Removal of Colloidal Suspension through Coagulation – Flocculation Process In Water Purification – A Review

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ABSTRACT

Received: 30/01/2022 Accepted: 15/11/2023 Online: 13/08/2024

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Background: In water treatment, the most challenging problem that environmental engineers encounter is the separation and settling of colloids from water. **Objective:** The study aims to present various aspects of coagulation-flocculation in the removal of colloidal impurities from water. The shape, size, and significant characteristics of colloidal particles have been discussed in the present study. The colloidal systems, origin and nature of charge on colloids, and the stability theories including the concept of zeta potential have also been explained. The study has also been extended for conventional coagulants along with natural coagulants. The effectiveness of various coagulants has been extensively reviewed and presented herein. The review indicates that natural coagulants are equally effective as conventional salt-based coagulants. **Conclusion:** It is concluded from the present study that polymer-based coagulants are more effective than conventional coagulants. The removal of colloidal impurities from water is an important aspect. The separation of colloidal impurities by coagulation and flocculation is an effective method of water treatment that is safe and sustainable.

 Keywords: Colloids, Coagulants, Polymers, water treatment, Turbidity, flocculation. DOI:<https://doi.org/10.24126/jobrc.2024.18.2.827>

1-INTRODUCTION

 Impurities of colloidal size are the most challenging problem for engineers in the separation and settling of colloids from water and wastewater. The colloids are composed of particles having at least one dimension lying within the range of 0.0000001 to 0.0001 cm (ten Angstrom to one micron). Biswas has reported that colloids of size, 1.0 micron require 10 days of settling time (1). In the water treatment process, it is not possible to provide such a long detention period for settling the colloidal suspensions. It has, therefore, been important to agglomerate two or more colloids to form flocs resulting in the increase of mass concentration, which makes quicker settlement possible. Coagulation is a physio-chemical process termed as coagulation achieves this agglomeration of colloids into larger size flocs. In general inorganic coagulants are used for water and wastewater treatment. When coagulants are used, particle coagulation arises mainly by 2 mechanisms: (i) adsorption of positively charged colloid particles surface in the water/wastewater, which sources charge neutralization; and (ii) entrapment of colloidal particles and precipitate. In the present study, an effort has been made to compare the effectiveness of the removal of colloidal impurities from water and wastewater using a variety of coagulants. The study has further provided a better understanding application of the coagulation and flocculation process and has solved many complexities of the physio-chemical treatment of water and wastewater.

2. COAGULATION

 Coagulation is a process of destabilizing colloidal particles so that particle growth can occur as a result of particle collisions. The aluminum salts can be effective as coagulants in two ways: (1) by adsorption to produce charge neutralization, and (2) by enmeshment in 'Sweep Floc', the required coagulant dose depends upon how the destabilization is achieved (2). In the former, the required coagulant dose is lower which increases with increasing colloid concentration. In 'Sweep Floc' a high dose of coagulant is necessary to produce a gelatinous metal hydroxide precipitate which can be effective in situations where colloidal concentrations are low. According to Packham, the coagulant dose is markedly pH-dependent for the concentration of colloidal suspension in terms of turbidity which has a cation exchange capacity (CEC) (3). For colloid concentration normally encountered in natural waters, coagulation is most difficult when alkalinity is low (2). The studies on colloids, coagulants, and flocculation are reviewed in the subsequent sections.

3. COLLOIDS

 In water generally, impurities are presented in the form of suspension, colloids, and dissolved particles. The colloidal suspensions have at least one dimension lying within the range of 10 (ten) Angstrom to 1 (one) micron i.e., 0.000,0001 to 0.001cm (4). Individual molecules or ions present in the water/wastewater in dissolved forms are presented in Fig. 3.1 shows the size and range of impurities in water. The time of settlement of such particles takes a longer duration in comparison to coarser particles, which is not possible to provide such a long detention period for settling. Biswas (1) has reported that colloids of size, 1.0 micron require 10 days of settling time in a one-meterdeep column. The settling characteristics of discrete suspended particles have been given in Table (3.1).

Figure (3.1): Size and Range of Impurities in Water

 It has, therefore, been important to combine two or more of the colloids to form flocs resulting in the increase of mass density through a physicochemical process, which makes quicker settlement possible. Therefore, thorough knowledge of the colloids present in the natural turbid waters is essential to accomplish the above tasks. The characteristics of colloids, the development of theoretical approaches using the salient characteristics, and their numerous applications are reviewed in the subsequent sections.

 The characteristics of colloidal particles depend upon the size, shape, and structure of particles along with the distribution of their quantities. The microscopic visibility does not show the sharp dividing line between giant molecules and small colloids; and between large colloids and fine heterogeneous suspensions. The relation between the particle size and the specific surface area is more important in view of colloidal settlement. As the particle size is reduced, the specific surface area gradually increases and a stage comes when further reduction in the particle size can destroy the interface between the disperse phase and dispersion medium, and also the free surface energy disappears. Therefore, the lower limit of the colloidal state is reached and the system becomes homogeneous. The change in shape and the specific surface result in simultaneous changes in the boundary energy of the system. Due to it, the adsorption, in turn, will cause a change in the electric double-layer. It also considerably influences the forces and their area of action between dispersed particles. The most important of these are (1) the frictional resistance between dispersed particles and dispersion medium, and (2) the viscosity of a dispersed system. The relationship between the frictional resistance between dispersed particles and the dispersion medium and the form of the colloidal particles can be stated by Stoke's Law.

 The structure of the colloid is characterized by three coordinating factors; A) Particles; B) Continuous medium and C) Stabilizing agent (4). Particles are the distinctive units of colloids (5). They are not in general molecules but commonly consist of several molecules. They may be wholly separated from each other, and therefore, independent; or more often they may be grouped to form still larger structures, some of which may have a high degree of organization, whereas others may be merely loose aggregates. The spherical particles are the densest aggregates due to the most balanced force. Polyhedral particles generally give loose structures. The *continuous medium* may be a gas, a liquid, or a solid (water in the present case). Its continuity is broken only by the particles themselves or by their points or surfaces of attachment to each other. The *stabilizing agent* must be of a dual nature, having affinity to both the particles and the medium, which are united by it. In some cases, it may be supplied by polar chemical groups on the surface of the particle itself.

