



# Water Treatment Coagulation: Dares and Trends

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## Abstract

Coagulation remains a technique by which finely dispersed solids are efficiently eliminated. It has been largely expanded and remains the most unavoidable method for treating water. This review focuses on colloid stability, coagulation mechanisms, and coagulant types. It presents electrocoagulation as an option of conventional coagulation and discusses challenges in coagulation technology especially health hazards in used chemicals toxicity. As promising solutions, new developments in terms of using coagulants are presented. Micropollutants are inorganic and organic substances that could disturb negatively nature even at very low levels. Microplastics are also observed. Coagulation could retain different micropollutants and microplastics at varying efficiencies even if there is a need to determine running circumstances that could increase their reduction. As a perspective, coagulation may be combined with additional processes, such as ultrafiltration. Further, traditional water treatment should be deeply revised.

## Subject Areas

Chemical Engineering & Technology

## Keywords

Coagulation, Water Treatment, Dissolved Organic Matter (DOM), Disinfection By-Products (DBPs), Chemicals Toxicity

## 1. Introduction

Coagulation remains a technique by which finely dispersed solids are efficiently eliminated [1] [2] [3]. Firstly employed by the ancient Egyptians as early as 2000 B.C.E [4] [5] [6], coagulation process has been largely expanded, espe-

cially during the last century. Nowadays, it remains the most unavoidable method for treating water [7] [8] [9].

Pursued by clarification stages (mostly decantation and (sand) filtration processes), coagulation remains the most performant for retaining particulate matter (it carries both colloids, mostly 10 nm to 1  $\mu\text{m}$ , and small particles, usually  $> 1 \mu\text{m}$ ) from water (**Table 1**) [10] [11] [12]. Further, it eliminates dissolved portions of some matters such as natural organic matter (NOM, like humic substances) from surface water [7] [13] [14].

This review focuses on colloid stability, coagulation mechanisms, and coagulant types. It presents electrocoagulation as an option of conventional coagulation and discusses challenges in coagulation technology especially health hazards in used chemicals toxicity. As promising solutions, new developments in terms of using coagulants are presented.

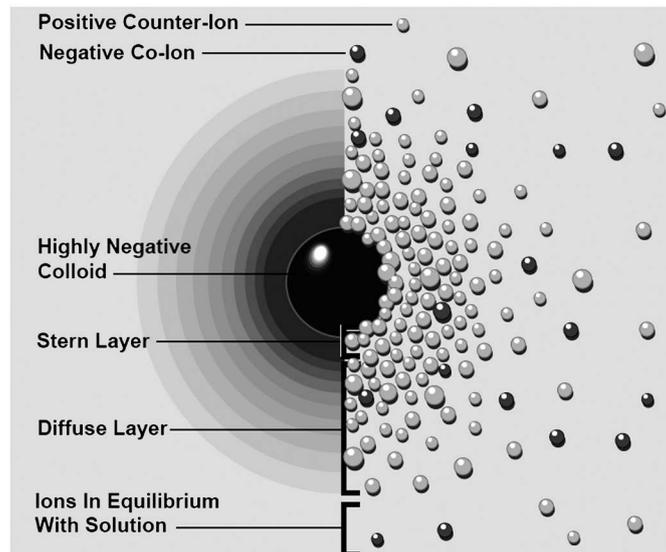
## 2. Colloid Stability

In water, colloids are negatively charged [7]. The cations in water, known as *counter-ions*, are strongly fixed to the colloid's surface and constitute the *Stern layer*. In addition to such counter-ions, many other positive ions are also pulled to the same colloid, because of their positive charge and the negative charge of the colloid, even if somewhat loosely because of the repulsion from the cations in the Stern layer, as well as because of the competition for attachment by other cations. This leads to a dynamic equilibrium producing the *diffuse layer*. In such layer, the level of the counter-ions gradually reduces with the distance from the colloid. In water, the anions, as well known as *co-ions*, gradually augment their occurrence in the diffuse layer, generating an equilibrium. The Stern layer and the diffuse layer constitute the so-named *double layer*. Such layers are presented in **Figure 1**.

