

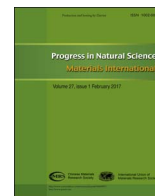
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Original Research

Application of mesoporous silica nanocontainers as an intelligent host of molybdate corrosion inhibitor embedded in the epoxy coated steel[☆]

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ABSTRACT

In this study, mesoporous silica served as a host for corrosion inhibitor. This material could adsorb and release corrosion inhibitor in different aqueous media. However, the extent of corrosion inhibitor release in the alkaline media was higher. By dispersing 1 wt% mesoporous silica loaded with sodium molybdate in the epoxy layer, a protective composite coating was produced. The corrosion properties of the composite coatings were assessed by electrochemical impedance spectroscopy. Results showed higher corrosion resistance of epoxy/mesoporous silica loaded with inhibitor in the NaCl media for eight weeks of immersion in comparison with epoxy/mesoporous silica. Corrosion inhibitors released from nano-containers in the response to corrosion damage at the interface zone.

1. Introduction

The application of organic coatings is one of the most conventional methods to protect of many metallic structures in the corrosive environments. However, degradation processes lead to deterioration of the protective coatings [1–3]. Recently, a new generation of smart anticorrosion coatings has been proposed and investigated, which are composed of two functional components. One part is passive coatings, including sol–gel or polymer coatings which act as a barrier against corrosive species [4,5]. The other part consists of containers which are distributed in the passive coatings and can respond to the local environmental changes associated with the corrosion damage. They can release incorporated corrosion inhibitors to hinder the corrosion process [6–8]. These containers consist of micro or nanocapsules [9–11], ion exchange substances [12–14], layer-by-layer polyelectrolyte shells [15–17], and porous materials [18–24]. Mesoporous silica which is a porous material has been applied as a corrosion inhibitor host [21,23]. This material has highly regular arrays of uniform-sized cylindrical pores with the diameter in the range of 2–50 nm, high surface area (700–1500 m² g⁻¹), large pore volume (1 cm³ g⁻¹), high chemical and thermal stability, and easy functionalization [25]. Well control of pore size, narrow pore size distribution, and controllable functionality of nanoporous silica indicate its application as the encapsulating material for controlled release of corrosion inhibitors in the protective coatings [25].

Falcón and his co-workers evaluated anticorrosive performance of carbon steel coated with alkyd primer loaded with 15 wt% of mesoporous silica incorporated with dodecylamine. Examinations showed that the release of dodecylamine at lower pHs (2.0) was faster than higher ones (6.2 and 9). In addition, due to the release of inhibitor during the corrosion process, active anticorrosion properties and self-healing were provided and detected by electrochemical techniques [18]. Epoxy coatings with both epoxy-encapsulated TiO₂ nanotubes and amine immobilized mesoporous silica were prepared on the carbon steel [19]. The self-healing ability of composite was confirmed by the electrochemical impedance spectroscopy-tests. The self-healing ability of composite coating has been proposed by the release of the epoxy prepolymer and its cross linking with the immobilized amine curing agent in the mesoporous silica [19]. Shchukina et al. [20] loaded halo sites and mesoporous silica with 8-hydroxyquinoline to reduce the corrosion of the metal substrate. A local increase of the pH due to the corrosion process immediately accelerated the solubility of the encapsulated 8-hydroxyquinoline and therefore the corrosion inhibitor release from nanocontainers was provoked [20]. In other work which was conducted by Borisova et al. [21], 1H-benzotriazole loaded in the mesoporous silica and then embedded these containers in the sol-gel films to protect Al-2024 alloy. They reported enhanced corrosion protection of Al alloys in comparison with neat sol-gel coating [21]. Moreover, they also found that the distribution of nanocontainers in any percent was not always

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favorable for the anticorrosion performance of the coating [26]. Besides, introducing the nanocontainers close to the metal surface enhanced the transport of released inhibitor upon corrosion attack and improved the active corrosion resistance and self-healing ability [27,28]. In a previously published paper, mesoporous silica nanocontainers as the corrosion inhibitor hosts dispersed in the conductive polymer matrix (polypyrrole). Corrosion investigations revealed that the addition of mesoporous silica nanocontainer loaded with sodium molybdate in the coating produced a better protected surface in comparison with polypyrrole/mesoporous silica coated steel [29].

However, the application of immobilized inhibitors in the nanocontainer host is still in its beginning, but it is undoubtedly a promising approach for research guidelines. In the present study, molybdate ion immobilized mesoporous silica was dispersed in the epoxy coatings as a corrosion-sensitive nanocontainer in the chloride media. However, mesoporous silica did not possess active sites for functional applications in the case of smart release. Therefore, it would be functionalized by different groups including alkoxy silane molecules. Afterward, cations can facilitate anion (molybdate as a corrosion inhibitor) adsorption on the surface of mesoporous silica. In this study, corrosion inhibitor was immobilized in the mesoporous silica by an electrostatic force resulted from the interaction between corrosion inhibitor and a cation like Fe^{+3} . Therefore, corrosion inhibitor was permitted to release in the corrosive media due to weak interaction between it and cation. Electrochemical behavior of composited coated steels was examined during 8 weeks of immersion in the chloride media to understand protective behavior of coatings in the aggressive media. By immersing epoxy/molybdate ion immobilized mesoporous silica coated steel in the corrosive media, it was expected that the corrosion inhibitors were released and subsequently could hinder the corrosion damage. The main aim of this study is to investigate of long term corrosion behavior of epoxy/molybdate ion immobilized mesoporous silica composite coated steel to understand the ability of smart coating in the aggressive media.

