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AN OVERVIEW OF NATURAL ORGANIC MATTER REMOVAL BY COAGULATION IN DRINKING WATER TREATMENT

BY

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Abstract. Natural organic matter (NOM) is equivalent to the total organic substances resulting from bacterial decomposition of animal and vegetal matter. NOM is naturally found in most surface water sources used for drinking water supply, and can have significant impacts on human health if it is not removed. Apart from the fact that they create problems with taste, odour and color of raw water, NOM species are precursors of disinfection by-products, which in turn have a negative effect on human health. Most of the NOM can be removed by coagulation and flocculation followed by sedimentation and filtration, processes that are considered the most common and economically feasible drinking water treatments. This study presents an overview of recently published investigations regarding NOM removal in drinking water treatment with different coagulant types and treatment techniques in relation to coagulation.

Keywords: coagulation, flocculation, natural organic matter.

Abbreviations: **AOPs** – advanced oxidation processes; **AC** – activated carbon; **AS** – aluminium sulphate; **BOD_5** – biological oxygen demand; **CF** – coagulation and flocculation; **COD** – chemical oxygen demand; **DBPs** – disinfection by-products; **DOC** – dissolved organic carbon;

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EC – electrocoagulation; **HA** – humic acids; **HAAs** – haloacetic acids; **HRT** – hydraulic residence time; **IE** – ion exchange; **MF** – microfiltration; **UF** – ultrafiltration; **NF** – nanofiltration; **RO** – reverse osmosis; **MIEX® -** magnetic anion exchange resin; **MBR** – membrane bioreactor; **NOM** – natural organic matter; **NTU** – nephelometric turbidity unit; **PA** – polyamide; **PAC** – polyaluminium chloride; **PAFC** – polyaluminium ferric chloride; **PATC** – polyaluminum titanium silicate chloride; **PASiC** – polyaluminium silicate chloride; **PCF** – ferric polychloride; **PD** – polydiallyldimethylammonium; **PDADMAC** – polydiallyldimethyl ammonium chloride; **PFA** – polyferric acetate; **PFC** – polyferric chloride; **PFS** – polyferric sulfate; **UV** – ultraviolet radiation; **SUVA** – specific UV absorbance; **THMs** – trihalomethanes; **TOC** – total organic carbon; **TS** – total solids; **TSS** – total suspended solids; **RE** – removal efficiency; **PCPs** – plant based products.

1. Introduction

Aquifers and surface waters are considered to be the major sources of drinking water supply. The quality of water resources is one of the main challenges that the whole humanity faces, and the raw water must correspond to quality standards that ensure the physical, chemical and biological safety of drinking water. Most of the water resources contain, along with suspended solids, pathogenic microorganisms (algae, protozoa and fungi), inorganic compounds and natural organic matter (NOM) (Dayarathne *et al*., 2021). The occurrence of NOM in waters appears because of the interactions between the hydrologic cycle and the biosphere and geosphere, namely from the decaying of plant and animal residues (Matilainen *et al*., 2010). NOM is defined as a mixture of heterogeneous hydrophobic acids, such as humic substances represented by humic acids, fulvic acids and humins (less soluble in water, high molecular weight, rich aromatic carbon, with conjugated double bonds and phenolic structures), and hydrophilic components less reactive represented by carbon and nitrogenous compounds (carbohydrates and proteins, sugars and amino acids) and others (Matilainen *et al*., 2010; Abu Hasan *et al*., 2020). The classification of NOM is presented in Fig. 1.

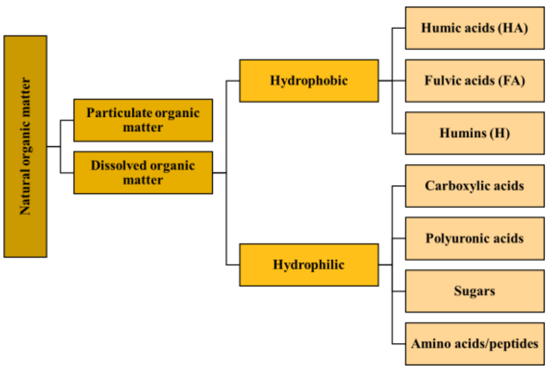

Fig. 1 – NOM classification (adapted from Abu Hasan *et al*., 2020).

The characteristics of NOM are profoundly affected by the type of water source and the surrounding environmental conditions (Dayarathne *et al*., 2021). A series of recent studies show the importance of NOM composition and concentration in the water source on the efficiency of the water treatment processes and drinking water quality, respectively (Krzeminski *et al*., 2019; Dayarathne *et al*., 2021; Mazhar *et al*., 2020) and the importance of the coagulant's aggregation mechanism (Ang and Mohammad, 2020; Dayarathne *et al*., 2021). To understand the physico-chemical behavior of NOM in water treatment processes, a characterization of NOM in the raw water is required by analyzing some quality parameters such as dissolved organic carbon (DOC), chemical oxygen demand (COD by Potassium Permanganate Method), specific UV absorbance (SUVA), pH, turbidity and color (Pan *et al*., 2016; Hua *et al*., 2020; Musteret *et al*., 2021). The certain components present in the NOM structure are recorded at different UV absorbance wavelengths. For instance, the absorbance recorded at 254 nm (UV_{254}) is corresponding to the aromatic groups with different activation degrees (Hua *et al*., 2020), the UV_{280} is associated with the presence of trihalomethanes (THMs) and haloacetic acids (HAAs), and UV_{365} is correlated with the presence of aquatic humic compounds (Musteret *et al*., 2021). SUVA, which is defined as the UV absorbance at 254 nm divided by the DOC concentration, has been widely used to characterize drinking water. Thus, high values of SUVA indicate that the NOM is mainly composed of hydrophobic, high molar mass organic compounds, whilst low SUVA values suggest the existence of mainly hydrophilic, low molar mass NOM compounds in raw water and drinking water (Pan *et al*., 2016; Hua *et al*., 2020; Musteret *et al*., 2021).

