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Characterization, Physicomechanical and Corrosion Properties of Reinforced Microcrystalline Cellulose Treated Silane Unsaturated Polyester Primer Coatings

Adzrie Baharudin, Zuliahani Ahmad*, Ayu Azellia Anuar, Nur Dina Adlina Azmani, Angelene Betsy Anak Mura, Siti Hana Humaira Rustam, Mohamed Syaheen Azhar, Siti Nor Din

Faculty of Applied Sciences, Universiti Teknologi MARA Cawangan Perlis, Kampus Arau, 02600 Arau, Perlis, Malaysia

**Corresponding Author's E-mail: zuliahani@uitm.edu.my*

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ABSTRACT

Corrosion has been a major universal issue due to failure of building structures and high maintenance. The utilization of unsaturated polyester (UPE) as primer coating is insufficient to inhibit the corrosion. Hence, microcrystalline cellulose (MCC) had been incorporated into UPE as a reinforcement filler for the production of primer coatings, due to its high crystalline region in the structure. The objectives of this study were to investigate the physical, mechanical, and corrosive properties of various MCC loadings (0, 3, 6, 9, 12 and 15 wt%) reinforced UPE applied on metal substrates and determine the effect of silane treatment (MCC-APTES) on MCC reinforcement used in UPE-MCC coating for primer application. The coating properties were studied for adhesion, mechanical and corrosive properties using pencil hardness, adhesion tape and immersion tests. At 6% MCC-APTES loading showed improvement in mechanical and adhesion properties at 6H grade and less than 5 % pull out test for pencil hardness and adhesion tests respectively. Whilst the immersion test demonstrated that at 6% MCC-APTES of loading gives optimum corrosion resistant where no corrosion found as compared to the control sample as supported by the improved interaction shown in FTIR spectrum. Therefore, it was evidenced that at 6% of MCC-APTES has significant potential as a reinforcement filler *and promotes enhanced mechanical, adhesion, and corrosion properties as compared to other formulations.*

Keywords: Polyester Primer Coating; Microcrystalline Cellulose; APTES Silane Coupling Agent; Immersion Test

INTRODUCTION

Corrosion occurs when metals undergo a chemical reaction in response to a certain external environmental factor such as temperature, acidity, and humidity, which determines the extent of the material's deterioration/damage that can be costly to fix [1]. According to Anwo *et al.* [2] metal corrosion faces various obstacles including decreased yield, ductility, downtime of operational facilities, ultimate strength, and collapsed constructions with disastrous consequences. In addition to its detrimental impact on the environment, corrosion also presents a significant safety hazard that has widespread implications for many industries worldwide [3-5]. To prevent metal corrosion, the use of vital materials, inhibitors, and optimal preventive measures such as coating is required. Based on Tiwa *et al.* [6], coating consists of metallic such as zinc alloy or chromium coatings, inorganic like phosphate or organic like primers and paints. Coatings provide a variety of benefits such as corrosion or wear resistance, improved surface roughness, thermal or electrical insulation and improved wettability [7]. Meanwhile, primer coating is a base coat that is applied to the paint surface before painting. Its purpose is to adhere the finish coats to the surface, resulting in a strong bond between the substrate and the paint and acting as the last barrier protection. Many based materials of primers are commonly used such as epoxy, polyester, and polyurethane primers.

UPE resin is a co-polyester consists of ester group in the main chain that gained interest on the coating application that can provide good chemical resistance to protect the surface from degradation and corrosion [8]. It also easy to be process and the same time low cost material compare with another polymer [9]. Nevertheless, it possesses certain disadvantages, including a low modulus, brittleness, non-biodegradable and susceptibility to breaking deformation due to its poor heat stability. To overcome these issues, UPE resin reinforced MCC was proposed. It is believed that MCC addition leads to enhance strength, stiffness, and dimensional stability of the composite material as well as facilitates the dispersion of the subsequent materials [8]. Previous studies have shown that the addition of MCC as a filler improves the mechanical adhesion and corrosion resistance of the UPE composite [10]. Following Platnieks *et al.* [11], due to its great biodegradability, high strength (young's modulus of 86-163 GPa) and low cost, MCC is recognized as an ideal reinforcement filler to obtain enhanced composites.