3.1 Characteristics of Colloids

 The characteristics of colloids are attributed to their size, shape, and structure when they are dispersed in the dispersion medium. The size, shape, and structure of the colloid differ from those of the suspension and the solution and so also the characteristics (5,6). Their respective characteristics are presented in Table (3.2).

Table (3.2): Characteristics of Colloids

 The characteristics indicated in Table (3.2), the last four i.e. settle ability, coagulation, surface phenomenon, and presence of electric charge have vital importance to water treatment processes.

3.2 ORIGIN AND NATURE OF CHARGE

 The aluminum and iron salts are generally described by two diverse systems i.e., charge neutralization and sweep flocculation. The pH and coagulant dosages have a significant impact on these mechanisms. Generally, colloids are electrically charged and the system is electrically neutral due to the availability of an equal number of free positive and negative charges. The charge on the surface of the particle may be derived from the dissociation of surface molecules. It may also arise from the dissociation of molecules adsorbed on the particle or the preferential sorption of one ion rather than another. Another possible alternative is a negatively charged particle, which sorbs trivalent aluminum ions resulting sorption of chloride ions (4). The total effective charges on any particle or a surface are the net algebraic sum of all the charges thereon.

 The colloidal particles acquire charges from the dispersion medium through electrophoresis and electroosmosis. Agglomeration of the charged particles is the sole aim of the process of flocculation in raw water (surface) and wastewater treatment processes. Agglomeration of the particles depends upon three primary parameters namely, Van der Waals interaction (usually attractive forces bringing the particles together), frequency of collision between particles, and the electrostatic interaction (usually repulsive forces which prevent aggregation or collision). The systematic understanding of flocculation and its development requires up-to-date knowledge of colloidal

stability. Helmholtz's double-layer theory and Gouy's diffuse double-layer theory provide basic ideas about colloidal stability (5,7).

 Cornwell and Bishop reported that this could be accomplished by particle destabilization due to doublelayer compression or physical entrapment of particles (8). Flocculation is a method of moving the coagulated particles into contact to form bigger particles (9,10). The development of particle destabilization and its collection is complex. Therefore, it is considered to be a two-stage process of particle movement and particle entrapment. Agglomerating of particles must stick together with each other and adhere upon collision (11,12). Generally, flocs do not combine with each other, which reach a steady-state size for a given condition. The floc growth is to be checked by floc splintering so that the rate of aggregation is considered a balance between floc development and floc splintering (13,14,15,16,17). In suspension, the stability of flocs is dependent upon the breakage. Generally, aggregate breakages are directly related to the strength of the bond and the number of the bonds holding the floc together. The floc breakage may be irreversible to some extent so that broken fragments do not readily re-form (14,18).

 The Zeta potential or electrokinetic potential is a measure of the stability of a colloidal system and is of great practical importance in the purification of water. When it decreases, the stability decreases as well (19). In case, colloids' charge with a negative charge has a large zeta potential, the stability of the colloids increases due to increased Brownian motion (20). Particles tend to amass together to form a bigger floc, when zeta potential approaches zero. This is the neutralization of charge in the process of coagulation. When the floc size is developed into a bigger size in comparison to the initial colloidal particle size, the effect of shear-induced diffusivity will increase.

 In order to make the coagulation of colloids effective, the distance between them must be such that the energy of their mutual attraction (due to Van der Waals' forces) is greater than the energy of heat movement. The coagulating action of these coagulating ions depends on their valency: the higher the valency, the stronger the coagulating action of the ions (21). However, Howe *et al.,* indicated that membrane fouling is increased by low coagulant doses instead of no coagulation (22). The jar test provides the optimal coagulation condition, which results in minimizing the fouling. In addition, Irene et al*.* discussed the effects of pH on zeta potentials and reported for pH 5 to 8 which is dependent on pH (23). A similar phenomenon has also been reported by a host of researchers (11,24,25,26) in the presence of natural organic matter.

4. COAGULATION

 Coagulation is a two-step process involving particle destabilization and particle collisions by moving towards destabilized particles. Destabilization of particles occurred due to the addition of a suitable coagulant, and particle contact through rapid mixing. In a typical water treatment plant coagulation occurs in the rapid mixing and the flocculation units. Particle collisions develop the formation of the floc and coagulation process which destabilizes colloidal particles. The negatively charged surfaces of colloidal particles are coated by positively charged species in the coagulation. Coagulation is primarily influenced by three factors: coagulant dose, pH, and colloid concentration (2). The coagulant dose necessary for destabilization is seen to be a function of the concentration of the colloid to be aggregated. The characteristics of the polymers, which are formed in such systems, are affected by the degree of over-saturation (coagulant dose and pH) in the system. When pH decreases below the iso-electric point, the average positive charge of the metal species tends to increase, thereby enhancing both destabilizations by charge neutralization and the possibility of re-stabilization by overdosing. The aluminum salts can be effective as coagulants in two ways: (1) by adsorption to produce charge neutralization, and (2) by entrapment in 'Sweep Flocculation', the required coagulant dose depends upon how the destabilization is achieved (2). In the former, the required coagulant dose is lower which increases with increasing colloid concentration. In 'Sweep Floc' a high dose of coagulant is necessary to produce a gelatinous metal hydroxide precipitate which can

be effective in situations where colloidal concentrations are low. The salt composition of water has a significant effect on the process of coagulation. Anions of weak acids account for the capacity of the buffer and promote hydrolysis of the coagulant. Cation can alter the charge on colloidal particles. Charged neutralization is, however, a vast over-simplification of the coagulation forces in which, for example, kinetic consideration may outweigh other factors such as Zeta-potential.

4.1 Coagulants

- (i) Aluminum Sulphate (Alum)
- (ii) Poly-aluminum-chloride (PAC)
- (iii) Poly-aluminum silicate (PASiC)
- (iv) Poly-aluminum-hydroxyl-Sulphate (PAHS)
- (v) Ferric chloride (FC)
- (vi) Fe (III)
- (vii)Polymers
- (viii) Magnesium
- (ix) Others and Natural coagulants

4.1.1 Coagulation with Aluminum Sulphate (Alum)

 Edzwald, reported that the alum is the most frequently used coagulant for water and wastewater treatment because of its effective proficiency and small cost (27). Usually, Al^{3+} is mixed directly to the water for the treatment of colloidal suspension and Al^{3+} ions hydrolyze swiftly producing a range of Al^{3+} species (28). The charge neutralization is foremost in a lower pH range $(4.5) and generally sweeps flocculation in a neutral pH (6–$ 8) in the coagulation process (29,30). Licsko´ reported, that the positively charged aluminum species are most important for destabilization (31). The optimum coagulant dose and development of flocs depend upon physical and chemical properties in the coagulation process (32,33,34,35). The efficiency was reduced for colloidal suspension removal in cold water (36,37), therefore, Epstein found that the requirement of coagulant would be increased than the optimal dosages (38). Koether et al., compared alum and PAHS coagulants and their performance on drinking water in batch scale operation with a modified jar filtration test and found satisfactory turbidity removal from raw water (39). The use of alum proved that the color and dissolved organic matter are removed from wastewater to such an extent that the water can be reused for daily needs with the insignificance of health risk (40).

