



Assessing the effectiveness of magnetic nanoparticles coagulation/flocculation in water treatment: a systematic literature review

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Abstract

In this study, a systematic review was performed based on the publications, which report applications of magnetic nanoparticles in coagulation/flocculation technology for water treatment. Through a series of screening steps, eight themes were developed, which composed of 62 relevant articles. The application of magnetic coagulant/flocculant and their effectiveness in water treatment were discussed based on the themes. The findings explained the roles and benefits of integrating magnetic nanoparticles in coagulation/flocculation in water treatment process, as follows: (1) ability to reduce the utilization of conventional coagulant/flocculant due to high charge density provides by magnetic nanoparticles (Fe^{2+} and Fe^{3+}); (2) shorten the duration of coagulation/flocculation process due to magnetic dipole attraction between magnetic nanoparticles and destabilized pollutants; and (3) recovery of used/exhausted magnetic coagulant/flocculant can be achieved using an external magnetic field, thus reducing materials cost. In-depth studies on magnetic flocs physico-chemical characteristics are recommended for further studies to enable the understanding of the fundamental mechanism of the coagulation/flocculation process. The toxicological and economic implications related to magnetic coagulation/flocculation applications in wastewater treatment are also recommended for investigation.

Keywords Coagulation · Flocculation · Magnetic nanoparticle · Systematic literature review · Water treatment

Introduction

Water is an important element to human survival and sustainability of the ecosystem. About 70% of the Earth's surface is covered by water, but only 2.5% of the Earth's water is freshwater (Mohd-Salleh et al. 2019); thus, preservation of such a great gift from nature is crucial. In addition to the municipal and agricultural uses, the rapid growth in global industrialization since the last decade also causes increasing water demands. According to Shah et al. (2018), the industrial activities are responsible for 19% of global

water consumption, and such figure will increase to 55% by 2050. The human and industrial activities also generate large amount of wastewater. In a broad perspective, wastewater can be interpreted as a combination of more than one effluent containing different components, such as pathogens, organic matters and heavy metals, which can diminish surface water quality (Mohd-Salleh et al. 2019). The lack of proper management and treatment of such wastewater will lead to disastrous effects such as deficiency of clean water for municipal, agricultural and industrial uses (i.e. drinking, washing, cooling, heating), which subsequently threaten the human well-being and quality of manufactured products.

Various separation technologies and management strategies have been developed for the removal of undesired pollutants prior to the effluent discharge into water bodies (Gerba and Brusseau 2019). The well-developed techniques have been incorporated in domestic and industrial wastewater treatment plants, while several advanced treatment technologies are being tested at pilot scale. Depending on the characteristics of pollutants to be removed, separation technologies including filtration (Hamoda et al. 2004; Ostad-Ali-Askari 2019), liquid–liquid extraction (LLE) (Mansur

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et al. 2008; Somerset and Luzi-Thafeni 2019), coagulation/flocculation (C–F) (Oladoja 2015; Salehizadeh et al. 2018), ion exchange (Soyluoglu et al. 2020), adsorption (Shen et al. 2019) and membrane (Asad et al. 2020; Moslehyani et al. 2019), as well as different combination of these techniques, are commonly used in wastewater treatment units. While some of these established techniques require high maintenance and operational costs, treatment via C–F with simple operating conditions and lower costs is a popular option in developing countries (Crini and Lichtfouse 2019; Ho et al. 2020). The primary goal of the C–F process is to get rid of the colloidal impurities as well as turbidity present in wastewater, by adding a coagulant, such as potent chemicals, to alter the physical states of suspended or dissolved solids (Saravanan et al. 2017). Flocculants (usually polymers) are added after coagulation process to ensure the formation of larger and denser flocs for better separation (Wang et al. 2020). Selection of coagulants and flocculants is important in order to maximize the treatment efficiency. Scientists have therefore developed various alternatives, including the use of natural coagulants (such as chitosan, alginate, *Moringa oleifera* (MO), starch, etc.) (Oladoja 2015) and organic polymers (Lee et al. 2014). Apart from that, integrative approach between natural and synthetic materials (Lee et al. 2012) and surface modifications of existing coagulant/flocculant (Luo and Nguyen 2017; Tang et al. 2018) have also been developed, and these could potentially replace the conventional coagulants (alum, ferric chloride and polyaluminium chloride).

As a part of the industrial revolution 4.0 component, nanotechnology has dramatically influenced social and environmental sustainable development. This technology has found numerous applications in various fields, including electronic, energy, medicine, environment and agriculture (Rai and Rai 2015). Magnetic nanoparticles (MNPs, Fe_3O_4) are among the nanomaterials that have become research hot-spot in different disciplines (Jiang et al. 2018). MNPs has portrayed unique characteristics such as high magnetism, low cost and toxicity, along with high durability and biocompatibility related to their applications (Medvedeva et al. 2015). MNPs also exhibit distinctive features that improve wastewater treatment efficiency via different processes. For instance, MNPs could act as photocatalysts for the removal of *E. coli* and *S. aureus* in water disinfection (Najafpoor et al. 2020; Zhang et al. 2019a). Modification of MNPs for adsorption of organic materials (e.g. dyes, pharmaceutical, etc.) (Chen et al. 2014; Hayasi and Saadatjoo 2018; Ojemaye et al. 2017) and reduction of heavy metals (Chen et al. 2014; Hayasi and Saadatjoo 2018; Ojemaye et al. 2017; Su 2017) were also reported. Similarly, the incorporation of MNPs in C–F technology also upgrades the water treatment process. Coagulant/flocculant(s) functionalized with MNPs are proven to improve the water treatment efficiency even

at small dosage. The settling time can also be shortened, and total recovery of used/exhausted magnetic coagulant/flocculant can be achieved using an external magnetic field (Chen et al. 2019, 2016; Zhang et al. 2016). With all these advantages, there are growing interests among scientists and practitioners in water treatment technology using magnetic coagulant/flocculants.

Despite the abundance of works on C–F technologies and applications of MNPs in water/wastewater treatments, a systematic literature review (SLR) on this topic is unavailable. Therefore, the SLR carried out in this study will fill in the gap by providing a body of knowledge based on evidences on the effectiveness of magnetic coagulant/flocculant application in water treatment. This study provides vital information to researchers on the recent development in the studied area. It also provides guidance for future improvement, especially on the adaptation of magnetic coagulation/flocculation (MCF) in water purification. The theoretical framework focuses mainly on evaluating the efficiencies of MCF in water treatment as well as the roles of MNPs in C–F mechanism that are still less explored.

The review process was conducted on March 2020 at Universiti Teknologi Malaysia, Malaysia.

Methodology

Systematic review framework

In general, SLR is produced based on the related scientific literature with a view to selecting, differentiating and analysing specific published studies. The measures required are outlined, concentrating on potential prospects in relation to the issues being posed. This study utilizes the Protocol, Search, Appraisal, Synthesis, Analysis and Report (PSAL-SAR) framework, which is commonly adopted in environmental science, technology, engineering and social science fields (Mengist et al. 2020), including knowledge transfer in maintenance applications (del Amo et al. 2018) and ecosystem services (Malinauskaite et al. 2019; Perevochtchikova et al. 2019). Various scientific works were adopted this methodological approach as it reduces the risks related to publication bias and to increase its acceptability of the task (del Amo et al. 2018; Grant and Booth 2009; Malinauskaite et al. 2019; Mengist et al. 2020; Perevochtchikova et al. 2019).

The aforementioned use of a review protocol is important in formulating an effective research question in addition to guaranteeing the transparency, transferability and replicability of the work, which are the characteristics of a SLR (Booth et al. 2016). Moreover, adoption of this method minimizes the bias by conducting exhaustive literature searches. Throughout this analysis, the SLR has been driven by

concepts of “Problem or Population, Interest and Context (PICO)” (Jones 2004). Based on these concepts, the authors have defined three main aspects in the systematic review, namely magnetic coagulation/flocculation (population), its roles and efficacy (interest) and water treatment (context). The central research question generated from these aspects is “what are the performances of MCF in water treatment?”.

Systematic review process

The article retrieval process used in this SLR is adapted from the works by Shaffril et al. (2019) and Moher et al. (2010). Two world-leading citation databases, namely Scopus and Web of Science (WoS), were used for this SLR, as both databases are robust and contain more than 256 fields of studies including engineering and environmental science fields (Vera-Baceta et al. 2019). With such prominence, these two indexed databases were chosen to ensure the quality of the articles reviewed in the present study. By using the complete search string shown in Table 1 in the first phase, a total of 1,101 and 836 documents were retrieved from Scopus and WoS, respectively.

In screening phase, all the 1937 articles were screened to extract appropriate articles based on specific inclusion

criteria designed based on the research question as recommended by Kitchenham and Charters (2007). For this study, only research articles written in English, which were published within 2009–2019, were selected. The number of studies related to magnetic nanoparticles in C–F technology with its application in water treatment has shown a healthy growth since 2009. Based on the criteria, 853 articles were removed. A total of 293 duplicate articles were also identified and removed at this stage.

At eligibility stage, the titles and abstracts of all 791 articles were manually skimmed to ensure their relevance to the research question. A total of 729 articles were eliminated due to the following reasons: (1) only C–F process was reported without the application of MNPs; (2) MCF was used for the applications other than water treatment; (3) the effectiveness of MCF in water treatment is not reported; (4) article content focused on MCF synthesis and characterization; and (5) non-accessible articles. Finally, 62 articles were selected for further analysis.

