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# Study on the characteristics of metal corrosion and sterilizing effect of chlorine dioxide

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**ABSTRACT**

The sterilizing effect of heterotrophic bacteria, sulfur bacteria and iron bacteria treated with chlorine dioxide is examined experimentally in this paper. The weight loss method was used to simultaneously study the comparative corrosion behavior of four different metals under treatments of chlorine dioxide, liquid chlorine and their mixture. The experimental results indicated that chlorine dioxide had a good sterilization effect against heterotrophic, sulfur and iron bacteria at different dosages, pH, temperatures and reaction conditions. Chlorine dioxide is used as a high-efficiency circulating water oxidizing sterilizer; its sterilizing effect on heterotrophic, sulfur and iron bacteria exceeds 90% at 0.6 mg/L dosage, pH 7, 20°C and 15 min reaction. Considering its strong oxidizing effect and chemical corrosion of metal pipes, the corrosion behavior of $ClO_2$, liquid chlorine and their mixture to carbon steel, aluminum, red copper and stainless steel were investigated. It was concluded that $ClO_2$, liquid chlorine and their mixture could all cause metal corrosion: liquid chlorine caused the greatest metal corrosion and $ClO_2$ gave the weakest metal corrosion; moreover, corrosion inhibition was observed under low concentrations of $ClO_2$. Therefore, the corrosion behavior of $ClO_2$ needs to be considered when it used as a sterilant for circulating cooling water.

*Keywords:* Chlorine dioxide; Liquid chlorine; Mixture of chlorine dioxide and liquid chlorine; Sterilization rate; Metal corrosion

## 1. Introduction

In industry, one of the major processes that consumes large volumes of water is the circulation of cooling water. In petrochemical, electric, iron and steel, metallurgy and other industries, the amount of circulating cooling water accounts for 60%–90% of total water consumption [1–3]. However, the long-term operation of industrial equipment leads to problems such as scaling, corrosion, microbial growth and other issues, which cannot be ignored [4–6]. Since the early 1930s, when corrosion and scale inhibitors were used in circulating cooling water, the development of nitrogen-based corrosion and scale inhibitors was relatively mature. However, these inhibitors led to the intensification of biological corrosion since the introduction of nitrogen and carbon species creates an environment that is conducive to microbial growth [7–9]. Since this time, non-oxidizing bactericides have been extensively used for sterilizing circulating cooling water; however, bioaccumulation in circulating cooling water persists as microbial tolerance increases [10–12]. Therefore, numerous studies began to explore the use of oxidation-type bactericides (chlorine bactericides) to inhibit biological growth and corrosion in circulating cooling water. Deyab [13] found that sulfate-reducing bacteria were the main cause of microbiological corrosion of carbon steel in petroleum field, and that quaternary ammonium salts (e.g., DDAC) could restrict the corrosion rate of carbon steel. Wu et al. [14] found that chlorine dioxide sterilization effectively restrained biochemical reactions in the circulating cooling water system and alleviated the serious corrosion problems of medium

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carbon steel and cement components in a company. Wen et al. [15] evaluated the effectiveness of chlorine dioxide at inactivation of three dominant genera of fungal spores isolated from drinking groundwater, looking at the effects of pH, temperature, chlorine dioxide concentration and humic acid. The results showed that chlorine dioxide was remarkably effective at inactivation of fungal spores.

In this study, a static experiment was conducted to study the bactericidal effect of $ClO_2$ on heterotrophic bacteria, sulfur bacteria and iron bacteria. The effect of chlorine bactericides on metal corrosion arising from biological activity in circulating cooling water was also investigated: $ClO_2$, $Cl_2$ and their mixture were chosen as different chlorine bactericides to study the corrosion of different metal materials (carbon steel, aluminum, stainless steel and copper).

