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# In-Situ Coagulant Generation in Electrocoagulation for Textile Dye Effluent Treatment: Comparative Performance of Fe–Fe, Al–Al, and Cu–Cu Electrode Systems

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oxidation–coagulation synergy

**ABSTRACT:**

Electrocoagulation (EC) has emerged as an effective and sustainable treatment technology for textile dye effluents due to its ability to generate coagulants *in situ* through anodic dissolution. The nature of the generated coagulant species, their hydrolysis products, and associated electrochemical intermediates strongly govern pollutant removal efficiency. This review critically evaluates the role of *in situ* coagulant generation in EC using iron (Fe-Fe), aluminum (Al-Al), and copper (Cu–Cu) electrode systems for the treatment of textile dye wastewater. Emphasis is placed on understanding how electrode-dependent coagulant chemistry and redox behavior influence dye removal mechanisms, energy consumption, and sludge characteristics. Evidence from high-impact literature demonstrates that Fe-Fe systems consistently outperform Al-Al and Cu-Cu configurations due to their dual coagulation–oxidation functionality, broader pH tolerance, and enhanced removal of recalcitrant dye molecules. The review highlights mechanistic insights into the formation of iron hydroxides and reactive iron intermediates, discusses comparative treatment efficiencies, and identifies future research directions for advancing electrocoagulation-based textile wastewater treatment.

## 1. Introduction

The textile industry is a major contributor to industrial water pollution, primarily due to the discharge of dye-laden effluents generated during dyeing and finishing operations (Can et al. 2006; Tezcan 2013; Kobya, Gengec, and Demirbas 2016; Syam Babu et al. 2020). Synthetic dyes, particularly azo and xanthene dyes, are designed to resist fading, rendering them persistent in aquatic environments (P. V. Nidheesh, Gandhimathi, and Ramesh 2013; A. 2015). Even at low concentrations, dyes impart intense coloration, inhibit light penetration, disrupt photosynthesis, and pose toxic and mutagenic risks to aquatic organisms (Sharpless, Page, and Linden 2003; Üstün, Solmaz, and Birgül 2007). Recent studies revealed that dye pollutants often remain as microplastics in water bodies. Conventional biological treatment processes often exhibit limited efficiency due to the complex aromatic structure and low biodegradability of textile dyes (Olayinka and Alo 2004; Villegas-Navarro et al. 2001). (Advances in Unit Operations and Processes for Wastewater Treatment: A Comprehensive Review) https://doi.org/10.33472/AFJBS.6.Si3.2024.2608-2613.

Physicochemical methods such as chemical coagulation, adsorption, and advanced oxidation processes (AOPs) have been widely investigated; however, these methods often involve high chemical consumption, secondary sludge generation, and operational complexity. Electrocoagulation (EC) has gained prominence as a promising alternative owing to its operational simplicity(Bayramoglu, Eyvaz, and Kobya 2007), reduced chemical requirement, and capacity for *in-situ* coagulant generation(Syam Babu et al. 2020). In EC, sacrificial electrodes dissolve under an applied electric field, producing metal ions that hydrolyze to form metal hydroxides capable of destabilizing and removing contaminants (Chanikya et al. 2020; P. V. Nidheesh and Gökkuş 2023; Davuluri et al. 2024).

Among various electrode materials, iron (Fe), aluminum (Al), and copper (Cu) have been most frequently studied.

While Al and Cu primarily function as sources of coagulant species, iron electrodes uniquely combine coagulation with redox-driven transformation pathways. This review synthesizes recent advances in EC for textile dye effluent treatment, focusing on how electrode material and generated coagulant chemistry govern treatment efficacy.

## 2. Fundamentals of In-Situ Coagulant Generation in Electrocoagulation

Electrocoagulation involves the application of a direct current between sacrificial electrodes immersed in wastewater. At the anode, metal dissolution releases cationic species ($M^{n+}$), while cathodic reactions generate hydroxide ions and hydrogen gas. The metal ions hydrolyze to form amorphous metal hydroxides, which act as coagulants.

For Fe–Fe systems:

- Anodic reactions generate $Fe^{2+}$ and $Fe^{3+}$.
- Hydrolysis leads to $Fe(OH)_2$ and $Fe(OH)_3$ flocs with high surface area.
- Redox cycling between $Fe^{2+}/Fe^{3+}$ introduces oxidative pathways under appropriate conditions.

For Al–Al systems:

- $Al^{3+}$ ions form $Al(OH)_3$ and polymeric Al-hydroxo complexes.
- Pollutant removal is dominated by adsorption and sweep flocculation.

For Cu–Cu systems:

- $Cu^{2+}$ species can form hydroxides and surface complexes.
- Coagulation is possible, but redox participation and stability are limited.

