle size, and type of magnetic nuclei, there are also numerous other factors that influence the magnetic flocculation process, such as the amount of magnetic nuclei added, the stirring intensity, etc. [24]. These factors together determine the effectiveness and efficiency of magnetic flocculation. The dosage of magnetic seeds is one of the important factors affecting the magnetic flocculation effect. The dosage amount has a direct influence on the formation speed, size, and settling performance of the magnetic flocks. Insufficient dosage may result in insufficient contact between pollutants in the water and magnetic seeds, thereby reducing the efficiency of pollutant removal. On the contrary, excessive addition of magnetic nuclei can lead to the magnetic flocks becoming too large and loose, the settling time being extended, and difficulties occurring in subsequent magnetic separation processes due to the excessive amount of magnetic nuclei themselves. In addition to the aforementioned dosage, an excessively large quantity may result in flocculation, loose disintegration, and separation difficulties. However, it is also imperative to consider the dissolution of the magnetic species, such as Fe_3O_4 magnetic species. For instance, the 'Surface Water Environmental Quality Standards' (GB3838-2002) [25] stipulates that the iron content in water environments should not exceed 0.3 mg/L for magnetic flocculation. In the event of dissolution, the surrounding

environment will be polluted, and the magnetic particles will become unusable. In practical applications, magnetic seed dosage needs to be optimised based on specific water quality conditions and treatment goals. In addition, the dosage of magnetic seeds is also affected by factors such as the design of the magnetic flocculation system and the efficiency of the magnetic separation equipment. Stirring intensity is another operating parameter that significantly influences the magnetic flocculation effect [26]. The stirring intensity determines the degree of uniformity of mixing magnetic seeds with pollutants in the water and thereby influences the formation of flocks and the efficiency of magnetic flocculation. Appropriate stirring intensity can promote complete contact between magnetic seeds and pollutants, accelerate the formation of flocs, and improve the sedimentation rate and water purification degree. However, excessive stirring intensity can cause the flocks formed to be dispersed, their stability to decrease, and, thus, the settling effect to be impaired. If the stirring intensity is too low, this can lead to insufficient mixing of magnetic seeds and pollutants and thus impair the flocculation effect. Therefore, it is necessary to determine the optimal stirring intensity through experiments based on specific treatment systems and water quality conditions.

3. Classification of Magnetic Flocculants

3.1. Inorganic Composite Magnetic Flocculant

Inorganic composite magnetic flocculants are obtained by compounding and reacting inorganic flocculants or flocculant materials with magnetic materials. Table 1 illustrates the complexing of inorganic compounds, namely, sodium alginate, iron, aluminium, and silicon, with magnetic particles. The majority of inorganic magnetic flocculants exhibit a removal rate exceeding 90% for turbidity, while demonstrating a comparatively lower removal rate for COD. Zhao et al. [27] employed a magnetic seed comprising natural magnetite coated with polymeric aluminium chloride (PAC) and a coating polymer, polyacrylamide (PAM), to facilitate the removal of a microalgae species, Chlorella vulgaris. The process was completed in less than 0.5 min, with 99% of the microalgae harvested. In comparison to the removal of microalgae with only Fe_3O_4 (pH: 7.0), a broader pH range was attained, with effective microalgae harvesting at pH 6.04 to 10.1 and a more extensive inclusivity of algal organisms. Ding et al. [28] prepared a polysilicate containing an iron borate and zinc sulphate (PSBFZ) flocculant and a new magnetic flocculant PSBFZ-Fe_3O_4 for the treatment of aqueous ink wastewater. The turbidity removal and decolourisation rate of PSBFZ-Fe_3O_4 at room temperature can reach 96% and 97%, respectively, which represents an increase of 3% and 2% in comparison to the removal rates achieved by PSBFZ alone. Furthermore, the flocculation waste can be recycled on two occasions through the utilisation of a magnetic recovery process, which can be readily separated from the wastewater. This facilitates the potential reuse of the magnetic flocculant. The magnetically recycled PSBFZ-Fe_3O_4 flocculant can be reused by adding a small amount of PSBFZ, and its removal rate of turbidity and chromaticity of wastewater can still reach more than 95%. However, it is not an effective method for degrading COD; thus, it requires combination with a variety of treatment techniques, including physical, chemical, and biological methods, in order to achieve a more comprehensive pollutant removal. Dai et al. [29] combined the environmental properties of calcium-based flocculants with the convenience of magnetic flocculation through a simple composite method to prepare $Ca(OH)_2/Fe_3O_4$ magnetic flocculants for efficient flocculation of black liquor lignin in papermaking. This flocculant can be used directly in aqueous sodium lignin solution without pH neutralisation and maintains a high lignin removal rate of 91% to 95% in the pH range of 9 to 12.5. In addition, the flocculant has a high tolerance to salts (0–1 mol/L NaCl) and pH values (9–12.5). Due to the magnetic properties of Fe_3O_4, the flocculant $Ca(OH)_2/Fe_3O_4$ can aggregate quickly under the influence of an external magnetic field. This magnetically controlled aggregation facilitates the contact and binding of lignin molecules with flocculants, thereby improving flocculation efficiency. After the flocculation process is complete, the flocks formed can be quickly separated by an external magnetic field. Due to the superparamagnetic property of

$Ca(OH)_2/Fe_3O_4$ flocculants, the flocs settle quickly under the effect of a magnetic field, making the separation process between flocks and aqueous solutions more efficient and faster. Inorganic composite flocculants mainly rely on van der Waals forces due to the interaction between their inorganic components and magnetic particles, which are relatively weak and not as strong as chemical bonds. This means that the flocs formed during the flocculation process may not be stable enough and may easily separate from the magnetic particles during the treatment process, weakening the magnetic responsiveness of the magnetic flocculant, affecting the flocculation effect and subsequent magnetic separation efficiency.