Despite the inconsistency observed in 2014, a steady growth in number of articles published on this topic from 1 (in 2009) to 11 (in 2017) was observed, followed by the decline after 2017. All the 62 articles selected for this study were published by authors from different countries around the globe. As shown in Fig. 1b, most of the studies reviewed were conducted in Asia region (Iraq, Iran, China, Taiwan, South Korea, Japan, Malaysia), followed by Europe continent (Russia, Sweden and Germany) and US region (Canada and Brazil). South Africa is the only country in Africa continent, which reported the MCF synthesis for C–F applications. Four articles were published as a result of international collaborations (Australia–China (Li et al. 2017b), France–China (Liu et al. 2013), Sweden–Mexico–Spain (Okoli et al. 2012b), Singapore–China (Zhang et al. 2015)). The journals related to the MCF synthesis for C–F application were also analysed based on the journal quartiles that reflect the journal qualities (García et al. 2011). As portrayed in Fig. 1(c), MCF research is considered a significant and impactful topic by the researchers, as about 34% of selected articles were published in the first quartile (Q1) journals. Meanwhile, sixteen articles were published in Q2 and Q3 journals, respectively, while four articles were published in Q4 journals.

The 62 articles were grouped into eight themes according to the categories of wastewater treated. Within review timeline, MCF has been applied in: (1) turbidity reduction; (2) heavy metal removal; (3) organic-based wastewater; (4) minerals and microbes; (5) effluents containing ultrafine particles; (6) textiles/dyeing wastewater; (7) pharmaceutical wastewater; and (8) others. The distributions of articles for each theme are summarized in Fig. 2.

Table 1 The search strings used for documents retrieval

Database	Keywords used
WoS	TS=((magnetic* OR "magnetic nanoparticle*" OR magnetite OR "magnetic powder*" OR "magnetite powder*" OR fe3o4 OR "iron oxide" OR "magnetic nanomaterial*" OR "iron oxide* nano*" OR mnp* OR mion* OR "superparamagnetic* nano*" OR spion* OR "superparamagnetic iron*" OR "magnet* seed*" OR "magnet* iron oxide* nano*" OR "nano&magneti*" OR "nano*&fe3o4") AND ((coag* OR aggrome*) OR (floc* OR "coagulation&flocculation" OR "coagulation-flocculation" OR "coag*-floc*")) AND (water* OR wastewater* OR pollutant* OR contaminant*) AND (treat* OR effluent* OR application* OR remed* OR removal*))
Scopus	TITLE-ABS-KEY((magnetic* OR "magnetic nanoparticle*" OR magnetite OR "magnetic powder*" OR "magnetite powder*" OR fe3o4 OR "iron oxide" OR "magnetic nanomaterial*" OR "iron oxide* nano*" OR mnp* OR mion* OR "superparamagnetic* nano*" OR spion* OR "superparamagnetic iron*" OR "magnet* seed*" OR "magnet* iron oxide* nano*" OR "nano&magneti*" OR "nano*&fe3o4") AND ((coag* OR aggrome*) OR (floc* OR "coagulation&flocculation" OR "coagulation-flocculation" OR "coag*-floc*")) AND (water* OR wastewater* OR pollutant* OR contaminant*) AND (treat* OR effluent* OR application* OR remed* OR removal*))



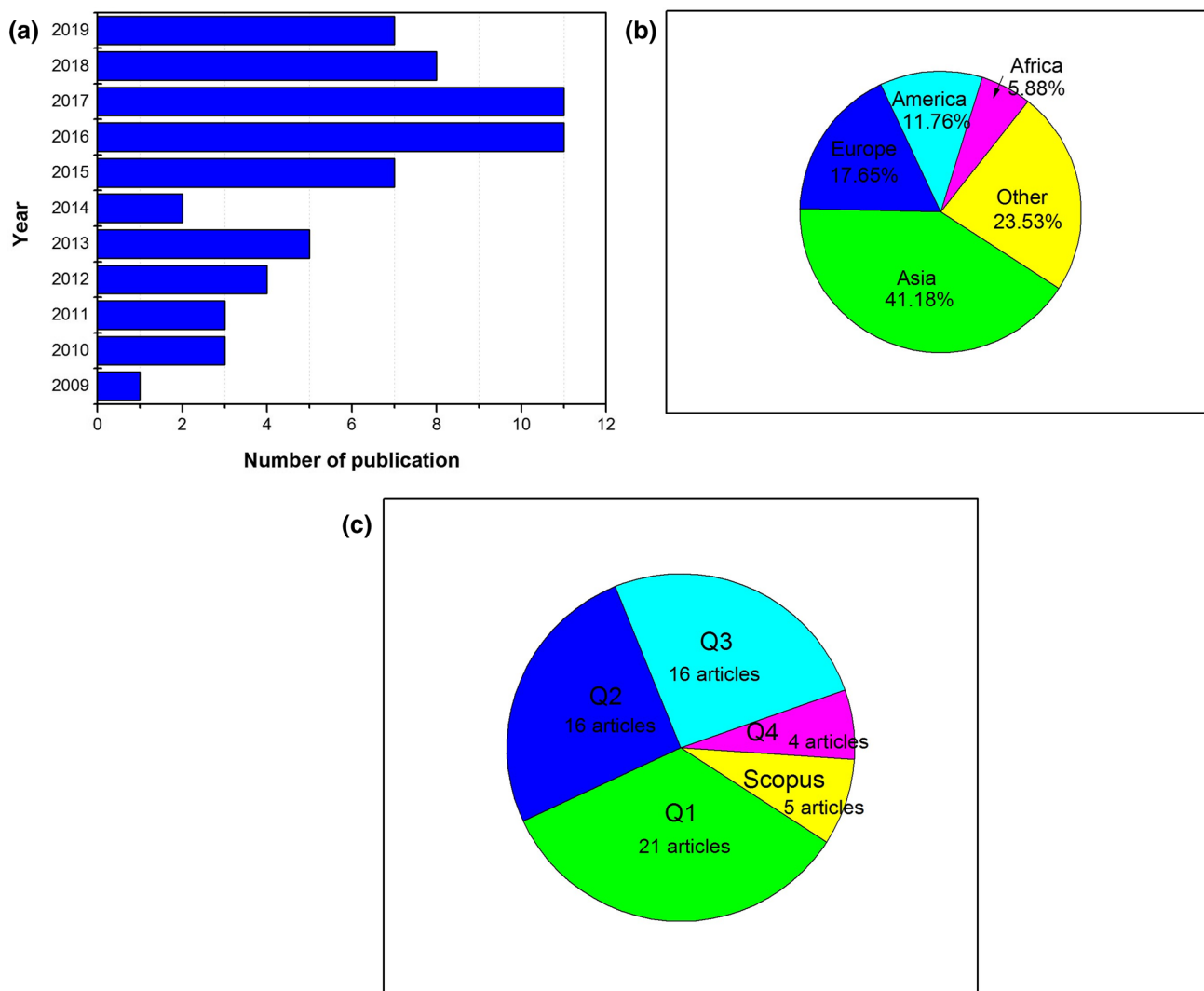


Fig. 1 Articles distribution based on **a** year of publication, **b** affiliation region and **c** indexed categories

Discussion

This section focuses on assessment of the effectiveness of MCF based on the 62 publications in treatment of various wastewater types based on the themes stated in the previous section.

Turbidity reduction

A total of 20 documents analysed in this study describes the application of MCF in turbidity reduction, making it the biggest cluster in the current review. This is probably due to the fact that the primary goal in C–F is to get rid of the turbidity as well as other colloidal impurities present in the water (Mohd-Salleh et al. 2019). All papers with their main objectives to remove turbidity in synthetic or actual wastewater employing MCF are summarized in Table 2.

A study carried out by Zhang et al. (2012) using magnetic polyaluminium chloride (PAC) nanocomposite in kaolin suspension revealed that the presence of MNPs could affect the hydrolysis of Al (from PAC) by increasing Al species–nanoparticle cluster and hence the effective collision rate. In addition, the large specific surface areas and magnetic dipole–dipole attraction of MNPs enhanced the adsorption capability, leading to higher turbidity removal rate (Zeng et al. 2010). Thus, the study concluded that instantaneous adsorption, enmeshment and sweep are the key factors for the higher performance of magnetic PAC (MPAC) when compared to PAC. The performance of dual coagulation (simultaneous addition of MNPs and PAC) was also tested in actual surface water by Lohwacharin et al. (2014). The sequence of MNPs and PAC introduction into the water sample did not show any influence on the performance. The same observation was also made by Zhang et al.



