

Research Article

Barrier Corrosion Protection Properties of Metakaolin Clay-Kadilux Epoxy Coatings on Galvanized Steel

Fantaye Tasew  and Ganesh Thothadri 

Department of Materials Science and Engineering, School of Mechanical, Chemical and Materials Engineering, Adama Science and Technology University, P. O. Box 1888, Adama, Ethiopia

Correspondence should be addressed to Ganesh Thothadri; ganesh_reliez@yahoo.co.in

Received 19 May 2020; Revised 8 February 2021; Accepted 27 February 2021; Published 15 March 2021

Academic Editor: Ihsan ul Haq Toor

Copyright © 2021 Fantaye Tasew and Ganesh Thothadri. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Epoxy polymer, an illustrious barrier corrosion protective coating, was reinforced with metakaolin clay, an eco-friendly inorganic filler to enhance the barrier corrosion protection properties in water and in acidic environment on galvanized steel plates. Various proportions 0, 1, 3, 5, and 7 wt.% of metakaolin fillers were mixed intrinsically with kadilux epoxy and characterized for thermal stability, water absorption according to ASTM G31, and acid immersion according to ASTM D-570 standards, respectively. The reinforced coatings minimized the pore size and density, lower water absorption, and better acid resistance properties especially at 7 wt.% of the fillers. The thermal stability of the films improved beyond 5 wt.% of filler composition.

1. Introduction

Corrosion is an intrinsic problem in many industries like automobiles, sugarcane industries, maritime, petrochemical, and aerospace. To protect the metal substrate, many methods had been established such as barrier protection, anodic protection, and cathodic protection [1–4]. Even though lots of advanced corrosion protection methodologies are available currently like cathodic protection, anodic protection, and alloys to suppress the corrosion processes, nevertheless a barrier corrosion protective coating is mandatory. Among many polymeric coatings, epoxy coating has strong adhesion capability to metallic substrates and excellent metallic resistance. Most of the polymers tend to absorb certain amount of water molecules when immersed in aqueous environment and in high humidity conditions [5]. Epoxy tends to absorb polar water molecules during curing and also on specific polar groups in the epoxy system [6]. This water molecules aid in corrosion and also deadhesion of the coatings [7]. The absorbed water molecules can cause various forms of corro-

sion like pitting, crevice corrosion, intergranular corrosion, and selective leaching.

Duplex system, a painting or powder coating over hot-dip galvanized steel, slows down the rapid deterioration of zinc coating. Epoxy paint or coating, widely used for its excellent mechanical and thermal properties, nevertheless scums due to pore network formation while cross linking, acting as a path for water and corrosive species. These defects (pore networks) lead to ions diffusing through the coating and attacking the metal substrate [8]. There are various reports concerning improving corrosion resistance of coatings using nanoparticles such as TiO_2 [9], ZnO [10], SiO_2 [11], and ZrO_2 [12]. Fillers like graphene, nanoclay, zinc oxide (ZnO), montmorillonite, and molybdenum sulfide (MoS_2) were used so far to increase the diffusion path of corrosive liquid beneath the coating and further inhibit the metal corrosion reaction. For an effective corrosion protection, the barrier coating of high density, highly cross-linked, thicker, and self-healing coating is needed [13]. Metal oxide fillers in to a polymer matrix are expected to improve the mechanical, chemical, and thermal

properties [14]. Among various fillers, clay fillers are most abundantly available at an economical price.

Clay minerals can form layered structures either two, three, or four sheets of tetrahedral silica $[\text{SiO}_4]^{4-}$ or octahedral alumina $[\text{AlO}_3(\text{OH})_3]_6$. Kaolin is one kind of clay minerals with chemical formula $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$, layered and electronically neutral (uncharged) material. It is found mostly in Bombowha and Kombolcha areas of Ethiopia [15] and also usually as byproducts of granite and pegmatites. In this study, both kaolin $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ and metakaolin $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ obtained by the heat treatment of kaolin (550-850°C) are used.

In this study, kaolin clay materials, metakaolin (MK), and modified clay particles are used as fillers in the epoxy matrix, and their acid resistance, water absorption, pore size reduction, and thermal properties had been studied.

2. Materials and Method

2.1. Materials Used. Kadilux epoxy is a two-part epoxy resin, consisting of a prime hardener as a curing agent and epoxy resin, manufactured by Kadisco Paints and Adhesive Industry S.C. (Asian Paint's Group), Addis Ababa, Ethiopia. Kaolin clay was received from Jimma Science and Technology University, MK clay was prepared by heating the kaolin clay at temperature of 600°C for 3 hours in a high temperature box furnace, and galvanized steel sheet was manufactured by Adama sheet and steel factory. The galvanized steel are cut into pieces with a dimension of 8×3 cm using a steel cutter. Liquid silicone rubber was used to make molds.

2.2. Equipment and Characterization Used. Grinder (RRH-A350), a high-speed multifunctional comminutor with a mortar speed 28000 r/min and degree of grinding 50-300 mesh size, is used to grind the kaolin clay material into fine fragments. Forced convection drying oven was used for drying wet samples with a temperature range of room temperature to 300°C. Ultrahigh frequency (up to 40 kHz) ultrasonicator from Jeiotech UC-20 was used to disperse fillers in polymer matrix and also for breaking the intermolecular interaction.

Optical microscope (Huvitz HR-300 series; magnification: x50) was used to examine the morphology of the samples and coatings. Shimadzu Maxima_X XRD-7000 X-ray diffractometer was used to study and identify the crystallographic phases, with Cu target (40 kV voltage; 30 mA current), divergence slit of 1°, scatter slit of 1°, and receiving slit of 0.3 mm and drive axis 2θ , scan range of 10-80°, scan speed of 3°/min, and a sampling pitch of 0.02°.