Table 1. Research progress on inorganic composite magnetic flocculants.

Number	Magnetic Flocculant	Removing Substances	Flocculation Conditions	Removal Effect	References
1	Magnetic ferric oxide poly (aluminium titanium silicate)–sodium alginate (SA) composite flocculant (Fe_3O_4–PSAT–SA)	Congo red (Congo red)	Dosage: 40 mg/L pH: 5 Flocculation time: 10 min Settling time: 10 min	95.43%	[30]
2	Fly ash magnetic coagulant (FAMC)	Fine mud full tailings	Magnetic field intensity: 0.3 T Dosage: 30 g/t Feeding speed: 0.6 t/(m^2·h)	•Reduce flocculant dosage by 50%; •The processing capacity per unit area (HCPU) increased by approximately 20%.	[31]
3	Magnetic coagulant (FMC)	Accelerate electron negative sludge and ultrafine tailings (ESUT)	Tailings concentration of 20%, tailings pH value of 7, magnetic field strength of 0.24 T, FMC dosage of 215 mL/t, NPAM dosage of 38 g/t, NPAM concentration of 0.2%, temperature of 25 °C	•Increase in settlement height (SH): increased by 65%, reaching 181.3 mm; •The solid content in tailings slurry can be reduced by 80%; •Under optimal conditions, the amount of non-ionic polyacrylamide (NPAM) can be reduced by 37%.	[32]
4	Magnetite nanoparticle/inorganic flocculant (MNP/IF)	Suspended soil particles; Sr^{2+}	MNP/IF: 0.3 g/g; Stir at 300 rpm for 30 s, then let it stand for 10 min	The turbidity removal rate of suspended particles is 82.5%, and the Sr^{2+} adsorption capacity reaches 163.6 mg/g.	[33]
5	Organic silicon iron hybrid flocculant	Compounds with turbidity and UV 254 absorption	First add magnetic powder, then add flocculant. The optimal particle size of magnetic powder is 20–30 microns, with a dosage of 300 mg/L	The turbidity and UV 254 removal rates reached 99.26% and 84.03%, respectively	[34]
6	Magnetic composite flocculant nanoFe_3O_4 and polyaluminium chloride (MFPAC)	Chemical oxygen demand (COD) and chromaticity	300 rpm rapid stirring; 70 rpm slow stirring; the dose of MFPAC reaches 1.5 g/L	The maximum COD removal rate reached 59%, and the colour removal rate reached 67%	[35]

3.2. Organic Composite Magnetic Flocculant

Composite organic flocculants are prepared by crosslinking and polymerizing high-molecular-weight organic compounds, such as chitosan, starch, cellulose, polyacrylamide,

etc., with magnetic materials. They combine the flocculation properties of organic polymers and the magnetic properties of magnetic nanoparticles to efficiently remove pollutants from water. They have a high intrinsic viscosity and good flocculation properties. Examples of organic compounds that can be used for compounding with magnetic particles include chitosan, moringa seeds, and other seeds, as illustrated in Table 2. Hena et al. [36] first dispersed Fe_3O_4 nanoparticles in deionized water and achieved better dispersion by ultrasonic treatment. Then, $FeCl_3$ was added as an oxidizing agent to the solution and stirred continuously at room temperature. Next, pyrrole monomer (Py) was added and the reaction mixture was stirred at room temperature for 2.5 h. Finally, the reaction was stopped by adding acetone, and the resulting nanocomposite material was purified by filtration and washing. The flocculant achieved a recovery efficiency of nearly 99%, 92.4%, and 90.8% for the microalgae Botryococcus braunii, Chlorella protothecides, and Chlorella vulgaris, respectively. In the study by Xie et al. [37], self-made magnetic Fe_3O_4, self-made magnetic sodium silicate, $Al_2(SO_4)_3$, $MgSO_4$, and starch were used to prepare magnetic organic loaded Fe_3O_4-PSAM St flocculants through a series of chemical reactions and treatment steps. The experimental results showed that under the optimal conditions (Fe_3O_4-PSAM St addition amount of 40 mg/L, simulated wastewater pH of 5–12, flocculation time of 10 min, settling time of 10 min), the highest removal rate of Congo red could be achieved (95.0%). After six repeated flocculation trials using 50% (*w*/*w*) magnetic flocs and 50% (*w*/*w*) Fe_3O_4-PSAM St as flocculants, the removal rate of Congo red still reached about 93%, showing good reusability and stability. Reck et al. [38] employed the natural material moringa seed protein in conjunction with iron oxide nanoparticle technology for the treatment of aqueous solutions containing synthetic dyes. The application of a magnetic field during the precipitation process resulted in a notable acceleration of the precipitation rate and an enhanced removal efficiency of the dyes. For instance, the removal of cochineal dye increased from 45% to 86% with the utilization of a magnetic field, while the removal of sunset yellow increased from 15% to 69%. Furthermore, the removal of reactive black 5 and bright blue achieved 94% and 52%, respectively. Furthermore, the article indicated that the optimal protein concentration differed for various dyes, with 150 mg/L for cochineal and sunset yellow, 115 mg/L for reactive black 5, and 75 mg/L for brilliant blue. This approach not only eliminates hazardous dyes from industrial wastewater but also enhances the removal efficiency by optimising the flocculation conditions (e.g., protein concentration, pH, and application of the magnetic field). There are various types of organic polymer compounds, and the properties of flocculants can be customized through various synthesis methods and modification techniques to meet the needs of different wastewater treatments. Due to the stronger bonding effects such as coordination bonds that can form between organic and inorganic polymer components, magnetic particles are less likely to detach from the flocks during the flocculation process, improving the stability of the flocks. But the high costs are also a factor to consider.

