Journal of Tropical Forest Science 36(3): 274–279 (2024)
https://doi.org/10.26525/jtfs2024.36.3.274
ISSN: 0128-1283, eISSN: 2521-9847

DEVELOPMENT OF DEFATTED SOY FLOUR AND CASEIN-BASED BIO ADHESIVES FOR WOOD JOINT

Patel DS*, Dholakia AB, Patel VC & Patel AJ

Department of Organic Chemistry, Shri Alpesh N. Patel Post Graduate Institute of Science & Research, Anand, Gujarat 388001, India

**darshanswami@gmail.com*

Submitted July 2023; accepted March 2024

Wood adhesives were made using defatted soy flour, casein, sodium hydroxide (NaOH) and ethylene diamine. These eco-friendly wood adhesives are used to substitute formaldehyde-derived adhesives in order to decrease formaldehyde and volatile organic compounds emissions from the adhesives that are utilised in plywood. The features of adhesively-glued wood joints in terms of performance such as tensile strength, water resistance and chemical resistance were gauged in this study. The results revealed that adhesives showing good performance and eco-friendly adhesive system was prepared for wood joints.

Keywords: Bio adhesive, defatted soy flour, casein, wooden joints, acidic and basic chemical resistance, water resistance

INTRODUCTION

Environmentally friendly materials have been utilised increasingly in the past few decades as people have grown aware of the negative effects of using items made from petrochemicals. Moreover, two key concerns of the 21st century are health and the environment (Milota & Wilson 1985, Andrews et al. 1986, Pizzi & Mittal 2011). Replacing the formaldehyde-based resins used in the production of wood-based panels with resins of natural origin, is one of the biggest challenges associated with the panel's quality. Typically, formaldehyde-based resins consist of urea-, melamine-, and phenol-formaldehyde. However, other phenolic chemicals such as resorcinol can also react with formaldehyde to produce polymers of the same kind but with varying adhesive costs and reactivity (Alma & Kelley 2000). Particleboards (PBs), fiberboards such as medium density fiberboards (MDFs), oriented strand boards (OSBs), plywood (PW) and other wood-based panels are made by gluing together wood components of different sizes using these resins at the necessary pressure and temperature. However, the development of natural resource-based substitute materials for petroleum-derived phenolic compounds in the wood adhesives industry was prompted by the increase in oil prices, concerns about volatile organic compounds (VOCs), and the need to minimize fuel use (Kim 2009).

The development of new synthetic materials using natural sources is currently one of the most interesting new trends in natural products research (Nasir et al. 2014). Defatted soy flour is utilised with several kinds of substances in the preparation of adhesive for wood joints (Allen et al. 2010, Hunt et al. 2009). Standard urea-formaldehyde resins produced according to Chimar Hellas technology and soy protein was added as natural additives (1% $s1^{-1}$) at various stages of the synthesis. Soy protein was in the form of flour (SF) or isolated soy protein (SPI) (Papadopoulou et al. 2008). Casein is also utilised with several kinds of substances in the preparation of adhesives for wood joints (Pizzi & Mittal 1994, Petrie 2007). In this work, casein and defatted soy flour—two natural and environmental-friendly ingredients—are used to formulate wood adhesives. This is an effort to investigate renewable materials for value-added applications such as wood adhesives that addresses the problem of availability of unprocessed material and VOC emissions.

MATERIALS AND METHODS

Experimental materials

Casein (60%, 75% and 90%) was procured from Anand Casein Udyog, Vithal Udyognagar, Anand, Gujarat, India. Soy flour was procured from local grocery store. Ethylene diamine, sodium hydroxide and concentrated sulfuric acid were laboratory-grade reagents. All other chemicals and materials were laboratory-grade reagents and used without any modification.

Formation of defatted soy flour

The Soxhlet method is used to remove oil from soy flour to prepare defatted soy flour. The nitrogen and protein contents in defatted soy flour and different percentages of casein were measured using Kjeldahl method. The nitrogen and protein contents in defatted soy flour and casein, and oil content in defatted soy flour were given in Table 1 (IS 548: Part I 1964).

Formation of adhesive from defatted soy flour and ethylene diamine

Adhesives were prepared by mixing defatted soy flour, ethylene diamine and NaOH in different proportions in water as solvent. Solutions of NaOH and ethylene diamine were prepared at room temperature. Defatted soy flour (9 g) was suspended in NaOH and ethylene diamine solution (25 mL), and stirred for 6 h (Figure 1).

Preparation of adhesive from casein and ethylene diamine

Adhesive systems were formulated by mixing casein, ethylene diamine and NaOH in different proportions in water as solvent. Solutions of NaOH and ethylene diamine were prepared at room temperature. Casein (9 g) was suspended in NaOH and ethylene diamine solution (25 mL), and stirred for 6 h (Figure 2).