NOMs present in raw water may be suspended or dissolved and, if not removed, rise problems in the production of drinking water, such as: (i) modification of organoleptic properties (taste, odour, and color) of drinking water, (ii) the increasing chemical reagents demand in oxidation, coagulation, and disinfection, (iii) the forming of disinfection by-products (DBPs), such as THMs HAAs (Golea *et al*., 2017; Gilca *et al*., 2020), (iv) the fouling of separation membranes (Marais *et al*., 2018), (v) biological growth in water distribution systems, and (vi) enabling the transport of heavy metals and hydrophobic organic chemicals (Bhatnagar and Sillanpää, 2017; Mazhar *et al*., 2020).

Numerous technologies and methods have been employed to remove NOM in the treatment of drinking water, such as coagulation (Dayarathne *et al*., 2021) and adsorption processes (Joseph *et al*., 2012; Bhatnagar and Sillanpää, 2017), membrane filtration (Marais *et al*., 2018), advanced oxidation processes (AOPs) (Sillanpää *et al*., 2018a) as well as biological (Abu Hasan *et al*., 2020) and ion exchange (IE) processes (Levchuk *et al*., 2018). Fig. 2 shows a schematic representation of the main processes for NOM removal in drinking water.

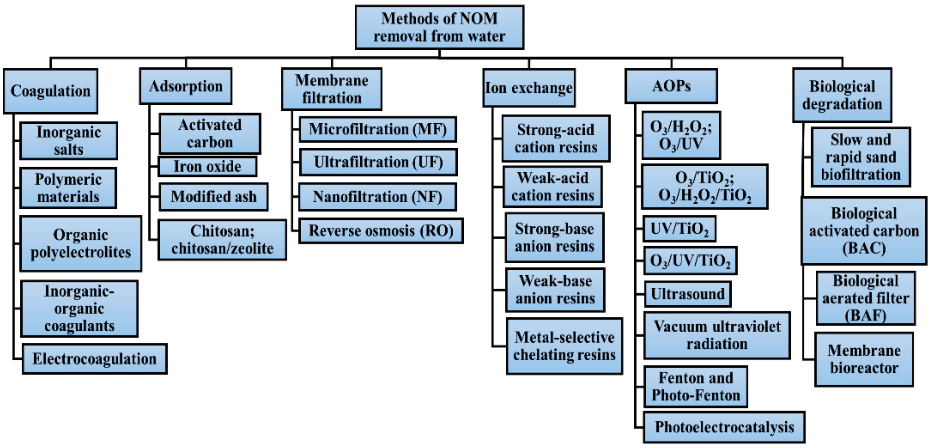

Fig. 2 – Main processes for NOM removal from water.

Advanced oxidation processes (AOPs) are powerful techniques generally applied prior to the coagulation stage to remove NOMs by using oxidants, mainly ozone (O_3) and/or hydrogen peroxide (H_2O_2), with different catalysts and/or radiation (UV, sunlight or artificial light). Thus, advanced oxidation processes include O_3/H_2O_2 peroxone process, UV/H_2O_2, UV/O_3, UV/TiO_2 photocatalytic oxidation process, Fe^{2+}/H_2O_2, Fenton and $Fe^{2+}/H_2O_2 + h\upsilon$ photo-Fenton processes, vacuum ultraviolet (VUV) and ultrasonic radiations (Sillanpää *et al*., 2018a). Membrane filtration technologies (microfiltration – MF, ultrafiltration – UF, nanofiltration – NF, reverse osmosis – RO) are also capable of reducing NOM. UF followed by RO has been proven as the best treatment process for removing natural organic compounds in raw water; UF also significantly reduces fouling of the RO membranes (Marais *et al*., 2018). Nonetheless, despite membrane processes high efficiencies to remove NOM, the implementation cost cannot usually be justified without additional benefits such as pathogen removal or salinity reduction (Sillanpää *et al*., 2018b). Another technique which is widely used in the field of water treatment is based on ion exchange. One of the most investigated and applied resin is the magnetic anion exchange resin MIEX®, which consists of a macroporous polyacrylic matrix in chloride form that contains magnetic iron oxide particles within its core (Sillanpää *et al*., 2018b; Levchuk *et al*., 2018). The usage of MIEX® resin to remove NOM in drinking water treatment demonstrated a good prevention of DBPs formation, a capability to remove hydrophobic and hydrophilic acids, to reduce membrane fouling, a decrease of coagulants and other chemicals required. However, after NOM removal from water, the regeneration and reuse of ion exchange resin represents an important issue (Levchuk *et al*., 2018). Adsorption process in water treatment field is widely used, NOM removal being usually realized by activated carbon, a well-known and used adsorbent. The problem is usually its cost and the regeneration and disposal of spent activated carbon (Kastl *et al*., 2015).