 Temperature affects turbidity and particle counts during coagulation through the removal of turbidity (41,42). Aluminum and iron slats effectively remove maximum impurities from water and wastewater, which include colloidal and dissolved organic substances (43). Ahmad *et al.* reported that alum, PAC, and Chitoson have the same percentage of removal of sediments (95%) in the treatment of wastewater (44). Chang *et al.,* achieve 95% turbidity removal efficiency which includes various organics (45) while the study of Feng *et al.,* showed the effects of low temperature on Aluminum (III) (Al) hydrolysis by constructing a solubility diagram for amorphous aluminum hydroxide (Al(OH)3(am)) and a distribution diagram of hydrolyzed Al species. The distribution diagram indicates that the monomeric Al, Al₁₃, and solid-phase Al (OH)₃ were alternately the predominant species with the increase of pH. At low-temperature coagulation process slowed down which decreased in accumulation rate and rate constants (46). The Alum demonstrated much better results in phosphorus removal than Calcium Chloride (CaCl2) (47). Zhu *et al.,* studied the floc characteristics and found that the floc size, the density increased with molecular weight (48). The alum has maximum efficiency for the removal of sorbed DAX-8 fraction (SDF)

compounds from surface water (49). Ferrous sulphate and alum are used in the chemical treatment of dairy wastewater (50). Alum is more effective than ferrous sulphate in the treatment of synthetic dairy wastewater (51). Researchers encounter solid-liquid separation problems as biogenic selenium, Se (0), has colloidal properties which leads to poor settling and membrane fouling (52). A compilation of some studies on the removal of colloidal suspension by alum with different experimental conditions is presented in Table (4.1).

Sl. No.	Experimental Conditions	Removal Efficiency	Reference
1.	$pH - 4 - 10$	80%	Ching et al., 1994
2.	pH - 6 - 8, true color unit-20-30, Turbidity	90%	Koether et al., 1997
3.	pH= 5 - 7.65, turbidity, Temp., DOC, TDS, color	$60 \pm 30\%$	Annalie Roux et al., 1997
4.	pH 6-8	\overline{a}	Lee et al., 2000
5.	$pH = 5 - 9$, Turbidity = 80 NTU, Temp. = 1.5 - 200C, Color	86-98%	Larry Braul et al., 2001
6.	$pH = 5 - 9$, Turbidity = 3 NTU, Temp. = $1.5 - 200C$	80%	Larry <i>et al.</i> , 2002
7.	pH and Turbidity, coagulant dosage	\overline{a}	Gao, <i>et al.</i> , 2002
8.	$pH = 5 - 9$, Turbidity, Temp. = 5 - 200C, Color	$-$	Jinming Duan et. al. 2003
9.	pH 4.5 6.5, Turbidity,	--	Dampsey, 1989
10.	Temp., Turbidity	90%,	Tao et. al, 2006
11.	pH, turbidity	95%	Ahmad et. al. 2006
12.	pH 6-8		Kim et. al, 2007
13.	pH - 5-8.5, TOC, DOC, turbidity, and UV254	95%,	Chang et. al, 2007
14.	$pH = 3-9$, Temp= 4 -250C	$\mathbb{L}^{\mathbb{L}}$	Feng et. al. 2008
15.	$pH = 3-9$, Temp= 2 -220C, Turbidity = 3 -16.5 NTU	75%	Feng Xiao et. al. 2009
16.	$pH = 3 - 11$, Phosphorus	83%	Sawsan et. al. 2009
17.	pH, turbidity	-1	Kokila et. al. 2011
18.	pH, turbidity	\overline{a}	Mahshid et. al. 2013
19.	NOM, pH, Turbidity	95%	Zeeshan et. al. 2013
20.	$pH(2-12)$, Turbidity,	$92 \pm 2\%$	Lucian et. al 2015

Table (4.1): Removal of Colloidal Suspension by Alum

4.1.2 Poly-aluminum-chloride (PAC)

Poly-aluminum-chloride (PAC) is a pre-polymerized Al^{3+} chemical, containing a range of hydrolysis and polymeric species. The rate of hydroxide precipitation is generally slow after dilution addition of preformed polymers (53). The polymers have a high cationic charge which improves charged neutralizing ability and becomes more successful at a smaller dose as compared to conventional coagulants. Poly Aluminum Chloride (PAC) is one of the inorganic polymeric coagulants (IPO), which is most useful under unstable conditions. In comparison to Al^{3+} salts, PAC has many advantages: 1) brisk aggregation velocity, 2) larger and heavy-weights, and 3) smaller coagulant dose. Therefore, PAC is also used widely in place of alum in the coagulation-flocculation process throughout the world (54). High removal efficiency by using PAC as a coagulant in the treatment of high-turbidity storm water (55). The benefit of using a PACl chemical is intangible at a higher dosage of coagulant (56,57). The use of PAC can remove the color and dissolved organic matter from wastewater and water can be reused for daily needs with the insignificance of health risk (40). The effect of pH and various coagulants and their doses on dye wastewater for removal of color COD and suspended solid and found removal efficiency of 90%, 88%, and 95%

respectively (58). The PAC showed constantly satisfactory results for turbidity and colloidal particle removal at various temperatures and dosages (42).

Gao *et al.*, found in the study that PAC enhances accumulating efficiency and provides excellent coagulating effects, when kept in store for a longer duration may weaken charge effectiveness in the coagulation process (28). The PAC was expected to have high-efficiency antimony removal because it is similar to arsenic (59). Uses of PAC as a coagulant for water containing clay suspension and humic acids and found the uniform blanket behavior in an up-flow bed (60).

