Mühendislik Bilimleri ve Tasarım Dergisi
9(2), 389 – 395, 2020
e-ISSN: 1308-6693

Journal of Engineering Sciences and Design
DOI: 10.21923/jesd.878299

Araştırma Makalesi
Research Article

SURFACE FREE ENERGY AND FLEXURAL PROPERTIES OF WOLLASTONITE FILLED POLYPROPYLENE COMPOSITES

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Keywords

Polymer Composites,
Surface Free Energy,
Contact Angle,
Wollastonite,
Flexural Properties.

Abstract

Nowadays, appearance is an important factor in vehicles. For this reason, paint application is widely used in plastic parts. Surface energy is a key parameter for the adhesion of paint on the surface of polymers. Among thermoplastic polymers, polycarbonate and acrylonitrile butadiene styrene are commonly used materials due to their higher surface energies. For low surface energy materials, surface energies can be increased using streamer, plasma, corona, mechanical etching and chemical etching methods. The aim of this work is to eliminate the pre-treatments applied to parts produced with polypropylene (PP) polymer using wollastonite ($CaSiO_3$) additive. For this purpose, $CaSiO_3$ filled PP composites containing 40 wt.% wollastonite with different sizes and coatings were produced by extrusion process. Afterwards, plate samples were prepared from granules using a hot pressing device. Flexural properties and surface free energy measurements were performed on these composites produced in plate form. From the results, an increased surface free energy was observed for small particle size and aminosilane coated wollastonite added PP composite with an increase by 23% of the total surface free energy. Flexural strengths were well correlated with the surface free energy results and showed an increase by 1.2% for the same additive.

VOLLASTONİT KATKILI POLİPROPİLEN KOMPOZİTLERİN YÜZEY SERBEST ENERJİSİ VE EĞİLME ÖZELLİKLERİ

Anahtar Kelimeler

Polimer Kompozitler,
Yüzey Serbest Enerjisi,
Temas Açısı,
Vollastonit,
Eğilme Özellikleri.

Öz

Günümüzde, araçlarda görsellik önemli bir faktördür. Bu nedenle, plastik parçalarda boya uygulaması yaygın olarak kullanılmaktadır. Boyanın polimer yüzeyine yapışması için yüzey enerjisi önemli bir etkendir. Termoplastik polimerler arasında, yüzey enerjileri daha yüksek olduğundan dolayı genellikle polikarbonat ve akrilonitril bütadien stiren polimerleri yaygın olarak kullanılmaktadırlar. Düşük yüzey enerjili malzemelerde ise flamaj, plazma, korona, mekanik dağlama ve kimyasal dağlama yöntemleri kullanılarak yüzey enerjileri arttırılmaktadır. Bu çalışmanın amacı vollastonit ($CaSiO_3$) katkı malzemesi kullanılarak, polipropilen (PP) polimeri ile üretilen parçalara uygulanan ön işlemlerin ortadan kaldırılmasını sağlayabilmektir. Bu amaçla, ağırlıkça %40 katkı içeren ve farklı boyut ve kaplamalardan oluşan $CaSiO_3$ katkılı PP kompozitler ekstrüzyon yöntemi ile üretilmiştir. Daha sonra, sıcak presleme cihazı kullanılarak granüllerden plakalar elde edilmiştir. Plaka halinde üretilen kompozitlerin eğilme özellikleri ve yüzey serbest enerjisi ölçümleri yapılmıştır. Sonuçlara bakıldığında, küçük partikül boyutlu ve aminosilan kaplı vollastonit katkılı PP kompozitin toplam yüzey serbest enerjisinin %23 değerinde bir artışıyla, yüzey enerjisini arttırıcı bir özelliği olduğu görülmüştür. Eğilme mukavemetleri yüzey serbest enerjisi sonuçlarıyla örtüşmektedir ve aynı katkı için %1.2 oranında bir artış elde edilmiştir.

