ZEOLITE/POLYANILINE BASED SELF-HEALING AND SILICON OXIDE COATINGS FOR MICROBIALLY INDUCED CORROSION INHIBITION

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“To my beloved ones”

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ABSTRACT

Microbially induced corrosion (MIC) occurs due to the presence of microorganisms such as bacteria, which form biofilms on the metal surface that can cause corrosion. Among the different methods that have been used to protect against MIC, coating has gained more attention because of its ease of application, low-cost and high effectiveness. Recent research has shown that self-healing coatings concept based on releasing healing agent when micro-cracks are initiated in the coating surface and hydrophobic silicon oxide based organic and inorganic coatings have great potential for use as antifouling coating. The aim of this research is to investigate the effects of self-healing and silicon oxide (SiO) coatings on inhibiting MIC in saline environment. The self-healing coating was prepared via interfacial polymerization of zeolite, polyaniline, and zeolite/polyaniline composite and then encapsulated with urea fomaldehyde as a shell material to form the microcapsules and embedded in epoxy to form coating material which was applied on mild steel substrate. The SiO coating, on the other hand, was deposited on mild steel substrate using radio frequency (RF) magnetron sputtering physical vapor deposition (PVD) method with different parameters of RF power, temperature, pressure and deposition time in order to achieve optimum parameters based on minimum surface roughness and good adhesion. The surface topography and roughness were examined by atomic force microscope (AFM), while the thickness and morphology of the coatings were observed using field emission scanning electron microscope (FESEM) equipped with energy dispersive spectrometer (EDS). The adhesion test was performed using nano scratch test for SiO coating and Pull off test for self-healing coating and supported by Rockwell C test. The corrosion behavior was investigated through salt spray test for 28 days and immersion tests in nutrient rich simulated seawater (NRSS) medium with *pseudomonas aeruginosa* bacteria for 70 days. The *Tafel* electrochemical test and electrochemical impedance spectroscopy (EIS) was performed on both bare and coated steel samples. AFM results clearly revealed that by varying the sputtering parameters has a strong influence on the surface roughness of the deposited SiO coating in which its thickness varied between 30 nm to 50 nm. The thickness for self-healing coating was between 50 µm to 175 µm. From the adhesion results, both coating methods produced superior adhesion on steel substrates. Fourier transform infrared spectroscopy (FTIR) results show the successful encapsulation of the three synthesized materials. The total self-healing occurred after the release of the core material when the capsule was ruptured after 21 days left at room temperature. The specimen exposed in salt spray chamber exhibited excellent corrosion resistance for all investigated coating materials, while, the specimens immersed in NRSS medium with *pseudomonas aeruginosa* bacteria showed varying anti-corrosion properties. *Tafel* results show that the lowest corrosion rate was observed for SiO coating with a value of 0.219 mm/yr, followed by encapsulated zeolite/polyaniline composite self-healing embedded in epoxy of 0.334 mm/yr. EIS results show that among all the coatings, encapsulated zeolite/polyaniline composite self-healing embedded in epoxy coating has the highest impedance modulus (Z) at a frequency of 0.01 of 7800 ohms. In conclusion, zeolite/polyaniline composite self-healing coating is the best among all the coating materials which shows superior anticorrosive and MIC inhibition property.

