

REVISED RESEARCH ARTICLE

Effects of MAPI/HMDA dual Compatibilizer and Filler loading on Physico-Mechanical and Water Sorption Properties of Natural Rubber/Poly (ethylene terephthalate) (PET)/Dikanut Shell Powder Bio-Composites

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Authors' contributions

This work was carried out in collaboration between all authors. Author GNO designed the study, selected the materials, performed the experiments and data analysis, wrote the first draft and finalized the manuscript. Authors EGO, IOA, MJU, UOE and COU performed the experiments, collected the data, and conducted the analyses of the study and literature search. All authors read and approved the final manuscript.

ABSTRACT

The effects of maleated polyisoprene (MAPI)/hexamethylene diamine (HMDA) dual compatibilizers and filler loading on physico-mechanical properties of natural rubber (NR)/poly(ethylene terephthalate) (PET)/dikanut shell powder (DSP) bio-composites have been investigated at filler loadings, 0 to 8 wt. %. NR/PET composition was fixed at 60/40 wt. %. Dikanut shell powder was prepared at three particle sizes of 120, 135, and 150 μm . The composites were prepared by reactive compatibilization in a two-roll mill. Results obtained have demonstrated that a combination of MAPI and HMDA is an excellent dual reactive compatibilizer for NR/PET/DSP bio-composites as shown by marked improvement in the properties investigated. The tensile strength, tensile elongation, unnotched impact strength, and melt flow rate of the composites were found to decrease with increase in filler loadings. On the other hand, the hardness of the composites increased with increases in filler loadings. However, the incorporation of MAPI/HMDA into the composites was found to improve these properties. Both the compatibilized and uncompatibilized composites absorbed water, the amount of which increased with increase in filler loading. The amount of water absorbed by the composites was substantially decreased on addition of MAPI/HMDA into the composites.

Keywords: Bio-composites; natural rubber; poly (ethylene terephthalate); dikanut shell powder; reactive compatibilization.

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1. INTRODUCTION

Fillers are intentionally added to polymeric materials to reduce the cost of the material and also improve the mechanical and dynamic properties of the products. The fillers used in polymeric materials can be classified into

reinforcing and non-reinforcing types. Reinforcing fillers are fillers which increase the tensile strength, hardness and abrasion resistance of the polymer product. They achieve performance enhancement by forming strong chemical bonds with the polymer, that is, strong filler-polymer interactions. Carbon black and silica are the most commonly consumed reinforcing fillers in the rubber industry. For example, carbon black imparts strength and toughness to rubbers, improves the rubber's resistance to tearing, abrasion and flex-fatigue, and also increases traction and durability [1]. Silica and other types of fillers have a weaker polymer-filler interaction and are extensively used where a high degree of reinforcement is not essential [2].

Non-reinforcing fillers cause reduction in strength properties but may increase hardness and modulus of the polymer product. They are generally applied as extenders to reduce cost; examples include calcium carbonate and china clay [1]. However, a major drawback of these conventional fillers is their non-renewability and non-biodegradability, and this has prompted research into seeking an adequate alternative. Consequently, the past decades have witnessed a growing interest in the use of renewable resources as fillers in polymeric systems. This is due to strong environmental regulations, high rate of depletion of petroleum resources, as well as increased interest in the proper utilization of renewable resources to develop eco-friendly materials [3]. Thus, research on the development of polymer composites prepared using various recycled materials is being actively pursued.

Among the possible alternatives, the development of composites using agro-wastes or lignocellulosic materials as reinforcing fillers and thermoplastic elastomers as matrices is currently receiving significant attention [4]. The growing trend in the use of lignocellulosic filler in the manufacture of polymer composite systems is due to their abundance, low density, high specific properties; and non-abrasive nature which offers decreased wear in the machine used for their production. Furthermore, they are non-toxic, renewable and biodegradable [3 – 5]. Dika nutshell powders (DSP) are natural fillers and represent an environmentally friendly alternative to conventional reinforcing fillers.

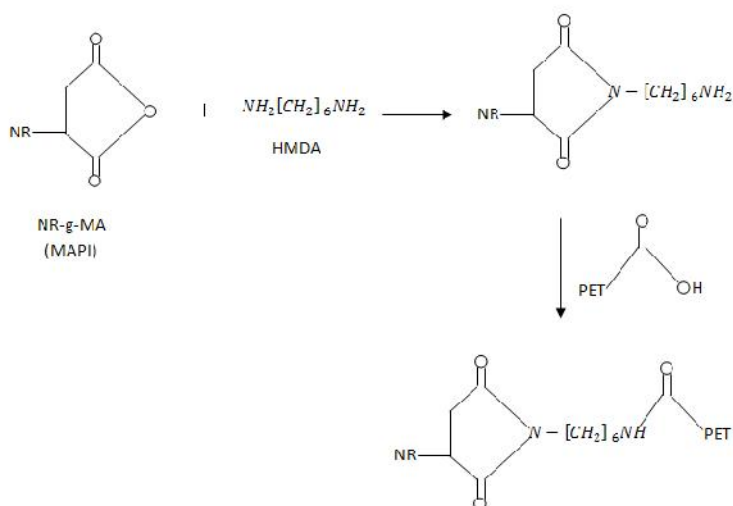
Dikanut (*Irvingia Gabonensis*), known locally as “ogbono” is a genus of African trees, valued for its fatty and protein-rich nuts. It has high content of mucilage which enables it to be used as thickening agent in foods. Among dikanut producers in Africa are Nigeria, Senegal, Uganda, Democratic Republic of Congo (DRC) and Angola [6 – 8]. The shells are a by-product of the nut processing and do not have any known domestic/industrial use. They quickly become pollutants as they are presently discarded as waste as could be unsightly found littering waste bins in our big cities and farm yards in most localities. The conversion of dikanut shell to polymeric filler has several benefits. First, it provides a cheap, renewable, abundant and biodegradable source of reinforcement to polymeric systems. Second, it offers an effective, innovative, and alternative solution to indiscriminate dumping of agro-wastes, thereby preventing them from polluting the environment either through incineration or filling up landfills and waterways.

Vast majority of polymer pairs are thermodynamically immiscible and, when blended, usually display poor properties, owing to their unstable morphology and poor adhesion between the phases. The problem is further compounded by the incompatibility of lignocellulosic fillers with non-polar polymer matrices which are mostly hydrophobic. This is due to the high polarity and hydrophilicity of lignocellulosic fillers. Thus, the main disadvantage of using DSP as reinforcing filler is the poor interaction between the DSP particle and polymer matrix [9 – 11]. Such problems have been alleviated by the introduction of a proper interfacial modifier in a process often called compatibilization. A more effective and less expensive compatibilization technique more frequently used for industrial applications is the reactive blending in which the modifier is in-situ-formed at the interface between the phases by the coupling reaction of functionalized components [12]. Some few examples of natural rubber – based thermoplastic elastomer blends are reported in the literature and include blends with linear low density polyethylene (LLDPE) [13], polypropylene (PP) [14, 15], polystyrene (PS)/natural rubber-graft-polystyrene [16], poly (vinyl alcohol) [17], and ultra-low-density polyethylene [18].

Poly (ethylene terephthalate) (PET) is a low cost commodity polymer with excellent properties, such as solvent resistance and electrical insulation, and therefore has found wide applications in the packaging industry. However, low impact resistance particularly at low temperature is the major disadvantage for PET, and this has limited its applications. The usual method to overcome this drawback is to add a second rubber phase to the PET matrix so that the rubber particles act as effective stress dissipating agents [19]. However, natural rubber and PET are

immiscible and incompatible. Thus, the resultant blends do not attain optimal properties. This is due to the polar nature of PET and non-polar character of natural rubber, which require that blends of this type be compatibilized in order to obtain a useful material that combines the strength of PET with the impact resistance of natural rubber [20]. In compatibilized polyester blends, blends of PET and polystyrene (PS) compatibilized by styrene-graft-maleic anhydride (SMA) and epoxy resin have been reported [21]. The authors revealed the non-reactivity of PET carboxyl groups with the anhydride of SMA. In a previous study [3] on the properties of reactive compatibilized dikanut shell powder-filled recycled polypropylene (PP)/PET bio-composites using maleated polypropylene (MAPP) and epoxy resin dual compatibilizers, we reported insignificant property improvements when MAPP alone was used as compatibilizer due to the low reactivity between MAPP and PET. Effective compatibilization was achieved on addition of epoxy resin (EPR) as a second component of the compatibilizer system as suggested by marked improvement in the properties investigated.