Wood joint binding process

Using a nylon brush, the prepared adhesives were applied on two pieces of teak wood at a thickness of 0.1 mm and lap-jointed with a 25 mm by 30 mm area of overlap. A pressure of about 1.3 MPa was applied to the joint for 48 h (Patel et al. 2012).

Measurement of bonding strength

Tensile strength

The fixed wood joints were tested for tensile strength according to the methods of American Society for Testing and Materials (ASTM D897-08 2016) using a Dutron Tensile Tester, Dutron Products, Ahmedabad, India.

Table 1 Analysis of defatted soy flour and casein

Sr. No.	Raw material	Oil content (%)	Nitrogen content (%)	Protein content (%)
1.	Defatted soy flour	10.2	6.18	38.64
2.	60% casein	-	10.92	68.26
3.	75% casein	-	12.45	77.86
4.	90% casein	-	14.76	92.26

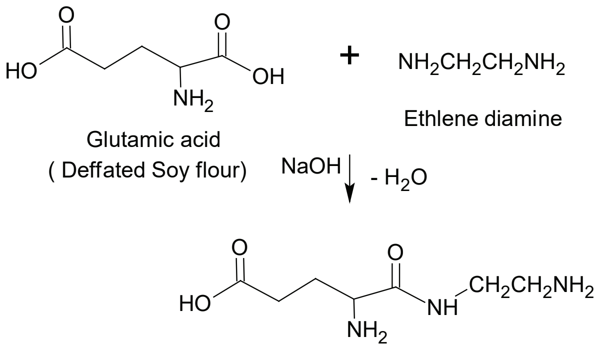

Figure 1 Probable scheme of defatted soy flour and ethylene diamine-based wood adhesive

Chemical resistance

The fixed wood joints were kept in cold water for one day and dried for one day at room temperature before measuring the tensile strength. Similarly, bonded joints were submerged in acidic pH 2 and alkaline pH 10, both at 80ºC, for 1 h (John & Joseph 1998). After drying, tensile strength was measured.

RESULTS AND DISCUSSION

Adhesive system was successfully formulated by mixing defatted soy flour, ethylene diamine and NaOH (Figure 1), which was characterised by infrared (IR) spectra (Figure 3). A band at 3411 cm^{-1} indicated N–H stretching in primary and secondary amine groups. A band around 2992–3000 cm^{-1} indicated O–H stretching in the carboxylic acid group. A band at 1738 cm^{-1} indicated C=O stretching in a carboxylic acid group. While a band at 1632 cm^{-1} indicated C=O stretching in the secondary amide group and band at 2854 cm^{-1} indicated C–H stretching vibration in the alkyl group.

Another adhesive system from casein, ethylene diamine and NaOH was also successfully formulated (Figure 2) and characterised by IR spectra (Figure 4). The N–H stretching in primary and secondary amine groups was indicated by a band at 3381 cm^{-1}. While a band at 1628 cm^{-1} represented C=O stretching in the secondary amide group and the band around 2853–2925 cm^{-1} represented C–H stretching vibration in the alkyl group.

The principal components in the wood adhesive, defatted soy flour and casein, are derived from natural products indicating that the synthesised adhesives are eco-friendly. The effects of NaOH and ethylene diamine on defatted soy flour-based wood adhesives was shown in Table 2.

Wood adhesive prepared from defatted soy flour with NaOH gave better tensile strength than defatted soy flour with ethylene diamine. The effects of NaOH and ethylene diamine on different percentages (60%, 75% and 90%) of casein-based wood adhesives were shown in Table 3. Wood adhesives prepared from casein with ethylene diamine gave higher tensile strength than wood adhesives prepared from casein with NaOH at room temperature. The adhesives prepared from casein with NaOH produced a two-pack system which made it difficult to apply to wood species. Wood adhesives prepared from casein, ethylene

Table 2 Effect of NaOH and ethylene diamine on defatted soy flour-based wood adhesives

Sr. no.	Formulation	Tensile strength ($kg\ cm^{-2}$)
1.	9 g defatted soy flour + 1 mL ethylene diamine + 25 mL water	Bond failed
2.	9 g defatted soy flour + 25 mL 1% NaOH	31.4
3.	9 g defatted soy flour + 1 mL ethylene diamine + 25 mL 1% NaOH	29.5