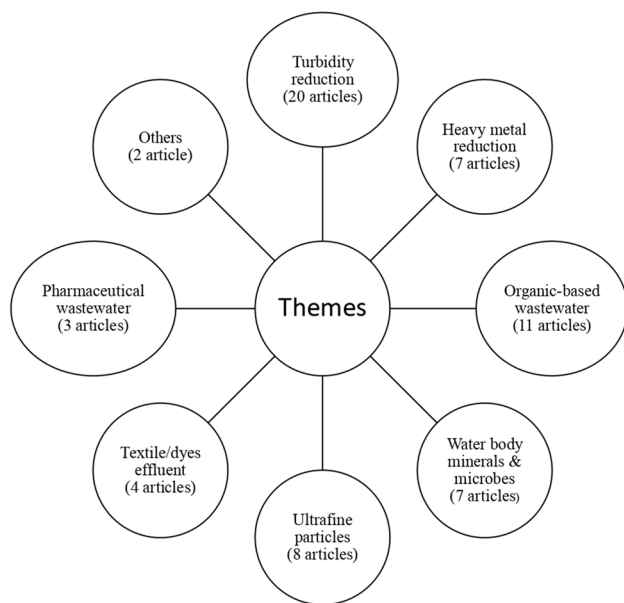


Fig. 2 Distribution of themes analysed

(2017b). Besides that, the MNPs–PAC combination could reduce the PAC dosage while achieving satisfactory turbidity removal efficiency. Addition of PAC increased the collision frequency and aggregation among MNPs and suspended solid (SS) particles.

The separation process after MCF application was also conducted by Zeng et al. (2015) by introducing a high gradient superconducting magnetic separation (HGSMs) as part of the treatment framework. The study highlighted that magnetized flocs were efficiently separated and eventually non-magnetic pollutants could be treated using HGSMs when MNPs were used in the process. The study also suggested that HGSMs can be used when there is limited space for placing traditional wastewater treatment unit, especially sedimentation tank, due to its smaller capacity, which eases the transport and installation processes.

The effects of flocculants, such as polyacrylamide (PAM), in MCF were also investigated by Chen et al. (2016). The result revealed that the turbidity removal was directly proportional to the PAM dosage until a saturation level was attained. Further addition of PAM at a fixed amount of MNPs beyond the saturation point did not produce any significant improvement in the removal performance. The research findings also showed that the presence of MNPs slightly reduced the amount of flocculants required from 18 mg/L (92% removal) to 17 mg/L (~93% removal) without compromising the turbidity reduction. Even though the performance seems insignificant, a huge difference can be seen from the settling speed of the flocs. At the optimum conditions, the settling speed of flocs by PAM with MNPs was approximately 3.64 times higher than that of without MNPs.

Turbidity removal was also performed using protein (extracted from MO seed), which was functionalized with MNPs (MMO) synthesized via the microemulsion method (Okoli et al. 2012b). The protein bound by MNPs showed higher coagulation activity due to the significant interactions between MNPs and protein. The functionality of adsorbed protein was retained even after binding to the MNPs. As the coagulation activity is a function of flocs sedimentation, the weight effect of MMO caused by electrostatic interactions increased the separation velocity of flocs as the magnetized flocs formed around them were much denser than suspended particles. The bare MNPs could also aggregate and flocculate, leading to formation of larger flocs due to collision between MNPs and suspended particles in turbid water sample (Hwang and Han 2015; Okoli et al. 2012a). However, the performance of MMO in removing turbidity from the actual surface water (lake) showed less promising results, probably due to the low turbidity level of the sample (< 16 NTU) (Lakshmanan et al. 2013).

In another study conducted by Santos et al. (2016), MNPs modification with oleic acid (AO) prior to functionalization with MO did not benefit the turbidity reduction (20% at settling time of 30 min in the presence of magnet) when compared to 90% reduction without AO at the same C–F condition. The reason behind this observation was that the presence of fatty acids in MO hindered the compatibilization of AO with MNPs and MO. This was demonstrated by a low removal due to the increased amount of organic content in water contributed by fatty acids (Zhu et al. 2011). However, AO was a good surface modifier to MNPs during its hybridization with PAM, as the presence of AO prevented agglomeration occurrence among MNP particles (Zhou et al. 2017). Moreover, oil extraction from MO seed using either ethanol or hexane prior to MNPs functionalization did not benefit the turbidity removal (dos Santos et al. 2018b). Ma et al. (2018) attempted the synthesis of magnetic cationic PAM via low-pressure UV-initiation grafting methods. Compared to conventional grafting method, this method increased the length and branching degree of PAM onto MNPs, which enhanced the entrapment of colloid particles and effective bridging effect during MCF process.

Magnetically enhanced coagulation membrane filtration process was also introduced, whereby the supernatant after C–F was allowed to pass through a membrane filter. Compared to normal C–F, MCF led to better permeation with significantly lower coagulant dosage needed. Addition of MNPs increased particle content and collision probability, thus resulting in the formation of magnetic flocs with stronger moving ability (Wang et al. 2016a). The process performance was dependent on the magnetization time and Fe^{3+} concentration in MNPs, as these factors influenced the distribution of Fe^{3+} ions and charge neutralization of pollutants during MCF (Wang et al. 2016b).

Table 2 The performance of MCF on turbidity removal

Magnetic coagulant/flocculant	Wastewater/effluent	Optimum parameter(s)	Turbidity removal (%)	References
PAC	Synthetic wastewater (kaolin)	Concentration: 0.08 mmol/L as Al	< 90	Zhang et al. (2012)
MPAC		Concentration: 0.08 mmol/L as Al; dosage of MNPs: 50 mg/L	92	
Crude MO protein MNPs	Synthetic wastewater (kaolin)	NR	70	Okoli et al. (2012b)
MO protein-functionalized MNPs (MMO)			45–50	
Crude MO protein MNPs	Synthetic wastewater (kaolin)	Settling time: 60 min	70	Okoli et al. (2012a)
MMO			< 40	
MMO	Orlangen and Brunnsviken Lake, Sweden	NR	> 80	Lakshmanan et al. (2013)
MNPs + PAC	Ara River, Japan	Dosage: 6.3 mg/L of MNPs and 20 mg/L of PAC; pH: 7.4	> 50	Lakshmanan et al. (2013)
MNPs	Municipal wastewater	Dosage: 8.85 mg/L; mixing time: 16.8 min; settling time: 10 min	> 98	Lohwacharin et al. (2014)
MNPs + PAC followed by HGMS	Raw water sample	Dosage: 8 mg/L of PAC and 30 mg/L of MNPs; magnetic intensity: 5.0 T; flow rate: 1 cm/s	29.4	Hwang and Han (2015)
MNPs + FeCl ₃ then membrane filtration	Luan River, China	Dosage: 15 mg/L; magnetization time: 6 min	90	Zeng et al. (2015)
MNPs + FeCl ₃ then membrane ultrafiltration	Luanhe River, China	Dosage: 20 mg/L; magnetization time: 4 min	95.4	Wang et al. (2016a)
MNPs + PAC + PAM	Synthetic copper wastewater	Dosage: 2.0 mg/L of MNPs, 1.8 g/L of PAC, 15 mg/L of PAM; pH: 9.0; MNPs size: 300–400 mesh	90	Wang et al. (2016b)
MNPs + PAC (optimization study)	Synthetic wastewater (kaolin)	Dosage: 0.6 mg/L of MNPs, 22.7 mg/L of PAC; dosing time: 1.2 min of MNPs, 1.5 min of PAC	93.25	Chen et al. (2016)
MNPs as nano-ferrofluid coagulant	Karun River, Iran	Dosage: 0.15 g; slow mixing rate: 40 rpm; slow mixing time: 10 min	98	Su et al. (2016)
MNP-coated oleic acid (AO)-functionalized MO	Pirapo River, Brazil	Dosage: 10 mg/L of MNPs, 200 mg/L of MO; settling time: 30 min	90	Hatamie et al. (2016)
MMO		Dosage: 10 mg/L of MNPs; 400 mg/L of MO; settling time: 30 min	20	Santos et al. (2016)
Magnetic PAM (MPAM)	Synthetic wastewater (kaolin)	Dosage: 2 g/L; settling time: 5 min	90	
PAM		Dosage: 0.4 mL of 2 g/L	82.8	Zhou et al. (2017)
Poly (acrylamide-co acryloyloxyethyl thimethylammonium chloride) grafted gelatin-coated MNPs [magneticpoly (AM-co-DAC)-g-gelatin]	Synthetic wastewater (kaolin)	Dosage: 10 mL; settling time: 3 min	82	Wang et al. (2017a)
poly (AM-co-DAC)-g-gelatin]			99	
MPAC	Synthetic wastewater (kaolin)	Dosage: 0.12 mmol/L as Al, 50 mg/L of MNPs; pH: 4; settling time: 30 min	92	
			89	Zhang et al. (2017b)



Table 2 (continued)

Magnetic coagulant/flocculant	Wastewater/effluent	Optimum parameter(s)	Turbidity removal (%)	References
PAC + MNPs			94	
MNPs coated with cationic PAM (MPAM)	Synthetic wastewater (kaolin)	Dosage: 0.24 g/L; rapid mixing rate: 300 rpm; rapid mixing time: 20 min; slow mixing time: 20 min	92.4	Ma et al. (2018)
MMO	Pirapo River, Brazil	Dosage: 20 mg of MNPs, 1 wt% of MO saline extract; settling time: 30 min	94.4	dos Santos et al. (2018b)
MMO	Pirapo River, Brazil	Dosage: 20 mg of MNPs, 1 wt% of MO saline extract; settling time: 10 min	96.8	Mateus et al. (2018)
PAC + MNPs followed by anionic PAM	Coal mine water, China	Dosage: 100 mg/L of PAC, 200 mesh 1 g/L of MNPs, 4 mg/L of anionic PAM; stirring intensity: 100–300 rpm	> 97	Zhang et al. (2019b)

Another study on the removal of turbidity from coal mine water (Zhang et al. 2019b) showed that pH significantly affected MCF efficiency, as neutral and alkaline environments were beneficial to magnetic flocculation. On the other hand, the size and dosage of MNPs contributed minor effects to the residual turbidity. However, the presence of MNPs increased the flocs precipitation speed. The findings also suggested that the stirring intensity should be reduced stepwise for development of a good flocs growth profile.

