Journal of Civil Engineering and Environmental Technology
p-ISSN: 2349-8404; e-ISSN: 2349-879X; Volume 10, Issue 2; April-June, 2023, pp. 63-67

http://www.krishisanskriti.org/Publication.html

Eco-Friendly Adhesive from Natural Rubber

Mohamed Faheem Faisal[1], Mohamed Abdul Jaleel[2], Abdullah S[3] and David K Daniel[4*]

[1,2,3]UG Student, Department of Chemical Engineering, Amal Jyothi College of Engineering, Kanjirappally, Koovappally P.O., Kottayam District, Kerala-686518
[4]Professor, Department of Chemical Engineering, Amal Jyothi College of Engineering, Kanjirappally, Koovappally P.O., Kottayam District, Kerala-686518*
E-mail: [1]mohamedfaheemfaisal2023@ch.ajce.in, [2]mohamedabduljaleel2023@ch.ajce.in
[3]abdullas2023@ch.ajce.in, [4]davidkdaniel@gmail.com

Abstract—*Growth in synthetic adhesive use has contributed to environmental problems like release of volatile organic compounds to the atmosphere, which contribute to the formation of photochemical smog, and many are implicated in the aggravation of lung diseases such as asthma. Almost all organic vapors absorb infrared radiation and therefore act as greenhouse gases. But this impact can be reduced by using adhesives made from established renewable resources such as natural rubber. Natural rubber is an important commercial material and can be used to make adhesives and a rubber plantation is very effective as a forest in carbon dioxide utilization. Natural rubber-based adhesives can be employed to bond non-metallic materials such as leather, fabrics, paper, and other rubber products and the depletion of fossil fuels can be prevented by their use. Therefore, the goal of this work was to develop a natural rubber-based bio-adhesive capable of competing with the currently used formaldehyde-based synthetic adhesives. From the experiments carried out, it was found that the relative viscosity and the solubility parameter play a major role in the selection of the best combination of natural rubber, tackifier and solvent to form the bio adhesive. The adhesive mix with solvent blend of hexane and heptane (80:20) was found to yield better adhesion. It was observed that with increase in the concentration of natural rubber and rosin, the viscosity increased. This bio-adhesive was tested for its bonding quality with different materials like paper, veneer, chart papers and rubber tubes. The properties of these materials were then compared with materials bonded using other synthetic adhesives available.*

INTRODUCTION

Wood adhesives currently being used are mainly synthetic polymers especially formaldehyde based and among them the most important are phenol formaldehyde (PF)-urea formaldehyde (UF), urea melamine formaldehyde (UMF), melamine formaldehyde (MF), resorcinol formaldehyde (RF) and phenol-resorcinol formaldehyde (PRF) resins [7,8,11,21]. They are the most widely used in panel industries for making wood-based panels and wood laminates. Although these types of resins have excellent characteristics, the formaldehyde release is their critical defect. With growing demand for green and clean processes the limits of use of formaldehyde-based adhesives is a growing concern. Numerous methods have been reported in literature [2,3, 20,23] such as lowering the resin final mold ratio, adding different amine compounds to the resin and adding formaldehyde scavenger, however, in spite of this progress, the potential harm of formaldehyde has not been completely eliminated. Due to the potential harm of formaldehyde-based resins, bio-based adhesives including protein, starch, tannin, carbohydrate, walnut meal, coconut shell and so on have attracted increased attention in the wood panel industry. An exhaustive review about non-formaldehyde, bio-based adhesives for use in wood based panel manufacturing industry was carried out on the development of formaldehyde-free adhesive systems based on renewable resources [6,9,22]. The current wood panel market as focused in this article, emphasizes the challenges that new and alternative bio-based adhesives must face. Another review discusses recent developments in the synthesis of PF resins, particularly those created from sustainable raw material substitutes [20] along with modifications applied to the synthetic route in order to improve the mechanical properties. They concluded that although bio-based alternatives to phenol and formaldehyde are thought to be ecologically beneficial, switching to bio-based materials occasionally results in a reduction in the resin's mechanical qualities when compared to phenolic resin. Coconut shell flour was found to be effective for partial replacement for phenol in phenol formaldehyde resin [10]. The effect of the partial replacement on tensile strength and morphology were investigated and it was revealed that 40-50% off phenol can be replaced by using coconut shell powder. A biobased phenolic adhesive was successfully developed by entirely substituting both petroleum-based phenol and formaldehyde with an unmodified corn stover biorefinery lignin and glyoxal respectively. The lignin glyoxal resin was found to have a 3-fold higher average molecular weight than the starting lignin, demonstrating the integration of lignin into polymeric resin network [12]. Therefore, because of the toxicity, volatility and restrictions on use, there is a strong need to develop and implement appropriate alternatives to replace formaldehyde based adhesive systems. Viable formaldehyde free adhesive systems based on renewable sources either at the technical or economical level have to be encouraged. In this context natural rubber-based adhesives have good ability to bond a wide variety of substrates and retain excellent adhesion when

