

Treatment of Printing Ink Wastewater Using Natural and Synthetic Coagulants



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1 Introduction

Water is one of the necessities of life, and preserving it and developing its sources deserves attention [1–4]. Numerous contaminants are found in our water sources due to the development of industry and the widespread usage of complicated chemicals. These compounds have a serious hazard to our ecology since they are difficult to break down. They have significantly grown in number over time, posing a grave threat to the next generations [5–7]. The procedures of printing, color matching, cleaning printing machines, etc. generate printing ink wastewater (PIW). The primary ingredients of ink are pigments, fillers, auxiliaries, solvents, and color carriers (often certain resin compounds with hydrophilic groups, etc.) [8] as shown in Fig. 1. Wastewater from printing ink comprises recalcitrant substances, traces of metals (such as Cd, Hg), adhesives, and pigments. In addition to their vibrant color, these wastewaters have high chemical oxygen demand (COD) values, typically up to 20,000 mg/L. To avoid major environmental issues, printing ink wastewater must be treated before being released [9–11]. Even very low concentrations of dyes in the effluent (less than 1 mg/L for some dyes, for example) cause the water to turn a highly noticeable and unwelcome shade of color. Additionally, it harms water bodies like rivers

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and lakes by obstructing sunlight or promoting eutrophication [12]. Effective treatment is required for (PIW) so it could be disposed of safely in aquatic systems. For the treatment of this sort of extremely colored wastewater, advanced oxidation processes (AOPs), electrochemical techniques, coagulation/sedimentation, and either separately or in combination with other physicochemical methods, are typically preferred [13, 14]. It was reported that utilizing coagulation/flocculation for processing wastewater showed satisfactory results using either organic or inorganic flocculants/coagulants such as aluminum, iron (III) salts, or organic polyelectrolytes [15]. Dyeing processes often involve the use of chemicals and may lead to changes in the pH range of the surrounding environment. Pipe lines and tanks made of aluminum alloys are more susceptible to corrosion in both highly acidic and highly alkaline environments. If the dyeing process alters the pH of the surrounding medium significantly, it could affect the corrosion rate of aluminum alloys. Water and wastewater treatment was conducted using flocculation and coagulation for a very long time. This treatment method is advantageous since it is inexpensive, simple to use, and effective at eliminating water contaminants. The type of coagulant has a significant impact on how well coagulation works. Wastewater treatment is essential for ensuring access to clean water and sanitation for all. By treating wastewater before its discharge into water bodies or reuse, water resources will be protected, improve water quality, and enhance sanitation achieving SDG (6).