Heavy metal removal

Excessive exposures of human body to hazardous heavy metals in water such as arsenic (As), lead (Pb), mercury (Hg) and chromium (Cr) could lead to fatality upon long-term accumulation of these metals ion in the bodies (Abdullah et al. 2019). MCF has become one of the options to remove these metals from the wastewater due to its simplicity in operation (Fu and Wang 2011). In this SLR, seven research publications related to the applications of MCF on removal of different heavy metals are identified.

As shown in Table 3, application of MCF technology resulted in 90% heavy metal removals. Although many studies have echoed the high surface adsorption by MNPs, removal of metal ions using MNPs alone did not produce satisfactory results when compared to MCF. The research works conducted by Li et al. (2010) and Ma et al. (2015) indicated removal efficiency below 20% when MNP was used alone, and a combination of MNPs and coagulants (iron(III) chloride (FeCl_3) and polyferric sulphate (PFS)) resulted in a more efficient adsorption–coagulation process for heavy metal removal. In general, MNPs act as core cells, which play important roles for adsorption of metal ions. The stable nanoclusters formed by MNPs with ferric salt

(coagulant) acted as bridging agents, which promoted the coagulation process (Zhao et al. 2012a). These nanoclusters accelerated the sedimentation velocity of metal ion flocs while promoting the occurrence of second flocculation owed to magnetic attraction. In other words, the combination of heavy metals, ferric salt (coagulant) and MNPs was induced by the interactions of surface charge, capture of particles in the magnetic flocculant mesh structure, and/or interaction between heavy metals and coagulant. These interactions caused precipitation on the magnetic particles. Research on the removal of various types of heavy metals via MCF conducted by Mandel et al. (2013) showed less favourable performance for Hg (< 60% removal). The difficulty to remove such metal ion could be related to a prevailing polyhydroxy complex for this element, which hardly precipitates with the ferric-based flocs (Ambashta and Sillanpää 2010).

Other than incorporating MNPs with the conventional inorganic coagulants in MCF, synthesis of effective magnetic coagulants derived from the natural resources such as gum karaya, starch and chitosan was also performed to evaluate their performances in heavy metal remediation. Most works showed promising results that are attributed to the abundance of functional groups, which attract the metal ions in the water. Most of the heavy metal removal mechanisms are pH-dependent (Ngah and Hanafiah 2008). The metal ions are more soluble in acidic solutions due to the high hydroxocomplex stability. The removal of metal ions under such conditions was not optimal, since H^+ would occupy most of the adsorption sites on the surface of magnetic flocculants, and fewer cationic contaminant ions could be bare because of electrical repulsion with H^+ ions (Liu et al. 2018). Higher uptake of metal ions was observed at higher pHs, which was ascribed to the increased degree of the metal complexation. These removals were likely due to

Table 3 Removal of metals in water using MCF

Type of MCF	Optimum conditions	Performance		References
		Metal	Removal (%)	
MNPs	Dosage: 150 mg/L of MNPs, 60 mg/L of PFS; slow mixing time: 10 min	As (V)	< 10	Li et al. (2010)
MNPs + PFS			> 95	
MNPs@SiO ₂ + FeCl ₃	Dosage: 10 mL/L of FeCl ₃ , 1 g/L of MNPs@SiO ₂	Cr As Pb Cadmium (Cd) Copper (Cu) Zinc (Zn)	> 95	Mandel et al. (2013)
MNPs	Dosage: 0.45 mmol/L; ratio of MNPs to FeCl ₃ : 0.25: 1.0; pH: 7	Hg	60	
FeCl ₃		Molybdenum (Mo)	< 20	Ma et al. (2015)
MNPs + FeCl ₃			80	
Gum karaya (GK)-grafted poly(acrylamide-co-acrylic acid)-incorporated MNPs hydrogel nanocomposite [GK-cl-P(AAm-co-AA)/MNPs]	Dosage: 1 g; settling time: 15 min	Pb	95.8	Fosso-Kankeu et al. (2015)
		Cr	98	
		Nickle (Ni)	96.3	
MNPs + N,Nbis-(dithiocarboxy) ethanediamine (EDTC)	Dosage: 80 mg/L; hydraulic retention time: 3 min; electric current: 3.5 A	Ni	100	Qiu et al. (2017)
Magnetic cross-linked starch-graft poly(acrylamide)-co-sodium xanthate (M-CSAX) nanocomposite	Dosage: 40 mg/L; pH: 5	Pb	78	Wang et al. (2017b)
		Cu	63	
Chitosan-coated MNPs (CS@MNPs)	Dosage: 250 mg/L; pH: 5	Ni	74	Liu et al. (2018)
Poly(itaconic acid-g-CS)-coated MNPs (MNPs@PIA-g-CS)			98	

a combination of hydroxylation and colloids entrapment, which produced net negative charge in addition to the bridging effects (Fosso-Kankeu et al. 2014). The use of natural polymers such as CS or starch was more beneficial because they possess long loop-forming chain structures that attach to the other particles in water, resulting in metals bridging (Kolya and Tripathy 2013).

Organic-based wastewater treatment

Oilfields, landfills and palm oil industries discharge voluminous amount of effluents containing organic contents, such as oil and grease and chemical oxygen demand (COD). The lack of proper treatments for such effluents could cause interferences to aquatic life. One of the approaches to overcome the problem taken by scholars is by implementation of MCF on the wastewater containing these pollutants.

Oil contaminants from oilfields, industries or even oil spill accidents pose serious threats to surface water (Demirbas et al. 2017; Zhou et al. 2019). In the current review,

seven articles were identified with an objective to efficiently separate oil–water mixture by using MCF technique. The performances of the investigated approaches are summarized in Table 5. It is also worthy to mention that real oily wastewater collected from offshore oilfields was used in four out of seven studies (Al-Rubaie et al. 2013; Cai et al. 2017; Duan et al. 2017; Fang et al. 2016); meanwhile, the remaining studies used simulated oil emulsion (diesel-water mixtures) (Lü et al. 2017; Lu et al. 2018; Tang et al. 2019).

As shown in Table 4, the presence of MNPs enhanced the oil removal beyond 85% in most studies. The usage of MNPs alone without modification, however, demoted the performance, as demonstrated by Lü et al. (2017). Zeta potential analysis result revealed the negative charge of MNPs at all pH levels; thus, it is presumed that repulsion occurred between the MNPs and oil droplets, as the latter were always negative at a broad pH range. The performance was significantly improved upon the anchoring of aminopropyl (APFS) at acidic and neutral media. However, the separation performance was unsatisfactory in an alkaline

Table 4 Oily wastewater treatment via MCF

Type of MCF or its combination	Wastewater	Optimum conditions	Oil reduction (%)	References
FeCl ₃ + PAM	Synthetic oily water	Dosage: 20 mg/L of FeCl ₃ , 5 mg/L of PAM, 200 mg/L of MNPs; settling time: 4 min	> 83	Al-Rubaie et al. (2013)
FeCl ₃ + PAM + MNPs			> 98	
Poly(silicate aluminium) (PSA)	Fuling Coke Dam, China	Dosage: 0.6 g/L	85	Cai et al. (2017)
Composite MNPs/PSA (MPSA)			98	
PAC	Synthetic oily water	Dosage: 50 mg/L of PAC, 50 mg/L of MNPs, 10 mg/L of PAM; settling time: 3 min	80	Tang et al. (2019)
PAC + MNPs			85	
PAC + MNPs + PAM			> 90	
Magnetic N,N-dimethylethanolamine (MDMEA)	Offshore oilfield in China	Dosage: 2.0 g/L;	90	Fang et al. (2016)
Magnetic polyethylenimine (MPEI)		temperature: 65 °C; sonication time: 6 min	90	
MDMEA	Offshore oilfield in China	Dosage: 2.5 g/L; temperature: 65 °C	90	Duan et al. (2017)
Chitosan	0.2 wt% of diesel–water emulsion	Dosage: 0.5 g/L; pH: 4	< 10% transmittance	Lü et al. (2017)
MNPs			< 5% transmittance	
aminopropyl-functionalized MNPs (MNPs@APFS)			> 80% transmittance	
MNPs@APFS-grafted chitosan (MNPs@APFS-g-CS)			> 90% transmittance	
Quaternized chitosan (QC)	0.2 wt% of diesel–water emulsion of QC, 14 mg/L of MNPs-g-QC; pH= 4	Dosage: (0–1.8) g/L	0% transmittance	Lu et al. (2018)
MNPs-grafted QC (MNPs-g-QC)			> 90% transmittance	

solution. The problem was solved by grafting aminopropyl-functionalized MNPs (MNPs@APFS) with chitosan layer, which dramatically enhanced the oil separation under various pH conditions. The grafting step provided more positive charges onto the MCF; hence, negatively charged oil droplets could be effectively attached to MNPs and further aggregated with each other via electrostatic attraction. Accordingly, bigger oil droplets and magnetic flocs with a size bigger than 100 µm were formed rapidly and could be easily collected by magnet in less than 20 s. Different oil separation mechanisms were used to explain the reactions that took place in different media. In alkaline environment, MNPs@APFS-grafted chitosan (MNPs@APFS-g-CS) could still be attached onto oil droplets surface. However, hydrophobic interactions occurred when MNPs@APFS-g-CS acted like magnetic surfactant and tended to accumulate at the oil–water interface, imparting magnetic properties on emulsified oil droplets for magnetic separation. In another research done by Lu et al. (2018), MNPs grafted with

quaternized chitosan (MNPs-g-QC) did not benefit the oil separation efficiency when compared to the previous study. However, great advantages were seen in minimizing usage of chemicals, synthesis route of magnetic flocculants as well as dosage reduction throughout the treatment.

