

## NOVEL METHOD OF BREAKING DOWN RUBBER MOLECULES USING PEPTIZER AND THE PROPERTIES OF RESULTANT NATURAL RUBBER SOLVENT-BASED ADHESIVE

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### Abstract

*The huge cost of energy associated with the breaking down of high molecular weight rubber during adhesive production has placed a demand for alternative processing technology. In this study, the use of benzoyl peroxide as a peptizer to lower the molecular weight of natural rubber has been reported. The properties of natural rubber-based solvent adhesive produced by mechanical mastication and peptizer were also compared. Natural rubber (NR) was masticated in a two-roll mill at 80°C for 30 mins and was processed into adhesive using xylene as solvent. Similarly, adhesives were formulated by chemical break down of the heavy natural rubber molecules using benzoyl peroxide as peptizer. The peptizer loading was varied from 0.2 – 1.0 parts per hundred rubber (phr) of rubber at 100°C in a water bath while other compounding additives were kept constants for all samples under study. The adhesives produced were subjected to the following test: viscosity, lap shear test, T-peel test and drying time test. Other properties studied are weathering resistance, heat resistance and effect of moisture on the adhesives bond on different substrates. The result shows that the viscosity of the adhesives increases with increasing peptizer loading and the masticated sample has a viscosity slightly lower than that of 0.8phr and 1.0phr peptizer loading. Furthermore, T-peel test shows that 0.4 phr peptizer loading has a better average T-peel strength for the bonds while the masticated sample had the least average T-peel strength when compared with all samples. The sample with 1.0phr peptizer loading had the least decrease in shear strength while the 0.2 peptizer loading had the maximum decrease in shear strength. Finally, the FT-IR spectra shows similarities in spectra but with difference in the intensity of the peaks, indicating the ability of the benzoyl peroxide to effectively oxidize the natural rubber as compared with the mechanical masticated sample. The peak of 1080 cm<sup>-1</sup> in the peptized sample indicates a major difference with the spectra of mechanical masticated sample. Generally, the benzoyl peroxide effectively lowered the molecular weight of the natural rubber which is evident in the viscosity, reduced process time and ease of the adhesive formulation.*

**Keywords:** solvent-based adhesive; natural rubber; mastication; peptizer; adhesive properties.

### Introduction

The significance of adhesives or sealants in our daily life cannot be overemphasized, being a class of material used for the bonding of materials physically, dissolution of plastics similar to the case of welding two materials after which materials get hardened later, soldering of electronic devices (that is, a conductive adhesive) [1–3], and a lot more. This material is sometimes referred

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to as cementing or binding agents, glue or gum [4] which often find applications in the packaging processes, product assembly, woodwork, transportation or automobiles, footwear, building constructions, electrical systems [5–7], medical and dental applications [4], [8], and many others. A further survey of reports in the literature indicates that the demand for adhesives is continually rising across various continents from North America to Africa following the report of the Statista Research Department (in Fig. 1) [9]. Fernández [10] further indicated that the demand for the adhesive would continue to rise irrespective of the pandemic, as shown in Fig. 2 graphically.

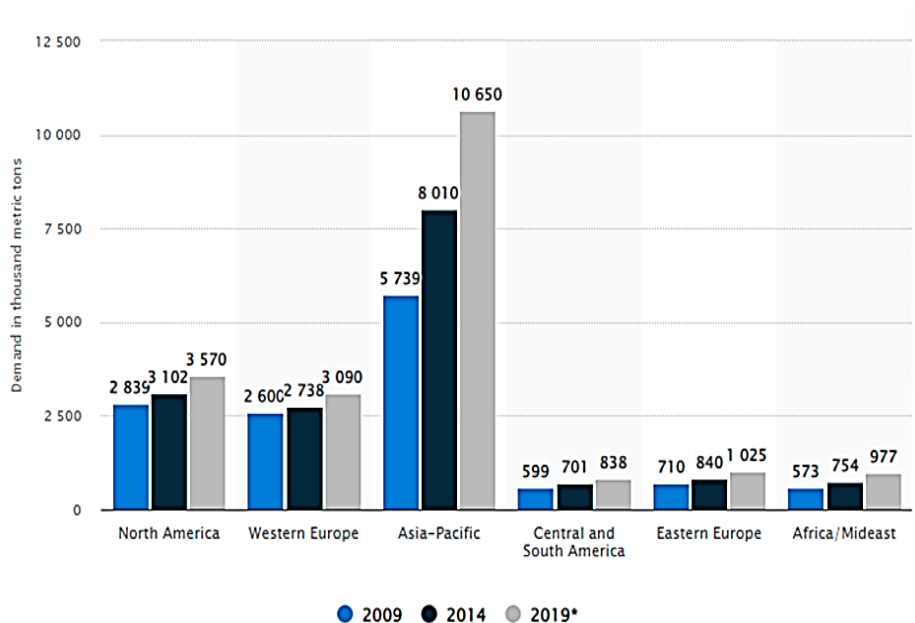


Fig. 1. Adhesive market demand across continents in 2009-2019 [9]