## 2. Materials and methods

### *2.1. Weight-loss method*

The corrosive effect of $ClO_2$, $Cl_2$ and their mixtures on different metals was compared using the weight-loss method test. The method involved suspending the metal specimen in aqueous solution. According to the principle of metal corrosion, metals corrode through the process of positive polarization. Therefore, the result of metal corrosion is weight loss; this is, of course, once the corrosion products that form on the metal surfaces are cleaned off, otherwise weight will appear to have been gained instead. Using the weight of metal lost per unit area and unit time as a measure of metal corrosion rate in the weight loss method, the corrosion rate ($V$, mm/a) can be calculated according to Eq. (1).

$$V = K \times \frac{\left(W_1 - W_2\right)}{\left(F \times t \times \gamma\right)} \quad (1)$$

where $W_1$ – specimen weight before corrosion (mg); $W_2$ – specimen weight after being corroded and corrosion product removed from the specimen surface (mg); $F$ – surface area of the specimen exposed to cooling water (cm$^2$); $K$ – constant (87.6); $t$ – specimen immersion corrosion time (h); $\gamma$ – metal density (g/cm$^3$).

In this experiment, four kinds of metal materials were used with the following densities: carbon steel, 7.84 g/cm$^3$; copper, 8.80 g/cm$^3$; stainless steel, 7.92 g/cm$^3$ and aluminum, 2.73 g/cm$^3$. An adapted weight-loss method was used to measure the corrosion rate. Measured corrosion data should have a uniform corrosion rate according to the "Code for design of industrial recirculating cooling water treatment" (GB50050-17) [16], which states that, generally, the corrosion rate of carbon steel should be controlled at <0.075 mm/a, and the corrosion rate of copper alloy and stainless steel at <0.005 mm/a.

To eliminate the influence of the boundary layer effect and reduce the amount of exposed terminal crystal, and thereby reduce experimental error, it was required that the coupon unit weight should have a large surface area and a small proportion of edges, which required thin test pieces of 1–3 mm average thickness. The two types of dimensions of the standard corrosion test pieces recommended by the Ministry of Chemical Industry (HG5-1526-83) (length × width × thickness) include:

Type I: 50.0 ± 0.1 × 25.0 ± 0.1 × 2.0 ± 0.1 mm

Type II: 72.4 ± 0.1 × 11.5 ± 0.1 × 2.0 ± 0.1 mm

All surface finishes were Δ7, Φ4.0 ± 0.1 mm hanging hole and smooth finish Δ4. Because of predetermined processing tolerances, the coupon area was a fixed value and an integer, facilitating calculation of the corrosion rate. Type I was used in the experiment and the area was 28.00 cm$^2$.

Surface pretreatment: The specimen was cleaned by placing in anhydrous ethanol and skimming using cotton wool (not less than 50 mL anhydrous ethanol reagent for every 10 test pieces). The specimen was blotted with filter paper to dry, placed in a desiccator for more than 4 h, weighed, then placed into the drier prior to use.

### *2.2. Determination of $ClO_2$ concentration*

$ClO_2$ was prepared by the hydrochloric acid method [17]. In this experiment, the concentration of chlorine dioxide was determined by the continuous iodometric method.

### *2.3. Calculation of sterilization rate*

Calculation of the sterilization rate was according to Eq. (2) as follows:

$$\eta = \frac{N_t - N_0}{N_0} \times 100 \quad (2)$$

where η – sterilization rate (%); $N_0$ – number of bacterial colonies in the original water before adding the fungicide (cfu/mL); $N_t$ – number of bacterial colonies after bactericidal treatment (cfu/mL).

## 3. Experimental results and discussion

### *3.1. Static sterilization experiments of $ClO_2$ on heterotrophic, sulfur and iron bacteria*

To provide a valuable $ClO_2$ dosage concentration, pH, reaction temperature and reaction time for a production plant, the bactericidal effect on heterotrophic, sulfur and iron bacteria was determined taking these four aspects into account. Circulating cooling pipe water from Harbin Gasification Plant located in Harbin, Heilongjiang Province was used.

#### *3.1.1. Effect of $ClO_2$ concentration on sterilization rate*

The bactericidal effect on heterotrophic, sulfur and iron bacteria was determined at different $ClO_2$ concentrations (0.2–1.0 mg/L). The pH was maintained at 7.0, temperature at 20.0°C and the reaction time was 15 min. The results are shown in Fig. 1.

