

# The Effect of Surface Modifiers on the Mechanical and Morphological Properties of Waste Silicon Carbide Filled High-Density Polyethylene

R. Dangtungee, A. Rattanapan, S. Siengchin

**Abstract**—Waste silicon carbide (waste SiC) filled high-density polyethylene (HDPE) with and without surface modifiers were studied. Two types of surface modifiers namely; high-density polyethylene-grafted-maleic anhydride (HDPE-g-MA) and 3-aminopropyltriethoxysilane have been used in this study. The composites were produced using a two roll mill, extruder and shaped in a hydraulic compression molding machine. The mechanical properties of polymer composites such as flexural strength and modulus, impact strength, tensile strength, stiffness and hardness were investigated over a range of compositions. It was found that, flexural strength and modulus, tensile modulus and hardness increased, whereas impact strength and tensile strength decreased with the increasing in filler contents, compared to the neat HDPE. At similar filler content, the effect of both surface modifiers increased flexural modulus, impact strength, tensile strength and stiffness but reduced the flexural strength. Morphological investigation using SEM revealed that the improvement in mechanical properties was due to enhancement of the interfacial adhesion between waste SiC and HDPE.

**Keywords**—High-density polyethylene, HDPE-g-MA, mechanical properties, morphological properties, silicon carbide, waste silicon carbide.

## I. INTRODUCTION

POLYOLEFINS are interesting materials that find application in various fields. Polyolefin polymer, such as High-density polyethylene (HDPE) is widely used in the automotive industry and for domestic applications where ductility, low cost and recyclable has to be combined. High-density polyethylene has been used for research and development, both on laboratory and on industrial scale. In addition, fillers are more extensively used in the plastic industry to achieve desired properties or to reduce the price of the finished product. Incorporation of fillers in plastics is a common practice, as it typically reduces the costs and modifies their physical and mechanical properties. This

alteration of properties in plastics depends on the nature of fillers as well as of the plastics [1]. Composites made from particulate fillers are macroscopically isotropic, and the fillers can be either organic or inorganic. The inclusion of stiff inorganic fillers into polymeric materials enhances their physical and mechanical properties, such as modulus, hardness, creep resistance, process ability and improves the dimension stability of the polymer. In some case it was verified that the worsening interfacial bonding between filler (hydrophilic) and polymer matrix (hydrophobic) decreased the tensile strength. The interaction between the component phases may be brought about either by addition of an agent which interacts with both phases and renders them compatible. There are different ways to improving the polymer-filler interaction and dispersion, the following can be cited; the use of compatibilizer [2]-[9], the use of coupling agent [10]-[17]. Maleic anhydride grafting turned out to be the most efficient way to make HDPE sufficiently compatible with organic filler. Thus, HDPE-g-MA was extensively used either as the compatibilizer for HDPE-based composites [18], [19]. It is known that that small amounts of HDPE modified with maleic anhydride to the composites significantly increased the mechanical properties and several studied showed that filler-polymer bonding can be improved by the used coupling agent. Due to many desirable properties, such as light weight, high strength, high thermal conductivity, high hardness, high corrosion resistance, high abrasion resistance, good heat resistance and low thermal expansion. Thus, waste silicon carbide (waste SiC), the by product from abrasive industry, was selected as a reinforcing material. Several studied showed that SiC/polymer composites can increase mechanical properties, thermal conductivity, decrease thermal degradation, and abrasion [20], [21].

In this study, waste SiC was used as a filler in HDPE. The effect of waste SiC content and a surface modifiers, i.e high density polyethylene-grafted-maleic anhydride copolymer (HDPE-g-MA) as a compatibilizer, 3-aminopropyltriethoxysilane as a coupling agent, on mechanical properties and morphology were carried out.

## II. EXPERIMENTAL

### A. Materials

High density polyethylene (HDPE) was supplied by IRPC Public Company Limited (Thailand) and had a melt flow index of 6 g/10 min at 190°C. The density of the polymer is

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0.957 g/cm<sup>3</sup>. The waste SiC particle from abrasive industry was produced by Kyocera Kinseki (Thailand) Company Limited, which has an average particle diameter of 13  $\mu$ m. The maleic anhydride grafted high density polyethylene (HDPE-g-MA) with a melt flow index of 2.05 g/10 min and 0.89% maleic anhydride content was supplied by Creative Polymer Limited (USA). Under the trade name Fusabond E MB-100D. The 3-aminopropyltriethoxysilane with a density 0.95 g/cm<sup>3</sup> was supplied by Jebsen & Jessen Chemicals (Singapore) Ltd. Under the trade name Dynasylan® AMEO.

