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# Reaction Mechanisms and Research Advances of Iron-Carbon Micro-Electrolysis Systems in Industrial Wastewater Treatment

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## Abstract

**Iron-carbon micro-electrolysis (ICME) technology has been widely applied in the treatment of coking wastewater, pharmaceutical effluents, and electroplating wastewater due to its simplicity of operation, cost-effectiveness, and broad applicability. Rooted in metal corrosion electrochemistry, this technology leverages a micro-galvanic cell system formed by iron (Fe) and carbon (C), which drives synergistic pollutant removal through multiple mechanisms including redox reactions, adsorption, and co-precipitation. System performance is jointly regulated by factors such as solution pH, Fe/C ratio, aeration intensity, and reaction time. Furthermore, ICME is frequently integrated with advanced processes like Fenton oxidation and biological treatment to enhance efficiency. Nevertheless, practical applications face challenges including filler compaction, iron sludge disposal, and strong pH dependency. Future research should focus on optimizing electrode stability and environmental adaptability through strategies such as electrode modification and process parameter refinement. Addressing these limitations will advance the technology's scalability and reliability, ultimately promoting its large-scale implementation in industrial wastewater remediation.**

## Keywords

**Water treatment; Iron-carbon micro-electrolysis; Industrial wastewater; Reaction mechanisms.**

## 1. Introduction

With the continuous advancement of industrialization, the variety and discharge volume of industrial wastewater have increased rapidly. Wastewater from chemical, pharmaceutical, and textile industries contains persistent accumulations of refractory organic compounds, heavy metals, and high-salinity substances. These complex components exhibit strong biological toxicity, poor biodegradability, and have exacerbated global water contamination. Consequently, there is an urgent need to develop wastewater treatment technologies with broad applicability and cost-effectiveness. Iron-carbon micro-electrolysis, based on the electrochemical principles of metal corrosion, utilizes the galvanic cell system formed by Fe and C to generate active substances such as [H] and •OH without the need for an external power source. The mechanism achieves pollutant removal through redox reactions, adsorption, and co-precipitation effects. Iron-carbon micro-electrolysis technology has garnered significant attention owing to its operational simplicity, low operating costs, and broad applicability. This technique has found extensive applications in systems characterized by high pollutant concentrations and complex compositions, including textile dyeing wastewater[1], pharmaceutical effluent[2], and pesticide-containing wastewater[3]. Recent research advancements have progressively elucidated the reaction mechanisms and influencing factors of iron-carbon micro-electrolysis (ICME). Numerous scholars have explored its synergistic integration with complementary technologies to enhance contaminant removal efficiency.

However, persistent challenges such as filler compaction, excessive sludge production, and operational instability continue to hinder its industrial scalability. This paper systematically reviews the reaction mechanisms, critical operational parameters, and application progress of ICME in industrial wastewater treatment. It further compares optimization strategies across diverse industrial scenarios, aiming to provide theoretical insights and engineering references for the treatment of high-toxicity, refractory wastewater.

## 2. Reaction Mechanisms of ICME

To optimize the application of ICME, a thorough understanding of its reaction mechanisms is essential. The process relies on the inherent electrode potential difference between iron and carbon to establish numerous microscopic galvanic cells within the system, leveraging their electrochemical properties for pollutant degradation. This core mechanism synergistically combines with auxiliary processes including adsorption, flocculation, and redox reactions, collectively enabling effective elimination of contaminants from aqueous environments.

### 2.1. Galvanic Cell Reaction

The spontaneous formation of galvanic cells in conductive wastewater is driven by the inherent electrode potential difference between iron (Fe) and carbon (C). Within this ICME, electrons autonomously migrate from the Fe anode to the C cathode under the intrinsic potential gradient, eliminating the need for external power input. Here, iron serves as the anode material, undergoing oxidative corrosion to release electrons, while carbon acts as the cathode to facilitate electron acceptance and subsequent redox reactions. The primary redox reactions under varying operational conditions are expressed as follows[4,5]:

Anodic:

$$Fe - 2\mathrm{e}^{-} \rightarrow \mathrm{Fe}^{2+} \quad \mathrm{E}^{0}(\mathrm{Fe}^{2+}/\mathrm{Fe}) = -0.44\mathrm{V} \tag{1}$$