Besides that, the scholars were also interested in the replacement of conventional coagulants by synthetic polymers (with dense active sites) incorporated with MNPs for oily wastewater treatment (Sun et al. 2011). Since the density of the oil droplets is less than that of water, formation of large flocs is hindered when the amount of coagulant such as FeCl₃, poly(silicate aluminium) (PSA), PAM or PAC is too small. MNPs coupled with coagulant polymer do not only lead to higher oil removal efficiency, but also shorter flocs sedimentation time (Al-Rubaie et al. 2013; Tang et al. 2019). Moreover, Cai et al. (2017) explained that polyelectrolytes attracted abundant Fe²⁺ (from MNPs) species to form a three-dimensional porous mesh polymer. Such polymer strengthened the sweep flocculation and bridging adsorption



Table 5 Water body nutrients and microbes treatment via MCF

Type of MCF or its combination	Wastewater	Optimum conditions	Performance	References
MNPs Red mud + MNPs	Synthetic phosphate water	Dosage: 50 mg/L of red mud, 100 mg/L of MNPs; magnet working current: 160 A; retention time: 0.5 min; flow rate: 2 L/min	Phosphate almost 100%	Zhao et al. (2012b)
MNPs + PAC (MPAC) MNPs + PAC + PAM	Simulated water composed of humic acid, kaolin and 5% domestic sewage	Dosage: 15 mg/L of MPAC; 0.4 mg/L PAM; reaction time: 5 min; magnetic field: 0.5 T	Phosphate removal 95.7%	Lv et al. (2019)
PFS + MNPs	Cassava starch processing plant in China	Dosage: 30 mg/L of PFS, 7.5 g/L of MNPs; pH: 10	Phosphate removal > 80%	Du et al. (2019)
MNPs MNPs + polyglutamic acid	Phytoplankton in model seawater	Dosage: 0.2 g/L of MNPs, 0.2 g/L of polyglutamic acid	Phytoplankton separation: < 10% (MNPs) ~ 100% (MNPs + polyglutamic acid)	Sakaguchi et al. (2010)
PFC MNPs + PFC (MPFC)	Freshwater Algal Culture Collection of Institute of Hydrobiology	Dosage: 5 mg/L of PFC, 4 mg/L of MPFC; settling time: 60 min; pH: 7	<i>M. aeruginosa</i> removal: 30% (PFC) ~ 100% (MPFC)	Jiang et al. (2010)
PAC MNPs + PAC	Freshwater Algal Culture Collection of Institute of Hydrobiology	Mass ratio PAC to MNPs: 4:1; pH: 4; settling time: 10 min	<i>M. aeruginosa</i> removal: 33% (PAC) 73% (MNPs + PAC)	Zhang et al. (2015)
MNPs from fly ash	Chaohu Lake, China	Dosage: 200 mg/L of MNPs; settling time: 5 min	Chlorophyll- <i>a</i> removal > 99%	Liu et al. (2013)

to the fullest, in addition to the high surface energy of MNPs conducive to remarkable oil separation in oily wastewater.

By taking different approaches, Duan et al. (2017) and Fang et al. (2016) investigated the effects of ultrasonic time and temperature on the removal of oil. The oil removal had a dramatic increase when the ultrasonic time exceeded 4 min, which is from 20% to 89.4% at 6 min at elevated temperature (55 °C). At these conditions, the adsorption of oil pigments on the magnetic *N,N*-dimethylethanolamine (MDMEA) surface was sped up. The type of polyether, either *N,N*-dimethylethanolamine (DMEA) or polyethylenimine (PEI), did not play significant effect in this case. As long as they possess the flocculation function, the MCF can be endowed with the same operation.

Landfilling inevitably implies the generation of leachate, which is a strongly polluted wastewater that must be treated prior to its discharge into receiving waters, and the removal of organic matter is always a prerequisite (Ribera-Pi et al. 2020). In the present review, authors identified two studies, which recommended the adoption of MCF for leachate treatment. Both approaches used a conventional flocculant, namely PFS and PAC, coated with MNPs to form a flocculant unit. Knowing the oxidation ability possessed by MNPs, Liu et al. (2016) integrated the MCF and advanced oxidation process (AOPs) (mainly persulphate, $S_2O_8^{2-}$) for the removal of COD and colour of leachate. They believed such

method is the most suitable treatment method for wastewater containing different organic compounds that are non-biodegradable and/or toxic to microorganisms. The result showed that combination of AOPs and MCF boosted the leachate treatment performance when compared to when MCF alone was used. This is because oxidation by sulphate radicals activated Fe^{2+} in MNPs during C-F process (Asha et al. 2017). Due to the higher availability of Fe^{2+} combined with the coagulation ability of PFS, the charge density of flocculant is increased for the entrapment of organic pollutants. Based on a comparison with leachate treatment without AOPs, they concluded that combination of MCF and AOPs could decrease the dosage required for the treatment that meets the discharge limits. Finally, the studied process efficiently reduced the residual contaminants, COD and colour of leachate to 73% and 95% of the original values, respectively.

Liu et al. (2017) synthesized a magnetic coagulant by introducing MNPs particles into PAC (MFPAC), which showed significantly better coagulation performance for COD and colour removals from landfill leachate, when compared to the use of PAC alone. The advantages related to such combination are as follows: (1) MNPs acted as cores of floc in the flocculation process, consequently improved the density and settling property of the flocculation body; (2) the magnetic property possessed by MNPs favours the MNPs combination with the PAC body during C-F process to form



more compact magnetic flocs; and (3) MFPAC enhanced the mutual attractions between magnetic particles and thus more particles form large aggregates, ultimately leading to high good coagulation performance (Kushida et al. 2013). However, the dosage of MFPAC had to be controlled precisely, as further increase over optimum dosage resulted in undesired effects. Increased MFPAC dosage signified the presence of more MNPs, which eventually led to self-coagulation among MNPs. Thus, these particles could no longer combine with the flocculation body in order to facilitate heterogeneous nucleation, thus leading to poor coagulation performance (Liu et al. 2017). The final results obtained using MFPAC were 60 and 68% for COD and colour removals, respectively, which were 12 and 13% higher compared to the flocculation when only PAC was tested.

Palm oil mill effluents (POME) are common wastewaters produced in the edible oil industry. It is claimed to be one of the most difficult wastewater to be handled due to its massive production loaded with high suspended solid (up to 80,000 mg/L) and organic content (50,000 mg/L) (Lee et al. 2019). Direct release of POME to water streams or rivers without treatment causes water quality deterioration and aquatic pollution. Despite many studies utilizing C-F for POME treatment, Saifuddin and Dinara (2011) successfully developed chitosan–MNPs nanocomposites (magnetic chitosan) for turbidity, TSS, COD and oil removals, and the performances were also compared to the process where chitosan was used alone. The magnetic chitosan produced better performance than chitosan due to the increment of charge density. Polymer adsorption increased as the charge density increased, and this signified the rapid destabilization of particles (Bhatia et al. 2006). The performance of magnetic chitosan coagulant was also largely dependent on the nature pH of POME, and the highest removal was attained in acidic environment below isoelectric point. The amine functional groups in chitosan and the positively charged MNPs (at pH 4.8) helped to coagulate and adsorb the anionic pollutants in POME (Wang et al. 2018). When the charges were neutralized, the small suspended particles were able to interact with each other through rapid mixing, and macroflocs produced were subjected to the sedimentation process.

Noor et al. (2018) also developed an approach for the synthesis of cellulose cross-linked MNPs (MagCell) as a novel flocculant. The study focussed on optimizing preparation condition of MagCell and its effect towards the removal of turbidity, TSS, colour and COD of POME. Their findings showed that equivalent ratio between MNPs and cellulose, combined with moderate concentration (1.5 mL) of cross-linkers, was adequate to produce the most sophisticated POME treatment performance. Under such optimum conditions, MagCell possessed satisfactory capability to destabilize colloidal particles while providing sufficient active sites for adsorption of pollutants onto the MagCell flocculant. As

expected, MagCell showed higher removals with an average of 35% higher than cellulose under the same conditions. Thus, it is concluded that the presence of MNPs and cross-linkers between both materials improved the C–F performance. As pH plays an important role in POME treatment, a study on the effects of this parameter in POME treatment will offer clear insights on the MCF mechanism.