ABSTRAK

Kakisan dipengaruhi mikrob (MIC) berlaku disebabkan oleh kehadiran mikroorganisma seperti bakteria, yang membentuk biofilem pada permukaan logam yang boleh menyebabkan kakisan. Terdapat kaedah berbeza yang telah digunakan untuk melindungi permukaan daripada MIC, kaedah salutan yang menjadi perhatian kerana mudah untuk digunakan, kos yang rendah dan keberkesanan yang tinggi. Kajian terkini menunjukkan bahawa konsep penyembuhan sendiri salutan melalui pelepaskan ejen-ejen penyembuhan apabila retakan mikro bermula di permukaan lapisan dan silikon oksida (SiO) hidrofobik berasaskan lapisan organik dan bukan organik mempunyai potensi yang besar untuk digunakan sebagai salutan anti-cemar. Tujuan kajian ini adalah untuk mengkaji kesan salutan penyembuhan sendiri dan salutan silikon oksida (SiO) bagi menghalang kesan MIC dalam persekitaran masin. Lapisan penyembuhan sendiri telah disediakan melalui pempolimeran antara muka dengan menggunakan zeolit, polyanilina dan komposit zeolit/ polianilina dan kemudian disalut dengan urea formaldehid sebagai cengkerang untuk membentuk kapsul mikro yang dimasukkan ke dalam epoksi untuk membentuk salutan yang digunakan pada permukaan keluli. Manakala lapisan SiO telah disadur pada permukaan keluli sederhana menggunakan frekuensi radio (RF) kaedah pengenapan wap fizikal (PVD) permercikan magnetron dengan parameter kuasa RF, suhu, tekanan dan masa pemendapan untuk mencapai parameter optimum berdasarkan kekasaran permukaan minimum dan lekatan yang baik. Topografi permukaan dan kekasaran telah diuji dengan mikroskop daya atom (AFM), manakala ketebalan dan morfologi lapisan dianalisis menggunakan mikroskop elektron imbasan medan pancaran (FESEM) yang dilengkapi dengan spektroskopi tenaga-serakan sinar-x (EDS). Ujian rekatan dilakukan dengan menggunakan ujian calar nano untuk lapisan SiO dan ujian tarikan untuk salutan penyembuhan sendiri dan disokong dengan ujian kekerasan *Rockwell C*. Kadar kakisan ditentukan melalui ujian semburan garam selama empat minggu dan ujian rendaman di dalam larutan nutrien air laut simulasi (NRSS) dengan *Pseudomonas aeruginosa* bakteria selama 10 minggu. Ujian elektrokimia *Tafel* dan spectroskopi impedans elektrokimia (EIS) telah dilakukan ke atas kedua-dua sampel keluli dan keluli tersalut. Keputusan AFM jelas menunjukkan bahawa dengan parameter pemercikan yang berbeza-beza memberi pengaruh yang besar ke atas kekasaran permukaan salutan SiO yang dienapkan dengan tebal yang berbeza diantara 30 hingga 50 nm. Ketebalan untuk lapisan pemulihan sendiri adalah antara 50 hingga 175 mikron. Daripada keputusan ujian rekat, kedua-dua kaedah salutan telah menghasilkan rekatan yang baik pada substrak keluli. Spektroskopi Fourier penjelmaan inframerah (FTIR) menunjukkan keputusan terbaik bagi ketiga-tiga bahan mikro kapsul yang disintesis. Penyembuhan penuh terjadi selepas pembebasan bahan teras apabila kapsul pecah selepas 21 hari dibiarkan pada suhu bilik.Spesimen ujian semburan garam menunjukkan ketahanan kakisan yang sangat baik untuk semua bahan salutan manakala, spesimen ujian rendaman dalam NRSS dengan bakteria *Pseudomonas aeruginosa* menunjukkan pelbagai ciri-ciri anti-kakisan. Keputusan ujian *Tafel* menunjukkan bahawa kadar kakisan terendah telah diperhatikan untuk salutan SiO dengan nilai 0.219 mm/tahun, diikuti salutan penyembuhan sendiri komposit zeolit/ polianilina di dalam epoksi dengan nilai 0.334 mm/tahun. Keputusan EIS menunjukkan bahawa di kalangan semua salutan, salutan penyembuhan sendiri komposit zeolit/ polianilina di dalam epoksi mempunyai modulus impedans (Z) yang tertinggi pada frekuensi 0.01 daripada 7800 ohms. Sebagai kesimpulan, salutan penyembuhan sendiri komposit zeolit/ polianilina adalah yang terbaik dikalangan semua bahan salutan, iaitu menunjukkan anti-kakisan dan sifat perencatan MIC yang unggul.

