

EFFECTS OF SODIUM BENTONITE ON THE CHARACTERISTICS OF RECYCLED PLASTIC COMPOSITES AS POTENTIAL GEOMEMBRANE

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Abstract

This paper investigates the utilization of post-consumer waste plastic sachets as potential geomembrane in landfills. This study promotes recycling of plastic wastes in Adamson University premises and supports the government advocacy in Ecological Solid Waste Management Act. To confirm the plastic compositions, the plastic waste sachets were subjected to Fourier Transformed Infrared (FTIR) Spectroscopy analysis. FTIR result shows that plastics were made of polypropylene (PP), polyethylene (PE), and polyester (PET). Subsequently, sodium bentonite (SB) was added to shredded waste plastics at varying percentages of 0, 5, 10 and 15 percent by weight. Then, it was subjected to melt intercalation and compression molding process to produce recycled plastic composites (RPC). Differential scanning calorimetry (DSC) was performed to determine the melt-intercalation and compression molding temperatures. Consequently, RPC was subjected to different physico-mechanical tests such as density, water absorption, flexural, tensile, izod impact and abrasion tests. Maximum water absorption capacity and tensile strength were observed in RPC with 15% SB. The highest density and flexural strength were observed in RPC with 10% SB content. Moreover, RPC with 5% SB exhibited the highest impact strength and abrasive resistance. Scanning Electron Microscopy (SEM) divulged the poor interfacial adhesion between the polymer blends and unequal dispersion of SB with recycled plastics. Overall, the surface response optimization corroborates that the ultimate percentage of SB was achieved at 10.4% content which has a great potential as geomembrane.

Keywords: Physico-mechanical Properties, Recycled Plastic Wastes, Sachets, Sodium Bentonite

1.0 INTRODUCTION

Plastics have wide applications such as bags, disposable drink bottles, plastic plates, cups and cup lids, plastic straws and many others. In Philippines, sachets are used to retail merchandises to provide affordable price to consumers. Due to its massive use and low cost, plastic voluminous wastes became a rising problem. Plastics are threat in the environment for it takes time to biodegrade (Tokiwa et al., 2009, pp. 3722--3723). In Metro Manila alone, where the estimated rate of daily solid waste generation is pegged at 0.7 kg per capita, this metropolis generates an estimated amount of 8,400 to 8600 tons daily (Alave, 2011, p. 1). Currently, Ecological Solid Waste Management Act of the Philippines (Republic Act 9003), imposes stricter requirements in municipal solid waste management. This mandates local government units to avoid waste dumping and use properly designed sanitary landfills instead. In addition, the legislation puts emphasis on the 3R policy reduction, reuse and recycling (Paul et al., 2012, p. 2019). Utilization of waste plastic sachets may contribute to volume reduction of post--consumer plastic wastes.

Many studies showed that recycling of plastic waste can turn into various kinds of cost effective material that is useful in controlling the continuous growth of solid wastes in the country without sacrificing the quality of the product. Senoro et al. (2012, p. 389) and Anselmo et al. (2016, p. 4) studied recycled plastics were utilized to enhance its properties that can be used in many ways such as in geotechnical engineering applications such as geomembrane.

Geomembrane, a synthetic membrane liner made from relatively thin continuous polymeric sheets with a very low permeability, is used in various application in geotechnical engineering, such as landfills, reservoir, dams, canals, or tunnels, to reduce or prevent fluids (liquid or gas) to pass through the human made project, structures or system (Touze--Foltz and Farcas, 2017, p. 1). The commonly used geomembranes are usually made up of either High--Density Polyethylene (HDPE) or Flexible Polypropylene (fPP). However, it is quite expensive especially for large scale area installation.

Plastics like HDPE geomembrane are low permeable material. Combining with right additives is an effective way to strengthen its plastics physico--mechanical properties. Conversely, nanotechnology has become a research priority in the Philippines because of its expanding potential solution to country's need of sustainable products and increasing demand for technologies to improve the plastic physico--mechanical properties using nanomaterials like SB. SB is known as "the clay of 1000 uses" is commercially used in cosmetics, pharmaceuticals and the treatment of hazardous wastes. It is also used in construction in drilling operations and many environmental retention applications (Schenning, 2004, p. 4). SB clay has low hydraulic conductivity, high swelling potential upon wetting and high capacity of cation exchange. In the Philippines, geological deposits of SB can be found in Mangatarem and Bigbiga, Pangasinan and Poon Bato, Zambales (Alexander et al., 2008, p. 4). Relative to the above--mentioned, this study aims to investigate the effect of SB in the recycled plastic waste as potential geomembrane.

