Enhancement of Barrier and Mechanical Performance of Steel Coated with Epoxy Filled with Micron and Nano Alumina Fillers

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Steel is an essential part of our life as it is used in wide applications as food equipment and heavy construction. The polymeric coating prevents the deterioration of the properties of metals due to rust and corrosion. This study investigated a reinforced polymeric coating to steel for enhancing barrier and mechanical properties. A comparison between different configurations of double-layered polymeric coatings was attained. The results showed that a maximum enhancement of 16.7%, 18.9%, 32.6%, 8.5%, and 5.7% in tensile strength, tensile strain, toughness, flexural strength, and flexural strain, respectively were achieved with a coating of epoxy filled with 1wt% Al2O3 microparticles before 1wt% Al2O3 nanoparticles on both sides as compared with pure epoxy coating. Adding micro/nanoparticles to epoxy coating enhanced the barrier properties of the coated steel against salt solution and citric acid environment as compared to pure epoxy coated steel.

Keywords: Micro/nanocomposites coating, Alumina, Steel, Mechanical properties, Food equipment, Barrier properties.

1. Introduction

Steel as a type of metal has become an essential part of our life as it is used in wide applications like automotive, household appliances, food equipment. The mild steel material was used for food equipment manufacturing purposes because it has good mechanical properties like its strength, ductility, and weldability. Corrosion of steel is considered as a sustained matter that attracted interest as it is the main cause of industrial accidents and also the consumption of metal resources. Metal with polymeric composite barrier liners was used to reduce oxygen and moisture diffusion produced in food packaging. Epoxy resin is one of the widely used polymers utilized to protect mild steel due to its strong adhesion, good chemical resistance, and low shrinkage. To get benefit from its good properties, inorganic particles may improve the stiffness and other properties of the epoxy coatings. Many studies have attempted to improve the toughness of epoxy and other polymers by reinforcing them with micro and nanofiller to promote extrinsic toughening procedures.

Nanocomposite coating is being investigated to enhance barrier properties for steel. Adding nanofillers to epoxy coatings leads to less coating blistering and delamination. This can be attributed to the high capability of fine particles to fill the cavities. Moreover, the transparency of the polymeric coating will not be disturbed with the existence of nanoparticles with dimensions less than 100 nm. The anticorrosion performance and mechanical properties of metals are highly improved by the incorporation of metal oxide nanofillers to epoxy matrices. In addition, alumina (Al2O3) fillers characterized by their high mechanical properties have been widely used to enhance related properties of epoxy matrices. Different nanomaterials such as nanopolymers and nanocomposites are included at various levels in the food industry. The coatings containing Al2O3 nanofillers showed enhancement in mechanical properties as compared to pure polymer coating.

Behzadnasab et al. reported that the simultaneous adding of layered nanoclay and small amounts of APS-treated zirconia nanoparticles to epoxy coatings considerably improves the corrosion resistance of the nanocomposite coatings. The optimum amount of nanoparticles was 1 wt% nanoclay and 1 wt% zirconia nanoparticles. However, other combinations reduce the corrosion performance, due to flaws introduced in the coating film. Similarly, epoxy/Molybdenum oxide nanocomposite coating enhanced the mechanical, adhesion, and anticorrosion properties as compared to epoxy coating. Furthermore, Abd El-Lateef and Khalef reported that the optimum percent of ZrO2-TiO2 is 10wt% but the inclusion of a higher weight
percent of ZrO2 resulted in the formation of a weak film with poor barrier properties. Furthermore, Ramezanazadeh and Attar\textsuperscript{3} stated that the epoxy coating resistance was significantly enhanced using nano and micro-sized ZnO particles. The barrier properties of the nanocomposite were considerably higher than the microcomposite. Golru et al.\textsuperscript{18} studied the impacts of the addition of 1, 2.5 and 3.5 wt\% Al2O3 nanoparticles of an epoxy/polyamide coated AA-1050 on the anticorrosion properties immersed in a 3.5 wt\% NaCl solution and salt spray test. Results revealed that Al2O3 nanoparticles could significantly improve the corrosion resistance of the epoxy coating. The rationale for this study is to create a new protective polymeric coating to steel using microparticles and nanoparticles embedded in an adhesive polymer which is the epoxy matrix. Alumina particles in micron size were used with two percentages (1wt\% and 2wt\%) for their low cost and good mechanical and barrier properties. However, alumina nanoparticles are more expensive, nanocomposite coatings were used with a low percentage (1wt\%) for their high mechanical and barrier properties. Combining both nanoparticles and microparticles embedded in an epoxy matrix to get benefit from both reinforcements. The mechanical and barrier performance of micro/nanocomposite double-layers coated steel with different configurations was evaluated. Three different alternating double-layer coatings were designed. The outer layers for all conditions were 1 wt\% Al2O3 nanoparticles. However, the first layers were differentiated with epoxy, 1wt\% Al2O3 microparticles, and 2wt\% Al2O3 nanoparticles. Mechanical and barrier resistance properties were conducted on the nano/microparticles coated specimens and compared with pure epoxy coated steel.