A similar problem is expected when maleic anhydride-grafted-polyisoprene (MAPI) alone is used as compatibilizer in natural rubber/PET blends in which only secondary interaction is expected. If used as the second component of the compatibilizer system, hexamethylene diamine (HMDA) is capable of producing PET-co-HMDA-co-MAPI copolymer at the interface, which is able to function as an effective compatibilizer. This study evaluates the dual compatibilizing effect of MAPI and HMDA as the reactive compatibilizer system for natural rubber/PET/dikanut shell powder (DSP) bio-composite to enhance its physico-mechanical and water sorption properties. Thus, the effects of HMDA compatibilizer on the composites were evaluated by physico-mechanical property measurements and water sorption tests. The use of MAPI/HMDA as a dual compatibilizer in polymer composite systems has not been reported in the scientific literature to our knowledge. Again, dikanut shells constitute a major part of agricultural wastes in Nigeria with no known domestic/industrial applications. Thus, it is hoped that the present study will help to convert waste to wealth by harnessing the industrial potentials of dikanut shells as filler in polymer systems. Scheme 1 illustrates the possible reactions that should take place at the interface of natural rubber- PET, during the melt blending process.



Scheme 1. Plausible reactions that occur at the interface of the compatibilized NR/PET blends

2. EXPERIMENTAL

2.1 Materials

The dikanut shells were sourced from different farm yards in Auchi, Nigeria. The natural rubber which conforms to Standard Nigerian Rubber (SNR 10) with Mooney Viscosity of 76 [ML (1+4) @ 100⁰C] was kindly supplied by Rubber Research Institute of Nigeria (RRIN), Iyanomo, Benin City, Nigeria. The rubber properties have been determined and reported in a previous work [1]. The compounding ingredients and chemicals used were industrial grades.

The compatibilizer system, maleic anhydride-grafted-polyisoprene (MAPI) containing 10 wt% maleic anhydride (Samsung Chemical Co.) and hexamethylene diamine (HMDA) (Shell Chemical Co.), and the poly (ethylene terephthalate) (PET) with a melt flow index of 20g/10min (Shell Chemical Co.) were purchased from Rovet Chemicals Ltd, Benin City, Nigeria.

The standardized compounding and test equipment were kindly provided by Ceeplast Ind. Ltd, Aba, Nigeria, the Department of Polymer Technology, Auchi, Nigeria, and the Processing Laboratory of the Department of Polymer and Textile Engineering, Federal University of Technology, Owerri, Nigeria.

2.2 Preparation of dikanut shell powder

The preparation of dikanut shell powder (DSP) was carried out according to the procedure described in our previous work [22]. Dikanut shells were washed and dried in air to remove sand particles and moisture. The dried dikanut shells were milled to fine powder and sieved through mesh sizes of 120 μ m, 135 μ m and 150 μ m. The fine particles that passed through the mesh were collected, stored in sealed plastic containers and kept in a desiccator until required.

2.3 Preparation of composite samples

The formulation given in Table 1 was used to prepare NR/PET/DSP biocomposites. The NR/PET composition and DSP filler content were fixed at 60/40 and 5 wt. %, respectively. MAPI/HMDA was used as the dual compatibilizer system. Prior to compounding, PET and MAPI were dried at 120⁰C and 90⁰C, respectively for 24 hours in separate vacuum ovens. All other ingredients were used as obtained. Compounding of the ingredients with the exception of the vulcanizing agent and accelerators was done on a laboratory size (160 x 320mm) two-roll mill at 170⁰C to produce suitable sheets. The temperature of the mill was reduced from 170⁰C to 90⁰C before introducing the vulcanizing agent and the accelerators to prevent premature curing of the compound mix. The milled samples were then cured into sheets of 2mm thick on a hot press at 5MPa pressure and temperature of 145⁰C for 20mins.

Table 1. Compounding recipe of the NR/PET/DSP biocomposites

Compounding Ingredients (wt. %)	Uncompatibilized MAPI/HMDA = 0/0	Compatibilized			
		MAPI = 2.0	MAPI/HMDA = 2.0/0.2	MAPI/HMDA = 2.0/0.4	MAPI/HMDA = 2.0/0.6
NR	60	60	60	60	60
PET	40	40	40	40	40
Zinc Oxide	4	4	4	4	4
Stearic acid	2	2	2	2	2
Wax	3	3	3	3	3
MBT ¹	0.5	0.5	0.5	0.5	0.5
TMTD ²	1	1	1	1	1
Sulphur	1.5	1.5	1.5	1.5	1.5
DSP Filler 0, 2, 4, 6, 8					

¹MBT: Mercaptobenzoylthiazole

²TMTD: tetramethylthiuram disulphide

2.4 Determination of composite properties

The physico-mechanical properties of NR/PET/DSP bio-composites were determined. The tensile strength and tensile elongation (ASTM D 638), tear strength (ASTM D 642), unnotched impact strength (ASTM D 256), Rockwell Hardness (ASTM D 785), and water-uptake [at room temperature (27°C), 24 hours] were determined by standard procedure. Melt flow rates (MFR) were measured at 280°C with a 2.16 Kg loading by using an automatic flow rate timer (Ray – Ran Co. Britain).

3. RESULTS AND DISCUSSION

3.1. Physico-mechanical properties

3.1.1 Tensile Strength

The central objective in the development of high-performance polymer composites based on immiscible components is to raise the compatibility of the component phases, so that a given stress can be transferred efficiently between phase domains. This is because compatibilized polymer composites have finer phase size and greater interfacial contact area and interfacial adhesion than the corresponding uncompatibilized samples [25].

Figure 1 shows the effect of MAPI and HMDA content on the tensile strength of the composites.

As shown in Figure 1, the tensile strength of the uncompatibilized composite was noticeably low due to poor compatibility between NR and PET on one hand and NR and DSP on the other. Clearly, the incompatibility of the composite phases has been improved significantly through compatibilization as shown by marked improvement in tensile strength. The tensile strength of the composites increased progressively with increasing content of MAPI, which is in agreement with previous study [4].

Figure 1 also showed that the tensile strength of the compatibilized composites could be enhanced progressively by increasing the HMDA content. Composite containing the smallest amount of MAPI did not exhibit additional improvement in tensile strength even with further increase in HMDA content. This observation is attributed to the small amount of MAPI (2 wt. %) in the composite which limited the quantity of the desirable copolymer required to be produced at the interface, even with progressive increase in HMDA content in the composite.

On the other hand, progressive improvement in tensile strength was obtained at higher levels of MAPI (4 and 8 wt. %). This showed that both the desired quantity of MAPI and presence of HMDA were essential for effective compatibilization of the composite phases.