 The blanket behaved inconsistently when the dose of Coagulant (PAC) increased, it is due to the amassing of Aluminum salts at the bottom of the blanket and meager mixing, the hydrolysis of which salts produced a low pH environment, reversing the charge of particles in the blanket. The PAC along with other coagulants has similar percentage of removal of colloidal suspension (95%) in wastewater treatment (44,57).

The turbidity and natural organic matter (NOM) do not affect the coagulant; however, alkalinity and temperature are the most important variables for selecting coagulants (61). The bio-coagulant and PAC performed well in turbidity removal (62). The effective removal of turbidity is also related to the organic matter in water (63). Cheng *et al.,* measured the floc diameter by monitoring the turbidity oscillation amplitude with a Nephelometric turbid meter monitoring system (NTMS) (64). The optimum dosage of PAC (16mg/l) in the optimum temperature (220 C°) and pH (6.5 – 8) range (65). Ozone (O₃) pretreatment could enhance the coagulation effect of the secondary effluent. Pollutant removal efficiency was recorded higher by the addition of Ozone with PAC (4 mg/l) as compared to regular dosage of PAC (6 mg/l) (66). The removal efficiency of colloidal suspension at $4 < pH > 7$ from surface water source (67). The color removal 85.62 % by PAC in the treatment of dye and humic acid wastewater (68). Éric *et al.*, examine the effect of temperature and water characteristics on the efficiency of a dairy wastewater Coagulation-Flocculation using poly-aluminum chloride (PAC) as a coagulant and found that the settling time is required more at lower temperatures below $10^{0}C$ (69). A compilation of some studies on the removal of colloidal suspension by poly-aluminum-chloride (PAC) with different experimental conditions as in Table (4.2).

Sl. No.	Experimental Conditions	Removal Efficiency	Reference
1.	pH, Turbidity		Viraraghavan and Wimmer, 1988
2.	$pH = 5 - 9$, Turbidity = 3 NTU, Temp. = 1.5 - 200C	80%	Larry <i>et. al.</i> 2002
3.	pH and Turbidity, coagulant dosage	--	Gao, et. al 2002
4.	pH - 5-9	95%,	Annadurai et. al, 2003
5.	$pH = 5 - 10$, Turbidity = 17 NTU, Temp. = $1.5 - 200C$	98%	Kang, et. al. 2003
6.	$pH - 5-7.5$, turbidity, TOC	90%,	Chang et. al., 2004
7.	Turbidity = $40-1000$ NTU, pH	--	Sung, et. al. 2005
8.	pH - 6.9 -7.6, Turbidity - 6 - 12, TOC	90%	Chen et. al, 2006
9.	pH, turbidity	95%	Ahmad, et. al. 2006
10.	pH, turbidity		Hong-Zhang et. al, (2007)
11.	$pH = 3 - 10$, Turbidity = 600 NTU,	--	Cheng, et. al. 2008
12.	pH, turbidity, Temp.	--	Cheng, et. al. 2010
13.	pH, Temp. Turbidity (NTU),	95%	Junguo He et. al. 2011
14.	pH, Temp. turbidity, COD,	79%	Cheng L H et. al. 2011
15.	pH, Temp. Turbidity		Zhonglian Yang, et. al. 2013
16.	COD, Color, pH, Turbidity	71-88%	Zhanmeng Liu, et. al, 2013

Table (4.2): Removal of Colloidal Suspension by Poly Aluminum Chloride (PAC)

4.1.3 Poly-aluminum silicate (PASiC)

 PASiC coagulant can be prepared by adding Aluminum salts and activated silica. The coagulation performance of PASiC coagulants was evaluated to optimize the preparation conditions and control of Al/Si ratio (28). As a result, a stable and effective composite coagulant developed. Particle size and coagulant dosages along with pH values were observed during the coagulation. A variety of organic polymers were used and also observed that poly anions and uncharged hydroxylated polymers have been most successful in colloidal suspension removal. Gao et al, reported that the manufacturing and use of inorganic polymer flocculants (IPF) have increased very rapidly all over the world (70). These compounds behave differently from usual water treatment coagulants in various respects (71). The advantages of IPF include superior efficiency and low cost as compared to traditional organic polymer flocculants. The IPF has coagulation-flocculation characteristics that hold similarities with common salts and organic flocculants (72). Studies reported from Japan and China confirmed that the addition of Al or other salts can delay the formation time of activated silica (73). Several other investigations show that composite prepared by aluminum-silicate polymer is enormously active for the removal of colloidal suspension and humics in cold water treatment (74,75).

 The preparation of polymers involves aluminum-based compounds by controlled environments and other numerous parameters that affect the level of polymerization (54). In some cases, the cost of polymeric coagulants increases significantly due to synthetic requirements. Only 10% of the cost is added in comparison to alum, as it is required to find out the polymeric coagulant has a low cost. The effective removal of colloidal suspension was not only dependent upon the type of coagulant but also depend on organic matter (63). Zhang *et al.,* prepared a new poly-silicate coagulant named poly-aluminum-calcium-silicate sulphate coagulant (PACSS) for the removal of suspended solids and organic matter (76). The experiment shows that the removal efficiency of turbidity is 96.2% and COD is 84.3% from wastewater. This coagulant has efficient colloidal suspension removal efficiency as compared with alum and polymeric aluminum sulphate (PAC). The recent works on the removal of colloidal suspension from water by Poly-aluminum silicate (PASiC) with variable experimental conditions are shown in Table (4.3).

Sl. No.	Experimental Conditions	Removal Efficiency	Reference
1.	pH, Turbidity		Viraraghavan and Wimmer, 1988
2.	Turbidity, pH		Gao et. al, 1990
3.	Turbidity		Hasegawa and Hashimoto et. al, 1991
4.	pH, Turbidity		Arnold-Smith et. al, 1992
5.	$pH = 7-7.4$, Turbidity 75 NTU,	94%	Gao <i>et. al</i> , 2002
6.	$pH = 4-9$, Turbidity 26.2 NTU, Color	92%	Gao et. al, 2003
7.	$pH = 3 - 10$, Turbidity = 600 NTU,		Cheng, et. al, 2008
8.	$pH = 5 - 10$, SS 161, COD 500, Turbidity 46.4, Temperatures = 26° C	96%	Zhang, et. al, 2012

Table (4.3): Removal of Colloidal Suspension by Poly-Aluminum Silicate (PASiC)

