

# An Effective Polyamine-Based Coagulation–Flocculation Approach for Cyanide and Color Removal from Steelmaking Wastewater

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## Research Article

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# Abstract

The present work demonstrates the usability of organic polymers as coagulant/flocculation aids for successful removal of free cyanide and color from steelmaking industrial wastewater. The effluent was characterized using various analytical instruments and contained a high cyanide concentration of about 13.2 ppm and color intensity of 7350 Pt-Co due to the presence of suspended solids, iron-containing species, and other lipophilic colloids. The treatment process involved the sequential addition of 60 ppm copper sulfate catalyst, 300 ppm sodium metabisulfite, and 400 ppm ferrous sulfate to convert toxic cyanide into less toxic cyanate, followed by coagulation and flocculation. Four different polyamine coagulants with varying molecular architectures were evaluated for treatment performance. Among them, the linear polyamine coagulant PA-L exhibited superior performance compared with branched and highly branched analogues, owing to its enhanced bridging capability and improved accessibility of cationic functional groups. Using an optimum dosage of 100 ppm coagulant followed by 4 ppm anionic flocculant aid, the cyanide concentration was reduced from 13.2 ppm to 0.2 ppm and color from 7350 Pt-Co to 90 Pt-Co, corresponding to removal efficiencies of 98.5% and 98.8%, respectively. The treated effluent satisfied the prescribed discharge limits for cyanide and exhibited excellent clarification characteristics. The study further highlights the influence of polymer architecture on coagulation performance and demonstrates that polyamine-based coagulation–flocculation is a simple, scalable, and economically attractive treatment approach for steelmaking wastewater. Based on the optimized treatment conditions, a scale-up study was successfully carried out for the treatment of actual steel plant wastewater.

## 1. Introduction

Industrialization has placed at the heart for the social and economic development of countries like India and China, which consistently increases the wastewater generation at staggering proportions due to the raised production levels. The growth of steelmaking industry can be one of the major important and key to achieving sustainable development, however it consumes 25–186 m<sup>3</sup> of water per tonne of steel which has caused the exploitation of water resources to meet the water requirements if not treated properly [1–3]. The volume of waste generated by the steel industry in India is at the rate of 5 m<sup>3</sup>/h. The types of wastewater differ in accordance to their origin in the manufacturing process where water is used and discharged. The blast furnace (BF) process in steel industry typically releases wastewater containing cyanide, a cytotoxic contaminant for animals and humans [4]. The water consumption per tonne of ingot steel produced in BF is 7.6 m<sup>3</sup> of water, thereby accounting one of the highest quantity of wastewater discharge [5]. The central and the state pollution control boards of India fix the stringent standards for surface discharge of cyanide not exceeding 1.0 ppm [6]. Hence, the development of innovative technologies for removal of cyanide from wastewaters is a matter of alarming concern for steel industries to recycle the water in the process itself. This practice facilitates the use of treated water in other applications including coke manufacturing process, cooling water systems, and housekeeping purposes.

Cyanide, in steelmaking wastewater, mainly available in free cyanide form. The free cyanide (FCN) includes the most toxic forms as cyanide anion ( $\text{CN}^-$ ) and hydrogen cyanide (HCN) [6]. Diverse processes are employed for cyanide removal from industrial wastewater including alkaline chlorination [7], biological oxidation [8], electrodialysis [9, 10], wet oxidation [11], ozonation assisted electrocoagulation [12], coke breeze adsorption process [1], anodic oxidation [13], layered double hydroxide adsorption [14], hydrogen peroxide oxidation [15], chemical precipitation [16, 17], reverse osmosis [18], coagulation/precipitation [19],  $\text{SO}_2$ /air (INCO) process [15, 20], electrowinning [21], hydrolysis/distillation [22], caro's acid [23], and use of resin [24]. Although several chemical and physical processes can be made available to remove cyanide, they are rather expensive and complex to operate due to sensitive operating conditions, larger time of operation, low-removal efficiency, and generation of large amounts of secondary wastes. Electrocoagulation is an efficient alternative for removal of cyanide; however, this can be affected by disadvantage of the increasing operating cost. In this context, coagulation/flocculation by inorganic or organic polymers can be a potentially preferred alternative for the removal of cyanide from steelmaking wastewater. This process conveys the flexibility of no added hazardous chemicals and even the polymer usage can be significantly lowered. Water-based synthetic polymers are widely as coagulant and flocculant aids in the treatment of different industrial wastewaters [25–36]. This method relies upon addition of organic polymer coagulant that helps in aggregating the contaminates followed by introducing organic polymer as flocculant aid to enhance the effectiveness of cyanide removal by enlarging the size of flocs and consequently causing rapid settling [37].