In Fig. 1, the sterilizing effects of $ClO_2$ on heterotrophic, sulfur and iron bacteria were positively related to dosage. The sterilization rates of $ClO_2$ on all three bacteria exceeded 90% when the dosage was 0.6 mg/L, and was highest for heterotrophic bacteria (>99%). $ClO_2$ also showed a strong sterilizing effect on iron bacteria, with the sterilization rate reaching 99% when the dosage was 0.8 mg/L. In contrast, $ClO_2$ showed the poorest sterilization effect on sulfur bacteria, with the

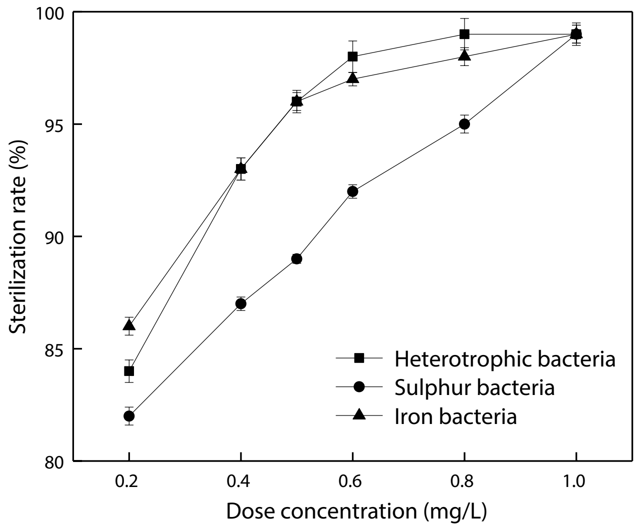


Fig. 1. Influence of $ClO_2$ concentration on bactericidal effect.

sterilization rate only reaching 99% when the dosage was increased to 1 mg/L. Comparison of the sterilization rates on the three bacteria at low dosages showed that when the dosage was 0.2 mg/L, the sterilization rate of $ClO_2$ on all three bacteria exceeded 80%, which indicated the good sterilizing effect of $ClO_2$ at low concentrations. In this experiment, the optimal dosage of $ClO_2$ was chosen as 0.6 mg/L in order to achieve a satisfactory sterilizing effect.

### 3.1.2. Effect of pH on sterilization rate of $ClO_2$

At a constant $ClO_2$ concentration of 0.6 mg/L, a temperature of 20.0°C and a reaction time of 15 min, the effect of $ClO_2$ on the three types of bacteria at pH 6.0, 7.0, 8.0, 9.0 and 10.0 was determined. The results are shown in Fig. 2.

It can be seen from Fig. 2 that under slightly acidic to slightly alkaline conditions, the sterilization rates of $ClO_2$ to heterotrophic, sulfur and iron bacteria increased with increasing pH. However, the sterilization rates decreased slightly with increasing pH at higher alkalinity. The sterilization rates of $ClO_2$ to all three bacteria were higher than 85% in the 6–10 pH range, and even exceeded 93% when the pH ranged from 7 to 9. This was because $ClO_2$ exists as free molecules in water, neither reacting chemically nor forming dimer or polymer structures. Instead, the molecules could readily penetrate the microbial cytomembrane to react with amino acids in proteins and thereby inhibit microorganism anabolism [18]. As $ClO_2$ achieved a satisfactory sterilizing effect in the 6–10 pH range, pH 7 was chosen as optimal in this experiment.

### 3.1.3. Effect of reaction temperature on sterilization rate of $ClO_2$

The effect of temperature on the bactericidal effect of $ClO_2$ on heterotrophic, sulfur and iron bacteria was studied further with a $ClO_2$ concentration of 0.6 mg/L, pH of 7.0 and a reaction time of 15 min. The results are shown in Fig. 3.

Fig. 3 shows that the sterilizing effects of $ClO_2$ on heterotrophic, sulfur and iron bacteria were influenced slightly by temperature. In general, high temperature was conducive to increased sterilizing effect. The sterilization rate of $ClO_2$ on heterotrophic and iron bacteria reached about 95% at 20°C, which approached 100% at 50°C. This could be due to heat facilitating the dispersion of $ClO_2$ molecules into microbial cells, thus strengthening the sterilizing effect. The sterilizing effect of $ClO_2$ on heterotrophic and iron bacteria was greater than on sulfur bacteria at the same temperature.

### 3.1.4. Effect of reaction time on sterilization rate of $ClO_2$

Finally, the sterilization rate of $ClO_2$ on the three groups of bacteria at varying reaction times of 3, 15, 30, 60 and 120 min and a fixed pH of 7.0, $ClO_2$ concentration of 0.6 mg/L and temperature of 20°C was determined. The results are shown in Fig. 4.