Natural rubber is one of the major production feed stocks among several rubber types used in adhesives production. The volume of natural rubber production across the globe cannot be overemphasized due to its wide range of applications. This application includes medical gloves, baby bottle nipples, diaphragms, membranes, balloons, pacifiers, gas tubes, toys, insulators, tires, blood pressure cuff coils [11, 12], and many others where sealants, otherwise known as adhesives, is one of the essential materials produced from the natural rubber. A recent natural rubber market trend survey indicates that worldwide production has risen by 2.1% to about 13 million tonnes as of November 2021 [13].

As a way of utilizing the existing advantage of the rising production of natural rubber [13] to meet the rising demand of adhesives across the globe [9], [10], several research works have been exploring the potential of developing a novel approach for obtaining the desired adhesive with a target property and area of application across different sectors. Some of the studies include Hermiati *et al.* [14] that employed the use of central composite design to determine the optimum condition of adhesive that would yield the optimum shear strength (8.8 MPa) of defined wood to be laminated using the prepared adhesive, where 16.4% crosslinker, glue spread rate of 298 g/m<sup>2</sup> for pressing time of 6 hrs. Another is the Boonrasri [15] report, which explores the potential of improving the quality of hot-melt adhesives obtained from natural rubber. The author explored the impact of different components of the composite from the resulting best hot-melt adhesive with a shear strength of 82.49±1.4 lbf/in<sup>2</sup> and cleavage peel strength of 2.06±0.02 kN/m, which were obtained at 100 phr of natural rubber, 25 phr of wax, 150 phr of ethylene vinyl acetate, and

150 phr of resin. Vaithanomsat et al. [16] studies further established the possibility of producing an eco-friendly adhesive via the interaction of furfural (from sugarcane bagasse) to natural latex, which was reported to have yielded good properties with no need for any risky solvent.

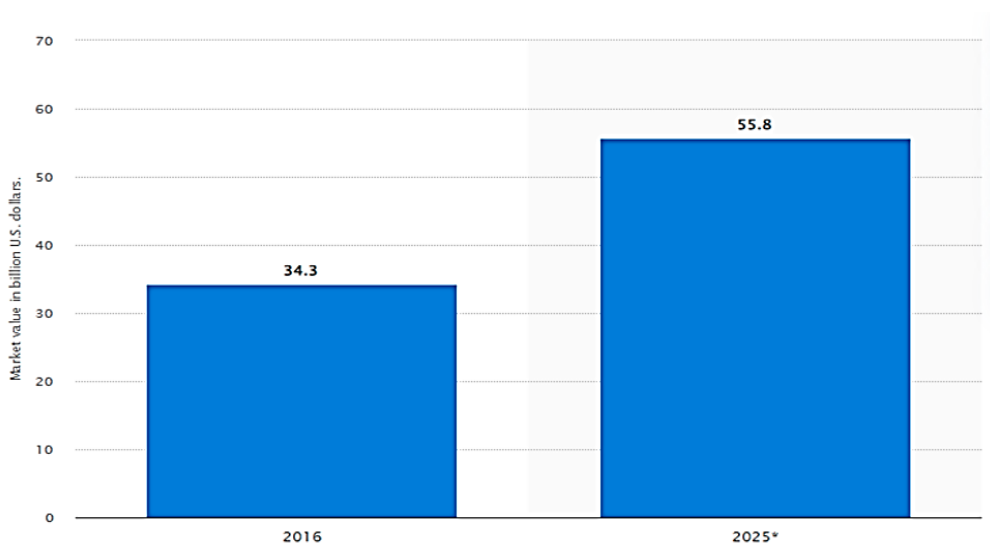


Fig. 2. Adhesive market value in 2016 and 2025 (projected) worldwide [10]

Other research looked into the blending of natural rubber latex with lignin in the production of adhesive, producing adhesives from cassava starch [17], the effect of utilizing bio-based adhesives on wood joint's tensile strength [18] and many other issues. A literature report survey further indicated that the research and development units of the industries are not left out in searching for the way forward. Among the industries' efforts, include the recent Rubber News [19] that revealed the effort of MICHELIN in investing in adhesive production through its new goal of aiming 80% sustainable adhesive that would be used in 2025.

To further the goal of meeting this rising demand of adhesives globally through value-addition to natural rubber, which is available globally across the different continent, this study, therefore, attempts to showcase a novel alternative to breaking the molecular weight of rubber by the use of benzoyl peroxide (that is, known to be cheaply available) as a peptizer for further processing into adhesive. In this study, benzoyl peroxide was not used as a pre-mastication additive but as a sole masticating agent to circumvent the conventional energy-demanding operation in the rolling mill. This study accounted for the potential of benzoyl peroxide as a peptizer to effectively lower the viscosity of the natural rubber for homogenous processing into adhesive.

