Characterization of hydrodynamics and electrochemical treatment of dye wastewater in two types of tubular electrochemical reactors

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Abstract. Electrochemical treatment is an environmentally friendly method of removing pollutants from industrial wastewater, and tubular electrochemical reactor is one kind of electrochemical reactor. However, the flow field on the electrode surface in a traditional concentric tubular reactor is not homogeneous, leading to low treatment efficiency of the reactors. Therefore, a new tubular electrochemical reactor based on spiral flow caused by a turbulence promoter is presented. In this work, fluid flow and hydrodynamics of the new tubular electrochemical reactor using computational fluid dynamics (CFD) method are studied by comparing them to the traditional one. The electro-oxidation of methylene blue simulation wastewater treatment was developed to analyze the processing efficiency of the two types of electrochemical reactors. In the new tubular electrochemical reactor, due to the presence of spiral turbulence promoter, the mean flow rate and turbulent intensity were clearly increased by 500-700% and nearly 200% compared to that of the traditional one, respectively. Under the same inlet-velocity, the removal rates of wastewater in the new tubular electrochemical reactor were also improved.

1. Introduction

Recently electrochemical oxidation treatment is of great interest in the industrial wastewater treatment process [1]. Various types of electrochemical reactors are employed in electrochemical reactor systems. Among them, tubular electrochemical reactor which consists of a rod type of anodes and tube type of cathodes are most popular[2], and has a wide range of applications in energy, metallurgy, environmental protection and other fields, for its various advantages such as improvement in the pollutant removal rate and lower energy consumption[3].

However, the distribution of flow field is not homogeneous in the reactor, and the mass transfer is relatively weak [4]. In order to solve these problems, we proposed a new tubular electrochemical reactor with a spiral turbulence promoter perpendicular to the axial velocity of the fluid.

Hydrodynamics of reactors is one of the most important factors determining the treatment efficiency. Computational fluid dynamics (CFD) is an effective approach to simulating the flow field and optimizing the configuration of reactors because of its advantages such as reduced time and costs compared to the traditional research methods [5].

In this work, we analyze velocity distribution and turbulence intensity of the new tubular electrochemical reactor with a spiral turbulence promoter by CFD method and compare the results with that of the traditional one. And then we investigate the removal rates of methylene blue simulation wastewater in two types of electrochemical reactors by electro-oxidation.
2. Materials and methods

2.1. Geometric models
The spiral-flow tubular electrochemical reactor with a spiral turbulence promoter is shown in Figure 1B. A Ti/TiO$_2$ tube (outer diameter = 2 cm) was used as the anode material and placed at the center of the reactor, and the cathode (outer diameter = 7 cm) was constructed from stainless steel. Besides, a spiral turbulence promoter 3D-printed by Acrylonitrile Butadiene Styrene was placed between anode and cathode. The traditional tubular electrochemical reactor had the same design except the turbulence-promoting mixer, as Figure 1A shown.

In this research, the total geometric area of anodes in these two types of reactors was 188.5 cm$^2$, and had a net working volume of 950 cm$^3$.

2.2. Numerical simulations in CFD
Both of the reactor models were created through AutoCAD software and then meshed by The Integrated Computer Engineering and Manufacturing code (ICEM) with approximately 10 million cells. The computation was carried out with three dimensional single precision and the maximum residual tolerance was $10^{-4}$ for the continuity equation, momentum equations and energy conservation equation. Realizable κ-ε model was chosen as the computing model. A type velocity of inlet boundary condition was 1.1 cm/s. The type outflow boundary condition was assumed for the outlet. The type solid and type wall boundary condition was assumed for the electrodes.

2.3. Experimental Setup and Procedures
In this work, methylene blue simulation wastewater was selected as a treatment object to assess the performance of both reactors. The electrolysis system consisted of a reaction cell, a peristaltic pump, a feed tank and a power supply. According to previous works, the initial concentration of methylene blue was 50 mg/L, concentration of the supporting electrolyte Na$_2$SO$_4$ was 0.5 mol/L. For comparing the electro-oxidation results of the new tubular reactor with that of the traditional one, the runs were carried out in the range 30-150 min residence time with an interval of 60 min, at current density of 8 mA/cm$^2$. 10 mL liquid samples were withdrawn at given time intervals from outlet and then were immediately analyzed. The concentration of methylene blue and TOC were measured by UV-Visible Spectrophotometer and Non Dispersive Infrared Absorption Method, respectively.