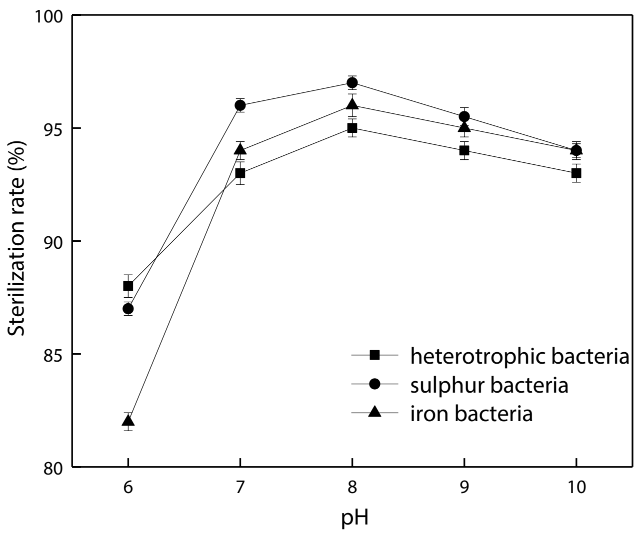


Fig. 2. Effect of pH on the bactericidal rate of $ClO_2$.

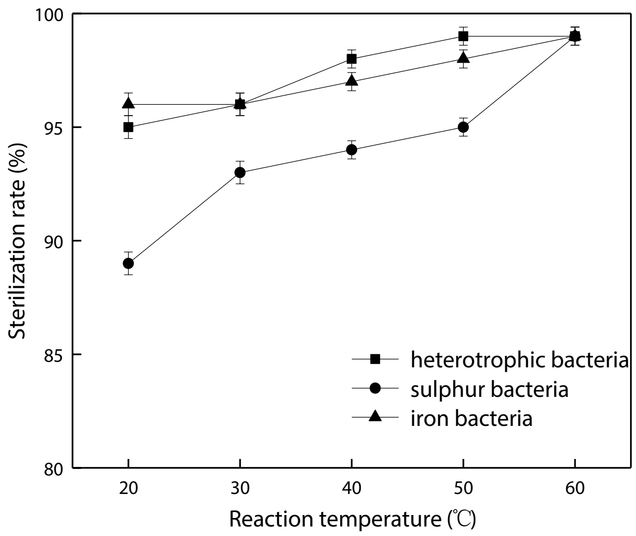


Fig. 3. Effect of reaction temperature on sterilization rate.

In Fig. 4, the sterilization rate of $ClO_2$ on heterotrophic, sulfur and iron bacteria all increased as reaction time increased. The sterilization rate of $ClO_2$ on heterotrophic and iron bacteria exceeded 90% after 15 min. However, it took longer to inactivate sulfur bacteria, with the sterilization rate exceeding 90% at 30 min. Sterilization rates of $ClO_2$ on all three bacteria were basically stable (>95%) after 60 min. This indicated that $ClO_2$ could kill bacteria very quickly; again, likely because $ClO_2$ was neither hydrolyzed nor polymerized in water, but could easily disperse into microbial cells to inactivate bacteria.

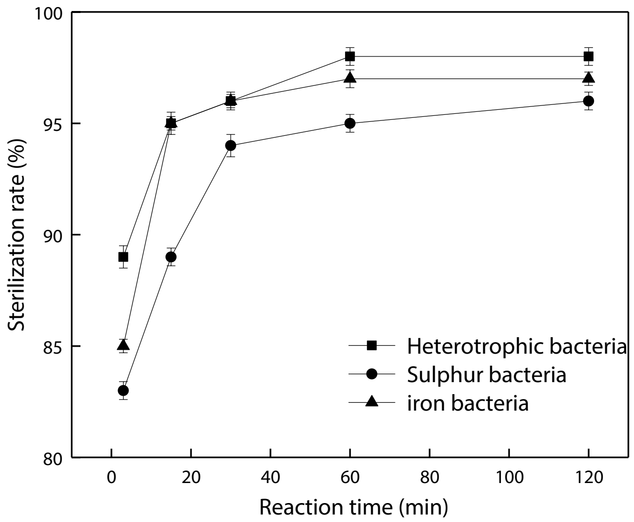


Fig. 4. Effect of reaction time on sterilization rate of $ClO_2$.