## Materials and Methods

The crumb rubber was obtained from the rubber research Institute Edo State, Nigeria. The solvent, xylene is an analytical grade; a product of Exxon Mobil Chemical with 98% purity was used as received. Other reagents are stearic acid manufactured by Acme-Hardesty with 98% purity, peptizer (benzoyl peroxide), a product of King Ind. Ltd with 97.5% purity. Zinc oxide, sulphur, TMQ and rosin powder with 98% purity manufactured by Manskhilal and Company

and calcium carbonate with 85% purity a product of Charlotte, NC. All these chemicals were used as supplied.

### Mechanical Mastication of Natural rubber

The two-roll mill machine model 5183 manufactured by North Bergen, New Jersey, USA was used to masticate the natural rubber samples. The roll temperature was set to 80°C. Natural rubber was masticated for twenty minutes to form a band on the roll. The masticated natural rubber was then sheeted out of the mill.

### Preparation of adhesive from mechanical masticated rubber

The adhesives were prepared using the formulation in Table 1. The masticated rubber and 400 ml of xylene were introduced into a conical flask and stirred until a solution of natural rubber was formed, followed by sequential addition of other additives with continuous stirring until a homogeneous mixture of the natural rubber base adhesive was obtained and was labelled as Sample A. The entire time for the adhesive production by mechanical mastication was thirty minutes.

**Table 1:** Formulation of the adhesive

Materials	Sample A phr	Sample B phr	Sample C phr	Sample D phr	Sample E phr	Sample F phr
NR	100	100	100	100	100	100
Calcium Carbonate	32	32	32	32	32	32
Zinc Oxide	4.0	4.0	4.0	4.0	4.0	4.0
TMQ	1.2	1.2	1.2	1.2	1.2	1.2
MBTS	-	0.2	0.4	0.6	0.8	1.0
Peptizer	2.0	2.0	2.0	2.0	2.0	2.0
Sulphur	1.6	1.6	1.6	1.6	1.6	1.6
Stearic acid						

### Preparation of Adhesive using Peptizer as Masticating Agent

The same procedure was followed for the adhesives produced using the peptizer (benzoyl peroxide) (Sample B- Sample F). However, in this case, no mechanical mastication was required. The average time for formulating each sample was ten minutes. The adhesives were prepared according to the formulation in Table 1. 400 ml of xylene was introduced into a conical flask for each sample. Then the natural rubber and peptizer were added and stirred at 100°C in a water bath until a peptized natural rubber solution was formed.



**Fig. 3.** Natural rubber base adhesive produced using mechanical mastication and chemical peptizer

Other reagents were sequentially introduced with continuous stirring until a homogeneous mixture of the natural rubber base adhesive was obtained. This step was followed to formulate the entire sample B, C, D, E, and F corresponding to 0.2phr, 0.4phr, 0.6phr, 0.8phr and 1.0 phr of the peptizer, respectively. However, for sample A, the rubber was masticated in two roll mills to reduce its molecular weight, followed by dissolution in xylene at 80°C in a water bath and a sequential introduction of other additives. Fig. 3 shows the various samples of the adhesive formulated.

## Characterization of the Adhesives produced

### FT-IR Analysis of the Adhesive

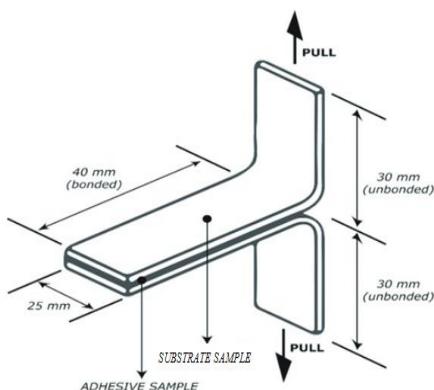
The adhesive was characterized for FT-IR to determine the functional groups present according to ASTM E1252.

### Viscosity Test

The test was conducted according to ASTM D1084. The rotor size of the digital viscometer was 4, and the rotor speed 600rpm. The rotor was inserted perpendicular to the adhesive, and the average viscosity of the adhesive was recorded from the screen of the digital viscometer.

### Adhesion Tests

The NR adhesives produced by mechanical mastication and peptizer were used to bond different substrates commonly used in footwear and leather products. The substrate used for bonding was leather material, polymeric flexible insole, slightly hard rubber outsole (Maco outsole) and flexible rubber outsole. Various adhesive bonding tests were carried out on different combinations of substrates.



