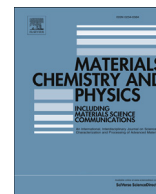




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Glutamine-reinforced silica gel microassembly as protective coating for aluminium surface

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HIGHLIGHTS

- Novel glutamine reinforced silica gel microassembly over aluminium.
- Monolayer and bilayer thin films examined by severe corrosive environment.
- Maximum inhibition efficiency in monolayer was = 98.3% using 1200 ppm at 10 °C.
- The adsorption process demonstrated appropriate Langmuir isotherm fit.
- The bilayer formation comprises firm layer ~99% inhibition for prolonged time.

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ABSTRACT

The present article comprises the formation of novel glutamine reinforced silica gel microassembly over aluminium and the use thereof as promising protective coating. The aluminium sheets were immersed in a glutamine solution and then in silica gel solutions with four different concentrations. Epichlorohydrin as coupling agent and Ca^{2+} ions as crosslinking agents were introduced in dropwise addition. Formation of monolayer and bilayer thin films, verified by inhibition efficiency and surface morphology, demonstrated successful protective coating of aluminium surface against severe corrosive environment for prolonged time. In monolayer, maximum inhibition efficiency obtained was 98.3% at 10 °C using silica gel concentration = 1200 ppm. Higher activation energy, enthalpy and entropy of activation obtained at higher silica gel concentration. The adsorption process demonstrated appropriate Langmuir isotherm fit. The formation of layer-by-layer comprises firm and stable thin films over aluminium surface with inhibition efficiency ~99%. Such coatings performed by building-up construction have some guideline specifics that include simplicity, low cost manufacturing, and superior corrosion protection.

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1. Introduction

The significance of coating surfaces as water-repellent surfaces and/or superior oxidation resistance surfaces has appeared in significant amount of industrial implementations. Their potential uses include but not limited to; self-cleaning and superhydrophobic surfaces [1–10], anti-icing [11–14], anti-fogging [15], and corrosion resistance surfaces [16–20]. On the other hand, corrosion leads to structural failures and huge economic damages in various industries [21–24]. Many techniques employed for resisting corrosion through formation of different coating deposition that include polymer and ceramic coatings [25–32], electro-deposition

materials [33,34], and self-assembled nanocoatings [35,36]. More specifically, layer-by-layer (LBL) technique comprises coating of multiple functionality groups deposited over the surface of the metal. Such simultaneous deposition occurs in cycles and involves stepwise electrostatic deposition of mediator followed by passivation of the surface with polyelectrolyte [37–39]. The layer-by-layer (LBL) deposition gains its significance from high impact inhibition efficiency, low cost, and superior oxidation resistance. Such coating materials possess the capacity to form physical barrier that prevent corrosion electrolyte from penetrating into metal substrate [40–44].

Herein, we report novel layer-by-layer (LBL) glutamine reinforced silica gel microassembly as superior coating material for aluminium surface and its use as promising protective layer-by-layer (LBL) coating. Kinetic and thermodynamic parameters of corrosion process were evaluated and verified. Surface morphology

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pre- and post exposure to severe acidic environment (1.0 M HCl solution) for prolonged period of time were investigated using scanning electron microscopy (SEM) technique.

2. Experimental

2.1. Materials & techniques

Glutamine (Janssen Chimica, 99%), sorbsil-C60-M (40–60 μm) silica gel (May & Baker LTD), epichlorohydrin (Aldrich) have been used as received without further purifications, calcium chloride (CaCl_2) and all other chemicals were of analytical grade and used as received. Aluminium sheets of 99.7% purity and dimensions of $30 \times 10 \times 1.5$ mm were used. The aluminium sheets, pre-assumingly, abraded with emery paper up to 1200 grade, washed with acetone, and dried between two filter paper prior uses. Scanning Electron Microscope (SEM): Samples mounted on the specimen stabs, and coated with gold ion by sputtering method with DSM 950 (ZEISS) model, Polaron (E6100) model. Sartorius 2024 MPb semi-microbalance used for weight measurements with precision of $\pm 10^{-5}$ g and triplicate replications.

2.2. Layer-by-layer (LBL) protective coating over aluminium surface

The aluminium sheets of $30 \times 10 \times 1.5$ mm dimensions were dipped and maintained in $\text{pH} = 10$ aqueous basic solution for 20 min to activate the surface. The activated aluminium sheets were macerated in 500 ppm glutamine solution adjusted to $\text{pH} = 8.5$ for another 20 min. In a parallel experiment, four different concentrations of silica gel solutions were prepared and adjusted to $\text{pH} = 12.5$ aqueous basic solution; 300, 600, 900, and 1200 ppm. Later on, the pH was adjusted to $\text{pH} = 10$, and 50% (w/w) epichlorohydrin coupling agent, with respect to silica gel, added in dropwise addition into the silica gel solution. The solution was allowed for gentle stirring for 20 min. The aluminum sheets covered with glutamine layer were transferred to the Sorbsil silica gel solution. Simultaneously, 20% (w/w) calcium ions (Ca^{+2}) added, used as crosslinking agent, under gentle stirring for 20 min. Experiments were carried out at four temperatures in the range of 283–313 K. After time completion, the aluminium sheets covered with monolayer of glutamine–reinforced silica gel were removed and allowed to dry at ambient temperature. The same procedure was repeated to form bilayer. The formed mono- and bilayers tested and verified via inhibition efficiency and scanning electron microscopy after been exposed to severe 1.0 M HCl corrosive environment for 1 h unless stated elsewhere.