2.2.1. Acid Immersion Test. Sulfuric acid is one of the most destructive acids to metal, and depending on its concentration and formation manner, it can cause severe degradation and damage to metal structures which come into contact with it. The rate of this action is dependent on factors such as porosity and pore structure and most importantly the pH and concentration of the sulfuric acid. In this work, the corrosion resistance data relates to the corrosion resistance of various coated steel materials immersed in a 2% sulfuric



FIGURE 1: Magnetic beads with adsorbed impurities while purification of kaolin clay.

acid solution at room temperature in accordance with ASTM G31 [16].

2.2.2. Water Absorption Test. The hydrogen bonds among polymer segments are, however, disrupted by any water that is absorbed into the polymer network. Water has higher electron density than the functional centers of the network, and this disruption of hydrogen bonds can swell and plasticize the polymer. The water absorption test was carried out based on ASTM D-570. The samples were cut, cleaned, and weighed before immersion in distilled water at room temperature. The specimens were removed from the water after some time, and the surface was wiped off and weighed immediately. The difference between the weight before and after immersion was noted. This procedure was repeated every 24 h until it reaches saturation level. The percent water absorption was determined based on the expression.

$$\text{Percent water absorption} = \frac{\text{Wet weight} - \text{Dry weight}}{\text{Dry weight}} \times 100, \quad (1)$$

where “Wet weight” and “Dry weight” are the final and initial weights of the samples after and before immersion, respectively.

2.3. Experimental Procedure

2.3.1. Sample Preparation for Reducing Impurities of Kaolin Clay. The received kaolin clay was grinded by RRH-A350 high-speed multifunction comminutor with speed of 28000 r/min. After grinding, the powder was sieved by 45 μm sieves to get kaolin clay with a particle size $\leq 45 \mu\text{m}$. Two types of treatment process were performed: (1) removal of moisture or water content in the kaolin clay and heat treatment at 600°C for 3 h and (2) heat treatment for removing moisture present in kaolin clay. In the former case, kaolin



FIGURE 2: Photo images of (a) kaolin clay before and (b) after purification.

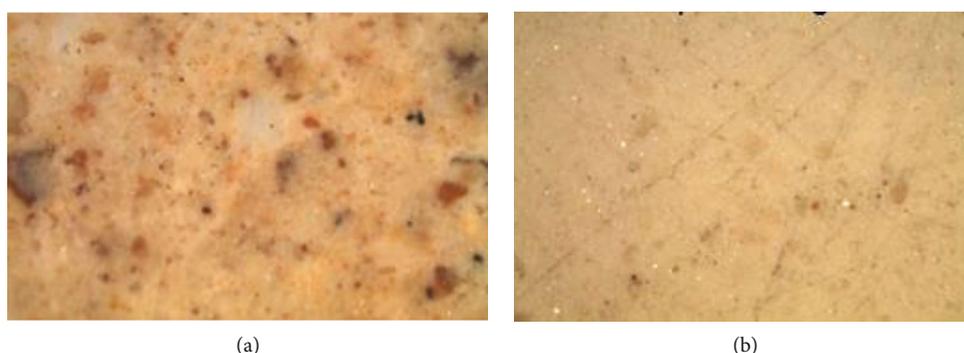


FIGURE 3: The optical images of (a) raw kaolin and (b) MK clay after heat treatment at 600°C.

was first purified by distilled water by dispersing 5 g of clay in to 100 ml of water followed by magnetic stirring for 1 h, then leave the solution for 2 h for sedimentation of heavy components and impurities like quartz and iron. Purification was followed by decantation of solution by using filter papers and then dried in an oven. After preparation of powder samples, X-ray characterization was done.

Kaolin clay powder was washed in water and dried many times to remove the impurities like quartz and iron and. During the purification process, while stirring the kaolin clay in distilled water, the impurities are agglomerated or adsorbed on the magnetic beads, as shown in Figure 1. Photo images of kaolin clay before and after purification are shown in Figures 2(a) and 2(b). The optical images of raw kaolin and MK clay are shown in Figures 3(a) and 3(b).

2.3.2. The Difference in Water Absorption between Kaolin and Metakaolin (MK). To determine the water absorption capacity of epoxy/kaolin clay and epoxy/MK clay composites, both the composites were immersed in distilled water for 3 days. The conversion of kaolin into metakaolin (MK)

TABLE 1: Epoxy samples specification.

Symbol	Name
GZ/GS	Bare steel (galvanized steel)
EP0	Epoxy without the addition of filler
EP1	Epoxy with the addition of 1 wt.% filler
EP3	Epoxy with the addition of 3 wt.% filler
EP5	Epoxy with the addition of 5 wt.% filler
EP7	Epoxy without the addition of 7 wt.% filler

was carried out by heating the kaolin clay at 600°C for 3 h. Preparation of the epoxy composite started by weighing 50 g of resin and heated at 60°C for 10 min to reduce the viscosity of resin. Then, MK clay was added and mixed by using magnetic stir for 15 min then 10 min ultrasonication followed by addition of hardener and mixed for 10 min, poured the prepared composite in to silicon mold, and then cured for 7 days (360 h).