2.0 METHODOLOGY

Gathering of materials, preliminary tests and fabrication of RPC

Post-consumer plastic wastes such as sachets were collected from Adamson University. Plastic wastes were cleaned, cut, shredded and sieved to obtain 1mm diameter particle size. The shredding and sieving of plastic wastes was performed in the Chemical Process Industry Laboratory and Civil Engineering Laboratory at Adamson University. On the other hand, SB was provided by Saile Industries. The batch formulations of the shredded plastic wastes with varying percentages by weight of SB is presented on Table 1.

Table 1: Batch Formulation

Sample Number	Shredded plastic wastes (%)	SB (%)
1	100	0
2	95	5
3	90	10
4	85	15

Fabrication of RPC was adopted from the study of Senoro (2006, p. 3). The shredded plastics underwent melt-intercalation process using two-roll mill. Then, it was molded into laminates using Shinto Compression Molding Machine at the Department of Science and Technology (DOST).

a. FTIR Analysis

The plastic strips were subjected to FTIR analysis at the Chemical Laboratory of Adamson University using PerkinElmer Spectrum 2 following ASTM E168. Its wavenumbers ranged from 550 cm^{-1} to 4000 cm^{-1} using 20 scans per sample with a resolution of 4 cm^{-1} . Plastic strips were examined back to back to evaluate and identify its composition. The unknown spectra scans were compared to the known spectra scan stored in a computer-based library to determine its base materials.

b. DSC Analysis

Four (4) samples of RPC were subjected to DSC test to determine the sachets melting temperature following ASTM E1356. This test was conducted using PerkinElmer DSC 4000 at the Advanced Device and Materials Testing Laboratory of DOST.

SEM Analysis

SEM was used to determine the composition and topography of the RPC using HITACHI Tabletop Microscope 3000, a focused electron beam, which creates an image of the sample in four (4) different magnifications (x250, x300, x400 and x500). Four (4) different batches of RPC samples with SB were analyzed at the Analytical Services Laboratory of University of Santo Tomas.

Physico-mechanical properties of RPC

In each batch, three (3) specimens were subjected to density and water absorption tests at the Standard and Testing Division of DOST following ASTM D792 and ASTM D570, respectively. In density test, method B was used wherein instead of water, RPC

samples were submerged to ethanol whose density is 0.7877 g/cm³. To determine its density at a standard temperature of 23°C which can be calculated using Eq. 1. The instrument used was Alfamirage Electronic Densimeter SD200L. For water absorption test, Mettler Toledo Balance and Mitutoyo Digital Caliper were used for the determination of water absorption of plastics when immersed in water at controlled temperature of 23±1°C (73.4±1.8°F) using Eq. 2.

$$\rho = \frac{W_{\text{§}}}{\text{§\%}} (\rho_{\text{ethanol}}) \quad \text{Eq. 1}$$

where:

ρ = Density of RPC, g/cm³
 ρ_{ethanol} = Density of ethanol, g/cm³
 W_A = Weight in air, g
 W_L = Weight in liquid, g

$$\frac{W}{W_*} \% = \frac{W_C}{W_*} (100\%) \quad \text{Eq. 2}$$

where:

W% = Water absorption percentage
 W_w = Wet weight, g
 W_c = Conditioned weight, g

Laboratory tests for Flexural, Tensile, Izod Impact and Abrasion were simultaneously performed to obtain the mechanical properties of RPC. For flexural test, five (5) specimen per batch of 127mm x 15mm x 4mm were subjected to a three--point loading to determine the force that can withstand by the RPC samples before breaking by using Eq. 3. In tensile test, five (5) specimen per batch of 140mm x 15mm x 4mm were subjected to a tensile tester that continuously pulls the bar constant until the sample breaks into two at a constant speed of 5mm/min. Hence, the tensile strength is measured by dividing the maximum load by the original cross--sectional area of the sample can be calculated using Eq. 4. Both tests were performed at the Geotechnical Engineering Laboratory of Adamson University using a 5kN capacity Universal Testing Machine by Zwick Roell following ASTM D790 and ASTM D638, respectively.

$$\sigma_f = \frac{3PL^2}{2bd^2} \quad \text{Eq. 3}$$

where:

σ = stress in the outer fibers at midpoint, MPa
 P = load at a given point on the load--deflection curve, N
 L = support span, mm
 b = width of beam tested, mm
 d = depth of beam tested, mm

$$\mu = - \frac{(1s_3)}{(1_4)} \quad \text{Eq. 4}$$

where:

- μ = Poisson's ratio
- dε₇ = change in transverse strain
- dε₈ = change in axial strain