Among the available treatment methods, coagulation–flocculation offers a simple and cost-effective option for industrial implementation. In addition to cyanide removal, the process can simultaneously reduce suspended solids, turbidity and color. However, limited information is available on the use of polyamine coagulants for treatment of cyanide-containing steelmaking wastewater. Therefore, the present work evaluates the performance of different polyamine architectures for cyanide and color removal from actual steel plant effluent and investigates their potential for industrial-scale implementation.

Overall, this study aims to evaluate the effectiveness of coagulation/flocculation method to remove the cyanide contaminant from industrial wastewater collected from clarifier tank of a leading steel making company in India. The commercially available synthetic polymer is used as flocculant aid with polyamine as coagulant. Effects of primary process parameters such as polymer concentration and pH of the wastewater were investigated.

## 2. Experimental methods

### 2.1. Materials

The cyanide containing effluent was obtained from a leading steel manufacturing plant in eastern India. Deionized water was obtained by passing house deionized water through a Borosil water system followed by 0.2-micron filter. The resulting deionized water has a measured resistance of 20 M $\Omega$ -cm. The

analytical grade sulfuric acid and sodium hydroxide were procured from M/s, Sigma Aldrich, USA for pH adjustment of the effluent. Three commercial polymer coagulants of polyamine series PA-L, PA-B1, PA-B2, and PA-HB generously given by SNF SAS (France) were tested, and an anionic polymer flocculant aid AN-1 from SNF was used for settling application.

## 2.2. Characterization of Effluent

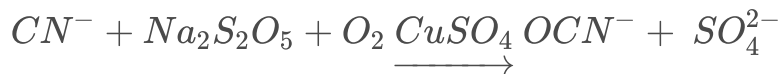
The characterization of steelmaking effluent was done in the laboratory prior to treatment methods. The effluent characteristics such as pH was measured by Mettler-Toledo pH meter (Model No: InLab Expert Pro-ISM, US.), conductivity and total dissolved solids were extracted by Mettler-Toledo conductivity meter (Model No: InLab 731-ISM, US.), color measurement was done by spectrophotometer (Model No: DR 3900, HACH, US.), turbidity was seen by nephelometer (Model No: DR 2100Q, HACH, US.), COD was captured by spectrophotometer with COD digester (Model No: DRB 200, HACH, US.), cyanide content was analyzed by a titration method using silver nitrate. The effluent characteristics are delineated in Table. 1.

| Table 1                                             |             |
|-----------------------------------------------------|-------------|
| Characteristics of steelmaking industry wastewater. |             |
| Type                                                | Quantity    |
| pH                                                  | 7.8 ± 0.3   |
| Total dissolved solids, TDS (mS/cm)                 | 7120 ± 30   |
| Color, (Pt/Co)                                      | 7350 ± 40   |
| Turbidity (NTU)                                     | 323 ± 10    |
| COD                                                 | < 90        |
| Cyanide (mg/l)                                      | 13.2 ± 0.03 |

## 2.3. Effluent Treatment

In order to find the polymers as coagulant and flocculant aids, Jar-tests were performed for the removal of cyanide and color from the effluent. The two parameters studied were pH of the effluent and dosages of the coagulant. It is observed from Table. 1 that pH of the wastewater is 7.8 and is varied up to 8.8 based on its origin in the steel plant [22, 38]. The solution pH has a major impact on cyanide specification in the wastewater. At pH above 8.6–8.7, most of the cyanide exists in free form (CN<sup>-</sup>) whereas available as a soluble HCN form at lower pH [38–40]. Thus, the pH of the steel plant effluent is in the range of 7.8–8.8, the cyanide exists in both free cyanide and HCN forms. In this study, prior to coagulation/flocculation process, sodium metabisulphite is used as a source of sulfite to convert toxic cyanide into less-toxic cyanate in the presence of air using copper sulfate as the catalyst [41]. The iron complexed cyanides in the effluent are reduced to the ferrous state and precipitated with copper as their

hydroxides using ferrous sulfate and sodium hydroxide. The anticipated pathway for cyanide to cyanate has been shown below:



