



Finite element analysis of the water diffusion behaviour in pigmented epoxy coatings under alternating hydrostatic pressure

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ABSTRACT

The water diffusion behaviour in pigmented epoxy coatings under alternating hydrostatic pressure (AHP) has been calculated by the finite element method. The water diffusion in the coating under AHP follows the non-Fickian diffusion, which is caused by cracks and superposed by the occurrences of many independent Fickian diffusion processes. The cracks largely contribute to the water absorption of the coating. If the occurrence time of the cracks is measured by the experiment, the lifetime of the coating under AHP can be quickly determined.

1. Introduction

Epoxy resin based organic coatings have been widely and effectively used in marine environments to protect structure materials from corrosion [1–3]. Unfortunately, organic coatings are confronted with the tough challenges and dramatic deterioration served in deep-ocean environments because of significant water uptake. The diffusion of water marks the start of coating failure and it has been demonstrated to induce swelling [4], cracking [5] and so on. In order to solve these problems, a lot of researchers have studied the behaviour of water diffusion in organic coatings.

For Fickian diffusion, many researchers discussed the free volume in a polymer which contributed to water diffusion in coatings [6–8]. Wong and Broutman [9] draw attention to the water diffusion mechanism in epoxy resins and proposed a model where epoxy network was consisted of two regions in which water molecules possessed different mobilities by the differences of free volume.

However, the non-Fickian diffusion is more common in the water diffusion process of coatings, because of the swelling and cracking of the coatings [10–12]. Van Westing et al. [13] proposed the swelling coefficient to adjust the ideal Fickian relation for water-epoxy system. After that Nguyen et al. [10] and Zhu et al. [14] studied the transition from Fickian to non-Fickian behaviour during water uptake. They suggested that the diffusion was rate-determined first in the ideal Fickian sorption then the amorphisation crystallisation of the polymer

started and water transport processes got disordered inducing non-Fickian sorption. Based on the studies of Berens [15] and Van Westing et al. [13], Reichinger et al. [16] pointed out the substrate-dependent water transport with a focus on the polymer-pigment coating by quantifying the transport rates and found that the diffusion rate of the pigmented coating on steel was five times slower than the clear coating.

In deep-ocean environments, one of the principal determinants of the coating failure is the high hydrostatic pressure (HP) [17–20]. The coupled effects of water diffusion and stress lead to the premature failure of the coatings [12]. In recent years, a series of studies on water diffusion of epoxy coatings under HP and alternating hydrostatic pressure (AHP) have been performed [21–23]. Liu et al. [21] have studied the water diffusion behaviour of the epoxy coating under HP and indicated that its ability of water absorption is enhanced by high HP. While under AHP, the water diffusion becomes more complex and is totally different, because the cracks in the coating contribute largely to the water absorption [24]. Meanwhile, AHP also makes the water diffusion coefficient of the coating change frequently [25]. Therefore, it is difficult to describe the water diffusion process under AHP by a simple equation originating from the theories proposed by Berens and Van Westing [13,15]. A two-Fickian-stage absorption curve decomposed into two simultaneous or successive Fickian sub-curves is able to provide useful insights into the mechanism of water diffusion within organic coatings [26] and it was used in this study.

Due to the complex structure inside epoxy coatings and variable

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HPs, it is helpful to solve the water diffusion process in the organic coating under AHP by means of the finite element method (FEM). Numerical investigations have been done with the FEM to study Fickian and non-Fickian process in composites [27–29]. Gagani and Echtermeyer [30] studied the effect of cracks on water diffusion of the glass fiber/epoxy composite by FEM and found that the cracks on the external layer of the sample increase the rate of water uptake, while cracks in the internal layer do not influence diffusivity. However, the cracks in their simulations were caused by tensile stress, which is different from HP in deep-sea environments. The nature of water diffusion under AHP is still ambiguous, because the high HP limits the in-situ observation of the changes of coating morphology and the in-situ measurement of the water uptake. In this study, a detailed description on the non-Fickian behaviour of water diffusion in epoxy coatings under AHP was made by FEM according to Wong's model and Coniglio's idea. Based on the model, the lifetime of a coating under AHP can be quickly determined.

2. Experiment

2.1. Sample preparation

The epoxy glass flake (EG) coating consisted of E-44 epoxy resin (biphenol A, Wuxi Resin Factory, China) as the binder, polyamide (TY650, Tianjin Yanhai Chemical Co., Ltd., China) as the hardening agent, dimethylbenzene as the solvent, and glass flake (produced by Wen'an Huaxing Glass Flake Factory, China) as a single component pigmented (thickness: 2–5 μm, diameter: 30–40 μm). They were mixed in a weight proportion of 1:0.8:0.3:0.3 for stoichiometric reaction, and then stirred using a commercial magnetic stirrer machine (Thermostat magnetic stirrer hotplate 85-2 of Guohua Electric Appliance Co., Ltd.) for 2 h to mix sufficiently. Finally, the coating was left for 0.5 h to make them partially cure before painting.