The efficiency of EC is therefore dictated not only by coagulant quantity but also by the physicochemical properties and reactivity of the generated species.

## 3. Impact of Iron-Based In-Situ Coagulants (Fe–Fe Systems)

### 3.1 Coagulation and Oxidation Synergy

Iron-based EC systems demonstrate superior dye removal performance due to the coexistence of coagulation and oxidative mechanisms. Iron hydroxide flocs effectively adsorb dye molecules through electrostatic interactions, while dissolved iron species participate in redox reactions that promote partial or complete dye degradation.

Under aerated or mildly oxidative conditions, ferrous ions may be oxidized to higher-valent iron species, enhancing transformation of chromophoric groups such as azo (–N=N–) linkages. This dual functionality results in higher color and COD removal compared with purely coagulative systems.

### 3.2 Treatment Performance

Multiple studies report color removal efficiencies exceeding 90–95% for azo and xanthene dyes using Fe–Fe electrodes, with relatively low specific energy consumption. Iron hydroxide flocs exhibit favorable settling behavior, reducing sludge handling challenges.

### 3.3 Sludge and Operational Considerations

Iron-based sludge is generally denser and easier to dewater than aluminum-based sludge. Moreover, Fe–Fe systems operate efficiently across a wider pH range, reducing the need for extensive pH adjustment.

### 4.1 Aluminum-Based In-Situ Coagulants (Al–Al Systems)

Aluminum electrodes are widely used due to their strong coagulation capability and formation of polymeric aluminum hydroxides. These species effectively remove dyes via adsorption and sweep flocculation. However, Al–Al systems often require higher current densities to achieve comparable removal efficiencies, resulting in increased energy consumption.

Additionally, the absence of intrinsic redox activity limits the degradation of dye molecules, often leaving structurally intact compounds within the sludge matrix rather than transforming them.

### 4.2 Copper-Based In-Situ Coagulants (Cu–Cu Systems)

Copper electrodes have been explored to a lesser extent for textile dye treatment. While $Cu^{2+}$ species can facilitate coagulation, concerns regarding copper leaching, secondary toxicity, and limited oxidation capability restrict their applicability. Existing studies suggest that Cu–Cu systems may be suitable for niche applications but are less competitive than Fe–Fe and Al–Al configurations for dye-rich effluents.

### 4.3 Comparative Evaluation of Electrode Systems

A systematic comparison reveals that:

- **Fe–Fe electrodes** provide the highest overall treatment efficiency due to combined coagulation and oxidation mechanisms.
- **Al–Al electrodes** rely solely on physical coagulation, leading to lower degradation of complex dyes.
- **Cu–Cu electrodes** show inconsistent performance and limited applicability in textile dye systems.

The superior performance of Fe–Fe systems is attributed to the nature and reactivity of the *in situ* generated iron coagulants and intermediates.

## 4. Influence of Generated Intermediates on Treatment Efficacy

The formation of reactive intermediates during iron EC significantly enhances treatment performance. Dissolved iron species can interact with dissolved oxygen or electrochemically generated oxidants, facilitating partial oxidation of dye molecules before or during coagulation. This synergistic interaction improves decolorization efficiency and reduces residual COD.

## 5. Proposed Mechanistic Schematics

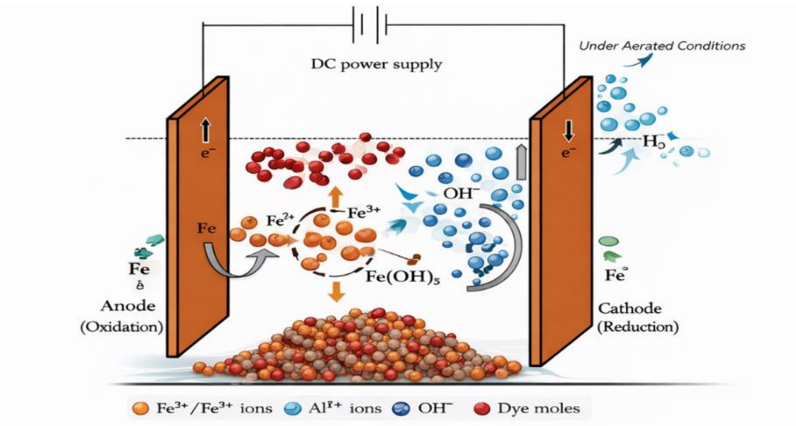

Figure 1. Mechanism of In-Situ Coagulant Generation in Fe–Fe Electrocoagulation

A schematic illustrating iron anodic dissolution ($Fe^{2+}/Fe^{3+}$ release), hydroxide floc formation, interaction with dye molecules, and simultaneous oxidative transformation pathways under aerated conditions.