### B. Sample Preparation

The dried HDPE and HDPE-g-MA were mixed with waste SiC powder. The mass ratio of HDPE to waste SiC was set as 100/0, 95/5, 90/10, 85/15, 80/20 and 70/30. The mass ratio of HDPE/waste SiC/HDPE-g-MA was set as 90/10/0, 85/15/1, 90/10/3, 90/10/5, 90/10/7 and 90/10/1. And the mass ratio of HDPE/waste SiC/3-aminopropyltriethoxysilane was set as 90/10/0, 90/10/0.4, 90/10/0.6, 90/10/0.8, 90/10/1.0 and 90/10/1.2. The polymer composites were melt compounded in two roll mill (LRM-S-110/3E, Labtech Engineering) at a temperature 175°C, then fed into the extruder (2525 SP, Betol). The screw speed was maintained at 50 rpm. The barrel temperatures from hopper to die were kept at 120–175°C. After extrusion, the samples were pelletized and then oven dried before hot compression. The polymer composite samples were prepared by compression molded (LP20-B, Labtech Engineering) for tensile, flexural, hardness and impact testing. The lower and upper mould temperatures were set up at 175°C with the hydraulic pressure 120 bar. The molding process was carried out using a pre heating time of 5 min, pressure was applied for 5 min, and the mould was immediately transferred to a cold press with applied pressure for cooling in 5 min.

### C. Characterization Methods

Tensile tests were carried out according to ASTM D638 on an Instron universal testing machine. Dumbbell specimens of 3 mm thick were cut from the molded sheets. A crosshead speed of 50 mm/min and gauge length of 50 mm were used and the tests were performed at room temperature. Five specimens were used and the average was calculated in each case.

Flexural modulus and strength of the specimens were determined by the same Instron universal testing machine at a crosshead speed of 1.3 mm/min. The dimensions of the samples were 12.7 x 3.2 x 63 mm. Five samples were tested from each case and the average results were recorded. All the tests were done at room temperature.

The notched Izod impact test was carried out according to ASTM D256 using a Ceast impact testing model 6957. The dimensions of the samples were 12.7 x 3.2 x 63 mm. The hammer used to strike the samples was 2.75 J. Five samples were tested for each case. The entire tests were conducted at room temperature.

The hardness was tested by shore durometer in accordance with ASTM D2240 type D. The data obtained represented the average value from 5 test specimens.

The dispersion morphology of waste SiC in polymer composites was observed by scanning electron microscopy (SEM; S-4800/Hitachi). The microscope was operated at 20 kV. Before scanning, a conductive coating layer was spread on the surface of the sample to eliminate the electron charging during examination. The objective is to get some information regarding the filler dispersion and bonding quality between filler and matrix.

## III. RESULTS AND DISCUSSION

### A. Effect of filler content

Figs. 1 (a), (b) display the flexural properties of the composites at different filler content. Due to the presence of waste SiC particles in the polymer, the HDPE/waste SiC composites exhibit markedly higher flexural strength and flexural modulus than the neat HDPE. The flexural modulus could be increased significantly by adding waste SiC particles. The flexural modulus increases in approximately 77%, and the flexural strength increases in approximately 12%, compared to the neat HDPE (with the addition of 20 wt% of the waste SiC particles). These results are attributed to the fact that the polymeric matrix is stiffened by the particulated second phase of SiC, which better stress transfer to the SiC particles in the composites.

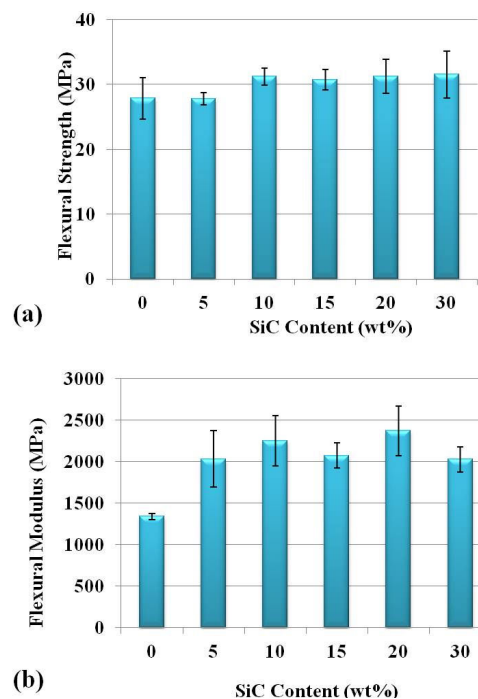


Fig. 1 Flexural strength (a) and flexural modulus (b) of the composites at different filler content