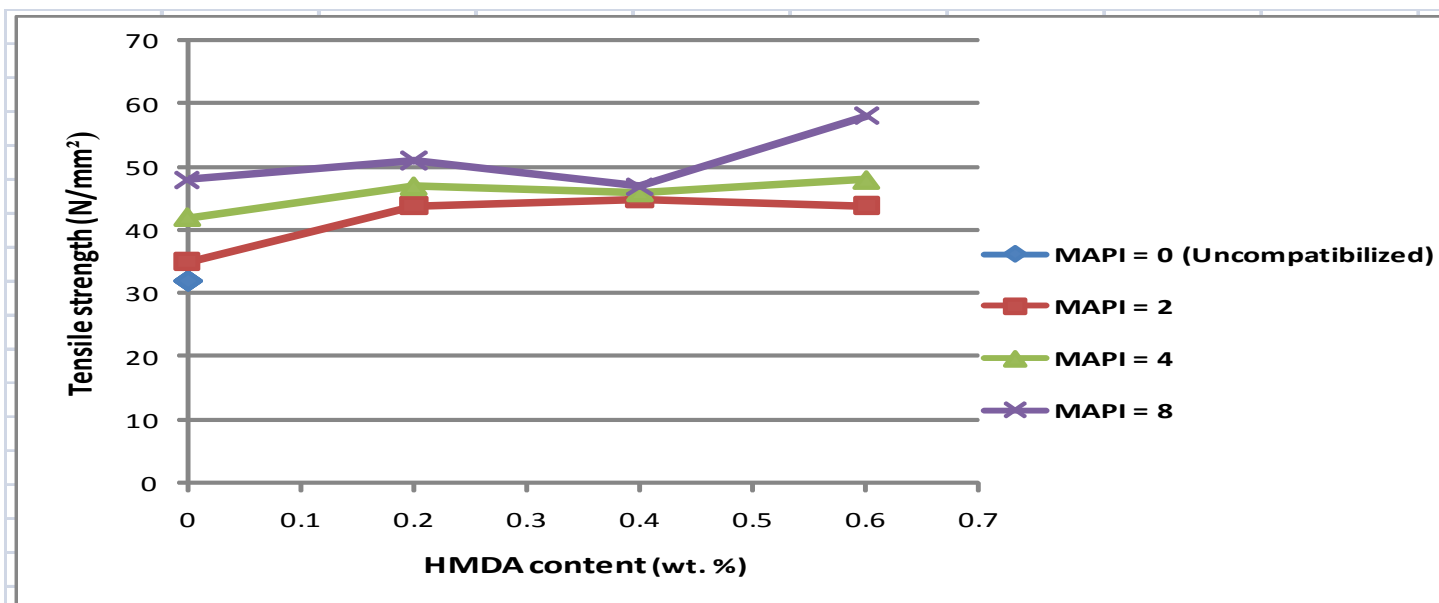


Figure 1. Plot of tensile strength versus HMDA content for compatibilized and uncompatibilized composites at 5 wt.% DSP filler loading (120µm filler particle size) and NR/PET = 60/40 composition

The effect of DSP filler loading and particle size on the tensile strength of the composites is shown in Figure 2. The result showed that the tensile strength of the composites decreased with increases in filler loading at all the filler particle sizes studied. Previous studies [4, 11, 27] have reported a decrease in tensile strength with increase in filler loading in other filled polymer systems.

Figure 2 also showed that the smaller the particle size of the filler, the higher the tensile strength of the composite at any filler loading investigated. The observed trend may be due to better dispersion of smaller sized filler particles, and improved filler-matrix interaction. Similar trend on the effect of filler particle size on composite strength had earlier been reported by some researchers while working with rice-husk flour reinforced polypropylene composites [4], oil palm wood flour reinforced epoxidized natural rubber [28], and snail shell powder-filled natural rubber [29].

3.1.2. Tensile Elongation

The effect of HMDA content on tensile elongation of compatibilized and uncompatibilized composites is presented in Figure 3. The result showed increases in tensile elongation with increasing quantity of HMDA. Again, for the composite containing the smallest amount of MAPI (2 wt. %) only limited additional enhancement in tensile elongation was observed indicating that the use of MAPI required the presence of HMDA to promote interfacial adhesion between otherwise gross-phase-separated composite phases by reducing the interfacial energy between the phases. The increased plasticization of the NR/PET blends and improved wetting of the filler-matrix interface are the considered factors responsible for the increases in tensile elongation with increasing HMDA content.

The effect of DSP filler loading and particle size on tensile elongation of the composites is presented in Figure 4. As clearly shown in Figure 4, tensile elongation decreased with increase in filler loading at all filler particle sizes

investigated. The filler particles which are encapsulated in the polymer matrix decreased the molecular mobility of the matrix which decreased further with increase in filler loading. This is considered to be responsible for the observed decrease in tensile elongation with increases in filler loading.

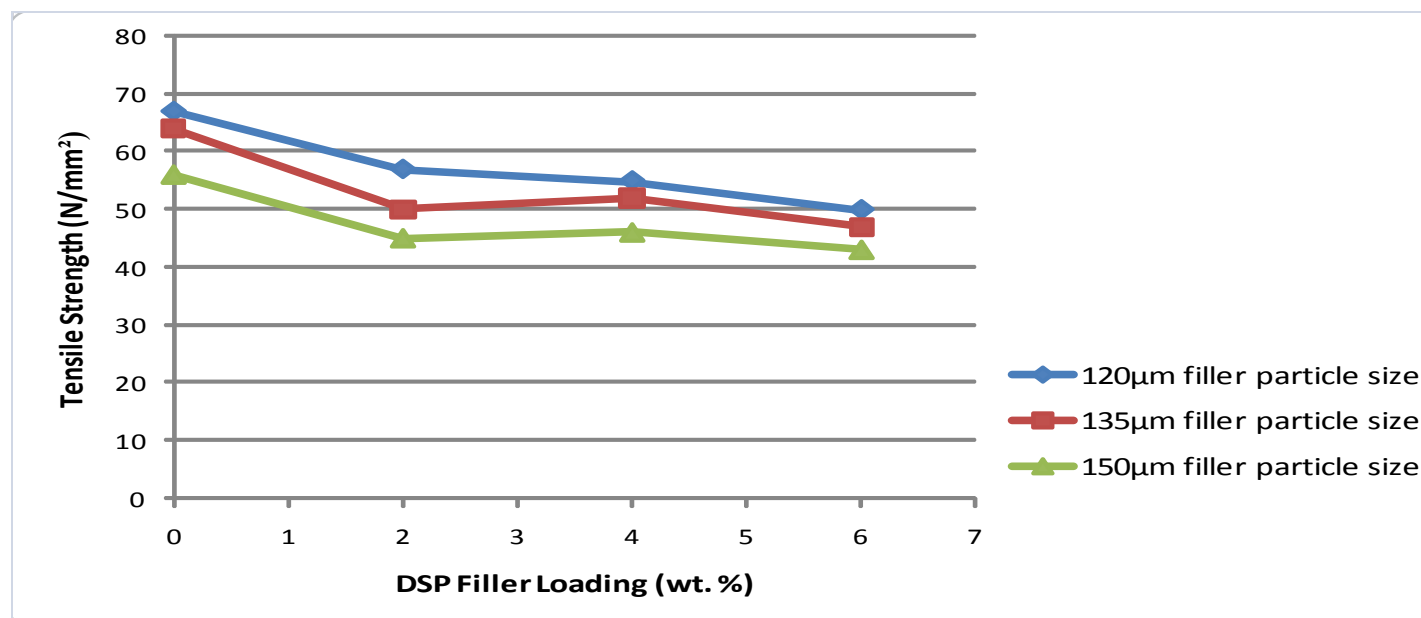


Figure 2. Plot of tensile strength versus DSP filler loading for compatibilized composites at MAPI/HMDA = 8/0.6 and NR/PET = 60/40 wt.% composition

3.1.3. Tear Strength

The effect of HMDA content on tear strength of compatibilized and uncompatibilized composites is presented in Figure 5. The result showed that tear strength increased with increases in the quantity of HMDA. Again for the composite containing the smallest amount of MAPI (2 wt. %) only limited additional increment in tear strength was observed. This result is in agreement with the envisaged plausible reactions which occur during processing of the compatibilized composites (Scheme 1). As shown in Scheme 1, the use of MAPI required the presence of HMDA to promote the coupling reaction. The amino groups of HMDA react with the anhydride groups of HMDA forming the imide group. The other amino groups in this new functionalized polyisoprene may react with the carboxyl groups of PET; and could also form intermolecular hydrogen bonding with the hydroxyl groups of DSP. These chemical and physical interactions give rise to a network at the interface, which is responsible for the observed marked increase in the tensile strength.

Go and Ha [30] in their studies of ethylene-propylene–diene terpolymer (EPDM)/butyl rubber (BR) blends had earlier reported that AAHR (a mixture of aliphatic and aromatic hydrocarbon resins) was very effective in plasticizing EPDM/BR and enhancing the compatibility between EPDM and BR. The authors revealed that the addition of AAHR increased the amount of bound rubbers, which improved the tear strength of the EPDM/BR blends.

