SEMARAK ILMU
PUBLISHING
202103268166(003316878-P)

Journal of Advanced Research in Applied
Sciences and Engineering Technology

Journal homepage:
https://semarakilmu.com.my/journals/index.php/applied_sciences_eng_tech/index
ISSN: 2462-1943

# Effects of Poly(vinylchloride)-Maleic Anhydride as Coupling Agent on Mechanical, Water Absorption, and Morphological Properties of Eggshell Powder Filled Recycled High Density Polyethylene/Ethylene Vinyl Acetate Composites

Nur Farahana Ramli[1], Supri Abdul Ghani[1], Teh Pei Leng[1,2,*], Yeoh Cheow Keat[1,2]

[1] BSL Technologies Sdn Bhd, Bandar Sri Damansara, PJU 9, 52200 Kuala Lumpur, Malaysia
[2] Faculty of Chemical Engineering & Technology, Universiti Malaysia Perlis (UniMAP), Arau, Perlis 02600, Malaysia
[3] Frontier Materials Research, Centre of Excellence (FrontMate), Universiti Malaysia Perlis (UniMAP), Perlis, Malaysia

**ABSTRACT**

The recycled high density polyethylene (rHDPE)/ethylene vinyl acetate (EVA)/ eggshell composites using poly(vinylchloride)-maleic anhydride (PVC-MA) as coupling agent was compounded using Brabender internal mixer. The results showed that the compatibilization of rHDPE/EVA blends was enhanced by the addition of 6 phr of PVC-MA. Furthermore, the tensile, water absorption resistance, and interfacial adhesion properties were studied. Tensile properties such as tensile strength and Young's modulus were improved, while the elongation at break was reduced by the addition of PVC-MA. The water absorption resistance was improved upon PVC-MA addition to the composites.

***Keywords:***
Compatibilization; Ethylene vinyl acetate; Fillers; Eggshell powder; Recycled HDPE

**Received: 14 June 2022** **Revised: 24 August 2022** **Accepted: 31 August 2022** **Published: 14 Sept. 2022**

## 1. Introduction

Polymer blending can provide the required properties than synthesizing new polymers. It has been recognized as an important route to novel and useful polymeric products with properties superior to those of the individual components. With increasing understanding of the science and engineering involved, polymer blend is expected to offer increasing contributions to the plastic industries [1-2]. High density polyethylene (HDPE) is a thermoplastic material which is a comprehensive resin and having unique properties such as excellent mechanical properties, ozone resistance, good electrical properties, and chemical resistance and has low price [3-4]. Ethylene vinyl acetate (EVA) is a thermoplastic resin made of ethylene monomer and vinyl acetate monomer. It shows high impact strength, stress crack resistance, good ageing resistance, low temperature

---

[*] *Corresponding author.*
*E-mail address: plteh@unimap.edu.my*

https://doi.org/10.37934/araset.28.1.3343

flexibility, improved clarity, permeability to oxygen and vapors, high moisture absorption, and high electrical resistance [3-4].

Blends of HDPE and EVA are widely used is shrinkable films, multilayer packaging, and wire and cable coating since this blend couples the superior properties of both materials. However, because of difference of chemical structure between HDPE and EVA, HDPE is difficult to compatibilize with EVA. Compared to HDPE, EVA is more polar and less crystalline due to polar vinyl acetate (VA) group and leads to better compatibility with polar polymers and fillers [5]. The common technique used is to add small amount of compatibilizer due to their ability to improve both physical and chemical interaction between the components [6].

Filler reinforced polymer composites find potentials over traditional engineering materials [7]. Composites based on polyolefins and mineral fillers have been extensively studied, specifically those ones based on polyethylene and inorganic fillers are of great research interest [8]. Inorganic fillers are reported to improve mechanical, thermal and electrical properties of some semicrystalline polymers, when they are used as reinforcing agents [9]. Besides, the purpose of incorporating fillers into polyolefin is to decrease cost and reduce the consumption of petroleum-based polymer.

The use of natural fillers as reinforcement to form biocomposite is attractive since it is based on abundant and renewable resources [10-12]. This is more interesting since in Malaysia there are abundant chicken eggshell which is a good source of calcium carbonate. Eggshells are generated in thousands or millions of tones every year. These characteristics qualify eggshell as excellent candidate for bulk quantity, inexpensive, low load bearing composite applications such as the automotive industry, trucks, homes, offices, and factories. Furthermore, using the eggshell from agricultural waste not only reduce waste but also reduce the energy consumed for manufacturing materials with readily available and inexpensive new reinforcement for thermoplastic polymer composites. A concern in polymer/eggshell composites have been started since the last few years as a new inorganic material that can be used as filler in polymer composite [13-17]. A dry eggshell has been reported to contain approximately 95% (by weight) with a typical mass of 5.5 grams in the form of calcite.

Attempts have been made to recycle the post-consumer plastics in order to reduce the environmental impact and consumption of virgin plastics. Previous studies show that the properties of the recycled high density polyethylene (rHDPE) with regenerated tire rubber as reinforcement thus could be used for various applications [18]. Also, the recycled plastics are cheaper than in the virgin form, for example, rHDPE pellets and flakes are 31-34% less expensive than the virgin HDPE. Thus, increased use of recycled plastics offers the prospect of lessening waste disposals and reducing the product costs.

Therefore, this paper presents the results of the effect of polyvinylchloride-maleic anhydride (PVC-MA) coupling agent to mechanical, water absorption, and morphological properties of the composites formed by mixing polymer blend of rHDPE and EVA with chicken eggshell powder (ESP).