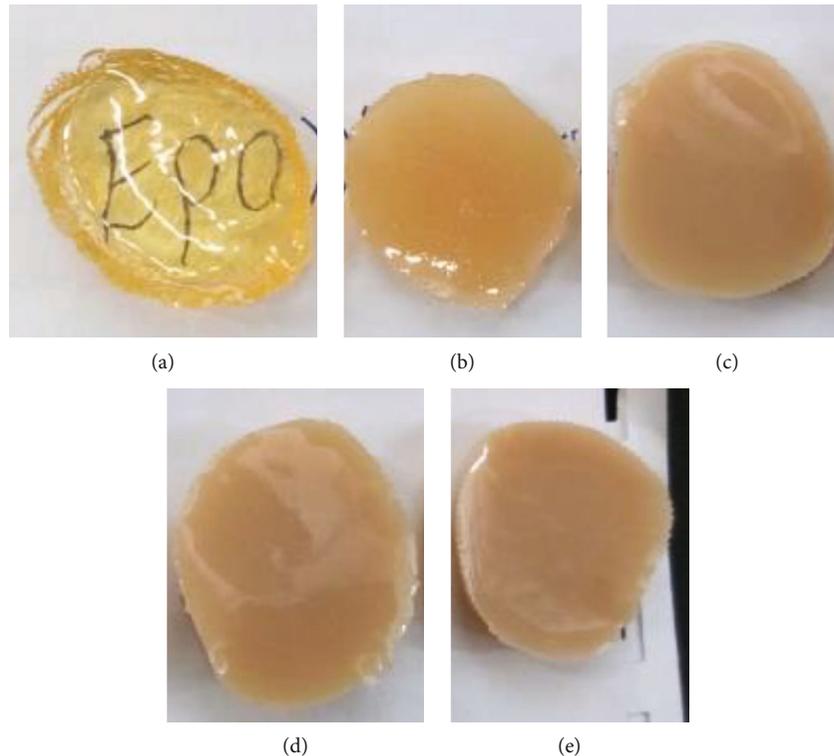


FIGURE 4: Shows the visual images of the epoxy-MK composite: (a) EP0, (b) EP1, (c) EP3, (d) EP5, and (e) EP7 (reduction in transparency due to the addition of MK fillers is shown and visually observed).

Weight of the samples were measured before and after (1, 2, 3, 4, 12, 16, 24, 48, and 72 h) immersion, and weight gain was calculated according to equation (1).

2.3.3. Sample Preparation for Studying Exfoliation and Corrosion Property of Epoxy/MK Composite. Epoxy resin was preheated at 60°C to lower the viscosity so that it was easier for the added MK clay to disperse, and this step was conducted by using magnetic stirrer. 0, 1, 3, 5, and 7 wt.% fillers were added into 50 parts of epoxy resin separately in different container and mixed by magnetic stirrer with speed of 400 rpm for 20 min. This speed assisted the clay to break into small particles and helped to increase the clay/epoxy interfaces; then, to increase homogeneity of the composite, ultrasonication for 10 min was done. The mixture was then added with 25 g of prime resin curing agents (2:1 ratio of resin to hardener based on factory specification data sheet) and mixed for 10 min by using magnetic stirrer. The obtained polymer composite was then poured into a silicon mold, pristine epoxy polymer, without the addition of filler, and was also prepared as reference sample. For acid corrosion testing purpose, sample was prepared by immersing the galvanized steel sheet in the prepared composite.

The steel specimen was washed with acetone and double distilled water; they were immersed into ethanol solution for 1 h for hydrolysis. Finally, sample was dried at 100°C for 20 min then immersed in to epoxy/filler composition for 5 sec. All samples were cured at room temperature for 7 days. The epoxy curing procedure was done according to kadisco

paint factory specification data sheet. The sample thickness before coating was 0.28 mm, and after coating the total thickness, it becomes 0.46 ± 0.03 mm. The sample specifications are shown in Table 1.

Optical transparency in polymer composite is limited by the particle size of filler since if the fillers are large enough to scatter the incident light, the composite becomes translucent or opaque depending on the amount of scattered light. According to this research, the composite sample was opaque since there was a total scattering of incident light due to the presence of microsized MK filler as is shown in Figure 4.

3. Results and Discussion

3.1. Structural Studies of Kaolin and Surface Modification of Clay by Using Triethanolamine (TEA). As mentioned in sample preparation technique, Section 2.3.1, the raw kaolin was undergone two different treatment processes: (1) removal of moisture or water content in the kaolin clay and heat treatment at 600°C for 3 h (heat-treated kaolin or MK) and (2) heat treatment for removing moisture only present in the kaolin clay (raw kaolin). In the heat-treated kaolin, most of the $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ present in the raw kaolin was removed after the heat treatment process at 600°C for 3 h, as shown in Figure 5, according to ICDD file number for kaolinite $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$, 80-885 and for α -quartz SiO_2 , 85-1054. Similar transformation of crystallographic structure on heat treatment of kaolin or kaolinite was also observed by Wang et al. [17].

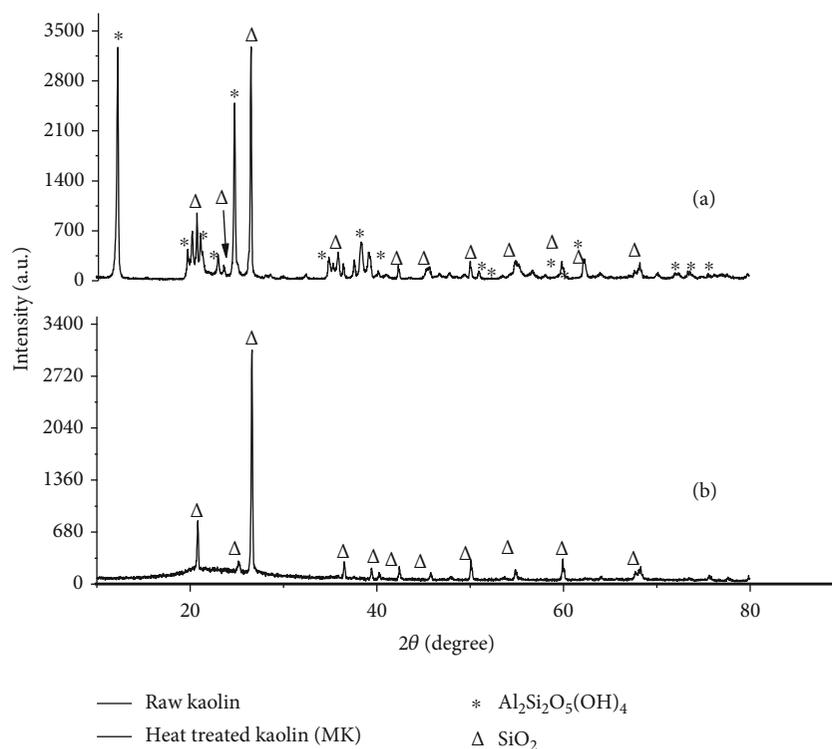


FIGURE 5: XRD spectra of (a) raw kaolin and (b) heat-treated kaolin (MK).