Cathodic:

(acidic condition, without O2)

$$2\mathrm{H}^{+} + 2\mathrm{e}^{-} \rightarrow 2[\mathrm{H}] \rightarrow \mathrm{H}_2 \qquad \mathrm{E}^{0}(\mathrm{H}_2/\mathrm{H}) = 0\mathrm{V} \tag{2}$$

(acidic condition, in the presence of O2)

$$\mathrm{O}_2 + 4\mathrm{H}^{+} + 4\mathrm{e}^{-} \rightarrow 2\mathrm{O}\cdot + 4[\mathrm{H}] \rightarrow 2\mathrm{H}_2\mathrm{O} \qquad \mathrm{E}^{0}(\mathrm{O}/\mathrm{H}_2\mathrm{O}) = 1.23\mathrm{V} \tag{3}$$

(alkaline or neutral condition, in the presence of O2)

$$\mathrm{O}_2 + 2\mathrm{H}_2\mathrm{O} + 4\mathrm{e}^{-} \rightarrow 4\mathrm{OH}^{-} \qquad \mathrm{E}^{0}(\mathrm{O}_2/\mathrm{OH}^{-}) = 0.40\mathrm{V} \tag{4}$$

This demonstrates that iron-carbon micro-electrolysis exhibits elevated reactivity under acidic and aerobic conditions, with the cathode potential reaching 1.23V. This elevated electrochemical activity stems from the synergistic coupling of microscopic galvanic units that collectively form an integrated ICME system. The system generates highly reactive intermediates, including atomic hydrogen ([H]) and ferrous ions ($Fe^{2+}$), which actively participate in redox reactions with diverse pollutants [6].

## 2.2. Reduction

The $H_2O_2$ and [H] generated during the ICME process exhibit strong reducing properties, effectively transforming contaminants in wastewater through redox reactions. Chen et al. achieved efficient removal of $Cr(pic)_3$ by introducing $H_2O_2$ into the ICME system and elucidated the associated redox pathways[7]. The study confirmed that the [H] generated on the carbon cathode surface synergistically interacts with $Fe^{2+}$ via electron transfer to suppress Cr (VI) formation, enabling its targeted reduction to Cr (III). The resulting Cr(III) is stabilized through the formation of inner-sphere complexes with iron oxyhydroxides, completely eliminating the risk of secondary Cr(VI) accumulation. The synergistic effect of $Fe^{2+}$ and [H] not only improves the removal efficiency of phosphorus but also effectively cleaves the carbon chain structure of ciprofloxacin (CIP), significantly enhancing the degradation effect of CIP[8]. Moreover, the presence of [H] allows nitrate to be rapidly reduced over a wider pH range, showing great potential for nitrate removal[9].

## 2.3. Flocculation Sedimentation

The $Fe^{2+}$ and $Fe^{3+}$ generated during the reaction undergo hydrolysis to form $Fe(OH)_2$ and $Fe(OH)_3$ with adsorption and complexation capabilities. These compounds facilitate organic pollutant removal through floc formation via charge neutralization, adsorption bridging, and sweep coagulation mechanisms, demonstrating effective coprecipitation. Ren et al. demonstrated the critical role of coprecipitation in persistent pollutant removal[10]. Research indicates significant precipitation effects on both hydrophobic phenanthrene and hydrophilic 2,4-dichlorophenol contaminants. XRD and SEM characterizations revealed that post-reaction iron surfaces were coated with layered FeOOH and $Fe_3O_4$-$Fe_2O_3$ mixed oxides, whose porous structures enhanced pollutant adsorption capacity. The coprecipitation efficiency was found to be influenced by Fe/C ratios. Additionally, $Fe(OH)_2$ and $Fe(OH)_3$ colloids play vital roles in removing surfactants[11] and carboxyl-functionalized organic pollutants[12].