## 2. Methodology

### *2.1 Materials*

rHDPE pellets with a melt flow index of 0.7 g/10 min (190°C) and a density of 939.9kg/m$^3$ and EVA contains 6.5 wt.% VA, with melt index of 2.5g/10 min (80°C; 2.16kg) and density of 0.93g/cm$^3$ was supplied from A.R. Alatan Sdn. Bhd., Kedah Darul Aman, Malaysia. The chicken eggshells were obtained from local food industry in Negeri Sembilan, Malaysia. Poly(vinylchloride) (PVC) and maleic anhydride (MA) powder was purchased from MERCK company while dicumyl peroxide (DCP, purity

98%) used as an initiator was purchased from Aldrich, and its half-lifetime at the melt grafting temperature (175°C) was about 1.45 min.

### *2.2 Preparation of Eggshell Powder*

The eggshells were washed and crushed into small pieces. Then they were dried in an oven at 80°C until the weight is constant to evaporate all moisture. The dried eggshells were grinded to powder using kitchen blender, which is designated ESP. Sieve was used to obtain an average filler size of 63**μ**m. ESP was dried in vacuum oven at 80°C till the constant weight to make it free from moisture as reported in Shuhadah and Supri [11].

### *2.3 Composite Preparation*

For the composite's preparation, the compounding of the blends was carried out by melt blending in an internal mixer (Plasticoder, Brabender). The rHDPE was first mixed in the internal mixer at 190°C, 50 rpm for 4 min and then pre-weighed amounts of EVA, PVC, MA, DCP, and ESP were added to the mixer and the mixing process continued for a further 10 minutes. Each of the molten samples was compression molded into sheets with a thickness of about 2 mm using a hydraulic press at 190°C for 2 minutes and cooled under pressure for 4 minutes. Table 1 presents the formulation and designation of the samples. All the formulations included a matrix of 50 phr rHDPE and 50 phr EVA. 6 wt.% of PVC and MA was added as compatibilizer together with 1% DCP as radical initiator to the compatibilized blends. The proposed interaction between the PVC and MA in the internal mixer is displayed in Figure 1.

**Table 1**
Formulation of rHDPE/EVA/ESP samples

| Composites | rHDPE (phr) | EVA (phr) | ESP (phr) | PVC-MA (phr) | DCP (phr) |
|---|---|---|---|---|---|
| rHDPE/EVA | 50 | 50 | - | - | - |
| rHDPE/EVA/ESP5 | 50 | 50 | 5 | - | - |
| rHDPE/EVA/ESP10 | 50 | 50 | 10 | - | - |
| rHDPE/EVA/ESP15 | 50 | 50 | 15 | - | - |
| rHDPE/EVA/ESP20 | 50 | 50 | 20 | - | - |
| rHDPE/EVA/ESP25 | 50 | 50 | 25 | - | - |
| rHDPE/EVA/ESP5$_{PVC\text{-}MA}$ | 50 | 50 | 5 | 6 | 1 |
| rHDPE/EVA/ESP10$_{PVC\text{-}MA}$ | 50 | 50 | 10 | 6 | 1 |
| rHDPE/EVA/ESP15$_{PVC\text{-}MA}$ | 50 | 50 | 15 | 6 | 1 |
| rHDPE/EVA/ESP20$_{PVC\text{-}MA}$ | 50 | 50 | 20 | 6 | 1 |
| rHDPE/EVA/ESP25$_{PVC\text{-}MA}$ | 50 | 50 | 25 | 6 | 1 |

rDHPE: recycled high-density polyethylene; EVA: ethylene vinyl acetate; ESP: eggshell powder; PVC-MA: poly(vinylchloride)-maleic anhydride.
Note: *6 phr of benzyl urea was added together with 1 phr of DCP and the filler loading

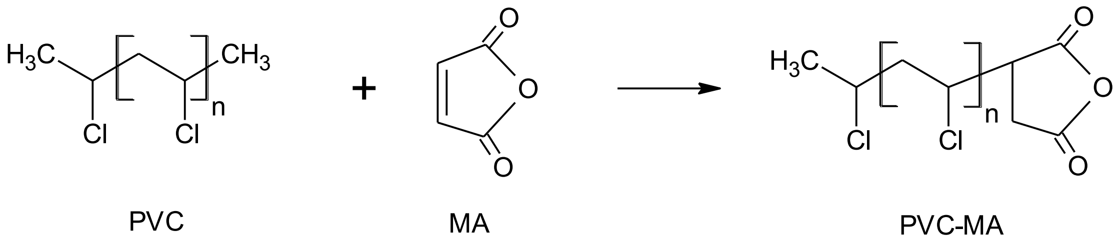


**Fig. 1.** Proposed interaction between the polyvinyl-chloride and maleic anhydride

### *2.4 Tensile Test*

Tensile properties were determined according to ASTM D638 by using the Instron 5569 with a crosshead speed of 50 mm/min. Five dumb-bell shaped samples were used for each blend composition. Tensile strength, elongation at break, and Young's modulus of each composite were obtained from the test.

### *2.5 Water Absorption Analysis*

Water absorption test was carried out according to ASTM D750-95 standard. It involved total immersion of five samples in distilled water at room temperature. All the specimens were previously dried in an oven at 50°C for 24 h and stored in desiccators. The water absorption was determined by weighing the samples at regular interval. A Mettler balance type AJ150 was used with precision of ±1 mg. The percentage of water absorption ($M_t$) was calculated using Equation 1, where $W_d$ and $W_N$ are original dry and weight after exposure, respectively.

$$M_t = (W_N - W_d)/W_d \times 100\% \qquad (1)$$

### *2.6 Scanning Electron Microscopy*

Morphology of the tensile fracture surface of rHDPE/EVA/ESP composites with and without maleic anhydride were carried out using a SEM, model JEOL JSM 6460LA. Surfaces of the samples were coated with a thin palladium layer of about 12 µm thickness using Auto Fine Coater, model JEOL JFC 1600.

