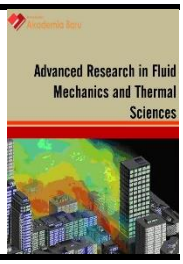
Penerbit Akademia Baru

Journal of Advanced Research in Fluid Mechanics and Thermal Sciences

Journal homepage: www.akademiabaru.com/arfmts.html
ISSN: 2289-7879



Thermal Aging Effect on the Mechanical and Morphological Properties of Polypropylene/Virgin Acrylonitrile Butadiene Rubber/Sago composites

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ARTICLE INFO

Article history:
Received 2 February 2019
Received in revised form 24 July 2019
Accepted 17 September 2019
Available online 30 November 2019

Keywords:
Thermal aging; sago starch; polypropylene; acrylonitrile butadiene rubber

ABSTRACT

This work investigates the effect of thermal aging on mechanical and morphological properties of polypropylene (PP)/ virgin acrylonitrile butadiene rubber (NBRv)/sago (Sago) composites. PP/NBR matrices at different weight fraction of 100/0, 70/30 and 40/60 with fix ratio of sago, were prepared by using a heated-two roll mill at 180 °C. For thermal aging test, dumbbell specimens were conditioned in an oven at 70 °C until 45 days. Mechanical and morphological properties of aged and unaged specimens were carried out by using Universal Testing Machine (UTM) and Scanning Electron Microscope (SEM), respectively. The results indicate that tensile strength and Young's modulus of aging specimens were optimized at day 15th. A better growing surface was observed after aging process showing an enhancement interfacial adhesion between the sago starch fibres and PP/NBRv matrices.

1. Introduction

Due to low density, environmentally friendly in nature, low in cost, non-toxic and recyclability, the development of polymer composites with natural fibres have gained too much interests [1-3]. The usage of natural fibres to replace synthetic fibre such as glass, have been applied in various applications mostly in automotive part, building construction and so on. Natural fibres likes kenaf [4,5], hemp, sisal, oil palm fibre, sugarcane baggase, banana skin [6,7], pineapple leaf, sago and many others have been incorporated into various polymer composites [8-10]. Sago is a starch extracted from the pith of tropical palm which known as *Metroxylon sagu.* In Malaysia, sago is abundantly grown in the state of Sarawak usually in swampy area. However, the over productions of sago have leads to river pollution since they were directly washed away to the nearby water body which might

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harm to the environment [11]. Thus, sago was implied as filler or reinforcement in numerous polymer composites [12-14].

Polypropylene (PP) is a kind of thermoplastic polymer with high heat resistance, high stiffness, good impact, recyclable and stretchable. Thermoplastic elastomers (TPE) prepared by melt mixing with rubbers like acrylonitrile butadiene rubber (NBR) are expected to have excellent oil resistant and high in strength. Previous studied on different types of TPEs materials are available in the literature [15-17]. Ismail *et al.,* [17] performed the blend of PP and NBR to investigate the tensile, thermal and morphological properties. PP/NBR blend has high in tensile strength and finer in morphology with better dispersion properties. In this work, sago was added into the PP/NBR matrices and act as filler in the TPE composites. The main objective in this study is to observe the thermal aging effect on mechanical and morphological properties of PP/NBRv/sago composites.

2. Methodology

Table 1 lists the coding and formulation of PP/NBRv/sago composites. As shown in the table, the sago content was fixed with 15 phr. PP (code 6331) with density 0.9 g/cm^3 was purchased from Titan PP Polymers Sdn Bhd. NBRv was purchased from Zarm Scientific & Supplies Sdn Bhd. Sago was supplied from Land Custody Development Authority (LCDA), Sarawak. Sago was grinded into fine powder with particle range from 150 µm to 300 µm. Then, sago was dried at 60 °C for 2 hours to remove excessive moisture content and prevented from agglomeration.

Table 1
Coding and formulation of PP/NBRv/sago composites

Sample code	Materials Ratio (phr)					
PP	100	80	70	60	50	40
NBRv	0	20	30	40	50	60
Sago	15	15	15	15	15	15

*phr: part per hundred resin

A heated two-roll mill machine (model DW-5110) from Fang Yuan Instrument (DG) Co. Ltd, China, was used to melt mixed the materials. Firstly, PP was discharged and melts for 4 minutes, followed by NBRv and mixed for another 3 minutes. Sago was then added and continued mixing until 10 minutes. Then, compounded samples were pressed using hot press machine (Go Tech model GT 7014) to get 1 mm thin molded sheet. Dumbell shape of sample was produced by cutting the molded sheet using Wallace Die Cutter S6/1/6.A.