The identical negative charge of the particles and the width of the double layers avoid agglomeration of particles with each other. This is why the colloids stay dispersed in water until their charges and the double layers are considerably diminished [7].

**Table 1.** Coagulation terminology [7] [15] [16].

Term	Description
<i>Coagulation</i>	<i>Coagulation</i> is the phenomenon by which colloids are destabilized, conducting to their agglomeration [17] [18] [19].
<i>Flocculation, clarification</i>	Practically, the coagulation technology is composed of coagulation, <i>flocculation</i> and <i>clarification</i> (separation unit stages) [20] [21] [22]. Following the coagulation, the destabilized particles and other precipitates produce agglomerates that require growing further into larger flocs. This stage is named flocculation. The agglomerated flocs can then be separated utilizing decantation (or flotation) and filtration methods [23] [24] [25].
<i>Coagulants, flocculants</i>	Throughout the coagulation and flocculation stages, chemical products are usually injected; such agents are known as coagulants and flocculants, respectively.



**Figure 1.** Electrical double layer and distribution of co-ions and counter-ions [26].

The colloid stability is defined by the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory. Such theory proposes that the colloid stability in water is a function of its total potential energy function  $V_T$ , which is the sum of three forces [7]:

$$V_T = V_A + V_R + V_S \quad (1)$$

where  $V_S$  is the potential energy attributed to the solvent (water). Practically, it is of minor importance. The attractive force is defined by

$$V_A = -\frac{A}{12\pi D^2} \quad (2)$$

where  $A$  is the Hamaker constant and  $D$  is the distance between the particles.  $V_A$  is also called the van der Waals force [7].

The repulsive force attributed to the electrical double layer is defined by

$$V_R = 2\pi\epsilon a\xi^2 e^{-\kappa D} \quad (3)$$

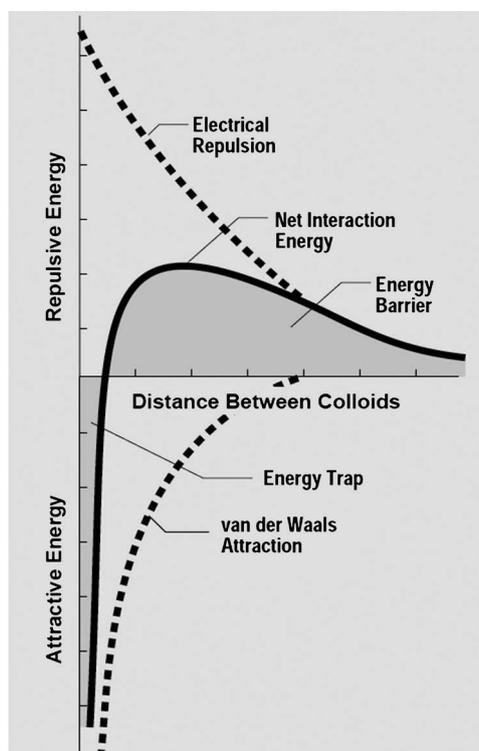
where  $a$  is the particle radius,  $\epsilon$  is the solvent permeability,  $\kappa$  is a function of the ionic composition, and  $\xi$  is the zeta potential [7].

The energy barrier resulting in the sum of forces (**Figure 2**) prohibits colloids, which are in Brownian motion, from approaching sufficiently closer where the attraction forces dominate [7].

### 3. Coagulation Pathways

As a rule, there are numerous classifications of coagulation routes encountered in the specialized references. **Table 2** lists the four routes including all classifications [7].