Water body mineral and microbes

Minerals such as phosphates are essential ions to promote plant growth and maintain soil freshness. However, excessive contents of these minerals in a water body could lead to eutrophication, characterized by rapid growth of algal and other aquatic microbes. This phenomenon destructs the marine system and deteriorates the quality of drinking water. Thus, a proper treatment is required to minimize such threat. In this SLR, the authors identified seven studies related to the removal of phosphate and microbial organisms from wastewater using MCF technology. The published works on the performance of MCF in mitigating water minerals and microbes are summarized in Table 5.

For the removal of phosphate, Zhao et al. (2012b) added MNPs as magnetic seed to the inorganic coagulants extracted from red mud. In the absence of magnetic seeds, the phosphate and inorganic coagulant yielded non-magnetic flocs that settled without the aid of magnetic force, resulting in very slow phosphate removal rate. The presence of magnetic seed increased the number of successful binding sites with non-magnetic flocs. Since only phosphate magnetic flocs could be separated during magnetic separation, the phosphate removal efficiency improved in the presence of MNPs. Addition of the MNPs/PAC composites followed by PAM improved the phosphate removal efficiency and thus was claimed to be the best dosing strategy (Lv et al. 2019). Al hydrolysis in PAC caused the formation of insoluble complexes of Al-hydroxy phosphates that dominated the phosphorus removal cycles in MCF. Moreover, MNPs served as nuclei that enhanced the formation of Al-hydroxy phosphate complexes with higher density, size and strength by allowing Al species to adsorb onto their surface. Meanwhile, PAM polymer coating formed an electrostatic barrier on the surface of MNPs and facilitated well embedding and uniform distribution of MNPs.

A recent study conducted by Du et al. (2019) proved that combination of PFS and MNPs as a new MCF system could replace traditional calcium oxide (CaO), which is widely used as a dephosphorization agent. The treatment using CaO precipitation method often produces unsatisfactory results owing to the production of ultrafine hydroxyapatite particles, which restrict the removal of phosphate. The new MCF system significantly reduced the phosphorus content from 134.34 to 10.88 mg/L



when 7.5 g/L of MNPs was applied. The improvement in this MCF performance is due to electrostatic attraction forces and van der Waals forces, which contributed to the agglomeration of the MNPs, and facilitated magnetic flocculation.

Excessive growth of marine microbes also reduces the water quality; thus, a proper treatment is required. Sakaguchi et al. (2009) established a high-speed magnetic separation treatment system for large-scale removal of adventive aquatic microorganism in seawater. They used phytoplankton in the study, and the performance was measured based on the number of cells per volume of simulated water. The use of MNPs alone as magnetic seeds did not produce satisfactory results. Addition of flocculant (polyglutamic acid), however, reduced the separation rate, indicating the important role of polyglutamic acid as a disperser. MNPs separation using superconducting magnet was identified as a possible method in separating plankton after MCF process. In the magnetic separation experiment, as much as 100% of magnetically seeded aquatic organisms were separated without flocculant. In the case with flocculant, partial floc drainage took place. The study proved that magnetic seeding of the aquatic organism is a possible strategy to achieve high efficiency in flocculation. However, flocculant amount optimization is necessary to control the re-dispersion or piling up of the particles on the magnetic separation unit.

Cyanobacterial *Microcystis aeruginosa* (*M. aeruginosa*) is known to produce several types of toxins as well as unpleasant tastes and odours, which cause severe water quality issues. Jiang et al. (2010) and Zhang et al. (2015) investigated the effects of MNPs combined with different inorganic flocculants, namely polyferric chloride (PFC) and PAC, respectively, on the removal of *M. aeruginosa*. The higher removal efficiencies shown by MCF when compared to C–F were mainly attributed to the adsorption effect of MNPs. It is postulated that in the MCF system, *M. aeruginosa* cells first reacted with positively charged MNPs to form large complexes. Then, the unoccupied cell surface reacted with PFC or PAC to achieve higher removal. In this case, pH played a major role in MCF mechanism, as such factor was responsible for the surface charge of *M. aeruginosa* and MNPs, as well as the hydrolysis degree of both PFC and PAC in the C–F process.

Lastly, Liu et al. (2013) converted fly ash (major waste of thermal power plants) to magnetic coagulant for rapid removal of algae from water body. A preliminary study on the mechanism indicates the roles of both physical adsorption and chemical coagulation effects exerted by the modified fly ash on the algal cells. MNPs became the nuclei of the floc cohesion, which enables the separation of algae under the action of magnet.

Ultrafine particles

Most of the iron mines in Asia produces tonnes of tailing slurry with a mass concentration of 20% (Zhang et al. 2017a). Due to the low-grade ore, serious argillation and high oxidation, the super-large-scale argillized ultrafine tailings slurry (SUT) is full of negatively charged slimes (Li et al. 2016). The repulsive interactions between these particles lead to many problems, such as low settling velocity, high overflow turbidity and solids content; therefore, large flocculant dosage is often needed for effective C–F.

Many research findings indicate that fly ash is a plentiful, convenient, and cheap magnetic seed owing to the abundant magnetic particles, which can accelerate the flocculation and sedimentation of the SUT within the magnetic field (Han et al. 2015). A fly-ash-based magnetic coagulant (FAMC) was developed by Li et al. (2016) to accelerate the SUT settling. The coagulant was prepared via acid leaching of fly ash to remove unnecessary surface structures to get better contact with SUT. FAMC did not display any effect on SUT flocculation in the absence of flocculant (i.e. PAM). The strong negative charge of SUT and its polarity towards water caused the formation of thick hydration shells. However, FAMC was rich in ground MNPs and cations (Fe^{3+} and Al^{3+}), which could neutralize the anionic shells of SUT and led to weaker repulsions, thinning of the hydration shells and lowering the zeta potentials. These changes in turn resulted in a better sedimentation of SUT, especially under the action of magnetic field. However, the excessive energy applied in magnetic field adversely affected the sedimentation process, as FAMC settled too quickly due to strong magnetic force. Such phenomenon limited the contact reaction between FAMC and SUT. The same observation was also discussed by Zhang et al. (2016). These studies also concluded that FAMC application increased the SUT sedimentation by 65%, and the usage of flocculant can be reduced by 37%. The use of FAMC is expected to promote the recycling of ~70,000 tons/year of waste fly ash, which provides a better method of waste recovery.

In recent years, the removal of titanium oxide nanoparticles (TiO_2) from water has emerged as an urgent issue, due to the associated acute toxicity and sub-lethal effects to the aquatic organisms upon long-term exposure to such pollutants (Skocaj et al. 2011). As a result of the expansion in TiO_2 applications, the increasing generation and irresponsible disposal of nano-sized particles in large quantities may be harmful to human and ecosystems. In a work performed by Bakhteeva et al. (2019), MNPs coated with silicon oxide (MNPs- SiO_2) and iron nanoparticles coated with carbon together with attached sulfonic acid groups ($\text{Fe-C-SO}_3\text{H}$) were used to remove non-magnetic TiO_2 nanoparticles from water. The study revealed that the separation efficiency and sedimentation were mainly due to the



hetero-aggregates formation and the related settling velocity in water. The aggregate sizes in the mixed (TiO₂)–(MNP-SiO₂) and (TiO₂)–(Fe–C–SO₃H) water suspensions were maximum in an acidic media. It was also discovered that the opposite surface charges of TiO₂ particles and MNPs with magnetic core facilitated the heteroaggregation during the magnetic sedimentation process. In another study conducted by Leshuk et al. (2018), separation of TiO₂ was performed using MNPs coated with different polymers, namely poly(diallyldimethylammonium chloride) (PDADMAC) and chitosan as positively charged polyelectrolytes (PEs) and poly(sodium 4-styrenesulfonate) (PSS) and poly(acrylic acid) (PAA) as negatively charged PEs as new formulation in MCF. The study concluded that MNPs coated with PDADMAC were capable of collecting ultrafine particles due to the loaded charge density and electrostatic association mechanism, without the overdosing issues associated with the flocculant.

Besides concerns on TiO₂, discharge of highly turbid wastewater containing high levels of extremely stable silica (SiO₂) nanoparticles from the backside grinder (BG) in the semiconductor manufacturing industry has also become a serious problem in the past few decades (Müller et al. 2017). Three articles identified in the present review report the effective removal of SiO₂ from the BG wastewater by adopting MCF technology. Ryu et al. (2016) used MNPs synthesized via co-precipitation method to evaluate the effectiveness in removing SiO₂ from the BG wastewater. The SiO₂ removal was significantly influenced by the particle size of the MNPs. The applications of nano-scaled MNPs dramatically reduced the amount of MNPs used in the removal of SiO₂. The pH was also an important parameter in SiO₂ removal, which remained at about 98% up to pH 8. At pH value higher than 8, the removal decreased drastically to zero removal (at pH 12) with an increasing residual Fe concentration due to the detachment of Fe from the magnetite surface. The stirring intensity and duration were the other factors affecting the enhancement of SiO₂ removal besides magnetic field strength and sedimentation.