The free film sample was made by brushing the coating on a silica gel plate. After being cured in an oven at 40 °C for 4 h and 60 °C for 2 h, the film was peeled off from the plate and cut into the dimension of 20 mm by 20 mm by 0.2 mm for gravimetric experiment. Subsequently, the film was cured at 60 °C for 20 h and room temperature (25 °C, 30% RH) for 7 days. The coating thickness was measured by a portable electronic gauge (PosiTector 6000 of Defelsko) and the average thickness of 200 ± 10 μm was obtained according to ISO 2808-2007 [31]. Prior to gravimetric tests, samples were stored in a desiccator to keep dry and avoid any change in properties due to adsorption of moisture from the atmosphere.

2.2. Experimental setup and gravimetric tests

The immersion experiments were separated in two parts according to the different experimental environments (HP and AHP). For the HP environments, the experiments under 0.1 MPa were conducted in atmospheric pressure. While for the HP of 3.5 MPa and AHP, the experiments were carried out in a specially designed deep ocean simulation system, shown in Fig. 1. The high HP was obtained by pumping high purity nitrogen into the autoclave, which was controlled by a pressure valve.

AHP was obtained by the instant alternation between HP at 3.5 MPa (equal to 350 m depth HP in the ocean) and atmospheric pressure at 0.1 MPa. Each alternating pressure cycle (12 h) consisted of high static pressure for 6 h and atmosphere pressure for 6 h, and started from 0.1 MPa. The testing solution was de-ionized water and the temperature kept 25 ± 1 °C.

Free films were immersed under the HP of 0.1 MPa and 3.5 MPa, and the AHP of 3.5 MPa, respectively. After removal from the test solution, each sample was dried quickly with a filter paper. Then, the mass gain was measured using a Sartorius MC5 microbalance (1 μg resolution) at different immersion time. Three replications were needed

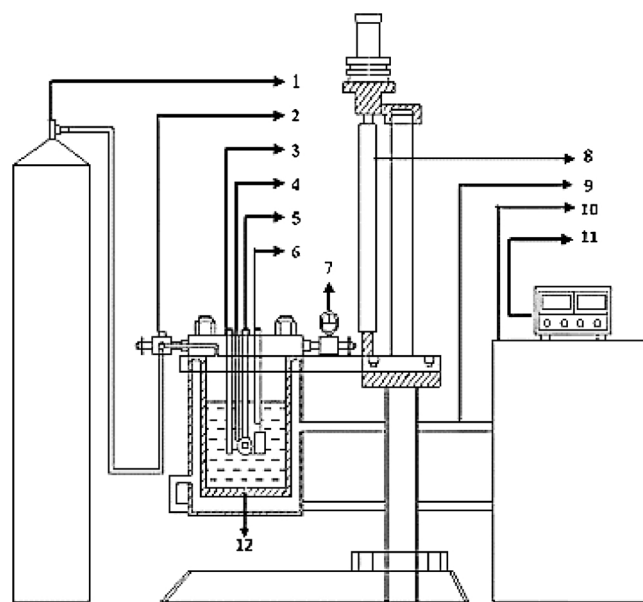


Fig. 1. Schematic diagram of the automatic deep ocean simulation device: 1) nitrogen cylinder, 2) valve, 3) solid reference electrode, 4) thermocouple, 5) working electrode, 6) counter electrode, 7) pressure controller, 8) automatic elevator, 9) circulation line, 10) temperature controller, 11) temperature measuring and 12) autoclave.

for each test and the average of the water absorption was calculated according to the following equation [32]:

$$Q_t = \frac{m_t - m_0}{m_0} \times 100\% \quad (1)$$

where m_t is the mass of free film at time t , m_0 is the initial mass before immersion and Q_t is the water absorption (mass%) at time t .

Different models have been developed to describe the water diffusion process inside materials [33–36]. If it follows an ideal Fickian process, water absorption data for a plane sheet geometry is represented by the equation [2,37,38]:

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left[-\frac{(2n+1)^2 D \pi^2 t}{l^2}\right] \quad (2)$$

where M_t is the mass of absorbed water, which equals to $m_t - m_0$, at time t , M_∞ is the mass of water absorbed at saturation, l is the thickness of the free film, D is the diffusion coefficient which is considered to be constant during the immersion time. For a free film, when $M_t/M_\infty < 0.6$, the water uptake is proportional to the square root of immersion, and Eq. (2) can be approximated as the following equation [10,39,40].

$$\frac{M_t}{M_\infty} = \frac{4\sqrt{D}}{l\sqrt{\pi}} \sqrt{t} \quad (3)$$

3. Water absorption modelling

In this work, COMSOL multiphysics was applied to establish a model to satisfy the experiment results of the water diffusion in the epoxy coating under AHP.

3.1. Water absorption modelling under HP

Due to the variation of the diffusion coefficient and the occurrence of cracks, the model of the water absorption under AHP is difficult to set up directly. Fortunately, the fact that water diffusion under HP follows Fick's law has been confirmed [22]. Thus, the water absorption of the EG coating under different HPs was firstly calculated by Eq. (3),

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