4.1.4 Poly-aluminum-hydroxyl-Sulphate (PAHS)

 The poly-aluminum- hydroxyl- Sulfate (PAHS) is prepared with alum and powdered limestone (77). A modified jar filtration test has been applied to compare bench operations (78) for more than 35 days WTP. It was observed that PAHS was formed using alum and limestone powder during the test at variable dosages of PAHS and alum for turbidity. The lime was added at the rate of 0.5 to 3 mg Al/ CaO (79). Paramasivam *et al.* discussed that PAHS works efficiently at low temperatures (80). The aluminum would be in a relatively insoluble particulate form in the lime (81). Hence, the researchers concluded that the alum can be replaced by the use of PAHS in water wastewater treatment plants. This would involve large quantity of lime consumption and aluminum in clarifier sludge. It has been observed that mostly, water treatment sludge discharge into surface waters (82). Many researchers are still investigating the use of coagulants for removing colloidal suspension from water, therefore compilation of some researches on the elimination of colloidal suspension from water by Poly-aluminum-hydroxyl-Sulphate (PAHS) with different investigations are presented in Table (4.4).

4.1.5 Coagulation with Ferric Chloride

The salts of Fe³⁺ are commonly used as coagulants in water and wastewater treatment. Fe³⁺ is hydrolyzing its role in coagulation. Usually, Fe^{3+} coagulant is added to raw water directly for treatment and Fe^{3+} ion hydrolyze rapidly producing a range of Fe³⁺ ions including mononuclear ions such as Fe(OH)²⁺ and Fe(OH)⁺², and polynuclear species (several Fe³⁺ ions) such as $Fe_2(OH)_2^{4+}$. At low coagulant doses, charge neutralization usually happens through the adsorption of dissolved metal ions and Sweep-flocculation happens at high dosages of coagulant (11,35). The sweep-flocculation of hydroxide precipitates encourages coagulation by increasing the inter-particle collision rate and floc formation of suspended particles (25,83). The adsorption method was applied to remove organic matter (humic substances) from raw water and precipitated as metal hydroxide (84,85,86,87,88,89). In the water treatment, pH and coagulant dosages also affect the process. Generally, at lower

pH conditions and low doses of coagulant precipitation of metal-humic compound take place; at pH 6.5 -7.5, optimal turbidity removal with metal coagulants (90), and at pH 5 optimal removal of organic matter was observed (85,91). However, at pH $4 - 5$, for coagulation of NOM with ferric chloride (FC) was optimized (92). It has been observed that pH (pH $5 - 6$) for the coagulation process of natural organic matter with aluminum salts due to the higher acidity of Fe³⁺ (85) (91). It requires to removal of both turbidity and NOM by enhanced coagulation which is the extension of traditional coagulants. The dosages and pH adjustment required to enhance coagulation which entails selection of the coagulants (87,88,93,94).

 The ferric chloride was more efficient than alum at 30C° water with low turbidity (2 NTU) (95). A coagulant such as ferric chloride can effectively remove the color and dissolved organic matter from wastewater for reuse for daily needs with insignificance of health risk (40). Ferric chloride can exceptional the removal of DOC, turbidity, and numbers of particles at optimum coagulant dosages (42). Xiao *et al.,* found in the study that Perfluorooctanesulfonate (PFOS) and perfluorooctanoate (PFOA) are organic pollutants that have removal efficiency by conventional coagulants \leq 20% (96). Coagulation with FeCl₃ and the subsequent photo-Fenton process shows the best results for the removal of COD of 92.4% was observed by (97).

 Ferric chloride and chitosan as coagulant aids had removed arsenate and arsenate removal in synthetic water (98). Many researchers are still studying the use of coagulants for the separation of colloidal impurities from raw water, the compilation of some recent studies on the removal of colloidal suspension from water and wastewater by ferric chloride with different experimental conditions is presented in Table (4.5).

Sl. No.	Experimental Conditions	Removal Efficiency	Reference
$\mathbf{1}$	pH 6.5-7.5	$-$	Hall and Packham, 1965
3	TOC, pH, coagulant doses, Temp.	$-$	Kavanaugh, 1978
$\overline{4}$	Color, turbidity, suspended solids, pH		Dampsey, 1984
5	Turbidity, pH, Temp		Montgomery, 1985
6	pH, Turbidity Humic Acid		Edwards and Amirtharajah, 1985
7	pH, Turbidity, Temp.		Randtke, 1988
8	$temperatures < 27$ degrees, turbidities < 2 NTU, pH		Haarhoff and Cleasby, 1988
9	pH, Turbidity, Temp.		Amy et. al, 1989
10	Turbidity, pH, Temp		Amirtharajah and O'Melia, 1990
11	pH and Turbidity	--	Amirtharajah, et. al, 1993,
12	pH - 3.0 - 9, Turbidity 5-40 NTU, Electrophoretic mobility (EM)	80%	Ching et. al, 1994
13	pH, Turbidity, Temp.		Cheng et. al, 1995
14	TOC = 2 to 11 mg/L, $pH = 5-8$,		Crozes et. al, 1995;
17	$pH = 5 - 9$, Turbidity = 3 NTU, Temp. $= 1.5 - 200C$	80%	Larry et. al. 2002
19	pH, Turbidity, arsenite		Farid Hesami et. al. 2012
20	COD, pH, Turbidity	92%	Jeremi Naumczyk, et. al, 2014

Table (4.5): Removal of colloidal Suspension by Ferric Chloride

4.1.6 Fe (III)

The use of $Fe³⁺$ salt as a coagulant quantity in which flocculation processes take place, and factors on which these depend, and the mechanism involved (99). Temperatures affect water and particle movement, particle interface, hydrolysis of coagulants, and adsorption and precipitation rates in the coagulation-flocculation process (11). This is known that when the particle diameter is more than 1µm, the ortho-kinetic collision rate greatly exceeds the rate due to Brownian diffusion even at a fairly low shear rate (100).

The low-temperature conditions can lead to low coagulation efficiency due to the slow mixing in water treatment (101,102). In the numerous studies of the turbulent mixing characteristics, (103,104,105) elevated queries about the effectiveness of velocity gradient or energy dissipation rate to describe inter-particle collision. At high temperatures and pH, the rate of hydrolysis of Fe^{3+} salt is also accelerated and the time for the formation of soluble polymeric iron is found to decrease rapidly (106,107). In a study done by Bartosz *et al.,* it was observed that coagulation involving Fe (III) salts in a standard coagulation test COD, color, turbidity, and suspended solid [SS] reduction efficiency observed more than 95% (108).