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Alıntı / Cite

Soncu, S., Akkoyun, M., (2021). Surface Free Energy and Flexural Properties of Wollastonite Filled Polypropylene Composites, Journal of Engineering Sciences and Design, 9(2), 389-395.

Yazar Kimliği / Author ID (ORCID Number)	**Makale Süreci / Article Process**	
S. Soncu, 0000-0002-7464-3768	**Başvuru Tarihi / Submission Date**	10.02.2021
M. Akkoyun, 0000-0002-8113-5534	**Revizyon Tarihi / Revision Date**	19.04.2021
	Kabul Tarihi / Accepted Date	29.04.2021
	Yayım Tarihi / Published Date	20.06.2021

1. Introduction

Among the various plastic materials classified as thermoplastics, thermosets and elastomers, thermoplastics are widely preferred materials in the industry and have broad usage areas due to their microstructure and particularly to their high formability. For this reason, the adhesion and surface treatment of thermoplastic materials were largely investigated in the literature (Cantero et al., 2003; Lee et al., 2018; Khoshkava and Kamal, 2013; Demjen et al., 1998; Drelich and Miller, 1995; Brandl et al., 2004; Rudawska et al., 2017; Zapata-Massot and Le Bolay, 2007). The process of painting the surface of plastic parts used in the automotive industry is gaining importance day by day. Although plastic surfaces can be produced in the desired color and brightness, they are also required to be painted in many applications for aesthetics or function.

The aim of this work is to prepare and investigate the surface free energy and mechanical properties of highly wollastonite filled PP composites in order to remove the pre-treatment step, which is generally applied to parts produced with polypropylene. For this purpose, $CaSiO_3$ filled PP composites containing 40 wt.% additives with different sizes and coatings were produced by extrusion process. To the best of our knowledge, no studies on the investigation of the surface free energy of $CaSiO_3$ reinforced PP composites exist in the literature. Afterwards, plate samples were prepared from granules using a hot pressing device. Mechanical properties and surface energy measurements were performed on these composites produced in plate form.

The literature survey and materials and methods of this work were given in Section 2 and Section 3, respectively. Section 4 presents the experimental results and discussion concerning the surface free energy and flexural properties of PP/$CaSiO_3$ composites. In the last Section 5, a conclusion of the study can be found.

2. Literature Survey

The improvement of the interactions between filler and polymer materials is most widely realized by non-reactive and reactive surface treatment of the additives (Demjén et al., 1998; Cantero et al., 2003; Drelich and Miller, 1995; Khoshkava and Kamal, 2013; Rudawska et al., 2017). In the case of non-reactive treatment, the interactions between fillers and polymer/filler are affected due to the presence of a substance inducing a drop of the surface free energy of the filler and then enhancing the homogeneity and processing of polymer composites. The other technique called reactive treatment allows the coverage of the filler by an organic substance resulting again in the diminution of the surface free energy of the additives. However, this method is not suitable for apolar polymers without reactive groups in the main chain, due to the weak interaction between the coupling agent and the polymer. The use of silane coupling agents with various reactive groups result in highly improved properties of polymer composites especially in the case of thermoset polymer matrices due to the presence of reactive groups in the polymer chain (Shokoohi et al., 2008; Yang et al., 2013). These coupling agents were also used for low reactive polyolefin materials such as polypropylene in order to improve their final properties (Cantero et al., 2003; Demjén et al., 1998; Khoshkava and Kamal, 2013; Yang et al., 2013).

The properties such as hardness, strength, thermal and electrical conductivity and thermal resistance of polymeric materials can be enhanced using additives of different nature and size (Brandl, Marginean, Chirila, & Warschewski, 2004) (Cantero et al., 2003; Khoshkava and Kamal, 2013; Rothon, 1999). However, for the painting of the surface of plastic parts used in the automotive industry, outside of all these properties, the surface free energy property, which is desired to rise, remains important. In this way, the adhesion strength of the material can be improved. The surface energy of polymeric materials is determined using the intermolecular forces taking into account the various attraction forces occurring between the surface and the adhesive during the bonding of polymeric surfaces (Şekercioğlu and Kaner, 2014; Drelich and Miller, 1995; Shimizu and Demarquette, 2000). In the literature, the surface free energy of polymer composites are widely investigated for fillers of different nature and type such as calcium carbonate, talc and glass fibers (Cantero et al., 2003; Demjen et al., 1998; Drelich and Miller, 1995; Khoshkava and Kamal, 2013; Lee et al., 2018; Rudawska et al., 2017; Shokoohi et al., 2008).

