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A study on the mixed corrosion inhibitor with a dominant cathodic inhibitor for mild steel in aqueous chloride solution

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ABSTRACT

Lanthanum diphenylphosphate compound ($\text{La}(\text{dpp})_3$) has been developed as an effective corrosion inhibitor system for controlling the dominant cathodic process of the electrochemical reaction on mild steel surface immersed in an aqueous chloride solution. The study has clearly demonstrated that less corrosion attack has been shown on the steel surface of steel coupons immersed in solutions containing $\text{La}(\text{dpp})_3$ compound even at low concentrations. This research has also proved that the $\text{La}(\text{dpp})_3$ compound improves the inhibition effectiveness with an increase in inhibitor concentrations, resulting in the superior protective film and charge transfer resistances, as well as protective film and double layer CPE magnitude values.

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1. Introduction

Corrosion of metals and alloys always have a significant effect on the economy, they pollute the environment and are also hazardous to human health and life [1]. This is one of the main factors which can lead to premature failure of infrastructure in critical sectors. Despite of the developments in corrosion resistant alloys over the past few decades, carbon steel still constitutes 99% of the material used in industrial applications [2] due to the most cost effective option and easier processing than other alloys such as stainless steel. Unfortunately, due to poor corrosion resistance in such aggressive environments, this concern can only be realized by adding a corrosion inhibitor to the environment [3–5] or applying a cathodic protection [6] or applying a protective coating [7] or using alloying element [8–10] or controlling microstructure [11,12]. Among these methods, inhibition is the most flexible mean of corrosion control due to its advantage which can be implemented changes in situ without disrupting a process, as well as possibly controlled both internal and external corrosions [13–18].

The inhibition performance of molasses and vegetable oils for protecting steel in acidic environment was evaluated by experimental researches in the 18th century and firstly published in 1895 as a patent by Baldwin [18]. In 1908, inhibitors were extracted

from glue, gelatin, and bran to protect iron in an acidic environment by Marangoni and Stefanelli [19,20]. Furthermore, organic compounds were then applied to inhibit corrosion in oil & gas industry in 1950 and concrete in 1990. Based on the huge benefits of corrosion inhibitors, an increase in research activities and relentless improvement of science in the use of inhibitors are clear evidences of the importance of corrosion inhibitors [21,22]. Each of these types has its own properties depending on the way it affects anodic and cathodic or both reactions, classifying anodic, cathodic, and mixed inhibitors, respectively.

Typical cathodic inhibitors are ZnSO_4 salt and metal chloride salts of which actions depend on zinc (or metals) hydroxide being precipitated at the cathode, where the pH increases, thus making the cathode reaction more difficult [23]. In addition, phosphate salts such as sodium pyrophosphate ($\text{Na}_4\text{P}_2\text{O}_7$) [24], sodium tripolyphosphate ($\text{Na}_5\text{P}_3\text{O}_{10}$), and sodium hexametaphosphate ($(\text{NaPO}_3)_6$) in the presence of divalent metal ions, can form a protective coating on the metal surface which can be used as cathodic inhibitors [25,26]. Furthermore, furfuryl alcohol has been found as an effectively mixed inhibitor with the predominant cathodic inhibition for steel by Vishwanatham and Haldar. They reported that inhibitor molecules were adsorbed on the active sites of the metal surface via Temkin adsorption isotherm. The formation of a compact protective film on the metal surface was attributed to the lone pair of electrons in O-atom and/or p-electrons of the furan ring of the inhibitor molecules. The anodic reaction might be controlled via electrostatic interaction of the

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Table 1
AS1020 steel compositions were checked by optical emission spectroscopy.

AS1020	Chemical elements (%)															
	C	Mn	Ni	Cu	V	Ni	Cr	Al	Co	S	P	Si	Mo	Nb	Ti	Fe
	0.146	0.408	0.0084	0.0227	0.0013	0.0048	0.0158	0.0511	0.0015	<0.0005	<0.0005	<0.001	<0.001	<0.004	<0.001	Bal.

outermost negative end of the furan ring and/or electron rich oxygen atoms in the inhibitor molecules, whereas, cathodic reaction could be controlled via adsorption of the protonated linkage polymer molecules on the negatively charged surface as the cathodic sites, resulting mixed inhibition mechanism. Cruz et al. has reported that 2-aminomethylbenzimidazole and bis(benzimidazol-2-ylethyl)sulphide could act as cathodic inhibitors at the first state for steel in the deaerated HCl solution [27]. The obtained effective inhibition could be attributed to the density of functional theoretical parameters including the frontier molecular orbitals, Mulliken charges and the electrostatic potential map. 1,12-bis(1,2,4-triazolyl)dodecane is also a good recommendation of cathodic inhibitor for steel in HCl solution [28] due to the synergistic effect between chloride ion and the positively charged quaternary ammonium ion moiety contained in the inhibitor molecule. But the effectiveness of this compound for steel might decrease if the immersion time of steel is prolonged or this compound used in the aerated solution. Elachouri et al. [29] changed the number of C atoms (n) in the side chain of 2-(alkyl(C_nH_{n+1}))dimethylammonium butanol bromides from 11 to 15, which showed an effective cathodic inhibition when concentration of these compounds was increased in the investigated solution. Recently, cathodic inhibitors have been developed via a way of synergistic properties of the ions and/or compounds [31–35]. The effective inhibition has been reached, resulting in the formation of protective film on the steel surface.