However, the hydrophilic nature of the MCC surface hinders the formation of uniform composites. To achieve the uniformity of the matrix surface, the tension between phases can be reduced so the unsaturated bonds can be introduced to the filler structure [12]. Hence, the addition of a silane coupling can improve the mechanical properties and adhesion between the resin and MCC. It can modify the surface properties of the filler and give a synergistic effect of a chemical bonding with physical interaction resulting in satisfactory bond properties at the interface [13]. Silane treatment strengthens the bond between the components by improving adhesion at the interface, resulting in improved mechanical strength and swelling behaviour of the composite material [1]. 3-aminopropyl trimethoxysilane (3-APTES) coupling agent is known as an amphiphilic surfactant where one end of the molecule can react with inorganic particles after hydrolysis while the other end can react with polymers leading to properties improvement between filler particles and polymers' interfaces [14].

In this work, UPE was filled with various loading of MCC and MCC treated with 3 APTES primer coating in order to enhance compatibility via sonication mixing method. The physicomechanical properties was determined using pencil hardness and cross-cut adhesion tests. Whilst, the corrosion protection was examined by the immersion test.

EXPERIMENTAL METHODOLOGY

Material and Chemicals

The chemicals and materials used in this research were UPE resin having average 2000 g/mol of molecular weight with 40% of styrene content, microcrystalline cellulose (MCC) approximately 50 µm of particle size, methyl ethyl ketone peroxide (MEKP) with 8.8-9% of active oxygen and 3- aminopropyl triethylene 3-APTES which is a clear to yellow pale colour liquid with 221.37 g/mol of molecular weight.

Surface Treatment of MCC

Firstly, 0.25 g of 3-APTES was dissolved in a 30:70 (v/v) acetone/water solvent and added with 5 g of MCC. The acetone solution was allowed to sit at room temperature for 24 hours before being filtered using distilled water. The treated MCC was allowed to dry at ambient temperature after an hour of drying at 100 °C in an oven [15].

Preparation of Wet Paint on Mild Steel Plate

The polyester primer coating with various loadings of MCC and MCC-APTES (3-15%) were mixed and homogenised using sonicator for 1 hour. The application of prepared primer coatings was performed using a hand brush and allowed to dry for a week at room temperature. The sample underwent a 24-hour conditioning period before further testing.

Characterization

The FT-IR spectra of the materials was acquired by means of Thermo Electron Corporation Nicholet 380 Spectrometer over a range of 400–4000 cm^{-1} with a resolution of 4 cm^{-1} and employing the KBr pressed pellet technique.

Physicomechanical Test

The hardness test was conducted to assess the scratch resistance and penetration of the surface layer following its application in accordance with ASTM D3363. A pencil with different types of leads ranged from 2B, B, HB, H, 2H, 3H, 4H, 5H and 6H was used. The adhesion test was performed following ASTM D3359. This technique involved covering cuts on the coating films with pressure-sensitive tape and then removing it to measure the coating's adherence to metal substrates. The rating scales generally used by this method range from 0B to 5B. Two replicates of steel plate used for each samples and cross cuts was created on coating of 0 to 5 scales in each direction to form a lattice pattern. The area was applied with Scotch tape and firmly pressed to ensure all the area was covered.

Corrosion Test

Immersion test was conducted based on the ASTM D6943 (standard practice for immersion of protective coating). First, each UPE primer coated plate was scratched with a 'X' mark before being immersed in a corrosion environment containing 3.5 % NaCl solution. The plate was placed in room condition for 9 days at room temperature. At three-day intervals, the coating condition for the corrosion rate of the plate was observed.

RESULTS AND DISCUSSION

FTIR

Based on the FTIR spectrum shown in Figure 1, MCC showed a broad O-H bonding in range of 3200-3600 cm^{-1}, while the C-O-C stretching was appeared at 1150 cm^{-1} from the glycosidic linkage in the MCC structure that improved the stiffness and mechanical strength of the composite. The strong linkage between glucose unit with well-ordered arrangement that increased crystallinity promoted increased mechanical properties as recorded [15]. Upon the association of MCC with unsaturated polyester resin, the C-H stretching peak of MCC showed the red shift of the peaks to 3500 cm^{-1} which indicates the presence of O-H groups in the UPE resin. While the present peak at 1740 cm^{-1} and 3150 cm^{-1} identified as C=O stretching and aromatic C-H stretching respectively as the UPE main backbone which had strong aromatic bonding exhibit hydrophobicity that produced crosslink network act as physical barrier toward electrolyte [16]. It was in line with the finding reported by Eivaz *et al.* [17] that polyester gives a good corrosion protection for the metal substrate despite of lower cost and easy to process.