 A change in pH at low temperatures causes a change in optimum coagulation dose of coagulants (109). The rate of flocculation and enhancing the charge neutralizing ability of $Fe³⁺$ coagulant (99). The kinetics of flocculation in water treatment plants are usually particle collisions by Brownian motion, fluid shear, and differential sedimentation (105). Sofia *et al.* attempted to evaluate the efficiency of arsenic removal processes, by Fe (III) coagulation and found that the removal ability of Fe (III) was more efficient than the adsorption process (110). Selenium (Se) is one of the contaminants required to be regulated during drinking water treatment, however, very little information was available on Se removal by coagulation. In the study, it was observed that Fe-based coagulant (98% at a dose of 0.4 mM Fe/l) is much more efficient than aluminum-based coagulants in Se removal (111). A compilation of some studies on the removal of colloidal suspension by Fe (III) with different experimental conditions is presented in Table (4.6).

4.1.7 Polymeric Ferric Sulphate (PFS)

 The Iron salts coagulants are mostly used in water treatment (112). Polymeric ferric salt has been developed with different techniques for coagulation as reported by Fan and Zhang (113). The polymeric ferric chloride (PFC) and polymeric ferric sulfate (PFS) are the most important polymeric iron compounds and the preparation, classification, and application of PFC have been already conducted (114,115). It was observed that PFS is less corrosive. It also observed that at high pH, the iron residue is stable and highly stable at pH-3, which

also reduces COD and color from water more effectively than aluminum and iron (116). PFS is extensively used in water treatment due to better efficiency in colloidal suspension removal as well as cost-effective. The new PFS synthesis method has the following advantages: a) PFS does not contain any toxic reagent; b) It reacts fast coagulation process, which significantly enhances the capacity of a plant in water treatment; c) Low capital cost investment as compared to existing treatment process; and d) The amalgamation proceeds at relatively soft and gentle reaction conditions (113). Fan et.al, reported that coagulation performance in terms of colloidal suspension removal by PFS and ferric sulphate, showed that PFS is better than FS in a reduction in a colloidal suspension of Kaolinite (112). Song *et al.,* reported that after coagulation, with the help of filtration units, a very high arsenic removal (more than 99%) from high-arsenic water (5 mg/l arsenic concentration), producing a cleaned water with a residual arsenic concentration of 13 µg/l (117). Jian *et al.,* observed that coagulation efficiency by the addition of polymers provided better coagulation/flocculation efficiency in turbidity removal, which enhanced the flocculation index and reduced the use of residual ferric (118). The poly-ferric flocculant has good performance to clean the polluted water (119). A compilation of some studies on the removal of colloidal suspension by Polymeric Ferric Sulphate (PFS) with different experimental conditions is presented in Table (4.7).

Sl. No.	Experimental Conditions	Removal Efficiency	Reference
1.	pH , Temp 250 c		Knight and Sylva, 1974;
2.	pH, Turbidity		Mikami et. al, 1980;
3.	pH, Turbidity		Stumm et. al ,1981
4.	pH, Turbidity		Fan and Zhang et. al, 1997
5.	$pH - 5 - 9.2$	80%	Fan <i>et. al</i> , 2002
6.	pH, Arsenic, Temp.	99%	Song, et. al, 2006
7.	$pH = 4 - 8$, Turbidity, DOC	90%	Jian et. al , 2011
8.	$pH = 7 - 8$, Temp., COD,	91%	Guo, et. al, 2012

Table (4.7): Removal of Colloidal Suspension by PFS

4.1.8 Poly-Alumino–Iron Sulphate (PAFS)

 The mostly inorganic salts of ferric iron are used as coagulants for water and wastewater treatment (120). The performance of pre-hydrolyzed metal–ion coagulants extensive interest and attention seen in recent years, based on ferric iron, and some of these performed better than conventional coagulants (FS) (75,121,122,123,124). It has been observed superior performance of pre-polymerized coagulants with a wider range of pH.

 Jiang and Graham reported that excessive cationic charge on polymeric substances is presented long enough in solution, which extremely enhances the charge neutralization and chemical precipitation (125). The PAFS removed the higher organic matter as compared to conventional coagulants (Aluminum Sulfate and Ferric Sulfate) and the less sludge production in the raw water treatment at the laboratory. The addition of several additives in different combinations with the commonly coagulants can obtain the maximum colloidal suspension removal efficiency for water (126). A compilation of some studies on removal of colloidal suspension by Poly-Alumino–Iron Sulphate (PAFS) with different experimental conditions is presented in Table (4.8).

4.1.9 Coagulation with Polymers

 Synthetic organic polymers can be effective as coagulants or coagulant aids. These polymers are made of long-chain molecules comprised of several subunits called monomers. Ruehrwein and Ward studied the mechanism by which clay particles were aggregated by polyelectrolytes and proposed the concept of chemical bridging to explain the behavior of polymer-clay systems (127). It suggested that bridges were formed when the polymers became attached to the surface of clay particles by the process of electrostatic attraction or ion exchange. The size and shape of the polymer in solution are important in determining its effectiveness and are influenced by ionic strength, ionic valences, and pH. Generally, polymers have a helical molecular structure that contains carbon chains with ionizing groups attached together (128). As the ionized groups become attached to the surface of colloidal particles. Hence, the charges of colloids are neutralized and the polymer starts to coil and form a dense floc for settling. An overdose of polymer may result in an excess of neutralized sites and cause the stable, straight-chain structure to persist. A low ionic strength or a pH that favors a high degree of ionization may also cause the polymer to remain extended and thus adversely affect coagulation. Polymers are not acidic and do not lower pH as alum does, thus, their use offers a decided advantage for treating low alkalinity waters similar to those found in the Southeastern United States, particularly if the waters are high in turbidity. Other advantages of polymers over alum include reducing the volume of sludge produced, providing a sludge that is easier to dewater, preventing the carryover of soluble aluminum into the distribution system, and preventing the carryover of light floc, (129). Jian *et al.,* reported that the addition of polymers provided better coagulation/flocculation efficiency in turbidity removal (118). A compilation of some studies on the removal of colloidal suspension by Polymers with different experimental conditions is presented in Table (4.9).