Five (5) samples per batch were subjected to izod impact test to analyze the impact strength of RPC using a 1 Joule capacity of Zwick/Roell 5.5P Izod Pendulum Tester. Moreover, three (3) samples were subjected to abrasion test to determine the abrasive resistance of RPC using Teledyne Taber 5130 Abrader with Calibrase CS--10 wheel wherein it was set up to 1000 cycles which can be determine. The wear resistance of RPC was calculated using Eq. 5. ASTM D256 and ASTM D4060 were followed during the tests. Both tests were performed at STD in DOST with 23±2°C temperature.

$$\text{Weight Index} = \frac{(9\%)}{;} (1000) \quad \text{Eq.5}$$

where:

- A = weight of specimen before, g
- B = weight of specimen after, g
- C = number of cycles of abrasion recorded

Statistical Analysis

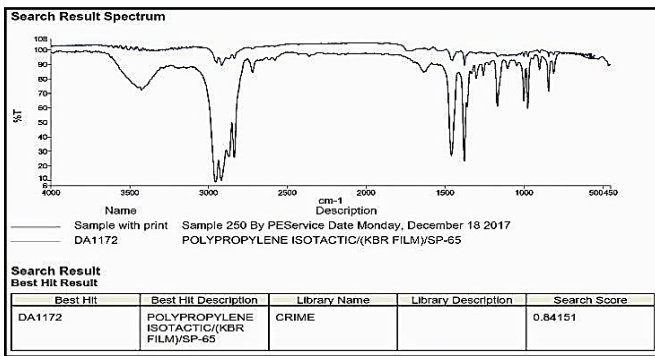
The statistical software XLSTAT 18 (Addinsort, NY, USA) was used for analyzing the data using the Pearson's correlation between the physico--mechanical properties of RPC. Meanwhile, Design Expert 11 software was used to determine the optimal percentage of SB using the Response Surface Methodology (RSM). The physico--mechanical properties functioned as the responses. Using the software, several solutions of optimum responses were found. The highest desirability was used to analyze the physico--mechanical test results.

3.0 RESULTS AND DISCUSSION

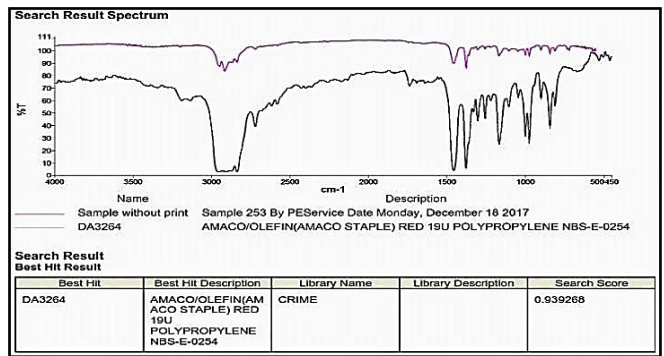
FTIR Analysis

FTIR is a qualitative analysis to classify the type of chemical constituents for identification of polymer and quality control. The peaks between 3000 cm⁻¹ to 2800 cm⁻¹ indicates that the material present in the sample was either PE or PP. However, the distinct characteristics for PE were observed in wavenumbers 2845 cm⁻¹ and 717 cm⁻¹ while for PP were in wavenumbers 2950 cm⁻¹ and 1455 cm⁻¹.

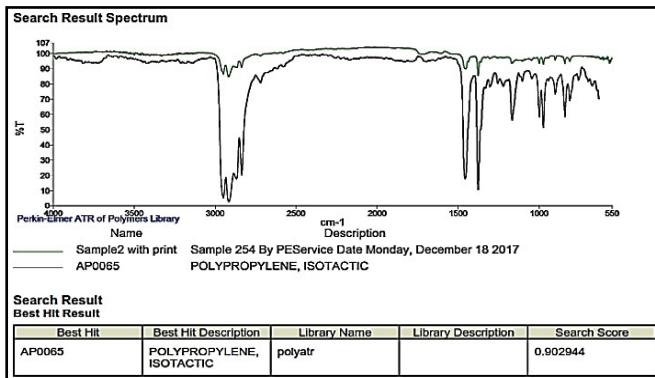
The FTIR results in Figure 1a, 1b, 1c, and 1d showed the presence of peaks in wavenumbers which ranges from 1376 - 2950 cm⁻¹. These wavenumbers suggested that the samples tested were identified as PP material. In Figure 1e, the profound peaks ranges from 723 - 2970 cm⁻¹ which indicate that the sample was a PET material. PET is a kind of polyester that is widely used in many applications (Bach, Dauchy and Etienne, 2009, p. 2). In Figure 1f, the presence of wavenumbers from 718 - 2916 cm⁻¹ suggest that the sample tested was a PE material.