MCC was treated with APTES in order to enhance the interaction between MCC and the polyester. The presence of spectrum in range of 3300-3500 cm^{-1} indicate N-H stretching from the APTES and acting as active site for interaction with oxygen to produce hydrogen bonding in between the UPE and APTES [18]. The presence of amine in the composite also act as corrosion inhibitor by blocking the transfer of electron that preventing oxidation reaction as reported by Pourhashem *et al.* [19]. While peak was observed at 1070 cm^{-1} indicates reaction of APTES with MCC create bonding that can increased the interaction between filler and polyester as bridging in line with reported by Hasniraaiman et al. [20] that polyester reacted with filler in range of 1045-1095 cm^{-1}. Meanwhile the MCC treated with APTES was recorded having lower intensity of O-H bonding at 3500 cm^{-1} as compared to the untreated MCC due to interaction with the APTES as proposed in Figure 2.

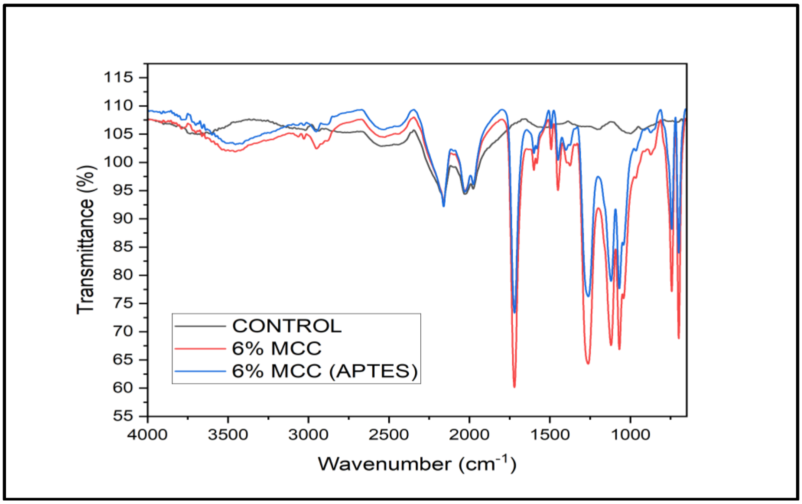


Figure 1: FTIR spectra of control, MCC before and after treated with silane.

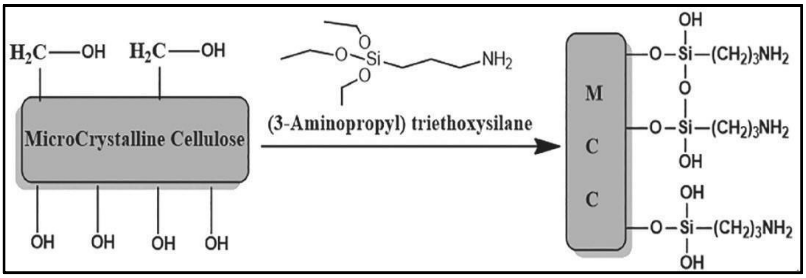


Figure 2: Interaction of MCC before and after treated with silane.

Hardness test

All the sample tested using the same pencil brand with different grades at constant loading for MCC and MCC-APTES at various loading is exhibited in Table 1. The control sample had a pencil hardness grade of H, which is the lowest level of hardness compared to the other formulations. The inclusion of MCC resulted in significant enhancement of the pencil hardness grades observed for all samples, ranging from H grade to 4H and 5H for samples containing 3% MCC and 6% MCC, respectively. This improvement can be attributed to the filler's ability to restrict the movement of the molecular chains within the structure, thereby preventing any displacement when subjected to external force [3]. In addition, the presence of high crystalline region of MCC gives improved mechanical performance of the coating. At an addition level of 6% MCC, the optimum hardness result observed was 5H, in comparison to the control sample. However, the hardness value decreased to 4H and 3H for 9% - 15% MCC, attributed to the increased filler loading that tends to agglomerate. It is believed, MCC is a low-density material which hard to be dispersed in high viscosity of liquid. Hence, the addition of silane coupling agent improved the compatibility of the UPE and MCC that shown better improvement for all treated MCC loading. The inclusion of MCC-APTES at 3% loading and 6-15% loadings recorded 5H and 6H of hardness respectively. It is due to a better interaction between UPE and MCC due to presence of APTES that create the bridge between these materials [4] as proposed in Figure 2.