subjected to varying environmental conditions [1,4]. Hence, in the present study an attempt has been made to develop natural rubber and solvent based adhesives which could meet the requirements of footwear, wood and automobile industry. The effects of natural rubber concentration, tackifier and solvent properties on the natural rubber adhesive were determined. The adhesive viscosity was found to increase with the concentration of natural rubber and tackifier. The relative viscosity was dependent on the solvency and the solution viscosity decreased with solvent viscosity.

MATERIALS AND METHODS

Natural rubber sheets were purchased from a local rubber dealer near the institute. n-Heptane (99%) and Hexane (85%) were procured from Simson Pharma Ltd., Mumbai. All other solvents used were of LR grade and purchased from Nice Chemicals, Kochi. Tackifier was procured from Punjab Rosin and Chemical works, Roorkee. Veneer sheets were provided as a gift from Kelachandra Wood Industries, Kottayam and TKM Ply and Boards, Muvattupuzha, Kochi.

Bio Adhesive Formulation

The natural rubber sheets were masticated into pieces. A mixture of 5% natural rubber and 1.5% tackifier were sequentially mixed in the solvent and mixed for 4 hours at 800 rpm using a mixer with cone shaped beaters (Philips, India) until a homogeneous mix is formed. The adhesive thus formed was stored at room temperature after addition of stabilizing and binding agents to ensure a uniform free flowing glue solution. The developed bio adhesive was then applied on different material surfaces and pressed using a hydraulic press to form specimens whose bonding characteristics were evaluated. Samples using conventional adhesives were physically compared with samples developed from this bio adhesive.

Bio Adhesive Property Estimation

The density of the solvents and the adhesive solution was found out using standard IS: 4730:1994. The dynamic viscosity was calculated using a Brookfield DVE (Ametek, USA) rotational viscometer. The speed of the rotor was calibrated such that the dynamic viscosity of the distilled water was 1 cP at 25ºC. The viscosity of the solutions and the solvent were measured at 25ºC. The samples of 500 mL were taken for each measurement. The instrument was switched on and the stress was applied on the adhesive and the viscosity was measured. The bio adhesive glue solution was applied on various surfaces and the surfaces bonded together using a temperature controlled hydraulic press (Mini Metal Industries, Kottayam). The work flowsheet for the preparation of sample is depicted in figure 1.

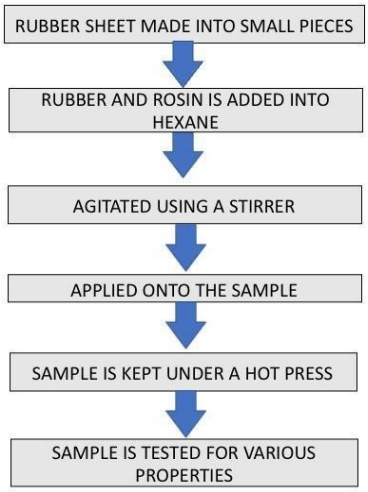

Figure 1: Methodology

RESULTS AND DISCUSSION

Properties of Natural Rubber and Tackifier

The density and the color of the natural rubber and the tackifier (gum rosin) used for the project are shown in the Table 1.

Table 1: Properties of NR and Gum Rosin

Sl. No	Material	Density (g/cm^3)	Color
1.	NR Sheet	0.95	Brown
2.	Rosin	0.57	Yellow

Along with the pure solvent varying blends of two different solvents were also tested. In order to find the optimum blend ratio, the two solvents (hexane and heptane) were mixed in different proportions and were tested for viscosity. The viscosity of the bio adhesive solution consisting of same percentage of solvents was found to be 9600 cP, whereas the viscosity for 80:20 blend of solvents was found to decrease by 10%. Hence considering the cost this blend was selected as the optimum one. The variation of relative viscosity with varying blends of solvents is depicted in figure 2.