XRD analysis was done on kaolin clay and kaolin clay treated with triethanolamine (TEA), as shown in Figure 6, to change the surface of kaolin. Apart from the characteristic peaks of kaolin clay, i.e., $2\theta = 12.36^\circ, 19.94^\circ, 24.90^\circ, 35.98^\circ, 38.46^\circ, 45.66^\circ, 55.12^\circ,$ and 62.34° , none other change in interplanar or d-spacing was observed. TEA is a strong base, and kaolin clay also carries a negative surface charge depending on pH. Under alkaline conditions, the kaolin clay represents either neutral or negative charge [18]; thereby, it is expected to repel against negative TEA.

3.2. Water Absorption in Kaolin and Metakaolin. The results of water absorption test of kaolin and MK, separately, showed that MK has low water absorption than kaolin which is a preferable filler in case of corrosion protection coatings. Blistering of the coating happens if it absorbs high amount of water which leads to the reduction in service time. Weight gain vs. time of water immersion of neat epoxy, epoxy/MK, and epoxy kaolin composite is shown in Figure 7.

Abdul Majeed and Sabar [19] showed that there is general increase in absorption rate with increase in kaolin content in PVC/kaolinite composites. Amit [20] showed the difference in water absorption ability between calcinated kaolin and row kaolin on their study of physicomechanical properties of composite materials of low-density polyethylene and raw/calcinated kaolin. According to their work, calcinated kaolin showed low water absorption than kaolin [12].

3.3. Optical Microscope Observation. During polymerization of epoxy, the hardener opens the C—O—C rings, and the

bonds are rearranged to join the monomers into a three-dimensional network of crosslinked chain-like molecules; this curing (shrinkage) leads to the formation of pore-like network in epoxy. Based on optical microscope image, the neat epoxy has high porosity than composites (the black colored spherical structures are the porous structures formed after epoxy curing process, as shown in the optical microscopic images in Figure 8); in case of composites, the fillers in the composite lead to reduction in porosity; this is due to the filling effect of the metakaolin clay. According to Figure 8(a), there were larger numbers of voids in EP0 which could be the reason for high water absorption by resin. But in case of composites, it showed the reduction of porosity with increase in concentration of MK and this was because of pore refinement properties of MK. The pore-size distribution curves indicated the marked influence that MK additions had in skewing pore-size distribution to the finer sizes compared with the samples without MK. It was also observed that the total porosity decreased substantially with increasing replacement level of MK; this is shown in Figure 8. Even though the pore refinement 100% was not achieved, high pore reduction performance showed by composite with 7 wt.% MK.

3.4. Corrosion Study. Corrosion is a chemical or electrochemical reaction between a material, usually a metal, and its environment that produces a deterioration of the material and its properties. It is also expressed as a destructive process developed by electrolyte action over a metal. Galvanized steel is most commonly used in applications such as buildings,

