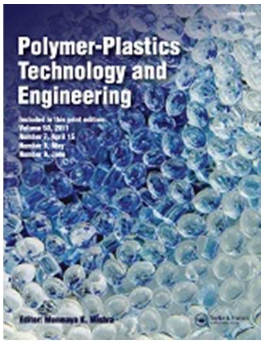

A Sustainable Polymer Composite from Recycled Polypropylene Filled with Shrimp Shell Waste

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Complete List of Authors:	Mohamad, Noraiham; Universiti Teknikal Malaysia Melaka, Department of Engineering Materials Abd Latiff, Anisah; UTeM, Ab Maulod, Hairul Effendy; UTeM, Azam, Mohd Asyadi; Universiti Teknikal Malaysia Melaka, Abd Manaf, Mohd Edeerozey; UTeM,
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A Sustainable Polymer Composite from Recycled Polypropylene Filled with Shrimp Shell Waste

[1]Noraiham Mohamad, [1]Anisah Abd Latiff, [2]Hairul Effendy Ab Maulod,
[1]Mohd Asyadi Azam, [1]Mohd Edeerozey Abd Manaf

[1]Department of Engineering Materials, Faculty of Manufacturing Engineering, Universiti Teknikal Malaysia Melaka, Durian Tunggal, 76100 Melaka, Malaysia

[2]Department of Manufacturing Engineering Technology, Faculty of Engineering Technology, Universiti Teknikal Malaysia Melaka, Durian Tunggal, 76100 Melaka, Malaysia

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*** Correspondence to**
Noraiham Mohamad
Faculty of Manufacturing Engineering,
Universiti Teknikal Malaysia Melaka,
Durian Tunggal, 76100 Melaka, MALAYSIA.
Phone: +606-3316976, Fax: +606-3316411
e-mail: noraiham@utem.edu.my

ABSTRACT

This research explores the potential of using recycled polypropylene (PP) incorporated with shrimp shell waste to produce a sustainable polymer composite. In this study, the mechanical and physical properties of recycled polypropylene/shrimp shell (rPP/SS) composites prepared by melt compounding and compression molding techniques were evaluated. The effects of SS loading were investigated by using various compositions of rPP/SS composites, ranging from 0 to 8 wt% SS that consists of two different sizes, i.e., fine and coarse SS. The composites were tested for their mechanical and physical properties using impact, tensile and water absorption tests. Furthermore, the morphology of the composites was examined by using a Scanning Electron Microscopy (SEM). Incorporation of SS was found to increase the Young's modulus of the rPP, but the impact and tensile strength showed a decrease. However, we observed that both the impact and tensile strength improve with the further increase of the SS content. In other words, composites with high shrimp shell loading were observed to exhibit better tensile and impact properties compared to composites with low shrimp shell loading. Moreover, at 8 wt% of SS, the value of tensile strength is comparable to that of neat rPP.

Key words: Waste-derived material, Shrimp shell waste, Recycled polypropylene

1. Introduction

Plastics consume approximately 8 per cent of world oil production, i.e., about 4 per cent as raw material for plastics and 4 per cent as energy for manufacture. However, about half of the total plastic produced is used only for single-use disposable applications such as packaging, agricultural films and disposable consumer items which eventually end up as plastic waste. [1,2] In Malaysia, there has been a rapid increase in municipal waste generation as reported by Othman [3], thus requires an urgent need for a better managed disposal option. The increasing waste commands for larger landfill sites that will directly contribute to noise, dust and odor as well as possibly bio-aerosols which are released soon after opening and possibly for several decades thereafter. Besides, it is a global issue where landfills contribute to the greenhouse effect due to gas emissions.

One of the methods to reduce the production of the everyday waste is through recycling, of which used materials are reprocessed into new products. It prevents the waste of potentially useful materials, reduces the consumption of raw materials, and reduces energy usage and hence greenhouse gas emissions, compared to virgin product. Recycling saves space in landfills and reduces the amount of virgin materials that must be mined or manufactured to make new products, saving energy and reducing global climate change in the process.