## **3. Results**

### *3.1 Tensile Properties*

Figure 2 shows the effect of filler loading on the tensile strength of rHDPE/EVA/ESP and rHDPE/EVA/ESPPVC-MA composites. The tensile strength for both composites gradually decreased from 25.30 MPa for the rHDPE/EVA blends without the filler to 20.09 MPa for rHDPE/EVA/ESP5, showing a 20.3% decrease. Further of the filler loading in the rHDPE/EVA/ESP composites caused a decrease of the tensile strength to 18.14 MPa, 15.80 MPa, 15.31 MPa, and 13.11 MPa, respectively. The decreased of tensile strength by the filler addition was probably attributed to the weak interfacial adhesion and low compatibility between the hydrophilic ESP and hydrophobic rHDPE/EVA. Supri et al [17] reported that as the amount of filler loading increased, the tendency for filler-filler interaction increased rather than filler-matrix interaction to form agglomeration due to the difficulties to achieve homogenous dispersion of filler at higher filler loading.

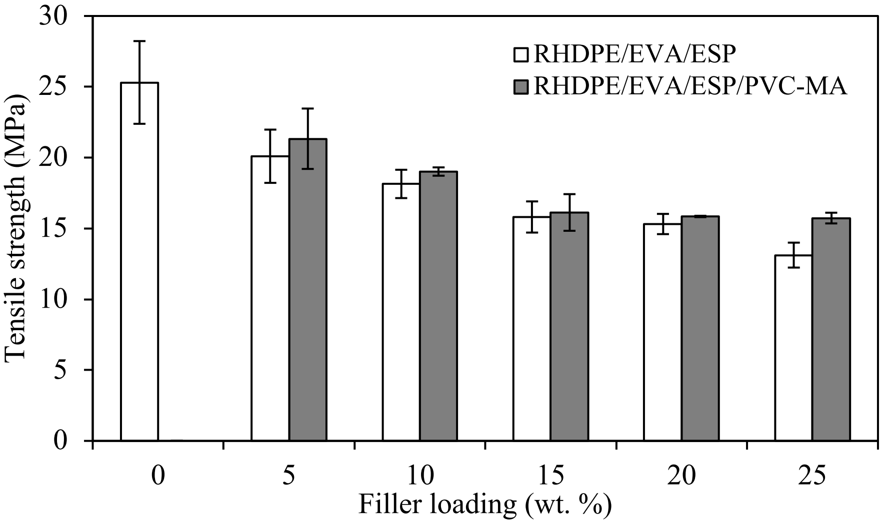


**Fig. 2.** Effect of filler loading on tensile strength of rHDPE/EVA/ESP composites and rHDPE/EVA/ESP$_{PVC-MA}$ composites

However, the utilization of PVC-MA as coupling agent produced some improvements, as the tensile strength of rHDPE/EVA/ESPPVC-MA composites was higher than that of rHDPE/EVA/ESP composites without PVC-MA at all filler loadings. The tensile strength increased from 20.097 MPa for rHDPE/EVA/ESP5 composites to 21.33 MPa, exhibiting an increase of 6.15%. Further increases in the filler loading to 10, 15, 20, and 25 phr enhanced the strength of the rHDPE/EVA/ESPPVC-MA¬ composites by 4.82, 2.02, 3.48, and 19.94%, respectively, compared with that of the rHDPE/EVA/ESP composites. These findings confirmed that the presence of PVC-MA acts as coupling agent which enhanced the interfacial adhesion between the rHDPE and EVA matrix, and hence increased the tensile strength of rHDPE/EVA/ESPPVC-MA composites. The PVC-MA has improved the mechanical capacity of the material by means of a chemical mechanism. This mechanism is based on the establishment of vinyl bonds of the PVC and the C-C aliphatic chains of the rHDPE besides the interaction between PVC and the acetate group of EVA [19]. The proposed interaction between the rHDPE/EVA/ESP with PVC-MA coupling agent is displayed in Figure 3.

Figure 4 shows the effect of filler loading on elongation at break of rHDPE/EVA/ESP and rHDPE/EVA/ESPPVC-MA composites. Upon filler addition, rHDPE/EVA/ESP5 exhibited the highest elongation at break of 625.9%. The composites with 10, 15, 20, and 25 phr of filler loading exhibited elongation at break values of 586.1, 569.76, 550.06, and 502.7%, respectively. Considering these results, the mobility of the polymer chains has been reduced by the addition of rigid fillers. Similar observation had been reported by Vandeginste [20].

On the other hand, a lower value of elongation at break can be observed for rHDPE/EVA/ESPPVC-MA composites compared to rHDPE/EVA/ESP composites at any filler loading. The elongation at break decreased from 625.9% for rHDPE/EVA/ESP5 composites to 612.575%, exhibiting a decrease of 2.13%. Further increases in the filler loading to 10, 15, 20, and 25 phr reduced the elongation at break of the rHDPE/EVA/ESPPVC-MA composites by 14.23, 9.27, 19.87, and 33.32%, respectively, compared with that of the rHDPE/EVA/ESP composites. These results might be attributed to the addition of PVC-MA which had improved the adhesion of rHDPE/EVA, thus increased the stiffness of the composites. Ismail *et al.* [21] reported that the addition of maleic anhydride to the waste PVC/NBR blends showed lower elongation at break, EB compared to the untreated blends.