2. Experimental Work

2.1. Materials

Mild steel was utilized as a substrate provided by Al Ezz-Dekheila Steel Company Alexandria. The chemical composition of the used steel is presented in Table 1. The sheets of steel were cut to the required dimension of the coupons by a laser machine. The coupons were polished to roughen the steel surface. After polishing, the top and bottom side coupons surfaces were cleaned with acetone before coating. Chemicals including sodium hydroxide, citric acid, and acetone were provided by El Nasr Pharmaceutical Chemicals, Egypt. The polymeric coating was Epoxy resin (Kemapoxy RGL150) and was provided by CNB Company, Egypt. The polymeric coating was prepared by adding hardener carefully to the epoxy and mixed thoroughly with a ratio of 1:2 by mass of epoxy matrix. Micro/nanoparticles were added to epoxy by the sonication method. Sonicing was conducted with Hielscher ultrasonic processor UP 200 S. The conditions of sonication were 0.5 cycles per second with an amplitude of 40\% for 2 hrs as recommended by\textsuperscript{26-27}.

To protect the epoxy resin from degradation, micro/nanoparticles with epoxy mixture were cooled by putting it on an ice water bath during operating sonication\textsuperscript{18}. Afterward, the blend and the hardener were mixed together with the recommended ratio. This preparation was performed at a temperature of 25°C. The coating layers were performed on steel by a metallic roller that was used to remove excess resin to reduce void content and any entrapped air bubbles. The polymeric coating on one side of the steel coupon is left for 24 hrs to cure. Subsequently, the second layer on the same side was laid and left for 24 hrs to cure. The same technique was done for the other two layers on the bottom side of the coupons. The quantity (volume) of each layer of epoxy resin and epoxy filled with micro/nanoparticles that was spread during the manufacturing process was the same. This indicates that the thickness of the composite layers is approximately the same. The film thickness of each layer is 110 µm. The final types of micro/nanocoatings on steel substrate were constructed as illustrated in Figure 1.

2.3. Materials characterizations

2.3.1. Tensile test

The tensile properties of the steel coated with micro/nanocomposites coupons were tested according to ASTM D3039. The various test coupons were cut into strips (250x25 mm\textsuperscript{2}). The tensile test was achieved with a computerized universal testing machine (Jinan Test Machine WDW 100 kN). The cross-head speed was set to 2 mm/min. The stress-strain curve was recorded by a computer data acquisition system. All tests were performed at ambient temperature.