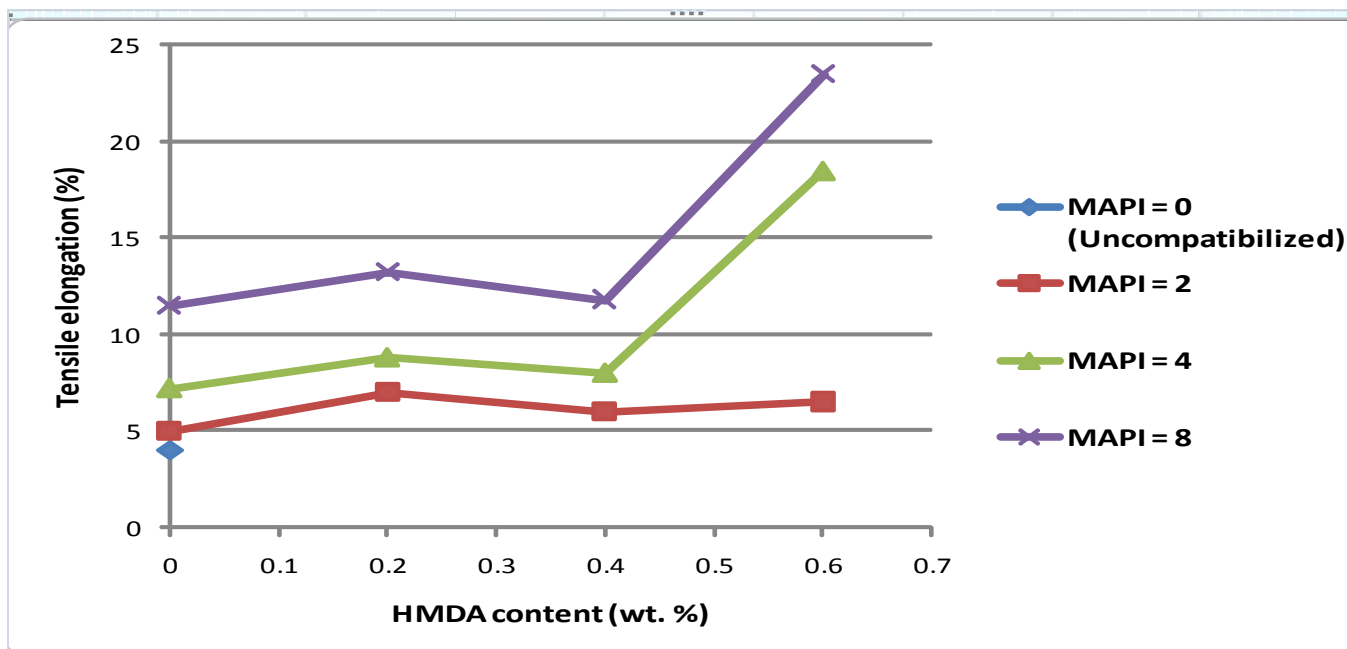


Figure 3. Plot of tensile elongation versus HMDA content for compatibilized and uncompatibilized composites at 5 wt.% DSP filler loading (120 μ m filler particle size) and NR/PET = 60/40 composition

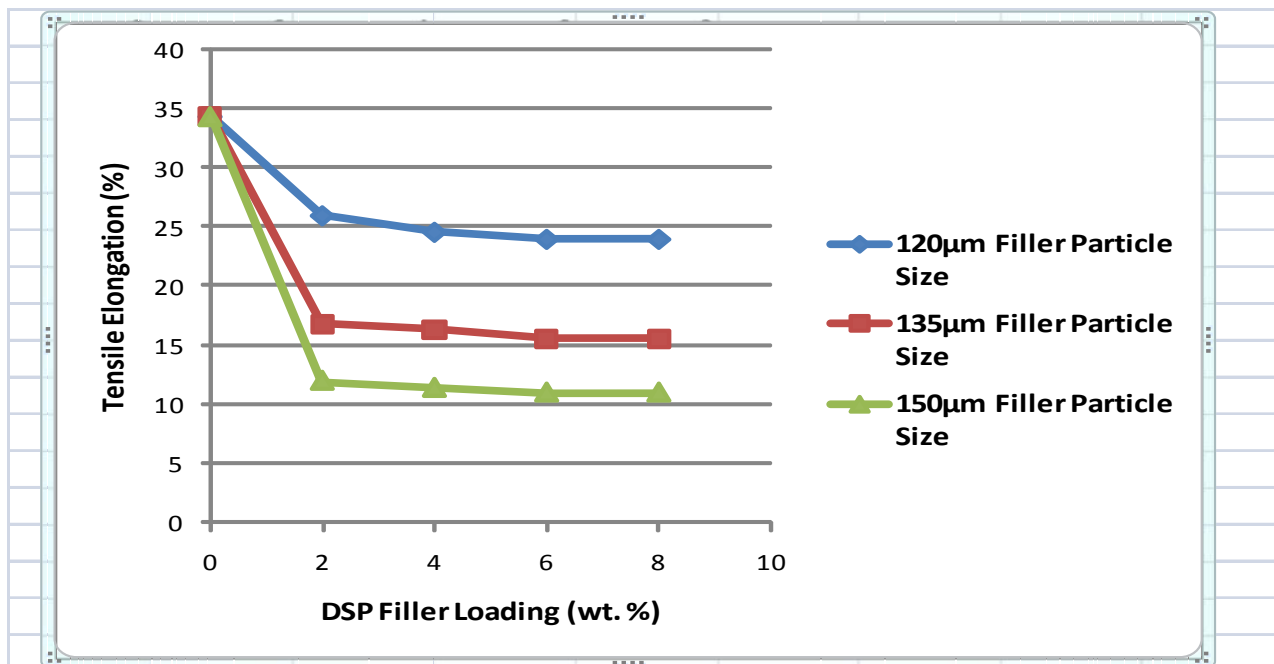


Figure 4. Plot of Tensile Elongation versus DSP Filler Loading for Compatibilized Composites at fixed MAPI/HMDA = 8/0.6 and NR/PET = 60/40 wt. % Compositions

3.1.4. Unnotched Impact Strength

Previous studies [19] have shown that unnotched impact strength is commonly used to differentiate toughness change resulting from compatibilization for notch sensitive polymeric materials. The effect of HMDA content on the unnotched impact strength of the composites is presented in Figure 6. It could be seen that the trend of unnotched impact strength exhibited by the composites with varying HMDA content was consistent with corresponding tensile properties (tensile strength, tensile elongation and tear strength).

Figures 1, 3, and 5 showed that the enhancement of the physico-mechanical properties of the composites by HMDA was significant. This is attributed to the in-situ-formed NR-co-HMDA-co-MAPI copolymer molecules which anchor along the interface, as well as the intermolecular hydrogen bonding formed between the amino groups of the newly functionalized polyisoprene and the hydroxyl groups of DSP as a result of improved wetting (Scheme 1). The number of the in-situ-formed copolymer molecules tended to increase with increasing MAPI and HMDA content.