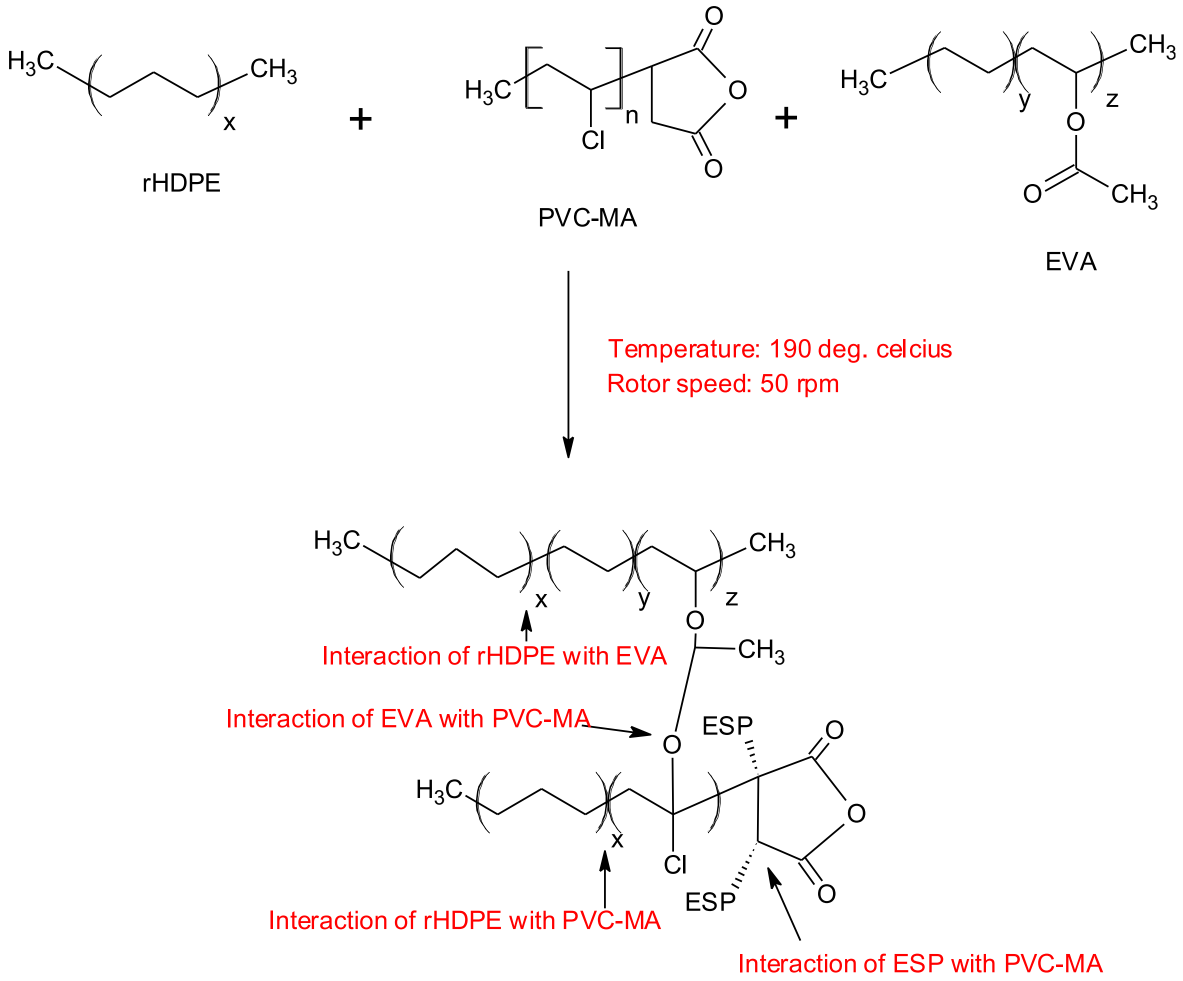


**Fig. 3.** Proposed interaction between the rHDPE/EVA/ESP composites with the PVC-MA coupling agent

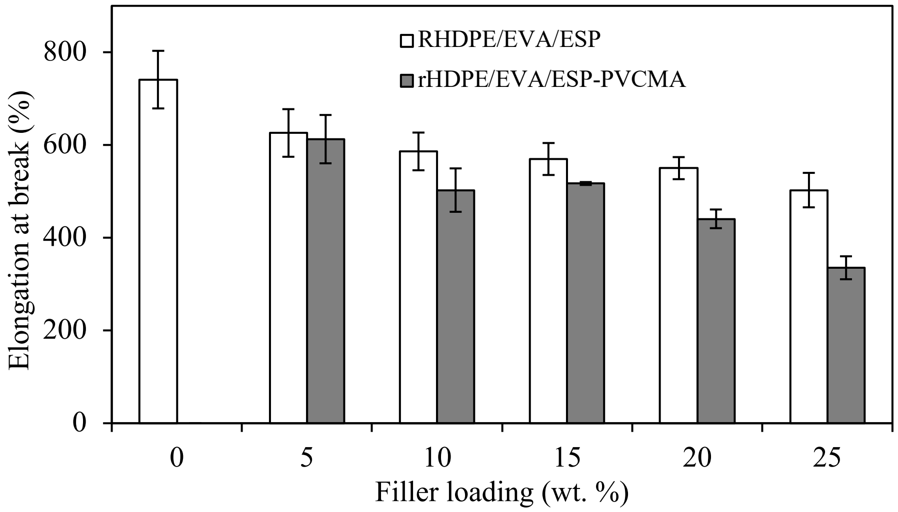


**Fig. 4.** Effect of filler loading on elongation at break of rHDPE/EVA/ESP composites and rHDPE/EVA/ESP$_{\text{PVC-MA}}$ composites

The effect of filler loading on Young's modulus of rHDPE/EVA/ESP and rHDPE/EVA/ESPPVC-MA composites is shown in Fig. 5. The Young's modulus of both composites increased as the filler loading increased. The rHDPE/EVA/ESP25 composite exhibited the highest value of Young's modulus (313.4 MPa). The composites with filler loadings of 5, 10, 15, and 20 phr exhibited values of 233.8, 255.48, 257.35, and 292.9 MPa, respectively. This might be due to the ability of ESP to impart greater stiffness to the rHDPE/EVA matrix. Moreover, ESP possessed higher modulus compared to rHDPE/EVA phases, as a result, the composites provided higher modulus than polymer blends.

