



Effects of highly crystalline and conductive polyaniline/graphene oxide composites on the corrosion protection performance of a zinc-rich epoxy coating

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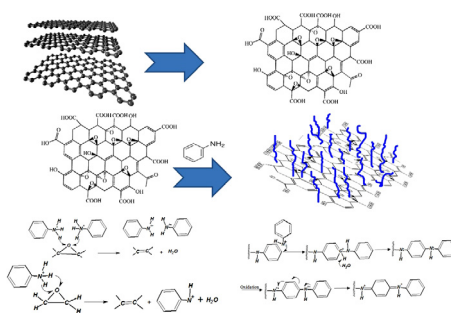
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HIGHLIGHTS

- Modification of GO by polyaniline nanofibers enhanced its electrical conductivity.
- Barrier properties of zinc rich paint remarkably increased after addition of GO-PANI.
- GO-PANI enhanced the electrical contact between zinc particles and steel substrate.
- The zinc rich sacrificial behavior noticeably enhanced after addition of GO-PANI.

GRAPHICAL ABSTRACT



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ABSTRACT

This study reports a new strategy for providing ZRC with enhanced cathodic and barrier protection mechanisms simultaneously. For this purpose, the graphene oxide (GO) nanosheets were modified by highly crystalline and conductive polyaniline (PANI) nanofibers in the form of Emeraldine salt (ES) through an in situ polymerization of aniline in the presence of GO as an oxidant. The aniline polymerization in the presence of GO and the PANI nanofibers deposition on the GO surface were exhibited by Fourier transform infrared (FT-IR) spectroscopy and high resolution-transmittance electron microscopy (HR-TEM). In addition to these the X-ray diffraction (XRD) patterns confirmed the deposition of highly crystalline PANI nanofibers on the GO and between the GO layers. Inclusion of 0.1 wt% GO and GO-PANI nanosheets into the ZRC sample remarkably enhanced its corrosion protection performance. Salt spray, open circuit potential (OCP) and electrochemical impedance spectroscopy (EIS) measurements revealed that both the cathodic protection properties and barrier performance of the ZRC were improved after addition of 0.1 wt% GO and GO-PANI nanosheets to the ZRC sample. The most pronounced improvement in the ZRC properties was obtained using GO-PANI. The results obtained from field-emission scanning electron microscopy (FE-SEM), energy dispersive spectroscopy and XRD analysis confirmed lower degree of zinc particles oxidation and steel substrate corrosion in the case of ZRC including GO-PANI nanosheets compared to other samples.

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1. Introduction

Several coating systems have been used to control or mitigate the corrosion processes that take place on the steel surface in

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exposure with severe environments including organic, inorganic, and hybrid protective coatings [1–3]. Among these methods application of organic coatings is the most popular and effective strategy to protect the steel structures against corrosion [4,5]. Depending on the chemical formulation and the type and amount of the ingredients of the organic coatings they can protect the steel from corrosion through three main mechanisms; (a) barrier, (b) sacrificial and (c) inhibiting features [6–11]. Organic coatings can provide a physical barrier against diffusion of corrosive agents such as H_2O , O_2 and ions i.e. Cl^- to the steel surface. However, they cannot show protective performance in the case of a slight mechanical damage creation on the coating. Zinc-rich coatings, i.e. epoxy zinc-rich, are another type of organic coatings that can protect the metals against corrosion for long exposure times in severe environments, even when there is mechanical damage. Addition of high amount of zinc particles to the epoxy resin provides a coating system with cathodic protection properties [12,13]. In this case the continuous electrical contact between the zinc metal particles and steel substrate provides a galvanic cell, where the steel substrate acts as cathode and zinc particles behave as sacrificial anodes. For a common zinc-rich coating with proper sacrificial properties the ratio of pigment volume concentration (PVC)/critical pigment volume concentration (CPVC) is >1 [12–17]. The dominant protection mechanism of zinc rich coating at earlier stages of its service life is cathodic protection but as the time passes the electrical contact between the zinc particles and steel substrate rapidly decreases as a result of fast oxidation of zinc particles. The zinc oxide corrosion products creation in the coating matrix results in the decrease of coating porosity and decrease in the coating sacrificial properties through reducing the amount of electrolyte that reaches the steel surface. In this case, at longer exposure times, the coating protection mechanism is a combination of barrier and sacrificial properties [16–20]. So the presence of high amount of zinc particles in the zinc-rich coating cannot guarantee its cathodic protection for a long time in severe environments. Inclusion of high amount of zinc particles increases the coating porosity and therefore corrosive agents easily diffuse into the coating, leading to the rapid loss of the electrical connection among zinc particles and with the carbon steel surface.