## 2.4. Adsorption

ICME fillers exhibit elevated specific surface area, with their abundant surface porosity effectively adsorbing aqueous contaminants, particularly hydrophobic pollutants[13]。Yang et al. investigated the degradation efficiency of organic contaminants in Fe-C micro-electrolysis systems under neutral pH conditions[14]. Their study confirmed that activated carbon with high specific surface area effectively adsorbs Sunset Yellow, while the Fe/C ratio critically influences the adsorption process. The research further revealed that linear alkanes (e.g., ethyl acetate) exhibit higher adsorption contribution rates compared to aromatic compounds due to their hydrophobic characteristics. Deng et al. explored the phosphorus adsorption properties of iron-rich substrates (IRS) based on ICME[15]. The research demonstrated that activated carbon serving as cathode simultaneously facilitates electron transfer and enhances contaminant enrichment on its surface. Physical adsorption mechanisms of activated carbon significantly improved dissolved phosphorus removal. Model fitting analysis identified maximum adsorption capacity at Fe/C ratio of 3:2, optimizing the system's phosphorus sequestration performance.

# 3. Key Influencing Factors of ICME

## 3.1. pH

pH is widely recognized as one of the most critical factors influencing the treatment efficacy of ICME. As indicated by Equations [1-4], the cathode exhibits higher electrochemical potential in acidic environments, which significantly enhances micro-electrolysis reactivity. However, excessively low pH levels (e.g., <3.0) induce overproduction of $Fe^{2+}$ ions, subsequently increasing sludge generation and complicating subsequent treatment processes. Yan et al.[16] demonstrated through pretreatment of organic peroxide wastewater that maximum chemical oxygen demand (COD) removal efficiency (34.5%) occurs at an initial pH of 3.0. Xu et al.[17] systematically investigated pH-dependent mechanisms, revealing that pH=4.0 achieves optimal balance with 31.8% COD removal and 458.5 mg/L $Fe^{2+}$ yield. Notably, neutral conditions (pH≈7.0) reduce COD removal to 8.8% due to diminished potential differences, while extreme acidity (pH=2.0) causes $Fe^{2+}$ accumulation (558.4 mg/L) but triggers anode polarization that inhibits pollutant degradation. Zhang et al. [18] systematically investigated the performance and mechanism of the persulfate-activated iron-carbon micro - electrolysis (PS - ICME) system for the pretreatment of landfill leachate under near - neutral pH conditions. Through optimization using the response surface method (RSM), the experiments revealed that when the initial pH was 7, the system achieved the highest COD removal rate of 62.91%, which was significantly better than the removal rates of 58.52% under strongly acidic conditions (pH = 3) and 59.67% under alkaline conditions (pH = 9). The research indicated that the neutral environment optimized the COD removal effect through the following two synergistic mechanisms: (1) $Fe^{2+}$ generated by micro - electrolysis activated persulfate to produce $SO_4{\bullet}^-$ and •OH radicals; (2) $Fe^{3+}$ hydrolyzed to form $Fe(OH)_3$ flocs, which removed humic - like macromolecular organic compounds through adsorption and co - precipitation. Generally, the initial pH of iron - carbon micro - electrolysis ranges from 2 to 7. Within this range, a balance can be achieved between the galvanic cell reaction and the formation of iron sludge, enabling the degradation of organic matter in water while maintaining a certain co - precipitation effect.

## 3.2. Fe/C Ratio

The Fe/C ratio primarily governs the ICME reaction by modulating the quantity of microscopic galvanic cells within the system. Deng et al.[15] demonstrated through controlled adjustment of volumetric ratios between iron filings ($Fe^0$) and activated carbon (AC) (2:3, 3:2, 4:1) that a Fe/C ratio of 3:2 achieved maximum phosphorus adsorption capacity. Mechanistic analyses revealed that the Fe/C ratio influences adsorption processes through dual pathways: (1) Elevated iron content enhances micro-galvanic reactions, where continuous $Fe^{2+}/Fe^{3+}$ release from the anode improves phosphorus removal via chemical precipitation, and (2) Increased AC proportion amplifies specific surface area for physical adsorption of dissolved phosphorus. However, excessive iron filings reduce specific surface area and porosity, thereby compromising AC's adsorption efficacy. Notably, the Fe/C=3:2 system exhibited optimal pH buffering capacity and strong adaptability to organic matter (OM) accumulation. Kong et al. [19] synthesized nano iron-carbon composites (CB-nZVI) through the liquid phase mixing-high temperature reduction carbothermal method. At a molar Fe/C ratio of 1:3.4, the composite achieved optimal performance, reducing residual As(V) concentration to 0.12 mg/L through a two-stage treatment process. Song et al.[20] further identified significant regulatory effects of Fe/C mass ratios on total nitrogen (TN) removal. TN removal efficiency increased from 16.8% to 38.9% as the nZVI/AC mass ratio rose from 1:2 to 2:1, but declined to 30% at 5:1 due to AC's adsorption capacity saturation. Nano zero-valent iron (nZVI) functions as a micro-anode, continuously releasing electrons to directly reduce nitrate into $NH_4^+$ and $N_2$, while AC serves as cathode to concentrate nitrate through adsorption and catalyze intermediate conversion to $N_2$. Nevertheless, excessively high Fe/C ratios diminish AC's adsorption site density and nitrate enrichment capability.