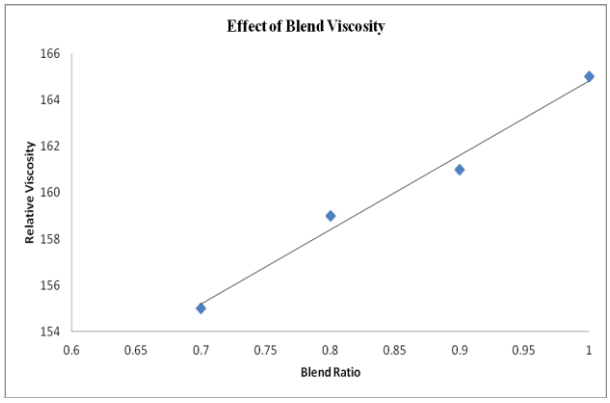

Fig. 2: Determination of optimum blend ratio

The viscosity of the hydrocarbon solvents and blends of solvent were tested and it was observed that for increase in viscosity of the solvent the solution viscosity decreased. This was mainly due to the solubility parameter of the solvent. It is in agreement with the literature that energetically favorable solvent has solubility parameter that closely matches with that of the solute [16-19] and is depicted in figure 3.

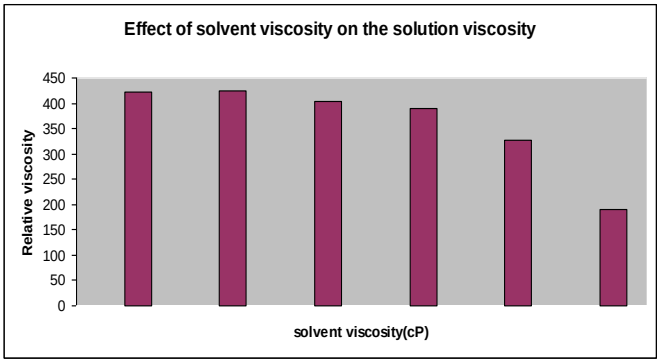

Fig. 3: Effect of solution viscosity on adhesive viscosity

Effect of Concentration of natural rubber on the viscosity of the adhesive

Tests were carried out for varying concentrations of natural rubber in the solution for same tackifier loading. Three different solvents were used for the study. It was found that as the concentration of the natural rubber in the mixture increased the viscosity also increased. It was found that in case of 3g of natural rubber, the dissolution of solute molecules in the solvent is incomplete, for all solvents (figure 4). This observation was associated to the increase in the solid content due to rubber particles in the mixture which is in agreement with the literature [13-15].

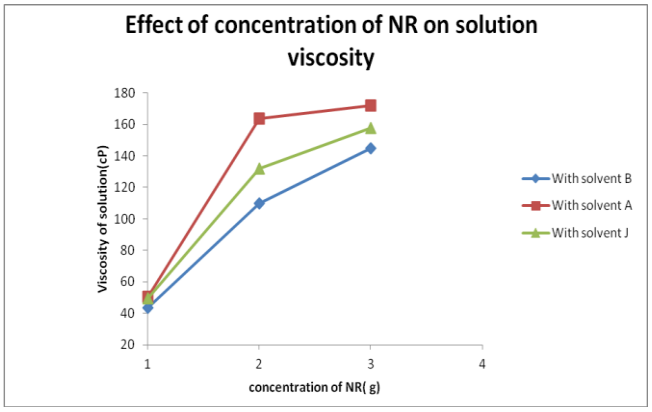

Fig. 4: Effect of concentration of natural rubber on the adhesive viscosity

Effect of concentration of gum rosin on the viscosity of the solution

In this study, the effect of tackifier was found on the viscosity of the solution and the results are shown in figure 5. The concentration of the gum rosin was found to increase for the given concentration of natural rubber and given amount of solvent. It was noted that the viscosity increased with increase in the concentration of the gum rosin. Similar observations were found earlier [14,24].

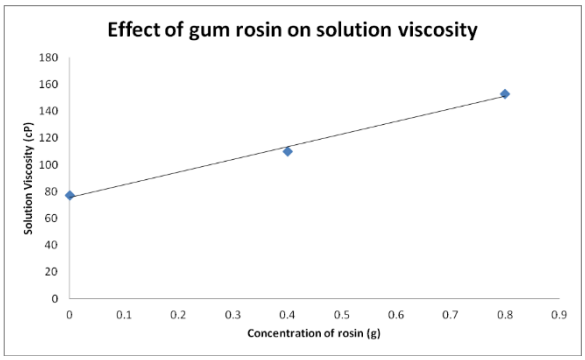

Fig. 5: Solution viscosity as a function of gum rosin concentration

Adhesive

An eco-friendly adhesive was developed from natural rubber using heptane as the organic solvent. The adhesive produced was moderately viscous (figure 6). In order to test the bonding properties of the adhesive, it was tested on various materials such as paper, veneer sheets, rubber and photographs. The physical properties of the adhesive were similar to that of synthetic adhesive available in the market. The adhesive was having good bonding property.