Wollastonite is a calcium silicate used as a functional filler in polymers and due to its specific acicular crystal form allowing high shape ratios, this naturally white mineral is an appropriate additive for polypropylene composites (Rothon, 1999). The structure of $CaSiO_3$ particles, namely their aspect ratios, depends not only on natural conditions but also largely on the preparation and size reduction techniques used. The mechanical properties such as tensile and flexural strengths of $CaSiO_3$ filled polypropylene composites are widely studied and usually an improvement was obtained for untreated and treated $CaSiO_3$ (Chan et al., 2020). In addition, for polypropylene/wollastonite composites the studies were mainly focused on the determination of the adhesion parameters such as the work of adhesion and interfacial free energy of the composites (Svab et al., 2005; Svab et al., 2007; Svab et al., 2009). To the best of our knowledge, no studies on the investigation of the surface free energy of $CaSiO_3$ reinforced PP composites exist in the literature.

3. Materials and Methods

3.1. Materials

Natural and unfilled polypropylene (Moplen HP500N) (density: 0.9 g/cm^3, MFR: 12 g/10min (230°C/2.16 kg)) provided from LyondellBasell was used in this study. Wollastonite powders were provided by Quarzwerke GmbH with the references TREMIN 939 300 AST (T1) (ρ=2.85 g/cm^3 and d (50%) = 30 µm), Tremin 939 300 FST (T2) (ρ=2.85 g/cm^3 and d (50%) = 37 µm), Tremin 283 010 AST (T3) (ρ=2.85 g/cm^3 and d (50%) = 19 µm) and Tremin 283 400 AST (T4) (ρ=2.85 g/cm^3 and d (50%) = 5 µm). All these wollastonite powders were surface treated materials. The applied treatments were aminosilane for T1, T3 and T4 and alkylsilane for T2. All materials were used as received.

3.2. Preparation of Wollastonite Filled Polypropylene Composites

$CaSiO_3$ reinforced PP composites were prepared at a high filler content of 40 wt.% for $CaSiO_3$ powders of different surface treatments and shape ratios (T1, T2, T3 and T4), in order to obtain an improvement in the surface free energy of the composites. Table 1 presents the formulations used for the preparation of these composites. $CaSiO_3$ particles were first pre-mixed with PP. Then, PP/$CaSiO_3$ mixtures were blended with a Polmak Plastik Lab. Extruder/18mm co-rotating twin screw extruder in order to have a homogeneous blend. The mixtures were prepared at a feeding rate of 4 rpm and a rotational speed of 60 rpm. Therefore, the temperatures were set at 90°C, 190°C, 195°C, 200°C, 205°C, 210°C, 210°C, 210°C, 210°C and 200°C, from the feeding zone to the exit die. During the next step, a water bath was used in order to cool the composite immediately after being extruded through the die which is finally granulated and dehydrated. In the last step, a CARVER 12-12 compression molding device was used for the preparation of plate samples (200x200x4 mm). During this process, the compression was carried out at a pressure of 3 tons at 200°C. For mechanical tests, appropriate samples were cut from the plates.