## 3.3. Aeration Control

As indicated by Equations [2,3], the iron-carbon galvanic cell exhibits higher cathode potential and intensified reaction kinetics under aerobic conditions. Practical experimental optimization reveals that enhanced aeration achieves dual benefits: it elevates dissolved oxygen concentration while simultaneously promoting hydraulic mixing, thereby improving contact efficiency between iron-carbon fillers and wastewater. Xu et al. [17] demonstrated that increased aeration facilitates $H_2O_2$ generation through enhanced cathodic reactions and intensifies redox processes at the Fe-C interface, elevating COD removal efficiency from ~15% (non-aerated) to 31.8%. Notably, COD removal efficiency plateaued when the air-to-water ratio exceeded 10:1. Complementary research by Ying et al. [21] on landfill leachate treatment revealed a significant COD removal improvement from 56.5% to 73.7% as airflow increased from 20 to 80 $L \cdot h^{-1}$, confirming the critical role of aeration in oxidation enhancement. However, excessive aeration (air-to-water ratio >15:1) may induce filler surface passivation and reduce hydraulic retention time, ultimately diminishing reaction efficiency through physical barrier formation and insufficient reactant contact.

### 3.4. Reaction Time

Reaction duration constitutes a critical operational parameter in iron-carbon micro-electrolysis systems, requiring precise optimization to balance organic pollutant degradation efficiency against practical implementation costs. Industrial applications prioritize identifying the temporal threshold that maximizes contaminant removal while preventing excessive energy expenditure and infrastructure wear from prolonged operation. Notably, process kinetics exhibit significant variability across different target pollutants and aqueous matrices. Luo et al. [22] achieved 95.71% COD removal within 90 minutes when employing an iron-carbon micro-electrolysis coupled persulfate-based advanced oxidation (ICEPS) system for fracturing flowback fluid (FPW) treatment. In refractory wastewater treatment scenarios, however, extended reaction durations upwards of 10 hours prove necessary to attain comparable remediation efficiency. This temporal disparity highlights the imperative for matrix-specific parameter optimization, particularly when addressing recalcitrant organic compounds or complex wastewater compositions

## 4. Application of ICME in Industrial Wastewater Treatment

### 4.1. Coking Wastewater

Coking wastewater contains high concentrations of ammonia, phenol, cyanide, thiocyanate, aromatic hydrocarbons, and various nitrogen-, oxygen-, and sulfur-containing heterocyclic compounds[23]. With its complex composition and diverse pollutants, it poses significant environmental and human health risks. Most wastewater treatment plants currently employ a combination of physicochemical pretreatment, biological treatment, and advanced treatment for coking wastewater[24,25]. Iron-carbon micro-electrolysis (ICME) is generally applied during the pretreatment stage. Lv et al. [26] established a micro-electrolysis system using waste iron scrap and activated carbon. Under optimized conditions—initial pH 3.0, iron dosage 73.5 g/L, reaction duration 70 min, Fe/C mass ratio 1:1.3, and coagulation pH 9.0—the system achieved removal efficiencies exceeding 50% for nitrite nitrogen ($NO_2^-$-N) and 45% for total nitrogen (TN). Xie et al. [27] developed an electric assisted micro-electrolysis filter (E-ME). Under operational conditions of 1 V applied voltage, initial pH 7.0, aeration rate 0.75 L/min, and 5-hour reaction time, the system achieved a COD removal rate of 70%, significantly surpassing the efficiencies of separate electrolysis (SE, 31%) and micro-electrolysis (41%). Zhu et al. [28] investigated the enhanced degradation of trichloroethylene (TCE) by integrating iron-carbon micro-electrolysis with a bioelectrochemical system (BES). At a Fe/C mass ratio of 1:2, dosage of 70 g/L, cathode potential of -0.3 V, and initial TCE concentration of 100 mg/L, the Fe/C+BES system achieved 97.8% TCE degradation within 120 hours, far exceeding the performance of individual processes. Experimental results demonstrated that the iron-carbon micro-electrolysis accelerated microbial dechlorination by providing electron donors and reducing system resistance, while the BES-generated electric field further intensified the micro-electrolysis reactions. Currently, the application of iron-carbon micro-electrolysis in coking wastewater treatment remains limited and non-mainstream, primarily employed in combination with other technologies for refractory organic contaminant removal.