Fig. 6: Bio Adhesive

Wood to Wood Bonding

The developed bio adhesive was applied on veneer sheets and the tear characteristics [5] were physically examined. Two different specimens have been made. The first specimen having 6 layers of 10x10 cm veneer sheets was bonded together using the adhesive made and the second one having 3 layers of 5x10 cm veneer sheets was bonded together with the adhesive. The bonding was strong and comparable with those reported in literature [6]. Both the specimens were subjected to a hot press machine and tested for the temperature resistance. It was having a comparatively good resistance to temperature and pressure. The bonding strength was also good because the veneer pieces were not getting separated when pressure was

applied. The specimen with 6 layers was found to have high strength when compared to the 3 layered specimen.

(a) 6 layer (b) 3 layer

Fig. 7: Samples of veneer sheets bonded together

Rubber to Rubber Bonding

A specimen was made by bonding two rubber pieces of 5x10 cm size and was kept under a press for a time of 10-12 hours. The bonding was done using the prepared adhesive and its properties were observed. It was observed that the bonding strength was comparatively less. The rubber sheets were easily separated when pressure was applied. The specimen was kept in a hot air oven for testing the temperature sensitivity. It was found that the specimen was separated after taking from the oven which proves that it was also sensitive towards high temperature.

Fig. 8: Rubber pieces bonded together using adhesive

Photo Paper Bonding

Two photo papers were bonded together using the adhesive to prepare a specimen. The face of the photo paper where the photo is printed was bonded together and was kept under a press for a time period of 4-6 hours. The specimen was taken and observed. The observation was that the bonding strength was good. And it was also observed that when the photos were separated by applying pressure there was no damage to both the photos, indicating that this adhesive solution could be very well used for craft paper bonding work.

Fig. 9: Photograph paper bonded together

CONCLUSION

Adhesive based on natural sources due to their eco-friendly properties and impressive glue strength, are becoming more popular and are being used for different applications. Their importance has been increasing over the years due to the problems created by synthetic based adhesive. The synthetic based adhesives such as formaldehyde based adhesive emit toxic gases to the atmosphere they are present. These toxic gases are dangerous to our health and cause several health problems. In order avoid this problem we have developed a bio adhesive from natural rubber. Experiments were carried out to find the properties of the blends. It was found that the relative viscosity and the solubility parameter play a major role in the selection of the best combination of natural rubber, rosin and solvent to form an adhesive. The developed adhesive was then applied to different material such as wood, rubber and photo paper. Then their properties were observed and studied. The veneer sheets were having good bonding strength, similar to that of synthetic adhesives. But the bonding between rubber sheets were not comparatively good. The bonding was also sensitive to high temperature. The bonding between photo papers were comparatively good and it was observed that the photos were not damaged when they peeled off.

The sample with solvent 80:20 blend of hexane and heptane was found to yield better adhesion. It was observed that with increase in the concentration of natural rubber and rosin, the viscosity increased. The effect of solvent viscosity was also determined in the process. It was observed that solvent viscosity directly contributes to the viscosity of the solution. The increase in concentration of natural rubber caused an increase in the solution viscosity. An increase in tackifier concentration increased the viscosity of the solution and was proportional to tackifier loading. The presence of the alcohol led to decrease in viscosity leaving a product which was not stable. Greater the solvency, better was the dissolution of the solute and thus lower was the relative viscosity. For the hydrocarbon solvents, the solution viscosity decreased with increase in solvent viscosity which was due to the increase in solubility parameter of the solvent. Further work is in progress to characterize the physical and mechanical properties of this adhesive solution.

ACKNOWLEDGEMENT

The equipment used for this work was supported by the funding support received from ***DST-FIST,*** *Grant No.*SR/FST/COLLEGE-346/2018.

REFERENCES

[1] Ansarifar, M.A., Zhang, B.J., Bell, A., Ellis, R.J., "Bonding properties of rubber to steel, aluminium and nylon6,6", *International Journal of Adhesion and Adhesives*, 2001, pp. 369-380.

[2] Boran, S., Usta, M., and Gumuskaya, E., "Decreasing formaldehyde emission from medium density fiberboard panels produced by adding different amine compounds to urea formaldehyde resin", *International Journal of Adhesion and Adhesives*, June 2011, pp. 674-678.