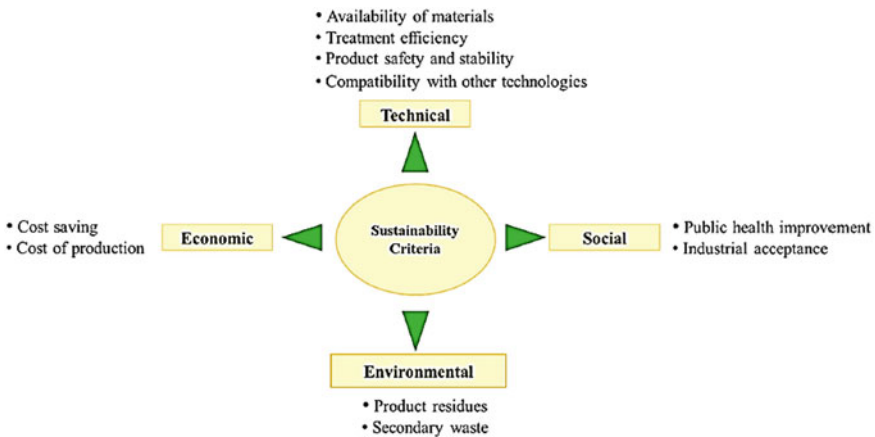


Fig. 1 Sustainability of natural coagulants

2 Printing Ink Wastewater Treatment Techniques

2.1 Coagulation

Chemical coagulation is often carried out in water treatment facilities by the addition of trivalent metallic salts such as ferric chloride (FeCl_3) or aluminum sulfate $\text{Al}_2(\text{SO}_4)_3$. Four mechanisms—ionic layer compression, adsorption and charge neutralization, sweep coagulation, and inter-particle bridging—are hypothesized to occur. Instead of utilizing a quantitative method, a jar test is used to choose the best coagulant dosages. Every water that needs to be coagulated must undergo the jar test, which must be repeated whenever a water's quality noticeably changes. The turbidity and chromaticity of ink wastewater can be decreased by coagulation. In the treatment of wastewater, coagulant selection is crucial [16]. It was found that COD treatment achieved 92.1%, decoloring rate obtained 97.4% following coagulation by polyferric chloride, taking sediment time, decoloring rate, and other aspects into consideration. Other study stated that employing ferrous sulfate as a coagulant resulted in low removal rates for both decoloring and COD, whereas polymerization aluminum chloride can produce decoloring removal rates of 99% and COD removal rates of 45–60% [17].

2.2 Adsorption

The findings showed that active carbon removes organic matters with good efficiency [18]. After adsorption, the wastewaters are transparent and colorless. Yet, active carbon is often expensive and saturated [19]. Remediate ink wastewater by first modifying zeolite with polyamidoamine (PAMAM). It was reported that fly ash and poly-dimethyl-diallylammonium chloride (PDMDAAC) were used in combination to treat ink wastewater and organic compounds with removal efficiency 94% and 74%, respectively [20].

2.3 Electrolysis Method

Iron was used as an anode and aluminum as a cathode in the electrolysis process to treat print ink wastewater. In the electrolytic process, iron gradually dissolved into Fe^{2+} , and hydrolysis produced $\text{Fe}(\text{OH})_2$, which precipitated. After being removed, COD_{cr} accomplished 47% removal, BOD_5 60% removal, and decoloring 84% following treatment [21].

2.4 Oxidation

Chemical oxidation frequently uses the oxidants NaClO , KMnO_4 , O_3 , and $\text{C}_2\text{H}_2\text{O}_4 \cdot \text{H}_2\text{O}$. Most organic materials can be reduced, but not completely, and the cost of treatment process is a substantial factor. Coagulation and Fenton mixing were employed to treat ink wastewater. Chroma and COD_{cr} can be completely removed at pH 4.5, H_2O_2 , 4.5 mg/L, FeSO_4 25 mg/L, and PAC 700 mg/L after a specified contact period. This is equivalent to 100% and 93.4% removal, respectively [22].

3 Biological Approach

Using biological methods for ink wastewater is a suitable approach considering the challenges associated with the biodegradability of ink compounds. The two-stage Sequencing Batch Reactor (SBR) is a commonly employed biological treatment process that can effectively remove organic contaminants from wastewater. The SBR process involves a sequence of fill, react, settle, and followed by decant phases within a single reactor. In the case of ink wastewater treatment, a two-stage SBR configuration is often applied to enhance the removal efficiency and achieve better treatment performance. In the pretreatment stage, physical and chemical processes are required to remove large particles, solids, and any potential inhibitors that could hinder the biological treatment. This step is required before biological treatment. The two-stage SBR method allows for a more comprehensive treatment of ink wastewater. The first stage focuses on the removal of easily biodegradable organic compounds and the initial reduction of chemical oxygen demand (COD). The wastewater is subjected to a controlled aerobic process where microorganisms metabolize the organic pollutants, breaking them down into simpler, more biodegradable forms. After the first stage, the wastewater undergoes a settling process to separate the biomass and any residual solids. The clarified wastewater is then transferred to the second stage, where more resistant and complex organic compounds are further degraded. This stage typically involves an anoxic or anaerobic process to target specific recalcitrant compounds, enhancing the overall removal efficiency. According to the findings, the COD removal rate stayed over 93% and the decoloring rate was 80% [23].

4 Ink Raw Materials

4.1 Pigments

The ink's color is a pigment's most evident function. Pigments can also be abrasive, glossy, and resistant to damage from light, heat, solvents, etc. Also, specialized pigments called extenders and opacifiers are utilized. Opacifiers are white pigments

that make the paint opaque so that the surface beneath the paint cannot be seen, whereas extenders are transparent pigments that make the colors of other pigments look less intense [24].

4.2 Resins

Resins are essentially binders; they combine the other ink components to form a coating that adheres the ink to the paper. They also support qualities like gloss and resistance. water, chemicals, and heat. There are many different types of resins utilized, and each ink often contains more than one resin. This is a list of resins that are most frequently used: Maleics, Formaldehydes, Phenolics, Acrylics, Alkyds, Cellulose derivatives, Rubber resins, and Ketones [25].