A study performed by Wan et al. (2011) incorporated MNPs with PAC to yield better reduction of turbidity and SiO₂ content in the BG wastewater. By using this method, the MNPs completely captured the silica nanoparticles via electrostatic attraction, due to the opposite zeta potentials of these compounds. Meanwhile, PAC attached to the nanoparticles has been combined with MNPs to form flocs for better separation. Taking different paths, Shen et al. (2013) investigated the effects of ultrasound regeneration on the reuse cycles of MNPs and application in BG wastewater. Prolonged regeneration time of magnetic seeds using ultrasound did not benefit the MCF performance. Such actions also reduced the particles or clump size of the MNPs and probably altered the characteristics of the MNPs. The

ultrasound application duration of 1 min was sufficient for the separation or desorption of MNPs and silica particles. Without the ultrasound application in MNPs regeneration, the reuse cycle of MNPs was only one time. In comparison, the reuse cycles of the samples regenerated by ultrasound could be increased up to five times. The results showed that enhancement of regeneration of MNPs enhanced by using ultrasound did not only increase the reuse cycles of the magnetic seeds, but also lessened the usages of coagulants and reduced the quantity of chemical waste sludge.

Textile/dye effluents

MCF has also been applied in textile/dye wastewater treatment, as indicated by the four publications analysed in this study. Wen-song et al. (2011) found that the addition of MNPs in Fenton oxidation—high gradient magnetic separation system—led to removal of colour (92.6%) and COD (79.5%) in the dye wastewater. The addition of magnetic powder enabled magnetic performance by non-magnetic pollutants to facilitate the purification by magnetic separation as long as the current intensity was controlled (Gómez-Pastora et al. 2017). The high-gradient magnetic separation technology showed several prominent characteristics such as small occupied area and large treatment amount. The experiment also showed that MNPs seed can be regenerated with recovery rate over 90%, and the effectiveness of the regenerated MNPs was in the reuse that was almost equivalent to those of the original MNPs. The pre-polymerized ion-based PFS combined with MNPs (MPFS) was synthesized by Chen et al. (2019) for removal of Congo Red, a typical anionic dye. The dye removal value increased from 29.5 to 64.8% with the increased MNPs dosage, indicating the significant positive effects exerted by MNPs on the C-F treatment for textile dye wastewater.

Two publications (dos Santos et al. 2018a; Reck et al. 2019) report the use of protein coagulant, namely MO seed saline extract functionalized with MNPs for textile wastewater treatment in the presence and absence of external magnetic field. The association of MNPs with the compounds present in MO seeds extract resulted in magnetic coagulation while maintaining the characteristics of both materials and facilitated the dye or colourant's sedimentation through aggregation to form magnetic flakes (Yogalakshmi et al. 2020). Such sedimentation was visible when the process was exposed to an external magnetic field. Moreover, the positive zeta potential of isolated MO suggested that the mechanisms involved in the C-F process were charge neutralization and adsorption (Mangale Sapana et al. 2012). In the meantime, similar zeta potential magnitude obtained by MO-functionalized MNPs indicated that possibly the same mechanisms were possibly involved.

Pharmaceutical wastewater

A great variety of pharmaceutical intermediates are being produced worldwide annually for medicine, extract and pesticide applications (Melero et al. 2009). Nearly half of the pharmaceutical wastewater produced is discharged without proper treatment, causing severe environmental problems (Vara et al. 2020). To solve this problem, a process integrating continuous coagulation, magnetic-enhanced flocculation and membrane filtration, denoted as recycling magnetic flocculation membrane filtration (RMFMF), was developed by Wang et al. (2017c). The aim of such process was to reduce the content of tetracycline, a micro-pollutant which is often used in pharmaceutical and personal care products, in the surface water. With the same target, Yu et al. (2015) developed a composite MNPs-ferric chloride followed by ultrafiltration as their treatment component. Meanwhile, Tian et al. (2015) adopted MCF to improve effluent quality after membrane-aerated biofilm reaction for the treatment of pharmaceutical wastewater, which contained high total nitrogen (TN) concentration. The performance was optimum at moderate MCF conditions (i.e. 1.2 g/L of PAC combined with 2 mg/L of PAM and 9 g/L MNPs) with the TN removal efficiencies varied in the range 90.1–90.7%. The results also met the effluent quality standard for industrial reuse in China (GB/T 19923-2005).

All of these studies depict that MCF application improved the quality of treated effluents either when being used alone or in hybrid system. MCF led to the appearance of destabilized colloids and small flocs fragments, which could be deposited and adsorbed, enhancing the collisions and aggregations of particles in water to form even larger flocs after the breakages of original ones (Vadasarukkai 2016). In addition, the magnetic force exhibited in the synthesized magnetic flocculant could force the suspended particles in water to generate larger flocs. While the presence of MNPs in filtration stage (ultra or membrane filtration) had no adverse impacts, their presence within residual coagulant flocs led to their accumulation in the filter cake layer for easy separation. Future studies are required to gain a better understanding on the interactions between MCF and pharmaceutical components, as well as to determine the effects of different coagulant properties and coagulation conditions (e.g. pH, temperature, mixing rate, etc.) towards the process performance.

Others

MCF also found applications in sludge water content reduction and drum cans wash water treatment, albeit with lower interest among researchers, as only one article describes such application. Sludge is a major by-product of municipal and industrial wastewater purification activities.

Improper control of water content complicates the handling and disposal of sludge and consequently increases the total treatment cost (Guyer 2018). Concerning this problem, Lakshmanan and Kuttuva Rajarao (2014) investigated the use of MNPs in C–F for reduction of water content in sludge sediment. This study also compared the performances of chemical (ferrous sulphate) and natural (protein from MO seed) coagulants. A maximum reduction of 95% sludge water content was achieved in 25 mg/L of MCF at 20-min mixing time, whereas only 20–30% sludge water reduction was achieved with the application of chemical and natural coagulants. Even though MNPs required higher dosage compared to the chemical- and protein-based coagulants, MNPs showed a stable decrease and the highest reduction in sludge water content at longer mixing time. The possible reason for the lowest sludge content via MCF was the catalytic activity possessed by the iron in MNPs, which increased the volatilization in sludge content during the treatment. In comparison, when chemical- or protein-based coagulants were used, the formation of complex molecules via precipitation was observed in C–F process and resulted in higher sludge water content. In addition, a comparative study of sedimentation kinetic also proved that the presence of MNPs in C–F shortens the treatment process time to < 10 min. In contrast, chemical and natural coagulants needed 60 min to attain a satisfactory performance. This study also proved that sludge dewatering via MCF is the new approach for a fast and easy large-scale sludge separation.

Among other methods of sludge water reduction process, electrokinetic showed 73% dewaterability in 4 h at constant applied voltage (Yuan and Weng 2003), while biodrying is another concept which is gaining interest due to the low electrical consumption (0.46 MW) required to reduce the total sludge weight by 73% at specific conditions (Winkler et al. 2013). These processes are far inferior to MCF, which achieves 95% sludge water content reduction in less than 10 min in the presence of external magnetic field.

Many factories use drum cans for storage purposes, and the used drum cans are often washed prior to reuse. The washing process generates large amount of water, approximately 70 L per drum, which requires treatment prior to discharge (Mishima et al. 2010). The washing wastewater consists various types of oil constituents, COD and chemicals depending on the sources; thus, a proper treatment is required to remove these pollutants. Mishima et al. (2009) introduced MNPs together with PFS in a C–F system to reduce COD level in the washing wastewater. The COD value was reduced from 500 to 100 ppm with the installation of the magnetically assisted separation device. It can be concluded that MNPs sped up the C–F process; however, effective capture of micro-size flocs remained an issue. To overcome such challenge, a permanent magnet was added to



generate higher magnetic field for better flocs capture without affecting the COD level of treated water.

Perspectives

Application of MCF in water treatment

The systematic review based on the 62 papers on MCF led to several important findings. Firstly, increasing number of annual publications related to application of MCF in wastewater treatment was observed. Such trend reflects the increasing attentions from the scholars on the sustainable wastewater treatment using innovative solutions. The existence of international collaborations within scientific community also drives the technology development in waste treatment, especially in C–F process.

In-depth investigations of MCF

The key information in the analysed publications enables prediction on the plausible C–F mechanism and influence of the coagulant/flocculants' structural features on the mechanism. One among the challenges in such studies is the difficulties in analysing the properties of magnetic-based flocculants, including their short- and long-range structure, which affects the flocculants interaction with the targeted pollutants. As different types of coagulants/flocculants are compounded with magnetic materials, the existence of various forces, such as covalent, electrostatic interactions, van der Waals forces, electromagnetic forces or highly specific forces involved in these interactions, could lead to the occurrence of C–F via different mechanistic routes (Gregory 2013). Moreover, the use of different synthesis approaches between MNPs and coagulant/flocculant(s) such as grafting, cross-linking, thermal-induction, and impregnation, could influence the architectures of the synthesized samples and subsequently the MCF performance. The development of advanced technologies and more sophisticated analytical instruments are required to uncover the influences of these factors. Nonetheless, by taking advantage of the well-known C–F mechanisms, the structural characteristics of magnetic coagulant/flocculants can be tailored to optimize the respective MCF performance. The interdependencies of preparation conditions, structural features, C–F performance and separation performance are illustrated in Fig. 3. In order to achieve the desired MCF performance and optimize C–F process, exact molecular control is important for the selection or postulation of the optimal magnetic coagulant/flocculants. This model was also suggested by Yang et al. (2016), and the strategy was applied to the preparation of high-performance chitosan-based flocculants.