4.1.10 Coagulation with Magnesium

Thompson *et al.*, have shown that magnesium precipitated as $Mg(OH)$ ₂ can be an effective coagulant for the removal of color and turbidity from natural waters (130,131). For waters high in naturally occurring magnesium, coagulation can be achieved by simply adding a sufficient amount of lime to precipitate Mg (OH) $_2$. For waters low in magnesium it is necessary to provide magnesium through the addition of a suitable salt. One advantage of the process is that both magnesium and lime can be recovered from the sludge and recycled. This

simplifies the sludge disposal problem and improves the overall economics of treatment. The chemistry of magnesium coagulation is a combination of water softening and coagulation chemistry. The so-called basic carbonate, $4MgCO₃Mg(OH)₂5H₂O$, has been used in some cases but is somewhat expensive and relatively insoluble. Dolomitic lime is a promising source of magnesium and is currently being studied on a pilot basis (132). Boon *et al.*, reported that coloring material can be removed (more than 90 %) by MgCl₂ (58). It shows better results in removing reactive dye than alum and PAC in terms of removal efficiency of COD and suspended solid are 88% and 95% respectively. The advantages, disadvantages, and process efficiency of high-pH using magnesium coagulation-flocculation process in relation to various sources of magnesium ions and reviewed the advantages and disadvantages of applicability of application of magnesium in wastewater treatment (133).

 The operational conditions can affect the floc formation time and rate in the magnesium hydroxide coagulation process and observed that floc time for formation generally decreases with the increase of coagulant dose, colloidal suspension, temperature, and pH (134). Zhanmeng *et al.,* prepared the Polyferric (III)- Magnesium(II) Sulphate (PFMS) which was more efficient than PFS, PAC, and PFC in the removal of color, turbidity, and COD from textile wastewater (68). The used of magnesium hydroxide to know the properties of slow mixing in preparation of floc size, and strength and also find out the removal efficiency of turbidity and total suspended solid (TSS) from extremely turbid water (135). A compilation of some studies on the removal of colloidal suspension by Magnesium with different experimental conditions is presented in Table (4.10).

Sl. No.	Experimental Conditions	Removal Efficiency	Reference
	pH, color and turbidity		Thompson <i>et. al.</i> 1972
3	pH, color and turbidity		Thompson, et. al, 1979
	pH, color and turbidity		Semerjian and Ayoub, 2003
6	Turbidity, Temperature and pH		Jianhai Zhao, et. al, 2012
⇁	COD, Color, pH, Turbidity	71-88%	Zhanmeng Liu, et. al, 2013
8	pH, Turbidity, Temp., TSS	98%	Ayoub, et. al, 2014

Table (4.10): Removal of Colloidal Suspension by Magnesium

4.1.11 Coagulation with other coagulants and Natural coagulants

 The chemical salts may exert an ill effect on health while in water treatment because they leave harmful aluminum in case of excessive usage (high dose) (136). Divakaran and Pillai used chitosan to surface water for colloidal suspension removal (137). It can effectively remove colloidal suspension at low dosages (20 to 80mg/l) and also specified that restabilization may appear when applying the excessive dosage. Ahmed *et al.,* used chitosan to treat palm oil mill effluent along with aluminum Sulphate and PAC coagulants (44). Chitosan is more efficient and economical as compared to alum and PAC with more than 95% removal efficiency colloidal suspension from the raw water.

 The Bio-CoA and PAC can remove colloidal suspension from raw water efficiently irrespective of initial turbidity, and no aluminum residue is collected (62). Ma *et al.,* found in the study that permanganate pre-oxidation enhanced both the processes of coagulation and filtration of surface water (138). In a study, removed lead ions from raw water samples using powdered marble wastes (PMW), which is low-cost, and easily available (139). Santhi *et al.*, used dried squamosal seed as a new low-cost activated carbon to remove malachite green and Methylene blue from replicated wastewater which shows 76% removal efficiency (140). Gupta *et al.*, successfully removed rhodamine B, fast green, and methylene blue from wastewater was achieved using red mud (Aluminum industry waste) with 92.5%, 94.0%, and 75.0% removal efficiency respectively (141).

 The used of fly ash from the sugar industry for the effective removal of lead and chromium as absorbents (142). The use of biodegradable coagulant Chitosana cationic biodegradable biopolymer produced by the extensive deacetylation of chitin obtained from shrimp shell wastes and shows the similar results as compare to conventional coagulants (44). Used of the Surjana seed powder (SSP), Maize seed powder (MSP) and Chitosan as a coagulant for removal of Congo Red (CR) dye from wastewater (143). The used of chitosan and Sulphate aluminum as coagulant for treatment of surface water with turbidity removal efficiency of 97% (144).

 The suitability of the coagulation-flocculation process using Moringa Oleifera seed as an environmentfriendly natural coagulant and antimicrobial agent for clarification of turbid water with turbidity removal efficiency up to 99.99% for higher turbid water (145). Optimized of water treatment parameters using Moringa Oleifera as coagulant (natural) for low turbidity water (146). Moringa Oleifera can be used as a potential alternative to alum for the treatment of water in rural and urban areas (147). Moringa Oleifera has been used for the treatment of hard waters and as a bactericidal agent too. Natural coagulant Moringa Oleifera is also used for the treatment of effluent from dairy plant. An optimum dose of MO seed powder as coagulant of 0.6gm/100ml and optimum time is one hour (148).

 The maximum percentage of turbidity removal was observed in oil free Moringa coagulants (149). The removal of turbidity and coliform from raw water by using locally available natural coagulants like Moringa Oleifera was about 89–96% (150,151). The combination of seed powder of Moringa Oleifera and sand filter in water treatment was found more effective for water purification than alone Moringa seed powder in water treatment. Combined treatment produced 99.97% reduction in *E. coli* (152). Other than Moringa Oleifera, use of banana stem juice as coagulant (natural) for treatment of washed-out coolant wastewater tested by Habsah *et al.,* (153).

 Moringa Oleifera is a locally available and alternative purification method in the improvement of water quality like turbidity, pH and hardness. After the application of Moringa Oleifera seeds in pond water treatment, they understood that Moringa is biodegradable, environmentally friendly and non-toxic alternative which can be used in purification of pond water in rural communities (154). The seed oil of Moringa Oleifera having a good physical and chemical properties and it could be utilized successfully for human consumption and for industrial applications (155,156). Oladoja, explored potential use of Cacao Husk Ash Extract (CHAE) as coagulant aid, in the removal of colloidal particles from simulated low alkalinity (pH= 2, 3 and 4) for various range of turbid water i.e. 50NTU, 100NTU & 300NTU (157). The study revealed the effects of the Cacao Husk Ash Extract (CHAE) on the kinetic s of flocculation, pH of the cleared water and sludge quantity. Fu Hong *et al.,* used the Zeolite powder to enhance the adsorption effect in removal of organic matter and turbidity (158).