**Fig. 4. a)** Schematic representation of T-peel test sample



**Fig. 4. b)** T-peel test samples of polymeric insole materials

Three different joints were bonded together for the shear test, involving leather to flexible rubber outsole, flexible polymeric insole to flexible rubber outsole (Nora), and a slightly rigid rubber outsole to flexible rubber outsole. For the T-peel test, three joints were also bonded, involving leather to leather, leather to the flexible polymeric insole and flexible polymeric insole to flexible polymeric insole. The weathering test involves joint flexible rubber outsole to a slightly rigid rubber outsole, while the effect of moisture and temperature test involving a joint of flexible polymeric insole to the flexible polymeric insole were evaluated. The procedure for the various tests is presented in the subsections.



Fig. 4. c) T-peel test samples of leather-to-leather materials



Fig. 4. d) T-peel test samples of leather to polymeric insoles

*T-Peel Test*

The test was conducted according to ASTM D1876 Figures 4a to 4d. Each end of the T-peel specimen was clamped on the universal tester's test grips. The specimen was separated at a rate of 127 mm.



Fig. 5. a) Schematic lap shear test sample

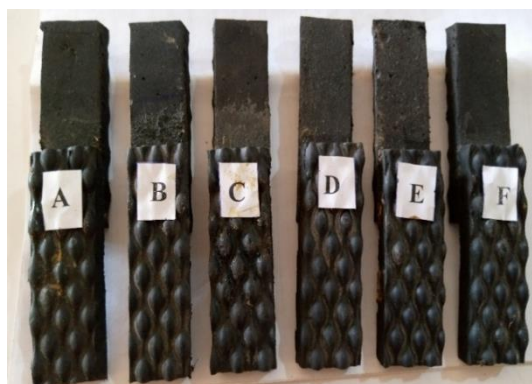


Fig. 5. b) Lap shear test samples of slightly rigid rubber outsole materials

*Lap shear strength Test*

The lap shear strength was conducted according to ASTM D 1002 (see Fig. 5a to 5b). The amount of shear area in square meters was measured. Each end of the specimen was loaded in the tensile grips. A force was applied at the controlled rate of the specimen until it broke. The maximum force was recorded, and the maximum shear stress was computed using equation (1).

$$\text{Maximum shear stress} = \text{maximum Force/Shear Area} \quad (1)$$

*Drying Time Test*

The test was conducted according to ASTM D1640. Equal quantities of the various adhesive samples were applied on a flat glass and allowed to air dry at room temperature. The time it took for the adhesive to dry uniformly and form a thin film was recorded for each sample under this study.





Fig. 5. c) Lap shear test samples of slightly rigid rubber



Fig. 5. d) Lap shear test samples of rubber to the leather outsole to insole material

#### *Accelerated Weathering Test*

The test was conducted according to ASTM D2919. The samples were prepared using the same dimension for the T-Peel strength test; the samples' initial strength before exposure was recorded. The samples were exposed in an open atmospheric environment for seven days. The samples were subjected to a peel test to determine the durability of the bond. The weather resistance was determined from the formula in equation (2).

$$\text{Percentage increase in peel strength} = (B - A) \times 100/A \quad (2)$$

where:

A is the initial peel strength and B is the final peel strength.

#### *Effect of Moisture and Temperature on Bond Strength*

The test was conducted according to ASTM D1151. The samples were prepared using the same dimension for the shear lap strength test. The samples were immersed in water at 23°C for 7 days, after which the shear strength of the exposed samples was then tested.

## **Results and discussions**

### **FT-IR result**

The plots in Fig. 6 a) and Fig. 6 b) represent the FT-IR analysis of the adhesive produced by mechanical masticated sample and using 0.4 phr of peptizer respectively. The characteristic peaks of mechanical mastication sample and 0.4 phr of peptizer loading at 3026 cm<sup>-1</sup>, 2922- 2926 cm<sup>-1</sup> and 2858 cm<sup>-1</sup> corresponding to the alkene stretching of -CH, asymmetric stretching of -CH<sub>3</sub> and symmetric stretching of -CH<sub>2</sub> were evident [20]. The differences in the intensity of the peaks in the 0.4 peptizer sample and mechanical masticated sample in this region is due to the ability of the peptizer to cause more breakdowns in the NR resulting in the hydrogen abstraction [21].

The 1606 cm<sup>-1</sup> -1602 cm<sup>-1</sup>, 1494 cm<sup>-1</sup>, 1453 cm<sup>-1</sup> and 730 cm<sup>-1</sup> – 693 cm<sup>-1</sup> peaks of the mechanical masticated and 0.4 peptizer sample indicates the evident of conjugated alkene stretching due to depolymerization of polyisoprene to isoprene unit, aromatic C=C, alkane methyl group bending of -CH, gem dimethyl bending of -CH and Cis disubstituted alkene bending of C=C [22].