[3] Berth, A.M., Jack. L.K., "A review of polymer dissolution, Progress in Polymer Science", 2003, pp. 1223-1270.

[4] Brydson J. A.,"Rubbery materials and their compounds, Elsevier Applied Science, 1988.

[5] Cook, J.W., Edge, S., and Packham, "The adhesion of natural rubber to steel and the use of peel test to study its nature", *International Journal of Adhesion and Adhesives*, 2007, pp. 333-337.

[6] Diogo, G., João, M.B., Ana C.M., "Non- Formaldehyde, Bio-Based Adhesives for Use in Wood-Based Panel Manufacturing Industry", 2021, pp. 50-100.

[7] Dunky, M., "Urea-formaldehyde (UF) adhesive resins for wood", *International Journal od Adhesion and Adhesive 1998,* pp. 95-107.

[8] Ganeshram, V., and Achudhan, M., "Synthesis and Characterization of Phenol Formaldehyde Resin as a Binder used for Coated Abrasives", *Indian Journal of Science and Technology,* 2013, pp. 511-516.

[9] Hui, W., Feng, W., and Guanben, D., "Walnut Meal as Formaldehyde-free Adhesive for Plywood panel", *Southwest Forest University,* Yunnan, China, 2018.

[10] Inamdar, S.S., Kiran, K.R., "Study of characteristics of partial replacement of phenol by coconut shell flour in a phenol formaldehyde resin", *Assistant Professor, Department of Chemical Engineering SDM College of Engineering and Technology,* 2013.

[11] Lee, Y. K., and Kim, H. J., "Relationship between curing activation energy and free formaldehyde content in urea-formaldehyde resin", *Journal of Adhesion Science and Technology,* 2013, pp. 5-6.

[12] Mohsen S., Sasha E., David B.H. and Mojgan N.,"Lignin-Glyoxal: A fully Biobased Formaldehyde free Wood Adhesive for Interior Engineered Wood Products", ACS Sustainable Chemistry & Engineering,2022, pp. 3430-3441.

[13] Nuruldiyanah, K., Dayang, R.A., Zurina, Z.A., Francisco, C., and Salit, M., **"**Rheological Study of Phenol Formaldehyde Resole Resin Synthesized for Laminate Application", *Institute of Advance Technology, University Putra Malaysia*, 2020, pp. 1-100.

[14] Palkin, S., Smith, W.C., "A new non- crystallizing gum rosin", *Journal of American Oil Chemist Society*, 1938, pp. 120-125.

[15] Pizzi, A., Mittal, K.L., *Handbook of Adhesive Technology, 1994, Marcel Dekker Inc*, New York.

[16] Poh, B.T., Lee, P.G., and Chuah, S.C., "Adhesion property of epoxidized natural rubber (ENR)-based adhesives containing calcium carbonate", *EXPRESS Polymer Letters,* 2008, pp. 398 - 403.

[17] Poh, and Ong, "Adhesion properties of styrene-butadiene rubber (SBR)/ Standard Malaysian Rubber (SMR L)-based adhesives in the presence of phenol formaldehyde resin", *EXPRESS Polymer Letters*, 2007, pp. 654-659.

[18] Pranee, C., and Oukrit, "Preparation, adhesive performance and stability of natural rubber latex grafted with n-butyl acrylate (BA) and methyl methacrylate (MMA)", *Suranaree Journal of Science and Technology*, 2007, pp. 269- 276.

[19] Reynolds, and Gebhart, Jr., "Effect of solvent properties on the viscosity of concentrated Natural Rubber Solutions", *Journal of Chemical and Engineering Data*, 1960, pp. 220-225.

[20] Sarik, P.R., Paul, N.A., and Taleb, I.,**"**Bio-Based Alternatives to Phenol and Formaldehyde for the Production of Resins", 2020, pp. 1-7.

[21] Satish, K. S., and Khali, D. P., "Synthesis and Comparison of Phenol-Urea- Formaldehyde (PUF) Thermosetting Resin with Commercial Synthetic Resins", *Asian Journal of Research in Chemistry,* 2015, pp. 1-20.

[22] Triveni, S., Arun. G.,and Saidatul, S.B., "Eco-Friendly Bio-Based Adhesive for Plyboard from Natural Rubber Latex (NRL) Blended Isocyanate Crosslinked Starch", *University Malaysia Pahang*, 2020, pp. 674-680.

[23] Ye, J., Quin, T., Wang, H., Guo, L., and Li, X., "Study of glycidyl ether as a new kind of modifier for urea- formaldehyde wood adhesives", *Journal of Applied Polymer Science,* 2013.