2. Experimental

2.1. Mesoporous silica nanocontainer synthesis

Fig. 1 shows the schematic procedure for the loading of corrosion inhibitor in the structure of mesoporous silica. At first, 500 mg of initial mesoporous silica powder (Fig. 1a) was added to toluene (50 mL) under the nitrogen atmosphere. After addition of 5 mmol 1-(2-aminoethyl)-3aminopropyltrimethoxysilane, the mixture was stirred at 400 K in an inert ambient for 16 h (Fig. 1b). The powder was filtered and rinsed with ethanol. After this step, 500 mg FeCl_3 (Fe^{3+}) was applied into the functionalized mesoporous silica in the propanol solution at ambient temperature for 6 h (Fig. 1c). The created positive ion on the surface of mesoporous silica can subsequently adsorb any anions such as molybdate. The obtained powder through filtration and washing was denoted as MS. Finally, the corrosion inhibitor adsorption process was performed (Fig. 1d). In this step, 250 mg of sodium molybdate powder was mixed with 500 mg of MS in 100 mL water and then stirred for 24 h, followed by filtering and rinsing by ethanol, and then drying. The obtained powder was denoted as MSInh, which could be applied as corrosion inhibitor nano-container in the epoxy coatings.

2.2. Mesoporous silica characterization

The size and morphologies of mesoporous silica powders were studied with a high resolution scanning electron microscope (CamScan Mira, 40 kV) and a transmission electron microscope (TEM Tecnai G2 F30 at 300 kV). The specific surface area, average pore diameter, and pore volume were obtained using N_2 adsorption–desorption isotherms (BELSORP mini-II). The specific surface area was calculated from the

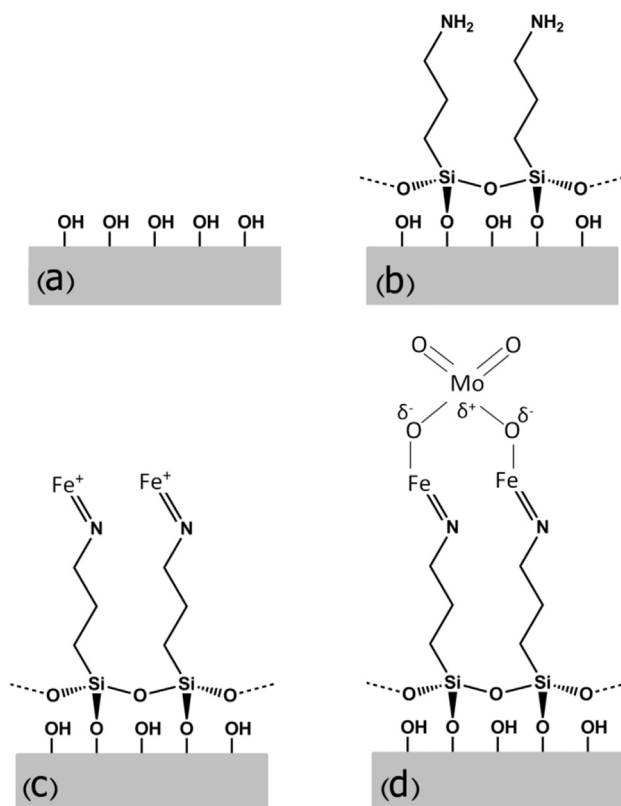


Fig. 1. Schematic procedure concerning the loading of corrosion inhibitor in the mesoporous silica (MS): (a) initial mesoporous silica, (b) functionalization of mesoporous silica by silane group, (c) adsorption of FeCl_3 compound on the mesoporous silica, and (d) adsorption process of corrosion inhibitor on the functionalized mesoporous silica.

adsorption data in the low-pressure range using the Brunauer–Emmett–Teller (BET) method. The average pore diameter and pore volume were determined using the Barret–Joyner–Halenda (BJH) method. The Fourier transform infrared (FT-IR) spectroscopic covered the range of 4000 cm^{-1} to 500 cm^{-1} . The corrosion inhibitor release from MSInh was measured in a range of acidic to alkaline pHs, adjusted by HCl and NaOH, respectively. The amount of molybdate ion release during three days of immersion was characterized by inductively coupled plasma optical emission spectrometry (ICP-OES) model Varian VISTA-MPX.

2.3. Application of coating

Mild steel panels were polished with SiCs and papers up to grade 1200 and carefully degreased with acetone, followed by water rinsing and drying. The epoxy resin (Shell) with EEW=450–550 was mixed with reactive diluent. MSInh or MS powders were added to the mixture. Then, the mixtures were sonicated with a high-power probe-type ultrasonicator (Hielscher, GmbH) for 1 h under power of 70 W and then mixed with hardener (Versamid 115, HEW=198, Saman Co.). The synthesized nanocomposite polymers (1 wt% MS or MSInh) were applied on steel panels by film applicator and cured at $60\text{ }^\circ\text{C}$ for 4 h. The final coating thickness was measured $50 \pm 3\text{ }\mu\text{m}$ by Elcometer 4563 coating thickness meter. The obtained coatings were denoted as epoxy/MS and epoxy/MSInh.

2.4. Study of corrosion behavior

The corrosion behavior of epoxy/MS and epoxy/MSInh coatings were examined in the 3 wt% NaCl solution. The surface area corre-

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