Therefore, most NOM can be removed by coagulation and flocculation (CF) followed by sedimentation and filtration, which are considered to be the most common and economically feasible processes to obtain drinking water (Dayarathne *et al*., 2021; Musteret *et al*., 2021). The main aim of this work was to provide an overview of recently published investigations regarding NOM removal with different coagulant types and treatment techniques in relation to coagulation for drinking water treatment.

2. Principles of coagulation-flocculation

In general, the CF process (Fig. 3) takes place in three stages: (1) charge neutralization by coagulation reagents addition; (2) formation of larger particles (flocs); (3) separation of flocs by sedimentation, filtration or flotation with dissolved air (Teodosiu, 2001; Jiang, 2015). The removal of NOM by coagulation from raw water for drinking purposes received attention worldwide, as it reduces the risk of formation of DBPs (i.e., THMs and HAAs), due to the lack of side effects with chlorine during the chlorination process (Liu *et al*., 2012; Bhatnagar *et al*., 2017). During CF processes, a combination of mechanisms is involved towards NOM removal such as charge neutralization, entrapment, adsorption and complexation (Okoro *et al*., 2021). The removal mechanism will be different for each type of NOM molecules in water, due to the different composition from one water source to the other, and within the same source due to seasonal variations (Matilainen *et al*., 2010).

By introducing chemical reagents into the raw water, conventional CF removes colloidal particles (turbidity) and partially reduces color, taste, odour, respectively the content of microorganisms and NOM. The dose of coagulant used in the conventional processes is not satisfactory for the simultaneous removal of turbidity and NOM, requiring the addition of an excessive amount of coagulant; this is a concept called enhanced coagulation (EC). By EC and flocculation, the removal effect of NOM and the precursors of DBPs are maximally improved in conventional treatment (Liu *et al*., 2012; Sun *et al*., 2019), and ensuring that drinking water DBPs concentrations comply with drinking water standards (Law 458/2002).

The most used coagulation reagents in drinking water production have been aluminium-based coagulants (alum, $Al_2(SO_4)_3$; aluminium chloride, $AlCl_3$; sodium aluminate, $NaAlO_2$) and iron-based coagulants (ferric chloride, $FeCl_3$, ferric sulphate, $Fe_2(SO_4)_3$), as well as the pre-polymerized inorganic compounds, calcium and magnesium salts, synthetic organic coagulants and natural-based coagulants. Regarding of flocculation reagents, activated silica, clays, and polyelectrolytes have been used frequently (Moud, 2022).

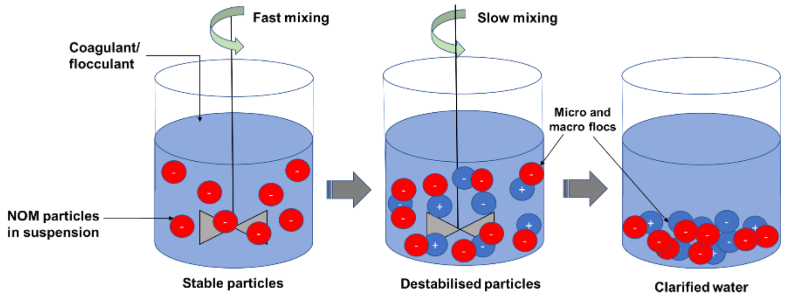

Fig. 3 – NOM removal during CF processes.

In terms of efficiency, NOM removal *via* CF is mainly affected by coagulant and flocculant type and dosage, mixing conditions, pH value, temperature of water, as well as the NOM properties (such as size, functionality, charge and hydrophobicity) (Dayarathne *et al*., 2021; Musteret *et al*., 2021). The nature of NOM has a considerable consequence on the coagulant dose. The hydrophilic fraction has a lower degree of removal as compared to the hydrophobic fraction and a higher coagulant dose is required (Bhatnagar and Sillanpää, 2017; Levchuk *et al*., 2018). Most studies in the literature show a high efficiency for the removal of NOM from raw water with high concentrations through the EC process. Remarkable results were obtained when combining the coagulation process with other processes, such as: advanced oxidation (Fenton processes, photocatalytic processes), ion exchange, filtration by activated carbon and membranes processes (RO/NF /UF/MF). Table 1 summarizes the efficiencies of the aforementioned processes (Liu *et al*., 2012; Sillanpää *et al*., 2018a; Sun *et al*., 2019). Pre-oxidation processes have been used effectively for the removal of the hydrophilic fraction, as well as in the case of pre-treatment with ion exchange resins. The coagulation process followed by the filtration processes proved high degrees of efficiency for the removal of NOM from raw waters.

