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Effect of diesel on corrosion inhibitors and application of bio-enzyme corrosion inhibitors in the laboratory cooling water system

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ABSTRACT

Negative effects of diesel on corrosion inhibition performances of 1-hydroxy ethylidene-1, 1-diphosphonic acid (HEDP), amino trimethylene phosphonic acid (ATMP), lysozyme and laccase were investigated. Orthogonal array design (OAD) was used for achieving the optimal composition of a combined inhibitor. The applied conditions of the combined inhibitor were also optimized. The results show that diesel can badly affect corrosion inhibition performances of HEDP, ATMP, lysozyme and laccase. The optimal combined inhibitor consisting of 30 mg/L ATMP, 30 mg/L lysozyme and 40 mg/L laccase can make the corrosion rate of carbon steel below 0.0075 mm/y and the corrosion inhibition efficiency above 96%.

1. Introduction

Poor quality of heat exchangers, outdated hermetic sealing technique, aging pipelines, improper operation and other reasons can all lead to the leakage of petroleum products. In order to reduce leakage hazards, refineries usually increase emissions and reduce the concentration index [1]. The above operation modes undoubtedly intensify the contradiction between water consumption increase and water resources shortages. In addition, the leakage of petroleum products easily introduces abundant organic substrates into circulating cooling water systems, which easily enhance biofilm formation. Geesey and Bryers [2] found that almost half of defects in cooling water systems were due to biofilm formation.

Besides biofilm problems, corrosion problems should be considered equally. Corrosion is an interfacial process leading to surface degradation. It involves electrochemical reactions between the material and its environment. In aqueous media, these reactions are governed by physicochemical parameters (pH, redox potential, conductivity, etc.). Microorganisms may also deeply influence this process, initiating or accelerating surface degradation. A microbial layer forms as a result of the adhesion and growth of microorganisms on metal surfaces. It corresponds to the formation of a complex hydrated matrix including polysaccharides and proteins. Cell metabolites accumulate and enhance corrosion by changing local physicochemical conditions. This phenomenon is commonly called "microbial influenced corrosion" (MIC) [3].

Significant scientific and technological efforts have been made to control corrosion problems. Chemical inhibitors play an important role in the protection and mitigation strategies for retarding corrosion. The most effective and efficient inhibitors are organic compounds that have π bonds, heteroatoms (P, S, N and O) and inorganic compounds [4–10]. The inhibitory efficiency of organic molecules mainly depends on their adsorption ability on metal surface, which can markedly change corrosion resistance of metals [11]. However, the use of these compounds has been questioned lately, due to their toxicities and insufficient inhibitor efficiencies at low dosages [12–15]. Thus, the development of novel corrosion inhibitors which are natural source and non-toxic type has been considered to be more important and desirable. Because of their natural origin, non-toxic features and negligible negative impacts on the aquatic environment, bio-enzymes seem to be ideal candidates to replace traditional toxic corrosion inhibitors.

Lysozyme (N-acetylmuramide glycanhydrolase, EC 3.2.1.17) contains a total of 29 charged groups located on the surface of molecules: 11 arginines, 6 lysines, 1 histidine, 7 aspartic acids, 2 glutamic acids and the N- and C-terminus which can all be protonated or deprotonated depending on solution pH and their intrinsic pKa [16]. The bactericidal activity of lysozyme is muramidase-dependent and uses cation-dependent or structure-related mechanisms [17,18]. The antifungal activity is also well known [19]. However, no studies have been done to test its corrosion inhibition performance.

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Laccase (benzenediol:oxygen oxidoreductase, EC 1.10.3.2) belongs to the group of blue oxidase and represents the largest subgroup of multicopper oxidase. Laccase can catalyze the oxidation of a wide variety of substrates, including mono-, di-, and polyphenols, aminophenols, methoxyphenols, aromatic amines and ascorbate, with the concomitant four-electron reduction of oxygen to water [20]. The use of laccase in micro-pollutant elimination, xenobiotic degradation and decolorization has been extensively studied in the last decades [21–23]. Similarly, little research has focused on laccase corrosion inhibition performance.

When the leakage of petroleum products happens, the cooling water quality will obviously change and it will lead to important effects on biofilm growth characteristics and on corrosion reactions. Because of this, the performance of corrosion inhibitors will be affected. This work was therefore carried out to study the effect of petroleum products on performances of common corrosion inhibitors and bio-enzymes. In order to eliminate this effect, the combined inhibitors containing both chemical inhibitor and bio-enzyme were optimized. In addition, the application conditions of the optimal combined inhibitor were also investigated.