4.3 Solvents

When ink is put on a printing plate or cylinder, solvents are employed to maintain the ink's liquid state until it has been transferred to the surface that will be printed. The solvent must now separate from the ink body for the image to dry and adhere to the surface [26].

5 Chemical Coagulants and Flocculants

Chitosan, starch, cellulose, tannin, microbiological raw materials, animal glue, and gelatin are the main sources of natural flocculants. Three categories can be made out of them: Depending on the type of functional groups that make up its chemical structure, flocculants are classified as (i) cationic flocculant, (ii) anionic flocculant, and (iii) non-ionic flocculant. Such flocculants are also non-toxic and biodegradable. Natural flocculants are thought to be the greatest option for eliminating dyes and/or heavy metals from wastewater because of their many inherent qualities. Long polymer chains and a high cationic charge density are some of these characteristics [27].

5.1 Inorganic Compounds

The TOC, AOX, and COD loading values supported the removal of turbidity and color as well as a significant portion of the inorganic content by adding aluminum and ferric chlorides to printing ink wastewater. From an economic and technological standpoint,

flocculation has been proven to be an appropriate, quick, and easy treatment for such effluent [28, 29] (Tables 1, 2 and 3).

Table 1 Values of water quality parameters for W1 and W2

Wastewater	COD, mg/L	BOD, mg/L	AOX, mg Cl/L	TOC, mg/L	pH
W1	3220	0.0896	9.323	941.971	7.6
W2	2320	0.0384	6.036	893.9	7.4

Table 2 Efficiency of studied flocculants expressed by the concentration of residual ink in treated water

	Flocculant, g/L	Turbidity, NTU ^a	Residual ink concentration, mg/L	pH	Sample of filtrate
Al₂(SO₄)₃					
W1	0.375	0.6	0.076	6.6	A
W2	0.450	0.69	0.08	6.6	B
FeCl₃					
W1	0.505	0.49	0	6.6	C
W2	0.650	0.39	0	6.5	D
AlCl₃					
W1	0.375	0.59	0	6.5	E
W2	0.415	0.49	0	6.4	F

Table 3 Treated water analysis

Sample of filtrate	COD mg/l	BOD ₅ mg/l	TOC mg/l	AOX mg/l Cl ₂	pH
W1					
A	840	0	313	2.102	6.7
C	740	0	323.1	1.398	6.6
E	560	0	305.7	1.955	6.6
W2					
B	220	0	146.4	0.674	6.6
D	160	0	139.3	0.510	6.5
F	110	0	118.2	0.474	6.4

5.2 *Chitosan and Tannin*

An effluent comprising ink that was created during the manufacturing of packaging was treated utilizing a coagulation/flocculation process using a variety of biopolymers (chitosan and tannin). The effectiveness of the procedure was examined in terms of how pH, coagulant and flocculant concentrations, and chitosan properties affected it (especially the molecular weight). The procedure was especially effective with acidic solutions since restricted the pH to 5 [30].

5.3 *Fenton Oxidation*

The Fenton process treated effluent was improved by the coagulation utilizing fenu-greek and iron sulfate (FeSO_4) by reducing the flocs settling time, enhancing turbidity, and increasing COD and BOD removal. Under particular circumstances, the elimination of COD, BOD, and total turbidity was 99%, 63%, and 39.5%, respectively. As a result, this work may provide a practical method for the printing industry and manufacturers of water-based inks to treat their wastewater [31].

6 Mechanism of Coagulation by Natural Coagulants

Natural coagulants have a similar mechanism of action to polyelectrolytes and contain a variety of functional groups, including $-\text{OH}$, $-\text{COOH}$, and $-\text{NH}_2$. The red circles are the negatively charged colloidal particles, while the green circles are the positively charged coagulants. Natural coagulants can be classified according to their mechanism of action into four categories: sweep flocculation, charge neutralization, double-layer compression, and antiparticle bridging [32]. Sweep flocculation/coagulation is a method for removing colloids by trapping or entangling them in a net-like structure that contains precipitates of the amorphous metal hydroxide that are produced during the hydrolysis of the colloids. The flocs formed using this method are relatively smaller in size with adequate settling capacity, but they are distinguished by a slow rate of floc formation, according to several evaluations, including initial floc-aggregation, the relative settling factor, and the flocculation index [32]. The great fractal dimension of the floc created by sweep flocculation demonstrates the complexity of flocs [33]. The high fractal dimension has theoretically stronger flocs that can withstand breaking. Nevertheless, sweep flocculation produces big, easily broken flocs despite having a higher floc generation rate. Adsorption between the oppositely charged surfaces of the coagulants and the colloid results in charge neutralization [34]. Chemical coagulants are hydrolyzed to produce some cationic species before reacting with the colloids. For the charge neutralization procedure, an electrostatic patch mechanism, a patch-wise medium, is used. The surface of the