Table 1. Formulations for PP/$CaSiO_3$ composites

Sample	Filler content (wt.%)	PP content (wt.%)
PP/T1 (aminosilane - 30 µm)	40	60
PP/T2 (alkylsilane - 37 µm)	40	60
PP/T3 (aminosilane - 19 µm)	40	60
PP/T4 (aminosilane - 5 µm)	40	60

3.3. Surface Free Energy Measurements

Surface free energies of PP/$CaSiO_3$ composites were obtained from the measurements of contact angles. These measurements were performed on a Biolin Scientific Attension Theta Flex Instrument at room temperature, immediately after the drop deposition from 10 to 30 s. The surface free energy of PP/$CaSiO_3$ composites were obtained using Owens Wendt Rabel and Kaelbel (OWRK) method (Owens and Wendt, 1969). In OWRK method, the surface free energy of the solid is calculated from the use of at least two liquids where the polar and dispersive components are known. The equation proposed in the OWRK model for the calculation of the surface free energy of a solid is given in Equation 1, where γ_L^D and γ_S^D are the dispersive parts and γ_L^P and γ_S^P are the polar parts of liquid and solid surface free energies, respectively whereas θ was the contact angle between the surface of the sample and the standard liquid.

$$\frac{\gamma_L(1 + cos\theta)}{2\sqrt{\gamma_L^D}} = \frac{\sqrt{\gamma_S^P}\sqrt{\gamma_L^P}}{\sqrt{\gamma_L^D}} + \sqrt{\gamma_S^D} \qquad (1)$$

The Equation 1 can be expressed in the linear form as presented in Equation 2. The parameters c, m and x were given as $c = \sqrt{\sigma_S^D}$, $m = \sqrt{\gamma_S^P}$ and $x = \frac{\sqrt{\gamma_L^P}}{\sqrt{\gamma_L^D}}$.

$$y = mx + c \qquad (2)$$

In this work, four different standard liquids (water, ethylene glycol, formamide and diiodomethane) were used and their total, dispersive and polar surface tensions were presented in Table 2. From these values, a determination of the dispersive and polar parts of the surface free energy of the different $CaSiO_3$ filled PP composites can be realized. From the slope of the OWRK graph plotted using the Equation 1, the polar part of the composites surface free energy is obtained whereas the dispersive part is determined with the vertical intercept.

Table 2. Surface tensions (SFT) of the standard liquids

Solvent	SFT - Total (mN/m)	SFT - Dispersive (mN/m)	SFT - Polar (mN/m)
Water	72.3	18.7	53.6
Formamide	59.4	39.4	20
Ethylene Glycol	47.5	29.3	18.2
Diiodomethane	50.8	50.8	0

3.4. Mechanical Properties Measurement

A three-point flexural test was performed according to ISO 178 using a Devotrans DVT GP D S60 N testing device at a cross-head speed of 10 mm/min. The specimens were prepared using the plate samples of 4 mm thickness obtained by compression molding.

4. Experimental Results and Discussion

4.1. Surface Free Energy Results

The images obtained during the contact angle measurements for the different PP/$CaSiO_3$ composites with water, ethylene glycol, diiodomethane and formamide were given in Figure 1. Table 3 summarizes the measured contact angles using these four standard liquids. In this work, contact angle measurements were performed for the surface characterization of PP/$CaSiO_3$ composites. Table 4 presents the total, dispersive and polar surface free energies determined using OWRK model. From Table 4, as expected the polar part value of the total surface free energy for pure PP sample is low compared to the dispersive one, because of highly apolar nature of the PP surface.

The comparison of all PP/$CaSiO_3$ composites revealed an improvement of the polar component value, particularly in the case of T3 and T4 filled PP composites. This behavior can be explained by the presence of H_2N- functional group in aminosilane coupling agent at the additive surface (Svab et al., 2005). However, the unexpected lower polar component of the surface free energy value obtained in the case of T1 and T2 filled PP composites is probably due to the higher particle size (30 μm) of fillers inducing a decrease of the surface free energy of the additives. As reported in the literature, the use of low sized particles leads commonly an improved surface free energy of the particles engendering enhanced adhesive properties (Holec et al., 2020; Rudawska et al., 2017).

