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The Influence of Starch Gelatinization on Mechanical Properties of Natural Rubber Latex Films

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Abstract. The addition of starch in natural rubber (NR) latex films tends to decrease the mechanical properties of NR latex films due to poor dispersion of the fillers. As an alternative, starch was gelatinized before being added into NR latex compounds. The dispersion of 10% starch was mechanically prepared through ball milling together with dispersing agent. The other dispersion of 10% starch was gelatinized using alkali. The influenced of ball milled starch and alkali gelatinized starch filled NR latex films were studied through scanning electron microscopy, tensile properties, tear strength, and swelling properties. The rough freeze fracture surface with less agglomeration of fillers was observed on the alkali gelatinized starch NR latex films. The NR latex films filled with alkali gelatinized starch showed improvement in tensile properties, tear strength, and swelling resistance compared to the ball milled starch NR latex films. It can be concluded that the alkali gelatinization improved the dispersion of starch in NR matrix thus enhanced the mechanical properties of NR latex films.

Introduction

Starch is a naturally renewable polysaccharide which is readily available, low cost, and able to biodegrade. However, starch also has disadvantages; large particle size, poor mechanical properties, poor dimensional stability, and hydrophilic in nature [1-3]. The addition of starch tends to reduce the mechanical properties of natural rubber latex films due to the hydrophilic nature of starch, attributed to abundant hydroxyl groups, decreases the interface compatibility between hydrophilic starch and hydrophobic rubber matrix [4].

Generally, filler use in NRL will be prepared in liquid medium during mixing process to improve their dispersion and homogeneity. Ball milling is a mechanical method to reduce the crystalline region and to modify the physicochemical properties of starch using friction, shear, impingement, collision or other mechanical actions [5]. This method generally related to the mechanical and physical process in preparation of dispersion. The other alternative is to gelatinize the starch with water and heat before being mixed into the NR latex compounds. However, this method is not recommended due to the retrogradation of starch after cooled at room temperature [6].

The starch gelatinization using alkali is a promising method to improve the dispersion of starch in matrix thus enhancing the mechanical properties of NR latex films. Alkali generally used as a stabilizer in the latex compounds, and it is able to gelatinize the starch under room temperature. According to Karim et al. [7], alkali treatment for starch gelatinization could prevent the retrogradation to occur. The aims of this study were to investigate the influence of alkali gelatinized starch on the starch dispersion in NR matrix as well as their effects on the mechanical properties of NR latex films.

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Materials and Method

NR latex used was high ammonia (HA) latex with 60% of dry rubber content (DRC) from Zarm Scientific and Supplies (Malaysia) Sdn. Bhd. Starch used was manufactured by Sago Link Sdn. Bhd. Other additives which are zinc oxide, antioxidant, zinc diethyldithiocarbamate (ZDEC), sulphur, pottasium hydroxide (KOH), and anchoid were supplied by Bayer (Malaysia) Sdn. Bhd in dispersion form and used as received.

The 10% ball milled starch was prepared by mixing the starch with anchoid, potassium hydroxide and distilled water before transferred into the ball mill jar and mixed at 25 hertz for 24 hours. Meanwhile, the 10% alkali gelatinized starch was obtained by starch (10g, dry basis) was mixed with 5.6% of diluted KOH was added in the mixture and stirred for 30 minutes under room temperature. Due to the low temperature, it takes about two days for alkali treated starch to fully gelatinize. The unfilled- and the starch filled- NR latex films were prepared in accordance to formulation in Table 1. The mixtures were constantly stirred for 2 hours under room temperature and was then transferred into water bath with temperature of 70°C and constantly stirred. The compound was sealed and left for 16 hours under room temperature for maturation process. The NR films were prepared through dipping process and cured in the air oven for 15 minutes at 100°C before being cooled under room temperature for 24 hours. For morphological analysis, the samples were cut into 2cm x 2cm and coated with gold for one hour and observed under scanning electron microscopy (SEM) with magnification at 300. Instron IX3366 machine was used to study the tensile properties (ASTM D412) and tear strength (ASTM D624) with the crosshead speed of 500mm/min. The swelling test was carried out according to ASTM 471.

Table 1: Formulation for unfilled- (control) and starch filled- NR latex.

