

# Polyelectrolytes as a Material of Value in Water Treatment: A Review

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**Abstract:** The use of polyelectrolytes in the treatment of water/wastewater has been studied with special emphasis on the characteristic impurities, which should be removed and the types of polyelectrolytes commonly available. Much attention is being on using polyelectrolyte as primary coagulation, their application as a coagulant aid, including the use of dual polyelectrolytes in the treatment of complex industrial wastewater. The optimum dosage of polyelectrolytes in all kinds of water and wastewater is very low when they are used as coagulant aids in conjunction with inorganic polyelectrolytes. Hence, the quantification of remaining polyelectrolytes in the water after their treatment is ruled out in this case. Polymer toxicity in aquatic animals has been assessed and the presence of residual polyelectrolytes in the treated water has been discussed. Keeping in view the applicability of these polyelectrolytes for the removal of contaminants, it is expected that this technique can be applied for further investigation in various types of water from different origins.

**Key words:** Polyelectrolytes, charge neutralization, polymer bridging, toxicity, coagulation, coagulant aid.

## Introduction

The well-being of mankind mainly depends upon the environment and ecological factors. However, environmental and ecological factors depend upon the quality of water available to be used for different purposes.

The literature data reveals that impurities in water are associated with suspension and dissolution (de Nardi et al., 2007; Vaughan et al., 2000). So, it is necessary to perform detailed analyses of impurities present in water and wastewater. Water has complex chemistry and contains different types of impurities ranging from suspended matter to dissolved matter. In conventional chemical treatment, inorganic coagulants like aluminum or iron salts are utilised majorly (Wei et al., 2016). However, handling a large volume of sludge produced from the use of metal coagulants is a difficult task

(Chang et al., 2005). With the establishment of different industries, organic polyelectrolytes were introduced for the treatment of different kinds of water and wastewater (Iakovides et al., 2014). Since then polyelectrolytes are being used in water treatment (Lourenço et al., 2017).

According to their ionic charge, polyelectrolytes are classified into different categories. Polyelectrolytes with a positive charge are cationic and the ones with negatively charged are anionic (Radoiu et al., 2004; Sarika et al., 2005). Different types of polyelectrolytes are available commercially. Most of them have been reviewed by Bolto and Gregory, (2007). They are available in synthetic and natural forms (Bing-hui et al., 2005; Choi et al., 2001; Selvapathy et al., 1992). Synthetic polyelectrolytes include polyamines, polyacrylamides, polyalkyleneimine, polyacrylonitrile, polydiallyl dimethyl ammonium chloride (PDADMAC), epichlorohydrine polymers and poly-acrilamides

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(Razali et al., 2011; Yap et al., 2014). The natural polyelectrolytes used in wastewater treatments include chitosan, glue, starch, gelatin galactomannans, cellulose derivatives and tannin (Elhakeem et al., 2014; Jabin and Kapoor, 2020).

The natural polyelectrolytes being biodegradable in nature are always preferred. An advantage of natural polyelectrolytes is that they are virtually toxicity-free (Ishak et al., 2020). So, natural polyelectrolytes and their derivatives can work as potential substitute in place of synthetic polyelectrolytes for minimising water pollution. Polyelectrolytes are used as primary as well as secondary coagulants in conjunction with inorganic coagulants

### Polyelectrolytes as a Primary Coagulant

The selection of coagulant is one of the most challenging task in the treatment of water and wastewater. There are many cases where polyelectrolytes have been used as a primary coagulant (Wilts et al., 2018), effectively replacing the use of inorganic coagulants; in many cases, they are used to reduce or remove suspended particles from water and wastewater (Blanco et al., 2002; Wu et al., 2018). Cationic polyelectrolytes with high charge density and low to medium molecular weight are more successfully applied as primary coagulants (Jabin and Kapoor, 2020). The most common examples of cationic polyelectrolytes used as primary coagulants are polydiallyl dimethyl ammonium chloride (PDADMAC) and poly methacrylate (Bharti and Mishra, 2016; Zhou and Franks, 2006). The literature also shows that polyelectrolytes, as primary coagulants, are also capable of removing dyes (Ahmad et al., 2008). It was also studied that the polyelectrolytes have the ability to remove colour from wastewater (Szyguła et al., 2009). As per literature, polyelectrolytes have been proven to be effective in the removal of microorganisms (Bolto and Xie, 2019) but it is more successfully applied in the removal of suspension present in different kinds of water.