Another explanation for the higher total surface free energy value determined in the case of T3 reinforced PP composites, with a rise by 23% may be a more homogeneous dispersion of these particles (Rudawska et al., 2017). In this case, a more detailed investigation of the microstructure can be valorized in a future paper. Furthermore, it can be stated that the particle shape can also have an effect on these results. While a higher polar component value of the surface free energy determined for anisotropic particle shaped T3 and T4 filled PP composites, needle-structured T1 and T2 filled PP composites exhibited lower values.

Table 3. Contact angles of PP/CaSiO3 composites obtained with four different standard liquids

Sample	Water Contact angle (°)	Formamide Contact angle (°)	Ethylene Glycol Contact angle (°)	Diiodomethane Contact angle (°)
PP	78.5	63.6	69.8	55.1
PP/T1	80.4	87.2	75.7	56.3
PP/T2	84.5	68.8	57.4	59.8
PP/T3	101.1	80.1	76.8	54.4
PP/T4	95.1	81.7	63.7	52.0

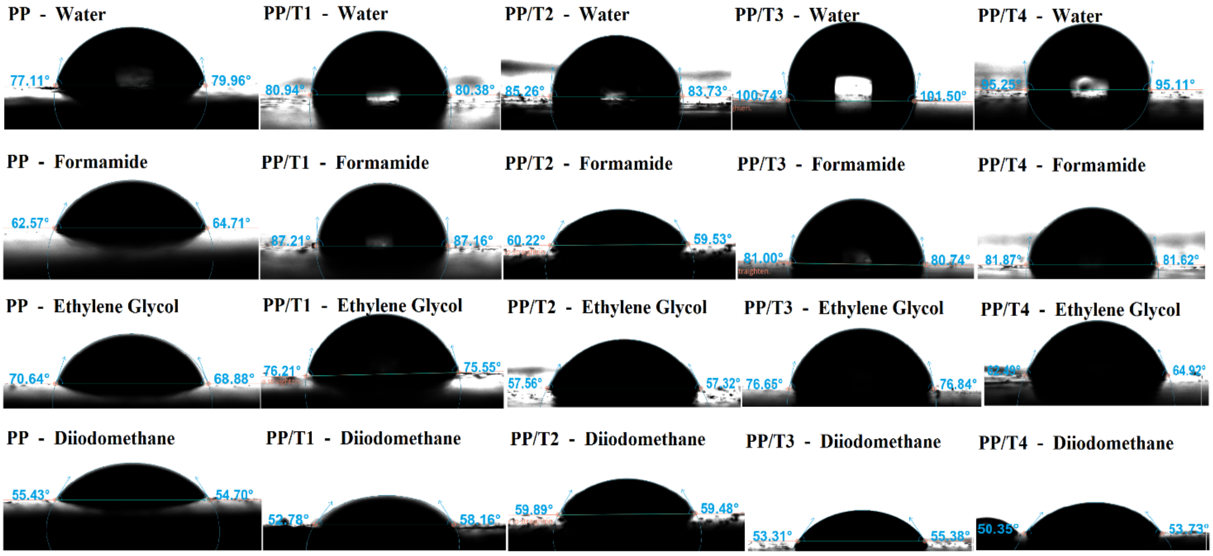

Figure 1. Pictures of contact angles obtained for the different PP/CaSiO3 composites with water, ethylene glycol, diiodomethane and formamide

Table 4. Total surface free energies (SFE) of PP/CaSiO3 composites with their polar and dispersive components

Solvent	γ_S^P (mN/m)	γ_S^D (mN/m)	γ_S^T (mN/m)
PP	7.420	50.79	58.21
PP/T1	5.240	46.44	51.68
PP/T2	0.120	20.02	20.14
PP/T3	71.55	0.001	71.55
PP/T4	39.90	7.310	47.21

4.2. Mechanical Properties Results

The flexural strength values obtained for pure PP and PP/CaSiO3 composites prepared with fillers of different size and surface treatment were gathered in Table 5 and presented in Figure 2. The results showed a slight increase by 1.2% of the flexural strength only in the case of T3 reinforced PP composite compared to the unfilled PP sample. This behavior is in correlation with the surface free energy results where an increase was obtained only in the case of PP/T3 composite. The slight improvement of flexural strength in the case of PP/T3 composites is mainly due to the surface treatment of the particles with aminosilane. As largely reported in the literature, the surface treatment of wollastonite allows the improvement of mechanical properties due to the stronger interfacial adhesion between the filler and the polymer matrix and then the facilitated load transfer from the polymer matrix to the filler during the mechanical test (Meng and Dou, 2008; Chen et al., 2008; Qu et al., 2009).