Polypropylene (PP) is a common thermoplastic, which is utilized in a wide variety of industrial applications due to its impressive range of properties exhibited and its ease of processing. Various studies have been carried out to explore the ability of this polymeric material [4-6]. However, PP is non-degradable due to its chemically stabilized state for long service life; hence the disposal of PP plastic waste causes an environmental issue. Therefore, the recycling of PP has increased significantly due to the economic and environmental factors. Recycled PP (rPP) has been used by mixing with other virgin thermoplastic materials to achieve low cost end products and this approach is almost similar to that of recycled polyethylene [7, 8].

As the properties of rPP are lower compared to virgin PP, incorporation of fillers such as coir, rice husk, bamboo and calcium carbonate, [9-13] is effective to produce rPP-based composites for intended technical applications utilizing its ecological and economic advantages as a recycled material. Such biomass-derived natural fiber reinforcements have received considerable attention during the recent past due to the cost effectiveness and increased environmental awareness and ecological concerns [14]. Generally plant-derived organic fillers such as coir, rice husk and bamboo show poor compatibility with the matrix polymer due to the polarity different between hydrophilic natural filler and hydrophobic polymer. Therefore, in order to improve the compatibility with the matrix polymer, filler modification such as alkaline treatment [15-17] and silane treatment [18,19] are required.

Shrimp shell is a biomass-derived material and totally biodegradable in the natural environment. According to Ravichandran et al. [20], there are many inorganic elements in the body of shrimp associated with the skeletal structure and biochemical involved in vital physiological functions. Shrimp shell contains chitin which is the main component of the cell walls of the exoskeletons of arthropods like shrimp [21, 22]. Chitin may be described as cellulose with one hydroxyl group on each monomer substituted with an acetyl amine group. The reinforcing effect of chitin results from the formation of a percolating network based on hydrogen bonding forces which allows good interaction with polymer matrix [23]. Utilization of shrimp shell as a biofiller potentially gives an added-value to shrimp shell waste as well as reduces the cost of the composite. Incorporation of shrimp shell powder into low density polyethylene (LDPE) was reported to increase its Young's modulus and yield stress [24].

This study evaluates the effectiveness of the shrimp shell (SS) waste as filler in recycled polypropylene (rPP) composite. There is no study reported on such composite to the best of our knowledge. The composites of rPP and SS are prepared by melt mixing and compression molding methods. Two different types of SS are used in this study, i.e., fine and coarse. The mechanical and physical properties are investigated by performing tensile, impact and water absorption tests. The morphologies of various compositions of rPP/SS composites are analyzed using Scanning Electron Microscopy (SEM).

2. Experimental

2.1 Materials

Recycled Polypropylene (rPP) was obtained from polypropylene based food container waste. It was confirmed by functional groups analysis using FTIR spectroscopy. The rPP was transformed into pellets by crushing the plastic waste into small pieces using an industrial crusher. Shrimp shell (SS) used in this study belongs to a family of P.indicus and was obtained from domestic waste. In order to eliminate the foul odor, the raw SS was boiled in water at 100 ^{o}C and subsequently dried in a vacuum drying oven (O Lab Tech, LVO-2030) at 100 ^{o}C for 1.5 hour to remove the water content. The SS in this study was prepared in two different sizes, i.e., coarse and fine. The coarse SS was obtained by manually tearing up the SS waste, while the fine SS was obtained by chopping using a conventional blender. The average size of the coarse SS is about 3-5 mm while the fine SS is in the range of 100-300 μm.

Compounding of rPP and shrimp shell was carried out using internal mixer (Thermo Electron, Haake Rheomix OS) at 190 ^{o}C and rotor speed of 50 rpm for 15 minutes. Initially, the unmixed rPP was compounded alone for 12 min. Then, the SS was added into the chamber and further mixed for 3 min. The mixture was left to cool down to room temperature before crushed into pellet size using a crusher. Subsequently, the mixture was compression molded using a hydraulic presser to obtain a composite sheet. The compression molding process involved pre-heating at 185 ^{o}C for 6 min, followed by compression for 5 min at the high temperature and subsequently, cooling under pressure for 5 min. Three different compositions of rPP/SS composites were prepared i.e., 97/3, 95/5 and 92/8, in addition to neat rPP.