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The treatment strategy adopted in this study involves a combination of oxidation, precipitation, coagulation and flocculation mechanisms. Sodium metabisulfite oxidizes free cyanide into less toxic cyanate in the presence of copper sulfate catalyst. Ferrous sulfate assists in destabilization and precipitation of iron-complexed cyanides, while the polyamine coagulant neutralizes negatively charged colloidal particles. The subsequent addition of an anionic flocculant enhances particle aggregation and settling.

### 3. Results and discussion

#### 3.1. Jar tests

An in-house square shaped jar-test apparatus by SNF SA was used for coagulation and flocculation tests as shown in Fig. 1. The apparatus contains four chambers of 500 ml capacity each, stainless steel paddles, and stirrer for maintaining uniform speed.

The steelmaking effluent was charged into the jar and the pH of the effluent was confirmed in the range of 7.8–8.8. The copper sulfate solution of required quantity was added into the jar under vigorous stirring. Then, the sodium metabisulfite of required concentration as a sulfite source was added to the above aqueous solution and mixed at a paddle speed of 300 rpm for 1 min followed by ferrous sulfate addition under agitation for 30 seconds. The pH of the solution in the jar was adjusted to 8.6–8.7 with 2M NaOH prior to coagulant addition. The polyamine series coagulant of known dosage was added and stirred for 1 min at 100 ppm followed by flocculant addition at slow mixing of 45 rpm for 2 min. This reduced speed of mixing during flocculation process is to avoid the breaking of flocs growth [42–43]. After the flocculation, the suspension was allowed to settle for 5 min. The supernatant after coagulation was decanted for residual cyanide content and color measurement. The cyanide concentration was found using silver nitrate titration for concentrations above 10 ppm, and colorimetric analysis for concentrations below 10 ppm. All the tests were repeated thrice for accuracy and reproducibility. The experiments were done at room temperature of 25–30 °C.

#### 3.2. Mechanism of Cyanide and Color Removal

The removal of cyanide and color from steelmaking wastewater in the present study occurs through a combination of oxidation, precipitation, coagulation, and flocculation mechanisms. Initially, sodium metabisulfite acts as an oxidizing agent in the presence of dissolved oxygen and copper sulfate catalyst. Under alkaline conditions (pH 8.6–8.7), free cyanide is converted into the significantly less toxic cyanate

ion. The copper sulfate accelerates the sulfite-mediated oxidation reaction, thereby improving cyanide destruction efficiency. The oxidation of cyanide can be represented by the simplified reaction as shown in Eq. (1)

After cyanide oxidation, ferrous sulfate is added to facilitate the precipitation of iron-complexed cyanides and the formation of metal hydroxide flocs. These precipitates provide additional adsorption sites for residual cyanide species and suspended impurities present in wastewater. The adjustment of pH to 8.6–8.7 further enhances hydroxide precipitation and improves subsequent coagulation performance.

Later, the polyamine coagulant may destabilize negatively charged colloidal particles through charge neutralization. The cationic nature of the polyamine promotes adsorption onto suspended solids, organic colloids, and finely dispersed color-causing species, resulting in particle aggregation. The addition of the anionic flocculant further increases floc size through polymer bridging, producing larger and denser aggregates that settle rapidly under gravity.

Therefore, the overall treatment results from the synergistic action of chemical oxidation, precipitation, coagulation, and flocculation, which collectively contribute to the substantial reduction of cyanide concentration and color in the treated effluent.

### **3.3. Effect of Coagulant Type**

The experiments were conducted with a one percent solution of coagulant, as recommended by the manufacturer. The coagulants tested are polyamine series of PA-L, PA-B1, PA-B2, and PA-HB. In addition, the 0.1% solution of anionic polymer AN-1 as flocculant aids was also studied. The photograph of the steelmaking effluent is shown in Fig. 1. The initial color and TDS of the effluent are 7350 Pt-Co and 7120 mS/cm, respectively.