2.3.2. Three-point flexural test

Three-point flexural tests were conducted according to JISK7055. Different test coupons were cut into strips (170x15 mm\textsuperscript{2}). The flexural strength and flexural strain, \(\sigma_f\) and \(\varepsilon_f\) were calculated, respectively as follows\textsuperscript{28}:

\[
\sigma_f = \frac{3P_f L}{2bh^2} \quad (1)
\]

\[
\varepsilon_f = \frac{6D}{L^2} \quad (2)
\]

where L denotes the support span length, \(P_f\) is the maximum flexural load and D represents the maximum deflection of the middle of the strips.

| Table 1. Chemical composition of steel in mass %. |
|----------------|----------------|-----------|----------|----------|----------|---------------|
| C               | Si            | Mn        | P        | S        | Fe       | Remainder     |
| 0.19            | 0.03          | 0.85      | 0.01     | 0.005    | Remainder|               |
2.2.3. Hardness

Barcol hardness test is a means of evaluating the hardness of reinforced and non-reinforced rigid plastics. The hardness value determines the resistance of polymeric materials as penetrated by the indenter. The composite layers are placed beneath the indenter of the Barcol hardness tester and a uniform pressure is applied to the coupons until the dial indicator reaches a maximum value. The hardness was determined via the PCE-1000N Hardness instrument at ten different places of the micro/nanocomposite coated steel and the average value was taken.

2.3.4. Barrier resistance

Some test coupons were immersed in salt solution and in citric acid solution to estimate the corrosion performance of the micro/nanocomposite coated steel. Citric acid is one of the most versatile, inexpensive, and widely used organic acidulates, and it is commonly applied to the production of fruit-flavored beverages, lemon juice, and food. Citric acid serves to adjust the pH of jellies, jams, or preserve mixtures to the optimum range, where pectin can act most effectively. Certain foods such as guava, mangos, blackberries, cherries, sweet peaches, and sweet plums would not naturally contain sufficient acids to give the proper pH. In particular, citric acid is highly favored by the food industry on account of its light fruity taste, solubility, low cost, and abundant supply. Citric acid solution with a concentration of 2 N was prepared by double distilled water. The salt solution was performed as 3.5 wt% NaCl dissolved in water. Uptake tests were carried out according to ASTM D5229 / D5229M - 14. The coupons were periodically withdrawn from the solutions, wiped dry, and weighed using an analytical balance of accuracy up to 10^-4 g to observe the weight change during the absorption process. The solution content M(t) absorbed by micro/nanocomposite coating was then calculated as the mass gain percent refer to its initial weight (w0) as follows:

\[ M(t) = \left( \frac{w_t - w_0}{w_0} \right) \times 100 \]  

where \( w_t \) is the coupon mass after time t. Coupons were immersed for up to 21 days.

Composite materials absorb water when exposed to a wet environment that is assumed to enter into the composite material obeying the diffusion laws as Fick's laws. Fick's law predicts the linearity of the quantity of water uptake with the square root of time, and then gradually a decrease is attained until an equilibrium is achieved. The diffusion of water in composite material was studied using Fick's model by applying Equation 4:

\[ D = \pi \left( \frac{h}{4M_e} \frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}} \right)^2 \]  

where \( D \) is the diffusion coefficient, \( M_e \) is the weight of absorbed water when fully saturated, \( h \) is the specimen thickness, \( M_1 \) and \( M_2 \) are the solution uptake contents at times \( t_1 \) and \( t_2 \), respectively. These times are selected at an early stage of the uptake process, where the weight change can vary linearly with the square root of time.

3. Results and Discussions

3.1. Tensile properties

Figure 2 shows the tensile stress-strain curves of pure epoxy and steel lined with epoxy filled with Al₂O₃ micro/nanoparticles. These curves revealed that the various coating constituents led to several tensile behaviors. Also, adding either nanofillers or microfillers to epoxy coating increased both the tensile strength and tensile strain. The addition of 1wt% Al₂O₃ microparticles before 1wt% Al₂O₃ nanoparticles to the epoxy coating exhibited the highest tensile strength and tensile strain. Figure 3 shows the ultimate tensile strength of steel coated with micro/nanocomposites. Peak improvement of 16.7% in tensile strength was achieved with epoxy filled with 1wt% Al₂O₃ microparticles before 1wt%


Al₂O₃ nanoparticles on both sides (M1N1) as compared to pure epoxy coating. Moreover, a maximum tensile strain of 18.9% was obtained with the same specimen M1N1. However, increasing the weight percentage of the first layers to 2wt% micron Al₂O₃ particles before 1wt% nano Al₂O₃ particles (M2N1) decreased the tensile strain as compared to M1N1. However, the tensile strain of M2N1 increased as compared to M1N1 as shown in Figure 4. An enhancement of 7% and 3.2% in tensile strength was attained with EN1 and M2N1, respectively as compared to pure epoxy coating. However, an enhancement of 9.7% and 13.7% were attained in the tensile strain with EN1 and M2N1, respectively as compared to pure epoxy coating.