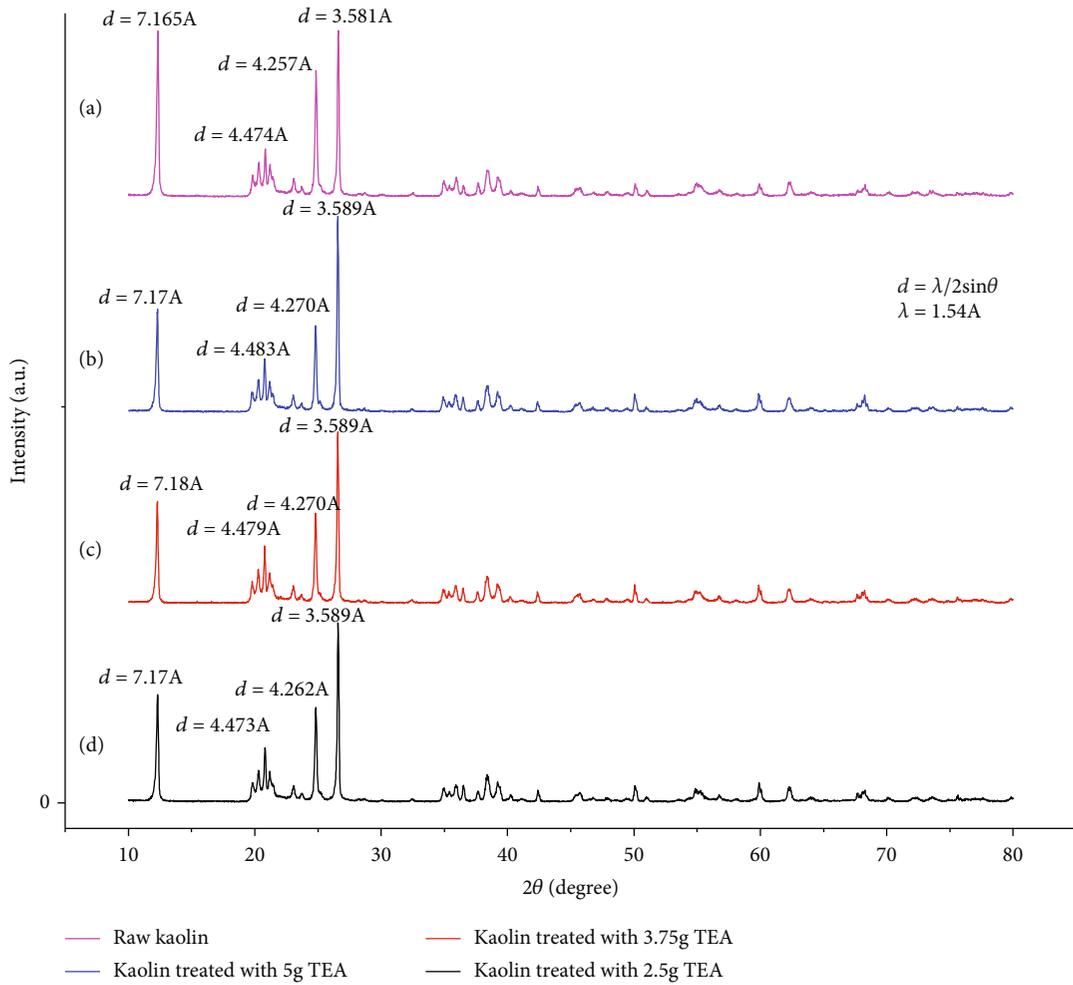


FIGURE 6: Comparison of interplanar or d-spacing of kaolin clay modified by (a) kaolin as received, (b) 5 g, (c) 3.75 g, and (d) 2.5 g of TEA modifier based on XRD analysis.

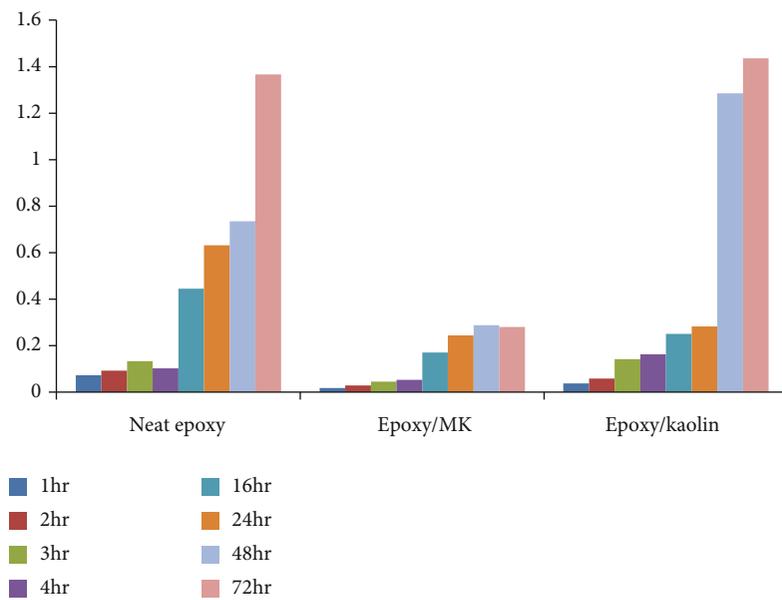


FIGURE 7: Weight gain vs. time of water immersion of neat epoxy, epoxy/MK, and epoxy kaolin composite.

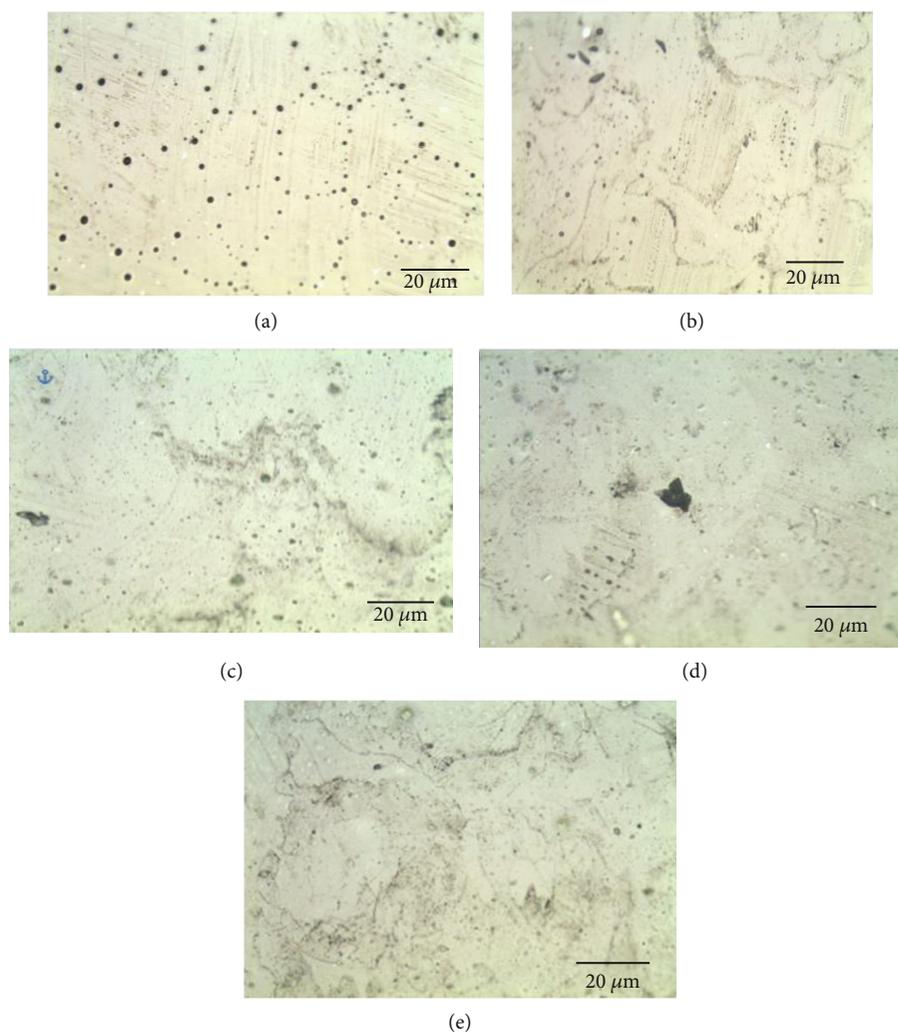


FIGURE 8: Optical image of (a) neat epoxy, (b) EP1, (c) EP3, (d) EP5, and (e) EP7 composite.

roofing, automotive parts, and water pipeline systems owing to its good resistance to corrosion. Corrosion resistance of galvanized sheet is largely dependent on the protection obtained from the zinc coating technique. A galvanized coating provides steel parts with reliable and long-term protection from corrosion if they are exposed in a suitable environment. Galvanized parts perform best when exposed to climatic influences in locations with low air pollution and where wetting of their surface is only temporary. If the corrosion conditions are unsuitable for hot-dip galvanized coating, the service life of the coating may be very short under certain conditions. Corrosion attack of zinc is substantially supported by high air pollution, especially an increased concentration of sulfur dioxide or chlorides in the atmosphere.