At similar filler loadings, the rHDPE/EVA/ESPPVC-MA composites exhibited higher Young's modulus compared to rHDPE/EVA/ESP composites. Young's modulus of rHDPE/EVA/ESPPVC-MA composites increased by 4.64, 2.59, 2.49, 3.0, and 4.08% for filler loadings of 5, 10, 15, 20, and 25 phr, respectively, compared with that of the rHDPE/EVA/ESP composites. As mentioned earlier, the function of PVC-MA as a compatibilizer which enhanced the interaction between the rHDPE and EVA thus produced higher stiffness to the composites. Similar observation found in our previous study using the 3-aminopropyltriethoxysilane as coupling agent for rHDPE/EVA/ESP composites [22].

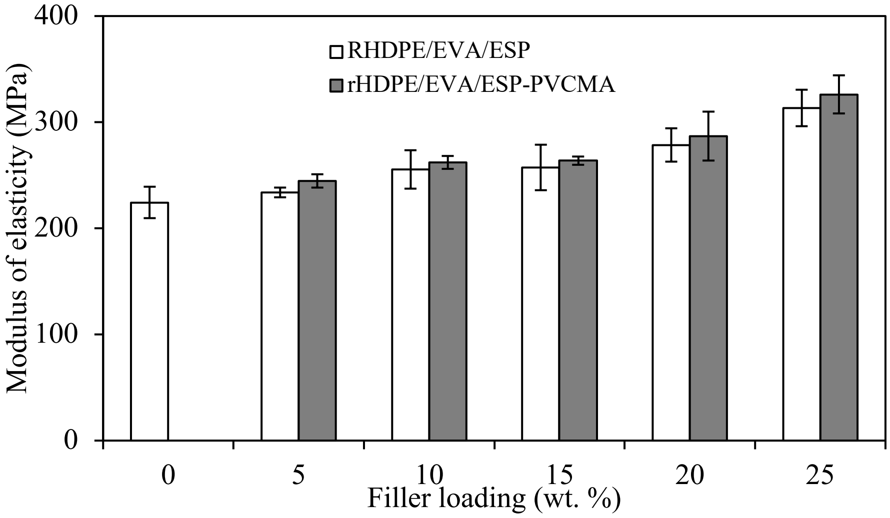


**Fig. 5.** Effect of filler loading on Young's modulus of rHDPE/EVA/ESP composites and rHDPE/EVA/ESP$_{PVC-MA}$ composites

### *3.2 Water Absorption Analysis*

Figure 6 shows the water absorption of rHDPE/EVA/ESP composites. As can be seen, the equilibrium water absorption of both composites increased as filler loading increased. The equilibrium water absorption of rHDPE/EVA/ESP composites increased to 0.08, 0.17, 0.19, 0.26, and 0.33% for filler addition of 5, 10, 15, 20, and 25 phr, respectively. As more filler were added into the polymer matrix, they caused absorption of more water due to the increasing content of the ESP. This is because ESP is a natural material, and its main constituents have hydroxyl groups. These groups are hydrophilic in nature.

Meanwhile, the percentages of the equilibrium water absorption for rHDPE/EVA/ESPPVC-MA composites are lower than uncompatibilized rHDPE/EVA/ESP composites. The equilibrium water absorption of rHDPE/EVA/ESPPVC-MA¬ composites was lower by 50.89, 30.12, 24.42, 24.26, and 12.89% for filler loadings of 5, 10, 15, 20, and 25 phr, respectively, compared with that of the composites without PVC-MA. After compatibilization, some hydroxyl groups of ESP reacted with maleic anhydride and increased compatibility with rHDPE/EVA matrix which can be referred in the proposed interaction as in Figure 3. Therefore, fewer hydroxyl groups were present in the resulting composites, which resulted in less water absorption. This difference was due to good interfacial adhesion between rHDPE/EVA and ESP with the presence of PVC-MA. Consequently, composites with PVC-MA compatibilizer exhibit better tensile strength compared to uncompatibilized rHDPE/EVA/ESP composites. Sellamia et al [23] found that the interaction between PVC and EVA will reduce water and gas permeability due to the low segmental mobility of PVC which enables it to create hydrogen bonds between the hydrogen atoms and the Cl-substituted carbon of PVC with VA carbonyls, which decreases the diffusivity.