colloids will patch with different cationic species, resulting in particle surfaces with positive and negative charges. surfaces of colloids with a mixed charge will weaken repelling forces and strengthen van der Waals forces between the particles [35].

Double-layer compression is a technique that penetrates the double layer that surrounds the colloidal particles by using ions that have the opposite charge to that of the colloidal particles. The volume and thickness of the double layer will decrease due to the counterions. As the electrolyte is continuously compressed, electrostatic repulsion is decreased and van der Waals forces are increased, which makes it easier for the two destabilized colloids to unite.

7 Sustainability of Natural Coagulants

Sustainable development meets the demands of both present and future generations. According to the United Nations' idea of sustainability, the reliability of technology is just as important as performance efficiency when it comes to the treatment of water. As a result, the idea of sustainability combines social, environmental, and economic considerations [36] as shown in Fig. 1. Industrial acceptance and advancements in public health are two societal aspects of the sustainability of natural coagulants. The ability of natural coagulants to provide outcomes comparable to those of chemical coagulants and be employed as an alternative is a requirement for industrial acceptability. Because there aren't any regulatory or approved standards for the treatment of potable water, Businesses are reluctant to use natural coagulants. Natural coagulants may help with health and cleanliness and raise everyone's standards of living, especially in rural regions. The technological side of sustainability includes product and treatment stability, material accessibility, and compatibility with other methods. The effectiveness of several natural coagulants in the treatment of water and wastewater has been well-established over time. Natural coagulants are regarded as harmless and non-toxic due to their natural origin. Organic coagulants' environmental safety needs to be confirmed because it is uncertain whether they are dangerous to humans and the environment [37].

Hence, careful choice and dose optimization of effective natural coagulants could offer encouraging outcomes in WT and possibly serve as a replacement for pharmaceutical coagulants. As was previously said, natural coagulants are readily available, reliable, resilient, and can be derived from a variety of sources, including plants, microbes, or animals [38–40]. Yet their vulnerability to microbial or other environmental biodegradation has a negative impact on their long-term storage (shelf life) and commercialization [41, 42]. Environmental sustainability criteria call for the use of plant-based, biodegradable coagulants that are safe for the environment and can produce biodegradable sludge [43], which can be used for a variety of other things like agricultural practices, landfills, and the civil engineering sector [44, 45].

8 Conclusion

The ink industry is one of the important industries at present, and despite its importance, the industry's output of liquids represents an environmental threat that must be addressed. The resulting wastewater contains many recalcitrant chemicals and heavy metals. Many researchers have dealt with the technology of removing these pollutants. One of the promising technologies for treatment is the use of the coagulation process. In this review, the composites of ink were discussed. Also, printing ink treatment technologies were illustrated.

9 Recommendation

The coagulation/flocculation technique is commonly used in water treatment and can be effective in removing pollutants from ink-contaminated water. However, one of its significant drawbacks is that the pollutants are concentrated during the process, requiring a subsequent treatment method for their proper disposal. This emphasizes the need for a comprehensive approach that ensures the safe and complete removal of pollutants. To address this issue, researchers and scientists should focus on developing or improving post-treatment methods that can safely dispose of the concentrated pollutants. These methods could include techniques such as adsorption, advanced oxidation processes, membrane filtration, or biological treatments, depending on the specific characteristics of the concentrated pollutants and the environmental requirements. Furthermore, it's essential to study different types of dyes present in inks to understand the variations in their toxicity and identify the most appropriate treatment methods for each type. Some dyes may be more harmful to living organisms or more resistant to conventional treatment techniques than others. By evaluating the toxicity of various dyes and assessing their chemical composition, researchers can determine the most effective and environmentally friendly treatment options.

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