3.4.1. Acid Immersion Test. Galvanized steel performs best in solutions with a pH in the range of 5.5 to 12 according to Pourbaix diagram. pHs between 3 and 5.5 (acidic) or 12 and 13.5 (basic) are corrosive to galvanized steel, but the gal-

vanized coating will still give corrosion protection to bare steel, although the protection will only last for a few years. To determine the effect of sulfuric acid (pH) on corrosion, 2 ml H_2SO_4 and 100 ml of distilled water were first mixed and then immerse the coated and reference samples in to acid/water solution. The samples were immersed for 2 and 24 h, separately in different containers.

It should be noted that sulfuric acid migrates from the outside to the interior. Therefore, the outer surface which exposed to the acid must play an important role in neutralizing and preventing the solution from penetrating the inside or substrate. According to the result, the specimens immersed in a 2 wt.% sulfuric acid solution were not much affected after the exposure period for 2 h except uncoated GZ but on the contrary, all of the specimens had inferior performances against the acidic solution after 24 h. Specimens which consisted 7 wt.% MK had better resistance against a highly aggressive sulfuric acid solution than the reference (GZ) and other coated specimens; this is shown in Figures 9 and 10. The rate of this action is dependent on

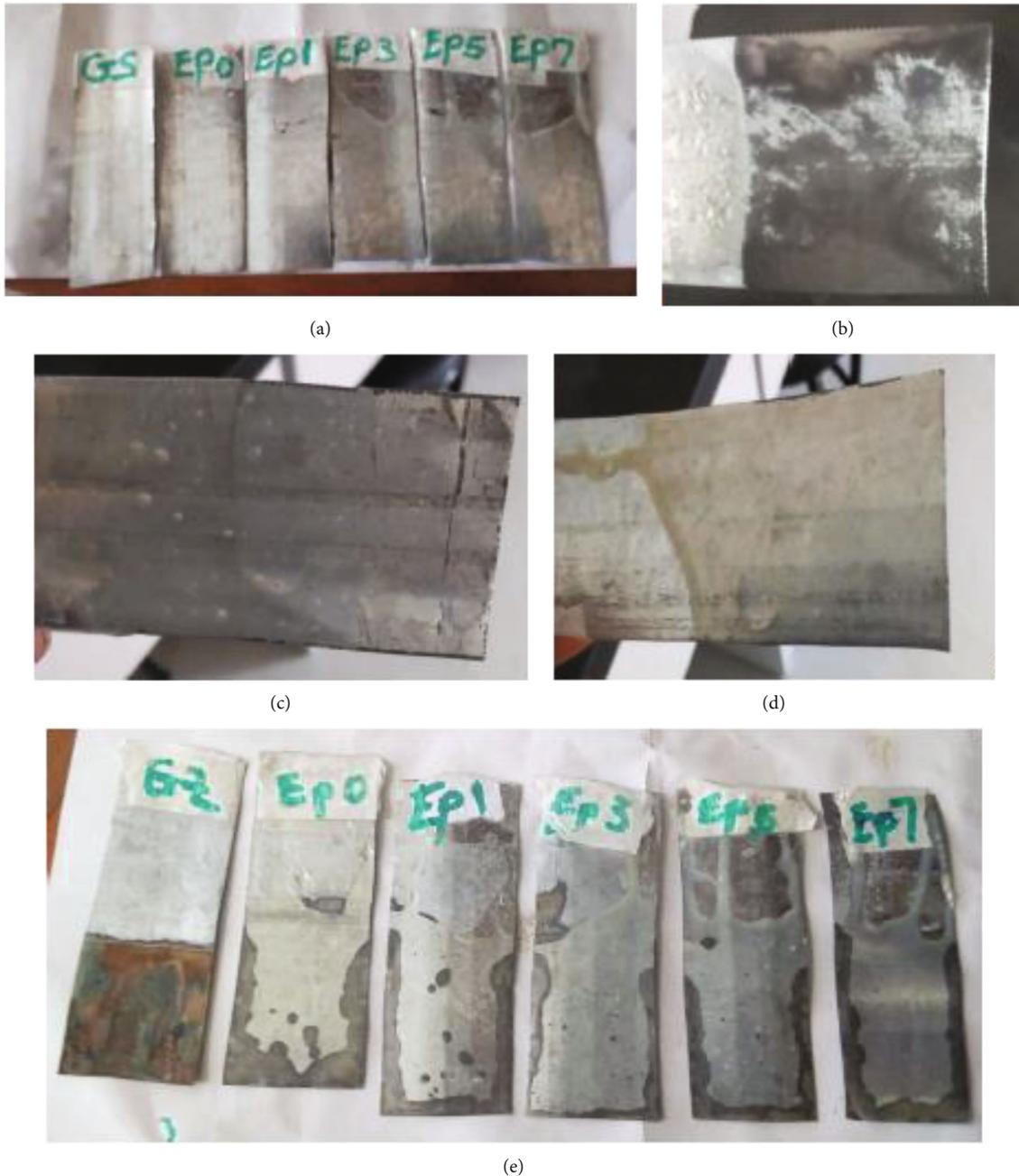


FIGURE 9: The effect of strong acid on galvanized steel (a) before immersion, (b–d) GZ, EP5, and EP7, respectively, after 2 h immersions, and (e) after 24 h immersions in 2 wt.% H_2SO_4 .

factors such as number of pore and the pore structure of coating and most importantly, the pH and concentration of the sulfuric acid.