To study the effect of thermal aging, dumbbell samples were conditioned in an oven at 70 °C for 45 days. PP/NBRv/sago with formulations of 100/0/15, 70/30/15 and 40/60/15 were selected to perform tensile test. Tensile test was done after 3, 5, 10, 15, 30 and 45 days with five sample repetitions.

Tensile strength, Young's modulus and elongation at break was examined using Universal Testing Machine (UTM) Instron (Model 3366) machine, followed the ASTM 638. The jaw separation distance was adjusted to 50 mm with speed of 5mm/min. The morphological fracture tensile surface of specimens was observed using Scanning Electron Microscope (SEM) machine. Prior testing, all the samples were coated with platinum using Auto Fine Coater series JFC 1600 in order to avoid electrostatic discharge.

3. Results

3.1 Tensile Strength

Figure 1 shows the tensile strength of 100/0/15, 70/30/15 and 40/60/15 of PP/NBRv/sago composites after thermal aging. From the data obtained, tensile strength was slightly increased until reached maximum at day 15. The optimum tensile strength for 100/0/15, 70/30/15 and 40/60/15 of PP/NBRv/sago composites are 29.402MPa, 13.854MPa and 4.452MPa, respectively. There are two phases involves in this aging process. In phase 1, the tensile strength continuously increased from day 0 to day 15 and then slightly decreased until day 45. According to [18], the increasing of tensile strength at the initial phase was due to the crosslinking of the materials which corresponds to the positions of chain segments are stable and decrease in the movement of network chains. High tensile strength was observed as the external stress can disperse and transfer accordingly. Whereas, in phase 2, after 15 days, tensile strength was decreased due to exceed certain critical value of the crosslinks density and the average molar mass of the NBR chain between two crosslinks points decreased which cause the limitation of effective mobility of the chain segment. As the consequence, it caused to disturb the normal orientation of the network chains and turned out to stress concentration. This situation will reduce the number of effective network chains and led to reduce of tensile strength value.

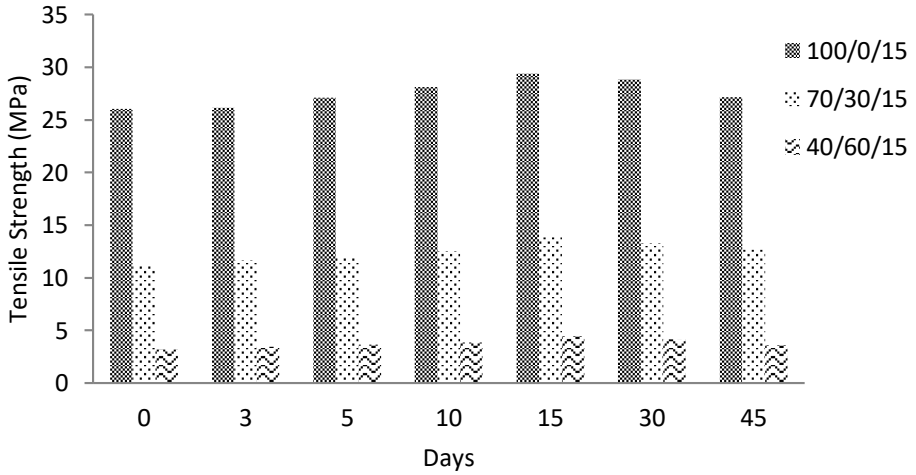


Fig. 1. Tensile strength of PP/NBRv/sago composites after thermal aging

Figure 2 shows the Young's modulus of 100/0/15, 70/30/15 and 40/60/15 of PP/NBRv/sago composites after thermal aging. It was gradually increased from day 0 until day 15. The optimum value gained for 100/0/15, 70/30/15 and 40/60/15 of PP/NBRv/sago composites are 1359.5MPa, 366.4MPa and 102.34 MPa, respectively. After thermal aging, the performance of mechanical properties for the composite with formulation 100/0/15 is 25.96% better than unaged composite up to day 15. For the formulation of 70/30/15 and 40/60/15, the mechanical results perform better than unaged composite with 48.4% and 27.61%, respectively. This is because of the loss of volatile matter after thermal aging such as water molecule that might contain in the filler. Without aging, the present of volatile are able to reduce the plasticity of material in the composites [19]. If the thermal aging is continued for the next days, the behaviour resulting in opposite manners. The increase of the Young's modulus can be briefly explained by oxidization reaction in polymer that causes molecular recombination. Thermal aging helps the formation of free radicals and consequently will increase the degree of network reticulation of the composites. Besides, crosslinks might cause the modulus to increase when expose to thermal. This research indicates that polypropylene (PP) and its blend, NBRv

become stiff and lead to enhance the tensile strength after thermal aging until it reach the optimum condition. As the amount of PP increase, the stiffness of the material becomes high. Thus, it leads to the high value of Young's modulus [20].