2.2 Measurements

Tensile tests were performed using Universal Testing Machine (Shimadzu, Autograph) at a crosshead speed of 5.0 mm/min. The tests were carried out at 23 °C and 50 % of humidity. For each formulation, 8 samples were tested. The rPP/SS composite sheets were cut into dog bone shape specimens according to the ASTM D638-03 standard, using a specimen cutter machine (Gotech, GT 7016–H).

Impact tests were carried out using a pendulum impact tester by RKT Cooperation, Malaysia equipped with an impact force hammer of 25 J. For impact tests, samples were prepared according to ASTM D 256, of which the dimension of a standard specimen was 64.0 x 12.7 x 3.2 mm.

Water absorption tests were carried out according to ASTM D 570-98. Samples were cut into a rectangular shape with a dimension of 76.2 × 25.4 × 3.2 mm. Three samples were tested for each formulation. The samples were initially conditioned by drying in an oven at 105 ºC for 24 hours, before cooled down to room temperature and weighed. Then, the samples were immersed in distilled water for 24 hours at room temperature. After a certain period, they were removed from the water and dried by softly wiping with a clean cloth before being weighed. Percentage of water absorption was calculated using Eq. 1, as below:

$$Water\ Absorbtion, \% = \frac{Wet\ weight - Conditioned\ weight}{Conditioned\ weight} \times 100\% \qquad (1)$$

The examination of the tensile fracture surfaces were carried out using a Scanning Electron Microscope (Zeiss, EVO 50) at magnifications of 100x and 1000x. The tensile and impact fractured surfaces were coated with gold prior to examination under the electron beam. For each sample, a minimum of three micrographs were taken using the same magnification. The distribution, shape and size of the dispersed shrimp shell particles were also observed and analyzed qualitatively. The micrographs were captured under variable pressure operated at 20 kV.

3. Results and Discussion

3.1 Tensile Properties

Tensile tests of the rPP/SS composites were performed to evaluate the effects of the SS content on their mechanical properties. The ultimate tensile stress, Young's modulus and percentage of elongation at break for each formulation were obtained from the resulted stress-strain curves and depicted in Fig. 1, 2 and 3 respectively.

As shown in Fig. 1, it is found that the addition of 3 wt% SS reduces the ultimate tensile strength of the rPP, irrespective of the SS size. For all compositions, composites with fine SS consistently show a higher tensile strength compared to those incorporated with coarse SS of the same amount. The result indicates that the fine SS is more effective as reinforcement in rPP than the coarse SS. The finer SS has a higher total surface area for a given particle loading that provides a more efficient stress transfer mechanism, thus results in the higher tensile strength. These results are in agreement with the study of mechanical properties in kaolin filled nylon 6, 6 composites by Buggy et al. [25]. The study reported that the composites strength increases with decreasing of the particle size.

Moreover, it is observed that composites with coarse SS do not show any significant improvement in tensile strength, even when the amount of filler is increased up to 8 wt%. On the other hand, composites with fine SS show a steady increase in the tensile strength with the increase of the amount of fine SS. This trend is similar to that exhibited by recycled low-density polyethylene (rLDPE) reinforced with fine SS powder as reported by Hussein et al. [24] Furthermore, at 8 wt% of fine SS, the value of tensile strength is found to be comparable to the neat rPP.

[Figure 1][Figure 2]

The increase of tensile with the increase of filler loading strength in the rPP/fine SS composites suggests good interfacial bonding between the filler and the polymer matrix interface, which will increase the difficulty to form the crack propagation in the material. The good interfacial bonding can be attributed to the high compatibility between the elements exist in shrimp shell such as chitin and calcium carbonate with the rPP. The molecular structure of chitin is similar to cellulose with one hydroxyl group on each monomer substituted with an acetyl amine. Furthermore, chitin also can act as natural coupling agent [26]. This allows for increase hydrogen bonding between the rPP and chitin, thus provides a better interfacial bonding between the SS-rPP interface for stress-transfer [27], and consequently improve the mechanical properties of the fabricated composites.