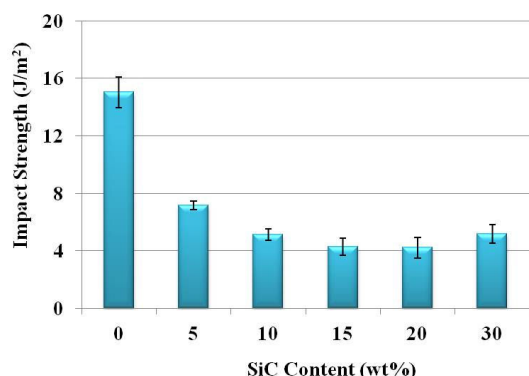


Fig. 2 Impact strength of the composites at different filler content

Fig. 2 displays the impact strength of the composites at different filler content. The impact strength of the composites decreases as the filler content increased. The reduction in toughness was found to be due to the limited plastic flow of the HDPE matrix, because the more ductile matrix was replaced by the more rigid dispersed SiC particles. Poor interfacial bonding induces microspace between the filler and polymer matrix, and these cause numerous micro-cracks when impact occurred, which induce crack propagation easily and decrease the impact strength of the composites. Especially, the impact strength of the neat HDPE showed significantly high toughness, which was drastically decreased at a filler content of 5-30 wt. %.

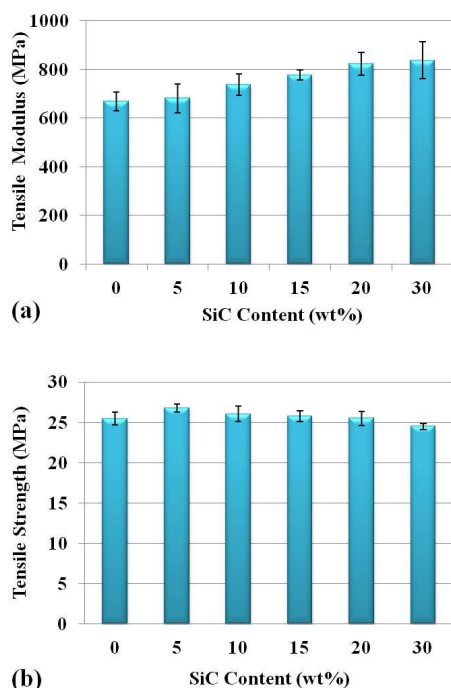


Fig. 3 Tensile strength (a) and tensile modulus (b) of the composites at different filler content

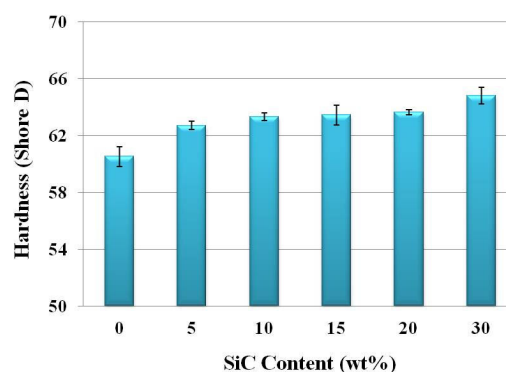


Fig. 4 Hardness of the composites at different filler content

Figs. 3 (a), (b) display the tensile properties of the composites at different filler content. The tensile modulus improved with increasing filler content. For example, the composites containing 30 wt.% of waste SiC, the tensile modulus is by approximately 25 % higher than the neat HDPE. These results are attributed to the fact that the polymeric matrix is stiffened by the particulated second phase of SiC, and the SiC particles restrict the mobility and deformability of the matrix. Tensile strength of the composites slightly decreased with increasing filler content. As the filler content increased, thereby increasing the interfacial area, the worsening interfacial bonding between filler (hydrophilic) and polymer matrix (hydrophobic) decreased the tensile strength. The resistance of composites to plastic deformation can be defined as hardness.

Fig. 4 demonstrates the effect of filler content on the hardness. Due to the presence of waste SiC particles in the HDPE matrix, the HDPE/waste SiC composites exhibit slightly higher hardness than the neat HDPE due to the rigid phase of SiC.