### 3.2. Experiment on corrosion properties of different metal materials by chlorination solution

From the above study results, it was deduced that $ClO_2$ has an efficient bactericidal effect; however, due to its strong oxidizing effect, it is corrosive to metals in recirculation cooling water. In this experiment, corrosion effect of chlorine disinfection liquids (i.e., $ClO_2$, liquid chlorine and $ClO_2$–liquid chlorine mixture) on carbon steel, aluminum, stainless steel and red copper were investigated by the weight loss method.

#### 3.2.1. Corrosion rates of materials at different $ClO_2$ concentrations

Each of the four specimens, carbon steel, aluminum, stainless steel and copper were weighed in 0.2, 0.3, 0.5, 0.8 and 1.0 mg/L $ClO_2$ solutions using the method described in Section 2.1. The area of the specimens was 28 cm$^2$ and the immersion time was 720 h. Each experiment was repeated three times and the corrosion rate was calculated separately. The results are shown in Fig. 5.

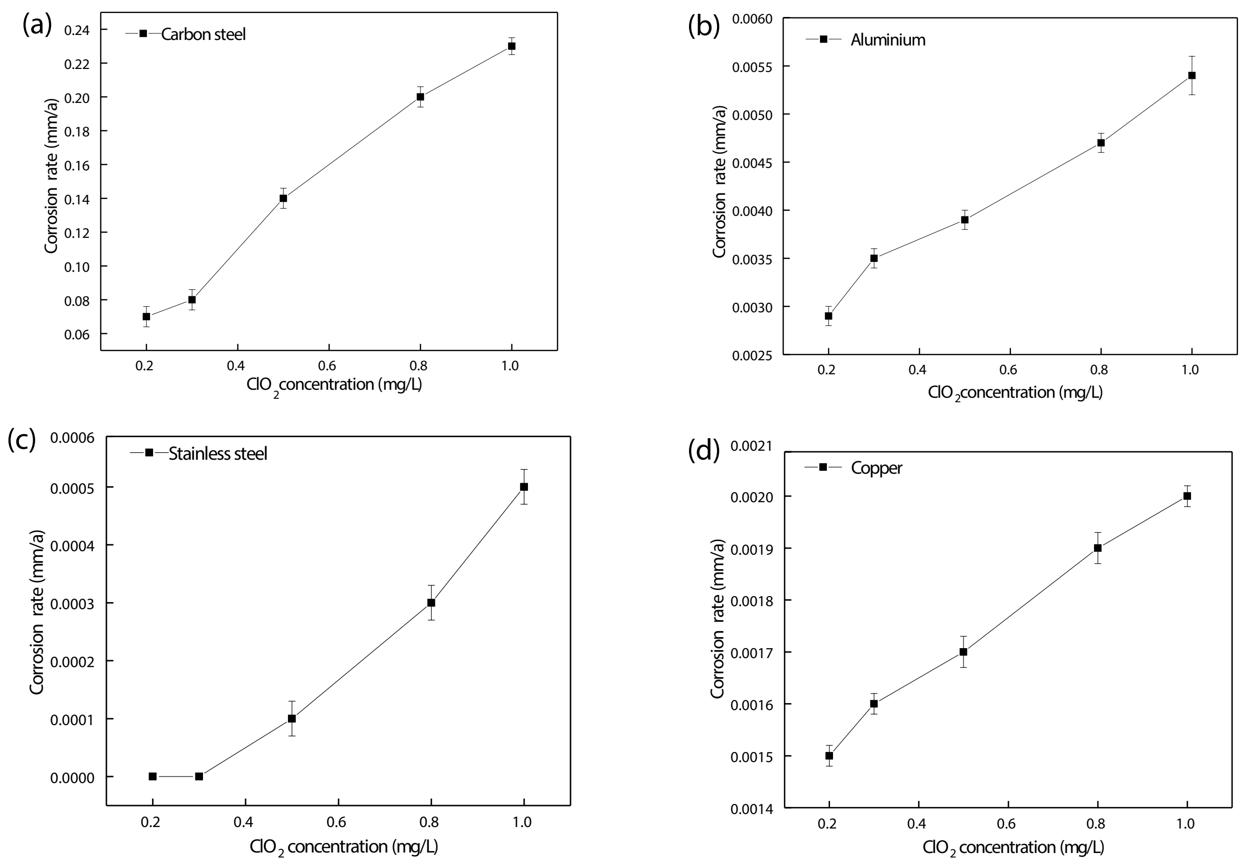


Fig. 5. Effect of different $ClO_2$ concentrations on corrosion rate of: (a) carbon steel, (b) aluminum, (c) stainless steel and (d) copper.