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LIST OF ABBREVIATIONS

Al	-	Aluminum
AA	-	Aluminum alloy
ATRP	-	Atom transfer radical polymerisation
Cu	-	Copper
DNA	-	Deoxyribonucleic acid
EPS	-	Extracellular polymeric substances
E	-	Elastic modulus
E_{corr}	-	Corrosion potential
I_{corr}	-	Corrosion current density
IOB	-	Iron oxidizing bacteria
IRB	-	Iron reducing bacteria
MIC	-	Microbial-Induced Corrosion
MOB	-	Manganese oxidizing bacteria
MS	-	Mild steel
N^+	-	Positively charged nitrogroups
NPs	-	Nanoparticles
NPVP	-	Poly (4- vinylpyridine)-co-poly (4-vinyl-N-hexylpyridinium
PANI	-	Polyaniline
PDA	-	Poly (dopamine)
PMOX	-	Poly (2-methyl-2-oxazoline)
PEG	-	Poly (ethylene glycol)
PEO	-	Polyethylene oxide
PFPEs	-	Perfluoropolyethers
PTFE	-	Polytetrafluoroethylene

QASs	-	Quaternary ammonium salts
SI-ATRP	-	Surface initiated atom transfer radical polymerisation
SRB	-	Sulphate reducing bacteria
SEM	-	Scanning electron microscopy
FESEM		Field immision scanning electron microscopy
TBT	-	Tributyltin
EDS		Energy dispersive spectroscopy
Ti	-	Titanium

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CHAPTER 1

INTRODUCTION

1.1 Research Background

In the majority of engineering structures operated in the marine environment, microbiological induced corrosion (MIC) is of great concern [1]. The issue of MIC could be particularly dangerous in marine structures such as ships and maritime platforms as well as offshore jetties and rigs [2]. Such constructions have to be protected against attack from the main components of the marine setting such as sea water, temperature, and biological attack, also referred to as “biofouling”. Indeed, biofouling is the colonization of submerged structure surfaces by organisms such as barnacles, bacteria, and algae. Maritime biofouling is a long-standing and pricey problem for the marine industry because the development of fouling assemblies on ship hulls, for instance, raises drag, decreases maneuverability, and increases fuel use and greenhouse gas release [3-6], leading to both high financial and ecological expenses [7]. Maritime fouling, or the settlement and growth of marine organisms on waterlogged structures, is predicted to have a worldwide expenditure of above $3 billion yearly [8]. Shipping accounts for approximately 90% of global business, and seaborne trade has increased by four-fold during the previous 40 years [9, 10]. Generally, MIC is not a novel kind of corrosion. Most commonly it appears in the shape of localized corrosion, whether pitting or crevice. MIC is instigated, extended, or proliferated because of the existence of microorganisms such as bacteria [11, 12].

Pseodomonas aeruginosa is one of the dominant bacterium found in the maritime surrounding, and is an aerobic slime-forming bacteria that creates a biofilm coat on the surface of the steel. The reaction of the biofilm layer with the steel surface and the generation of differential aeration cells provide conditions on the steel surface that instigate and hasten the corrosion process. The production of these concentration cells is harmful to the integrity of the oxide layer and increases the vulnerability of steels to corrosion [13-15].

Numerous conventional antifouling systems are in the form of paints, which is an inclusive word covering a diversity of substances: lacquers, enamels, varnishes, surfacers, undercoats, primers, fillers, sealers, plugs, and several others. The use of non-environmentally friendly and toxic antifoulants on ship hulls is one of the most widely used techniques of managing fouling even though biocides such as lead, mercury, arsenic, and their natural derivatives are banned owing to environmental concerns. Antifoulants comprise one of the numerous additives typically incorporated in the top-layer paint of a maritime defensive coating system [2].

In recent years, a new technique has shown a great promise for autonomic healing of micro-cracks and mechanical damage, this technique is the use of self-healing polymers [16]. Self-healing coatings are an extremely enhanced group of smart substances in which the aim is to repair the micro-cracks completely, in a passive way, with no necessity for detection or any kind of foreign interference [17-21].

A different method of mitigating against biofouling is the physical vapor deposition (PVD) coating approach, which features a set of diverse methods that can be employed to deposit silicon oxide coating onto steel substrates. PVD comprises several vacuum deposition approaches and is a universal term employed to explain a process that deposits thin films through the concentration of a vaporized form onto diverse substrate surfaces. The basic process of PVD falls into two universal classes: sputtering and evaporation. The commercial applications of PVD methods vary over a broad range of uses from decorative, to extreme temperature superconducting layers [22].