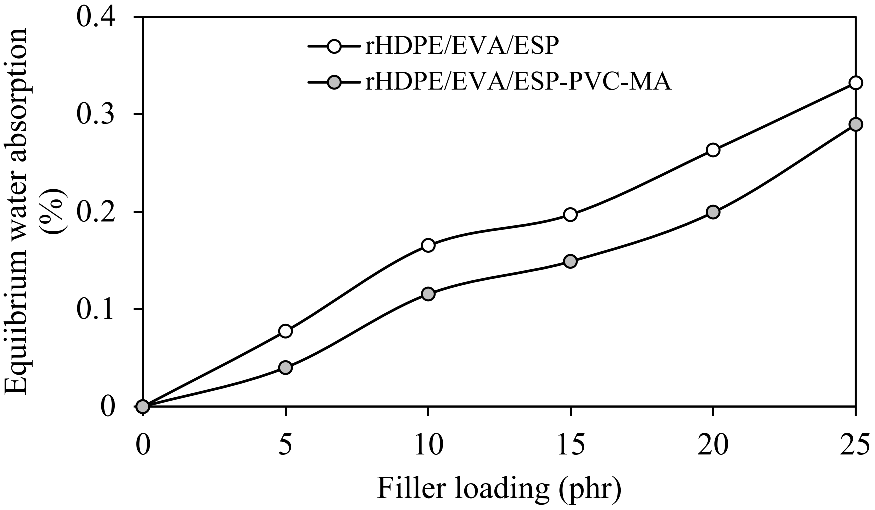
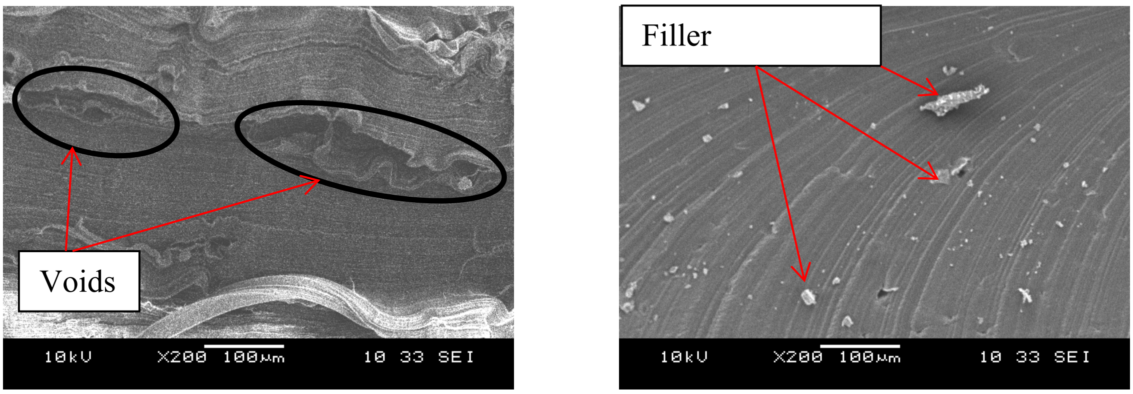


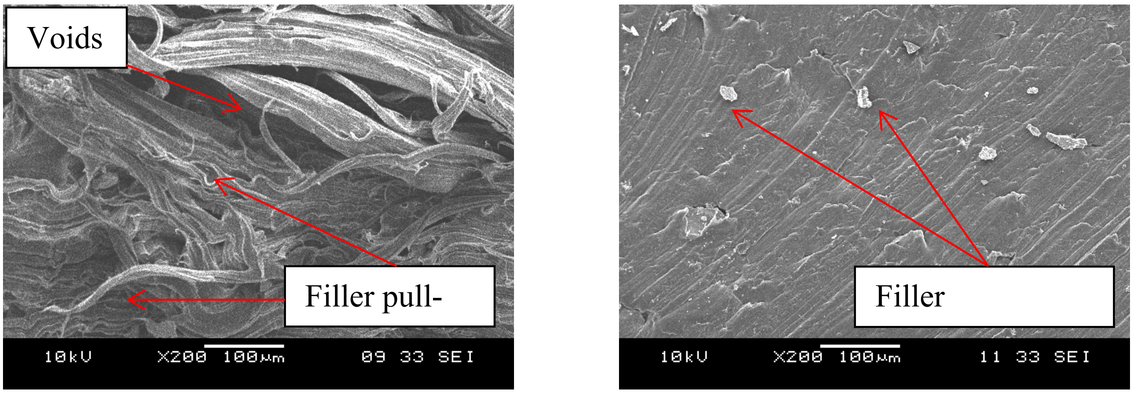
**Fig. 6.** Percentage of equilibrium water absorption versus filler loading for rHDPE/EVA/ESP composites

### *3.3 Scanning Electron Microscopy*

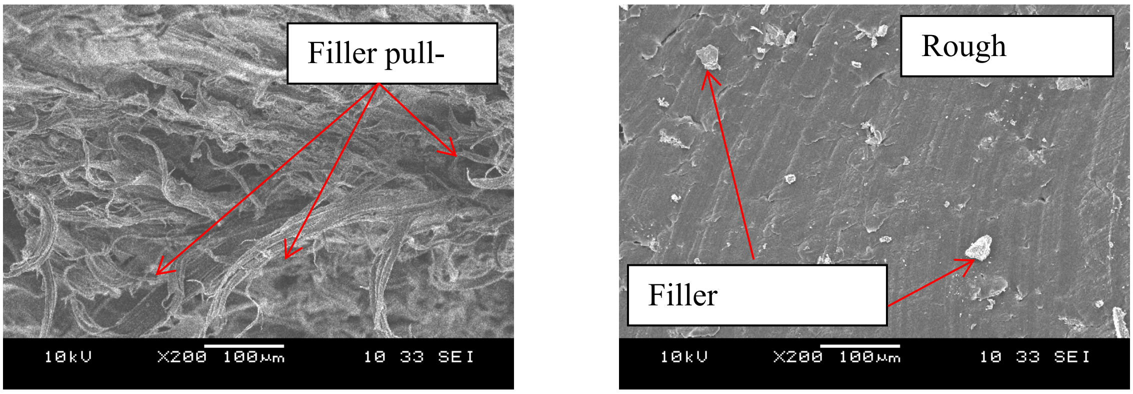
Scanning electron microscopic analysis reveals the surface morphology of the uncompatibilized and compatibilized composites. Figure 7 shows the tensile fracture surface of rHDPE/EVA/ESP and rHDPE/EVA/ESPPVC-MA composites at different filler loading. The uncompatibilized composites present broad interfaces between both components of the composite, characterized by the holes around the fillers and the holes left by them by pull out when the specimens were assayed. Filler pull-out could act as initial flaws leading to localized stress concentration during deformation, finally premature failure of composites occurs and decreases the tensile strength. This result is consistent with a low adhesion of the polymer matrix on the surface of the fillers due to the low compatibility [24]. Fig. 7(b), 7(d) and 7(f) show homogenous materials characterized by a well dispersed reinforcement inside the polymer matrix and by the absence of holes between the matrix and the fillers. This result clearly demonstrated that the compatibilization of the composites provided strong interfacial adhesion and good wetting. The improved interfacial adhesion attributed to the strong chemical interaction between the rHDPE/EVA matrix and PVC-MA and to the strong physical interaction between the PVC and the acetate group of EVA, and the vinyl group of PVC with the long chain of rHDPE. When the coupling agent was situated at the interface between the matrices and interacted with both, the interfacial tension was reduced, and compatibility was increased.