The result is supported by the image of the tensile fracture surface by SEM in Fig. 2, in which more filler breakage are observed compared to fiber pull-out. Filler breakage was occurred due to the stress of the external force is exceed the tensile strength between the filler and matrix, whereas the pull out occur due to the poor wettability between the filler and matrix. The SEM micrograph indicates a good wetting between SS filler and rPP in the composite containing 8 wt% fine SS.

Furthermore, the reduction of the tensile strength from 3 to 5 and 8 wt% of filler loading for the coarse SS possibly due to the non-uniformity of the particle distribution and the forming of agglomerations of the coarse SS filler. The agglomerates will act like foreign body in the composites and could initiate failure under stress. This main factor of reduction of the tensile strength is due to the presence of the agglomerations has also been reported elsewhere [28-31].

[Figure 3][Figure 4]

The result of the modulus of elasticity as shown in Fig. 3 clearly shows that the stiffness of the composites increases with the addition of 3 wt% SS filler compared to the virgin rPP, then decreases when the amount of the SS filler is increased. Despite the decrease, composites containing 3, 5, 8 wt% of SS filler shows a higher modulus of elasticity than neat rPP indicating that addition of SS increases the stiffness of the material. Moreover, it is revealed that composites containing coarse SS show higher Young’s modulus than composites with fine SS.

The effects of amount and filler size on the impact strength of rPP/SS composites are depicted in Fig. 4. It is obvious that SS filler addition decreases the impact strength in rPP significantly. However, the impact strength of the SS/rPP composites slightly improves when the filler content is increased, indicating the positive contribution of SS to increase toughness in rPP composites. Furthermore, it is found that composites incorporated with fine SS consistently show higher impact strength than those with coarse SS, indicating that the smaller particle size improves the interfacial bonding between the matrix and filler. As a result, the rPP/fine SS composites are capable of absorbing higher impact energy to stop crack propagation compared to the rPP/coarse SS composites.

3.2 Water Absorption

Water absorption is one of the key parameters in evaluating natural filler/thermoplastic composites. The results of the water absorption test of rPP/SS composites after 7 days of immersion is summarized in Fig. 5. As shown in the figure, the amount of water absorbed is higher in rPP/SS composites than in neat rPP, indicating the hydrophilic nature of the SS. Furthermore, it is found that water absorption increase with the increase of the SS content.

[Figure 5]

The hydrophilic nature observed in SS is associated to the presence of chitin in SS. Chitin is described as cellulose with one hydroxyl group on each monomer substituted with an acetyl amine group. The hydroxyl groups in chitin have a tendency to form hydrogen bonds with water molecules, thus have criteria of hydrophilic molecules [32]. Each hydroxyl (-OH) group on a chitin can make hydrogen bonds to three different water molecules. The hydrogen can bond to a pair of valence electron on the oxygen of water and each of the two pairs of valence electrons of the hydroxyl can bond to hydrogen of water. Most of the molecules within cells can form H-bonds and are hydrophilic. The water absorption the recycled PP was less than the composites materials due to its hydrophobic nature.

Furthermore, it is found that the amount of water absorbed is higher in composites reinforced by coarse SS, indicating a better water resistant property of the composites incorporated with fine SS. This result is attributed to the difference in size of the fillers. A smaller filler size provides a higher surface area, thus enables higher interaction between the filler and the matrix polymer as reflected in the slightly higher values of tensile strength of the composites with fine SS shown in Fig. 1. According to Akil *et*. al [33], one of the water absorption mechanism into composites materials is by capillary transport of water molecules into the gaps and flaws at the interface between dispersed phase and matrix. Thus, a higher interaction between filler and matrix lowers the water absorption at filler/matrix interfaces. Similar results regarding the effect of filler size on water absorption have been reported by other researchers elsewhere [34, 35].