Figure 5 indicates the toughness of steel lined with pure epoxy and epoxy filled with Al₂O₃ micro/nanoparticles. The toughness is the ability of the material to absorb energy without fracture which its value is obtained from the tensile test. All additives of either nanofillers or microfillers to epoxy coating increased the toughness of the coupons. A significant enhancement of 32.6% was observed with M1N1 coated specimens. Moreover, an improvement of 7.3% and 14.3% was attained with EN1 and M2N1, respectively as compared to pure epoxy coating.

The addition of 1wt% Al₂O₃ microparticles before 1wt% Al₂O₃ nanoparticles exhibited the highest tensile strength and tensile strain as compared to pure epoxy coating. This is followed by a decrease in tensile strength and strain with a further increase in Al₂O₃ microparticles to 2wt% before 1wt% Al₂O₃ nanoparticles. This may be attributed to the good dispersion of the first layers of 1wt% micron Al₂O₃ particles in epoxy coating. Enhancing the mechanical properties depends on the good dispersion of Al₂O₃ microparticles inside the epoxy matrix. Dispensing of Al₂O₃ microparticles into epoxy homogenously gains the full benefit of its interfacial area with the epoxy matrix. Due to epoxy highly cross-linked structure, poor durability was
demonstrated to crack initiation and propagation. The good interfacial bond oriented the crack growth path through the micro/nanophase epoxy matrix. This long traveling path of the propagation of crack consumed high energy and hence improved the mechanical properties as compared to 2E specimen. So, the Al$_2$O$_3$ micro/nanoparticles might act as crack stoppers and increased the capability of the material to absorb energy by forming tortuous pathways for crack growth which resulting in an increasing in tensile properties. In addition, when microcracks due to the tensile loads were met with Al$_2$O$_3$ micro/nanoparticles, they might be stabilized by crack bridging of Al$_2$O$_3$ micro/nanoparticles$^{39}$. Micro/Nanoparticles hindered the development and propagation of micro-cracks in epoxy coatings. Epoxy filled with micro/nanoparticles offered more effective stress transfer, hence reducing the local stress concentration throughout the epoxy matrix$^{39}$.

However, further increase in the weight percentage of Al$_2$O$_3$ microparticles to 2wt% cause the formation of agglomeration which in turn decreases the tensile strength and strain as compared to M1N1. These agglomerations possess a higher surface area that helps in the formation of enclosed air bubbles from the atmosphere. Consequently, this causes a reduction in the tensile properties of the polymeric matrix composite$^{39-41}$. Also, the presence of particles agglomerations leads to stress concentration and may cause premature failure$^{42}$. Moreover, an increase in matrix viscosity due to the increase in Al$_2$O$_3$ microparticles content, which in turn allowed small air-bubbles to be trapped in the matrix during the mixing process forming tiny voids in the samples. This in turn results in specimens failure at relatively low stress$^{38}$.

### 3.2. Flexural properties

Figure 6 shows the flexural stress-strain curves of steel lined with pure epoxy and epoxy filled with Al$_2$O$_3$ micro/nanoparticles. Also, adding either nanofillers or microfillers to epoxy coating increased both the flexural strength and strain. The addition of 1wt% Al$_2$O$_3$ microparticles before 1wt% Al$_2$O$_3$ nanoparticles (M2N1) led to a maximum significant enhancement of 39.4% in hardness as compared to 2E. This may be concluded as, during the hardness test, when the indenter goes downward, it faced increased resistance from the filled coated material$^{43}$. This high resistance was owing to the increase of the weight content of Al$_2$O$_3$ particles that have a high hardness value. As the mass percentage of filler increased, the filler particles filled in the gap and voids in the polymeric matrices and formed a denser structure, and therefore hardness increased$^{44}$. An enhancement of 12.3% and 14.9% in hardness was attained with EN1 and M1N1, respectively as compared to the 2E specimen.