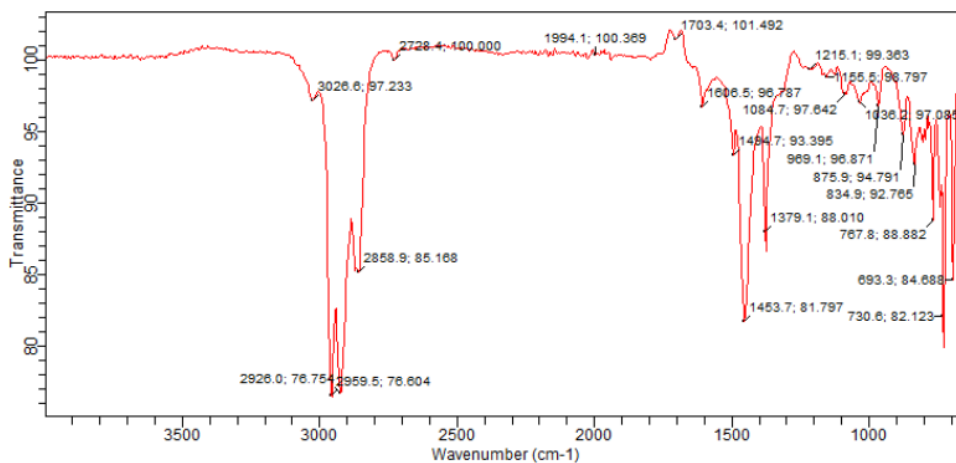


Fig. 6. a) FT-IR Spectra of Adhesive Produced by Mechanical Mastication

The  $2732\text{ cm}^{-1}$  and  $2728\text{ cm}^{-1}$  peaks represent in the 0.4 peptizer and mechanical masticated samples shows evident of aldehyde stretching of  $-\text{CH}$ . According to Naylor [23], the oxidative fission of the double bond of an isoprene unit may be expected to give rise to an aldehyde group. The difference in the peak could be as a result of shift due to hybridization changes or electronic distribution [24]. This shows the extent of oxidation by the benzoyl peroxide peptizer resulting in more unsaturation in the compound. The  $1080\text{ cm}^{-1}$  peak present in the chemical peptizer indicates the presence of secondary alcohol stretching of  $\text{C}-\text{O}$ . This functional group is formed due to reduction in the benzoyl peroxide which affects the oxidation [23].

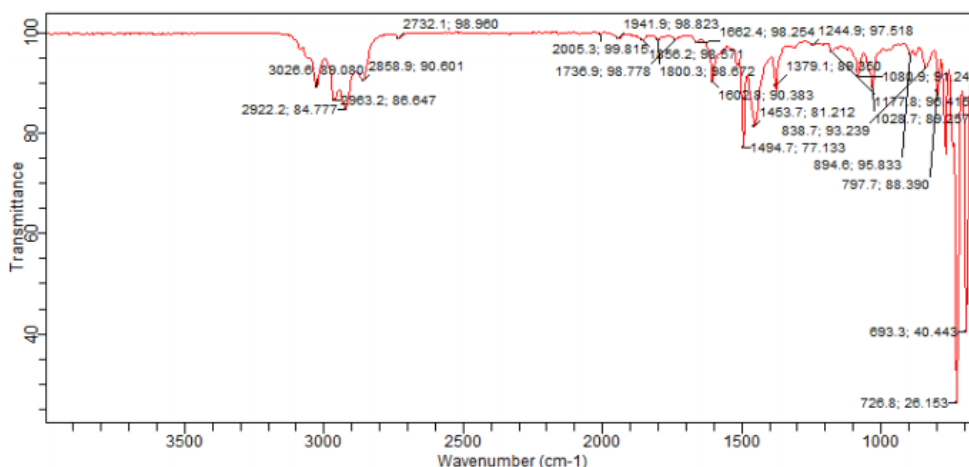


Fig. 6. b) FT-IR Spectra of adhesive produced by 0.4 phr of peptizer loading

The ability of the peptizer to effectively oxidize and lower the molecular weight of natural rubber result to better dispersion of other compounding additives, thus increase the shear strength, peel strength, and reduce the viscosity of the adhesive.



### Viscosity

The results presented in Fig. 7 showcase the effect of introducing peptizer in the adhesive formulation from 0 to 1 phr at an interval of 0.2 phr with corresponding viscosities for the various adhesive formulations. The viscosity of the adhesive is a measure of adhesive flow, which 136etermine show well it can wet the substrate surface area for adequate bonding. From the result in the chart, an increase in the peptizer loading significantly increases the adhesive's viscosity for peptizer loading up to 0.8 phr (sample E). However, the viscosity remained almost steady with the further addition of the peptizer from 0.8 phr (sample E) to 1.0 phr (sample F). The increase in viscosity of the adhesive between 0.2 – 0.8 phr could be attributed to the ability of the peptizer to cause a breakdown of more rubber molecules resulting in the dissolution of not only more solid rubber but also other additives. The maximum viscosity value was about 7000 mPa.s representing the maximum peptizer loading under this study.