### B. Effect of Surface Modification

TABLE I  
FORMULA AND DESIGNATIONS OF THE SYSTEMS

Sample designation	HDPE/waste SiC	HDPE-g-MA	3-aminopropyltriethoxysilane
C1	90/10	-	-
C2	90/10	1	-
C3	90/10	3	-
C4	90/10	5	-
C5	90/10	7	-
C6	90/10	10	-
C7	90/10	-	0.4
C8	90/10	-	0.6
C9	90/10	-	0.8
C10	90/10	-	1.0
C11	90/10	-	1.2

Agglomerates are formed as the waste SiC content in HDPE composites increases. Therefore, there are different ways of improving the polymer filler interaction and dispersion. Nowadays, many scientist and engineers are engaged in research on among the different ways to improving the polymer-filler interactions, such as the use of the use of compatibilizer [1], [2]-[9] and coupling agent [1], [10]-[17]. In

the present study we used a HDPE-g-MA compatibilization as the compatibilizer and a 3-aminopropyltriethoxysilane as a coupling agent to prepare a HDPE/waste SiC composites. The formulas and designations of the systems of HDPE/waste SiC composites with and without compatibilizer and silane coupling agent are shown in Table I.

Figs. 5 (a), (b) show the effect of surface modification on the flexural strength and flexural modulus of the systems of HDPE/waste SiC composites investigated. It was found that flexural strength was decreased with increasing amount of modification substance (Fig. 5 (a)). Additionally, the use of modifiers (HDPE-g-MA as compatibilizer and 3-aminopropyltriethoxysilane as coupling agent) in the HDPE/waste SiC composites improves the flexural modulus (Fig. 5 (b)). Incorporation of 3 wt.% of HDPE-g-MA, 1.0 wt.% of 3-aminopropyltriethoxysilane in related composites, show a highest flexural modulus. Due to the use of HDPE-g-MA can improve adhesion and thus enhance the properties. When HDPE-g-MA was used, interactions between the anhydride groups of the maleated HDPE and the hydroxyl groups of the SiC can overcome the incompatibility problem. Mechanical interlocking may occur between the SiC particles and compatibilizer (HDPE-g-MA) and between the polymer and compatibilizer. The use of 3-aminopropyltriethoxysilane as a coupling agent in the treatment of the SiC particles surface in HDPE/waste SiC composites improves their flexural modulus because it produces a better dispersion and a decrease in the amount of filler agglomerates. This can be explained by the difference in the morphology of the systems of HDPE/waste SiC composites as we discuss later.

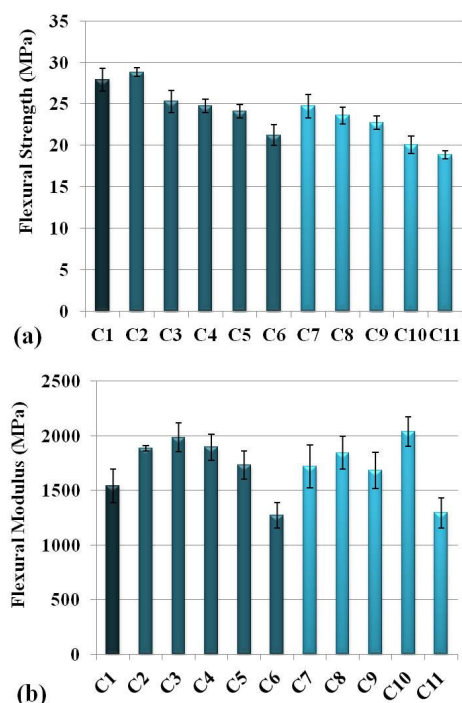


Fig. 5 Flexural strength (a) and flexural modulus (b) of the systems of HDPE/waste SiC composites

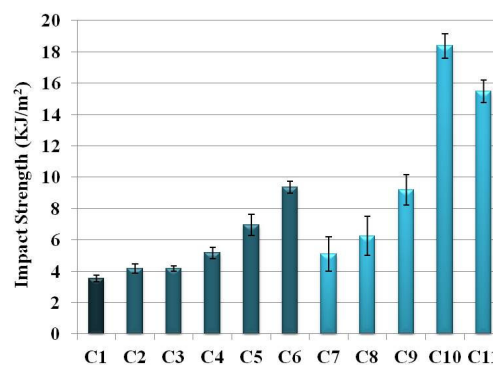


Fig. 6 Impact strength of the systems of HDPE/waste SiC composites

Fig. 6 shows the impact strength of HDPE/waste SiC composites with and without surface modification. It can be seen that both series of composites show an increasing trend as the surface modifier content increases. This indicates that the presence of HDPE-g-MA as compatibilizer and 3-aminopropyltriethoxysilane as coupling agent could improve the wettability between SiC and HDPE and consequently increase the impact strength. A similar finding was also observed by N.Othman et al. [3]. They found that the impact strength being greater for the polypropylene grafted maleic anhydride (PPMAH) treated bentonite filled polypropylene composites than untreated composites which indicate a good interfacial bonding between PPMAH and the filler and the polymer compatibility with the hydrocarbon of PPMAH.