(a) rHDPE/EVA/ESP5 (b) rHDPE/EVA/ESP5$_{PVC-MA}$



(c) rHDPE/EVA/ESP15 (d) rHDPE/EVA/ESP15$_{PVC-MA}$



(e) rHDPE/EVA/ESP25 (f) rHDPE/EVA/ESP25$_{PVC-MA}$

**Fig. 7.** SEM micrograph of tensile fracture surface of (a) rHDPE/EVA/ESP5, (b) rHDPE/EVA/ESP5$_{PVC-MA}$, (c) rHDPE/EVA/ESP15, (d) rHDPE/EVA/ESP15$_{PVC-MA}$, (e) rHDPE/EVA/ESP25, and (f) rHDPE/EVA/ESP25$_{PVC-MA}$ composites

## 4. Conclusion

The investigation on the effect of compatibilizer PVC-MA on the properties of rHDPE/EVA/ESP composites with different filler loadings was done in this study. PVC-MA was confirmed to act as the coupling agent in presence of DCP in rHDPE/EVA composites by creating physical interactions with the filler and new bonding with the rHDPE/EVA matrix. Consequently, the addition of PVC-MA improved the tensile strength, Young's modulus, water absorption resistance, and interfacial adhesion of the composites, although it reduced the elongation at break. From the SEM observations, the composites with PVC-MA seemed to have less pull-out filler, filler voids, and agglomeration, which explained the higher values of tensile strength. The results obtained in this study further support the use of PVC-MA as the coupling agent in rHDPE/EVA composites, showing that eggshell powder has a promising potential for use as reinforcing filler in rHDPE/EVA composites.

## Acknowledgement

The author would like to acknowledge the support from the Fundamental Research Grant Scheme (FRGS) under a grant number of FRGS/1/2018/TK05/UNIMAP/02/13 from the Ministry of Higher Education Malaysia.

## References

[1] Hassan, Azman, Abozar Akbari, Ngoo Kea Hing, and Chantara Thevy Ratnam. "Mechanical and thermal properties of ABS/PVC composites: Effect of particles size and surface treatment of ground calcium carbonate." *Polymer-Plastics Technology and Engineering* 51, no. 5 (2012): 473-479. https://doi.org/10.1080/03602559.2011.651242

[2] Kam, Ka-Wei, Pei-Leng Teh, Hakimah Osman, and Cheow-Keat Yeoh. "Comparison study: effect of un-vulcanized and vulcanized NR content on the properties of two-matrix filled epoxy/natural rubber/graphene nano-platelets system." *Journal of Polymer Research* 25, no. 1 (2018): 1-19. https://doi.org/10.1007/s10965-017-1418-x

[3] Liang, Ji-Zhao, and Fang Wang. "Flexural and impact properties of POM/EVA/HDPE blends and POM/EVA/HDPE/nano-CaCO3 composites." *Polymer Bulletin*72, no. 4 (2015): 915-929. https://doi.org/10.1007/s00289-015-1314-7

[4] Chen, Yang, Huawei Zou, Mei Liang, and Ya Cao. "Melting and crystallization behavior of partially miscible high density polyethylene/ethylene vinyl acetate copolymer (HDPE/EVA) blends." *Thermochimica Acta* 586 (2014): 1-8. https://doi.org/10.1016/j.tca.2014.04.007

[5] Khanal, Santosh, Yunhua Lu, Dandan Jin, and Shiai Xu. "Effects of layered double hydroxides on the thermal and flame retardant properties of intumescent flame retardant high density polyethylene composites." *Fire and Materials* 46, no. 1 (2022): 107-116. https://doi.org/10.1002/fam.2951

[6] Supri, A. G., S. J. Tan, H. Ismail, and P. L. Teh. "Enhancing interfacial adhesion performance by using poly (vinyl alcohol) in (low-density polyethylene)/(natural rubber)/(water hyacinth fiber) composites." *Journal of Vinyl and Additive Technology*19, no. 1 (2013): 47-54. https://doi.org/10.1002/vnl.20315

[7] Aqzna, S. S., C. K. Yeoh, M. S. Idris, P. L. Teh, Khairul Amali bin Hamzah, Y. Y. Aw, and T. N. Atiqah. "Effect of different filler content of ABS–zinc ferrite composites on mechanical, electrical and thermal conductivity by using 3D printing." *Journal of Vinyl and Additive Technology* 24 (2018): E217-E229. https://doi.org/10.1002/vnl.21640

[8] Rahman, Abdul Rozyanty, Pei Leng Teh, Keat Yeoh Cheow, and Kathiravan Suppiah. "Modified Carboxymethyl Cellulose/Halloysite Nanotube (CMC/HNT) Using Sodium Dodecyl Sulfate (SDS)." In *Mineral-Filled Polymer Composites*, pp. 211-229. CRC Press, 2022. https://doi.org/10.1201/9781003221012-11