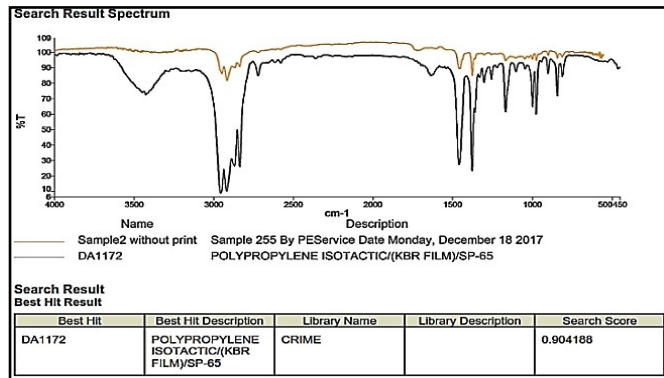
(1a)



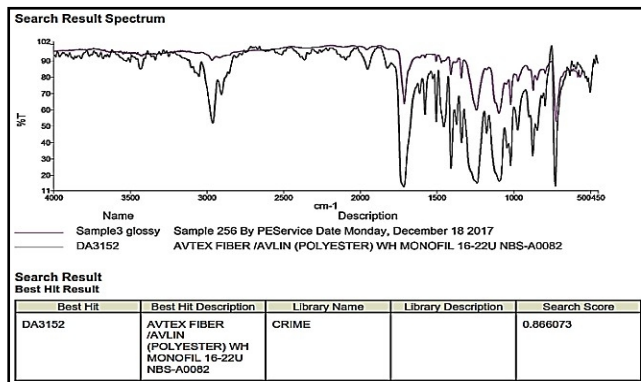
(1b)



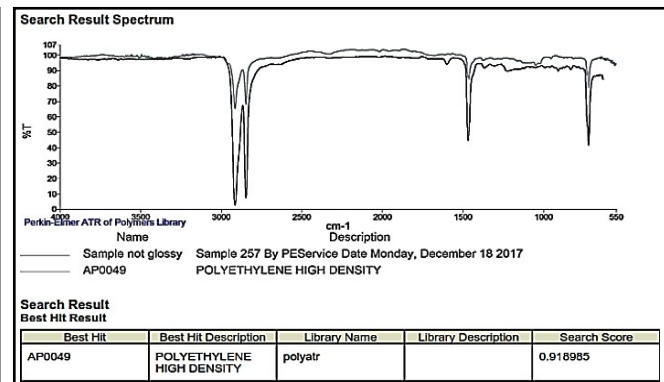
(1c)



(1d)



(1e)



(1f)

Figure 1: (a) Sample 1 print side of plastic film, (b) Sample 1 without print side of plastic film, (c) Sample 2 print side of plastic film, (d) Sample 2 without print side of plastic film, (e) Sample 3 glossy side of plastic film, and (f) Sample 3 not glossy side of plastic film.

Morphology of RPC

Micrographs of the surface of the RPC were shown on Figure 2. Figure 2a shows the SB powder with small particles of the clay; while on Figure 2b shows the intercalated plastic composites. Whereas, Figure 2c, 2d, and 2e show the RPC samples with varying percentage (5%, 10% and 15%) of SB. Holes, pores, and hollow spaces were observed in the intercalated plastics due to poor interfacial adhesion between polymer blends. On the other hand, it was observed that there is an unequal dispersion of SB to the intercalated plastics that resulted to an agglomeration of the clay. Based on the

study of Ramos (2005, p. 388), the presence of large amount of bentonite resulted to an agglomeration that causes poor adhesion and the uneven dispersion of the clay to the sample.