Several types of research have been reported on the coagulation–flocculation technique for the treatment of industrial effluent (Cao et al., 2011; Qin et al., 2006). The process of coagulation involves different steps which have been shown in Figure 1. After the collection of water/wastewater, different parameters are assessed. After optimisation of pH, destabilisation of coagulated flocs occurs by the addition of polyelectrolyte (Crittenden et al., 2005). Mixing of coagulant is done in two steps viz.: (i) rapid and (ii) slow mixing. It

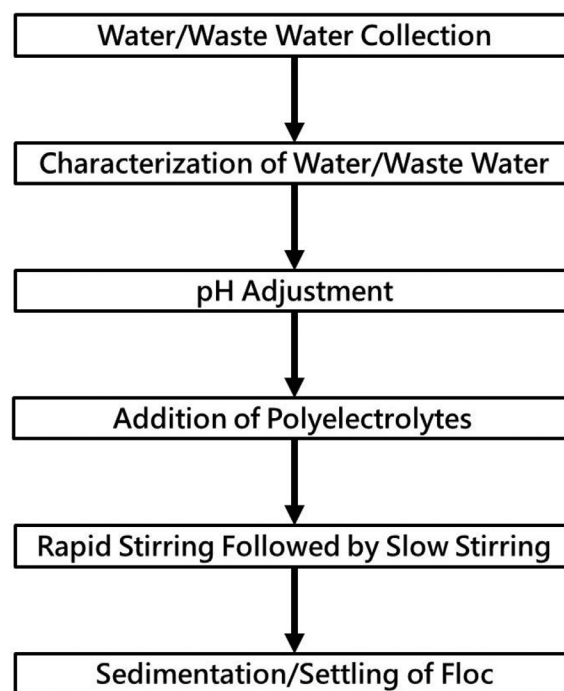


Figure 1: Flow chart showing the process of removal of impurities by polyelectrolyte as a primary coagulant.

requires proper turbulence to form homogenous mixing. The mixing is done slowly to encourage the formation of the homogenous mixture, and this process is known as “coagulation”. The final step involves the removal of the impurities with the process of sedimentation. A high dosage of polyelectrolyte is required when it is used as a primary coagulant (Lee et al., 1998). The coagulant dosage added also depends upon the types of wastewater and pH.

### Polyelectrolytes as a Coagulant Aid

Polyelectrolytes are majorly used as coagulant aids in water treatment in conjunction with inorganic coagulants, which are used as primary coagulants in this situation. The use of inorganic coagulants alone has its own limitations, such as a large amount of coagulant is required and as a result, a large volume of sludge is generated. In contrast, proper use of polyelectrolytes as a coagulant aid can successfully remove pollutants from different kinds of wastewater (de Sena et al., 2008). A low dosage of polyelectrolyte is required when it is used as a coagulant aid. Synthetic polyelectrolytes are more widely used as they are more effective as coagulant aids. Their efficiency depends upon the properties they possess such as the type of charge units, charge density and molecular weight. The charge density of a