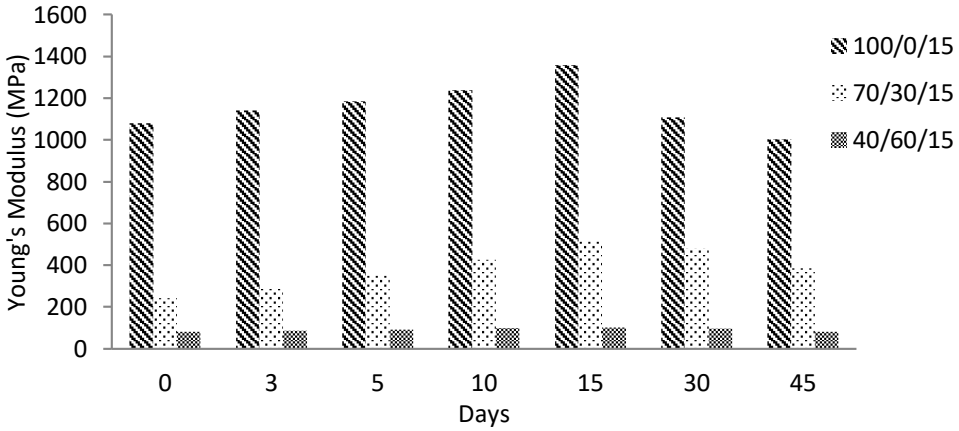


Fig. 2. Young's modulus of PP/NBRv/sago composites after thermal aging

Figure 3 shows the elongation at break of 100/0/15, 70/30/15 and 40/60/15 of PP/NBRv/sago composites after thermal aging. It shows uniform decreasing of elongation at break as the thermal aging is longer. Elongation at break becomes low after thermal aging due to high elasticity of composites. The result shows that the percentage of elongation at break decreased as the period for thermal aging is increase. The lowest elongation at break occurred at day 15 where tensile strength and Young's modulus are found to be optimum at this point. The percentage of the elongation at break for aged sample has reduce a lot compared to unaged sample with 53.49%, 49.06% and 24.17% at day 15 thermal aging for formulation of 100/0/15, 70/30/15 and 40/60/15, respectively. The removal of moisture during thermal aging also reduce the stress elongation until it meet the breaking points since removal of volatile matter may improve the elasticity of the composites. As a conclusion, percentage of elongation at break might produce the result that oppose the tensile strength and Young's modulus since good composites will performed with lowest stress of elongation from its original length.