Ingredients	Part per hundred rubber (phr)		
	Control	Ball milled starch	Alkali gelatinized starch
60% HA Latex	100	100	100
10% KOH	0.3	0.3	-
50% ZnO	0.25	0.25	0.25
50% Antioxidant	0.5	0.5	0.5
50% ZDEC	0.75	0.75	0.75
50% Sulfur	0.5	0.5	0.5
10% Starch *	-	10 w/w%	10 w/w%

*Measured using weight percentage, starch (w)/ latex (w) (%)

Results and Discussion

From Fig. 1 (a), control films showed smooth freeze fracture surface with a few tearing line. The voids were observed on the freeze fracture surface of NR latex films with the addition of 10% ball milled starch (Fig. 1 (b)) due to poor interaction between hydrophobic NR matrix and hydrophilic starch. The increased in number of tearing lines were observed on the freeze fracture surface of NR latex films filled with 10% alkali gelatinized starch (Fig. 1 (c)). The roughness of fracture surface with many tearing lines indicated that higher energy required to cause failure which also means strong interfacial adhesion between both phases [8].

Tensile modulus is related to the stiffness of a material during the elastic stage of a tensile test [9]. Fig. 2 shows the tensile modulus values of 10% ball milled starch filled NR latex films were decreased, while, the addition of 10% alkali gelatinized starch filled NR latex films retained the stiffness of the films which showed a comparable tensile modulus value compared to that on the control films. From Fig. 3, it can be observed that the addition of 10% alkali gelatinized starch showed a slight decreased in elongation at break percentages of NR latex films.

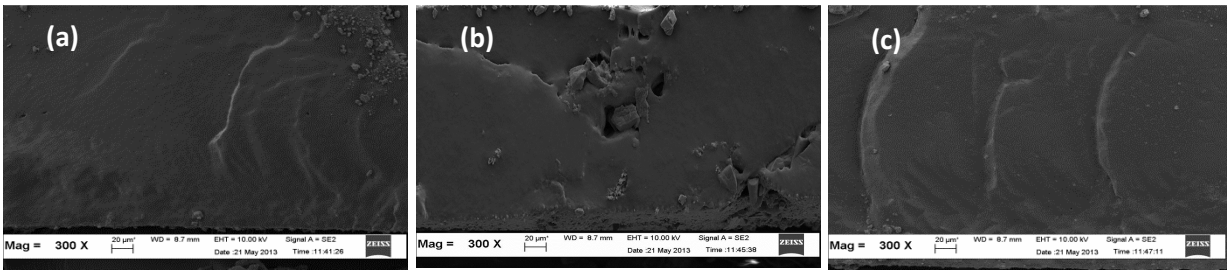

Fig. 1: Freeze fracture surfaces of (a) control, (b) 10% ball milled starch-, and (c) 10% alkali gelatinized starch- filled NR latex composites films (mag: x300).

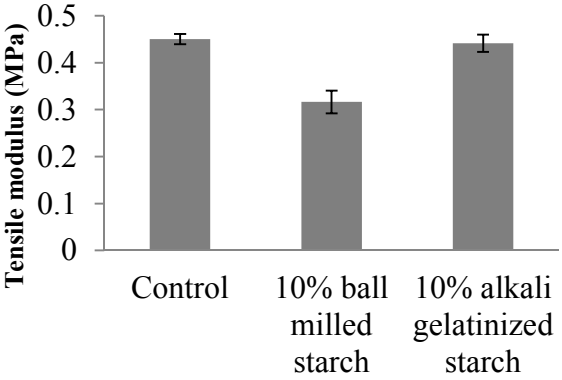

Fig. 2: Tensile modulus values of control, 10% ball milled starch-, and 10% alkali gelatinized starch- filled NR latex composites films.

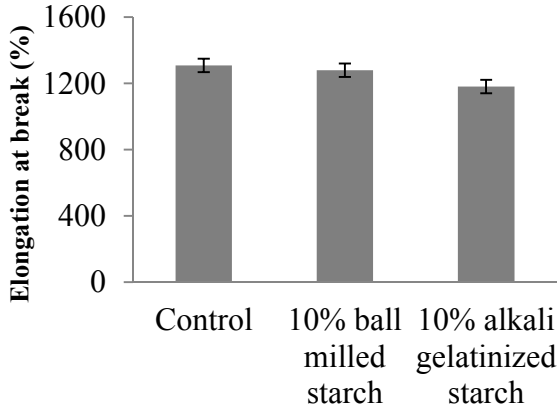

Fig. 3: Elongation at break of control, 10% ball milled starch-, and 10% alkali gelatinized starch- filled NR latex composites films.