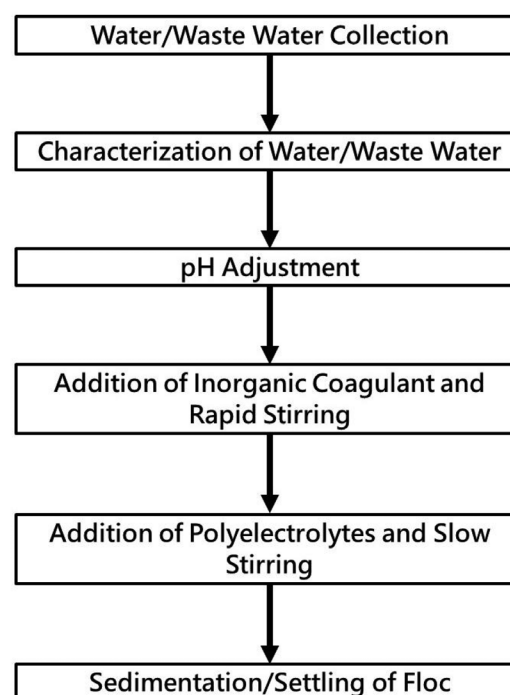
particular polyelectrolyte influences the destabilisation mechanism and leads to floc formation (Theodoro et al., 2013). Kam and Gregory (2001) have demonstrated that the charge density instead of the molecular weight is more important in selecting the polyelectrolytes as a coagulant aid. Synthetic polyelectrolytes, especially cationic polyelectrolytes are more effective in the removal of turbidity and suspension as compared to anionic and non-ionic polyelectrolytes (Kapoor et al., 2015). However, anionic polyelectrolytes are more effective in the removal of dissolved impurities (Clark and Stephenson, 1999). As per Bolto, the efficiency of cationic polyacrylamide with low molecular weight is better than a high molecular weight polyacrylamide. In general, the best results are found when polyelectrolyte is added after primary coagulant addition (Choong et al., 2007). In some cases, where polyelectrolyte and impurities are of opposite sign, then the addition of polyelectrolyte before metal coagulant could reduce the surface charge of particles on adsorption, thereby subsequently reducing the quantity of metal coagulant required to effect charge reduction.

A combination of inorganic coagulants and polyelectrolytes as a coagulant aid can be useful for minimising pathogens in water. Judicious choice of appropriate polyelectrolytes may go a long way to enhance the effectiveness of coagulation-flocculation. The schematic diagram of coagulation is presented in Figure 2.

In summary, each process has its positive and negative aspects. Overview on the usage of polyelectrolytes as a primary coagulant and as a coagulant aid is presented in Table 1.

### Use of Dual Polyelectrolytes in Single Wastewater Treatment

Nowadays many chemical treatment methods are available, but none is efficient enough to remove all kinds of impurities from different sources. A single polyelectrolyte cannot do it, so focussing on



**Figure 2:** Flow chart showing the process of removal of impurities by polyelectrolytes as a coagulant aid.

environmental sustainability can be done by applying dual polyelectrolytes in water and wastewater treatment. The use of dual polyelectrolytes for the treatment of industry effluent especially for the removal of suspended solids, oil and grease and organic matter is well established (Lee and Liu, 2000). Dual polyelectrolytes can enhance efficiency without adding to the solids load that occurs with inorganic coagulants. A combination of “low molecular weight with high charge density cationic” and “low-charge density with high molecular weight anionic” polyelectrolytes yield effective results (Petzold et al., 2001). Suspended particles are mostly negatively charged that’s why cationic polyelectrolytes are successfully attached with suspended particles by charge neutralisation. However, anionic polyelectrolytes work through an electrostatic patch mechanism. Vaughan et al. (2000) exploited dual polyelectrolytes

