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Effects of multiwall carbon nanotube addition on the corrosion resistance and underwater acoustic absorption properties of polyurethane coatings

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ABSTRACT

In this study, the effect of multiwall carbon nanotube (CNT) addition on the anticorrosion and underwater acoustic absorption properties of polyurethane (PU) coatings was studied. The corrosion and mechanical properties of various coatings were characterized through electrochemical impedance spectroscopy, polarization curve measurement, salt spray tests, pull-off tests, and dynamic mechanical analysis. Underwater acoustic absorption properties were evaluated using an underwater acoustic impedance tube system comprising two hydrophones, a sound and vibration data acquisition module, a power amplifier, and a speaker. The results indicated that the corrosion resistance and adhesion strength of the PU coating was enhanced by adding 0.4 wt% CNTs. When additional CNTs were added (1 wt%), however, the corrosion rate of the coating increased. Measurement of the underwater sound absorption coefficient revealed that the underwater acoustic absorption properties of the PU + CNT composite coatings increased as the CNT content increased to 1 wt%.

1. Introduction

Corrosion is a widely observed phenomenon in metallic substrates. Surveys conducted by NACE International in 2016 have revealed that the global cost of corrosion is US$ 2.5 trillion, which is equivalent to approximately 3.4% of global gross domestic product. Unlike weather-related disasters, corrosion can be controlled and prevented. For several years, numerous corrosion protection treatments, such as anodization, metal plating, and organic coating have been used to protect metallic substrates. Among these treatments, paint and organic coatings are the most prevalent and economical methods used in daily life. Organic coatings are generally used to prevent the corrosion of metallic substrates. The primary function of organic coatings is to provide a physical barrier between the metal substrate and a corrosive environment. However, organic coatings are subject to corrosive substances, such as Cl^-, O_2, and H_2O. The presence of water molecules at the coating–substrate interface may induce the electrochemical corrosion of metal under the coating, which reduces coating adhesion. Consequently, adding inorganic additives to the coating is considered to be an effective method for improving organic coating permeability [1–11]. For example, Shi et al. examined the influence of SiO_2, Zn, Fe_2O_3, and halloysite clay nanoparticles on the anticorrosion performance of an epoxy coating. They reported that the incorporation of a small amount of nanoparticles (1% of the total weight of resin and hardener) into the epoxy coating considerably reduced the corrosion rate of the epoxy-coated steel, and Fe_2O_3 and halloysite clay nanoparticles provided the optimal results [1]. Yen et al. revealed that well-dispersed nonconductive clay in polyaniline could increase the length of diffusion pathways for reactive gases, such as oxygen and water molecules, in polymer coatings. Therefore, the diffusion of oxygen and water in a nanocomposite coating was slower than that in a neat polymer coating [5]. Several carbon derivatives, including graphene (Gr), graphene oxide (GO), reduced graphene oxide (rGO), and carbon nanotubes (CNTs) have been considered to be optimal anticorrosive additives [12–22]. For example, Li et al. revealed that the protective properties of composite coatings were considerably improved by the addition of 0.2 wt% silanized GO [14]. Yu et al. prepared rGO-modified ZnAl-layered double hydroxide nanofillers used in epoxy coatings, which exhibited satisfactory corrosion resistance performance [18]. CNTs have received substantial attention because of their extraordinary electrical and physical properties. The addition of CNTs to a polymer coating can enhance the anticorrosion performance of the coating [23–31]. Song et al. reported that well-dispersed multiwall carbon nanotubes (MWCNTs) in a waterborne acrylic coating can reduce through-porosity in the acrylic matrix, increase the length of an ionic conduction pathway, and substantially enhance the anticorrosion properties compared with that of a neat acrylic coating [24]. Shen et al. introduced MWCNT-reinforced epoxy resin, which exhibited excellent adhesion

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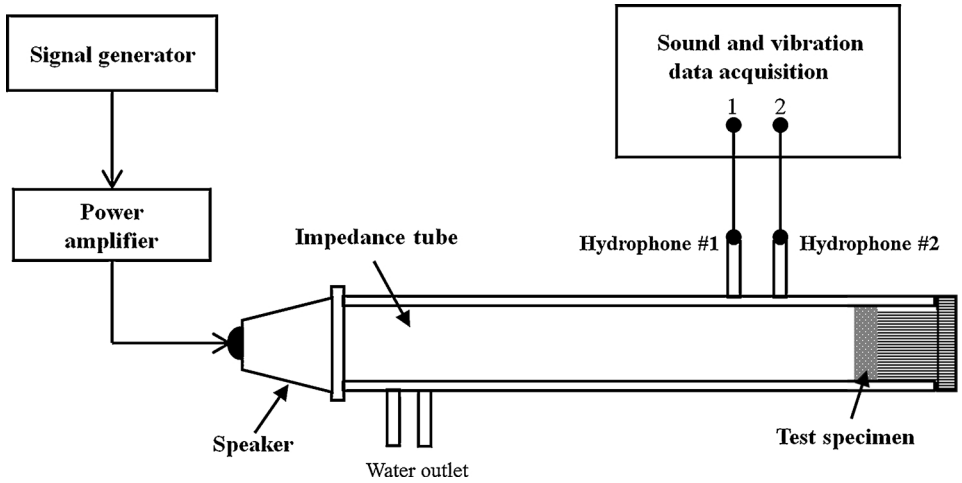