In Fig. 5, the corrosion rates of carbon steel, aluminum, red copper and stainless steel increased with increasing $ClO_2$ concentration, indicating that the metal corrosion was intensified by increasing the dosage of bactericide. Carbon steel showed the highest average corrosion rate (0.144 mm/a), followed by aluminum (0.004 mm/a), red copper (0.0017 mm/a) and stainless steel (0.00018 mm/a). The corrosion rates of $ClO_2$ to aluminum, red copper and stainless were within the standards of the Design Codes for Industrial Circulating Cooling Water Processing (GB50050-17); however, aluminum, red copper and stainless steel cannot be extensively applied due to their high cost. Currently, circulating water cooling pipes in China are all made of carbon steel. The corrosion rate of carbon steel was significantly influenced by $ClO_2$ concentration and it exceeded the standard value. Nevertheless, the corrosion rates for the other materials under different $ClO_2$ concentrations were controlled within the standard value despite fluctuating slightly at different $ClO_2$ concentrations. The application of $ClO_2$ would be certain to cause corrosion of the carbon steel pipes that are used in China.

### *3.2.2. Corrosion rate of liquid chlorine, $ClO_2$ solution and $ClO_2/Cl_2$ mixed liquid on carbon steel*

After the carbon steel was weighed and treated as described above, it was immersed in $ClO_2$ (0.5 mg/L), $Cl_2$ solution (0.5 mg/L) or $ClO_2$ and $Cl_2$ mixture (0.5 mg/L) to study the corrosion rates in the different chlorine oxidizer solutions. The area of the specimen was 28 $cm^2$, immersion time was 720 h and the experiment was repeated three times. The calculated corrosion rates are shown in Fig. 6.

Fig. 6 revealed that the minimum corrosion rate of carbon steel was in $ClO_2$ rather than in the same concentrations of liquid chlorine or $ClO_2$-liquid chlorine mixture. The average corrosion rate of carbon steel in $ClO_2$ was 0.0737 mm/a, and the average corrosion rates in the mixture and liquid chlorine reached 0.0917 and 0.1063 mm/a, respectively. In conclusion, $ClO_2$ was superior to liquid chlorine and the mixture in controlling the corrosion behavior of carbon steel; however, the corrosion rate of carbon steel in liquid chlorine was still significantly higher than the standard limits.

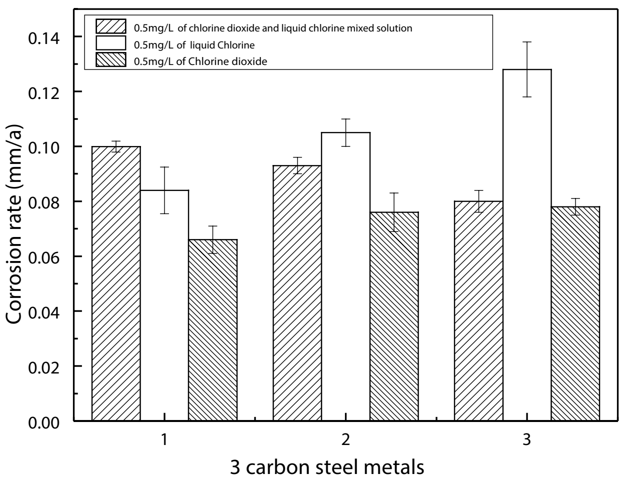


Fig. 6. Corrosion rate of carbon steel in different bactericides solution.