### 3.3. Hardness

Figure 9 shows the hardness of steel lined with micro/nanocomposites. The several coating configurations led to different hardness values. Increasing the weight percentage of the first layers to 2wt% Al$_2$O$_3$ microparticles before 1wt% Al$_2$O$_3$ nanoparticles (M2N1) led to a maximum significant enhancement of 39.4% in hardness as compared to 2E. This may be concluded as, during the hardness test, when the indenter goes downward, it faced increased resistance from the filled coated material$^{43}$. This high resistance was owing to the increase of the weight content of Al$_2$O$_3$ particles that have a high hardness value. As the mass percentage of filler increased, the filler particles filled in the gap and voids in the polymeric matrices and formed a denser structure, and therefore hardness increased$^{44}$. An enhancement of 12.3% and 14.9% in hardness was attained with EN1 and M1N1, respectively as compared to the 2E specimen. The enhancement in scratch and abrasive resistance was owing to the dispersion hardening of alumina nanoparticles in polymeric coatings$^{22}$. The high hardness value could
be attained for metallic coatings by inducing the hard nanocrystalline phases\textsuperscript{45}.

3.4. Failure mode

Figure 10 shows the damage mechanism in steel coated by epoxy and epoxy filled with Al\textsubscript{2}O\textsubscript{3} micro/nanoparticles after being subjected to tensile loading. The pure epoxy coating separated from the steel substrate during tensile fracture as shown in Figure 10. Adding 1 wt\% Al\textsubscript{2}O\textsubscript{3} nanoparticles into the epoxy matrix in the exterior layers improved slightly the adhesion of the coating to steel as compared to 2E specimen. However, adding 1 wt\% Al\textsubscript{2}O\textsubscript{3} microparticles into the epoxy matrix that represented the first layer coated steel on both sides followed by 1 wt\% Al\textsubscript{2}O\textsubscript{3} nanoparticles into the epoxy matrix as exterior layers enhanced the adhesion to steel substrate thus revealing the good tensile properties of this
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Adding 2 wt% Al₂O₃ microparticles into epoxy matrix that represented the first layer coating on both sides followed by 1 wt% Al₂O₃ nanoparticles into the epoxy matrix enhanced also the coating adhesion. The increase in micron-sized weight percentage to 2 wt% in the first layer decreased the adhesion of the coatings as compared to M1N1. Adding 2 wt% Al₂O₃ microparticles to the epoxy matrix as the first layer increased the adhesion of this layer to the steel substrate. The second layers from both sides of epoxy filled with 1 wt% Al₂O₃ nanoparticles were characterized by good adhesion to the first layers of epoxy filled with 2 wt% Al₂O₃ microparticles on both sides. So, after being subjected to tensile loadings, good bonding was observed for the two composite layers from both sides to steel. However, for EN1 specimens, the first layers of unfilled epoxy were separated from the steel surface after tensile loading. This in turn led to the separation of the second layers of epoxy filled with 1 wt% nanoparticles as the second layers had adhered to the first layers of epoxy coatings.