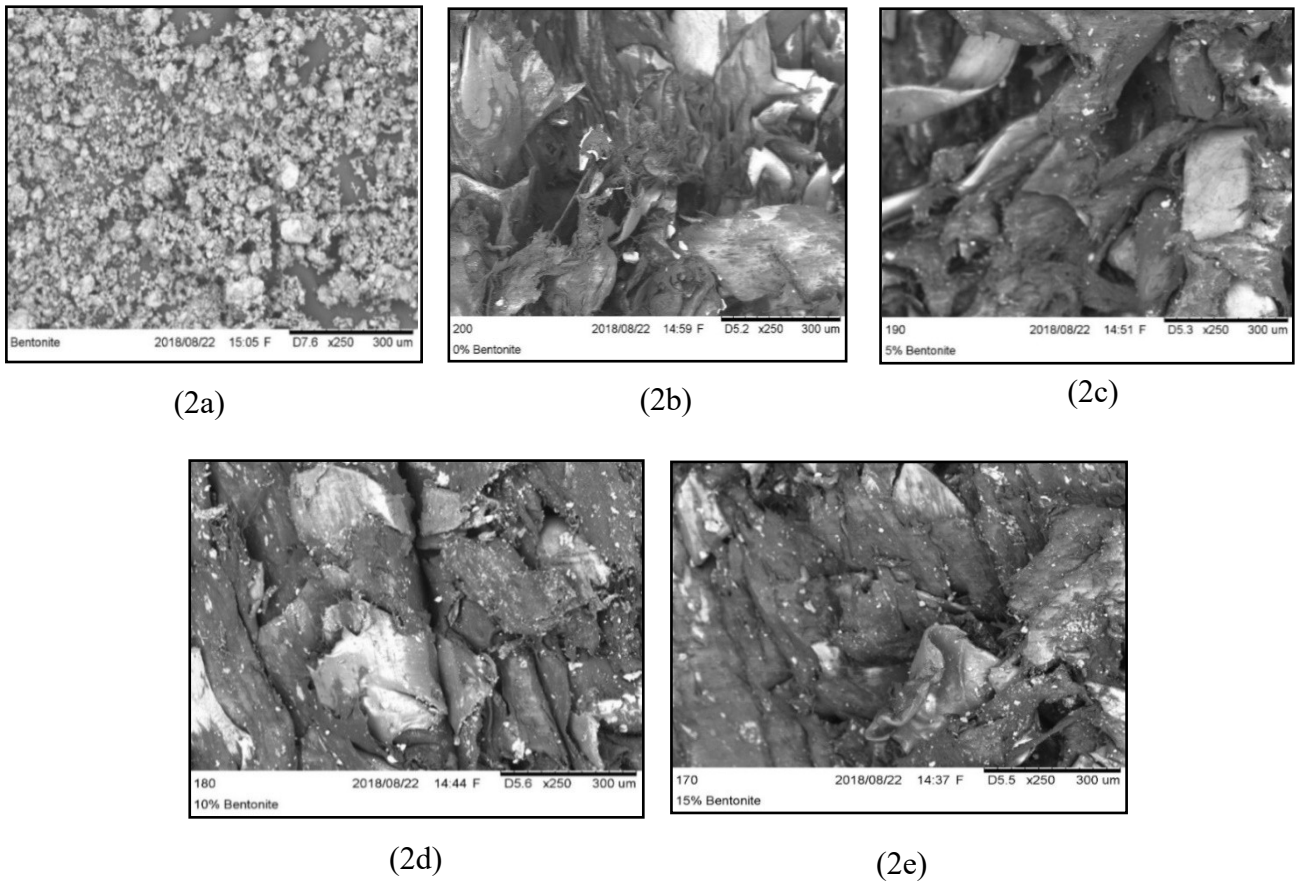


Figure 2: (a) SB, (b) RPC w/ 0% SB, (c) RPC w/ 5% SB, (d) RPC w/ 10% SB and (e) RPC w/ 15% SB

Melting temperature of identified plastics

Degradation of plastics usually occur at high temperature (Lee, 2007, p. 362). Relative to this, the sachets melting temperatures (T_m) were also identified. DSC test showed the melting temperature of the RPC samples on Table 3. The melting temperature of HDPE and PP were 122.23°C and 159.78°C, respectively. DSC results were relative to the study of Café (2007, p. 5) and Forrest (2002, p. 9). PL and low density polyethylene (LDPE) gave the highest and the lowest melting temperature with 251.63°C and 105.42°C, respectively.

Table 2: Differential Scanning Calorimetry Test Results

Plastic	Melting Temperature (T_m , °C)		
	This study	Based on Café (2007)	Based on Forrest (2002)
LDPE	105.42	107 – 124	105
HDPE	122.23	122 – 137	135
PP	159.78	158 – 168	160
PL	251.63	220 – 268	256

Based on the study of Yoshida and Rowe (2003, p. 8), the common sanitary landfill liner temperature as often observed is at 35 to 45°C and may still rise quickly depending on the different rates of landfilling, types of wastes, generation of heat due to decomposition of organic waste, effects of leachates and the different rate of chemical reactions. Also, the landfill temperature can increase as the wastes degrade (Yoshida and Rowe, 2003, p. 1). The estimated service life of HDPE geomembrane based on the 50% reduction of tensile strength ranges from 130--190 service life at 35 °C (Rowe, 2005, p. 663). The DSC results showed that the temperature of plastics has the capacity to withstand the temperature of landfills.