Table 2. Research progress of organic composite magnetic flocculants.

Number	Magnetic Flocculant	Removing Substances	Flocculation Conditions	Removal Effect	References
1	Magnetic flocculant γ-Fe_2O_3 nanoparticles and moringa oleifera seed extract (oil and non-oil) (γ-Fe_2O_3-Mo and γ-Fe_2O_3-MO(et))	*Escherichia coli*	120 rpm rapid stirring 15 rpm slow stirring 30 min settling Apply an external magnetic field (260 A/m, i.e., 260 amperes per meter)	•Original water sample: complete inactivation of *Escherichia coli*; •In the synthesised water sample, the large intestine rods of γ—Fe_2O_3-MO and γ—Fe_2O_3-MO (et) reached 88% and 93%, respectively.	[39]

Table 2. *Cont.*

Number	Magnetic Flocculant	Removing Substances	Flocculation Conditions	Removal Effect	References
2	Carboxymethyl chitosan magnetic flocculant (MC), carboxymethyl chitosan polyacrylamide magnetic flocculant (MC-g-PAM), carboxymethyl chitosan polyacrylamide 2-acrylamide-2-methylpropanesulfonic acid magnetic flocculant (MC-g-PAA)	Copper (II) containing wastewater	Using a flocculant dosage of 80 mg/L, pH value of 6, reaction time of 1.5 h, G value (stirring intensity) of 200 s^{-1}, and magnetic field intensity of 120 mT	•Under optimal flocculation conditions, the removal rates of copper ions by MC, MC-g-PAM, and MC-g-PAA were 93.39%, 88.64%, and 61.41%, respectively; •MC-g-PAA has good recyclability, with a recovery rate of 77.24% and a removal rate of 67.53% for Cu (II) after five uses.	[40]
3	Magnetic flocculant: Fe_3O_4-ZnO nanocomposites coated with stearic acid (SA)	Algal biomass, especially Scenedesmus dimorphus algae cells	Fe_3O_4-ZnO nanocomposites coated with SA, with a loading rate of 0.3 g-MNPs·g-algae^{-1}: the initial algae concentration is 0.8 g L^{-1}; mixing for 2 min and magnetic separation for 3 min; UV365 irradiation and mechanical stirring or mixing are used to increase the UV exposure of magnetic nanocomposite particles.	Under the combined action of ultraviolet light irradiation and mechanical stirring, algae gradually migrate from the aggregated suspension to the main body of the suspension.	[41]
4	Magnetic flocculant $CoFe_2O_4$/chitosan magnetic composite material	Indigo Blue Dye (IBD)	pH: 3.0: $CoFe_2O_4$/chitosan: 0.75 g/L	The time for adsorption to reach equilibrium is very short, only 15 min. Under these conditions, the maximum adsorption capacity reached 380.88 mg/g.	[42]
5	Magnetic flocculant: magnetic oxide nanoparticles combined with moringa oleifera seed extract	Compound with apparent colour, turbidity, and UV254 nm absorption	10 mg/L γ-30 min settling time of Fe_2O_3 and 400 mg/L prickly orange seed extract (MoFe-G combination)	Effectively remove over 90% turbidity, 85% apparent colour, and 50% absorption of UV254 nm compounds.	[43]

3.3. Biological Composite Magnetic Flocculant

A biological compound magnetic flocculant is a special type of flocculant that combines biologically generated flocculants with magnetic materials. This flocculant combines biopolymers produced by organisms with magnetic particles to enhance the flocculation effect and facilitate magnetic separation. Guo et al. [44] used chemical reagents such as chitosan, isopropanol, chloroacetic acid, and activated sludge as microbial flocculant MBFX-8 in their study. Magnetic properties were prepared by chemical modification and a co-precipitation method. Fe_3O_4@carboxymethyl chitosan (CMC) nanoparticles were

prepared, and MBFX-8 was prepared through freezing centrifugation and other steps. The production of composite flocculants involved the combination of MBFX-8 with magnetism Fe_3O_4@CMC. The mixture was performed according to different mass ratios, and stirring and static reactions were carried out. The results showed that under certain conditions (pH 6.5, flocculant dosage 5 g/L, standing reaction time 3 h), the removal rate of Cu^{2+} by the composite flocculant reached 98.9%. Yu et al. [45] synthesised the composite cellulose@iron oxide nanoparticles containing magnetic nanoparticles and cellulose by a one-step co-precipitation method. The cellulose was dissolved using an aqueous NaOH–thiourea–urea solution. The NaOH was used as a precipitant for iron oxide nanoparticles in the cellulose solution. Additionally, low-cost cellulose was used as a template to promote the growth of the nanoparticles in the cellulose matrix. The results of Langmuir adsorption isotherm modelling demonstrated that the adsorption capacities of the complexes for arsenite and arsenate were 23.16 and 32.11 mg/g, respectively. The Fe_2O_3 nanoparticles were uniformly dispersed in the cellulose matrix. The composite exhibited a sensitive magnetic response and superparamagnetic behaviour, which enabled its easy separation from the aqueous solution by an external magnetic field. This property is of great significance for flocculant recycling and reuse in practical water treatment processes. In a study conducted by Periyasamy et al. [46], environmentally friendly materials, including cellulose (Cel), hydrotalcite (HT), and hydroxyapatite (HAp), were used to prepare cellulose/hydrotalcite (Fe_3O_4@CelHT) and cellulose/hydroxyapatite (Fe_3O_4@CelHAp) for the removal of Cr(VI). The biocomposite magnetic flocculant combines the rapid settling properties of magnetic materials with the flocculation effect of biopolymers while being biodegradable and environmentally friendly. Due to these properties, biocomposite magnetic flocculants have enormous potential and research value in the field of water treatment. Due to its biodegradability, the impact of biomagnetic flocculants on the environment is relatively low. However, stability problems may arise with biological magnetic flocculants under certain conditions, such as pH changes or reduced flocculation efficiency in high-temperature environments. Compared to inorganic magnetic flocculants, the manufacturing process of biological magnetic flocculants may be more complex and require precise biotechnological operations.

