Journal of Hazardous Materials 357 (2018) 475–482

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

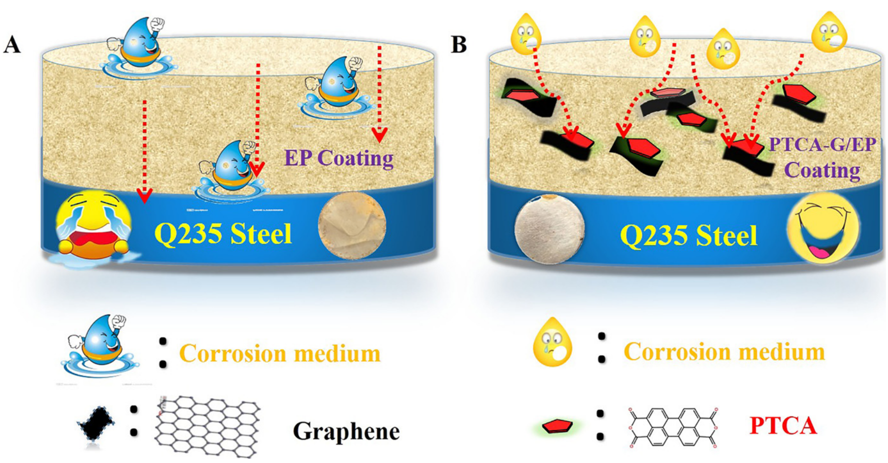
Enhancement of the corrosion resistance of epoxy coating by highly stable 3, 4, 9, 10-perylene tetracarboxylic acid functionalized graphene

Tao Yang[a,b,1], Yanan Cui[a,1], Zeshan Li[a], Hui Zeng[b], Shizhong Luo[a], Weihua Li[b,*]

[a] *Key Laboratory of Sensor Analysis of Tumor Marker of Education Ministry, College of Chemistry and Molecular Engineering, Qingdao University of Science and Technology, Qingdao, 266042, China*

[b] *College of Chemical Engineering and Technology, Sun Yat-sen University, Zhuhai, 519082, China*

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
3, 4, 9, 10-Perylene tetracarboxylic acid
Graphene
Electrochemical measurements
Corrosion protection

ABSTRACT

In this paper, the 3, 4, 9, 10-perylene tetracarboxylic acid-graphene (PTCA-G) composite was synthesized and the corrosion protection property of epoxy coating-coated Q235 steel containing PTCA-G composite was investigated. The results of Fourier transform infrared spectroscopy (FT-IR) proved that 3, 4, 9, 10-perylene tetracarboxylic acid (PTCA) and graphene (G) were combined via π-π interactions and hydrophobic forces between PTCA and G. The results of electrochemical tests indicated that with additives of sole PTCA, sole G and PTCA-G composite, the corrosion resistance of epoxy coating was increased compared with pure epoxy coating. And the most significant improved corrosion resistance of PTCA-G/epoxy coating might be attributed to the good dispersion and barrier performance of PTCA-G composite in the epoxy coating. Besides, compared with the corrosion protection property of PTCA-G/epoxy coating with other volume ratios, the corrosion resistance of epoxy coating containing PTCA-G composite with 10:4 vol ratio of PTCA and G was the best. It might be attributed to the excellent barrier and dispersion properties of PTCA-G composite with 10:4 vol ratio of PTCA and G.

* Corresponding author.
E-mail address: ytlwh666@163.com (W. Li).

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

https://doi.org/10.1016/j.jhazmat.2018.06.038
Received 7 November 2017; Received in revised form 1 June 2018; Accepted 18 June 2018
Available online 19 June 2018
0304-3894/

1. Introduction

In recent years, one of the major concerned problems around the world is corrosion of various metals, which not only leads to degradation of metal structures and mechanical strength, but also results in serious loss of economy and harms to human security [1]. Now the traditional organic coatings are applied to solve this problem [2]. But these coatings always have heavy metal toxic substances, which is not environmental-friendly and harms the human health. Therefore, the development tendency of coatings has transformed into the nontoxic organic and inorganic coatings. And the nontoxic, environmental friendly, cheaper, efficient protective measures and novel protective materials has become the hot spots of current research in reduction and prevention of corrosion.

Many researchers have focused on developing new materials with excellent corrosion resistance property. Graphene (G), as a rising and promising corrosion resistance material, has drew considerable attention. Since its discovery by Konstantin Novoselov and Andre Geim in 2004, due to its low density, super-conductivity, high light transmittance, excellent thermodynamic and chemical stability [3–5], G has been applied in many fields such as fuel cells [6,7], electrochemical biosensors [8–11], optical and electronic components, microwave absorbing materials [12,13]. In recent years, G has also applied to anti-corrosion coatings because of its unique characters, such as impermeability, flexibility, barrier property [14–17]. Prasai et al [18]. researched the influence of G on the corrosion resistance of copper and nickel, on which G was deposited on the surface of copper and nickel immediately by chemical vapor deposition. Zhong et al [19]. employed a special method to grow G on the carbon steel and stated the excellent anti-corrosion property of G on the surface of metals. But the researches also proved that the G coatings only could provide a short-term corrosion protection, which limited its use in corrosion protection coating.