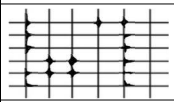
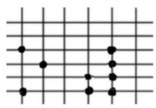
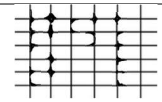
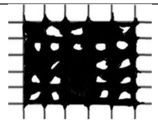
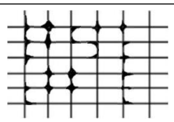
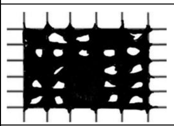
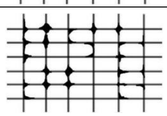
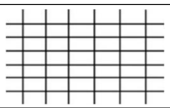
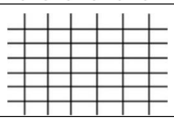
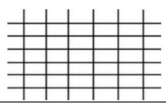
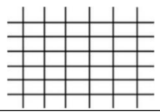
Table 1: The hardness test results of the coating on the steel plate.

Sample	Hardness Results	Sample	Hardness Results
Control	H	-	-
3% MCC	4H	3% MCC-APTES	5H
6% MCC	5H	6% MCC-APTES	6H
9% MCC	4H	9% MCC-APTES	6H
12% MCC	3H	12% MCC-APTES	6H
15% MCC	3H	15% MCC-APTES	6H

Adhesion test

Cross-cut adhesion was conducted to determine how strongly a coating adheres to a substrate. Table 2 shows the results of the samples of MCC and MCC-APTES at various loadings 0-15%. The untreated samples regardless of MCC loadings give a scale from 1B to 4B with the percentage of area removed up to 65% showed that untreated samples have poor interaction between the constituents' materials. It is believed that the absence of a coupling agent has led to the reduction of bonding strength between the coating and the substrate as supported by Sivakumar *et al.* [21]. Therefore, surface modification is crucial as it is possible to create a strong bonding between organic and inorganic materials with the silane coupling agent [21]. The absence of chemical interaction leads to the detachment of bigger flakes during adhesion testing, as a consequence of inadequate adhesion. This lack of adhesion hinders the consistent adherence of the coating. On the other hand, MCC-APTES filled UPE shows that the areas that were removed was between 5% and 15%, and the rating scale is from 3B to 5B, respectively. According to Parhizkar *et al.* [18] MCC modified with APTES allowed the surface modification process to occur on the MCC itself allowing enhanced crosslinking and make them more difficult to be removed from the substrate. This is in line with finding from Baharudin *et al.* [10], who reported that the increased amount of MCC, which is 4% wt of MCC, showed a low corrosion rate which is from 1B grade 35-65% pull out to 4B grade which is less than 5 % pull out as compared to the control sample. The MCC and the APTES interact strongly and effectively as a result of the increased availability of functional groups. Furthermore, silane interacts with polymers to generate chemical bonds and intermolecular polymers, enhancing polymer adherence [21]. Additionally, it reacts with inorganic surfaces to establish metal-siloxane covalent bond, resulting in superior adhesion between silanes and inorganic substrate [18]. Besides, silane coupling agent also frequently consists of reactive groups like alkoxy which can hydrolyse to produce silanol (Si-OH) groups [22-23]. On the MCC surface, the silanol groups then react with the hydroxyl groups to produce siloxane linkages (Si-O-Si). Subsequently, the MCC's surface is successfully activated by this reaction and generates more areas for further adhesion.

Table 2: The adhesion test result for each protective coating sample.

Sample	Result	Analysis	Grade
Control		Percent of removed area: 5% to 15%	1B
3% MCC		Percent of removed area: less than 5%	4B
6% MCC		Percent of removed area: 5% to 15%	3B
9% MCC		Percent of removed area: 35% to 55%	1B
12% MCC		Percent of removed area: 35% to 65% of the coating	1B
15% MCC		Percent of removed area: 35% to 55% of the coating	1B
3% MCC-APTES		Percent of removed area: 5% to 15% of the coating	3B
6% MCC-APTES		The edges of the cuts are completely smooth; none of the squares or the coating are detached.	5B
9% MCC-APTES		The edges of the cuts are completely smooth; none of the squares or the coating are detached.	5B
12% MCC-APTES		The edges of the cuts are completely smooth; none of the squares or the coating are detached.	5B
15% MCC-APTES		The edges of the cuts are completely smooth none of the squares or the coating are detached.	5B