Many investigations dealing with the comparison of coagulants effectiveness have been made. According to these studies, the use of Al-based coagulants has decreased due to the potential of Alzheimer's disease associated with residual aluminium (Exley, 2017), thus Fe-based coagulants are found more effective in removing NOM, especially for high and intermediate molecular mass compounds (1000 – 4000 g/mol) (Sillanpää *et al*., 2018b). Furthermore, the flocs formed during ferric coagulation are numerous and larger, about 710 μm as compared with 450 μm of flocs formed during aluminium coagulation (Jarvis *et al*., 2012), due the higher charge density of ferric coagulants (Umar *et al*., 2016). In order to overcome the limitations of metallic coagulants, polymeric coagulants have been developed. These polymeric coagulants showed better removal capacities towards NOM and other organic compounds from water (Lal and Garg, 2019; Adebayo *et al*., 2021). Lately, natural coagulants such as plant-based products (PCPs), have been studied and proposed as sustainable alternatives to synthetic coagulants due to their abundant availability, low cost, low sludge volume, disposal cost, and biodegradability (Okoro *et al*., 2021). However, the choice of adequate coagulant depends mainly on the characteristics of the raw water to be treated.

Table 1

Coagulation combined with other treatment processes and their removal efficiency

Treatment processes	Position in treatment train	NOM fraction removed	Removal efficiency, RE%
MIEX®	Before coagulation	Hydrophilic fraction, compounds with low molecular weights	10 – 30 % DOC
Oxidation processes	Before coagulation	Hydrophilic fraction, compounds with low molecular weights	5 – 32 % DOC 8 – 33 % UV_{254}
AC filtration	After coagulation	Hydrophilic fraction, compounds with low molecular weights	69 % DOC
Membrane filtration (UF)	After coagulation	Hydrophilic fraction, compounds with low molecular weights	73% DOC

Note: Removal efficiency (RE, %) was calculated by using Eq. (1):

$$RE = \frac{C_i - C_f}{C_i} \times 100, \% \quad (1)$$

where: C_i and C_f – the pollutant concentrations in influent and effluent expressed in mg/L.

The latest studies revealed that the use of coagulants in dual system (inorganic + organic) have higher effectiveness in removing turbidity and NOM, as compared to the single use of a coagulant. Table 2 shows the dose ratio of coagulants used in dual system (Matilainen *et al*., 2010; Sun *et al*., 2011; Lou *et al*., 2012; Jiang, 2015; Dayarathne *et al*., 2021).

Table 2

Dose ratio of dual coagulant system

Coagulants	Dose ratio
$Al_2(SO_4)_3$ + PA	7:8 mg/L
$TiCl_4$ + PD	0.5:0.3 mg/L
$Al_2(SO_4)_3$ + PD	8:3 mg/L
PCF + PD	1:14 mg/L
PAC + PD	3:0.5 mg/L
PAFC + $FeCl_3$	3:1 mg/L
$Al_2(SO_4)_3$ = aluminium sulphate; $TiCl_4$ = titanium tetrachloride	

3. Aluminium based coagulants

Alum [$Al_2(SO_4)$] and $AlCl_3$ are the most used coagulants within this group. The use of these coagulants was found to be sensitive to low temperature and low levels of pH. Moreover, aluminium residual concentration in the treated water may cause possible health diseases or other problems in distribution system. In order to avoid a low-quality of treated water, a pH control and an optimized coagulant dose are required (Matilainen *et al*., 2010; Sillanpää *et al*., 2018b). For instance, by increasing the coagulant dose NOM removal is not significantly improved because low molecular mass compounds are difficult to be removed. Verma and Kumar (2018) obtained high removal efficiencies at an alum optimal dose of 3.8 g/L, the dose increasing to 4.3 g/L conducting to a decrease of RE or it remained constant. Also, Lal and Garg (2019) used a higher dose of coagulant and no significant change in the removal degrees was observed. In another study, Wang *et al*. (2009) used as a coagulant $AlCl_3$ and observed that, by increasing the total hardness, the parameters UV_{254}, TOC and HA obtained good removal efficiencies.

Pre-hydrolyzed aluminium coagulants, *e.g.* polyaluminium chloride (PAC), have been developed by partially neutralizing $AlCl_3$ at different basicity ratios. These Al-species (A9 or A16) are considered to be efficient at floc formation due to their larger size and higher positive charges (Gkotsis *et al*., 2017). Furthermore, the pre-hydrolyzed polymer coagulants have been reported to enhance the removal efficiency of NOM (Gkotsis *et al*., 2017; Lal and Garg, 2019), although contradictory results have been found in some cases. When PAC coagulant was used for water with low value of DOC, the efficiency decreased, even if the dose was higher due to colloid restabilization (Musteret *et al*., 2021).

An overview of Al-based coagulants used on NOM removal from water or synthetic water in recent research studies is given in Table 3.

Table 3

Overview of recent research studies on NOM removal from water using Al-based coagulants

Coagulant type	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
$Al_2(SO_4)$	pH=5 dosage=0.5–3 mM	29% DOC 69% color 42% UV_{254}	Ferric-based coagulants removed a greater proportion of most of the DOC fractions, color, and UV_{254}.	Umar *et al*., 2016