4. Conclusions

Mechanical and water absorption properties of rPP filled with various amount of SS are studied. Two different types of SS are employed to study the effect of filler size on these properties. It is found that addition of a small amount of SS (3 wt%) reduces the ultimate tensile strength of the rPP. However, in composites filled with fine SS, the value of tensile strength increases steadily with the increase of the filler. Furthermore, at 8 wt% of fine SS, the tensile strength is found to be comparable to that of neat rPP. Young's modulus was found increased with the addition SS filler. Contrary to the results for tensile strength, the value of Young's modulus is found to decrease with the increase of SS. Impact strength shows a similar pattern to what observed in tensile strength, i.e., it decreases with the SS addition, but as the filler amount was increased the value shows an increase. The size of SS filler also influences the mechanical properties of the composites. Fine SS is found to be more effective as reinforcing filler than coarse SS as indicated by the results of tensile and impact strength. Furthermore, composites filled with fine SS also found to be more water resistant than composites with coarse SS as a result of higher interaction between the filler and the matrix polymer as reflected in their higher values in tensile strength.

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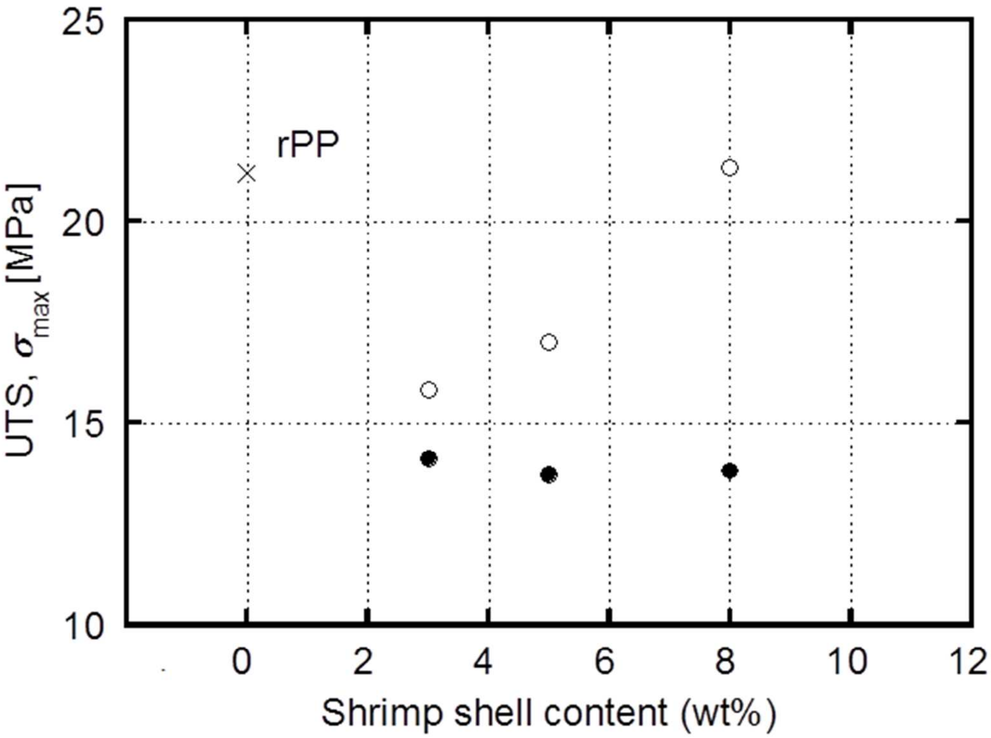
Figure Captions

Figure 1	Relation between shrimp shell content and tensile strength for rPP/fine SS (open diamonds) and rPP/coarse SS (closed diamonds) composites. The value of tensile strength for neat rPP (cross sign) is given for comparison.
Figure 2	SEM image of the fractured surface at 100x of magnification of rPP containing 8 wt% of fine SS.
Figure 3	Relation between shrimp shell content and Young's modulus for rPP/fine SS (open diamonds) and rPP/coarse SS (closed diamonds) composites. The value of Young's modulus for neat rPP (cross sign) is given for comparison.
Figure 4	Relation between shrimp shell content and impact strength for rPP/fine SS (open diamonds) and rPP/coarse SS (closed diamonds) composites. The value of impact strength for neat rPP (cross sign) is given for comparison.
Figure 5	Relation between shrimp shell content and water absorption for rPP/fine SS (open diamonds) and rPP/coarse SS (closed diamonds) composites. The water absorption for neat rPP (cross sign) is given for comparison.