Practically, there are two pathways dominant: adsorption-charge neutralization and colloidal entrapment. In addition to these two routes, the double layer compression pathway could affect the coagulation performance with the occurrence of



**Figure 2.** Total potential energy of two particles approaching each other [26].

**Table 2.** Four coagulation mechanisms [7].

Coagulation mechanism	Description
<i>Mechanism of Particle Removal</i>	<p><i>Double Layer Compression</i> The compression of the double layer decreases the energy barrier; thus, the colloids can come closer [27] [28] [29]. This is attained via injecting indifferent electrolytes to the water [30] [31] [32]. The Schultz–Hardy rule shows that the impact of the indifferent electrolytes augments with the valence of the ions by sixth exponential power [33] [34] [35]. As an illustration, one mole of <math>\text{Ca}^{2+}</math> possesses the identical impact as <math>2^6</math> ions of <math>\text{Na}^+</math> on the double layer compression [36] [37] [38].</p> <p><i>Adsorption-charge neutralization</i> In coagulation method, such a mechanism remains the most cost-efficient pathway [39] [40] [41]. During this route, the positively charged ions and species cover the negatively charged colloids, thereby decreasing the surface charge and by that the energy barrier [42] [43] [44]. This mechanism is cost-efficient tanks to the fact that the neutralization process occurs stoichiometrically between the positively and negatively charged ions [45] [46] [47]. Such a pathway could realize coagulation via inorganic coagulants or cationic organic polymers [48] [49] [50].</p> <p><i>Interparticle bridging</i> Such route takes place when organic coagulants or organic polymers are employed [51] [52] [53]. Polymers possess threads and fibers that bind the particles into big and compact agglomerates [54] [55] [56]. Polymers with higher molecular weight (MW) are frequently more efficacious, thanks to their long chains [57] [58] [59].</p> <p><i>Colloidal entrapment</i> or “sweep floc” The final product of the hydrolysis of inorganic coagulants is the hydroxide precipitates that could entrap colloids [60] [61] [62]. Such pathway consumes coagulants in excess of stoichiometry [63] [64] [65].</p>
<i>Mechanisms of Organic Matter Removal</i>	<p>Natural organic matter (NOM) is the source of natural color in surface waters [66] [67] [68]. NOM removal is fundamental in potable water treatment since it form carcinogenic disinfection by-products (DBPs) [69] [70] [71]. NOM is mostly composed of humic substances (their average size is 0.47 - 3.3 nm) [72] [73] [74]. NOM is quantified by color, ultraviolet (UV) absorption, and total organic carbon [75] [76] [77]. NOM is noted to be retained through all particle removal routes [78] [79] [80]. NOM could dominate coagulant injection and adoption of water treatment technology rather than colloidal particles [81] [82] [83].</p>

electrolytes. Colloidal entrapment only happens when inorganic coagulants are injected. The interparticle bridging happens only when organic coagulants are introduced. **Table 3** lists the operational dominance of coagulant pathways [7].

## 4. Coagulant Types

### 4.1. Mineral Coagulants

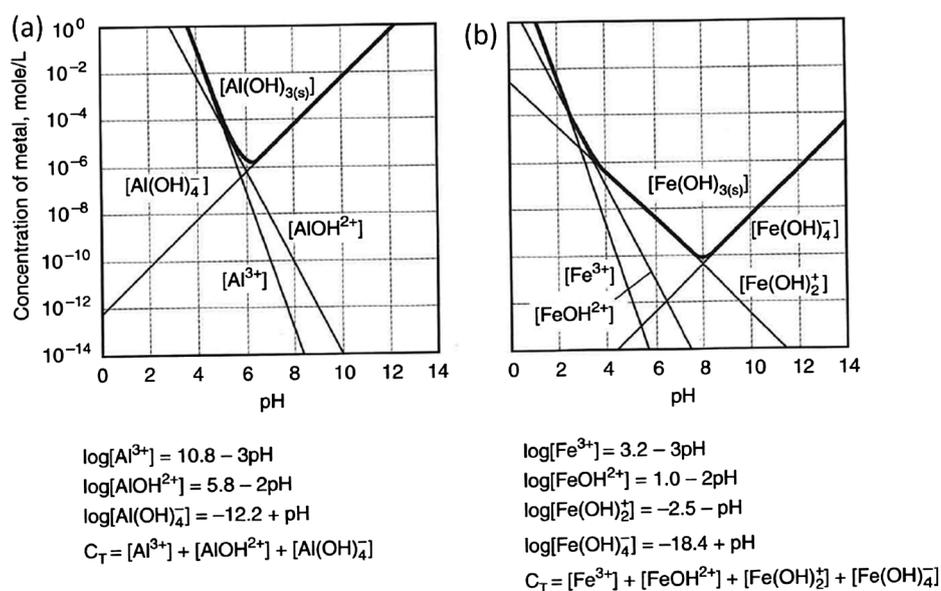
The colloids elimination remains mostly founded on the hydrolysis of mineral coagulants [7]:



Reaction (4) takes place during seconds and follows several steps that form numerous intermediate species, which are favorable for coagulation. Following the circumstances, there can be numerous mononuclear hydroxides (like  $\text{Al}(\text{OH})^{2+}$ ,  $\text{Al}(\text{OH})_2^+$ ,  $\text{Al}(\text{OH})_3$ , and  $\text{Al}(\text{OH})_4^-$ ) or polymerization reactions to polycations (such as  $\text{AlO}_4\text{Al}_{12}(\text{OH})_{24}(\text{OH}_2)_{12}^{7+}$ ) [7]. More than eighty Al species can be formed [84]. **Figure 3** shows instances of monomers in a solubility diagram and **Figure 4** presents the usual pathways related to the coagulants' injection and pH.

**Table 3.** Practical dominance of the coagulation pathway [7] [63].

Mechanism type	Coagulant type	
	Inorganic coagulants	Organic coagulants
Double layer compression	Occasionally	Not applicable
Adsorption-charge neutralization	Dominant	Happens with cationic polymers
Interparticle bridging	Not applicable	Dominant
Colloidal entrapment (sweep floc)	Occasionally	Not applicable



**Figure 3.** Solubility diagrams of (a) Al(III) and (b) Fe(III), with monomeric species [85].

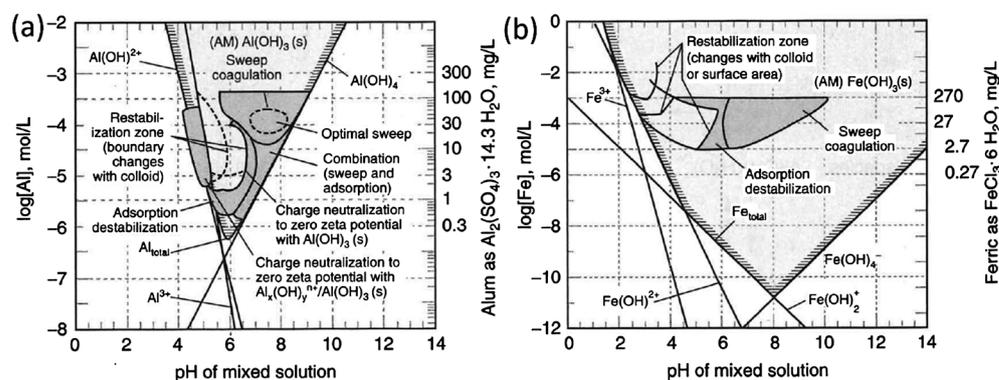


Figure 4. Usual coagulation working diagram. (a) Al(III) and (b) Fe(III) [86].

In the water treatment industry, the most frequent coagulants remain aluminum sulfate, ferric chloride, ferric sulfate chloride, and calcium hydroxide. Since the 1980s, investigation has conducted to the formation of pre-polymerized aluminum hydroxychlorides, enabling better utilization of mono- and polynuclear species. For such coagulants, the ratio of OH/Al stays a fundamental parameter [7].