Physico--mechanical properties of RPC

a. RPC Density results

The RPC density results shown in Table 3. The obtained SB density was 1.78 g/cm³ which falls for the range of density of SB from 1.6 to 1.8g/cm³ (Liu and Neretnieks, 2006, p. 11). The RPC densities fall under the general limits for the density of geomembrane polymers which are 0.85 to 1.5 g/ cm³ (Bag et al, 2003, p. 562). RPC without SB attained the lowest density among the batches with 0.998 g/cm³. The density of RPC increased with the addition of SB. However, the highest density of 1.041 g/cm³ was observed in RPC with 10% SB. This can be attributed to the uneven dispersion of SB in the shredded waste plastics during the manual mixing. Moreover, plastic densities may vary in different portions because of difference in crystallinity, porosity and compositions (i.e. types of resins, fillers, etc.) Furthermore, thermoplastics may have considerable deviations by adding fillers, pigments, and reinforcing agents in classifying elasticity, behavior on heating, density, and solubility (Bag et al, 2003, p. 561).

Table 3: Density of RPC with different percentages of SB

Batch No.	Weight in Air (g)	Weight in liquid (g)	Density (g/cm ³)
1	3.138	0.660	0.998
2	3.197	0.726	1.017
3	3.418	0.830	1.041
4	3.324	0.777	1.027

b. RPC Water Absorption Results

The average water absorption of RPC was shown in Table 4. It showed that with an increasing value of SB clay ratio, the water absorption value also increases. The results are consistently increasing because of SBs capability to absorb water on the huge specific surface of the particle and to allow and hold water into the interlayer (Schenning, 2004, p. 7).

Table 4: Water Absorption of RPC with different percentages of SB

Batch No.	Weight before (g)	Weight after (g)	Difference in Weight (g)	% Absorption
1	3.69	3.70	0.003	0.085
2	3.78	3.79	0.004	0.099
3	3.64	3.64	0.005	0.133
4	3.60	3.60	0.007	0.186

The percent water absorption of RPC samples is all less than 1% which corresponds to the allowable percentage of a commercially available geomembrane. Based on water absorption results, batch 4 shows the best result. With its high absorption and swelling capacity, SB helps the polymers to resist fractures and cracks especially during installation and desiccation (Allen, 2005, p. 10).

c. RPC Flexural, Tensile and Izod Impact Test Results

Figure 3 shows that 10% SB addition provides the highest flexural strength. This effect may be due to the polarity between clay and polymer matrix (Fermino et al., 2016, p. 662). However, RPC with 10% SB and 90% post-consumer wastes reached the maximum flexural strength of the overall RPC samples. The lowest flexural strength was recorded in the RPC samples with 15% SB.