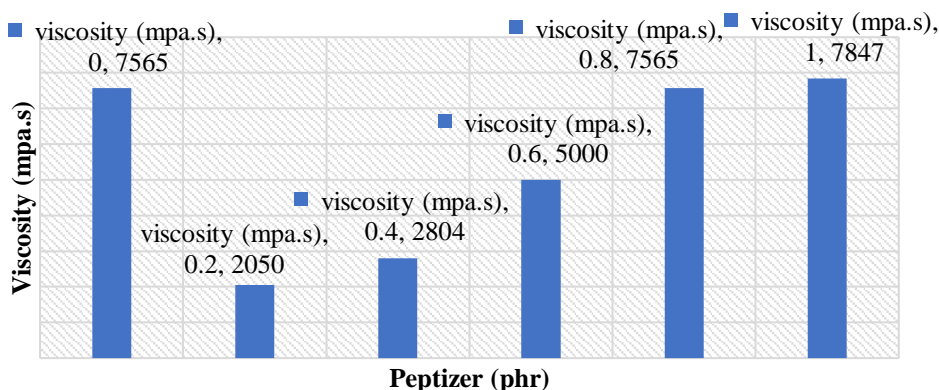


Fig. 7. Viscosity of natural rubber adhesive produced by mechanical mastication and peptizer

Similarly, the viscosity of the mechanically masticated sample was seen to be 7000 mPa.s. Generally, adhesive viscosity tends to increase with solid content [25], [26]. However, the ideal viscosity would strike a balance between good wetting and the enhanced strength of bonded joints.

The multiple bar charts (Fig. 8) show the T-peel test involving leather to leather joint, leather to insole joint, and insole joint. Peel tests intended for flexible adhesive measure resistance to highly localized stress. The insole-to-insole joint had better peel strength for all samples under study. This finding could be attributed to the nature of the substrates and their ability to form mechanical interlocking, thereby maximizing the bonding strength provided by the adhesive, consequently reducing the effect of stress concentration on the bonded joint. This was in line with the report of Ebnesajjad and Landrock [27] suggest that the higher the adhesive modulus and flexibility of adherend, the more nearly the stressed area is reduced to linearity. Poh and Chang [28] also reported that the higher the wettability, the higher the peel strength of adhesive joints.

The least peel strength was recorded with leather-to-leather surfaces, which could be attributed to poor absorption of the adhesive into the leather material resulting in poor interface strength. The highest peel strength was about 39N/25mm recorded with the 0.4 peptizer for the insole-to-insole material. This deduction indicates that more mechanical interlocking occurs as the adhesive flow into the pores of the substrate. The insole-to-insole joint has more pores; as such, more wetting occurs in the substrate. However, with further increase in peptizer loading, the T-peel strength of the adhesive joints was seen to decline irregularly; this could be possibly due to shifting from cohesive to adhesive failure as a result of the transition from viscous-like liquid to rubber-like elastic behavior arising from much rubber content in the adhesive [29].

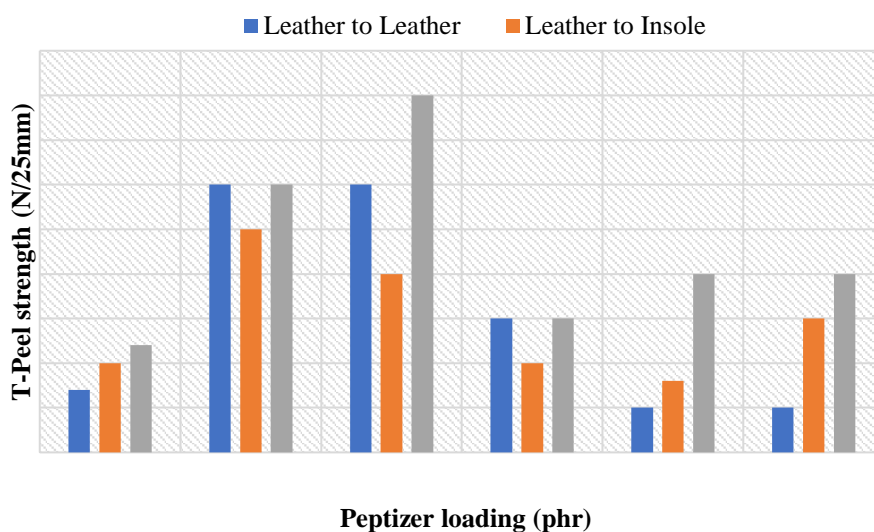


Fig. 8. T-peel strength for different joints bonded by natural rubber-based adhesive produced by mechanical mastication and peptizer

Furthermore, the mechanically masticated adhesive (sample A) exhibited relatively less T-peel strength for all adherend type. This clearly shows that the use of the benzoyl peroxide as a masticating agent also significantly improves the adhesive's strength performance by acting as radical acceptors in stabilizing the dissolved rubber molecules. Brendan & Walter [30] reported that to achieve significant improvement in compound processability, reduction in energy consumption during mixing and improvement in compound uniformity, peptizers such as pentachlorothiophenol are generally used at levels between 0.1 and 0.25 phr. However, higher levels can adversely affect the compound properties, as excess peptizer continues to catalyze polymer breakdown as the product is in service. To this end, the optimum benzoyl peroxide loading for natural rubber adhesive is 0.4 phr.