Attempts have been performed to enhance the cathodic protection duration of zinc-rich coating through inclusion of different types of fillers and additives. The effect of addition of zinc nanoparticles to the zinc-rich coating on its cathodic protection performance has been studied by Schaefer et al. [21]. In a work done by Arianpouya et al. [22] the effect of addition of a mixture of nanozinc and nanoclay on the zinc-rich coating protection performance was reported. They revealed that inclusion of a small quantity of nanozinc and nanoclay particles could remarkably enhance the coating performance through improving both sacrificial and barrier properties of the zinc-rich coating, simultaneously. Jagtap et al. [23] reported significant improvement of the barrier properties and service life of zinc-rich coating after addition of small amounts of zinc oxide particles. In another study Zhang et al. [24] reported the beneficial role of addition of the modified silicon-based vehicle and lamellar Zn (Al) pigments on the zinc rich coating protective properties. The effect of nano-size alumina modified by polypyrrole on the zinc-rich coating corrosion resistance enhancement has been reported by Gergely et al. [25]. In our previous studies the effects of addition of lamellar aluminum and iron oxide pigments [13] and surface modified aluminium nanoparticles [26] on the corrosion protection performance of the epoxy zinc-rich coating have been evaluated. In other studies carbon nanotube [27], polyaniline clay (PANIc) nanocomposites [28], nanoclay [29], propargyl alcohol impregnated mesoporous titanium dioxide [30] and zinc powders [31] modified with organosilanes were added to the zinc-rich coating to enhance its protection perfor-

mance. In all of these studies the improvement of corrosion protection performance of zinc-rich coating has been reported.

An attractive option for improving the zinc rich coating corrosion performance is inclusion of an advanced nanomaterial based on graphene oxide. GO is a single-atom-thick sheet of hexagonally arrayed sp^2 -bonded carbon atoms with many functional groups i.e. hydroxyl, carbonyl, carboxyl and epoxy groups [32–36]. These are reactive sites for covalent and/or non-covalent functionalization with organic and/or inorganic compounds, leading to the improvement of the interfacial interactions between the graphene oxide nanosheets and polymers [35–41]. It has been demonstrated in recent studies that inclusion of functionalized GO nanosheets into the polymeric coatings remarkably enhances their corrosion protection performance. GO sheets are impermeable against water, oxygen and ions diffusion and provide a good barrier as a result of their high surface area [42–58]. So this nanomaterial would be a good candidate for enhancing the protection performance of a zinc-rich coating. In addition, covalent functionalization of GO can be done by conducting polymers (CPs) which have received a considerable attention in recent years because of their unique electrochemical properties. It has been shown that CPs can effectively protect the metals from corrosion through passivation mechanism [59,60]. Armelin et al. [61] and Akbarinezhad et al. [28] have shown the beneficial role of PANIs on the corrosion protection performance improvement of the zinc-rich coating. However to the best of our knowledge there is no report on using GO and GO-PANI nanosheets on the corrosion protection performance of zinc rich coating.

In this study the influence of GO and GO-PANI nanosheets on the corrosion protection performance of a zinc rich coating has been studied. First the GO was synthesized using expendable graphite powder and then it was converted to graphene oxide by a Modified Hummers' method. In the next step, the highly crystalline and conductive PANI nanofibers were deposited on the GO surface through an in situ polymerization of aniline in the presence of GO as an oxidant. After characterization of GO and GO-PANI by FT-IR, XRD and HR-TEM analyses, 0.1 wt% GO and GO-PANI nanosheets were separately incorporated into the ZRC. Finally, the corrosion protection performance of the ZRC, GO/ZRC and GO-PANI/ZRC samples were examined by OCP, EIS and salt spray tests.

2. Experimental

2.1. Raw materials

The zinc dust with average particle size, oil absorption and density of $5\ \mu m$, $6.7\ g\ 100\ g^{-1}$ pigment, $6.95\ g\ cm^{-3}$, respectively was prepared from Iran Zinc Powder Co. Graphene oxide nanosheets were obtained through a modified Hummer's method from an expandable graphite powder, Kropfmuehl Graphite Co. Germany. The expansion rate and grain size of the graphite used were $350\text{--}700\ cm^3/g$ and $80\% >300\ \mu m$, respectively. Sulfuric acid (Merck 98%, pure), zinc nitrate (Merck), sodium nitrite (Merck), potassium permanganate (Merck), hydrogen peroxide (Merck) and dimethylformamide (DMF, Merck Co.) were used without further purification. The steel panels with dimension of $10\ cm \times 8\ cm \times 0.2\ cm$ and chemical composition (wt%) of: 0.05% S, 0.5% Mn, 0.12% C, 0.3% Si, 0.045% P and balanced Fe) were prepared from Foolad Mobarakeh Co (Iran). Epon 828 as a solvent free epoxy resin and Epikure F205 as a polyamine hardener were purchased from Shell, USA and Kian Co., Iran, respectively.

2.2. GO and GO-PANI synthesis procedures

GO nanosheets were synthesized through modified Hummer's method [62] from expandable graphite (EG). In the first step 1 g

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