### *3.2.3. Corrosion rates of $ClO_2$, $Cl_2$ and their mixture on four types of materials*

Carbon steel, aluminum, stainless steel and copper were weighed in the manner described earlier and immersed in $ClO_2$ (0.5 mg/L), $Cl_2$ (0.5 mg/L) or $ClO_2$ and $Cl_2$ mixture (0.5 mg/L). The immersion time was 720 h and the experiment was repeated three times. The calculated corrosion rates are shown in Fig. 7.

Fig. 7 shows that the order of corrosion rates of four metal materials was $ClO_2$ < $ClO_2$–liquid chlorine mixture < liquid chlorine. This reflects that $ClO_2$ can inhibit metal corrosion to some extent compared with disinfection liquid containing $Cl_2$. Liquid chlorine had a highly corrosive effect on carbon steel and its corrosion rate increased with increasing concentration; this was mainly because of the large amount of free $Cl^-$ ions in the solution, which easily destroyed the passivation film on the carbon steel surface and decreased the pH value. This, therefore, resulted in pitting to form small primary cell structures with free metal cations on the surface of the metal, which was then further corroded by acid or electrochemical processes. Generally, these processes act simultaneously to deteriorate the material life and shorten its lifespan in circulating cooling water structures [19,20].

The $ClO_2$ solution also has strong oxidizing properties that not only destroy bacteria but also cause metal corrosion in circulating cooling water structures. $ClO_2$ apparently exists in water in molecular form, and, theoretically, electrochemical processes can readily occur with carbon steel in cooling water: in the anodic region carbon steel is oxidized to form ferrous ions that are released into the water, leaving two electrons on the carbon steel metal substrate; at the same time, in the cathodic region $ClO_2$ molecules in the water accept the electrons left on the carbon steel and are reduced to form $Cl^-$ and $H_2O$. The reaction at the two electrodes is as follows:

Anode: $Fe - 2e^- \rightarrow Fe^{2+}$
Cathode: $ClO_2 + 4H^+ + 5e^- \rightarrow Cl^- + 2H_2O$

Higher concentrations of $ClO_2$ in circulating cooling water continue to oxidize the aqueous $Fe^{2+}$ to form $Fe^{3+}$, thereby forming a yellowish rust composed of FeOOH or $Fe_2O_3.H_2O$ [21,22].

By studying the corrosion rate and bactericidal effect of $ClO_2$ at a different concentration, it could be determined that the bactericidal effect increased gradually with increasing concentration, and the corrosion rate of metals also showed a gradual upward trend. However, the addition of a low concentration of $ClO_2$ could decrease the metal corrosion rate, mainly due to the oxidation of $ClO_2$ gradually forming a passive film on the surface of the metals, thus hindering their further corrosion. Results also showed that the addition of low concentrations of pure $ClO_2$ in circulating cooling water not only had a significant effect on alleviating the problem of biological corrosion but also had a certain degree of corrosion inhibition effect, which should be made widely known.