Coagulant type	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
$Al_2(SO_4)$	optimum dose = 3.8 g/L pH=6	80% COD; 81% TSS; 90% turbidity; 90% NH_3; 98% NO_3; 99% PO_4	As the alum dose increased to 4.3 g/L, COD and turbidity decrease while TSS remained constant	Verma and Kumar, 2018
$AlCl_3$	1g/L HA UV_{254}=0.572 TOC=24.3 mg/L pH=7.32 dosage=7 mg/L	95% UV_{254} 92% TOC	The UV_{254} removal efficiency increased with increasing total hardness. The TOC removal efficiency increased with increasing total hardness. $CaCl_2$ can bind with HA, the absorbance of HA solution increases with increasing total hardness.	Wang *et al.*, 2009
PAC, PAFC	water pH = 11.1 coagulant dose=100 – 600 mg-Al/L	At a dose of 300 mg-Al/L: 98% color; 86% lignin; 66% TOC	PAC exhibited the best performance for organics removal among all coagulants. No significant change in removal from these parameters was observed with higher coagulant dose.	Lal and Garg, 2019
PAC-A9 (9% Al_2O_3)	BOD_5=355 mg/L HRT=7h	96.6% BOD_5; 96.2% COD; 90.4% NH_4^+-N; 92.3% TOC; 84.3% $UV_{254;}$ 96.6% turbidity	C/F pretreatment used to mitigate membrane fouling in MBR system. Lab-scale MBR system.	Gkotsis *et al.*, 2017
PAC-A16 (16% Al_2O_3)	BOD_5=355 mg/L HRT=7h	97.5% BOD_5; 96.4% COD; 72% NH_4^+-N;		

Coagulant type	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
		84.6% TOC; 83.7% UV_{254}; 94.4% turbidity		
PATC	low-turbidity water=10 NTU coagulant dosage=9 mg/L pH=8 stirring speed=50 rpm settling time=50 min T=50 °C HA=10 mg/L	95.6% turbidity 0.51 NTU	The turbidity of water decreased with the increase of PATC dosage.	Liao and Zhang, 2018
PAC-1	Synthetic water 1g/L HA UV_{254}=0.572 TOC=24.3 mg/L pH=7.32 coagulant dose=13 mg/L	93.5% UV_{254} 90%TOC	The UV_{254} removal efficiency increased until total hardness was 4mmol/L. The TOC removal efficiency increased with increasing total hardness. $CaCl_2$ can bind with HA, the absorbance of HA solution increases with increasing total hardness.	Wang *et al*., 2009
PAC-2	Synthetic water 1g/L HA UV_{254}=0.572 TOC=24.3 mg/L pH=7.32 coagulant dose=8 mg/L	94.5% UV_{254} 91% TOC	The UV_{254} removal efficiency increased with increasing total hardness. The TOC removal efficiency increased with increasing total hardness.	Wang *et al*., 2009

Coagulant type	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
			$CaCl_2$ can bind with HA, the absorbance of HA solution increases with increasing total hardness.	
PACl/ polyacrilamide	optimum dose=7 mg/L at low temperature	100% turbidity, 65.68% COD, 24.94% DOC, 37.44% UV_{254}, 36.06% UV_{280}, 43.75% UV_{365}, and residual Al=0.09 mg/L	The residual aluminium concentration was affected by the PACl dose, mixing conditions, and temperature. The C/F process at a lower temperature was more efficient.	Musteret *et al.*, 2021
	optimum dose=4 mg/L at high temperature	100% turbidity, 53.85% COD, 46.79% DOC, 34.44% UV_{254}, 44.65% UV_{280}, 45.83% UV_{365}, and residual Al=0.12 mg/L		

4. Iron based coagulants

Herein, the most representative ferric salts, ferric chloride ($FeCl_3$) and ferric sulphate [$Fe_2(SO_4)_3$], will be discussed since they are commonly used in coagulation processes.

An important operating factor that affects the effectiveness of CF is pH. Only a slight pH variation may increase or decrease charged species that can influence colloids agglomeration rate. Determining the optimum pH and dosage of ferric coagulant are necessary to optimize the CF process for NOM removal (Sillanpää *et al*., 2018b). In the study of Heiderscheidt *et al*. (2016), pH adjustment from 4.5 to 6.5 had a strong influence on the coagulant optimum dose, and this increased from 71 mg/L to 80 mg/L. Overall, Fe-based coagulants achieve good performances in NOM removal. A comparative study founded that the Fe-based coagulants removed a greater proportion of the DOC fractions, color, and UV_{254} than Al-based coagulants (Umar *et al*., 2016).

Recently, polymeric iron coagulants, including polyferric sulphate (PFS), polyaluminium ferric chloride (PAFC) or polyferric chloride (PFC) received more attention. Pre-hydrolyzed coagulants are considered superior to monomeric forms of ferric salts due to some advantages, such as wider working pH range, lower sensitivity to water temperature, reduced amounts of coagulant and lower residual iron concentrations (Dayarathne *et al*., 2021). An overview of Al-based coagulants used on NOM removal from water or synthetic water in recent research studies are given in Table 4.