3.4. Advantages and Disadvantages of Composite Magnetic Flocculants

Magnetic flocculants are classified into three categories based on their composition, namely, inorganic, organic, and biological. Each category exhibits distinct characteristics. Inorganic composite magnetic flocculants are renowned for their affordability, yet they may exhibit inadequate biocompatibility and an elevated risk of environmental accumulation. Organic composite magnetic flocculants are diverse and adjustable, and they can be chemically structured to meet specific needs. However, they are more costly and complex to synthesise, and excessive amounts of some organics may be harmful to water bodies [47]. Biocomposite magnetic flocculants are distinguished by their exemplary environmental compatibility and biocompatibility, derived from renewable resources and readily biodegradable. However, they are associated with elevated production costs and process complexity, as well as more rigorous environmental standards. In practice, the combination of different types allows for the optimisation of the overall performance of the flocculant, thereby achieving more efficient water treatment. Specific advantages and disadvantages are shown in Table 3.

Table 3. Advantages and disadvantages of inorganic, organic, and biological magnetic flocculants.

Classification	Advantage	Disadvantage	References
Inorganic composite magnetic flocculant	•The price is cheap and the cost is low; •Wide application range and strong adaptability, including turbidity and dye wastewater;	•Secondary pollution issues; •The stability issue of inorganic magnetic flocculants requires further optimisation.	[48]
Organic composite magnetic flocculant	•Diversity: there are a wide variety of organic polymer compounds that can meet various wastewater treatment requirements; •The floc has better stability; •Low dosage and strong flocculation ability.	•Higher costs; •The synthesis process of organic magnetic flocculants is complex; •Flocculants may be toxic and cause pollution to water bodies.	[49]
Biological composite magnetic flocculant	•The raw material is natural biopolymers, which have less impact on the environment compared to inorganic and organic composite flocculants; •Good biocompatibility; •Good renewability.	•High environmental requirements, requiring appropriate pH and water temperature factors; •The preparation process is more complex, and the learning and application costs are higher.	[50]

4. Magnetic Flocculation Combined with Other Technologies

Magnetic flocculation technology, as an efficient physical and chemical water treatment method, is based on utilizing the magnetic properties of magnetic particles to enhance the flocculation process, thereby improving the removal efficiency of suspended solids, colloidal particles, and certain dissolved pollutants in water. This technology promotes the coagulation and sedimentation of pollutants by adding magnetic nuclei or magnetic flocculants into the treatment system, thus achieving rapid separation. Magnetic flocculation technology has been widely studied and used in the field of water treatment due to its high efficiency, low cost, and easy operation. In order to further improve the efficiency and adaptability of treatment, magnetic flocculation technology has been widely combined with other water treatment technologies in recent years, creating various comprehensive treatment processes. The majority of the co-located technologies presented in Table 4 are employed in the deep treatment stage, or in the pre-treatment stage, with varying degrees of phosphorus removal in both domestic wastewater and industrial wastewater.

In a study conducted by Liu et al. [51], a magnetic composite flocculant (MFPFS) was used, which was compounded with Fe_3O_4 nanoparticles and polymeric ferric sulphate (PFS). The results demonstrated that the flocculant was capable of achieving COD and chromaticity removals of up to 60% and 80%, respectively. Furthermore, the combination with sulphate radical oxidation technology effectively removed the residual pollutants following flocculation of MFPFS through Fe^{2+}-activated sulphate radical oxidation, significantly enhancing the removal rates of COD and chroma, which could be further increased to 75% and 95%, respectively. The efficacy of sulphate radical oxidation in treating recalcitrant organics was demonstrated. This combined approach not only enhanced the removal of organic matter but also improved the overall water treatment performance. Wang et al. [52] employed magnetic flocculation with response surface methodology (RSM) and artificial neural network (ANN) to optimise the magnetic flocculation process for the treatment of secondary effluent from municipal wastewater plants. Among the methodologies employed, Response Surface Methodology (RSM) was utilised, specifically the Box–Behnken Design (BBD), for the design of experiments. The concentrations of polymeric aluminium chloride (PAC), magnetic powder (Fe_3O_4), and flocculant (PAM) were selected as the controllable variables, with total phosphorus (TP) removal and turbidity removal employed as the responses to determine the optimal combination of the concentrations of PAC, magnetic powder, and PAM by RSM. A three-layer BP-ANN model was constructed using MATLAB

software and the back-propagation (BP) algorithm for the artificial neural network (ANN). The input layer comprised the concentrations of PAC, magnetic powder and PAM, while the output layer represented the removal rate of TP and turbidity. The TP and turbidity removal rates obtained experimentally under the optimal conditions determined by RSM were 88.79% ± 5.45% and 63.48% ± 9.60%, respectively. The combination of multiple models may prove a superior approach to the adaptation of the process to the inevitable variation of variables that will occur in the actual water treatment process.

Table 4. The application of magnetic flocculation in combination with other technologies.

