



## Protection of low carbon steel by oxadiazole derivatives and biocide against corrosion in simulated cooling water system



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### ABSTRACT

Electrochemical behavior of low carbon steel in simulated cooling water system in the absence and presence of oxadiazole derivatives, namely 2,5-bis(*n*-methylphenyl)-1,3,4-oxadiazole with ( $n=2,3,4$ ), denoted *n*-MPOX, have been investigated using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The polarizations measurements revealed that the oxadiazole derivatives inhibited both cathodic and anodic reactions. Inhibition efficiency depended on the methyl group position and followed the sequence: 3-MPOX  $\geq$  4-MPOX  $>$  2-MPOX. In addition, their inhibitions were enhanced by the addition of non-oxidizing biocide CTAB. So, a large plate of passivation was registered in the case of mixture 1 (3-MPOX+CTAB) which reinforced the low carbon steel resistance against pitting corrosion. Scanning electron microscopy (SEM) observations and energy dispersive X-ray (EDX) analysis of the low carbon steel surface illustrate that the surface homogeneity increases in the presence of mixtures and confirmed the inhibitor effect after 2 days of immersion.

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### Introduction

The use of water as thermal fluid in cooling systems usually leads to three phenomena: corrosion, scaling and biofouling processes, which can occur jointly, reducing the thermal efficiency, metal degradation and the proliferation of microorganisms. These problems are closely related to the chemical composition of the water, the nature of the metal and operating conditions of the system. In addition, the microorganisms drastically alter the physical chemistry interface of metal/environmental, thereby causing the initiation and the acceleration of severe localized corrosion (the order of  $1 \text{ cm year}^{-1}$ ) [1].

In order to limit these damages, many mixtures for cooling water treatment were investigated [2–5]. These formulations contain essentially corrosion, scaling inhibitors, and biocides to control the microorganism's growth [4–6]. So, in our previous works, many formulations, containing sodium gluconate [7,8], monosaccharides [9], phosphonate [10], tetrazole [6], or TDMTAA

[11], were used as corrosion and scale inhibitors using electrochemical measurements and SEM/EDX analysis. It is shown that these compounds gave a good inhibition against corrosion and scale phenomena. Furthermore, the inhibition properties of the formed film remained independent of the hydrodynamic conditions and reinforced with immersion time, and remained practically independent of the CTAB addition.

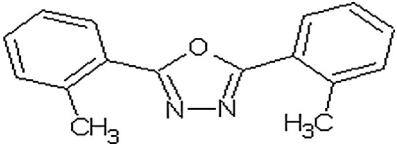
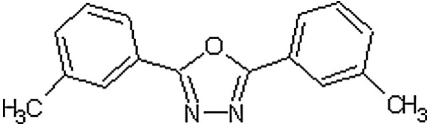
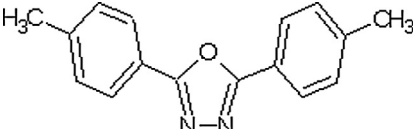
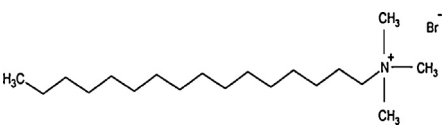
For instance, in our recently paper [5], the oxadiazoles derivatives effect on corrosion and scale inhibition of brass in simulated cooling water was compared. It was found that these compounds act as mixed inhibitors-type and their inhibition efficiency depended on their molecular structure and their concentrations. In addition, their inhibitions were enhanced by the non-oxidizing biocide CTAB addition.

In the present study, new formulations of water treatment in cooling systems for low carbon steel were developed. These formulations include oxadiazoles derivatives, namely 2,5-bis(*n*-methylphenyl)-1,3,4-oxadiazole ( $n=2,3,4$ ) (Table 1), associated with a non-oxidizing biocide cetyltrimethylammonium bromide (CTAB). This study was conducted by using potentiodynamic polarization curves and electrochemical impedance spectroscopy diagrams. Surface characterization studies were used to determine the nature of the formed protective film on the low carbon steel surface.

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**Table 1**  
Molecular structures, names and abbreviations of the studied oxadiazole derivatives and biocide.