3.4.2. Water Absorption Test. The prepared specimens were placed in water bath at room temperature for about 7 days until it reached saturation state, at regular intervals of 1 h for first 4 h and increasing to 24 h, then increased the immersion time to 48, 72, and 96 h. Each sample was first removed from water and dried with a tissue before weighting using electronic balance. Tendency of epoxy composites to absorb

water and water diffusion through the samples was investigated by the water uptake test at ambient temperature during 4 days. For the initial stage, the water uptake (W_t) increases with time, while for longer exposure time, the asymptotic behavior (saturation) was present (Figure 11). The negative reduction in weight showed in EP5 and EP7 may due to the loss of loosely bonded fillers from the composite due to sorption of water, and this was also shown by Omar [21]. Water sorption would result in elution of unreacted monomers and/or unreacted fillers, which in turn will result in loss of weight.

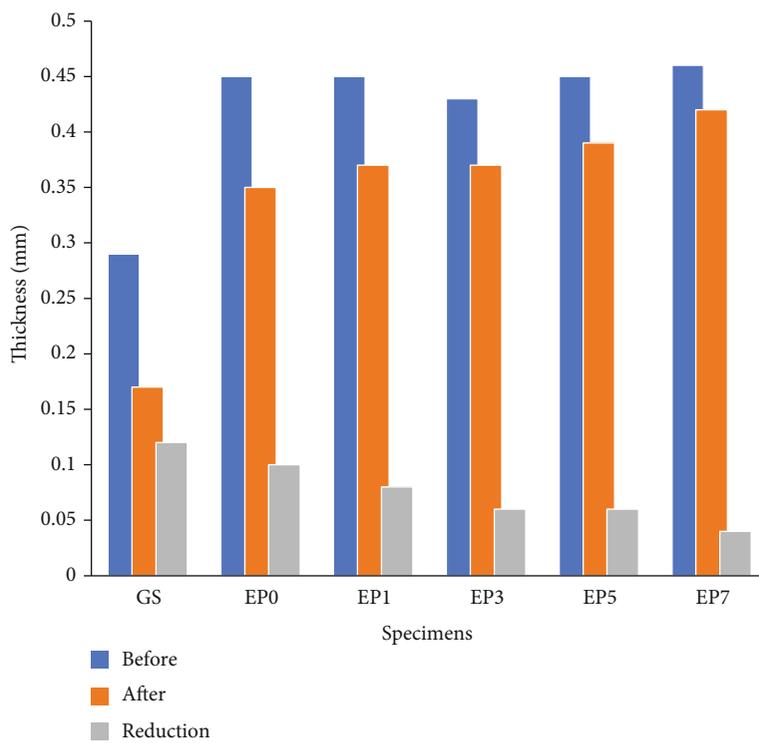


FIGURE 10: The reduction in thickness before and after immersions, in 2 wt.% of H₂SO₄, of uncoated and coated GS.

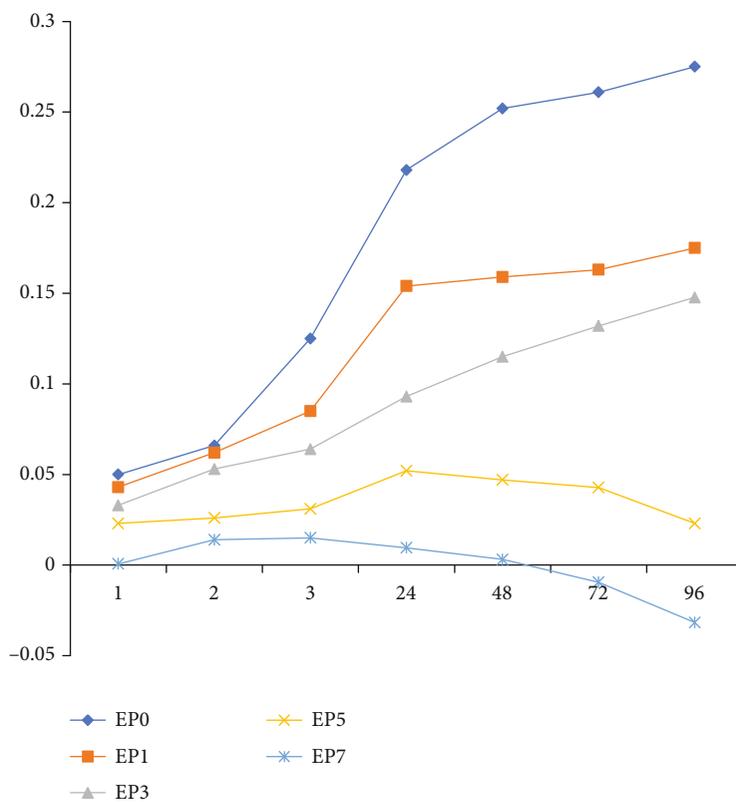


FIGURE 11: Percent weight gain of epoxy and epoxy composite after 96 h immersion in distilled water.

4. Conclusion

In this study, epoxy was reinforced with MK (with different concentration) in order to observe the effect of those MK reinforcements on the pore refinement and corrosion property (the water absorption and acid resistance property) of the composite, and through the processing methodology listed above, epoxy/MK composite was synthesized. The surface modification of MK was not improved significantly after TEA treatment, due to high basic nature of TEA and negative surface charge of filler. Based on water absorption and acid resistance test, all epoxy composites, especially epoxy/7 wt.% MK composite, showed a positive effect on both acid resistance and absorption property. The performance enhancement of MK is because of high pore refinement and acid protection ability and also low water absorption property due to removal of OH groups during calcination of kaolin. In general, filler concentration in the coatings, the thickness of the coatings, and surface preparation of substrates were the prime factors which affecting the corrosion resistance of composite coatings, and effective corrosion resistance was observed when the coating thickness increased. The epoxy/7 wt.% MK coated samples with a coating thickness of 250 μm showed better corrosion resistance properties when compared to the other samples. The authors recommend that for further enhancement of barrier protection properties of kadilux epoxy resin, using nanosized particles and thermal crosslinking of resin will have better performance.