2. Experimental

2.1. Experimental device

Fig. 1 shows the simplified schematic diagram of the rotary coupon corrosion test device, which was a laboratory cooling water system. Standard carbon steel coupons (Q235, $50 \times 25 \times 2$ mm^3) were used with a composition of C: 0.16%; Si: 0.30%; Mn: 0.53%; P: <0.035%; S: <0.04% and Fe balanced (in weight percentage). These coupons were purchased from Hangzhou Guanjie Industrial Clean Water Treatment Technology Corporation. The cleaned coupons were stored in desiccators. Representative industrial water from a circulating cooling water system was taken from Qingdao Refining & Chemical Corporation of China Petrochemical Corporation.

2.2. Diesel oil

Diesel oil is a good model for studying the leakage of petroleum products since it contains a variety of hydrocarbons. The diesel oil was added into the cooling water. The diesel concentrations in the cooling water were 0 mg/L, 80 mg/L, 160 mg/L, 320 mg/L, 480 mg/L, 800 mg/L and 1200 mg/L, respectively. After 72 h of adding diesel oil, coupons were taken out for analysis. The diesel density was 0.84 g/cm^3 at 20 °C and the viscosity was 3.26 mm^2/s at 35 °C. Zinc and iron contents in diesel were 2.38 μg/g and 0.91 μg/g, respectively.

2.3. Chemical inhibitors and bio-enzymes

The chemical inhibitors included amino trimethylene phosphonic acid (ATMP) and 1-hydroxy ethylidene-1, 1-diphosphonic acid (HEDP). The bio-enzymes included lysozyme (>20,000 units/mg protein) and laccase (>2000 units/mg protein). The molecular structures of ATMP, HEDP, lysozyme and laccase are shown in Fig. 2.

2.4. Experimental methods

The tests were carried out through four stages. In the first stage, diesel corrosiveness was investigated. The diesel concentrations in the cooling water were 0 mg/L, 80 mg/L, 160 mg/L, 320 mg/L, 480 mg/L, 800 mg/L and 1200 mg/L, without adding chemical inhibitors and bio-enzymes. After 72 h of adding diesel oil, coupons were taken out for analyzing corrosion rates. In the second stage, the effects of diesel on corrosion inhibition performances of HEDP, ATMP, lysozyme and laccase were investigated. HEDP, ATMP and lysozyme concentrations were 0 mg/L, 5 mg/L, 10 mg/L, 20 mg/L, 30 mg/L, 40 mg/L, 60 mg/L and 80 mg/L, respectively. Laccase concentration was 0 mg/L, 10 mg/L, 20 mg/L, 40 mg/L, 60 mg/L, 80 mg/L and 100 mg/L, respectively. In addition, the degradation of diesel by laccase was also investigated. In the third stage, the orthogonal array design (OAD) was used. In the fourth stage, some application conditions, such as pH value, temperature, and calcium ion content, were investigated in order to optimize corrosion performances of the combined inhibitor. In the above four stages, coupons were taken out for analyzing corrosion rate and corrosion inhibition efficiency after 72 h of adding HEDP, ATMP, lysozyme, laccase or the combined inhibitor. The steady rotating speed was kept at 80 rpm in the laboratory circulating cooling water system. The corrosion tests were repeated three times to obtain reproducible results.

2.5. Orthogonal array design

The orthogonal array ($L_9 3^4$) was designed with ATMP, lysozyme and laccase as factors. Nine experiments were completed in accordance with orthogonal array design (OAD). The experiment conditions of OAD are listed in Table 1. The immersion time of coupons into the cooling water containing combined inhibitors and 80 mg/L diesel was 72 h. The pH value of the cooling water was about 8 and the test temperature was 40 °C.

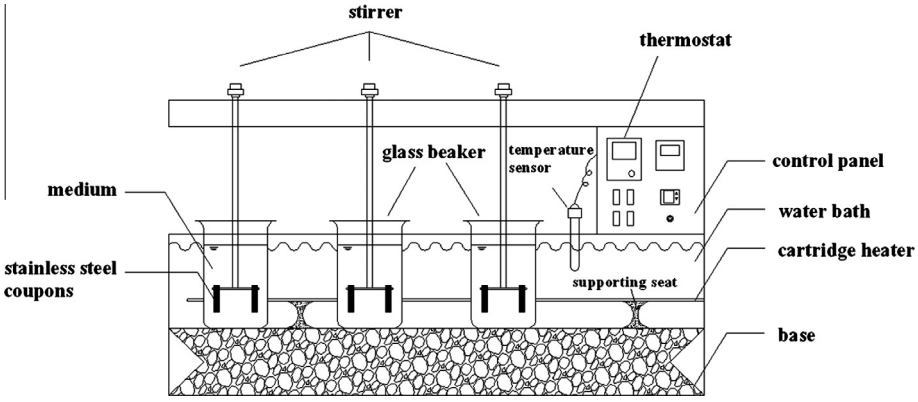

Fig. 1. Schematic diagram of the rotary coupon corrosion test device.