Fig. 9 represents the table of results for Lap shear strength of the viscosity joint of the different substrates. The lap shear test describes the ability of an adhesive to resist force in the plane of bonded surfaces. The lap shear strength value indicates the strength of an adhesive [31]. The adhesive joint resisted shearing with an increase in peptizer loading. At a force of 130N, there was substrate failure for all samples bonded with adhesive formulated using the peptizer (sample B –sample F). This could be attributed to the rigid nature of one of the adherends, i.e., the Nora material. However, the mechanically masticated adhesive bonded joints (sample A) were observed to shear at forces between 100N and 115N. The flexible polymeric rubber outsole (Nora) and leather had the highest shear strength, while the flexible polymer outsole and insole had the least shear strength for this sample. Again, this observation further validates the activity of the peptizer in the serving receptor by eliminating the possibly formed free radicals during mixing, inhibiting polymer recombination and consequently improving the overall strength of the adhesive [30].

The drying time is the time required for the solvent to evaporate from the adhesive leaving the solid content. The drying time of adhesive can be affected by many factors such as the solid content present in the adhesive, temperature, nature of the solvent, and many others. The result shows that the drying time reduces with an increase in peptizer loading except for the mechanical masticated sample, which has a drying time slightly higher than the least peptizer loading see Fig.

10. The reduction in drying time with increasing peptizer content indicates that the solid content of adhesive was able to dissolve more in the solvent, leading to more exposure to air and dry faster on surfaces when applied. Generally, the drying time of solvent-based adhesive is governed mainly by nature and solvent type [32].

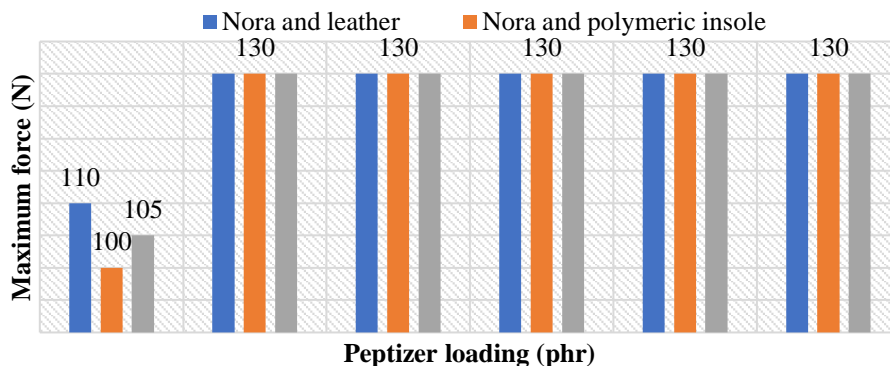


Fig. 9. Lap shear strength for different joints bonded by natural rubber adhesive produced by mechanical mastication and peptizer

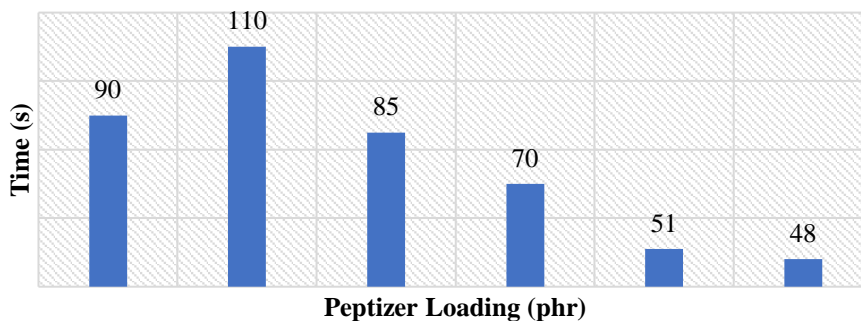


Fig. 10. The adhesive's drying time produced by mechanical mastication and peptizer