3.5. Barrier resistance

Figure 11 shows the barrier properties of steel coated by epoxy and epoxy filled with Al₂O₃ micro/nanoparticles when immersed in 3.5 wt% NaCl. Moreover, Figure 12 shows the barrier properties of steel coated by epoxy and epoxy filled with Al₂O₃ micro/nanoparticles when immersed in citric acid solution. The salt solution and citric acid absorption content increased as the immersion time increased until equilibrium saturation was achieved. It is clear that the coating with double layers filled with either nanoparticles or microparticles increased the barrier properties of steel against salt solution and citric acid solution as compared with pure epoxy coating. The least water absorption was detected for M1N1 composite coating, followed by M2N1 composite coating. This indicated that further increase of microparticles led to a slight decrease in barrier properties. Table 2 and Table 3 show Fickian Diffusion coefficient values of steel coated by epoxy and epoxy filled with Al₂O₃ micro/nanoparticles when immersed in 3.5 wt% NaCl and citric acid solution, respectively. It was observed that the addition of 1 wt% and 2 wt% of Al₂O₃ microparticles to the epoxy matrix in the first layers decreased the diffusion coefficient as compared to 2E as the specimens immersed in 3.5 wt% NaCl and citric acid solution. The highest diffusion coefficient was observed for steel coated with pure epoxy coating. From Tables 2 and 3, the diffusion coefficient of specimens immersed in citric acid was greater than ones immersed in the salt solution.

Figure 10. The damage mechanism in steel coated by epoxy and epoxy filled with micro/nano Al₂O₃ particles after being subjected to tensile loading.
Nanofillers as alumina, titania, silica are considered well-known materials utilized in coating\(^7\). As reported by Saji and Thomas that the epoxy filled with MoO\(_3\) nanofillers coated steel significantly improved the barrier capacity of epoxy by preventing the transportation of H\(_2\)O and ions into the epoxy matrix and reduce the opportunities of deterioration and blistering of the coating film\(^9\). The nano-sized inclusion reduce the rate of absorption of water due to the barrier properties of these nanoparticles thus improve the properties of these plasticized nanocomposites\(^27\),\(^46\). These nano-fillers close the pores inside the epoxy matrix, permitting interconnecting with molecule chains, thus increasing the density of polymer cross-linking hence reducing the free volume\(^47\). Similarly, reducing water absorption by adding nanoparticles to the polymeric matrix was attained by\(^48\),\(^49\),\(^50\). Moreover, Nguyen-Tri et al.\(^{45}\) attributed the good barrier properties to the inclusion of nanofillers into the organic polymeric that decreased the voids and zigzagging the path of diffusion for deleterious species. Therefore, the coating films including nanofillers were expected to have significant barrier behavior for corrosion protection and lower the trend for the coating film to blister or delaminate. Nanofillers modify the surface energy of the inherently hydrophobic siloxane polymers thus enhancing the performance of nanocomposite coating soaked in aggressive media\(^19\). The fillers efficiently blocked the corrosion medium and enhanced the anticorrosive performance of the composite coating\(^6\).

Figure 11. The barrier properties of steel coated by epoxy and epoxy filled with micro/nano Al\(_2\)O\(_3\) particles when immersed in 3.5wt\%NaCl.

Figure 12. The barrier properties of steel coated by epoxy and epoxy filled with micro/nano Al\(_2\)O\(_3\) particles when immersed in citric acid solution.

Figure 13. SEM showing the distribution of alumina nanoparticles in M2N1 composites.
of nanocomposites. Figure 14 shows the EDX of coated M2N1 composites on the steel substrate. The elements that are more presented in the composite coating are carbon and oxygen.

### 4. Conclusions

In this study, the mechanical and barrier properties of steel coated with epoxy filled with alumina particles in micron and nanosized were investigated. Double layers of pure epoxy coated steel were compared with the other three different sequences of double micro/nanocomposites layers coated steel. Tensile, flexural, hardness, and barrier properties of the coated coupons were studied. The results showed that a maximum enhancement of 16.7%, 18.9%, 32.6%, 8.5%, and 5.7% in tensile strength, tensile strain, toughness, flexural strength, and flexural strain, respectively, were achieved with a coating of epoxy filled with 1wt% Al₂O₃ microparticles before 1wt% Al₂O₃ nanoparticles on both sides as compared with pure epoxy coating. Adding micro/nanoparticles to epoxy coating enhanced the barrier properties of the coating against salt solution and citric acid environment as compared to pure epoxy coated.

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