Table 4
Overview of recent research studies on NOM removal from water using Fe-based coagulants

Coagulant type	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
$FeCl_3$	pH=5 coagulant dosage=80–480 mg/L	42% in DOC, 78% in color and 53% in UV_{254} reduction	Ferric-based coagulants removed a greater proportion of most of the DOC fractions, color, and UV_{254}. Using as a pre-treatment for the UVC/H_2O_2 treatment	Umar *et al*., 2016
$Fe_2(SO_4)_3$	pH=5 coagulant dosage=80–480 mg/L	40% in DOC, 80% in color and 52% in UV_{254} reduction		
$Fe_2(SO_4)_3$	coagulant dosage: 0-100 mg/L optimum dose=71 mg/L at 4.5 pH and 80 mg/L at 6.5 pH. pH=4.5–6.5 reaction time: around 45 min Stirring rate: 50-300 rpm	76% DOC SUVA reduced from 3.8 L/mg-m to 2.8 L/mg-m	High residual Fe and SO_4^{2-} concentrations in the treated water.	Heiderscheidt *et al*., 2016
$FeCl_3$	low turbidity (1.5 – 8 NTU), and low temperature (<10 °C) optimum dose = 40 mg/L	72.4% UV_{254}, 11.5% COD, and residual Fe=0.08 mg/L	The combined coagulants showed superior coagulation performance in terms of	Lou *et al*., 2012

Coagulant type	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
PAFC:$FeCl_3$ (3:1 by mass)	low turbidity (1.5 – 8 NTU), and low temperature (<10 °C) optimum dose=20 mg coagulant /L	84% UV_{254}, 43% COD, turbidity < 0.5 NTU, residual Al=0.09 mg/L, and residual Fe=0.01 mg/L	turbidity, UV_{254}, COD_{Mn}, iron, and aluminium removal.	
PAFC, PFC	natural pH of water=11.1 coagulant dosage= 200–1200 mg Fe/L	At a dose of 800 mg Fe/L 93% color; 81% lignin; 62% TOC	No significant change in removal from these parameters was observed with higher coagulant dose.	Lal and Garg, 2019
PFA	T=60°C Molar ratio Fe:CH_3COOH =1:4 t=6h pH=7–9 coagulant dose=24 mg/L settling time=5 min	Residual turbidity 5.3 NTU Phosphorus removal 96.1%	Promising coagulant in the process of water/wastewater containing phosphorus treatment.	Wei *et al.*, 2017
PFS	Coagulant dose=20 mg/L	Residual turbidity 11.7 NTU Phosphorus removal 92.2%	–	
PFS	BOD_5=355 mg/L HRT=7h	97.2% BOD_5 96.1% COD 78.6% NH_4^+-N 93.8% TOC 90.7% UV_{254} 96.6% turbidity	C/F pre-treatment used to mitigate membrane fouling in MBR system. Lab-scale MBR system.	Gkotsis *et al.*, 2017

5. Composite inorganic – organic coagulants

Various combination of inorganic and polymeric (synthetic or natural) coagulants have been made for developing composite coagulants which have advantages over the previously mentioned coagulants. An enhanced inorganic–organic composite, combining aluminium sulphate (AS) and polydiallyldimethylammonium chloride (PDADMAC) was tested and compared to conventional AS coagulant. The main results revealed that the composite AS-PDADMAC used small dosage with effective floc compactness (Adebayo *et al*., 2021). Another study investigated the efficiency of PAC – chitosan composite coagulant to remove NOM from synthetic and natural waters. It was found that PAC – chitosan was more effective than PAC alone in removing organic matter from the synthetic water, with close performances in the natural surface water. Indeed, for a low Al dosage (2.16 mg L^{-1}), a much higher removal of NOM from synthetic water, in terms of UV_{254} and DOC measurements, was achieved by the composite coagulants in comparison to that removed by PAC or PAC and chitosan added separately (Ng *et al*., 2012).

An overview of composite inorganic – organic coagulants used on NOM removal from water or synthetic water in recent research studies are given in Table 5.

Table 5

Overview of recent research studies on NOM removal from water using composite inorganic – organic coagulants

Coagulant type	Main operating conditions	Removal efficiencies	Other key results	Ref.
Composite inorganic salt - polyelectrolyte				
AS-PDADMAC	Mass ratio AS:PDADMAC = 10:1	COD_{Mn} 65.31–73.33% NH_3-N 25.5–73.08% turbidity 55.60 –97.26%	Enhanced composite AS/PDMDAAC coagulant performed better than AS coagulant. Mass ratio of 10:1 showed the best performance among all the composites.	Adebayo *et al.*, 2021
PAC-PDADMAC	PDADMAC intrinsic viscosity = 0.55 – 2.47 dL/g Mass percentage of PAC, AS, A-F = 5 – 20%	89.3 – 90.6% algae removal	Algae removal was monitored parameter. After sedimentation the residual turbidity reached 2 NTU.	Zhao and Zhang, 2011
AS-PDADMAC		84.7 – 85.5% algae removal		
A-F-PDADMAC		84.3 – 73.5% algae removal		
Composite inorganic salt + organic coagulants				

Coagulant type	Main operating conditions	Removal efficiencies	Other key results	Ref.
PAC-chitosan	Al dosage = 2.16 mg/L pH = 6	Maximum removal of 84% UV_{254} and 79% DOC at Al concentration of 4.32 mg/L	Chitosan in composite coagulant was shown to improve the coagulation performance.	Ng *et al.*, 2012

6. Natural coagulants

Natural coagulants have been studied and proposed as sustainable alternatives to chemical coagulants due to their availability, cost-effectiveness, low sludge volume and disposal cost, nontoxicity, biodegradability and their performance that is less affected by water pH (Ang *et al*., 2016; Okoro *et al*., 2021). Natural coagulants can be obtained from bacteria, fungi, algae, animals, and plants (Tomasi *et al*., 2022). As for now, chitosan, starch and tannin-based-coagulants are the commercially available natural coagulants (Ang *et al*., 2016; Choy *et al*., 2016; Tomasi *et al*., 2022). For instance, tannin-based coagulants exhibited good performance in removing turbidity, color, suspended solids, organic matter (expressed as chemical oxygen demand), total phosphate, algae, and heavy metals (Tomasi *et al*., 2022).