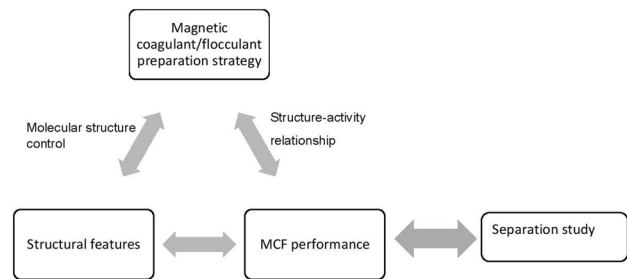


Fig. 3 The general relationship between magnetic coagulant/flocculant preparation, MCF performance and separation performance

Several types of magnetic coagulant/flocculants, which include synthetic magnetic polymer flocculants, natural magnetic coagulants and various combinations between those coagulants, have been developed to enhance the C–F features. The effectiveness of these magnetic coagulant/flocculants is highly dependent on the composition and characteristics of wastewater to be treated. Analysis of the retrieved publications in this study reveals two classes of magnetic coagulant/flocculant(s), namely dual magnetic coagulant/flocculant (DMCF) and surface-functionalized magnetic coagulant/flocculant (SMCF). In the MCF process, MNPs and coagulant/flocculant were added separately in the DMCF system. On the other hand, modification of the surface of MNPs and coagulant/flocculant using appropriate methods (e.g. composite, cross-linking, grafting) resulted in SMCF with distinguished characteristics. Investigations on DMCF could reveal the role of each material used in the C–F mechanism and how the materials affect the performance of C–F. However, this route is expected to consume larger space and longer treatment time as coagulation–flocculation has to precede the separation step. On the contrary, the dual functionalities of SMCF allow simultaneous execution of the C–F process. Many studies showed that SMCF produced a better treated water quality with less sludge formation when compared to DMCF. Moreover, investigations on SMCF synthesis provide researchers with new knowledge related to the chemistry of coagulant/flocculant, which is important to produce SMCF with high quality and efficiency in the MCF process (Maksoud et al. 2020).

Magnetic flocs properties analysis

While the main focus in this subject field lies in the correlation of molecular structures of magnetic coagulant/flocculant and the treated pollutants, the knowledge of the properties of the flocs formed (flocs size, flocs strength, flocs morphology as well as dynamic flocs formation scheme) during MCF process must be considered for better understanding of the C–F mechanisms. Image analysis or light scattering are the most applied techniques in

studying the said properties along with the fundamental of fractal theory (Glasgow and Kim 1989). In MCF process, fractal dimension is the most crucial parameter in the fractal concept of the flocs, which explains the space-filling capacity (i.e. flocs compactness), while compactness of the flocs is directly proportional to the fractal dimension. The two factors that are commonly used in fractal dimensions are two- (D_2) dimensions and three-dimensions (i.e. mass fractal dimension, D_F). D_2 shows a power law relationship between the projected area (A) and the total scattered light intensity (I) (Eq. 1) for the floc structure. Meanwhile, D_F depicts the power law relationship between mass (m) and I , which can be obtained by light-scattering technique, since I is linearly related to m , and the scatter vector (Q) is inversely proportional to I (Eq. 2) (Xiong et al. 2019).

$$A \propto I^{D_2} \quad (1)$$

$$I \propto Q^{-D_F} \quad (2)$$

A few studies on MCF included flocs characterization results and comparisons between magnetic and non-magnetic flocs to support the proposed MCF mechanisms. For example, a comparison of flocs characteristic between cationic PAM (CPAM) and magnetic CPAM (MCPAM) on the removal of turbidity under various flocculant dosage and pH levels was reported by Ma et al. (2018). The flocs size observed when using CPAM was obviously lower than that of MCPAM was used. Thus, the presence of MNPs in CPAM promoted the aggregation of the flocs. Besides flocs size, the flocs strength and recovery factor of MCPAM were also higher compared to when only CPAM was used. The MNPs in the system provide surfaces for flocs adsorption and bridging effect, which assisted the agglomeration of numerous tiny flocs particles (Zhao et al. 2018). The small size of the flocs produced by CPAM also indicates inadequate charge neutralization for high turbidity removal as well as flocs formation.

In addition to the factors discussed above, the effectiveness of MCF or conventional C–F process is associated with the effective collision aspects, hydrodynamic interactions, particle surface characteristics and flocs properties (Li and Chen 2012). The complexity of MCF is recognized not only in research on water treatment, but also other industrial applications, including oil extraction (Lee et al. 2015) and food and latex production (Yang et al. 2016). A comprehensive understanding of MCF is vital to achieve effective implementation. Multidisciplinary research involving the researchers in environmental science and engineering, nanomaterials field, and colloidal chemistry is necessary to expand the frontiers in this research topic.

Cost consideration of MCF

Cost is another bottleneck that restricts the practical application of MCF. To date, the investigation on the operating costs related to the MCF in water treatment is lacking. Studies have shown the benefits of MNPs in reducing dosage of conventional coagulant/flocculants (Medvedeva et al. 2015), which illustrate the attractiveness of coagulation/flocculation in water treatment. Li and Wang (2016) demonstrated that utilization of FAMC with PAM in MCF reduces the annual water treatment costs by 53 million Yuan. At the same time, PAM dosage can be reduced by more than 2100 t/a, which translates to 63 million Yuan saving. More costing analysis studies are required to evaluate the feasibility of the application of MCF in water treatment.

Another great advantage related to the use of magnetic coagulant/flocculants is the regenerability or reusability (Shen et al. 2013). Such property does not only reduce the usage of coagulant/flocculants, but also the cost. Various regeneration technologies, such as chemical extraction (Li et al. 2017a), bio-regeneration (Qi et al. 2017), as well as solvents (Ali et al. 2017) and ultrasound regeneration (Shen et al. 2013), have been introduced. Among these technologies, ultrasound regeneration is getting more attentions, as no additional chemical is required. Thus, secondary pollution can be avoided and the MCF process using the ultrasound-regenerated samples remains satisfactory even after 7–10 cycles (Duan et al. 2017; Lu et al. 2018; Mateus et al. 2018). However, in-depth studies on the economic aspect related to the ultrasound regeneration is required to verify such benefit.

Recommendations

The potential of MCF in wastewater remediation has been demonstrated by different independent research teams. As the food manufacturing industry and rubber industry are the largest contributors of organic pollutants according to United Nations Educational, Scientific and Cultural Organization (UNESCO) (Lapworth et al. 2012), feasibility of MCF in treatment of effluents from these industries should be investigated. It was also observed that about 26% of the analysed research works utilized coagulant/flocculant synthesized from natural sources such as chitosan, MO, and cellulose. Since green technology and sustainability approach play an important role in the environment and human well-being, the exploration of bio-magnetic-based coagulant/flocculants is highly recommended to minimize the usage of inorganic materials, which may lead to secondary pollution after MCF process. Similar to traditional C-F, the MCF process generates sludge as end products. Additional cost is necessary for the management of such sludge. Improper handling and

disposal of the sludge will contribute to secondary pollution, especially landfill contamination (Babatunde and Zhao 2007). Investigations on the ecotoxicology and economic factors of the MCF process are also needed to provide further insights on the selection of magnetic coagulant/flocculant prior to treatment and comparison with the traditional C–F. Sustainability assessment and lifecycle assessment can be executed to provide an intuition of the aspects that can be ameliorated for MCF using magnetic coagulant/flocculant. Lastly, the performance of MCF at pilot scale should be evaluated to reveal its potential in industrial water and wastewater treatment plants.

Limitation of this study

One limiting factor of this review is the criteria used to select relevant papers. Paper published in proceeding, non-peer-reviewed journals, books and papers written in languages other than English were excluded, which might result in omission of valuable data and findings. Similarly, the SLR was restricted to papers that focused on the application of MCF in water treatment as the key subject covered, and the studies on the other interrelated topics (e.g. application of MCF in different fields), which may, however, provide useful comprehensions on the MCF process. Likewise, the set of keywords used for the queries were based on existing works and the authors' knowledge on the topic. While it is presumed that such keywords cover the various aspects on this topic, the use of other keywords may lead to retrieval of different articles and hence different analysis and interpretations.

Conclusion

This paper presents a SLR on the applications of magnetic coagulation/flocculation in water treatment based on 62 articles retrieved from WoS and Scopus. The increasing number of annual publications demonstrates the scholars' interests in the potential of magnetic coagulants/flocculants in water treatment. The thematic analysis revealed eight themes, which reflect the major fields where MCF is applied. The "turbidity reduction" theme was placed in the first rank as the primary goal of MCF is to get rid of turbidity and SS. The remaining themes include heavy metals removal, organic-based wastewater, water bodies mineral and microbes, ultrafine particles, textile or dyes effluent. The existence of different themes show the diverse applications of MCF. An examination and discussions of the magnetic flocs properties and cost evaluation revealed that these are the important aspects that need to be considered to gain full understanding of MCF and its performance in water

treatment. This review offers several recommendations for future studies, including utilization of more biodegradable materials for the synthesis of magnetic coagulant/flocculants, as well as the need for more in-depth studies on flocs physico-chemical characteristics for further understanding of the fundamental C–F mechanism. Lastly, toxicological and economic implications related to MCF applications in wastewater treatment should be investigated.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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