Number	Combination Technology	Application Scenarios	Processing Effect	References
1	Carbon loading + magnetic coagulation for efficient precipitation	Printing and dyeing wastewater treatment	All indicators meet the requirements of the direct discharge standard of the Water Pollutant Discharge Standard for Textile Dyeing and Finishing Industry (GB 4287-2012) [53].	[54]
2	Improved Bardenpho bioreactor + magnetic coagulation high-efficiency sedimentation tank + denitrification deep bed filter process	Mainly domestic sewage	Magnetic coagulation enhances chemical phosphorus removal, resulting in effluent TP $\leq$ 0.4 mg/L	[55]
3	Moving Bed Biofilm Reactor (MBBR) + magnetic coagulation sedimentation tank + denitrification deep bed filter tank	Domestic sewage and some industrial wastewater	Make the effluent COD BOD_5; the ammonia nitrogen and total phosphorus indicators stably reached the Class IV standard of the Surface Water Environmental Quality Standard (GB3838-2002), reducing electricity consumption by about 7.8% and drug consumption by about 9%, saving operating costs	[56]
4	Improved Anaerobic-Anoxic-Oxic (AAO) magnetic coagulation precipitation ozone-activated carbon	Mainly industrial wastewater, accounting for over 60%	Magnetic particle removal of TP and SS achieved removal rates of 92.4% and 96.1%, respectively	[57]
5	Magnetic flocculation + ultrafiltration	High turbidity concentrated water	The turbidity is below 0.3 NTU, and the turbidity removal rate reaches 99.9%; the COD removal rate is 77.9%, and the UV254 removal rate is 32.4%	[58]

5. Conclusions and Perspectives

This article discusses the application and potential of magnetic flocculation technology in the field of water treatment. Magnetic flocculation in the field of water treatment can be divided into magnetic seed flocculation and magnetic flocculation. Magnetic seed particles are used to promote flocculation. Factors affecting magnetic seed flocculation include force, particle size, type, dosage, and agitation speed. The flocculation trajectory of magnetic species is influenced by magnetic force, gravity, van der Waals force, and electrostatic force. The classification and characteristic analysis of magnetic flocculants further expand our understanding of magnetic flocculation technology. There are other examples such as carbon loading and ultrafiltration. These combined-technology examples demonstrate the flexibility and effectiveness of magnetic flocculation in integrated water treatment solutions. Magnetic flocculation technology has shown great application prospects in the field of water treatment due to its high efficiency and environmental protection. Future research

should further address the renewal and expansion of magnetic species, the development of new magnetic materials, the optimisation of flocculant synthesis methods, and the improvement of synergies with other technologies to achieve broader applications and more profound improvements in water quality.

The potential of magnetic seeds and magnetic flocculants for application in water treatment technology is considerable, with several avenues of development emerging from this research. The initial focus of research will be on the multifunctionality of these materials. The precise control of the size, morphology, and magnetic properties of magnetic materials using nanotechnology will optimise their performance in specific applications. Furthermore, the introduction of intelligent response functions may enable the materials to automatically adjust their treatment strategies according to changes in the environment. Additionally, the incorporation of flocculation, adsorption, and catalytic properties may facilitate the highly efficient removal of a wide range of pollutants in a complex water environment. Secondly, the combination of magnetic materials with inorganic materials, organic polymers, or biological materials will become a focus of innovation, with the objective of enhancing the efficiency and selectivity of removal. The achievement of large-scale production and cost reduction of magnetic materials, with the aim of making them more widely used in practical water treatment projects, in addition to the development of non-toxic and recyclable magnetic materials, will become a trend in order to reduce the impact on the environment. Ultimately, future research should further investigate the renewal and expansion of magnetic species, the development of new magnetic materials, the optimisation of flocculant synthesis methods, and the enhancement of synergistic effects with other technologies. This will facilitate the achievement of wider applications and deeper water quality improvements.

Author Contributions: Conceptualization, Z.H., Z.W., K.W., K.J.S. and Y.S.; methodology, Z.H. and Y.S.; validation, K.J.S. and Y.S.; formal analysis, Z.H., Z.W. and K.W.; investigation, Z.H., Z.W. and K.W.; resources, K.J.S. and Y.S.; data curation, K.J.S. and Y.S.; writing—original draft preparation, Z.H. and Y.S.; writing—review and editing, Z.H., K.J.S. and Y.S.; visualization, Y.S.; supervision, Y.S. and K.J.S.; project administration, Y.S. and K.J.S.; funding acquisition, Y.S. All authors have read and agreed to the published version of the manuscript.

Funding: This research was supported by the National Natural Science Foundation of China (No. 51508268), Natural Science Foundation of Jiangsu Province in China (No. BK20201362), and 2018 Six Talent Peaks Project of Jiangsu Province (JNHB-038).

Data Availability Statement: Data are contained within the article.

Conflicts of Interest: The authors declare no conflicts of interest.

References

1. Dhakal, N.; Salinas-Rodriguez, S.G.; Hamdani, J.; Abushaban, A.; Sawalha, H.; Schippers, J.C.; Kennedy, M.D. Is Desalination a Solution to Freshwater Scarcity in Developing Countries? *Membranes* **2022**, *12*, 381. [CrossRef] [PubMed]
2. Dutta, D.; Arya, S.; Kumar, S. Industrial wastewater treatment: Current trends, bottlenecks, and best practices. *Chemosphere* **2021**, *285*, 131245. [CrossRef] [PubMed]
3. Ma, T.; Sun, S.; Fu, G.; Hall, J.W.; Ni, Y.; He, L.; Yi, J.; Zhao, N.; Du, Y.; Pei, T.; et al. Pollution exacerbates China's water scarcity and its regional inequality. *Nat. Commun.* **2020**, *11*, 650. [CrossRef]
4. Virpiranta, H.; Okyere Abayie, S.; Mäkikangas, J.; Puirava, M.; Koivula, K.; Leiviskä, T. Treatment of fish processing plant wastewater using dissolved air flotation and pilot-scale biochar column filtration. *J. Environ. Chem. Eng.* **2023**, *11*, 110853. [CrossRef]
5. Baghizade, A.; Farahbod, F.; Alizadeh, O. Experimental study of the biological treatment process of the exit wastewater from flocculation reactor. *Appl. Water Sci.* **2023**, *13*, 74–77. [CrossRef]
6. Babu Ponnusami, A.; Sinha, S.; Ashokan, H.; Paul, M.V.; Hariharan, S.P.; Arun, J.; Gopinath, K.P.; Hoang Le, Q.; Pugazhendhi, A. Advanced oxidation process (AOP) combined biological process for wastewater treatment: A review on advancements, feasibility and practicability of combined techniques. *Environ. Res.* **2023**, *237*, 116944. [CrossRef]
7. Xu, S.; Yan, Y.; Shuang, C.; Zhou, Q.; Ji, R.; Li, A. Biological magnetic ion exchange resin on advanced treatment of synthetic wastewater. *Bioresour. Technol.* **2023**, *372*, 128613. [CrossRef] [PubMed]