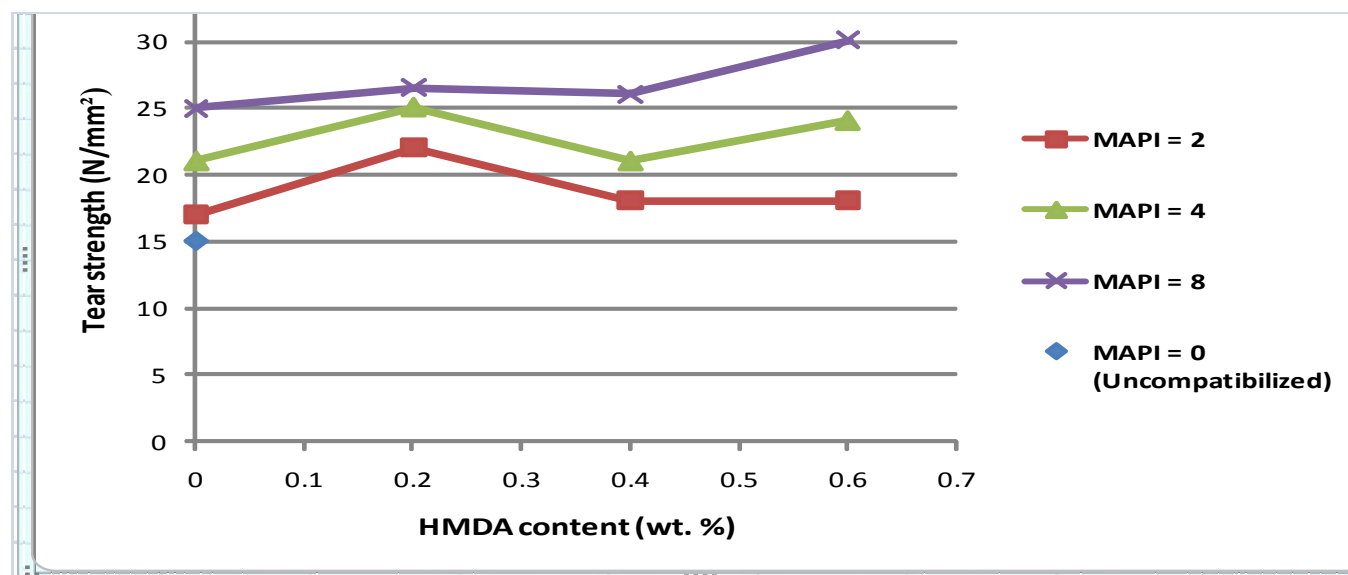


Figure 5. Plot of tear strength versus HMDA content for compatibilized and uncompatibilized composites at 5 wt.% DSP filler loading (120µm filler particle size) and NR/PET = 60/40 composition

Figure 7 showed the effect of filler loading and filler particle size on the unnotched impact strength of the compatibilized composites. As shown in Figure 7, the unnotched impact strength of the composites decreased as the filler loading increased. The decreasing trend in unnotched impact strength with increasing filler content may be due to the poor interfacial bonding between the filler and matrix polymer which caused micro-cracks to occur at the point of impact thereby permitting easy propagation of cracks in the composite.

The findings of this study are in agreement with the work of Yang et al. [4] who reported that the addition of rice-husk flour filler led to the creation of an interface between the filler and polypropylene matrix, which constituted stress concentrating and crack initiating point. This resulted in significantly reduced impact strength in the unnotched composite specimen. Figure 7 also revealed that the unnotched impact strength of the composite decreased with increasing filler particle size due to better filler dispersion and improved interfacial adhesion between filler and matrix polymer with decreases in filler particle size.

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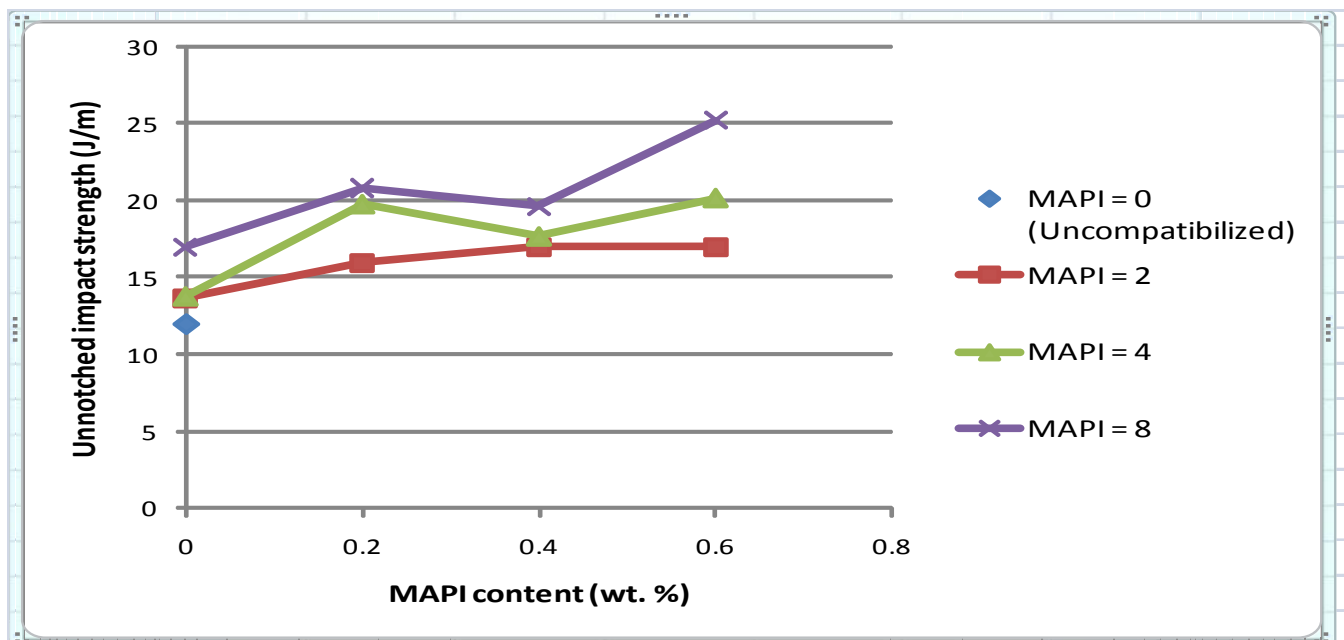


Figure 6. Plot of unnotched impact strength versus HMDA content for compatibilized and uncompatibilized composites at 5 wt.% DSP filler loading (120µm filler particle size) and NR/PET = 60/40 composition

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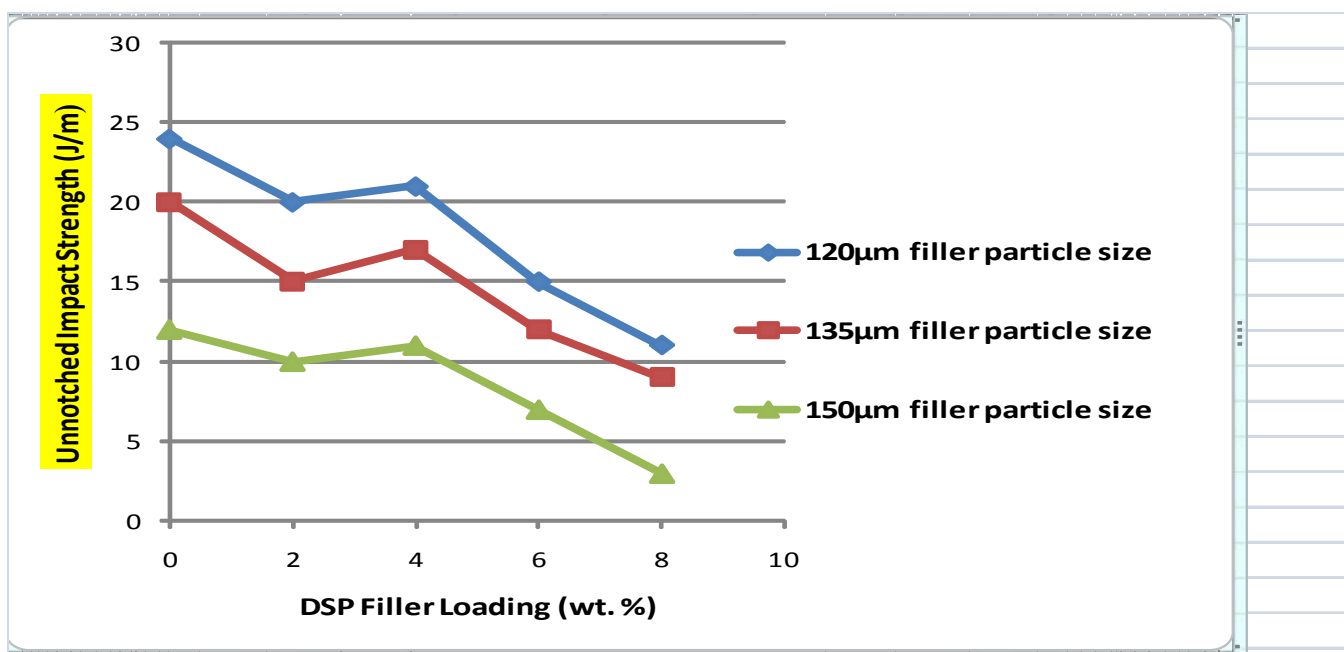


Figure 7. Plot of Unnotched Impact Strength versus DSP Filler Loading for Compatibilized Composites at fixed MAPI/HMDA = 8/0.6 and NR/PET = 60/40 wt. % Compositions

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3.1.5. Melt Flow Rates

Figure 8 showed the effect of HMDA content on melt flow rates of the composites. Figure 8 showed that in the absence of HMDA, the presence of MAPI in the composite resulted in higher melt flow rate when the MAPI content was less than 4 wt. %. This is attributed to insignificant reaction between PET and MAPI during melt compounding. At 8 wt. % MAPI content (HMDA = 0), the melt flow rate became lower which is believed to be due to appreciable reaction between PET and MAPI. This result indicates that portion of the HMDA was able to effectively couple with MAPI and PET simultaneously to produce the desirable PET-co-HMDA-co-MAPI copolymers which tended to anchor along the interface.