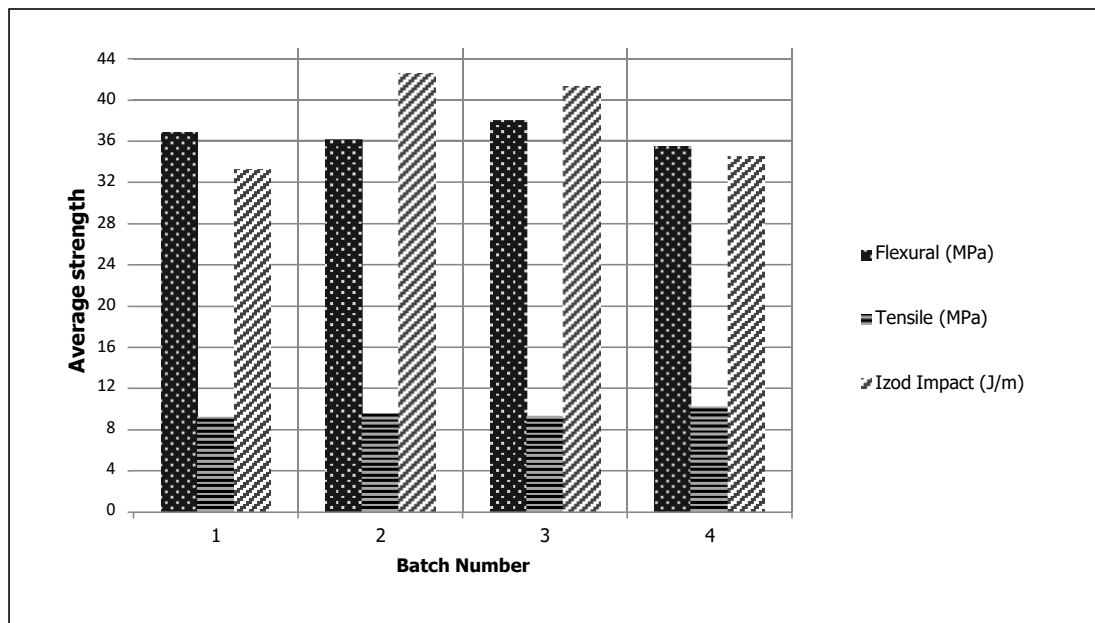


Figure 3: Mechanical Property Test Results

Conversely, tensile strength test (Figure 3) divulged that SB addition resulted to an increase of tensile strength. Anisotropy is one of the factors that influence the tensile properties of the polymer composites (Pukanszky, 1990, p. 260). The changes in the composite properties may be caused by the different adhesion of component, separation of layers, fibre breakdown and the orientation of the particles. Pukanszky elaborates that these reasons explain the differences observed in the reinforcing effect of the fillers to the polymer matrix. Moreover, the rupture of the composite originates if there is a high difference in chemical nature and physical morphology between matrix material and the fillers (Tong et al., 2003, p. 739). Unfortunately, the tensile strength of RPC samples did not meet the 15 MPa tensile strength of a commercially available geomembrane.

The average impact strength of the RPC (Figure 3) shows that the highest impact strength in all the samples was observed on the RPC with 5% SB and 95% post-consumer wastes. However, upon SB addition, the impact strength decreases. This result can be attributed to the increasing the percentage of clay on the sample that leads to large agglomeration resulting to higher possibility of debonding due to poor interfacial adhesion (Sarkar et al., 2008, p. 28).

d. RPC Abrasive Test Results

Table 5 presents the weight losses of RPC samples which denotes the abrasive resistance of the samples. The sample that has the highest weight loss was observed in RPC with 15% SB. Based from the results shown, RPC with 5% SB obtained the highest abrasive wear resistance since it has obtained the lowest weight loss after 1000 cycles. According to Tong et al. (2003, p. 734), the bond strength at the filler interface affects the free abrasive wear of a filler-reinforced composite. In this interface, the rupture of the composite easily originates particularly if there is a high difference in chemical nature and physical morphology between the matrix material and the fillers. In addition, the weight loss of RPC samples is less than 0.05g which corresponds to the allowable weight loss of a commercially available geomembrane.

Table 5: RPC Abrasion Test Results

Batch No.	Initial Weight (g)	Weight after (g)	Weight Loss after 1000 cycles (g)
1	47.876	47.860	0.017
2	46.403	46.392	0.011
3	46.262	46.249	0.013
4	47.691	47.670	0.021

Statistical Analyses Results

Table 6 shows the Pearson correlation matrix Pearson among the physico-mechanical properties considered. The density shows a moderate correlation with water absorption and izod impact strength similar to the study of Asuero et al. (2006, p. 56). Correspondingly, the density of RPC shows a moderate positive correlation with its water absorption. Meanwhile, water absorption shows a high correlation with tensile

strength but moderately correlated with abrasion. Small amounts of SB as a reinforcement improves the tensile strength and water absorption which is relative to the study of Najafabadi, Khorasani and Esfahani (2014, p. 412).

Meanwhile, the relationship within mechanical properties shows that the flexural strength portrays a negative high correlation with tensile strength which supports the study of Othman et al. (2004, p. 719), the tensile strength of PP composites with bentonite as a filler was decreased as the filler loading increases. Meanwhile, the flexural strength showed an inverse result. As for the tensile strength, it shows a moderate positive correlation with abrasion. Abrasion resistance was improved with the of the organoclays addition such as SB (Kim et al., 2006, p. 2065). On the other hand, izod impact shows a negative high correlation with abrasion. The relationship between impact strength and abrasion is inversely proportional to each other. This attributes to the study of Wetzel et al. (2002, p. 1922) that the addition of minerals such as calcium silicate in a polymer composite, improved the wear resistance while the impact strength was reduced.

Table 6: Pearson’s correlation of physico--mechanical properties

Variables	Density	Water Absorption	Flexural Strength	Tensile Strength	Izod Impact Strength	Abrasion
Density	1					
Water Absorption	0.636	1				
Flexural Strength	0.265	--0.392	1			
Tensile Strength	0.346	0.861	--0.789	1		
Izod Impact Strength	0.529	--0.186	0.309	--0.157	1	
Abrasion	0.139	0.623	--0.478	0.560	--0.883	1

The mechanical property test results support the study of Abd El--Fattah and Abd ElKader (2018, p. 1218) that reinforcing the sample with clay comes with two phenomena: (1) the distribution of clay at low contents results to a positive effect that can boost the mechanical properties and (2) a negative effect that caused by agglomeration of clays at high content generates a decrease in performance.