Inhibitor formula	Name	Abbreviation
	2,5-Bis(2-methylphenyl)-1,3,4-oxadiazole	2-MPOX
	2,5-Bis(3-methylphenyl)-1,3,4-oxadiazole	3-MPOX
	2,5-Bis(4-methylphenyl)-1,3,4-oxadiazole	4-MPOX
	Cetyltrimethylammonium bromide	CTAB

## Experimental procedure

### Electrochemical cell and materials

The working electrode was low carbon steel and had a chemical composition previously defined [7] (Table 2). It was mounted onto a rotating support to form a rotating disk electrode (RDE). The rotating electrode used was an INF-EL-EDI 101 type. Before each experiment, the working electrode was prepared by mechanical polishing with emery paper, rinsed thoroughly with distilled water, degreased with acetone, and finally dried with compressed air. A saturated calomel electrode (SCE) was used as a reference. All potentials were given with reference to this electrode. The potential was corrected from the ohmic drop due to the solution resistance. The reference electrode was located far enough from the disk electrode to avoid distortions in potential and velocity distributions [12]. The counter electrode was a platinum plate of large surface area. The simulated cooling water solution, with an electrical conductivity of  $1678 \mu\text{S cm}^{-1}$  measured at 299 K, was used previously [5] and its composition was displayed in Table 3. This composition represents the average amount of various ions present in the waters used in Moroccan cooling waters system.

2,5-Bis(2-methylphenyl)-1,3,4-oxadiazole (2-MPOX), 2,5-bis(3-methylphenyl)-1,3,4-oxadiazole (3-MPOX), and 2,5-bis(4-methylphenyl)-1,3,4-oxadiazole (4-MPOX) were used as corrosion inhibitors. The cetyltrimethylammonium bromide (CTAB) was added as corrosion and biofilm inhibitor. The biocide was evaluated using the minimal inhibitory concentration technique [13,14]. There structure formulas were presented in Table 1. All assessments were

carried out at temperature and pH of  $32 \pm 1^\circ\text{C}$  and  $7.35 \pm 0.02$ , respectively.

### Electrochemical methods

The working electrode was immersed in the test solution during 60 min until a steady state open circuit potential ( $E_{\text{ocp}}$ ) was obtained. For this study, we use a rotation speed of the disk of 1000 rpm. The cathodic polarization curve was recorded by polarization from  $E_{\text{ocp}}$  to negative direction under potentiodynamic conditions corresponding to  $1 \text{ mVs}^{-1}$  (sweep rate) and under air atmosphere. After this scan, the anodic polarization curve was recorded by polarization from  $E_{\text{ocp}}$  to positive direction under the same conditions as said before. To evaluate corrosion kinetic parameters, a fitting by Stern–Geary equation was used. So, the overall current density values,  $i$ , were considered as the sum of two contributions, anodic and cathodic current  $i_a$  and  $i_c$ , respectively. For the potential domain not too far from the open circuit, it may be considering that both processes follow the Tafel law [15]. Thus, it can be derived from Eq. (1):

$$i = i_a + i_c = i_{\text{corr}} \times \{ \exp[b_a \times (E - E_{\text{corr}})] - \exp[b_c \times (E - E_{\text{corr}})] \} \quad (1)$$

where  $i_{\text{corr}}$  is the corrosion current density ( $\text{A cm}^{-2}$ ),  $b_a$  and  $b_c$  are the Tafel constants of anodic and cathodic reactions ( $\text{V}^{-1}$ ), respectively. These constant are linked to the Tafel slope  $\beta$

**Table 2**  
Chemical composition of low carbon steel in wt%.

C	Si	Mn	Cr	Mo	Ni	Al	Cu	Co	V	W	Fe
0.11	0.24	0.47	0.12	0.02	0.1	0.03	0.14	<0.0012	<0.003	0.06	Balance

**Table 3**  
Composition of the studied simulated cooling water.

Salts	MgCl <sub>2</sub> , 6H <sub>2</sub> O	CaCl <sub>2</sub> , 2H <sub>2</sub> O	Ca(NO <sub>3</sub> ) <sub>2</sub> , 6H <sub>2</sub> O	MgSO <sub>4</sub> , 7H <sub>2</sub> O	NaHCO <sub>3</sub>
Concentration (mM)	2.85	3.64	0.566	1.56	3.77

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