The flowchart of the experimental method is shown in Fig. 2. By changing the type of the coagulant and flocculation aid, the optimum coagulant, which minimized cyanide concentration and colour, were determined. The details of the polymer coagulants and their corresponding properties are shown in Table 2.

There are three categories of polyamine series coagulants were tested based on their degree of branching whose percentage of non-volatile solids are same. The jar test results for the coagulation with a flocculation aid are presented in Fig. 3.

Table 2  
Details of the polymer coagulants used in the study.

| Polymer | Type      | Degree of branching | Molecular weight | pH  | Viscosity (cP) | Non-volatile solids (%) |
|---------|-----------|---------------------|------------------|-----|----------------|-------------------------|
| PA-L    | Coagulant | Linear              | Very high        | 6.5 | 30             | 50                      |
| PA-B1   | Coagulant | Branched            | Very high        | 5.0 | 150            | 50                      |
| PA-B2   | Coagulant | Branched            | Very high        | 5.0 | 400            | 50                      |
| PA-HB   | Coagulant | Highly branched     | Very high        | 5.0 | 850            | 50                      |

It can be observed that variation of coagulation process with type of polymer is clearly seen. The flocs sizes are good with a linear polymer coagulant. Table 3 shows the residual cyanide content, color and floc size of the supernatant. As mentioned before, the cyanide concentration was found using silver nitrate titration.

Table 3  
Details of the residual cyanide content, color and floc size of the supernatant.

| Coagulant | Flocculant | Floc size | Settling rate | Color (Pt-Co) | Residual cyanide content (ppm) |
|-----------|------------|-----------|---------------|---------------|--------------------------------|
| PA-L      | AN-1       | Good      | Good          | 90            | 0.2                            |
| PA-B1     |            | Moderate  | Good          | 241           | 1.8                            |
| PA-B2     |            | Moderate  | Moderate      | 321           | 4.3                            |
| PA-HB     |            | Moderate  | Moderate      | 104           | 3.9                            |

As can be seen from Table. 3, the cyanide removal is high for the liner polymer coagulant as compared to the branched, whereas highly branched polymer also shows better performance in yielding the better cyanide and color reduction of steel making effluent. The cyanide content in the effluent was reduced from 13.2 ppm to 0.2 ppm with a linear polymer coagulant PA-L followed by an anionic polyacrylamide flocculant aid. It also provides the best processing performance in terms of color from 7350 Pt-Co to 90 Pt-Co, settling speed and capture rate of suspended solids as shown in Table 4.

Table 4  
Overall treatment performance.

| Parameter      | Initial | Final | Removal (%) |
|----------------|---------|-------|-------------|
| Cyanide (mg/l) | 13.2    | 0.2   | 98.5        |
| Color (Pt-Co)  | 7350    | 90    | 98.8        |

The overall treatment efficiency exceeded 98% for both cyanide and color removal, demonstrating the effectiveness of the integrated treatment process. This reduction or eliminating suspended solids from

the process improves the efficiency of the purification process and reduces the risk of contamination of surface and groundwater.

### 3.4. Effect of Polymer Architecture

The experimental results clearly demonstrate that polymer architecture significantly influences cyanide and color removal efficiency. Among the polyamine coagulants tested, the linear polymer PA-L exhibited superior performance compared to branched and highly branched counterparts.

The enhanced performance of PA-L may be attributed to its extended chain configuration, which facilitates greater polymer adsorption and particle bridging. Linear polymer chains possess improved accessibility of cationic functional groups, allowing effective interaction with negatively charged colloidal particles and cyanide-containing complexes.

In contrast, highly branched polymers may suffer from steric hindrance, reducing the accessibility of charged sites and limiting the effective bridging distance between suspended particles. Although PA-HB exhibited good color removal, its cyanide reduction efficiency remained lower than that of PA-L.

The superior floc formation observed for PA-L further supports the hypothesis that polymer chain extension plays a dominant role in particle aggregation and subsequent settling. Similar observations have been reported for linear cationic polymers employed in wastewater treatment where long molecular chains yield larger and denser flocs.