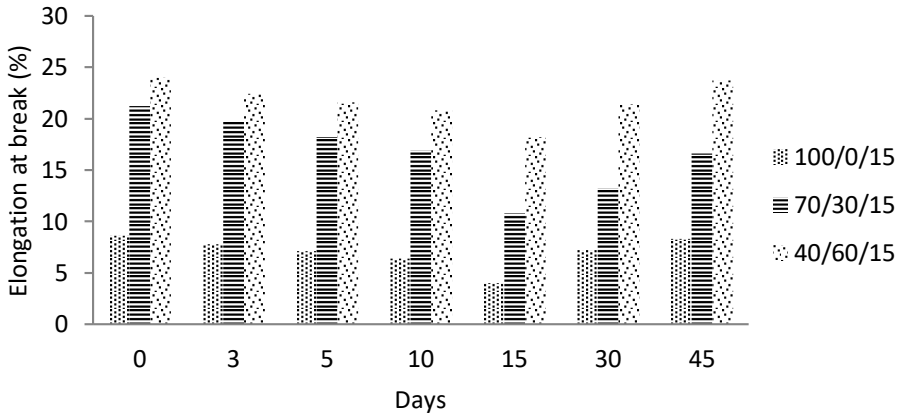


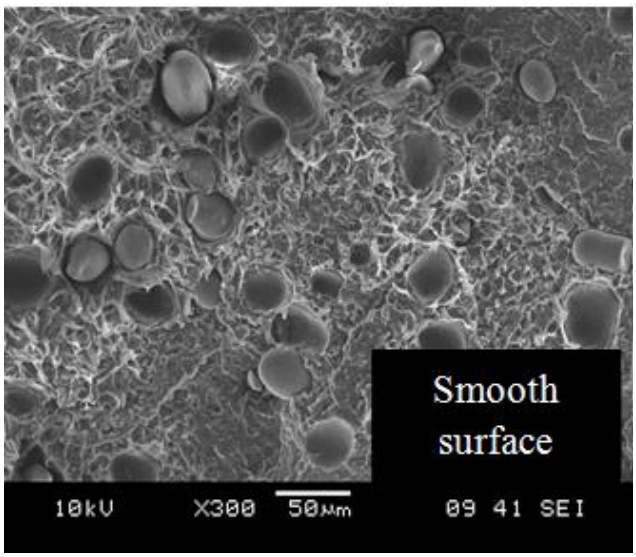
Fig. 3. Elongation at break of PP/NBRv/sago composites after thermal aging

3.1.1 Morphological properties

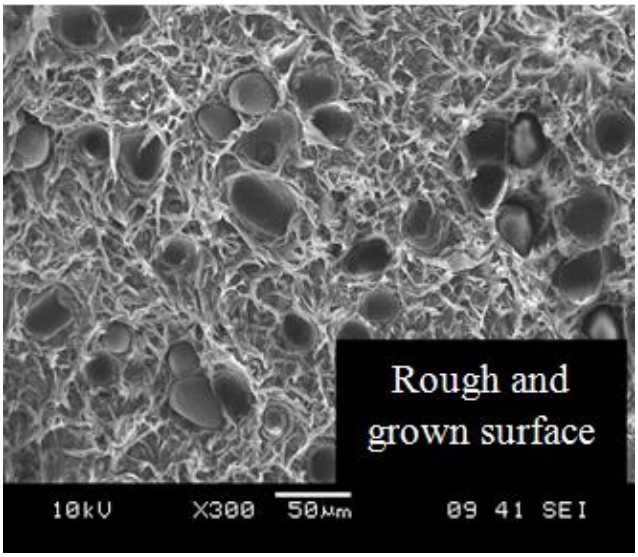
Figure 4 shows the micrographs of tensile fracture surface of aged and unaged PP/NBRr/sago composites with different formulation. Figure 4(a) shows the unaged composite of 100/0/15 of PP/NBRv/sago. The attachment sites of sago filler are clearly shown with smooth PP matrix which does not really expand. Figure 4(b) observed rough and grown surface after thermal aging for 15 days as per the mechanical strength are performed at maximum level. The findings convinced that the samples were having modification or enhancement after exposed to heat at significant period. That rough structure indicates that the sample change the properties from ductile to brittle which cause the material better in strength.

Figure 4(c) describes that unaged 70/30/15 of PP/NBRv/sago composite, show rough surface and formation of sinusoidal folding. Apparently, the fractured surface of unaged sample was quite ductile compared to the sample with thermal aging. The sinusoidal folding indicates high plastic deformation that led to fracture. The stated features explain the characteristic of ductile failure [19]. The aged composite of 70/30/15 specimen is shown in Figure 4(d). The fracture surfaces of samples show very surprisingly result of fractured topography. The area shows tufted or growing surface and rough texture formation. In the other hand, the formation of sinusoidal folding are no longer appear in this aged sample. Thermal aging had proved that the fracture meant the changing of properties from ductile to brittle one. This finding proved the mechanical testing that aged composites are performed better compared to unaged composites.

Figure 4(e) shows micrograph of unaged composite with formulation of 40/60/15. It barely shows more sinusoidal folding effect on NBRv matrix. Otherwise, Figure 4(f), shows the 40/60/15 aged composites. A rough surface was observed due to fracture of PP and rear line effect from the tensile test. However, these features enhance the interface strength of matrix. It will directly improve the effectiveness of the load transfer from matrix to filler. Thus, this result indicates that as the composites strength is initially increased, begin from day 0 up to 3, 5 and 10 days until it reach the maximum at day 15, the stress displacement response is simply shifted up to the greater level of strength. However, as the thermal aging is further, the stress displacement response at higher stress level but diminish very fast with increasing the displacement strength. This will lead to reduction of the strength since the percentage of stress elongation is increase. This reduction in fracture energy contributed to the transition in the fracture mechanism such as matrix cracking and filler scissoring [21].