3. Results & discussion

3.1. Monolayer coating

It is known that the outer oxide layer of aluminum surface can interact with water and form hydroxylated aluminium layer. Such hydroxylated aluminium groups could be rather protonated or deprotonated depending on pH value [45]. The zero charge potential for aluminium surface lies between $\text{pH} 6\text{--}9$ [46,47]. Therefore, at $\text{pH} = 10$ the hydroxylated aluminium surface is deprotonated and form negative charge ions (i.e. Al-O^-). On the other hand, glutamine amino acid solution maintained at $\text{pH} = 8.5$ is deprotonated from the α -carboxylic acid and negative charge carboxylate ions are formed. As a result, dipping of the activated aluminium with Al-O^- surface into glutamine solution, result with simultaneous attraction of partially positive hydrogen atoms of the amine groups in glutamine to the negative charge of aluminium surface ions. Consequently, the negative side carboxylate ion of the

glutamine oriented toward the solution as depicted. Furthermore, silanol groups of the silica gel microspheres can be protonated or deprotonated depends on pH value ($\text{p}K_a = 9.84$). Therefore, at $\text{pH} = 10$, almost 50% of the silanol groups of the silica gel microspheres are deprotonated leading to the formation of negative charge surface ions. Such negative ions can dimerize through the interaction with epichlorohydrin coupling agent and form silica gel dimer. The dimer interactions, mediated by epichlorohydrin, continue irregularly until reinforced silica gel clusters formed. Addition of positive charge calcium ions in solution allow immediate crosslinking of reinforced silica gel clusters with each other and with negative charge zwitterion layer over aluminium surface leading to the formation of glutamine–reinforced silica gel monolayer over aluminium surface (Fig. 1).

The firmness of monolayer and bilayer formed on the surface of aluminium were tested by exposure to 1.0 M HCl solution for different time intervals from one to 48 h, and verified by inhibition efficiency and SEM surface morphology. The inhibition efficiency calculated as follows

$$\text{Inhibition efficiency (\%)} = \frac{W_0 - W_i}{W_0} \times 100 \quad (1)$$

where W_0 and W_i were the weight loss of aluminium in absence and in presence of corrosion protecting mono- and bilayers, respectively. The rate of corrosion (R_c) of aluminum determined from as follows:

$$R_c = \frac{W_b - W_a}{St} \quad (2)$$

where W_b and W_a are weights of the aluminum sheet before and after been immersed in 1.0 M HCl solution for 1 h, respectively. The ($W_b - W_a$) was the weight loss (in mg), S was the surface area of the aluminium sheet (in cm^2) and t was the exposure time (in s). Fig. 2A and Table 1 summarized the normalized weight loss of aluminum versus the concentration of reinforced silica gel at different temperatures. The weight loss was inversely proportional to the concentration of reinforced silica gel at the same temperature. Whereas increasing temperature, increases the rate of dissolution of aluminium in HCl solution. On the other hand, the inhibition efficiency of glutamine–reinforced silica gel monolayer in Fig. 2B comprises exponential inhibition increase at higher reinforced silica gel concentration and lower temperatures. The higher inhibition efficiency denotes better-orientated adsorption of reinforced silica gel clusters over aluminium surface and describes successive building up layer-by-layer protective shield over the aluminium surface.

3.2. Surface topology

Scanning electron microscope images describe the morphologic changes that occur over the aluminium surface pre- and post exposure to corrosive 1 M HCl solution. Fig. 3A presents the image of the uncoated aluminium surface after exposure in severe molar HCl solution. Apparently, aluminium surface was damaged and many corrosive punctures appeared all over the surface. On the other hand, Fig. 3B demonstrated the formation of monolayer on the aluminium surface before been exposed to corrosive HCl. Clearly, globular microspheres were compacted and aligned near each other forming protective shield monolayer against the corrosive environment. The examination of monolayer against corrosive 1.0 M HCl environment reveal longitudinal and cross tearing and the formation regular size micro-islands with estimated thickness of ~ 1.5 μm (Fig. 3C and D). The tearing occurs in the weakest points of the monolayer structure as a result of unsuccessful accumulation over the aluminium surface.

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