### 3.5. Mechanism of Color Reduction

The significant reduction in color from 7350 Pt-Co to 90 Pt-Co indicates efficient removal of chromophoric contaminants present in the wastewater.

Color in steelmaking wastewater generally originates from suspended carbonaceous particles, iron-containing compounds, tar derivatives, and organic colloidal materials generated during coke oven operations and blast furnace processes [1]. These species are generally negatively charged and remain stable in suspension.

The positively charged polyamine coagulant neutralizes the surface charge of these particles, leading to destabilization and aggregation. The resulting aggregates are further enlarged with the aid of an anionic flocculant through bridging interactions. The removal of these suspended and colloidal contaminants leads to substantial reduction in apparent color and turbidity.

The color removal efficiency achieved in this study may be calculated as:  $\frac{7350-90}{7350} \times 100 = 98.8 \%$

The observed value demonstrates the effectiveness of polyamine-based coagulation systems for treatment of highly colored industrial effluents.

### 3.6. Comparison with Existing Cyanide Treatment Technologies

Several technologies have been reported for the treatment of cyanide-containing industrial wastewater, including alkaline chlorination, hydrogen peroxide oxidation, electrocoagulation, adsorption, membrane filtration, and biological treatment processes as shown in Table 5. Although many of these methods can achieve high cyanide removal efficiencies, their industrial application is often restricted by high operating costs, energy requirements, complex operation, or secondary waste generation.

Table 5  
Comparison of cyanide treatment technologies.

| Technology            | Advantages                                               | Limitations              |
|-----------------------|----------------------------------------------------------|--------------------------|
| Alkaline chlorination | Rapid cyanide oxidation                                  | Chlorinated by-products  |
| Electrocoagulation    | High removal efficiency                                  | High power cost          |
| Biological treatment  | Environmentally friendly                                 | Long retention time      |
| Reverse osmosis       | High water quality                                       | Membrane fouling         |
| Present study         | Simple, scalable, simultaneous cyanide and color removal | Requires chemical dosing |

In the present study, the combination of sodium metabisulfite oxidation, metal-assisted precipitation, and polymer-based coagulation–flocculation achieved a cyanide reduction from 13.2 mg/L to 0.2 mg/L, corresponding to a removal efficiency of approximately 98.5%. Simultaneously, a color reduction of approximately 98.8% was achieved. The process requires relatively short treatment time and employs commercially available treatment chemicals. Therefore, the proposed treatment methodology represents a practical and economically attractive alternative for steelmaking wastewater treatment.

### 3.7. Scale-up study

The scale up study for the effluent treatment of coke oven plant predicts the below stream of operation in several chambers to reduce cyanide and color as per the permissible limits before discharge into common effluent treatment plant (CETP). In chamber 1, the copper sulphate catalyst and sodium metabisulfite (SMBS) are added to the effluent to convert cyanide into cyanate. Then, the pH of the effluent will be adjusted to 8.6–8.7 followed by a ferrous sulphate addition and PA-L coagulant addition respectively in chamber 2. In chamber 3, tilted plate interceptors (TPI) can be used for water clarification with aid of anionic polymer flocculant addition. The treated supernatant can be collected in chamber 4 for discharge or further treatment in CETP.

The amount of coagulant and flocculant required is more for high flow rate of effluent and high cyanide concentration. For example, at steel making effluent flow rate of 20 kl/h with ~ 13 mg/l of initial cyanide

concentration, 48 kg of coagulant and 1.92 kg of flocculant is required per day of treatment cycle.

The proposed treatment process offers several advantages for industrial implementation. Unlike advanced treatment technologies such as electrocoagulation, reverse osmosis, and ozonation, the present approach utilizes commercially available chemicals and conventional wastewater treatment equipment. Therefore, the process can be readily integrated into existing steel plant effluent treatment facilities without significant capital investment.