Figure 8 also showed that at any MAPI content investigated, melt flow rate decreased with increasing HMDA content and approached zero for the composite NR/PET/MAPI/HMDA = 60/40/8/0.6. As expected, higher level of HMDA caused increased crosslinking of the NR/PET blend and resulted in increased viscosity which caused the decrease in melt flow rate. Similar observation on the variation of compatibilizer content with melt flow rate have been reported by Ju and Chang [21] and Soares et al [26] for other filled polymer systems.

The effect of filler loading and filler particle size on melt flow rate of the compatibilized composites is presented in Figure 9. It could be seen that melt flow rate decreased as the filler loading increased; but increased with increasing filler particle size. The observed trend variation in melt flow rate with increasing filler content is attributed to increase in interfacial friction due to hindrance in chain mobility of the matrix polymer under shear stress.

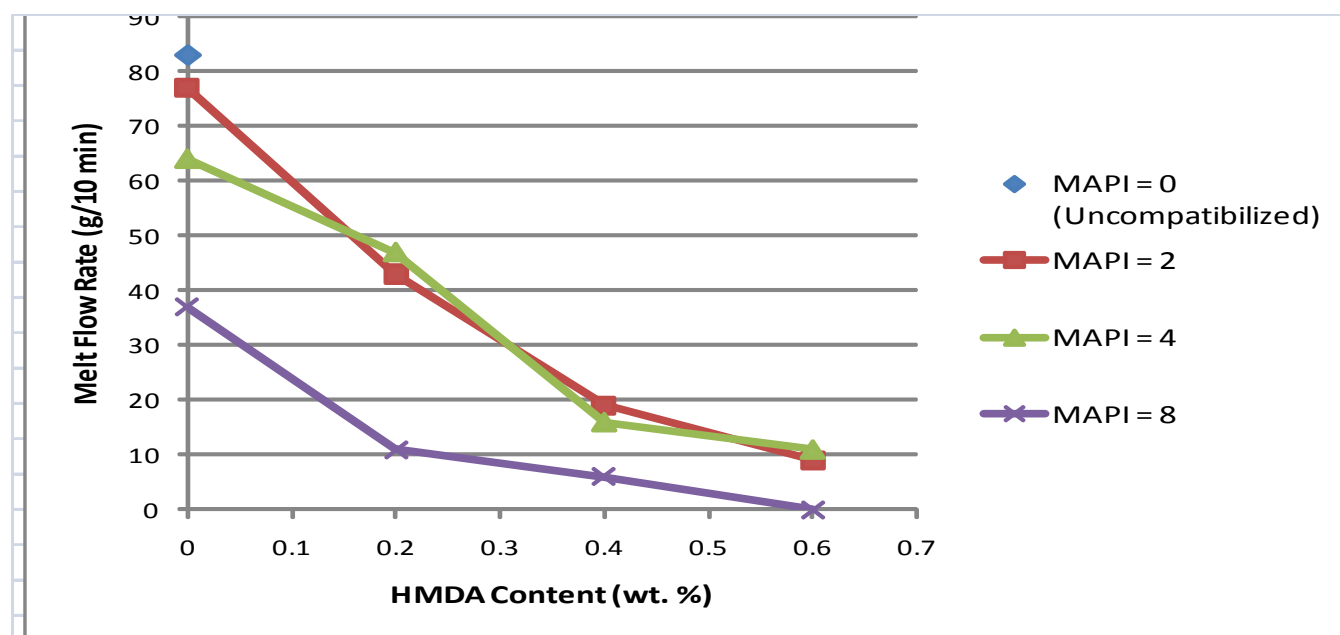


Figure 8. Plot of melt flow rate versus HMDA content for compatibilized and uncompatibilized composites at 5 wt.% DSP filler loading (120µm filler particle size) and NR/PET = 60/40 composition

3.1.6. Rockwell Hardness

The shore D hardness of the composites as a function of MAPI content is shown in Figure 10. Clearly, the shore D hardness of the uncompatibilized composites was lower than that of the compatibilized sample at the filler loading (5 wt. %) investigated. Figure 10 also showed a progressive increase in shore D hardness of the composites with increases in HMDA content. At any given MAPI content investigated, only a small quantity of HMDA (0.2 wt. %) in the composite could be able to increase the shore D hardness significantly. The improved interfacial adhesion

between the filler and matrix polymer, promoted by the addition of HMDA during melt compounding is the considered factor responsible for the increase in shore D hardness with increases in HMDA content.

Figure 10 also revealed that small amount of MAPI in the composite (2 wt. %) only resulted in insignificant increase in shore D hardness. Small amount of MAPI was found to limit the quantity of the desirable in-situ-formed copolymer during melt compounding, even with increasing HMDA content. This resulted to insignificant improvement in the filler-matrix interfacial adhesion and shore D hardness observed at low level of MAPI. This result indicated that a combination of MAPI and HMDA was required for effective reactive compatibilization of NR/PET/DSP bio-composites. The variation of shore D hardness of the composites with filler loading and filler particle size is presented in Figure 11. The figure showed that the hardness of all composites at any given particle size increased with increase in the filler loading. This result is an indication of the enhancement of stiffness of the composite with increasing filler content which increases the composite hardness.

Similar results in composite property with increasing filler loading have been reported in previous studies of filled polymer systems by Osabohien and Egboh [1], and Khalid et al. [31]. Also, Ewulonu and Igwe [11] who studied the properties of oil palm empty fruit bunch fibre filled high-density polyethylene reported that the addition of this filler increased the hardness of the high-density polyethylene which increased further with increasing filler content. Figure 11 also showed that at any given filler loading, the hardness of the composites decreased with increases in the filler particle size. The decreasing trend in the hardness of a composite polymer system had been reported by Yang et al. [32], Rana et al. [33], and Hattotuwa et al. [34].