On the other hand, T1, T2 and T4 filled PP composites revealed a drop by 24%, 23% and 14% compared to neat PP, respectively. The decrease of flexural strength observed for T1, T2 and T4 added PP composites is a result of the weak interactions between the fillers and the polymer matrix, where the load transfer at the interface is hindered. Similar results were obtained in the literature (Luyt et al., 2009; Svab et al., 2007). Moreover, this behavior can also be explained by the reduced motion of polymer chains inducing a decrease of the crystallinity of the polymer due to the presence of $CaSiO_3$ particles (Sohail et al., 2017; Sohail et al., 2012; Yuan et al., 2017).

Table 5. Flexural strength values of pure PP and PP/$CaSiO_3$ composites

Sample	PP	PP/T1	PP/T2	PP/T3	PP/T4
Flexural strength (MPa)	40.8 ± 0.50	31.10 ± 3.10	31.50 ± 2.60	41.30 ± 0.53	35.10 ± 2.40

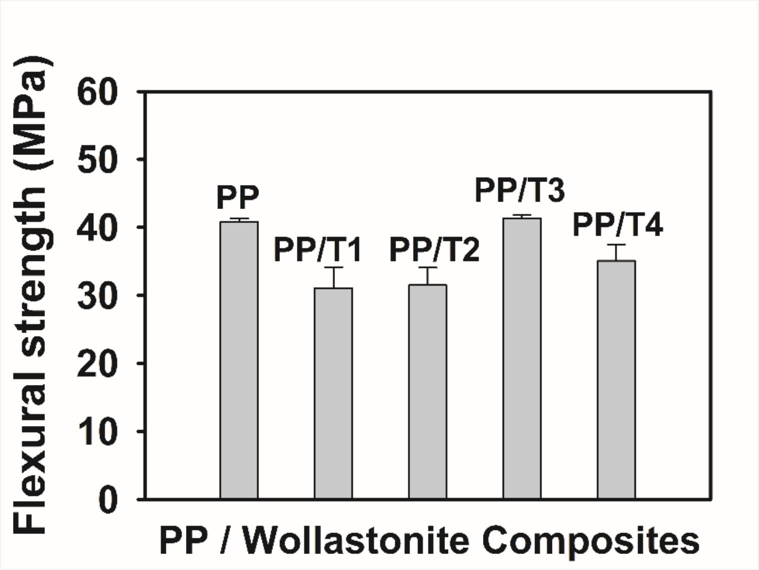

Figure 2. Flexural strength values of pure PP and PP/$CaSiO_3$ composites

5. Conclusion

PP/$CaSiO_3$ composites were produced using a twin-screw extruder and the surface free energy with the flexural properties were investigated. The results indicated an increase by 23% of the total surface free energy only in the case of PP/T3 composite probably due to the presence of H_2N- functional group in aminosilane coupling agent at the additive surface and a better filler-matrix interaction compared to the other composites. The flexural strength results of PP/$CaSiO_3$ composites also revealed a slight enhancement of the flexural properties only for PP/T3 composite with an augmentation by 1.2%. The other composites showed a maximum diminution by 24% for PP/T1 composite mainly due to the poor load transfer between polypropylene and wollastonite as a result of poor interactions between these two phases.

Acknowledgement

The authors gratefully acknowledge the financial support of Demo Plastik Ürünleri San. ve Tic. A.Ş. (Turkey).

Conflict of Interest

No conflict of interest was declared by the authors.

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