8. Zhao, C.; Zhou, J.; Yan, Y.; Yang, L.; Xing, G.; Li, H.; Wu, P.; Wang, M.; Zheng, H. Application of coagulation/flocculation in oily wastewater treatment: A review. *Sci. Total Environ.* **2021**, *765*, 142795. [CrossRef] [PubMed]
9. Huang, L.; He, W.; Zhang, Y.; Wang, X.; Wu, K.; Yang, Z.; Zhang, J. Chitosan enhances poly aluminum chloride flocculation system removal of microplastics: Effective, stable, and pollution free. *J. Water Process Eng.* **2023**, *54*, 103929. [CrossRef]
10. Li, S.; Kang, Y. Polymerization mechanism of polyferric aluminum phosphatic sulfate (PFAPS) and its flocculation effect on simulated dye wastewater. *Korean J. Chem. Eng.* **2022**, *39*, 1831–1838. [CrossRef]
11. Wang, J.; Chang, F.; Zheng, M. Advanced Treatment of Coking Wastewater by Polyaluminum Silicate Sulfate for Organic Compounds Removal. *Int. J. Environ. Res. Public. Health* **2023**, *20*, 6342. [CrossRef] [PubMed]
12. Zhang, M.; Xiao, F.; Wang, D.; Xu, X.; Zhou, Q. Comparison of novel magnetic polyaluminum chlorides involved coagulation with traditional magnetic seeding coagulation: Coagulant characteristics, treating effects, magnetic sedimentation efficiency and floc properties. *Sep. Purif. Technol.* **2017**, *182*, 118–127. [CrossRef]
13. Song, W.; Liu, M.; Hu, R.; Tan, X.; Li, J. Water-soluble polyacrylamide coated-Fe_3O_4 magnetic composites for high-efficient enrichment of U(VI) from radioactive wastewater. *Chem. Eng. J.* **2014**, *246*, 268–276. [CrossRef]
14. Li, R.; Li, P.; Cai, J.; Xiao, S.J.; Yang, H.; Li, A.M. Efficient adsorption of both methyl orange and chromium from their aqueous mixtures using a quaternary ammonium salt modified chitosan magnetic composite adsorbent. *Chemosphere* **2016**, *154*, 310–318. [CrossRef] [PubMed]
15. Wang, Y.; Zhang, Y.; Hou, C.; Qi, Z.; He, X.; Li, Y. Facile synthesis of monodisperse functional magnetic dialdehyde starch nano-composite and used for highly effective recovery of Hg(II). *Chemosphere* **2015**, *141*, 26–33. [CrossRef] [PubMed]
16. Lim, S.F.; Zheng, Y.M.; Zou, S.W.; Chen, J.P. Characterization of copper adsorption onto an alginate encapsulated magnetic sorbent by a combined FT-IR, XPS and mathematical modeling study. *Environ. Sci. Technol.* **2008**, *42*, 2551–2556. [CrossRef] [PubMed]
17. Luo, L.; Nguyen, A.V. A review of principles and applications of magnetic flocculation to separate ultrafine magnetic particles. *Sep. Purif. Technol.* **2017**, *172*, 85–99. [CrossRef]
18. Seo, J.Y.; Lee, K.; Lee, S.Y.; Jeon, S.G.; Na, J.; Oh, Y.; Park, S.B. Effect of barium ferrite particle size on detachment efficiency in magnetophoretic harvesting of oleaginous Chlorella sp. *Bioresour. Technol.* **2014**, *152*, 562–566. [CrossRef] [PubMed]
19. Liu, C.; Wang, X.; Qin, L.; Li, H.; Liang, W. Magnetic coagulation and flocculation of a kaolin suspension using Fe_3O_4 coated with SiO_2. *J. Environ. Chem. Eng.* **2021**, *9*, 105980. [CrossRef]
20. Dos Santos, T.R.T.; Mateus, G.A.P.; Silva, M.F.; Miyashiro, C.S.; Nishi, L.; de Andrade, M.B.; Fagundes-Klen, M.R.; Gomes, R.G.; Bergamasco, R. Evaluation of Magnetic Coagulant (α-Fe_2O_3-MO) and its Reuse in Textile Wastewater Treatment. *Water Air Soil Pollut.* **2018**, *229*, 92. [CrossRef]