## 4.2. Textile Dyeing Wastewater

With the progressive development of the printing and dyeing industry, wastewater discharge from dyeing processes continues to increase. The wastewater contains chemicals such as dyes, surfactants, dispersants, and emulsifiers, resulting in high concentrations of toxic, hazardous, and refractory organic compounds[29]. In practical treatment of dyeing wastewater, iron-carbon micro-electrolysis is typically employed as a pretreatment to reduce COD. Yang et al.[30] investigated ICME coupled with $H_2O_2$ for degrading Direct Blue 15 dye. Under conditions of an initial dye concentration of 300 mg/L, Fe/C mass ratio of 2:1, initial pH 3.0, and three-stage $H_2O_2$ addition (total dosage 2 mL/L), the system achieved 98% decolorization and 40% total organic carbon (TOC) removal within 60 minutes. The study further elucidated the mineralization pathways of azo bonds (–N=N–). Wang et al.[31] applied ICME-$H_2O_2$ pretreatment to real textile dyeing wastewater. At optimized conditions ($H_2O_2$ concentration 8.88 g/L, initial pH 1.5, Fe/C dosage 837 g/L, reaction time 186 min), COD removal reached 77.65%. UV-vis spectroscopy revealed a significant blue shift in the absorption peak at 220 nm, indicating effective destruction of polycyclic aromatic hydrocarbon structures. Three-dimensional fluorescence spectroscopy identified three dominant organic components: refractory humic acids (81.76% degradation), soluble microbial metabolites (53.78% degradation), and aromatic proteins (70.83% degradation). This process enhanced wastewater biodegradability by degrading macromolecular organics into smaller molecules, creating favorable conditions for subsequent biological treatment.

## 4.3. Pharmaceutical Effluents

Pharmaceutical wastewater primarily originates from pharmaceutical production facilities and medical institutions, typically exhibiting complex compositions containing refractory toxic substances such as pharmaceutical residues, antibiotics, and metabolites. It is characterized by significant water quality fluctuations, broad pH ranges, high COD levels, low BOD/COD ratios, poor biodegradability, and inhibitory components that suppress microbial activity[32]. Wang et al.[33] implemented ICME coupled with coagulation for pharmaceutical wastewater pretreatment. Results demonstrated COD removal of 66.9% and $S^{2-}$ elimination of 98.9% under optimal conditions. The B/C ratio improved from 0.16±0.02 to 0.41±0.02, indicating enhanced biodegradability. Subsequent treatment combining activated sludge, hydrolytic acidification, sequencing batch reactor (SBR), and activated carbon adsorption achieved 96% total COD removal, with effluent COD 300 mg/L. In another study, Wang et al.[34] developed an integrated process of Fe/C micro-electrolysis-anaerobic hydrolyze-microalgae integrated process for high-concentration pharmaceutical wastewater treatment. At pH 4.0 and Fe/C ratio 3:1, the system attained removal efficiencies of 55% COD, 36% $NH_3$-N, and 63% TP, while enhancing B/C ratio from 0.12 to 0.35. Liu et al.[35] designed a hybrid ICME-expanded granular sludge blanket (EGSB)-microalgae reactor (MR) system for mixed pharmaceutical wastewater. Optimized parameters (Fe/C ratio 2:1, ICME dosage 200 g/L, aeration rate 2 L/min) enabled 58.4% COD and 82.8% TSS removal in the micro-electrolysis unit, with chromaticity reduced from 720 C.U. to 133 C.U. Final effluent COD remained below 200 mg/L (99.2% total removal) after downstream processing. In practical applications, iron-carbon micro-electrolysis primarily serves as a pretreatment to degrade organics, TN, and TP in pharmaceutical wastewater, typically integrated with other technologies.