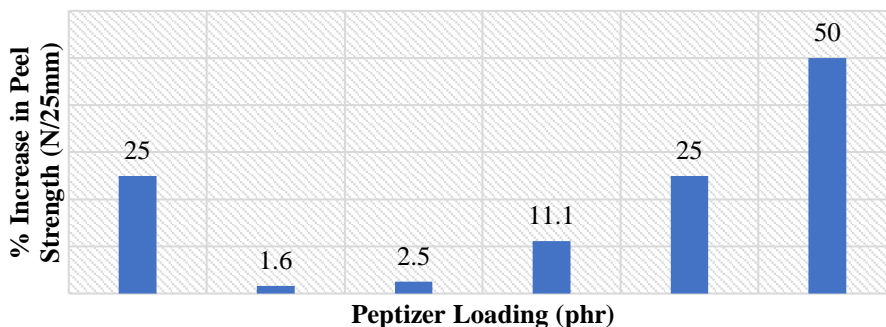


Fig. 11. Weathering resistance of insoles bonded by natural rubber-based adhesive produced using mechanical mastication and peptizer.

Fig. 11 shows the weather resistance of adhesive joint using the insole-to-insole adherends when exposed to the atmosphere for seven days. Weathering test determines the durability of adhesive when exposed to heat, UV from the sunlight, moisture, and many others. The result shows that the T-peel strength of all samples formulated using the peptizer (sample B – sample F) increases with an increase in the peptizer content after exposure to atmospheric weather. This may be attributed to the ability of the benzoyl peroxide peptizer to promote the curing activity of the sulphur, which resulted in cross-linking on exposure to elevated, thereby impacting more strength on the bonded joints. However, with more prolonged exposure to this adverse weather, the tendency for the adhesive to strain or break is increased. An adhesive's chemical makeup alters at higher temperatures, breaking down the components and causing the adhesive to crystallize and become unstable. The adhesive can then crack or shatter, no longer being useful.

The sample with the highest peptizer loading had the highest percentage increase in T-peel strength, while the least peptizer loading had the least increment.

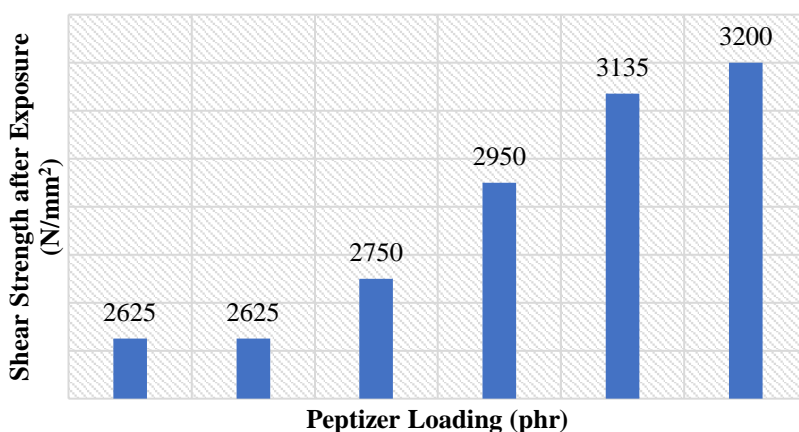


Fig. 12. The effect of moisture and temperature on adhesive joint bonded by natural rubber-based adhesive produced by mechanical mastication and peptizer

The results presented in Fig. 12 represent the outcomes obtained from the effect of moisture after seven days. Moisture is a substance that causes great difficulty in terms of the environmental stability of adhesive. The result obtained shows that moisture and temperature have an effect on the peptized sample except for the control sample. The shear strength of the exposed sample increases with an increase in peptizer loading. This could be due to the permeability of natural rubber due to the unsaturation present in its chain [33]. During peptization, thermo-oxidation occurs, resulting in hydrogen abstraction. This hydrogen abstraction gives rise to unsaturated polymer; therefore, the more effective the peptizing action, the higher the water vapour permeability, thus the lower the shear strength. When water permits the adhesive, it preferentially migrates to the interfacial region displacing the bulk polymer at the interface [34].

## Conclusions

In conclusion, the importance of chemical peptizer in natural rubber-based adhesive cannot be overemphasized; literature on the use of various chemical additives as peptizers has shown that they aid the breaking of rubber molecules and serve as radical acceptors. Still, these are used to complement mechanical mastication in roll mills. This study has shown that peptizer

such as benzoyl peroxide can effectively masticate natural rubber without employing a roll mill. Furthermore, important adhesive properties such as T-peel strength were significantly improved by the novel process of using peptizer as a masticating agent. In addition, the weathering resistance determined by the T-peel strength also showed significant weather resistance within the exposure time. Chemical peptizer has shown to be more effective in the mastication of natural rubber as it has better properties than the mechanical masticated adhesive sample. This study shows that temperature affects the efficiency of the chemical peptizer used. At 100°C, an increase in the peptizer slows down the breakdown efficiency. The optimum peptizer loading for this study was obtained at 0.4phr peptizer loadings (sample C); at this loading, there was a balance in the adhesive properties produced when subjected to testing.

### Acknowledgement

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