21. Liu, Y.; Jin, W.; Zhou, X.; Han, S.; Tu, R.; Feng, X.; Jensen, P.D.; Wang, Q. Efficient harvesting of Chlorella pyrenoidosa and Scenedesmus obliquus cultivated in urban sewage by magnetic flocculation using nano-Fe_3O_4 coated with polyethyleneimine. *Bioresour. Technol.* **2019**, *290*, 121771. [CrossRef] [PubMed]
22. Tang, J.; Wang, J.; Jia, H.; Wen, H.; Li, J.; Liu, W.; Li, J. The investigation on Fe_3O_4 magnetic flocculation for high efficiency treatment of oily micro-polluted water. *J. Environ. Manag.* **2019**, *244*, 399–407. [CrossRef] [PubMed]
23. Jiang, R.; Zhu, H.Y.; Fu, Y.Q.; Li, X.; Jiang, S.T.; Li, J.B. Adsorptive removal of anionic azo dye by Al(3^+)-modified magnetic biochar obtained from low pyrolysis temperatures of chitosan. *Environ. Sci. Pollut. R* **2023**, *30*, 44985–44998. [CrossRef] [PubMed]
24. Chen, Y.; Luo, M.; Cai, W. Influence of operating parameters on the performance of magnetic seeding flocculation. *Environ. Sci. Pollut. R* **2016**, *23*, 2873–2881. [CrossRef] [PubMed]
25. *GB3838-2002*; Surface Water Environmental Quality Standards. Ministry of Ecology and Environment: Beijing, China, 2002.
26. Zhang, X.; He, X.; Wei, M.; Li, F.; Hou, P.; Zhang, C. Magnetic Flocculation Treatment of Coal Mine Water and a Comparison of Water Quality Prediction Algorithms. *Mine Water Environ.* **2019**, *38*, 391–401. [CrossRef]
27. Zhao, Y.; Liang, W.Y.; Liu, L.J.; Li, F.Z.; Fan, Q.L.; Sun, X.L. Harvesting Chlorella vulgaris by magnetic flocculation using Fe3O4 coating with polyaluminium chloride and polyacrylamide. *Bioresour. Technol.* **2015**, *198*, 789–796. [CrossRef]
28. Ding, C.; Xie, A.; Yan, Z.; Li, X.; Zhang, H.; Tang, N.; Wang, X. Treatment of water-based ink wastewater by a novel magnetic flocculant of boron-containing polysilicic acid ferric and zinc sulfate. *J. Water Process Eng.* **2021**, *40*, 101899. [CrossRef]
29. Dai, K.; Chen, P.; Wang, Z.; Yang, P.; Li, M.; Tang, C.; Zhuang, W.; Zhu, C.; Ying, H.; Wu, J. Magnetic composite $Ca(OH)_2/Fe_3O_4$ for highly efficient flocculation in papermaking black liquor without pH neutralization. *Adv. Powder Technol. Int. J. Soc. Powder Technol. Jpn.* **2021**, *32*, 2457–2468. [CrossRef]
30. Song, L.; Zhang, H.; Wang, R.; Wang, X.; Zhu, B.; Sun, D.; Lou, D. Preparation of a Magnetic Polysilicate Composite Flocculant for Dye Wastewater Treatment. *Water Air Soil Pollut.* **2024**, *235*, 101. [CrossRef]
31. LI, S.; Wang, X.; Zhang, Q. Dynamic experiments on flocculation and sedimentation of argillized ultrafine tailings using fly-ash-based magnetic coagulant. *T Nonferr Metal. Soc.* **2016**, *26*, 1975–1984. [CrossRef]
32. Li, S.; Wang, X. Fly-ash-based magnetic coagulant for rapid sedimentation of electronegative slimes and ultrafine tailings. *Powder Technol.* **2016**, *303*, 20–26. [CrossRef]
33. Lee, H.K.; Kim, J.H.; Kim, I.; Jeon, H. Efficient separation performance of suspended soil and strontium from aqueous solution using magnetic flocculant. *J. Environ. Chem. Eng.* **2021**, *9*, 106810. [CrossRef]