Data Availability

Data are available upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

References

- [1] B. Ramezanzadeh, S. Niroumandrad, A. Ahmadi, M. Mahdavian, and M. H. M. Moghadam, "Enhancement of barrier and corrosion protection performance of an epoxy coating through wet transfer of amino functionalized graphene oxide," *Corrosion Science*, vol. 103, pp. 283–304, 2016.
- [2] T. Ohtsuka, "Corrosion Protection of Steels by Conducting Polymer Coating," *International Journal of Corrosion*, vol. 2012, no. 1, Article ID 915090, 7 pages, 2012.
- [3] G. M. Spinks, A. J. Dominis, G. G. Wallace, and D. E. Tallman, "Electroactive conducting polymers for corrosion control," *Journal of Solid State Electrochemistry*, vol. 6, no. 2, pp. 85–100, 2002.
- [4] A. D. King and J. R. Scully, "Sacrificial anode-based galvanic and barrier corrosion protection of 2024-T351 by a Mg-rich primer and development of test methods for remaining life assessment," *Corrosion*, vol. 67, no. 5, pp. 1–22, 2011.
- [5] H. Zheng, Y. Shao, Y. Wang, G. Meng, and B. Liu, "Reinforcing the corrosion protection property of epoxy coating by using graphene oxide-poly(urea-formaldehyde) composites," *Corrosion Science*, vol. 123, pp. 267–277, 2017.
- [6] P. B. Messersmith and E. P. Giannelis, "Synthesis and characterization of layered silicate-epoxy nanocomposites," *Chemistry of materials*, vol. 6, no. 10, pp. 1719–1725, 1994.
- [7] J. Ampudia, E. Larrauri, E. M. Gil, M. Rodríguez, and L. M. León, "Thermal scanning Rheometric analysis of curing kinetic of an epoxy resin. I. An anhydride as curing agent," *Applied Polymer Science*, vol. 71, no. 8, pp. 1239–1245, 1999.
- [8] S. H. Kim, *Improving water barrier properties of epoxy coatings with addition of graphene oxide*, Bachelor of Science, the College of William and Mary, 2017.
- [9] S. Radhakrishnan, C. R. Siju, D. Mahanta, S. Patil, and G. Madras, "Conducting polyaniline-nano-TiO₂ composites for smart corrosion resistant coatings," *Electrochimica Acta*, vol. 54, no. 4, pp. 1249–1254, 2009.
- [10] S. K. Dhoke, A. S. Khanna, and T. J. M. Sinha, "Effect of nano-ZnO particles on the corrosion behavior of alkyd-based waterborne coatings," *Progress in Organic Coating*, vol. 64, no. 4, pp. 371–382, 2009.
- [11] J. Xu, J. Tao, S. Jiang, and Z. Xu, "Investigation on corrosion and wear behaviors of nanoparticles reinforced Ni-based composite alloying layer," *Applied Surface Science*, vol. 254, no. 13, pp. 4036–4043, 2008.
- [12] M. Behzadnasab, S. M. Mirabedini, K. Kabiri, and S. Jamali, "Corrosion performance of epoxy coatings containing silane treated ZrO₂ nanoparticles on mild steel in 3.5% NaCl solution," *Corrosion Science*, vol. 53, no. 1, pp. 89–98, 2011.
- [13] B. Chen, J. Ma, and L. Gu, "Anticorrosive properties of oligoaniline containing photocured coatings," *International Journal of Electrochemical Science*, vol. 10, pp. 9154–9166, 2015.
- [14] X. Shi, T. A. Nguyen, Z. Suo, Y. Liu, and R. Avci, "Effect of nanoparticles on the anticorrosion and mechanical properties of epoxy coating," *Surface and Coatings Technology*, vol. 204, no. 3, pp. 237–245, 2009.
- [15] H. M. Fentaw and T. Mengistu, "Comparison of Kombelcha and Bombowha kaolins of Ethiopia," *Applied Clay Science*, vol. 13, no. 2, pp. 149–164, 1998.
- [16] ASTM, "G31 – Standard practice for laboratory immersion corrosion testing of metals," 2004.
- [17] M. R. Wang, D. C. Jia, P. G. He, and Y. Zhou, "Influence of calcination temperature of kaolin on the structure and properties of final geopolymer," *Materials Letters*, vol. 64, no. 22, pp. 2551–2554, 2010.
- [18] M. S. Nasser and A. E. James, "The effect of electrolyte concentration and pH on the flocculation and rheological behavior of kaolinite suspensions," *Journal of Engineering Science and Technology*, vol. 4, no. 4, pp. 430–446, 2009.
- [19] B. A. Abdul Majeed and D. A. Sabar, "Effect of kaolinite on the mechanical properties, thermal properties, flammability and water absorption percentage of poly (vinyl chloride) composite," *Iraqi Journal of Chemical and Petroleum Engineering*, vol. 18, no. 2, pp. 27–39, 2017.
- [20] A. Mallik, A. K. Barik, and B. Pal, "Comparative studies on physico-mechanical properties of composite materials of low density polyethylene and raw/calcined kaolin," *Journal of Asian Ceramic Societies*, vol. 3, no. 2, pp. 212–216, 2015.
- [21] H. A. A. Omar, "Water sorption and solubility of resin filled composites," in *A desertion for the degree of Master of Science in Dental Science in Restorative Dentistry at the Faculty of Dentistry*, pp. nn38–nn48, University of the Western Cape, 2015.