(a)



(b)

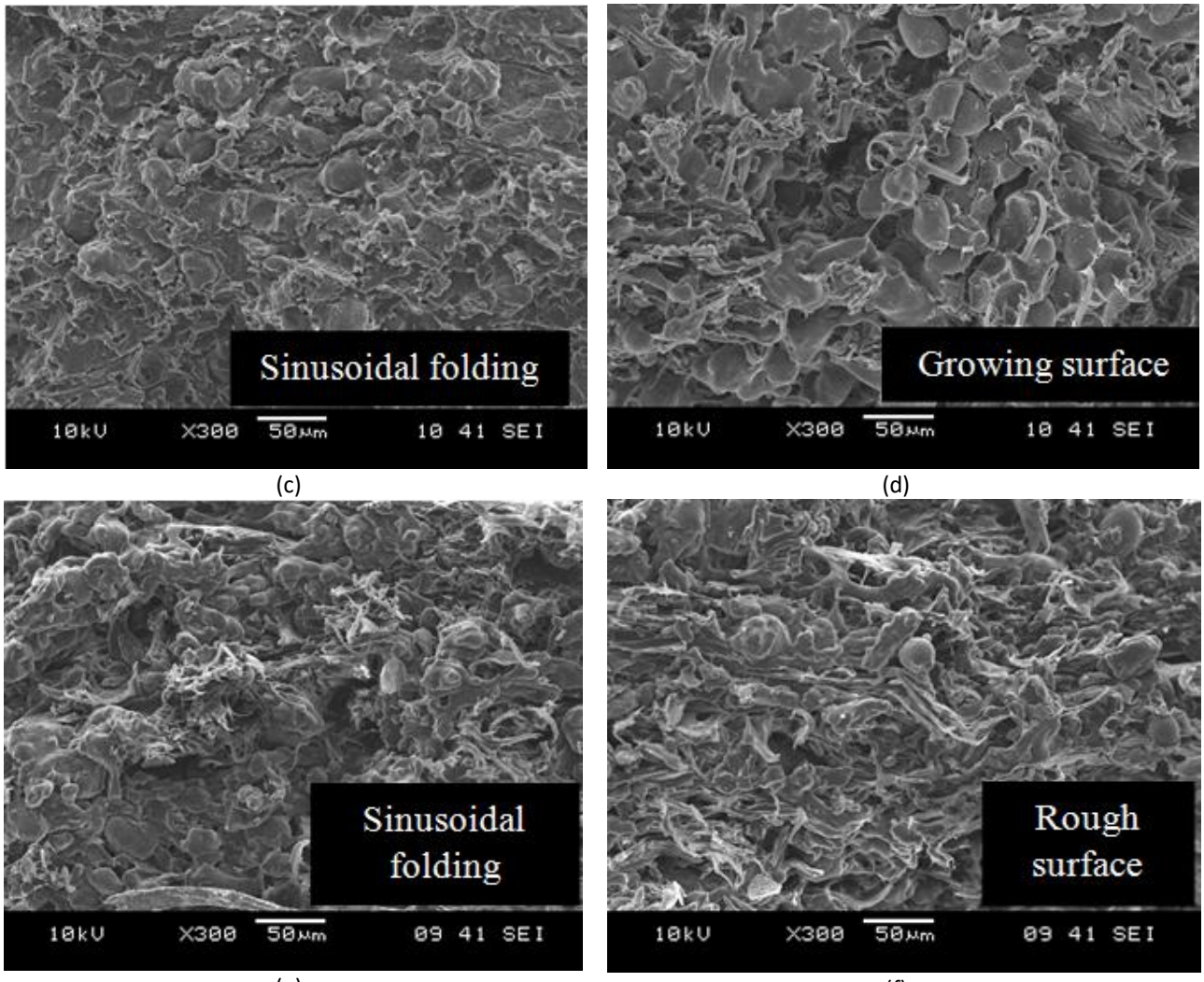


Fig. 4. Morphological fracture tensile surface of; (a) 100/0/15 unaged; (b) 100/0/15 aged; (c) 70/30/15 unaged; (d) 70/30/15 aged; (e) 40/60/15 unaged; (f) 40/60/15 aged at magnification X300

4. Conclusions

The mechanical properties and morphological structure of the composite had performed better and modified very well after thermal aging. Tensile strength and Young's modulus show highest values at day 15 of thermal aging. From the morphological study, the aged composites at day 15 show a quite rough, tufted and grown surface. They were resembling the modification of the composite from ductile to brittle which cause the materials better in strength.

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