25
20
15
10
UTS, σ_{max} [MPa]
rPP
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2
4
6
8
10
12
Shrimp shell content (wt%)

Figure 1

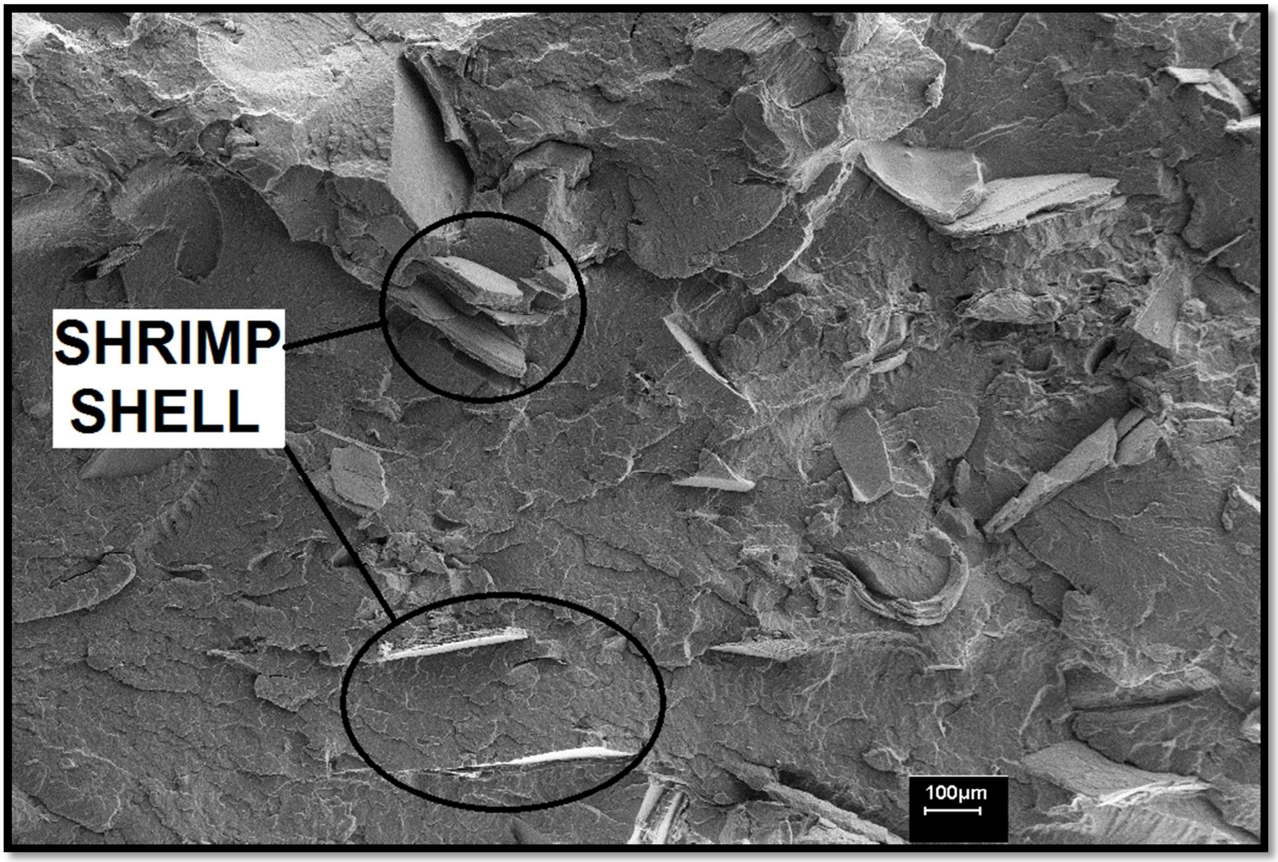


Figure 2

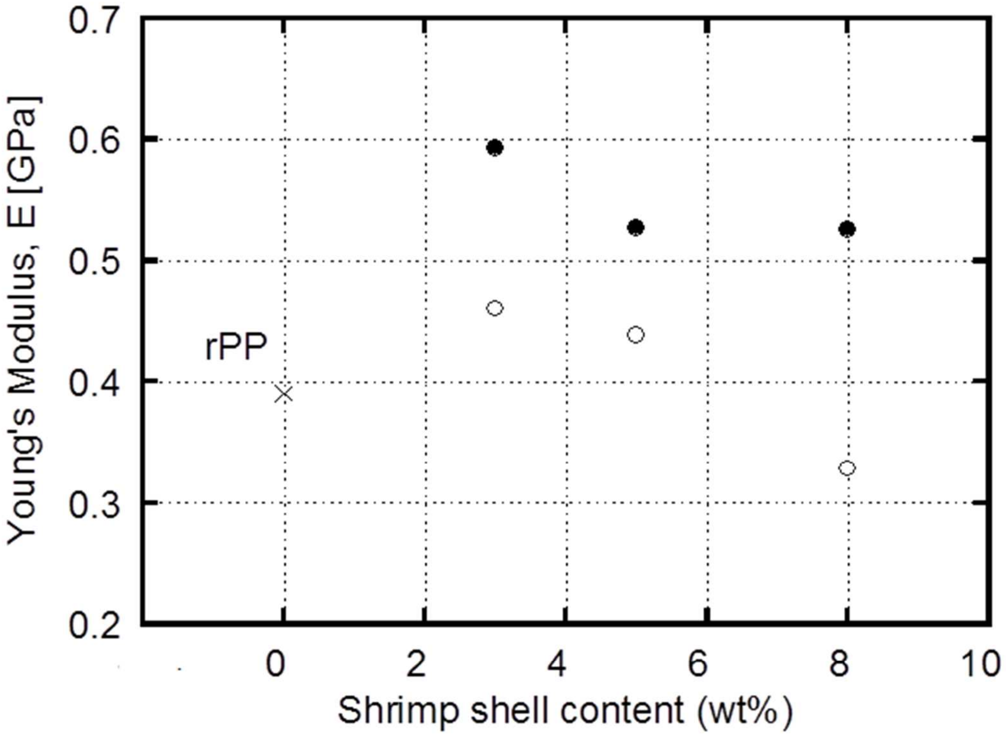
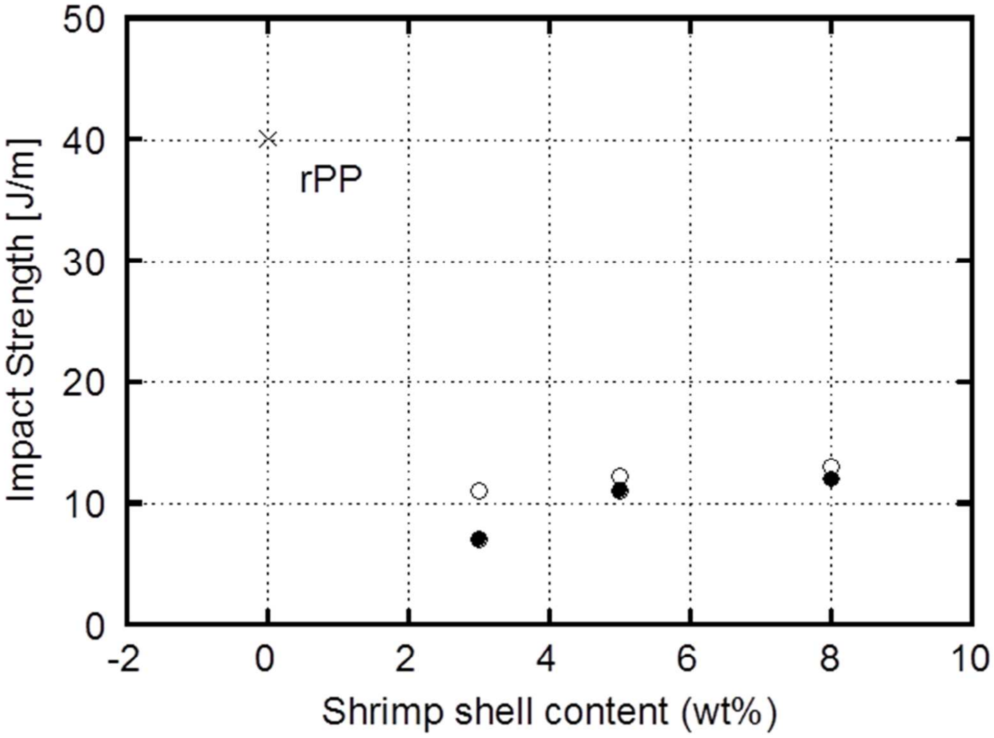