An overview of natural coagulants used on NOM removal from water or synthetic water in recent research studies are given in Table 6.

Table 6
Overview of recent research studies on NOM removal from water using natural coagulants

Coagulant type	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
Starch	optimal dose = 120 mg/L pH = 4 settling time = 30 min	50% turbidity removal	Reduced the amount of chemical-based sludge by 60%.	Choy *et al.*, 2016
Chitosan	pH = 4 – 7 synthetic water HA = 20 ppm	Remove 98% of turbidity and 91% UV_{254}	Reducing the turbidity to lesser than 1 NTU.	Ang *et al.*, 2016
Ca-Alginate	synthetic turbid water initial turbidity 150 – 10 NTU pH = 7.3	98% turbidity removal	Turbidity value of 1 NTU was achieved at a low dose of alginate 0.02 mg/L.	Devrimci *et al.*, 2012

Coagulant type	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
	optimal dose = 0.1 mg/L		Not very efficient floc formation at low turbidity values (10 NTU).	
Tannin-based coagulants	Surface water pH = 8 coagulant dosage = 1250 mg/L	99% turbidity removal 90% color 72% COD 95% TS	Water source contaminated with diazo dyes.	Tomasi *et al*., 2022

7. Electrocoagulation

Electrocoagulation (EC) is an electrochemical water treatment process, which uses soluble anodes made of metal coagulants, such as iron or aluminium – based coagulants. Coagulation shows up when these metal cations, Al^{3+} and Fe^{2+}, react with the negative charged NOM particles through various destabilization mechanisms and accompanied by pH change and hydrogen gas formation (Verma and Kumar, 2018). The method may have some advantages over the conventional coagulation, including sludge volume reduction and different chemicals required, adaptability to the existing treatment units, and efficiency removal for both hydrophobic and hydrophilic fractions (Ulu *et al*., 2015). Hence, the potential in NOM removal of EC has been observed, but it has a mechanism that is highly dependent on the chemistry of the aqueous medium, especially the conductivity (Matilainen *et al*., 2010). Table 7 presents the main operating conditions and other experimental findings of some studies that apply EC for NOM removal.

Table 7

Overview of recent research studies on NOM removal from water using EC

Electrodes	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
Al electrodes	current density = 386 A/m^2 at 12V 5 cm inter electrode spacing 0.7 g/L Al consumption reaction time 30 min pH = 9 – 10.5	73% COD 53% TSS 88% turbidity 87% NH_3 95% NO_3 85% PO_4	An increase in electrolysis time causes an increase in pH.	Verma and Kumar, 2018
Al, Fe and hybrid Al-	current density = 3mA/ cm^2	DOC reduction	The hybrid	Ulu *et al*., 2015

Electrodes	Main operating conditions	Removal efficiencies (%)	Other key results	Ref.
Fe electrodes	pH = 4 – 8	removal rates using Al, Fe and hybrid Al–Fe electrodes were 71.1%, 59.8%, and 68.6%, respectively.	electrodes were more effective in removing color (92.4%) than Al and Fe electrodes	
Anode: hot-rolled iron steel Cathode: stainless steel	Synthetic water DOC: 13.8 mg/L Conductivity: 300 μS/cm Current density: 2.43-26.8 mA/cm^2 pH: 7	73% DOC 88% UV_{254}	The highest removal efficiencies were reported at a current density optimum 10 mA/cm^2. At pH 6, was noted an enhancing the DOC and UV254 removals by 13.8% and 29%, respectively.	Dubrawski and Mohseni, 2013

8. Discussion and Conclusions

The presence of NOM in almost all surface raw water sources and their nature constitute the main challenge facing drinking water treatment techniques for their removal. In order to choose the appropriate treatment technology to achieve a high removal efficiency and to mitigate the formation of toxic by-products, rigorous characterization of NOM and water source quality indicators are required first. The most suitable and economically water treatment technology, which has the purpose of NOM removal, has been proven to be the CF process. Thus, several types of coagulants have been developed, such as metal salts, inorganic or organic (synthetic or natural) polymers and their composites. Regarding the proposal of an efficient coagulant, a summary based on research findings is presented in Table 8.

Table 8
Advantages and limitations of coagulant categories considered in this review

Coagulant category	Coagulant type	Advantages	Limitations	Highest removal efficiencies (%)	Ref.
Al-based coagulants	$Al_2(SO_4)$	Stable, easily handled, readily soluble. Better turbidity removal than with ferric salts in many cases. Can be more effective than ferric in low doses. Higher color removal efficiency.	Coagulant residuals in the finished water. Potential for Alzheimer's disease. Sensitivity to low temperature and low levels of pH. Ferric salts are found more effective in removing NOM than aluminium salts. High alkalinity consumption.	95% UV_{254} 92% TOC	Umar *et al*., 2016; Verma and Kumar, 2018
Fe-based coagulants	$FeCl_3$	Ferric salts are found more effective in removing NOM than aluminium salts. Especially for high and intermediate size NOM fractions molecular mass compounds (1000 – 4000 g/mol). Not so sensitive to temperature variations compared to alum. Larger and numerous flocs are formed.	Greater chemical addition for stabilization and corrosion control is required. High alkalinity consumption. Sulphate and/or chloride in finished water increases corrosivity.	42% DOC 72.4% UV_{254}	Lou *et al*., 2012; Umar *et al*., 2016;