**Table 1:** Comparative study of the usage of polyelectrolyte as a primary coagulant and as a coagulant aid

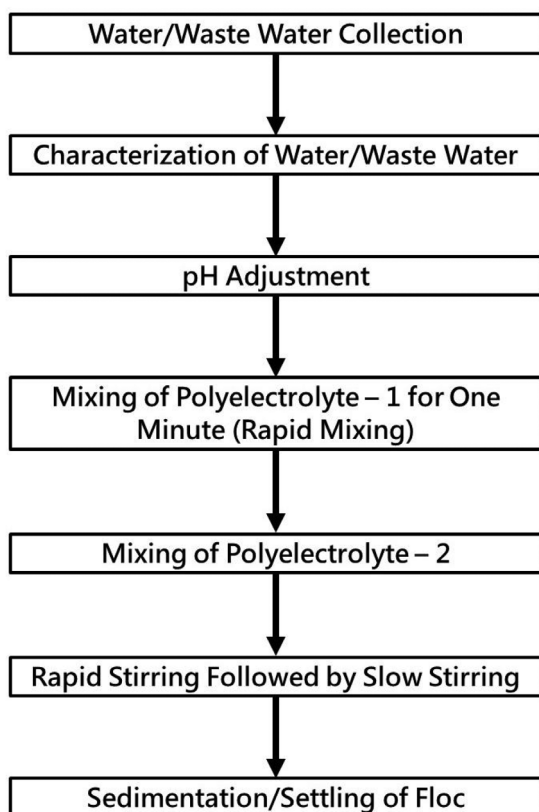
S. No	Parameters	Polyelectrolyte as a coagulant aid	Polyelectrolyte as primary coagulant
1	Treatment efficiency	Suspended/dissolved pollutants both	Suspended pollutants only
2	Types of coagulants	Inorganic coagulants and polyelectrolytes	Polyelectrolytes only
3	Overall chemical requirement	More	Less
4	pH adjustment	More pH adjustment is required	Less pH adjustment is required
5	Cost	More expensive	Less expensive

in oil industry effluent, they also explored the use of cationic and then anionic polyelectrolytes. Dual polyelectrolytes in the form of cationic hemicellulose and anionic tannin have shown better performance in the removal of oil and surfactant residues in biodiesel effluent. A deep understanding of dual polymers mechanism is important before utilising them for the treatment of any kind of wastewater.

Better results could be obtained if both polyelectrolytes are used one after another. Different steps of coagulation based on dual polyelectrolytes have been shown in Figure 3.

### Interaction of Suspended Impurities with Polyelectrolytes

Coagulation-flocculation is an important physicochemical operation used for the removal of suspension with the help of polyelectrolytes. It is used in the treatment of different kinds of water. Considering that this is a complex mechanism, a thorough understanding of suspended particles is important. Their surface is generally negatively charged and they repel each other to form a stable dispersed suspension. The concentration of polyelectrolytes required for coagulation is proportional



**Figure 3:** Flow chart showing the process of removal of impurities by dual polyelectrolytes as a coagulant aid.

to the concentration of impurities present in water. Nozaic et al. studied that optimisation of polyelectrolytes dosages is the most important parameter in the water industry's ability to meet the goal of suspension removal (Kapoor et al., 2011). They stressed the significance of adopting a treatment process and highlighted the necessity of adjustment of two important parameters: pH and polyelectrolytes dosage. The polyelectrolytes dosage depends upon the types of wastewater and pH.

Removal of suspension requires less amount of coagulant-flocculant. In the presence of suspended solids, low molecular mass polyelectrolytes preferentially react with dissolved organic matter and high molecular mass polyelectrolytes at low dosage with suspended solids. At higher dosage, the surface of the suspended solids is completely covered by polymer and then additional polyelectrolytes start interacting with humic substances. The presence of suspended solids increases the degree of humic substance removal. For removal of suspension, polyelectrolytes can be used as primary coagulants or as coagulants-aids in conjunction with inorganic coagulants.

Proper mixing of polyelectrolytes is an essential parameter for the removal of suspension. This is the first process wherein polyelectrolyte is distributed evenly throughout the suspension. Mixing is generally carried out swiftly, else extra concentration may cause uneven adsorption and some particles may re-establish as a result of adsorbing excess amount of polyelectrolyte. Improper mixing generally results in residual haze in water after polymer flocculation and sedimentation. It is desirable to have a considerable amount of turbulence for proper mixing, this is because polyelectrolyte chains may encounter some scission in this condition.

### Interaction of Dissolved Impurities with Polyelectrolytes

Dissolved organic matter (DOM) is generally divided into two categories: humic substances and non-humic substances. Humic substances are generally hydrophobic macromolecules and bound together by a hydrogen bond. It shows yellowish-brown colour in water. Non-humic materials are colourless, less hydrophobic and more easily defined such as polysaccharides, protein, nucleic acids and small molecules such as sugar and amino acids. The non-humic substances consist of biodegradable organic carbon which serves as the substrate for bacterial growth in water distribution systems.