 The seed and Pectin extracted from pith of orange peel as natural coagulants in removing laundry waste and found that Nirmali seeds are better in turbidity removal (159). A novel treatment process which was used for dye removal from water, by biosorption and photocatalysis under the visible light irradiation shows the 95% removal efficiency (160). Bodlund *et al.,* and Tassia *et al.,* also used the natural coagulant in treatment of surface water along with other natural coagulants (161). A compilation of some studies on removal of colloidal suspension with other coagulants with different experimental conditions is presented in Table (4.11).

Table (4.11): Removal of Colloidal Suspension by other and Natural Coagulants

5. FLOCCULATION

 The terms coagulation and flocculation are generally used to describe the process of removal of turbidity caused by fine suspensions, colloids, and organic color. Flocculation is the second stage of the formation of settleable particles (or flocs) from destabilized colloidal-sized particles and is achieved by gentle and prolonged mixing. In modern terminology, the combination of mixing (rapid) and stirring or agitation (slow mixing) that produces aggregation of particles is designated by the single term 'flocculation.' The slow mix intensity rate must be the minimal able to suspend particles without floc breakage. The process of coagulation is a complex phenomenon, which is influenced by several variable characteristics of colloids as well as dispersion medium – water. Understanding of the process, therefore, involves understanding of these variables independently, and their interactions before and after the addition of coagulants into the solution. Concerning colloidal particles physical properties, such as size-shape-and-structures, and concentration of particles per unit volume of the liquid, chemical, and surface properties (adsorption and electric double-layers) have been recognized. In the case of water, the turbidity, organic color, temperature, alkalinity, pH, dissolved ions, and salt compositions are considered important. Of these, the electro-kinetic phenomena due to the electric double-layer are probably more important concerning particle destabilization and certainly better understood.

6-RESULTS

 Colloidal suspension removal efficiency also varied with the type of coagulant used for water and wastewater treatment. The effectiveness of different types of turbid water using conventional coagulants is presented in Table (4.12).

Low turbidity <10 JTU low alkalinity ≤ 50 mg/l (as CaCO3) High turbidity >100JTU high alkalinity>250mg/l (as CaCO3)

Raw water/wastewater contains colloidal suspensions which are removed by coagulation and flocculation process, however, this is not a new concept for the treatment, and it remains a key option towards the decline of chemical usage in the water/wastewater treatment industry. Several concepts have experienced modifications and improvements over the years through research efforts. Mostly coagulant are used for water treatment such as Aluminum Sulphate (Alum), Poly-Aluminum-Chloride (PAC), Poly-Aluminum Silicate (PASiC), Poly-Aluminum-Hydroxyl-Sulphate (PAHS), Ferric Chloride (FC), Fe (III), Polymers, with maximum turbidity removal efficiency (99%) from water and wastewater.

7- CONCLUSIONS

 Surface water treatment contains colloidal suspensions which are removed by coagulation and flocculation process; however, this is not a new concept for water treatment and remains a key option towards the decline of chemical usage in the water treatment industry. The whole concept has experienced modifications and improvements over the years through research efforts. In this paper various coagulants are used for water treatment such as Aluminum Sulphate (Alum), Poly-Aluminum-Chloride (PAC), Poly-Aluminum Silicate (PASiC), Poly-Aluminum-Hydroxyl-Sulphate (PAHS), Ferric Chloride (FC), Fe (III), Polymers, Magnesium, and Other Naturals were reviewed. Poly-aluminum-chloride (PAC), Ferric chloride (FC), Polymeric Ferric Sulphate (PFS), Poly-Alumino–Iron Sulphate (PAFS), and other coagulants including Natural coagulants have recently been shown to have maximum removal efficiency (99%) in colloidal suspension from water. However, there is a need for further research on the economic viability and quantification of sludge produced which has been largely overlooked. The studies on recovery and recycling of coagulants from the treatment plant in view of protecting the environment and enhancing sustainability have not been performed so far.

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إزالة المعلق الغروي من خالل عملية التخثر - التلبد في تنقية المياه – مراجعة

عبذ السالم تىفيق داود حازم إسماعيل عبذالباري خالذ سهيل العساوي إسراء حسىن سلمان كفيل احمذ

مركز بحوث التقنيات الاحيائية ، جامعة النهرين ، بغداد- العراق قسم علْوم البيئة/جامعة ملية الاسلامية /نيودلهي/الهند

الخالصة

خلفية عن الموضوع: عند معالجة المياه تكون هناك مشكلة وهي الأكثر تحديا التي تواجه مهندسي البيئة وهي عملية فصل وتسوية الغرويات من الماء_. تمت مناقشة شكل وحجم وخصائص الجسيمات الغروية في هذه الدراسة كما تم شرح الأنظمة الغروية وأصل وطبيعة الشحنة في الغرويات ونظريات الاستقرار بما في ذلك مفهوم إمكانات زيتا وتم توسيع الدراسة لتشمل مواد التخثر التقليدية إلى جانب مواد التخثر الطبيعية حيث تم مراجعة فعالية مختلف مواد التخثر على نطاق واسع وعرضـها هنا. تشير المراجعة إلى أن عوامل التخثر الطبيعية لها نفس الفدر من الفعالية مثل عوامل التخثر التقليدية القائمة على الملح. **الهدف من الدراسة:** تهدف الدراسة إلى تقديم جوانب مختلفة من التخثر والتلبد في إزالة الشوائب الغروية من الماء. **الاستنتاجات:** يستنتج من هذه الدراسة أن مواد التخثر ذات الأساس البوليمري أكثر فعالية من مواد التخثر التقليدية وتعد إزالة الشوائب الغروية من الماء جانبًا مهمًا في عملية المعالجة ويعد فصل الشوائب الغزوية عن طريق التخثر والتلبد طريقة فعالة لمعالجة المياه وهي أمنة ومستدامة.

الكلمات المفتاحية: الغرويات، مواد التخثر، البوليمرات، معالجة المياه، العكارة، التلبد.