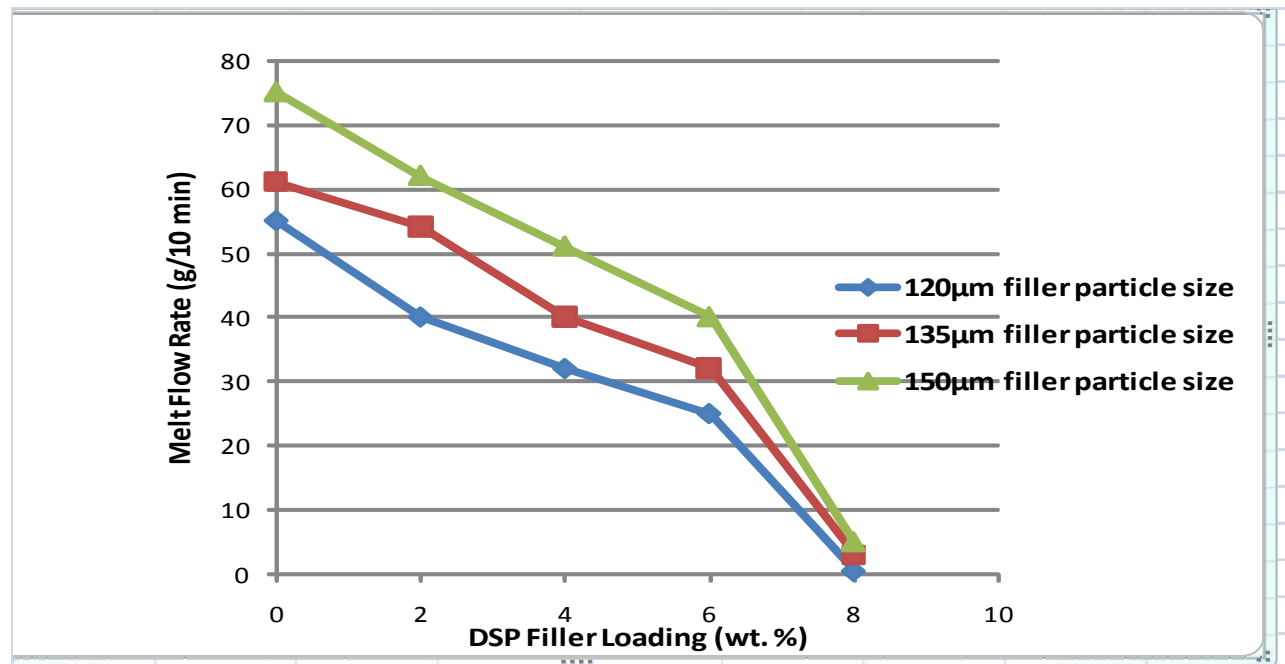


Figure 9. Plot of Melt Flow Rate versus DSP Filler Loading for Compatibilized Composites at fixed MAPI/HMDA = 8/0.6 and NR/PET = 60/40 wt. % Compositions

3.2. Water Uptake Properties

The water uptake indices of the composites as a function of HMDA content and DSP filler loading are illustrated in Figures 12 and 13, respectively. Figure 12 showed that the uncompatibilized composites absorbed more water than the compatibilized composites, showing that the amount of water absorbed by the composites was reduced by compatibilization. For the composite containing the smallest amount of MAPI, a further increase in HMDA did not cause appreciable reduction in water uptake. It is believed that small amount of MAPI in the composite (2 wt. %) limited the quantity of the desirable copolymers to be produced, even with increases in HMDA content. Figure 12 also showed that the amount of water absorbed decreased with increases in HMDA content and tended to level off (i.e, equilibrium water uptake) at 2 wt. % HMDA. The presence of HMDA promoted a decrease in the interfacial tension, an improvement in both the interfacial adhesion and crosslinking of the rubber particles which resulted to the observed decreases in water sorption with increases in HMDA content.

Figure 13 showed that at any given filler particle size considered, there was a progressive increase in water absorbed with increasing filler loading for the compatibilized composite. The hydrophilic nature of the DSP filler is considered to be responsible for the water absorbed by the composites which increased with increase in filler loading. Previous studies [11, 35] on other polymer composite systems have reported increases in water absorption with increasing filler loading. Again, the amount of water absorbed at any filler loading considered increased with increase in the filler particle size. The increase in the amount of water absorbed by the composite could be attributed to increases in the degree of polymer-filler interaction associated with smaller filler particle size.

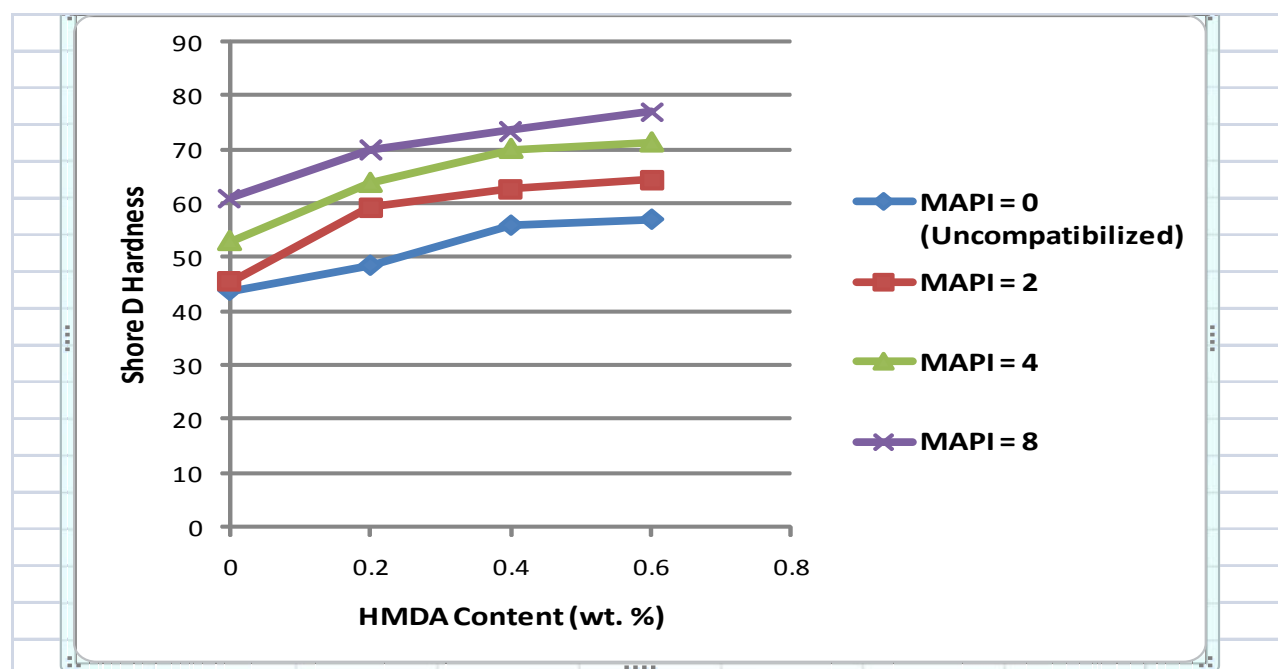


Figure 10. Plot of Shore D hardness versus HMDA content for compatibilized and uncompatibilized composites at 5 wt.% DSP filler loading (120µm filler particle size) and NR/PET = 60/40 composition

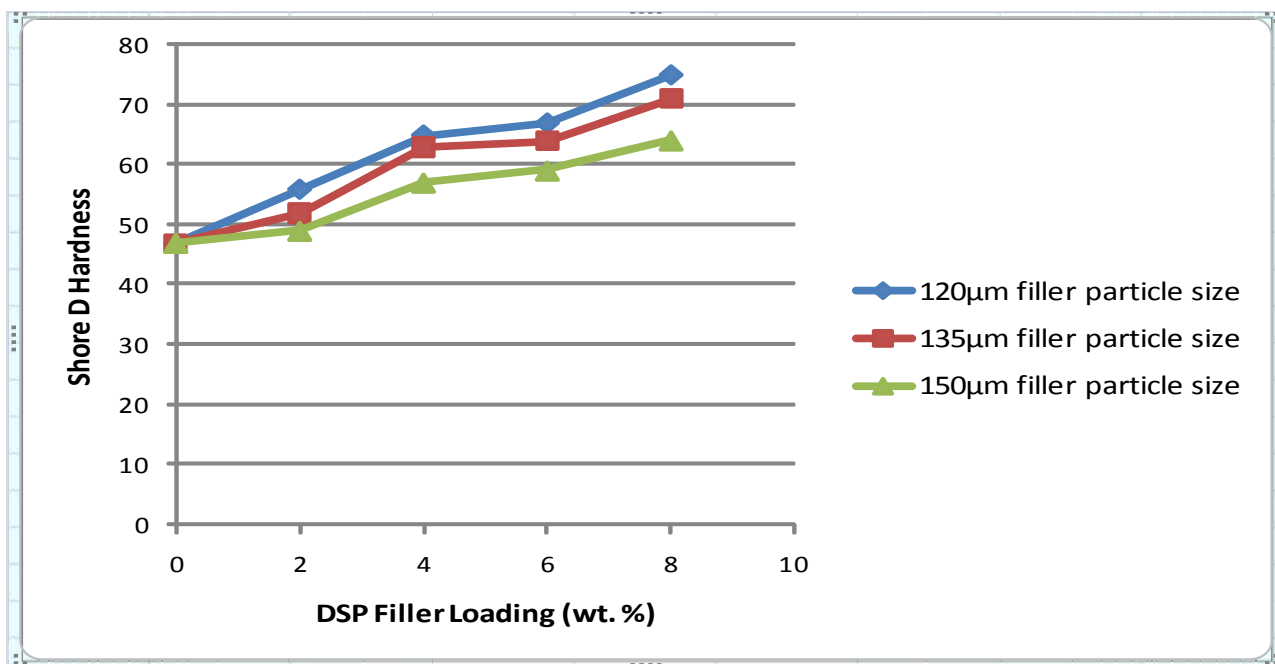


Figure 11. Plot of Shore D Hardness versus DSP Filler Loading for Compatibilized Composites at fixed MAPI/HMDA = 8/0.6 and NR/PET = 60/40 Compositions