[9] Aw, Yah Yun, Cheow Keat Yeoh, Muhammad Asri Idris, Pei Leng Teh, Wan Nurshaznie Elyne, Khairul Amali Hamzah, and Shulizawati Aqzna Sazali. "Influence of filler precoating and printing parameter on mechanical properties of 3D printed acrylonitrile butadiene styrene/zinc oxide composite." *Polymer-Plastics Technology and Materials* 58, no. 1 (2019): 1-13. https://doi.org/10.1080/03602559.2018.1455861

[10] Salmah, H., B. Y. Lim, and P. L. Teh. "Melt rheological behavior and thermal properties of low-density polyethylene/palm kernel shell composites: Effect of polyethylene acrylic acid." *International Journal of Polymeric Materials* 61, no. 14 (2012): 1091-1101. https://doi.org/10.1080/00914037.2011.617336

[11] Ilyas, Rushdan Ahmad, Salit Mohd Sapuan, Mohamad Ridzwan Ishak, and Edi Syams Zainudin. "Water transport properties of bio-nanocomposites reinforced by sugar palm (Arenga Pinnata) nanofibrillated cellulose." *Journal of Advanced Research in Fluid Mechanics and Thermal Sciences* 51, no. 2 (2018): 234-246.

[12] Mustafa, Zaleha, Anira Shahidah Razali, Thavinnesh Kumar Rajendran, Siti Hajar Sheikh Md Fadzullah, Sivakumar Dhar Malingam, Toibah Abd Rahim, and Thanate Ratanawilai. "Moisture Absorption and Thermal Properties of Unidirectional Pineapple Leaf Fibre/Polylactic Acid Composites under Hygrothermal Ageing Conditions." *Journal of Advanced Research in Fluid Mechanics and Thermal Sciences* 96, no. 2 (2022): 65-73. https://doi.org/10.37934/arfmts.96.2.6573

[13] Shuhadah, Siti, and A. G. Supri. "LDPE-isophthalic acid modified egg shell powder composites (LDPE/ESPI)." *Journal of physical science* 20, no. 1 (2009): 87-98.

[14] Hussein, Abdullah A., Rusel D. Salim, and Abdulwahab A. Sultan. "Water absorption and mechanical properties of high–density polyethylene/egg shell composite." *Journal of Basrah Researches (Sciences)* 37, no. 3A/15 (2011): 36-42.

[15] Lin, Zhidan, Zishou Zhang, and Kancheng Mai. "Preparation and properties of eggshell/β-polypropylene bio-composites." *Journal of applied polymer science* 125, no. 1 (2012): 61-66. https://doi.org/10.1002/app.34670

[16] Supri, Abd Ghani, H. Ismail, and S. Shuhadah. "Effect of polyethylene-grafted maleic anhydride (PE-g-MAH) on properties of low density polyethylene/eggshell powder (LDPE/ESP) composites." *Polymer-Plastics Technology and Engineering* 49, no. 4 (2010): 347-353. https://doi.org/10.1080/03602550903414035

[17] Ghani, Supri A., and Heah Chong Young. "Conductive polymer based on polyaniline-eggshell powder (PANI-ESP) composites." *J. Phys. Sci* 21, no. 2 (2010): 81-97.

[18] Fazli, Ali, and Denis Rodrigue. "Thermoplastic elastomer based on recycled HDPE/Ground tire rubber interfacially modified with an elastomer: effect of mixing sequence and elastomer type/content." *Polymer-Plastics Technology and Materials* 61, no. 9 (2022): 1021-1038.

[19] Hammiche, Dalila, Amar Boukerrou, Hocine Djidjelli, and Abderrahmane Djerrada. "Effects of some PVC-grafted maleic anhydrides (PVC-g-MAs) on the morphology, and the mechanical and thermal properties of (alfa fiber)-reinforced PVC composites." *Journal of Vinyl and Additive Technology*19, no. 4 (2013): 225-232. https://doi.org/10.1002/vnl.21317

[20] Vandeginste, Veerle. "Food waste eggshell valorization through development of new composites: A review." *Sustainable Materials and Technologies* 29 (2021): e00317. https://doi.org/10.1016/j.susmat.2021.e00317

[21] Ismail, H., and A. M. M. Yusof. "Blend of waste poly (vinylchloride)(PVCw)/acrylonitrile butadiene-rubber (NBR): the effect of maleic anhydride (MAH)." *Polymer Testing* 23, no. 6 (2004): 675-683. https://doi.org/10.1016/j.polymertesting.2004.01.008

[22] Farahana, R. N., A. G. Supri, and P. I. Teh. "Tensile and water absorption properties of eggshell powder filled recycled high-density polyethylene/ethylene vinyl acetate composites: effect of 3-aminopropyltriethoxysilane." *Journal of Advanced Research in Materials Science* 5, no. 1 (2015): 1-9.

[23] Sellami, Ferhat, Ounissa Kebiche-Senhadji, Stéphane Marais, and Kateryna Fatyeyeva. "PVC/EVA-based polymer inclusion membranes with improved stability and Cr (VI) extraction capacity: Water plasticization effect." *Journal of Hazardous Materials* 436 (2022): 129069. https://doi.org/10.1016/j.jhazmat.2022.129069

[24] Vicente, Ana I., João Campos, João Moura Bordado, and M. Rosario Ribeiro. "Maleic anhydride modified ethylene–diene copolymers: Synthesis and properties." *Reactive and Functional Polymers* 68, no. 2 (2008): 519-526. https://doi.org/10.1016/j.reactfunctpolym.2007.10.026