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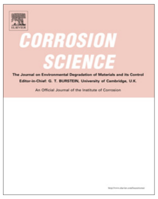

Synergistic corrosion inhibition of environment-friendly inhibitors on the corrosion of carbon steel in soft water

Bingru Zhang [a,c,1,*], Chengjun He [a,1], Cheng Wang [b], Peidi Sun [a], Fengting Li [a], Yu Lin [a]

[a] *State Key Lab of Pollution Control & Resource Reuse, College of Environmental Science and Engineering, Tongji University, 1239 Siping Rd, Shanghai 200092, China*
[b] *Water Supply and Dicharge Design and Research Institute, Yunnan Design Institute, Guangfu Rd, Kunming 650100, China*
[c] *Key Laboratory of Yangtze Aquatic Environment, Ministry of Education, College of Environmental Science and Engineering, Tongji University, 1239 Siping Rd, Shanghai 200092, China*

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ABSTRACT

The synergistic effect of the combination of polyaspartic acid (PASP), polyepoxysuccinic acid (PESA), polyamino polyether methylenephosphonate (PAPEMP), sodium gluconate (Glu) and Zn^{2+} on carbon steel corrosion was investigated using weight loss and electrochemical measurements. The combination of PASP, PESA, PAPEMP, Glu and Zn^{2+} is an environment-friendly inhibitor and exhibited mixed-type inhibition behaviour. The composite efficiently inhibited corrosion on carbon steel at relatively low dosages in severely corrosive soft water media. Atomic force microscopy (AFM) images and X-ray photoelectron spectroscopic (XPS) spectra further confirmed the formation of a protective film composed of the adsorbed inhibitor molecules on the carbon steel surface against corrosion attack.

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1. Introduction

Soft and low-alkalinity waters normally appear in open recirculating cooling systems and water distribution systems. These systems are known to cause relatively high levels of metal corrosion [1], and enable the release of iron corrosion by-products into potable water [2–4]. Carbon and low-alloy steels are the major construction materials extensively used in water distribution, petroleum, power production, as well as chemical and electrochemical industries [5–7]. However, iron-based alloys are inevitably subject to corrosion, especially pitting corrosion in neutral or acidic solutions containing aggressive anions [8]. Corrosion events result in economic loss and even serious operational problems [2,9].

Metals used in industrial applications are protected from corrosion in different ways apart from surface treatment such as coating [10]. The use of organic molecules as corrosion inhibitors is one of the most popular, efficient, and practical methods applied extensively to protect metals against corrosion damage [6,11,12], especially in aggressive acidic media [11,13].

For many years, chromate-based and nitrate-based corrosion inhibitors were considered the performance standard in industrial water treatment such cooling water. Unfortunately, chromates and nitrates are toxic, and accordingly have displaced by more environmentally acceptable chemicals, such as silicates, molybdates, phosphate/polyphosphates, and zinc salts have been recognised as the more environmentally acceptable carbon steel corrosion inhibitors replacing chromates [4–6].

From the 1960s to early 1970s, phosphates/polyphosphates such as sodium hexametaphosphate and sodium tripolyphosphate, which possess $-O-P(O)(OH)_2$, were the optimal choices due to their low cost, versatility, and performance. However, phosphate-based cooling water treatment programmes rely on relatively higher dosages for corrosion inhibition and require more careful control of system parameters than chromate-based programmes. High phosphate dosages can result not only in calcium phosphate precipitation and the fouling of heat exchanger surfaces, especially in alkaline cooling water systems, but also in the eutrophication of seas, rivers, and lakes. Consequently [2], given the environmental problems and tendencies of these compounds to form calcium phosphate scales, their use has been decreased in favour of other corrosion inhibitors, such as phosphonates and anionic polymers.