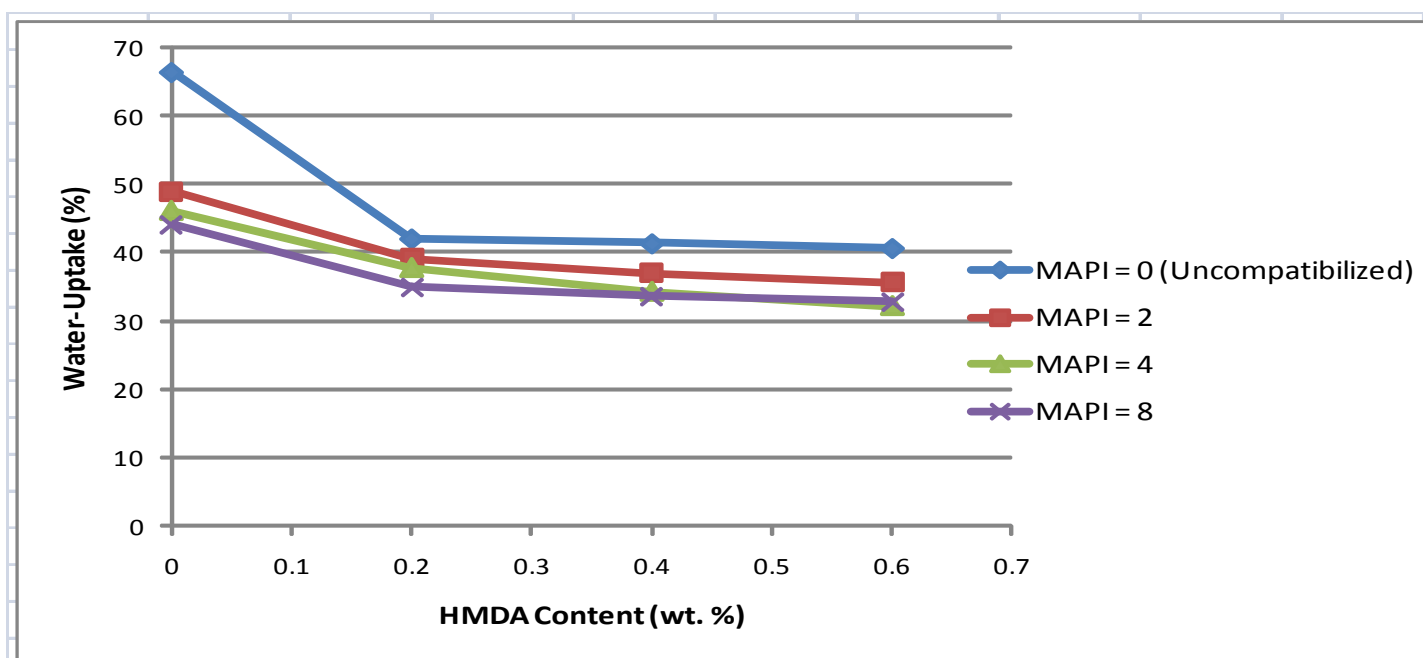


Figure 12. Plot of Water uptake versus HMDA content for compatibilized and uncompatibilized composites at 5 wt.% DSP filler loading (120µm filler particle size) and NR/PET = 60/40 composition

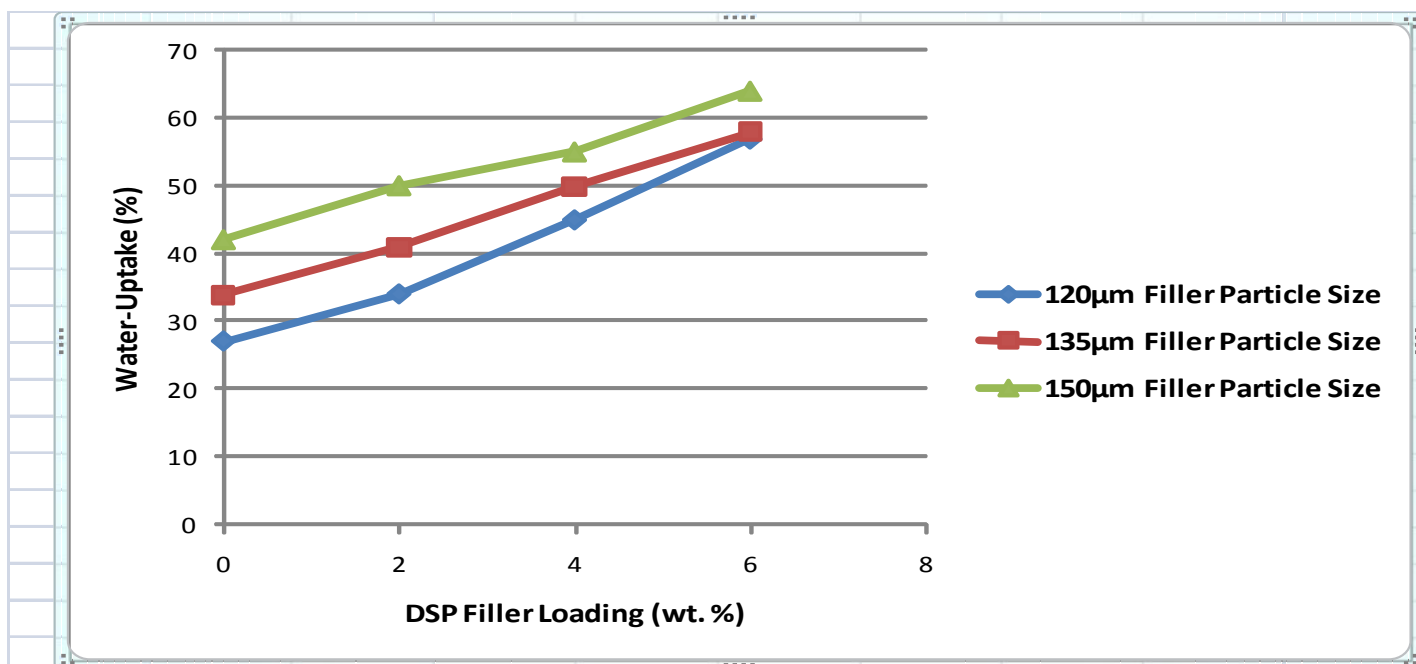


Figure 13. Plot of Water-Uptake versus DSP Filler Loading for Compatibilized Composites at fixed MAPI/HMDA = 8/0.6 and NR/PET = 60/40 wt. % Compositions

4. CONCLUSION

Compatibilized NR/PET/DNS powder biocomposites with better physico-mechanical and water uptake properties were prepared by using a combination of two compatibilizer systems. Due to the low reactivity between MAPI and PET in the absence of catalyst, the quantity of the in-situ-formed PET-co-MAPI copolymer is insignificant, and property improvement of the compatibilized composite is not substantial. Addition of a small amount of HMDA can act as an effective coupling agent to produce PET-co-HMDA-co-MAPI copolymers at the interface. This results to compatibilized composites with finer domain size of the dispersed phase and greater interfacial adhesion. Combination of MAPI and HMDA has been demonstrated to be an effective dual reactive compatibilizer for NR/PET/DNS powder biocomposites as shown by marked improvement in the properties investigated.

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COMPETING INTERESTS

The authors hereby declare that no competing interests exist.

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