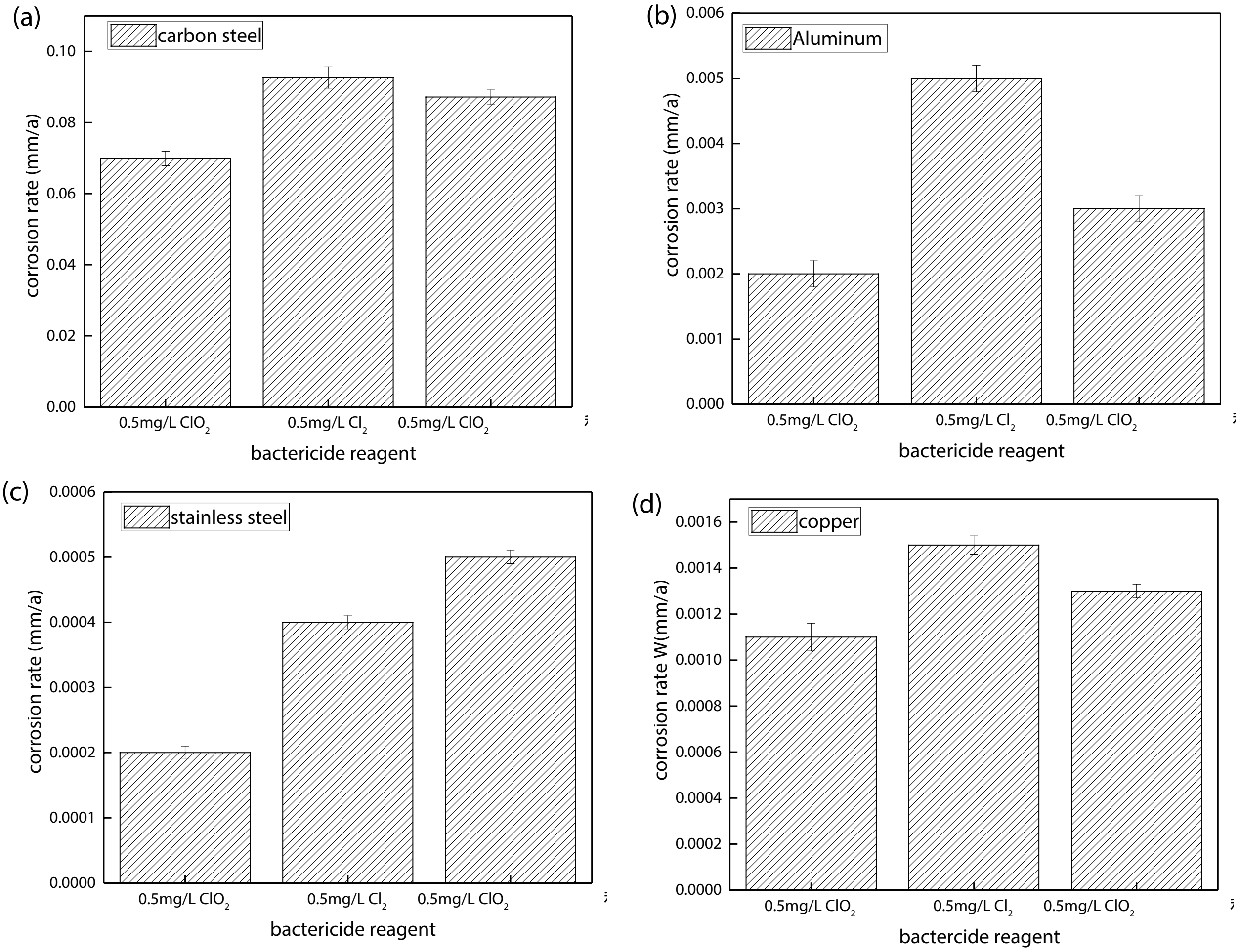


Fig. 7. Corrosion rate of different bactericides with different materials; (a) carbon steel, (b) aluminum, (c) stainless steel and (d) copper.

## 4. Conclusion

The following conclusions can be drawn from the experimental study on the bactericidal rate of chlorine-oxidizing bactericides on heterotrophic, sulfur and iron bacteria and the corrosive action of $ClO_2$, $Cl_2$ and their mixtures on different metal materials:

- $ClO_2$ had good bactericidal efficiency on heterotrophic, sulfur and iron bacteria. Increased $ClO_2$ concentration, pH, temperature and reaction time all increased the sterilization rate.
- In $ClO_2$ solution, carbon steel corroded more than the other three metal materials and the average corrosion rate was 0.144 mm/a, forming a reddish-brown scale; aluminum, copper and stainless steel had average corrosion rates of 0.004 mm/a, 0.0017 mm/a and 0.00018 mm/a, respectively.
- The corrosion rate of metals in $ClO_2$ solution was lower than those in liquid chlorine and $ClO_2$–liquid chlorine mixture. The average corrosion rate in $ClO_2$ was 0.0737 mm/a. The average corrosion rates in the mixture and liquid chlorine were higher, reaching 0.0917 and 0.1063 mm/a, respectively. This was mainly because $Cl^-$ in the $Cl_2$-containing disinfection liquid could cause point corrosion of metals, whereas $ClO_2$ is oxidized and formed a passivation film on the metal surface to somewhat inhibit metal corrosion.

In conclusion, by comparing the bactericidal effect of chlorine-based bactericides on heterotrophic, sulfur and iron bacteria, and on the corrosion of different metal materials, it can be seen that $ClO_2$ is an ideal bactericide for circulating cooling water. It can not only inhibit the growth of algae and bacteria but also control the corrosion of metals. It is, therefore, an ideal bactericide for circulating cooling water.

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## Conflicts of interest

Declarations of interest: none.

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