Immersion Test

Table 3 exhibits the corrosion properties of untreated and treated MCC UPE primer coatings samples. It is apparent that the addition of MCC at 3-9% contributed to less corrosion as compared to the control sample. It is because, MCC provides a homogeneous and cohesive layer, which could function as a barrier thus giving the coated surface some water resistance [24]. However, further addition of MCC at a concentration of 12-15% resulted in the formation of corrosion in comparison with the control sample. The uneven surface condition of the samples containing a significant amount of MCC is responsible for their greater susceptibility to agglomerate, hence, indirectly promote the corrosion activity on the sample's surfaces [11]. The indirect promote of the corrosion activity on the sample's surfaces was also supported by Hiremath *et al.* [23], who reported that a large amount of MCC had delayed disintegration as tablet hardness increases and also may drive the formation of agglomeration of MCC. The agglomerates also hinder the silane coupling agent from being able to fully treat the surface and reduced the corrosion resistance. The increased loading of MCC facilitated the direct penetration of water towards the metal surface due to incomplete reaction between UPE and MCC, resulting in the formation of agglomerates. Additionally, the corrosive media, such as oxygen, water, and chloride ions, can reach the substrate or coating interface through diffusion channels, therefore leading to low corrosion resistance in the sample [22].

However, with the addition of MCC-APTES resulting lower corrosion occurred at 6% MCC-APTES and 9% MCC-APTES due to improvement from the silane coupling agent that enhance the interaction between polyester and MCC that lowering the corrosion. The ability of MCC-APTES to form a film helps protect the substrate from moisture-related damage and inhibit water penetration in immersion tests where the coating is exposed to liquids. Therefore, any corrosion reaction on metal surfaces may be prevented as its protection layer can act as a corrosion barrier. Sample at 6% MCC-APTES loading exhibit optimum formulation on corrosion protection. The corrosion resistance of the samples was diminished by the presence of agglomerations caused by the higher loading of MCC-APTES above 9% MCC-APTES.

Table 3: The immersion of 3- and 9-days test result for each protective coating sample.

Sample	3 Days		9 Days	
3-MCC		There was a corrosion occurred on an "X" scratched and the edge of plate		There was a corrosion occurred on an "X" scratched and the edge of plate
6-MCC		There was a corrosion occurred on an end "X" scratched		There was a corrosion occurred on the edge of plate
9-MCC		There was a corrosion occurred on the end edge of plate		There was a small corrosion occurred on the edge of plate
12-MCC		There was a corrosion occurred on an "X" scratched and the edge of plate		There was a corrosion occurred on an "X" scratched and the edge of plate
15-MCC		There was a corrosion occurred on an "X" scratched and the edge of plate		There was a corrosion occurred on an "X" scratched and the edge of plate
3%MCC-APTES		There was a corrosion occurred on an "X" scratched.		There was a corrosion occurred on an "X" scratched.
6% MCC-APTES		There was no corrosion occurred.		There was no corrosion occurred.
9% MCC-APTES		There was a corrosion occurred on an "X" scratched and the edge of plate		There was a corrosion occurred on an "X" scratched and the edge of plate
12% MCC-APTES		There was a corrosion occurred on an "X" scratched and the edge of plate		There was a corrosion occurred on an "X" scratched and the edge of plate

CONCLUSION

In conclusion, the MCC treated silane coupling agent primer coatings were successfully developed for mechanical and corrosion protection of the metal substrates. It was observed that the loading effect (0-15 wt %) of MCC and silane treated MCC-APTES filler in UPE via mechanical mixing have an effect on physical, mechanical and corrosion properties of the primer coating. At 6% MCC-APTES loading showed improvement in mechanical properties where form H grade to 6H grade as compared to control sample. It was also recorded an improvement of adhesion test from 1B grade which is 35-65% removed to 4B grade which is less than 5 % removed for 6% MCC-APTES and control sample, respectively. Whilst immersion test exhibited that optimum loading at 6% MCC contributes to improved corrosion resistant as compared to the control sample. Thus, it was concluded that 6% MCC-APTES loading is the optimum formulation due to enhance mechanical, adhesion and corrosion resistant properties in comparison with other formulations.

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