From the mid-1970s to early 1990s, phosphonates were developed and extensively used as scale and corrosion inhibitors. Phosphonates such as 1-hydroxyethane-1,1-diphosphonic acid (HEDP) [14], amino (trimethylene phosphonic acid) (ATMP) [15], ethylenediaminetetra (methylenephosphonic acid) (EDTMP) [16], diethylenetriaminepentakis (methylenephosphonic acid) (DTPMP) [17], and 2-Phosphonobutane-1,2,4 tricarboxylic acid (PBTCA) [18] contain one or more $R_3C-P(O)(OH)_2$ group/s instead of an

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[*] Corresponding author at: State Key Lab of Pollution Control & Resource Reuse, College of Environmental Science and Engineering, Tongji University, 1239 Siping Rd, Shanghai 200092, China. Tel.: +86 21 65980567; fax: +86 21 65985059.
E-mail address: zhangbingru2000@126.com (B. Zhang).

[1] These two authors contributed equally to this work.

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$R_3C–O–P(O)(OH)_2$ group. Phosphonates possess a number of superior qualities, including high chemical stability under extreme pH and temperature, ability to complex metals, ability to adsorb strongly onto a metal surface, as well as dispersant activity against suspended matter.

Over the past three decades, many water-soluble anionic polymers have been developed as corrosion inhibitors. These polymers are mainly homo-, co-, or ter-polymers having acrylic or maleic acid as the initial monomer together with other monomers containing acrylamide, esters, sulphonic acid, phosphoric acid, etc. These polymers are excellent corrosion inhibitors. Acrylic acid-based scale and corrosion inhibitors, such as polyacrylic acid [23], acrylic acid/2-acrylamido-2-methypropane sulphonic acid copolymer, and acrylic acid/2-acrylamide-2-methypropane sulphonic acid/2-hydroxypropyl-acrylate copolymer [24], are generally low toxicity but are difficult to biodegrade.

In the 21st century, biodegradability has become an important mechanism for limiting chemical build-up in the environment. Biodegradability is the ability of a material to be broken down into simple and non-toxic components by the action of microorganisms and fungi.

Polyaspartic acid (PASP) and polyepoxysuccinic acid (PESA) are representative green scale inhibitors due to their non-phosphorus and biodegradable features [25–28]. Hence, they are valuable in terms of environmental acceptability and waste disposal [29]. Many relevant studies on PASP have focused on its synthesis, properties, and application as a mineral scale inhibitor in water treatment applications [30]. PASP can also be used as a dispersing agent in detergents, paints, and papermaking processes [31]. PESA, given its good chelating ability with various metal ions [32], exhibits a versatile anti-scaling performance [29,33,34] and is even used in the extraction of Cd from sewage sludge [35]. However, the application of PESA and PESA as corrosion inhibitors has not been investigated.

Polyamino polyether methylenephosphonate (PAPEMP), a newly discovered inhibitor, has been developed for scale control in water desalination. PAPEMP has a good performance on controlling both calcium carbonate as well as calcium sulphate scales at extremely high supersaturations [36–38]. PAPEMP is also capable of suppressing calcium phosphonate precipitation [19]. Numerous studies have focused on the mineral scale inhibition efficiency of PASP, PESA, and PAPEMP. However, studies on their corrosion inhibition performances are limited. Previous studies have indicated that the sodium and calcium salts of gluconic acid are effective corrosion inhibitors for mild steel immersed in near-neutral media [39,40] and chloride solutions [41,42,50]. Sodium gluconate (Glu) can be applied in simulated cooling water as a corrosion inhibitor owing to its non-poisonous characteristic [40]. Zinc ions are frequently used as cathodic inorganic corrosion inhibitor [80].

Electrochemical measurements by electrochemical impedance spectroscopy (EIS) and potentiodynamic polarisation techniques are commonly used to identify the inhibitor type, model the corrosion and film-forming processes on the metal/solution interface, and calculate the corrosion inhibition efficiency of inhibitors based on specific parameters [2,7,9,43–46]. Potentiodynamic polarisation and electrochemical impedance spectroscopy (EIS) are employed to evaluate the inhibition efficiency and mechanism of the inhibitors monolayers on the steel surface in test solution at 318 K. Atomic force microscopy (AFM), a powerful technique for investigating surface topography, has been recently used to study the effects of inhibitors on the metal/solution interface and the formation of the adsorbed barrier layer [5,6,9,47–49].