The treatment sequence involves oxidation, coagulation, flocculation, and clarification under near-neutral to mildly alkaline conditions, thereby minimizing operational complexity. The low polymer consumption together with rapid settling characteristics of the generated flocs indicates favourable operating economics. Furthermore, the treated effluent meets the discharge requirement for cyanide and can be further processed in a common effluent treatment plant or reused for non-contact industrial applications such as cooling water systems, dust suppression, and housekeeping operations. The simplicity and scalability of the process make it a practical solution for treatment of cyanide-containing wastewater generated in steel manufacturing operations.

Compared with advanced technologies such as electrocoagulation, reverse osmosis and ozonation, the present treatment methodology can be readily integrated into existing wastewater treatment facilities without significant capital investment. The process operates under near-neutral pH conditions and employs commercially available chemicals and conventional clarification equipment, making it attractive for large-scale operation.

## 4. Conclusions

A practical oxidation–coagulation–flocculation process was developed for the treatment of cyanide-containing steelmaking wastewater. Sodium metabisulfite oxidation catalyzed by copper sulfate converted cyanide into less toxic cyanate, while ferrous sulfate facilitated precipitation of metal-cyanide complexes. Among the four polyamine coagulants investigated, the linear polyamine PA-L demonstrated superior treatment performance due to its enhanced bridging capability and accessibility of cationic functional groups.

Using an optimum treatment sequence consisting of 60 ppm copper sulfate, 300 ppm sodium metabisulfite, 400 ppm ferrous sulfate, 100 ppm PA-L coagulant, and 4 ppm anionic flocculant, the cyanide concentration was reduced from 13.2 ppm to 0.2 ppm, corresponding to a removal efficiency of 98.5%. Simultaneously, color was reduced from 7350 Pt-Co to 90 Pt-Co, corresponding to a removal efficiency of 98.8%.

The process utilizes conventional treatment equipment, requires relatively low polymer dosages, and can be readily scaled for industrial wastewater treatment applications. These findings demonstrate that polyamine-based coagulation–flocculation represents an effective and economically attractive approach for treatment and potential reuse of steelmaking wastewater.

The simultaneous removal of cyanide (98.5%) and color (98.8%) demonstrates the potential of linear polyamine-based coagulation–flocculation as a practical treatment strategy for steelmaking wastewater. The results further highlight the importance of polymer architecture in determining contaminant removal efficiency and flocculation performance.

## **Declarations**

## **AI-Assisted Writing Disclosure**

The authors used Microsoft Copilot (GPT-5–based AI assistant) as an aid for language editing, manuscript organization, scientific writing improvement, manuscript title and abstract text analysis and development of supporting discussion sections. The research, experiments, data generation, interpretation, conclusions, and scale-up calculations are entirely our own work.

## **Data Availability Statement**

Some or all data that support the findings of this study are available from the corresponding author upon reasonable request. The data include the experimental measurements and analytical results generated during the coagulation–flocculation experiments.

## **Funding Declaration**

The authors received no financial support from any funding agency in the public, commercial, or not-for-profit sectors for the research, authorship, and/or publication of this article.

## **Ethics, Consent to Participate, and Consent to Publish**

Ethics, Consent to Participate, and Consent to Publish declarations: not applicable.

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## Figures



**Figure 1**

The photograph of the raw effluent whose color and TDS are 7350 Pt-Co and 7120 mS/cm, respectively.