In order to solve this problem, some studies used G-based composites to improve the corrosion resistance of a range of coatings. For example, polyaniline [20,21], carboxylated oligoanilines [16], and other additives [22–33] were added to G to form graphene-based nanocomposites and prepared a range of coatings. Zhao and his coworkers [34] have studied corrosion protection behaviors of functionalized G enhanced polyurethane nano-composite coatings. Liu et al. [16] investigated the corrosion protection property of carboxylated aniline trimer derivative -functionalized graphene to epoxy (EP) coating. Besides, our group [35] also researched the corrosion resistance of epoxy coating containing sulfonated aniline trimer-graphene oxide composite. It was discovered that the corrosion protection behavior of epoxy coating with sulfonated aniline trimer-graphene oxide composite was significantly improved. Although these composites modified coating can enhance the corrosion resistance of substrate, the preparation methods of composites are complex and the binding force of composites may be not strong due to the linear polymer [36,37]. In order to solve this problem, novel modification reagent with facile preparation procedure and simple molecular structure should be applied to modify G. We noticed that Huang's group [38] had prepared highly stable graphene oxide by adopting dopamine modified graphene oxide. So, we can infer that the molecule with plane and aromatic ring conjugate structure might be applied to modify G and improve the performance epoxy.

In this study, we chose 3, 4, 9, 10-perylene tetracarboxylic acid (PTCA) to functionalize G. PTCA is a bifunctional organic molecule possessing four carboxyl groups and an aromatic pyrenyl group [39–41]. Niu et al. [42] prepared PTCA-G super-molecular nanocomplex for an ultrafast supercapacitor. Li et al. [43] prepared chemically converted graphene/3,4,9,10-perylene tetrcarboxylic acid/Au-ionic liquid composites through a green chemical method.

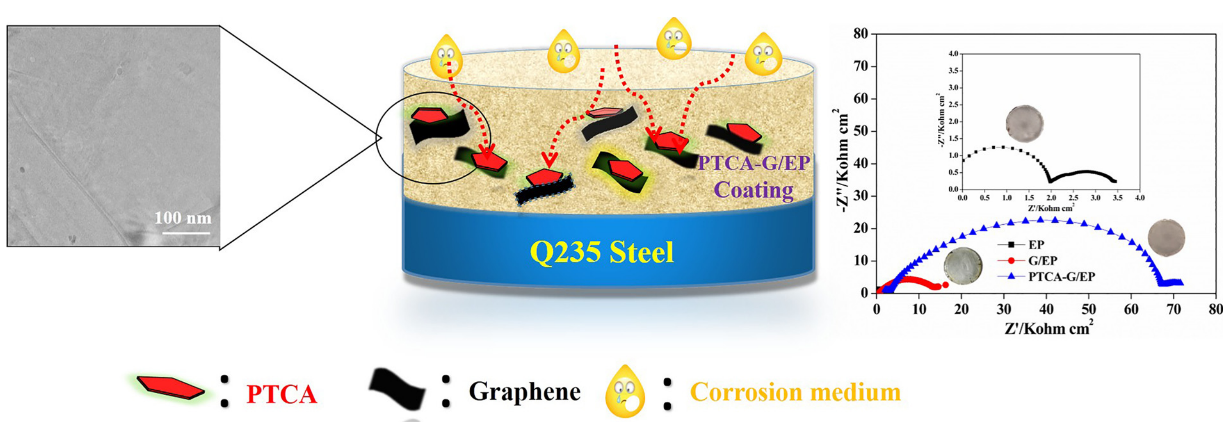
Here, PTCA-G was obtained by mechanically ultrasonicating and mixing. Then, a succession of epoxy with different concentration of PTCA-G, G and PTCA was prepared. The morphology and structural features were characterized by transmission electron microscopy (TEM), scanning electron microscopy (SEM), fourier transform infrared spectroscopy (FT-IR). In addition, anti-corrosion properties of coatings were detected by electrochemical technology. At last, anti-corrosion mechanisms of epoxy without and with different additives were speculated by the experimental data (Scheme. 1).

2. Experimental

2.1. Reagents and apparatus

3, 4, 9, 10- perylenetetracarboxylic dianhydride (PTCDA ≥ 97%) was purchased from SIGMA-ALDRICH, Co., 3050 Spruce Street, (St. Louis, Mo, USA); N, N-dimethylformamide (DMF, 99.5%) and ethyl alcohol (≥99.7%) were procured from Fuyu Fine Chemical Co., Ltd. (Tianjin, China); Epoxy coating (E-44) and corresponding curing agent Polyamide Resin were obtained from Danbao resin Co., Ltd. (Zhenjiang, China); Graphene was obtained from Jining Lite Nano technology Co., Ltd., China; Sodium Hydroxide (NaOH) was purchased from Tianjin Bodi Fine Chemical Co., Ltd., China. All the reagents were of analytical grade and used directly as received. All deionized water was obtained from a water purification device (Aquapro ultrapure water system, Ever Young Enterprises Development. Co. Ltd., Chongqing, China).

All the substance needed stripping and washer was ultrasonicated in KQ-500B ultrasonic cleaners (Kunshan ultrasonic instrument co., LTD, China). The change of functional groups the pre and post chemical modification were received by FT-IR spectra from Tensor 27 FT-IR spectrophotometer (Bruker Company, German). The morphology of sole graphene and PTCA-G were characterized with the SEM (JSM-6700 F machine, JEOL, Tokyo, Japan) and TEM (JEM-2100 machine).



Scheme 1. Schematic of PTCA-G/EP during corrosion process in 3.5% NaCl solution.