Fig. 1. Schematic of underwater acoustic absorption measurement.

and provided marked corrosion protection to substrates. They revealed that hybrid coatings exhibited the highest anticorrosion efficiency when the MWCNT content was 2 wt% [25]. Deyab reported that CNT addition improved the corrosion resistance of alkyd resin films, and no blisters, pinholes, or delaminations were observed for alkyd resin containing 0.5% CNTs [26]. Khun et al. prepared epoxy coatings containing MWCNTs and found that coating impedance increased with an increase in MWCNT content [27]. Rahman and coworkers demonstrated that adding 2.0 wt% functionalized MWCNTs considerably improved the degradation and corrosion protection efficiencies of the coating [30]. Wei et al. revealed that a protection efficiency of 97.70% was obtained when polyurethane nanocomposite coatings were contained MWCNTs [31].

The incorporation of CNTs into polymers exhibited excellent results in improving the mechanical properties of polymers, such as electrical conductivity [32], tribological properties [19,27], thermal stability, and mechanical strength [33,34]. Moreover, a few studies have recently revealed that acoustic absorption characteristics can be modified by adding inorganic and organic additives into polymer composite materials [35–37]. Bandarian et al. revealed that the incorporation of CNTs can increase the value of the absorption coefficient and improve the acoustic damping of polyurethane (PU) foam [35]. Verdejo and coworkers reported that the incorporation of only 0.1 wt% of MWCNTs can considerably improve acoustic damping [37]. Therefore, incorporating CNTs into polymer materials can simultaneously influence anticorrosion and acoustic absorption properties. The incorporation of particles into polymer coatings to enhance their anticorrosion and mechanical properties has been studied [1,7,12,19,25,27,31]. However, according to our review of the relevant literature, few studies have been conducted on anticorrosion and acoustic absorption properties of PU coatings filled with CNTs. For future applications, understanding the correlation between CNT addition and the corrosion and acoustic absorption performance of PU composite coatings is necessary. An understanding of the aforementioned factors is expected to provide guidance for the design of high-performance coatings with novel multifunctional properties for use in new applications, such as preventing corrosion of the surfaces of underwater vehicles. In this study, a PU composite coating was developed using CNTs as an additive, and the effect of CNT addition on the corrosion resistance and acoustic absorption properties was observed.

2. Material and methods

2.1. Materials

In this study, commercial PU resin and hardener were purchased from Yung Chi Paint & Varnish Manufacturing Co., Ltd. (Kaohsiung, Taiwan) and mixed at a weight ratio of 3:1. The CNTs were provided and dispersed by lLex Nanocoat Technologies Co. (Taipei, Taiwan). The average diameter and length of the CNTs were 20 nm and 10 μm, respectively. CNT powder with four concentrations (0, 0.4, 0.7, and 1 wt %) was mixed with a dispersant, which was then added to a base matrix. The particles were dispersed using a high-speed agitator bead mill.

Phosphated steel (R-46-I, Q-LabCo., USA) was used as the substrate. The substrates were cleaned in an ultrasonic bath with acetone for 10 min, air-dried, and subsequently coated with PU coatings containing 0, 0.4, 0.7, or 1 wt% CNTs using wire wound lab rod, which were designated as PU, 0.4CNT, 0.7CNT and 1CNT, respectively. The prepared coating samples were cured at room temperature overnight and were then postcured at 80 °C for 1 h in an oven to ensure that they were completely dry before taking measurements. The dry film thickness of all coatings was controlled in the range of 45–55 μm, measured using an Elcometer 456 dry film thickness gauge.

2.2. Corrosion resistance measurements and surface morphology analysis

Electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization curve measurements were performed in a 3.5 wt% NaCl solution using a Gamry Reference 600 potentiostat. A standard three-electrode system comprising a graphite counter electrode and a saturated calomel electrode (SCE) reference electrode was used in all electrochemical tests. The EIS measurements were recorded at open circuit potential (OCP) in a frequency range of 10^5–10^{-2} Hz by utilizing an alternating current amplitude of 10 mV. The potentiodynamic polarization curve measurements were obtained by sweeping the potential from −0.2 to 0.5 mV versus OCP at a scan rate of 0.5 mV/s. The corrosion potential (E_{cor}) and corrosion current density (i_{cor}) were determined through the Tafel extrapolation method. The tested area on the coating samples for all electrochemical tests was 7.6 cm^2. Moreover, the corrosion protection performance of various coating samples was evaluated through salt spray tests with 504 h exposure to a salt fog (5 wt%) environment in accordance with the ASTM-B117 standard [38]. The surface morphology of the coating samples was investigated through field emission gun scanning electron microscopy (FEG-SEM, JEOL-JSM-7800) by using facilities provided by National Taiwan University.

2.3. Adhesion tests

Pull-off adhesion tests were performed in accordance with the ASTM D4541-09 standard [39] to evaluate the adhesion strength of PU and PU + CNT composite coatings. An adhesion tester (PosiTest, USA)