Figure 1. Schematic diagram of traditional (A) and new concentric (B) tubular electrochemical reactors.
3. Results and discussion

3.1. CFD simulation
Mass transfer is a key factor influencing the efficiency of a reactor and is controlled by the distribution of flow fields [6]. Therefore, the velocity fields and turbulence intensity distribution of the new tubular electrochemical reactor were analyzed and compared with the traditional concentric tubular electrochemical reactor in this section.

3.1.1 Velocity fields distribution. In the simulation of velocity fields, the working volume was 950 cm$^3$, the residence time was 30 min and the cross-sectional area of inlet was about 0.48 cm$^2$, so that the inlet velocity was 1.1 cm/s. The velocity field contour of the new and traditional concentric electrochemical reactors are depicted in Figure 2. As described in the Figure 2, there was great difference between the flow field of new tubular electrochemical reactor and that of the traditional one, which was mainly caused by the spiral turbulence-promoting mixer. In the traditional concentric tubular electrochemical reactor, the velocity was relatively slow (<0.03 cm/s). However, in the new tubular electrochemical reactor, the reaction area was divided into a narrow, spiral channel because of the mixer. The flow velocity distribution was quite uniform after the first spiral channel, and the maximum velocity was nearly 0.15 cm/s, about 5 to 7 times that of the traditional concentric reactor.

![Velocity magnitude (cm/s) distribution](image)

**Figure 2.** Velocity magnitude (cm/s) distribution in the traditional (left) and new (right) tubular electrochemical reactors.

3.1.2 Turbulence intensity. Turbulence intensity was chosen as the indicator of turbulence strength and the turbulent intensity of both reactors was shown in Fig.3. For traditional tubular electrochemical reactor, the turbulent intensity was relatively low and distributed quite evenly in the whole reactor, which mainly because of wide flow channel without any mixer in the reactor. While for the new tubular electrochemical reactor, the flow channel was modified by a spiral turbulence-promoting...
mixer. As the fluid passed through the channel, the flow velocity was increased and so did the turbulent intensity.

As shown in Fig.3, the mixing was more intense in the new reactor than in the traditional one, which could accelerate mass transfer between anode surface and solution. Specifically, the turbulent intensity distribution around anode in the spiral-flow tubular electrochemical reactor was about 0.0009, 2 times more than that in the traditional tubular reactor.

![Turbulence intensity distribution](image)

**Figure 3.** Turbulence intensity distribution in traditional (left) and new (right) tubular electrochemical reactors.

### 3.2. Electro-oxidation of methylene blue in two reactors

As described in Figure 4, compared with the traditional concentric tubular electrochemical reactor, the new reactor with a spiral turbulence promoter can enhance the electro-oxidation of methylene blue simulation wastewater. For residence time of 30 min, almost 97% decolorization occurred in the new tubular electrochemical reactor within 20 min, whereas only 83% degradation of methylene blue was observed in the traditional one. In addition, with the spiral turbulence-promoting mixer, the removal of TOC can also been strengthened. For residence time of 90 min, the removal efficiency of TOC was increased by 71% in the new reactor compared to the traditional one.
Figure 4. The electro-oxidation efficiency in two tubular reactors (initial MB: 50 mg/L; current density: 8 mA/cm²; electrolyte Na₂SO₄: 0.5 mol/L)

4. Conclusion
The spiral turbulence promoter clearly enhanced the flow velocity, turbulence intensity and improved the removal rate of pollutants in the new tubular electrochemical reactor. By CFD simulation, it was possible to obtain the velocity distribution and turbulence intensity distribution within the reactor, and the new reactor performed better than the traditional one. In addition, higher removal rates of methylene blue and TOC were observed in the new tubular electrochemical reactor compared with the traditional concentric tubular electrochemical reactor.

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