Coagulant category	Coagulant type	Advantages	Limitations	Highest removal efficiencies (%)	Ref.
Polymeric coagulants	PAC/ PAFC	Larger size and higher positive charges flocs are formed. Wider working pH range. Lower sensitivity to water temperature. Lower dose requirement and less sludge produced. Lower residual aluminium or iron in treated water.	The coagulant hydrolysis species formed affected the effectiveness of the coagulant. Preformed Al species are stable and cannot be further hydrolysed during coagulation. Might not be so efficient in removing hydrophobic NOM.	PAC 46.79% DOC 34.44% UV_{254} PAFC 62% TOC	Lal and Garg, 2019; Musteret *et al.*, 2021
Composite coagulants	PAC-PDADMAC	Larger and stronger flocs are formed than with any other coagulant alone. Lower coagulant dose requirements. Smaller volume of sludge. Cost saving.	The cost is dependent to the polymer dose required. Toxic effects.	84% UV_{254} 79% DOC	Adebayo *et al.*, 2021; Zhao and Zhang, 2011
Natural coagulants	Chitosan	Abundant availability, cost-effectiveness, low sludge volume and disposal cost, nontoxicity, biodegradability. Lower sensitivity to water pH.	Formation of smaller flocs due of charge neutralization. The cost is dependent to the polymer dose required.	91% UV_{254}	Ang *et al.*, 2016

Coagulant category	Coagulant type	Advantages	Limitations	Highest removal efficiencies (%)	Ref.
		Exhibited good performance in removing turbidity, color, suspended solids, chemical oxygen demand, total phosphate, algae, and heavy metals.			
Electro-coagulation	Al, Fe or hybrid Al-Fe electrodes	Effective in all temperatures. Remove also the smallest charged particles. Produce small amounts of sludge.	Energy demand raises as initial NOM concentration increases.	73% DOC 88% UV_{254}	Ulu *et al*., 2015 Dubrawski and Mohseni, 2013

Among the six coagulants categories taken into consideration in this review, Al-based coagulants revealed the best results to remove NOM with the highest efficiencies in terms of TOC (92%) and UV_{254} (95%). With 73% and 88% reductions in DOC and UV_{254}, respectively, the electrocoagulation exhibits the potential for NOM removal. Also, the composite coagulants obtained a performance simultaneous highest removal of DOC (84%) and UV_{254} (79%). Within natural coagulants, the chitosan is showing promising removal rates of NOM (91% UV_{254}).

In terms of research, most studies are conducted using synthetic water that make it inappropriate to extrapolate the results to real case studies. Therefore, performing coagulation tests on natural waters, either immediately or after preliminary tests on synthetic waters, should be a rule for the future studies for developing new coagulants, which are expected to be mostly hybrid or natural.

Also, the CF processes applied in drinking water treatment have to be improved to fulfil both the sustainable concepts of circular economy and bioeconomy. In the context of circular economy, the future research and development studies should take into consideration two aspects: (i) improving the coagulation process without increasing the coagulant doses, but by replacing the conventional coagulant with more efficient hybrid coagulants and (ii) developing and applying recovery and reuse technologies for coagulants. Some studies have reported recovery options (Ahmad *et al*., 2016; Keeley *et al*., 2016; Mora-León *et al*., 2022; Kang *et al*., 2022) and more effort is needed in this direction.

Regarding the bioeconomy, the production and application of natural polymeric coagulants derived from plants, algae, or microorganisms are the most viable alternatives to accomplish the sustainability approach in water management. The goal for related research and development studies should be to develop biocoagulants capable of competing with conventional ones, but with increased cost efficiency and eco-friendly.

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ÎNDEPĂRTAREA MATERIEI ORGANICE NATURALE PRIN COAGULARE PENTRU TRATAREA APEI ÎN VEDEREA POTABILIZĂRII

(Rezumat)

Materia organică naturală (NOM) este echivalentă cu totalitatea substanţelor organice rezultate din descompunerea bacteriană a materiei animale și vegetale. NOM se găsește în mod natural în majoritatea surselor de apă de suprafață utilizate pentru alimentarea cu apă potabilă și poate avea un impact semnificativ asupra sănătății umane dacă nu este îndepărtată. În afara faptului că determină gustul, mirosul și culoarea apei brute, aceste substanțe sunt și precursori ai produșilor secundari de dezinfecție, care la rândul lor au efect negativ asupra sănătății umane. Cea mai mare parte a NOM poate fi îndepărtată prin coagulare și floculare urmată de sedimentare și filtrare, procese care din punct de vedere economic sunt considerate cele mai comune și fezabile tratamente pentru obținerea apei potabile. Acest studiu prezintă o abordare de ansamblu asupra studiilor publicate recent privind îndepărtarea NOM în tratarea apei pentru potabilizare cu diferite tipuri de coagulanți și alte procese de tratare având legătură cu coagularea.