### 4.4. Electroplating Wastewater

Electroplating involves depositing one metal onto another via electrochemical deposition. The process includes alkaline cleaning, electroplating, acid pickling, and rinsing. The wastewater typically contains refractory organic pollutants, heavy metal ions (e.g., arsenic [As], cobalt [Co], copper [Cu], chromium [Cr]), cyanides, sulfates, and high concentrations of nitrogen/phosphorus contaminants. It exhibits low biodegradability and significant treatment challenges due to its complex composition[36]. Gao et al.[37] treated electroplating wastewater using ICME combined with an anaerobic-anoxic-oxic (A2/O) process. The study first acclimatized microbial communities using domestic wastewater, then gradually increased the electroplating wastewater inflow ratio. As the inflow ratio rose, COD, $NH_4^+$-N, TN, and TP removal efficiencies declined. At a 50% electroplating wastewater inflow ratio, removal rates decreased to 73.6% COD, 55% $NH_4^+$-N, 55% TN, and 71.9% TP, meeting discharge standards. Li et al.[38] developed an Electrocatalytic internal micro-electrolysis (ECIME) fluidized bed system integrated with an external electric field (EEF) and iron-carbon particles, significantly enhancing Cu(II) removal efficiency. Under optimal conditions (polarization voltage 12 V, initial pH 4.0), the ECIME system reduced Cu(II) concentration from 100 mg/L to 0.25 mg/L within 60 minutes, outperforming single-process treatments. This approach offers a novel strategy for efficient heavy metal removal and resource recovery. Wang et al. [39] designed an interior microelectrolysis-Fenton-recycle ferrite (IM-Fenton-RF) process for treating copper-chelated electroplating wastewater. IM-Fenton pretreatment (Fe/C mass ratio 2:1, Fe/C-to-water volume ratio 5:6, initial pH 2.5, 60 min reaction) achieved 84.65% copper removal. Subsequent RF treatment (reflux ratio 0.37, pH 10.18, $Fe^{2+}$ concentration 9.20 g/L) further reduced effluent copper to 0.104 mg/L. Ferrite sludge demonstrated 3.86× faster settling velocity and 11-fold volume reduction compared to conventional metal hydroxide sludge. Magnetic separation enabled sludge reuse as micro-electrolysis filler, achieving resource circularity.

## 5. Discussion and Prospects

ICME demonstrates promising application potential due to its operational simplicity, no external power requirement, low running costs, and broad practicality. However, the following challenges remain for practical industrial implementation:

(1) pH sensitivity: Optimal performance occurs under acidic conditions ($pH < 4$), while efficiency markedly declines in neutral/alkaline environments. Frequent pH adjustments increase reagent costs.

(2) Iron sludge generation: $Fe(OH)_2$/$Fe(OH)_3$ flocs generated during ICME enhance pollutant coagulation but produce substantial iron sludge, necessitating costly disposal.

(3) Filler passivation: Prolonged operation leads to filler compaction and surface passivation, requiring periodic acid-wash reactivation. Continuous aeration further elevates energy consumption.

(4) Limited degradability: ICME alone exhibits insufficient removal efficiency for refractory organics, often requiring integration with complementary processes.

Future research should focus on addressing these limitations through:

(1) Develop nano iron-carbon composites or inert metal-doped fillers to enhance reactivity and stability.

(2) Mechanistic studies: Quantify contributions of redox reactions, co-precipitation, and adsorption to pollutant removal. Clarify degradation pathways for specific contaminants.

(3) Hybrid process development: Integrate ICME with Fenton, A2/O bioreactors, electric fields, or ozone ($O_3$) for synergistic effects.

(4) Smart control systems: Implement IoT sensors and machine learning for real-time pH/aeration control to reduce operational costs.

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