Innovative coagulants with combinations of calcium (to augment the double layer impact), water glass (to improve sedimentation properties), flocculants (to avert the request for two injecting systems), and so on are more and more mentioned even if their large usage is restricted [7].

## 4.2. Organic Coagulants

Organic coagulants possess synthetic and biological origins. The synthetic polymers are prevailing as both coagulants and flocculants. The synthetic polymers are mostly polyamines, polydiallyldimethylammonium chloride (poly-DADMACS), dicyandiamide resins, and melamine-formaldehyde resins. The polyacrylamides and poly-DADMACs are likely the most famous cationic coagulants. Such coagulants are known by their MW (3000 - 3,000,000) and cationic charge density (low to extra high) [7].

Extracted from crab and shrimp shells, chitosans are adopted as performant biological coagulants in potable water industry thanks to their numerous merits. They work over a larger pH span without changing the pH of the treated water and they do not generate any remaining aluminum. Nevertheless, their drawback remains the cost, since the organic polymers are considerably more expensive than inorganic coagulants, and biopolymers are even more expensive than synthetic organic polymers [7].

## 4.3. Flocculants

As flocculants, the synthetic organic polymers are utilized. They possess different degrees of anionic, non-ionic, or cationic charge and could have MWs of 3,000,000 to 20,000,000. The flocculants could greatly augment the floc production speed and the strength of the flocs and make them much heavier. Floccu-

lants are efficient products that increase the length of filtration (delayed breakthrough) in drinking water treatment [7] [87].

## 5. Coagulation Technique Options

### 5.1. Traditional Potable Water Treatment

**Figure 5** shows traditional potable water. It is frequent to insert sieves or micro-sieves before coagulation and disinfection stages of the final treated water [88] [89] [90]. If the water possesses low pH/alkalinity, the pH/alkalinity will be adjusted following the disinfection step [91] [92] [93]. Taking into account the components in between such two steps, a collection of coagulation methods have been adopted [94] [95] [96]. The most usual coagulation techniques are founded on a coagulant mixing zone followed by a flocculation step where the flocs are progressively formed [97] [98] [99]. Following flocculation, the flocs are separated via a decantation or flotation step [100] [101] [102]. It is frequent to insert a filtration step that also comprise a Granular or Powder Activated Carbon (GAC or PAC filter that could reduce any residual organic matters) [103] [104] [105]. If the raw water has low colloids amount, coagulation could happen without decantation or flotation, and the separation of the microflocs takes place indirectly in the filter [106] [107] [108]. If there is a flocculation step or not, the technique integrations are called *contact filtration* or *direct filtration* [109] [110] [111]. Keeping the coagulation pH inside the running spans of the respective coagulants stays fundamental. In numerous conditions, particularly if the raw water source is soft, the coagulants are injected simultaneously with CO<sub>2</sub> and lime to control the pH that also positively participates to dominating corrosion in the distribution system [7] [112] [113].

### 5.2. Electrocoagulation (EC) Process

Employing electrochemical technology, coagulation could also be realized. An electrochemical cell equipped with Fe or Al electrodes can generate *in situ* hydrolysis species, conducting to coagulation like when introducing inorganic salts [114]. Electrocoagulation (EC) presents the simplicity of injecting control via adjusting the electrical current flow across the device. EC is also well known for its disinfection features [115] [116]. EC is largely employed in industrial wastewater treatment and its large application in the water supply and urban wastewater treatment remains to be implemented [7] [117] [118].