The study of Dong and Bhattacharyya (2008, p. 1191) explained that the mechanical properties were affected by the interfacial bonding between the clay and polymer. As seen in the SEM micrographs, poor interfacial bonding effect that may further clarify the less enhancement in their tensile/flexural properties. Moreover, the excessive amount of clay decreases the impact strength because of agglomeration that can cause cracks easily. Thus, enhanced mechanical properties can achieve by pairing good clay dispersion and a strong filler/matrix with good interfacial bonding.

Response Surface Optimization

A statistical technique such as RSM, which is known, was used for developing, improving and optimizing methods (Ba and Boyaci, 2007, p. 836). Design Expert 11 was utilized to investigate the experimental data results.

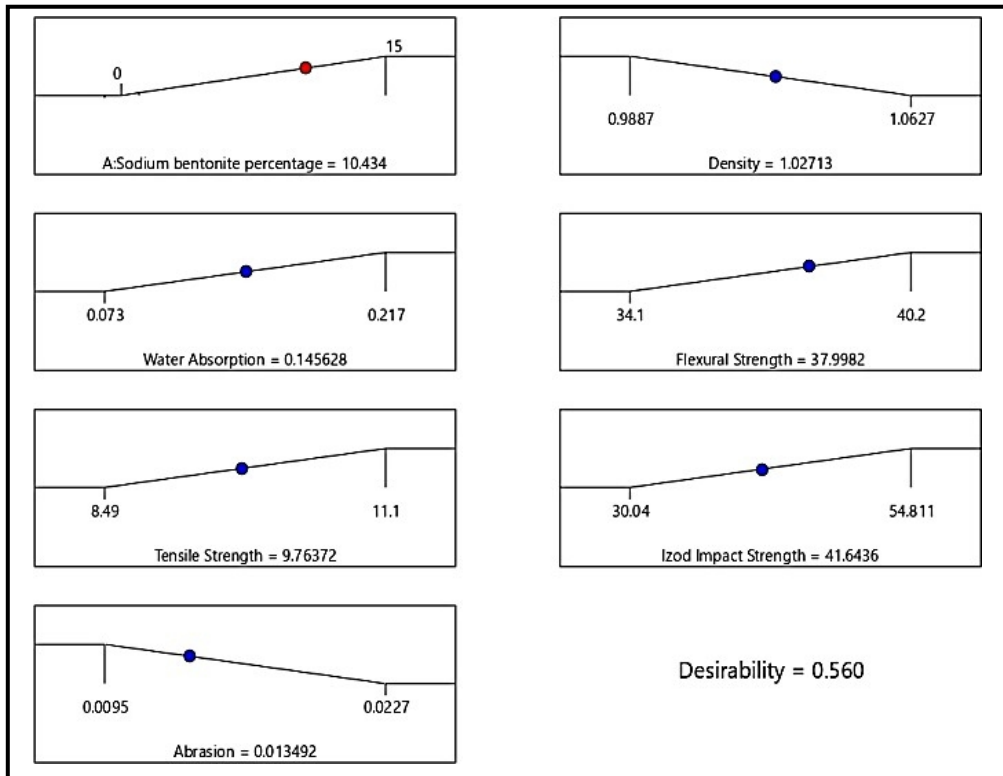


Figure 4: RSM optimal percentage of SB based on physico--mechanical properties

The desirability values of each response is shown in Figure 4. The dot on each function graph denotes the prediction value for that specific response (Adalarasan and Santhanakumar, 2015, p. 2629). Also, the height of the marks represents how desirable it is. The physical property test of the RPC conceded with 1.027g/cm³ and 0.146% for density and water absorption, respectively. Meanwhile, the mechanical property tests with 37.998 MPa, 9.764 MPa, 41.644 J/m and 0.013 g for flexural, tensile, izod impact strength, and abrasion, respectively. The responses were able to get a desirability of 0.560 together with the optimal percentage of SB at 10.434%.

4.0 CONCLUSION

Based on FTIR results, the post--consumer plastic wastes consisted of PP, PE and PL. DSC test results indicated that the polymers present in RPC are LDPE, HDPE, PP and PL with a melting temperature of 105.420°C;; 122.23°C;; 159.780°C;; and 251.630°C, respectively. The Pearson’s correlation matrix reveals the physico--mechanical properties relationship among each other. Water absorption and izod impact are both positively correlated to RPCs density. The RPCs tensile strength is strongly correlated to water absorption but negatively correlated to its flexural strength. RPCs abrasion test results are both positively correlated to water absorption and tensile strength but

negatively correlated to izod impact strength. Moreover, SEM results corroborates the SB uneven dispersion to the intercalated plastic and poor adhesion between SB and plastic wastes. Further, RSM suggests the SB optimum mix of 10.434% for all the tests considered. Therefore, RPC with 10.434% SB was determined to be acceptable to use as a geomembrane based on physico--mechanical properties considered.

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