0.7
0.6
0.5
0.4
0.3
0.2
Young's Modulus, E [GPa]
rPP
0
2
4
6
8
10
Shrimp shell content (wt%)

Figure 3


50
40
30
20
10
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-2
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2
4
6
8
10
Impact Strength [J/m]
Shrimp shell content (wt%)
rPP

Figure 4

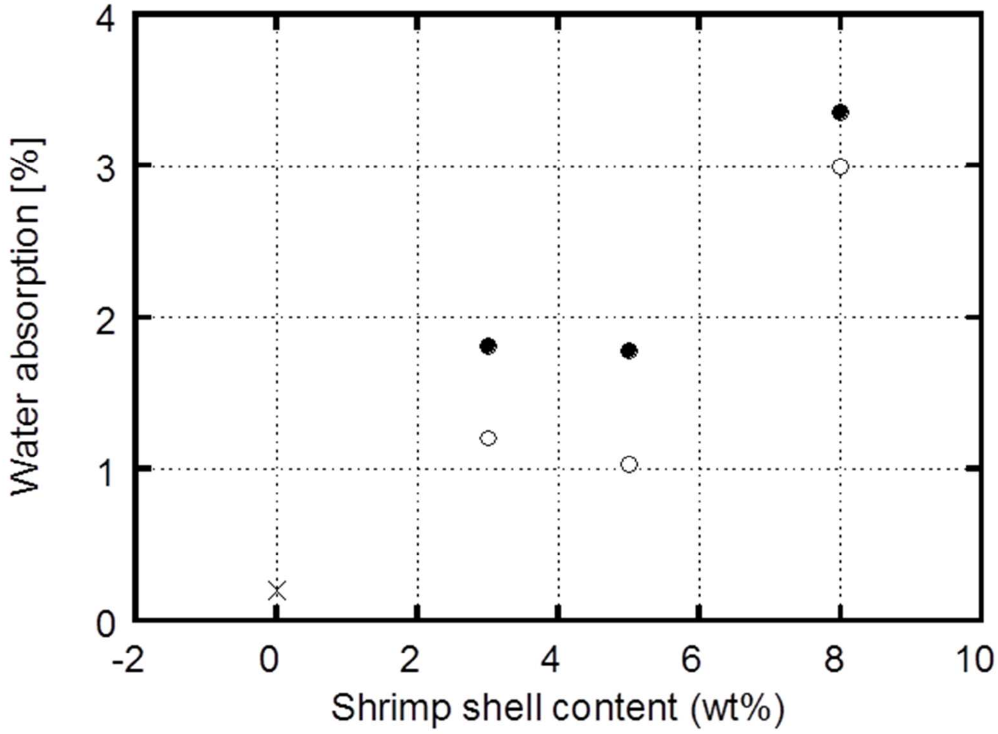

Figure 5