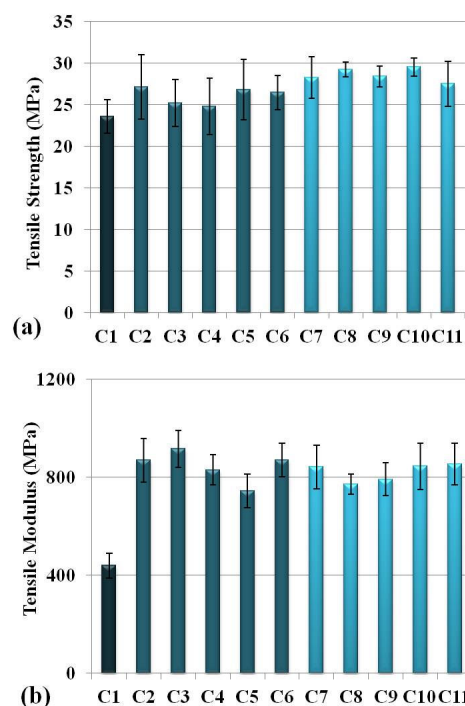


Fig. 7 Tensile strength (a) and tensile modulus (b) of the systems of HDPE/waste SiC composites



Figs. 7 (a), (b) show the tensile properties of the systems of HDPE/waste SiC composites. The effect of adding HDPE-g-MA as compatibilizer and 3-aminopropyltriethoxysilane as coupling agent on the mechanical properties of waste SiC filled HDPE have been studied. The tensile strength and tensile modulus of HDPE/waste SiC composites have been improved with the addition of surface modifier. This indicates that surface modifier improved the interaction between SiC and HDPE causing the stress transferred from HDPE to SiC to become more effective thus increasing the tensile strength and tensile modulus of the composites. Note that, more than 100% improvements of tensile modulus were occurred in all composition with both modified substance.

The effect of addition of the surface modifiers on the morphology of the HDPE/waste SiC composites was studied by SEM image (Figs. 8 (a), (b) and 9 (a), (b)). In case of composites without surface modifier, the particles were not attached to the matrix, as shown in Figs. 8 (a) and 9 (a). Addition of HDPE-g-MA and silane coupling agent improved the adhesion, the waste SiC particles were embedded in HDPE matrix (see in Figs. 8 (b) and 9 (b)). Improvement in both impact strength (Fig. 6) and tensile strength (Fig. 7 (a)) of the composites was obtained.

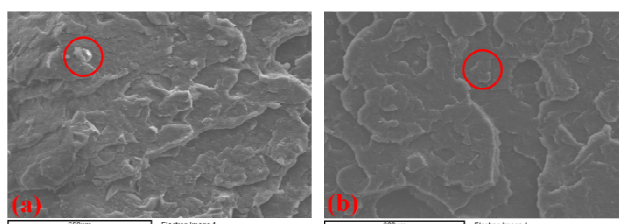


Fig. 8 SEM images of HDPE/waste SiC composites (without (a) and with HDPE-g-MA 5 wt% (b))

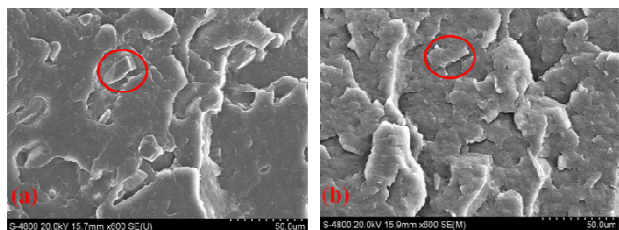


Fig. 9 SEM images of HDPE/waste SiC composites (untreated (a) and 1.0 wt% silane treated (b))

#### IV. CONCLUSION

The incorporation of waste SiC into HDPE matrix increases flexural strength, flexural modulus, stiffness and hardness increased but decreases the impact strength and tensile strength decreased with the increasing in SiC particles content. The addition of surface modifiers improves the interaction between SiC and HDPE to become more effective thus increasing the impact strength and tensile strength of the composites. These results indicate that SiC particles can be interesting filler for HDPE matrix due to the good properties determined for these materials. HDPE-g-MA and 3-

aminopropyltriethoxysilane are an effective surface modifier for HDPE/ waste SiC composites.

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