It means that the research on corrosion inhibitors to date has examined very few cathodic inhibitors and several mixed types of inhibitor with the predominant cathodic inhibition for steel. Therefore, significant progress has been achieved in understanding the inhibition phenomenon in cathodic inhibitors and several mixed types of inhibitor with the predominant cathodic inhibition [31–35], however a comprehensive understanding of the behavior and mechanisms of important corrosion inhibitors is still lacking. Over the past few decades, the primary improvements in inhibitor technology have been limited to the refinement of formulations and the development of improved methods for applying inhibitors [30]. There have been very few discoveries in the area of new corrosion inhibitors. In recent years, the increasing demands for environmentally friendly inhibitors and suppressing localized corrosion have been driving forces behind the advances in corrosion inhibition technology [30,35], but there is still a need to develop improved inhibitor systems, not only for combating corrosion in existing areas, but also to mitigate corrosion within new applications. Furthermore, controlling cathodic processes within corrosion systems by inhibitors have not been satisfied yet. Previous reports [36,37] indicated that rare earth organophosphate compounds can be partially dissociated into the polynuclear species or clustered complexes rather than merely separate ionic species in solution. These species initially form local surface films that could limit cathodic and/or anodic processes of electrochemical corrosion reaction, resulting in an increase of local pH at cathodic sites. This phenomena could cause the rare earth ions to be hydrolysis leading to the deposited film that entirely cover and then inhibit electrochemical corrosion over the metal surface. This possibility suggests that rare earth organophosphate compounds can be a potential candidate for inhibiting corrosion of steel in an aqueous solution. Therefore, this work has concentrated primarily on the characterization and evaluation of the lanthanum

diphenylphosphate compound to create a corrosion inhibitor with more efficient and environmentally benign as well as a mixed type of inhibitor with the predominant cathodic inhibition for protection of carbon steel in aqueous chloride solution systems.

2. Experimental procedures

2.1. Chemicals and materials

The reagents used in this work include lanthanum chloride ($LaCl_3$), diphenylphosphinic acid ($H(dpp)$), reagent grade sodium chloride ($NaCl$) (all purchased from Sigma Aldrich, $\geq 99\%$ pure and used without further purification) and distilled water. Lanthanum diphenylphosphate ($La(dpp)_3$) compound was prepared in detail as previously reported [38–40] and will be referred to as corrosion inhibitor. $LaCl_3 \cdot xH_2O$ was dissolved in 95% ethanol. A solution of $H(dpp)$ dissolved in 95% ethanol was added slowly, with stirring, to the lanthanum chloride/ethanol solution, where upon immediate formation of a precipitate was observed. The pH of the mixture was raised from 1.2 to 6.0 using 0.5 M $NaHCO_3$. The mixture was heated to 30 °C for 2–3 h and allowed to stand overnight. The precipitate was filtered and washed with 5% ethanol, followed by washings with distilled water until the pH of the filtrate was maintained around 6.5–7.0. The precipitate was air dried for 4 h, followed by further drying in a vacuum desiccator for 2 days. $La(dpp)_3$ was added to 0.1 M (mol/L) $NaCl$ solution to make final concentrations of 0.00, 0.11, 0.34 and 0.68 mM (mmol/L) using reagent grade sodium chloride, distilled water, and 12 h of stirring. The steel coupons of 1 cm \times 1 cm \times 0.3 cm used as working electrodes for the electrochemical tests were fabricated from steel sheet. These steel specimens were coated with a low viscosity epoxy cold mounting system with 17:1 ratio of EpoFix resin and EpoFix hardener curing at room temperature in 12 h for controlling 1 cm² of the exposed area, no shrinkage and attached to a Teflon holder. The steel compositions were characterized using optical emission spectroscopy and are presented in Table 1. The steel specimens for corrosion tests were finished by grinding with 1200-grit silicon carbide paper and then rinsed with deionized water and ethanol.

2.2. Electrochemical measurements

A standard three electrode system including a silver/silver chloride ($Ag/AgCl$) reference electrode, a titanium mesh counter electrode, and the working electrode was used for carrying out electrochemical experiments. To prevent electrical interference, the electrochemical cell assembly was placed in a Faraday cage. The electrochemical tests were conducted using a VSP system (BioLogic Scientific Instruments) consisting of a commercial software program for AC measurements. Before electrochemical testing, the specimens were kept in the natural aerated solution for 2 h to stabilize the open-circuit potential. The electrochemical impedance spectroscopy (EIS) test was carried out at open circuit potential (OCP) and conducted every 2 h over a period of 20 h. The 10 mV peak-to-peak amplitude of the sinusoidal perturbation and the frequency range from 10 kHz to 10 mHz were used for EIS testing. In addition, potentiodynamic polarization tests followed ASTM G5-94 were carried out after 20 h of immersion, the potential of

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