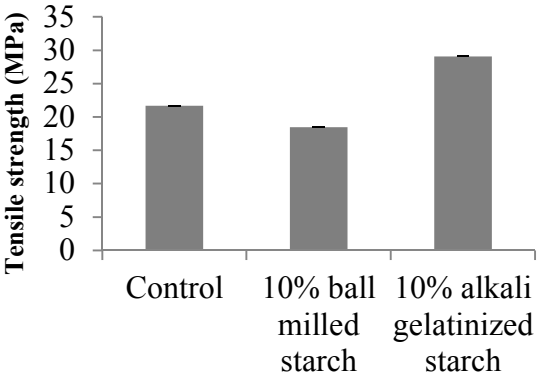

Fig. 4: Tensile strength of control, 10% ball milled starch-, and 10% alkali gelatinized starch- filled NR latex composites films.

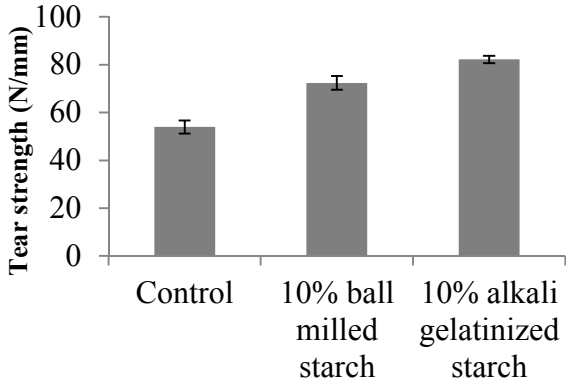

Fig. 5: Tear strength of control, 10% ball milled starch-, and 10% alkali gelatinized starch- filled NR latex composites films.

Fig. 4 shows the incorporation of 10% ball milled starch decreased the tensile strength of natural rubber latex films (18.44 MPa) due to the incompatibility of hydrophobic NRL and hydrophilic starch system [10]. The utilization of 10% alkali gelatinized starch increased the tensile strength up to 29.07 MPa of natural rubber latex films compared to control films (21.68 MPa). The homogenous gelatinized substrates offer more advantages over the heterogeneous of starch in granular form [11]. Wu et al. (2004) [12] suggested in an aqueous system, the rubber latex particles, which are generally smaller than 100 nm, can be mixed with gelatinized starch uniformly, giving starch/rubber composites with a fine dispersion. As shown in Fig. 5, the addition of starch

especially for 10% alkali gelatinized starch has increased the tear strength of NR latex films up to 82.2 N/mm. When the latex was mixed with filler, the increased in contact between rubber particles and filler will causing the formation of physical tangles. The ability to inhibit the movement of macromolecules of NR increased, resulting in good reinforcing effect [13].

Fig. 6 shows the swelling index percentages of 10% ball milled starch filled NR latex films were increased (519.3%) compared to the control films (465.8%) due to the poor rubber –filler interactions. The slight reduction of swelling upon 10% alkali gelatinized starch addition (420.9%) indicated the penetration of toluene was restricted in NR latex film. A good surface contact between rubber and filler inhibited the swelling of the polymeric chains located in the interfacial zone [14].

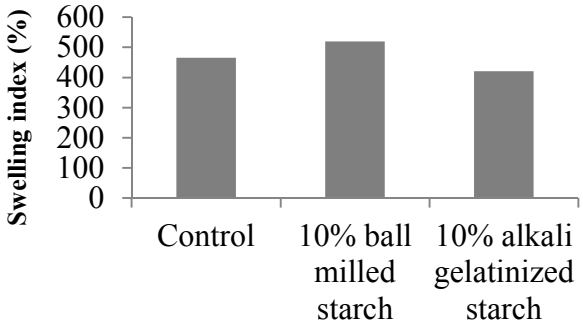

Fig. 6: Swelling index percentages of control, 10% ball milled starch-, and 10% alkali gelatinized starch- filled NR latex composites films.

Conclusions

The improvement of starch dispersion in NR matrix which obtained from the utilization of 10% alkali gelatinized starch as observed via SEM were reported to improved the tensile properties, tear strength, and swelling resistance of NR latex films. This indicated that the alkali gelatinization is an inexpensive and reliable alternative to improve the mixing of starch in NR matrix thus increased the mechanical properties of NR latex films.

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