## 6. Dares in Water Coagulation

### 6.1. Health Risks in Water Coagulation

As previously mentioned, coagulation stays the most usual technique in dealing with water treatment and Al salts are the most widely used coagulants. Al species are detected in dissolved forms beyond pH ranges relevant to their levels (**Figure 3** and **Figure 4**). Non-optimal injection of coagulants, particularly in water with

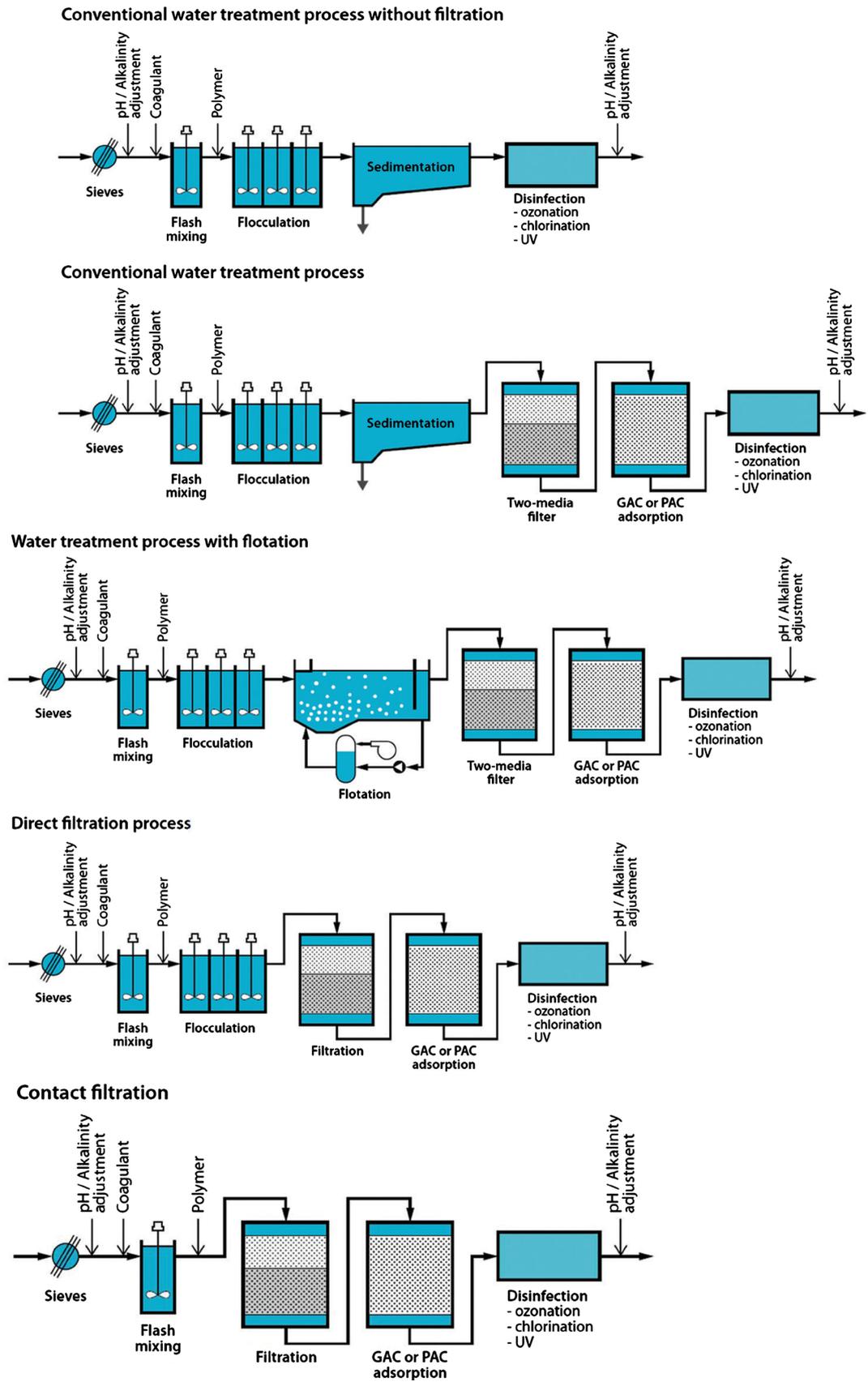


Figure 5. Coagulation method options in potable water treatment industry [7].

low alkalinity, could modify the coagulation pH to unwanted ranges where some of the aluminum will be in dissolved form. The usual separation techniques are unable to retain dissolved portions, and they may end up in the supplies to the consumers [119]. There are shreds of evidence of a linkage between the aluminum concentrations in drinking water and Alzheimer's disease. Employing Fe-based coagulants may avert this hazard, usually with favorable effects on denser flocs, conducting to better sedimentation features [7].