1.2 Problem Statement

MIC of immersed structures in maritime environments is the effect of biological organisms colonization and adhesion on the surface. Given that the significant bio-interfacial processes which lead to fouling are nano-scale or micro-scale in size, the surface properties of the structures to manage biofouling are assumed to be on a similar size scale. An area of specific interest in recent years is the use of nanotechnology in combating MIC. There is a necessity to find ecologically harmless coatings to hinder MIC successfully. Certainly, current research has demonstrated the significant of coating protection with minimal environmental impact of self-healing coatings based on the release of healing agents when micro-cracks are instigated in the coated surface. Silicon oxide-based organic and inorganic coatings in addition to diamond-like carbon coatings have shown great potentials to combat antifouling with fewer negative impacts on the environment. The new methods are based on “fouling release” and “contact killing”. The former method, does not involve the discharge of biocides in maritime water and therefore should be ecologically responsible. The “contact killing” method may be considered a favorable approach, and polycationic coatings are utilized to hinder MIC using this method. Nevertheless, to attain such an objective, the coating should be modified so its surface properties have excellent smoothness and corrosion resistance, high hardness, good thermal stability, and low cost. Zeloite and polyaniline have good adhesion to the steel substrate and only destroy the microorganisms that come into contact with them without leaking out, while silicon oxide owing to its unique characteristics of providing a smooth film which could minimize the adhesion of the microorganism on the steel substrates. Due to their biocide behavior and anticorrosive properties as well as environmental responsiveness, silicon oxide, polyaniline, and zeolite are excellent candidates to protect metal surfaces against MIC.

1.3 Objectives of the Research

The aim of this research work is to investigate on coatings that would be capable of inhibiting MIC. Foremost, the research induces an exploration on the mechanisms of MIC on steel surfaces in bacteria-inoculated medium, which could be helpful to facilitate the use of efficient procedures to mitigate against MIC. Next, the research is also aimed at exploring the capabilities of the encapsulated zeolite, polyaniline, zeolite/polyaniline composite self-healing embedded in epoxy and silicon oxide hydrophobic coatings strategies in inhibiting MIC. The output of this research is anticipated to enhance the properties of MIC inhibition of coated steel that is exposed to bacteria-inoculated medium, and also to provide an environmentally friendly and suitable coating for the mitigation against MIC of steels in the marine environment.

The specific objectives of the research include:

(i) To develop encapsulated zeolite, polyaniline, zeolite/polyaniline composite self-healing and silicon oxide coatings that can be utilized as coating materials to inhibit microbial-induced corrosion.

(ii) To charcterise the properties of newly developed self-healing and silicon oxide coatings on mild steel substrates.

(iii) To evaluate the performance of self-healing and silicon oxide coatings as MIC inhibitive coatings.

1.4 Scope of the Research

The scope of research includes:

(i) Synthesis of the self-healing core substances encompassing polyaniline, zeolite, and zeolite-polyaniline composites and their microcapsules by the in-situ chemical polymerization technique and applying them on steel substrates.

(ii) Deposition of silicon-oxide based coatings on steel substrates with enhanced properties using the physical vapor deposition method.

(iii) Characterization of the properties of synthesised and deposited coatings, including hardness and coating adhesion, using various characterization techniques such as Atomic Force Microscopy (AFM), X-Ray Diffraction (XRD), Field Emission Scanning Electron Microscopy (FESEM), X-ray, Energy Dispersive Spectroscopy (EDS), Fourier Transform Infrared Spectroscopy (FTIR), Electrochemical Impedance Spectroscopy (EIS), and Electrochemical Tafel Analysis.

(iv) Performance of biological assays to assess the antifouling capacities of the developed coatings versus *P. aeruginosa* microorganisms.

(v) Determine the corrosion rate using immersion tests

(vi) Microscopic examination of samples upon completion of immersion tests using standard characterization equipment.

1.5 Significance of Research

Many types of coatings used by the marine industries are toxic and affect the marine life. Therefore, many researchers are investigating on alternative coatings which are friendly to the environment but at the same time are effective on protecting the metal against MIC.

The fundamental goal of this research is to investigate the performance of encapsulated based self-healing and hydrophobic concept by physical vapor deposition coated steel substrates in inhibiting MIC when exposed to bacteria-inoculated medium. Therefore, the outcome of this study would benefit various sectors, particularly the marine, gas, and oil industries.

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