34. Tian, Y.; Li, Q.; Xu, G.; Zhang, Y.; Tu, Y.; Jin, Z.; Liang, Y.; Zhang, Y.; Dai, J.; Xie, X. Mechanistic investigation of an inorganic-organic hybrid coagulant with Fe3O4 magnetic loading-enhanced flocculation for water treatment. *J. Clean. Prod.* **2024**, *444*, 141214. [CrossRef]
35. Liu, Z.; Duan, X.; Zhan, P.; Liu, R.; Nie, F. Coagulation performance and microstructural morphology of a novel magnetic composite coagulant for pre-treating landfill leachate. *Int. J. Environ. Sci. Technol.* **2017**, *14*, 2507–2518. [CrossRef]
36. Hena, S.; Fatihah, N.; Tabassum, S.; Lalung, J.; Jing, S.Y. Magnetophoretic harvesting of freshwater microalgae using polypyrrole/Fe3O4 nanocomposite and its reusability. *J. Appl. Phycol.* **2016**, *28*, 1597–1609. [CrossRef]
37. Xie, F.; Zhang, H.; Wang, R.N.; Wang, X.Y.; Lou, D.W. Treatment of simulated wastewater containing Congo red using magnetic polysilicate containing aluminum and magnesium-starch flocculant. *Environ. Prot. Chem. Ind.* **2024**, *44*, 130–137.
38. Reck, I.M.; Baptista, A.; Paixao, R.M.; Bergamasco, R.; Vieira, M.F.; Vieira, A. Application of magnetic coagulant based on fractionated protein of Moringa oleifera Lam. seeds for aqueous solutions treatment containing synthetic dyes. *Environ. Sci. Pollut. R.* **2020**, *27*, 12192–12201. [CrossRef] [PubMed]
39. Dos Santos, T.R.T.; Mateus, G.A.P.; de Andrade, M.B.; Guerra, A.C.S.; Vieira, A.M.S.; Bergamasco, R. A simple and effective method for Escherichia coli inactivation in aqueous medium using natural based superparamagnetic coagulant. *Environ. Prog. Sustain.* **2021**, *40*, e13503. [CrossRef]
40. Sun, Y.; Yu, Y.; Zheng, X.; Chen, A.; Zheng, H. Magnetic flocculation of Cu(II) wastewater by chitosan-based magnetic composite flocculants with recyclable properties. *Carbohyd Polym.* **2021**, *261*, 117891. [CrossRef]
41. Ge, S.; Agbakpe, M.; Zhang, W.; Kuang, L.; Wu, Z.; Wang, X. Recovering Magnetic Fe_3O_4-ZnO Nanocomposites from Algal Biomass Based on Hydrophobicity Shift under UV Irradiation. *ACS Appl. Mater. Inter.* **2015**, *7*, 11677–11682. [CrossRef]
42. Dos, S.J.; Pereira, C.R.; Pinto, L.; Frantz, T.; Lima, E.C.; Foletto, E.L.; Dotto, G.L. Synthesis of a novel $CoFe_{(2)}O_{(4)}$/chitosan magnetic composite for fast adsorption of indigotine blue dye. *Carbohyd Polym.* **2019**, *217*, 6–14.
43. Santos, T.R.T.; Silva, M.F.; Nishi, L.; Vieira, A.M.S.; Klein, M.R.F.; Andrade, M.B.; Vieira, M.F.; Bergamasco, R. Development of a magnetic coagulant based on Moringa oleifera seed extract for water treatment. *Environ. Sci. Pollut. Res. Int.* **2016**, *23*, 7692–7700. [CrossRef] [PubMed]
44. Guo, Z.H.; Liu, Z.T.; Shen, B.X.; Ma, Y.L.; Sun, J.Q. Fabrication of MBFX-8 /magnetic Fe_3O_4@CMC composite flocculant and its removal towards $Cu2^+$. *Chin. J. Environ. Eng.* **2018**, *12*, 421–426.
45. Yu, X.L.; Tong, S.R.; Ge, M.F.; Zuo, J.C.; Cao, C.Y.; Song, W.G. One-step synthesis of magnetic composites of cellulose@iron oxide nanoparticles for arsenic removal. *J. Mater. Chem. A* **2013**, *1*, 959–965. [CrossRef]
46. Periyasamy, S.; Gopalakannan, V.; Viswanathan, N. Fabrication of magnetic particles imprinted cellulose based biocomposites for chromium(VI) removal. *Carbohyd Polym.* **2017**, *174*, 352–359. [CrossRef]
47. Xiao, X.F.; Yu, Y.Y.; Sun, Y.J.; Zheng, X.; Chen, A.W. Heavy metal removal from aqueous solutions by chitosan-based magnetic composite flocculants. *J. Environ. Sci.* **2021**, *108*, 22–32. [CrossRef]
48. Proskurina, V.E.; Kashina, E.S.; Rakhmatullina, A.P. Sedimentation of Titanium Dioxide Suspension under the Action of Magnetic Flocculants. *Colloid J.* **2023**, *85*, 72–79. [CrossRef]
49. Yang, J.J.; Wang, J.K.; Guo, J.P.; Zhang, Y.; Zhang, Z.F. Dendrimer modified composite magnetic nano-flocculant for efficient removal of graphene oxide. *Sep. Purif. Technol.* **2023**, *307*, 122851. [CrossRef]
50. Noor, M.; Kho, J.L.; Sundrammoorthy, D.; Ngadi, N. Phytotoxicity and Cost-effectiveness Analysis of Palm Oil Wastewater Treated with Magnetic Cationic Starch Flocculant. *Water Air Soil Poll.* **2022**, *233*, 482. [CrossRef]
51. Liu, Z.; Zhan, P.; Nie, F. Advanced Treatment of Leachate Secondary Effluent by a Combined Process of MFPFS Coagulation and Sulfate Radical Oxidation. *Pol. J. Environ. Stud.* **2016**, *25*, 1615–1622. [CrossRef]
52. Wang, K.; Mao, Y.; Wang, C.; Ke, Q.; Zhao, M.; Wang, Q. Application of a combined response surface methodology (RSM)-artificial neural network (ANN) for multiple target optimization and prediction in a magnetic coagulation process for secondary effluent from municipal wastewater treatment plants. *Environ. Sci. Pollut. Res. Int.* **2022**, *29*, 36075–36087. [CrossRef] [PubMed]
53. *GB 4287-2012*; Water Pollutant Discharge Standard for Textile Dyeing and Finishing Industry. Ministry of Ecology and Environment: Beijing, China, 2012.
54. Zhang, J.L.; Xie, W.; Cai, J.F.; Luo, C.; Ren, Y.; Ding, J. Application of Carbon-loaded Magnetic Coagulation and High Efficiency Sedimentation in Keqiao Jiangbin WWTP. *China Water Wastewater* **2023**, *39*, 120–125.
55. Gai, X. Design of a Wastewater Treatment Plant Phase II Upgrading and Expansion Project in Wuhu City. *China Water Wastewater* **2023**, *39*, 80–84.
56. Chu, Z.Y.; Li, J.F.; Lin, Y.; Dong, Q.Q. Application of MBBR, Magnetic Coagulation and Denitrification Deep Bed Filter Process in an Underground Wastewater Treatment Plant. *China Water Wastewater* **2023**, *39*, 80–84.
57. Chen, B.; Tang, J.; Ye, Q.H.; Gao, X.H. An application example of improved AAO-magnetic coagulation sedimentation-ozone-activated carbon process in urban sewage treatment plant. *Ind. Water Wastewater* **2021**, *52*, 81–84.
58. Chen, J.H.; Guan, Z.J.; Feng, S.Q.; Huang, M.Z.; Lin, X.Z.; Sun, S.Y. Study on treatment of high turbidity wastewater from a water supply plant by magnetic flocculation coupled ultrafiltration process. *Environ. Pollut. Control* **2023**, *45*, 1660–1665,1670.

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.