In water supply systems, the excessive Al levels could be efficaciously controlled via fixing optimal coagulant injections to aver over- and under-dosages integrated with overriding with coagulation pH range control to secure favorable pH ranges. A different strategy is to employ biopolymers (like chitosan), even if their usage is not yet economically feasible in bigger treatment facilities [7].

## 6.2. Over- and Under-Dosage Conducting to Disinfection By-Products (DBPs)

Many treatment plants prefer to employ raw water from lakes rather than from rivers, because of its more stable water qualities [7]. The drawback is that some lake waters mostly carry out an elevated level of NOM that produce carcinogenic compounds (e.g., trihalomethanes) during chlorination. Therefore, NOM should be reduced prior to chlorination. Nevertheless, the non-optimal coagulant dosing and unfavorable pH ranges could lead to poor reduction of NOM, conducting to health dangers. The dare has augmented lately with the augmentation of NOM levels in lake water because of climate change.

As a possible option, utilizing more optimal dosing control systems, which rapidly respond to variations in raw water quality and maintain favorable pH ranges, is suggested [7].

## 7. New Developments as Potential Options in Terms of Using Coagulants

There are recent enhancements in terms of utilizing coagulants. As a rule,  $\text{Ca}^{2+}$ ,  $\text{Fe}^{3+}$ , and  $\text{Al}^{3+}$  salts have been utilized as coagulants. Even if  $\text{Ca}^{2+}$  salts have become less frequent,  $\text{Al}^{3+}$  has become the dominant coagulant in the water treatment industry. In order to elevate the performance of such classical chemicals, pre-polymerized coagulants were suggested four decades ago. They are synthesized via carrying out partial hydrolysis, enabling more efficacious use of positively charged hydrolysis species (mostly in an adsorption-charge-neutralization mechanism). Whilst the first generation of pre-polymerized coagulants was polyaluminum chlorides and polyferric chlorides, a recent generation of coagulants like polyaluminum silicate sulfate, polyferric sulfate, and polyaluminum ferric sulfates are well examined. Prepolymerized coagulants possess numerous merits over conventional aluminum sulfate (alum) [7].

Lately,  $\text{Ti}^{4+}$  and  $\text{Zr}^{4+}$  salts have been suggested as highly efficient coagulants, considering their high valence. Nevertheless, they are not yet employed at large scales, mostly because of their elevated prices [7].

## 8. Conclusions

This review focused on colloid stability, coagulation mechanisms, and coagulant types. It presented EC as an option of conventional coagulation and discussed challenges in coagulation technology especially health hazards in used chemicals toxicity. As promising solutions, new developments in terms of using coagulants are presented. The main findings of this work are listed below:

Micropollutants are inorganic and organic substances that could disturb negatively nature even at very low levels. Heavy metals are traditionally known as micropollutants. Organic micropollutants comprise both classical (dichlorodiphenyltrichloro-ethane, polychlorinated biphenyls, polycyclic aromatic hydrocarbons, pesticides) and emerging contaminants (hormones, endocrine disruptors, pharmaceuticals, and personal care products). The traditional water treatment techniques are frequently not concentrating on the reduction of such matters. Nevertheless, there is a request to eliminate them during water treatment. Coagulation could retain different micropollutants at 6% - 90% [120]; however, there is still a necessity to determine running circumstances that could increase the reduction of micropollutants.

In potable water resources, microplastics are observed. Microplastics generate a hazard to human health and nature. For their elimination, coagulation could be efficaciously employed. Combining coagulation with, for instance, ultrafiltration, has illustrated great capacity for their elimination from water.

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## Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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