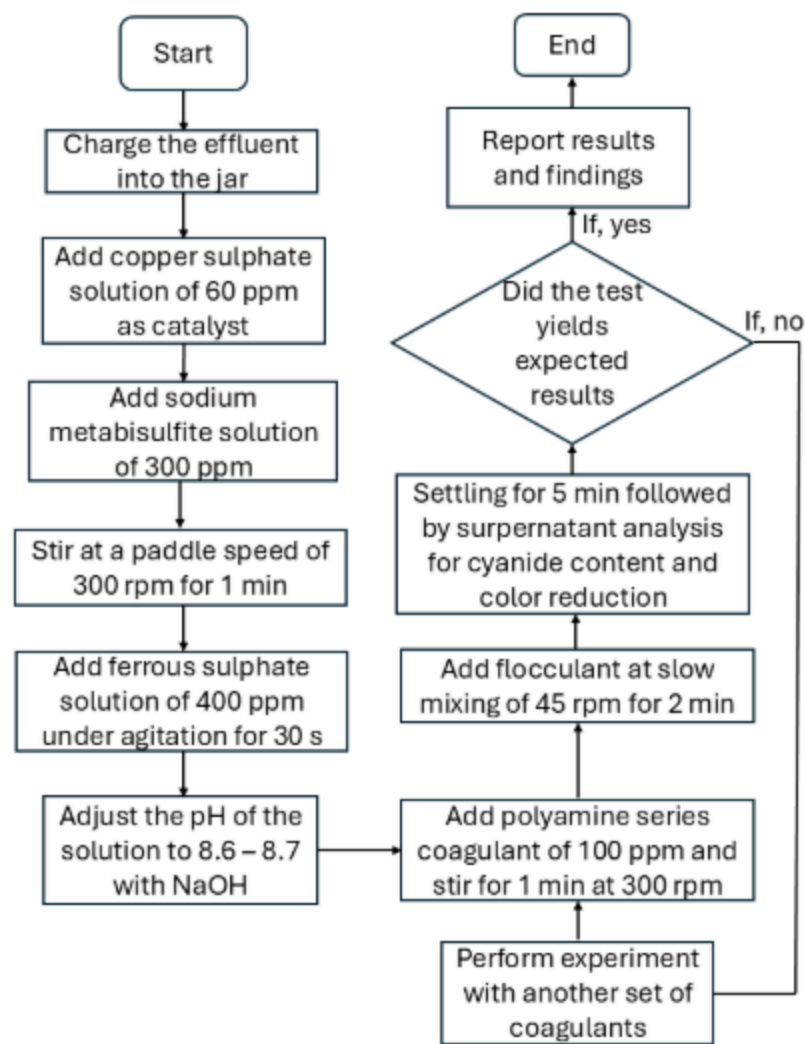


**Figure 2**

The flowchart of the experimental method.

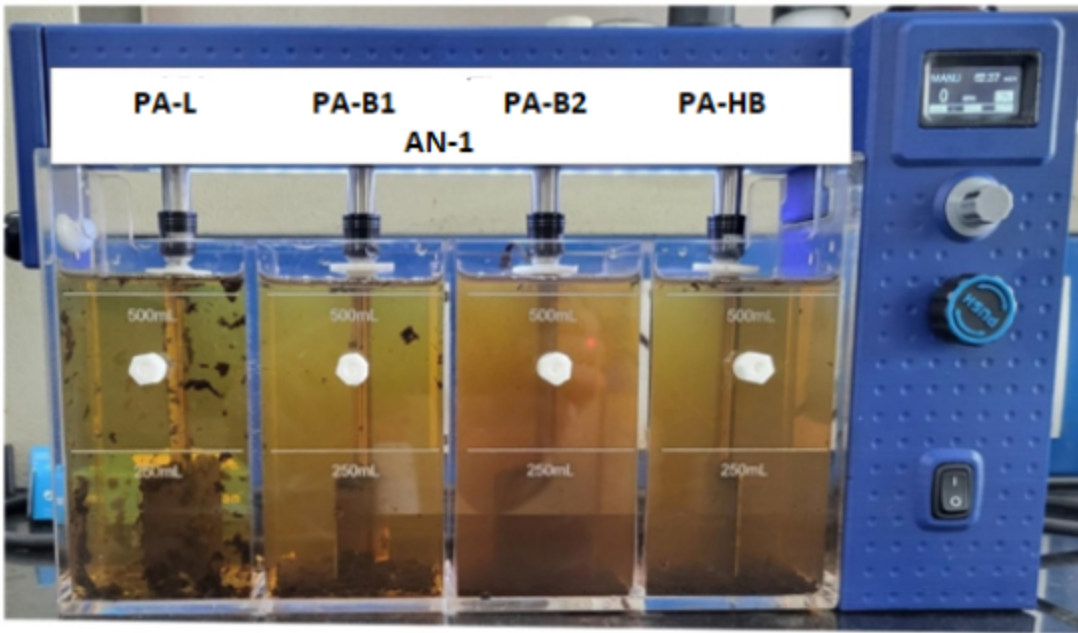


Figure 3

Jar test results of coagulation/flocculation process for cyanide and color removal.