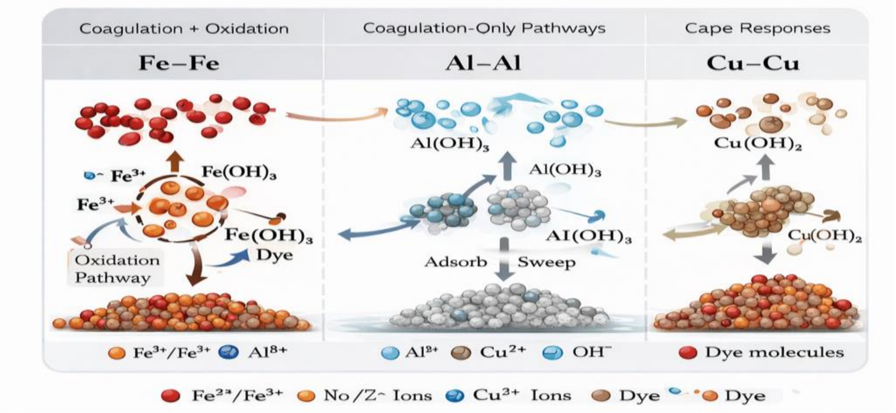

Figure 2. Comparative Pathways in Fe–Fe, Al–Al, and Cu–Cu Systems

A conceptual diagram comparing coagulation-only pathways (Al–Al, Cu–Cu) with combined coagulation–oxidation pathways in Fe–Fe systems, highlighting differences in pollutant fate and removal efficiency.

## 6. Conclusion

This review critically evaluates the role of in-situ generated coagulants during electrocoagulation (EC) for the treatment of textile dye effluents, with particular emphasis on the comparative performance of Fe–Fe, Al–Al, and Cu–Cu electrode configurations. Based on a comprehensive assessment of Q1 literature, it is evident that iron-based electrocoagulation (Fe–Fe) consistently outperforms aluminum and copper systems in terms of dye removal efficiency, reaction versatility, and mechanistic robustness.

The superior performance of Fe–Fe EC is primarily attributed to its dual-functional removal pathways. Unlike Al–Al and Cu–Cu systems, which are largely restricted to physical mechanisms such as charge neutralization, adsorption, and sweep flocculation, Fe–Fe EC uniquely enables simultaneous coagulation and oxidative degradation. The electrolytic dissolution of iron produces $Fe^{2+}/Fe^{3+}$ species that hydrolyze to form highly reactive amorphous iron hydroxides, providing extensive surface area for dye adsorption. More importantly, under aerated or oxygen-rich conditions, iron undergoes redox cycling to generate high-valent iron intermediates, particularly ferryl species (Fe(IV)=O), which introduce an additional advanced oxidation pathway capable of attacking chromophoric structures.

Accumulating evidence demonstrates that ferryl species exhibit strong electrophilic character and oxygen-atom transfer ability, making them particularly effective toward electron-rich azo (–N=N–) linkages and conjugated aromatic systems commonly present in textile dyes. This oxidative pathway is absent in Al–Al EC and is only marginally operative in Cu–Cu systems, where concerns related to copper toxicity and limited redox activity further constrain applicability. Consequently, Fe–Fe EC achieves not only higher color removal but also enhanced mineralization, reduced residual toxicity, and improved effluent biodegradability.

From a practical standpoint, Fe–Fe electrocoagulation offers additional advantages, including lower electrode cost, environmental compatibility of iron sludge, operational flexibility across a broad pH range, and adaptability to real textile wastewater matrices. While challenges such as sludge management and process optimization remain, these limitations are common to all EC systems and do not outweigh the fundamental mechanistic benefits associated with iron electrodes.

Overall, this review establishes Fe–Fe electrocoagulation as a chemically superior and mechanistically enriched platform for textile dye effluent treatment, bridging conventional coagulation with iron-based advanced oxidation chemistry. Among commonly studied systems, **Fe–Fe electrodes consistently offer superior removal efficiency, lower energy demand, and enhanced degradation of dye molecules** due to the unique chemistry of in situ-generated iron coagulants and intermediates. These advantages establish iron-based EC as a leading technology for sustainable textile wastewater treatment. Future research should focus on selective probing of ferryl species, reactor-scale validation, and integration with hybrid treatment systems, thereby unlocking the full potential of Fe-based EC as a sustainable and next-generation solution for complex industrial wastewater remediation.

### Research Gaps and Future Perspectives

Future research should focus on:

- Pilot-scale validation of Fe–Fe EC systems.
- Quantitative assessment of oxidative intermediates and their contribution.
- Integration of EC with complementary AOPs.
- Life-cycle and sustainability analyses.

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