The present study investigated a novel environment-friendly composite of inhibitors that can efficiently inhibit corrosion on carbon steel at relatively low dosages in soft water media. The purpose was to overcome the challenge of the inhibition efficiency of a single component being greatly dependent on a great quantity of inhibitor. The inhibition effects of a single material (including PASP, PESA, PAPEMP, Glu and Zn^{2+}) and their multi-component blends on carbon steel corrosion in soft water media were studied by weight loss experiments [6,20–23]. The underlying mechanisms of the inhibitors on carbon steel corrosion were examined by electrochemical measurements. The optimum additive concentrations of the inhibitors involved in the compound formula were determined by the orthogonal test. AFM images were utilised to further confirm the corrosion attack as well as inhibitor molecule adsorption and film-forming processes. In order to clarify the formation of adsorption layer, X-ray photoelectron spectroscopic (XPS) spectroscopy could be used for detecting the monolayer adsorption behaviour of inhibitors on the steel surface to clarify the molecular interaction mechanism [69,70].

2. Experimental

2.1. Reagents and materials

All chemicals used in this study were analytical reagent grade and obtained from the Sinopharm Chemical Reagent Shanghai Co., Ltd., China. Double-distilled water was used. Calcium stock solutions were prepared from calcium chloride dihydrate and filtered with 0.22 μm filter paper prior to analysis by EDTA complexometric titration.

The polymer inhibitors used were commercial grade, and their structures are shown in Table 2. PESA (average $Mw \approx 3800$) was obtained from Meijing Environmental Protection Material Co., Ltd., China. PASP (average $Mw \approx 8000$) was purchased from Zibo Leadbond Chemical Products Co., Ltd. PAPEMP was obtained from Jianghai Chemical Group. All polymer inhibitor concentrations were calculated on a dry polymer basis.

Corrosion tests were performed on carbon steel samples with the following composition (in wt.%): 0.17–0.24 C, 0.17–0.37 Si, 0.35–0.65 Mn, $\leqslant 0.035$ P, $\leqslant 0.035$ S, $\leqslant 0.025$ Ni, $\leqslant 0.025$ Cr, $\leqslant 0.025$ Cu, and balanced Fe. These components were used for the weight loss and electrochemical experiments. The specimens used for the weight loss test were rectangular with dimensions $5.0 \times 2.5 \times 0.2\ cm^3$. For the electrochemical tests, the working electrode (WE) was embedded in epoxy resin, leaving a 1 cm^2 geometrical surface area of carbon steel exposed to the electrolyte. Prior to all measurements, the carbon steel specimens were mechanically abraded with different types of emery papers (grade 320–400–600–800–1000–1200), degreased with acetone, and rinsed with distilled water before immersion in the experimental solution. Tap water was used as the test solution and a typical analysis of this electrolyte is given in Table 1.

Table 1
Analysis of tap water (electrolyte).

Parameter	Value
pH	7.1
Conductivity ($\mu S\ cm^{-1}$)	6700
Total hardness ($mg\ L^{-1}$)	142
Alkalinity ($mg\ L^{-1}$)	129
TDS ($mg\ L^{-1}$)	324
Sodium ions ($mg\ L^{-1}$)	8.20
Potassium ions ($mg\ L^{-1}$)	0.55
Calcium ions ($mg\ L^{-1}$)	51.60
Magnesium ions ($mg\ L^{-1}$)	3.12
Chloride ions ($mg\ L^{-1}$)	62
Sulphate ions ($mg\ L^{-1}$)	59
LSI	−1.71
RSI	10.62
PSI	9.83

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