Research on the removal of DOM with polyelectrolytes has also been done (Lurie and Rebhun, 1997). In



natural water, dissolved organic matters (both humic and non-humic) are generally anionic in nature. They are generally removed by the mechanism of charge neutralisation. There is stoichiometric relation between cationic polyelectrolyte and dissolved organic matter (Lee and Westerhoff et al., 2006). As per literature, cationic polyelectrolytes having high charge density are more effective in the removal of dissolved organic matter. It has been observed that the molecular weight of polyelectrolytes is not an important criterion for the removal of dissolved organic matter. It has also been observed from the literature that more hydrophobic DOM is easily removed as compared to low hydrophobic DOM. Inorganic coagulants showed poor results in the removal of DOM. They also reduce alkalinity leading to a decrease in the pH of effluent.

The technology for suspended and dissolved impurities so far has shown a great degree of variation in terms of their respective removal efficiencies. Variation in impurities in various types of water makes it difficult to handle. The impurities are both organic and inorganic in nature. The fine fraction of suspended solids carries a significantly high pollutants load compared to dissolved water pollutants.

### **Toxicity of Polyelectrolytes**

The issue of toxicity in polyelectrolytes cannot be ignored. If polyelectrolyte is used as a coagulant aid in conjunction with an inorganic coagulant, then a very low dosage of polyelectrolyte is required. So, this fact ruled out the possibility of toxicity in the treated water. As a primary coagulant, polyelectrolytes are considered more toxic (Goodrich et al., 1991).

If polyelectrolytes are used as primary coagulants to treat drinking water, then the quantification of polyelectrolytes in treated water is an important factor. The toxicity of the remaining polyelectrolytes depends upon the amount of monomers units present in treated water (Ghimire and Bhattarai, 2020). Quantification of epichlorohydrin and polyacrylamide polyelectrolytes in treated water have more health hazards as compared to other polyelectrolytes (Letterman and Pero, 1990). So, it's always safe to use polyelectrolytes as a coagulant aid since quantification of polyelectrolytes is negligible in such cases.

As per literature, polyelectrolytes having a positive charge are considered to be more toxic as compared to anionic and non-ionic polyelectrolytes; hence, cationic polyelectrolytes are more harmful to aquatic organism especially fishes (Goodrich et al., 1991). However, the presence of clay and humic acid reduces the toxicity of

polyelectrolytes in water (Weston et al., 2009) because humic acid and clay can adsorb the polyelectrolytes present in water.

As per Bolto, Japan and Switzerland do not permit the usage of polyelectrolyte in the treatment of drinking water. However, France and Germany also have stringent limits. As per Bolto, acrylamide polyelectrolytes are more toxic so their permissible limit is only 0.5mg/L. However, in USA, the limit for polydiallyl dimethyl ammonium chloride is <50mg/L, and for dimethyl amine, the limit is <20mg/L. Literature also reveals that polyelectrolytes with high charge density are more toxic than polyelectrolytes with low charge density (Bolto and Gregory, 2007).

### **Conclusion**

The study tries to find out treatment techniques for all kinds of water pollution viz, suspended and dissolved impurities with the help of polyelectrolytes. The study shows that polyelectrolyte as a primary coagulant is more successful in the removal of suspension from water. However, polyelectrolyte as a coagulant aid is efficient to remove both suspended and dissolved impurities in conjunction with an inorganic coagulant. The optimum dosage of polyelectrolyte is low when the amount of pollutants is high. It was found that the efficiency of polyelectrolyte was affected by molecular weight, charge density, characteristics of flocculants, of polyelectrolyte and polyelectrolyte dosages. In a complex situation, dual polyelectrolytes can be used for the removal of oil and grease, colour and other pollutants. Dual polyelectrolytes are not added simultaneously. After homogenous mixing of one polyelectrolyte, another one is added. When polyelectrolyte is used as coagulant aids then the optimum dosage is kept low. So, the toxicity of polyelectrolyte in the aquatic medium is ruled out in this case.

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