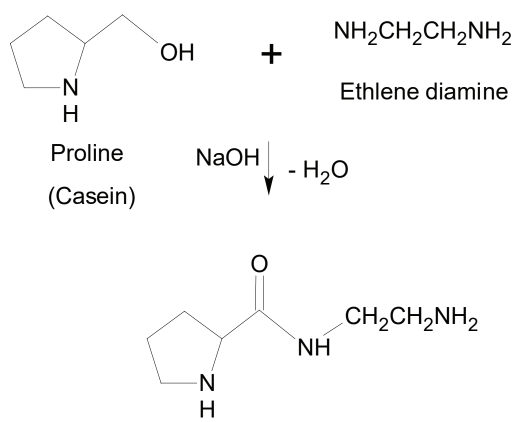

Figure 2 Probable scheme of casein and ethylene diamine-based wood adhesive

Table 3 Effect of NaOH and ethylene diamine on different percentages of casein-based wood adhesives

Sr. no.	Formulation	Tensile strength (kg cm^{-2})
1.	9 g 60% casein + 1 mL ethylene diamine + 25 mL water	40.5
2.	9 g 60% casein + 25 mL 1% NaOH	Two-pack system*
3.	9 g 60% casein + 1 mL ethylene diamine + 25 mL 1% NaOH	19.5
4.	9 g 75% casein + 1 mL ethylene diamine + 25 mL water	56.7
5.	9 g 75% casein + 25 mL 1% NaOH	Two-pack system*
6.	9 g 75% casein + 1 mL ethylene diamine + 25 mL 1% NaOH	25.5
7.	9 g 90% casein + 1 mL ethylene diamine + 25 mL water	83.4
8.	9 g 90% casein + 25 mL 1% NaOH	Two-pack system*
9.	9 g 90% casein + 1 mL ethylene diamine + 25 mL 1% NaOH	30.1

* = Cannot be applied to the wood species

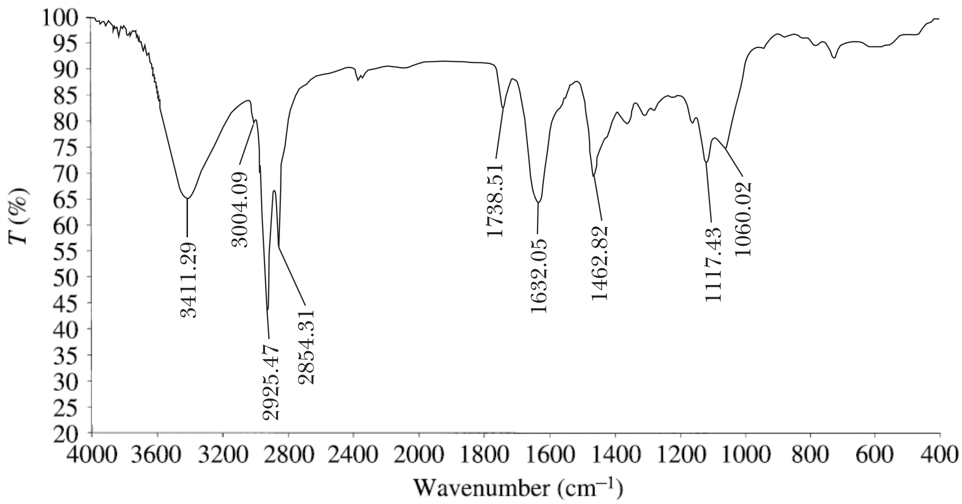

Figure 3 IR spectra of defatted soy flour and ethylene diamine-based wood adhesive

diamine and NaOH decreased tensile strength. It was also noted that as the percentage of casein increased in the wood adhesive, tensile strength also increased. The results indicated that wood adhesive prepared from 90% casein and ethylene diamine at room temperature gave the best results.

Strong acid and base, and cold water were used to treat bonded wood joints in order to examine the chemical resistance of the synthetic wood adhesives. The adhesives prepared from defatted soy flour with NaOH and ethylene diamine gave no chemical resistance. While wood adhesives prepared from casein (60%, 75% and 90%) with NaOH and ethylene diamine showed less chemical resistance compared to adhesives prepared from the different percentages of casein with ethylene diamine. Based on the results from this study, wood adhesive prepared from 90% casein with ethylene diamine exhibited excellent chemical resistance (Table 4).

CONCLUSION

Two different types of wood adhesive systems were successfully prepared, one from defatted soy flour, ethylene diamine and NaOH, and the other from casein, ethylene diamine and NaOH. Defatted soy flour and casein used to formulate the adhesives in this study are natural products. Thus, these types of wood adhesives are ecofriendly. Wood adhesive prepared from 90% casein and ethylene diamine gave the highest tensile strength at room temperature and showed excellent chemical resistance.

Table 4 Chemical and water resistances of wood adhesives

Sr. no.	Formulation	Alkaline resistance	Water resistance	Acid resistance
1.	Defatted soy flour + 1 mL ethylene diamine + 25 mL water	Bond failed	Bond failed	Bond failed
2.	Defatted soy flour + 1 mL ethylene diamine + 25 mL 1% NaOH	Bond failed	Bond failed	Bond failed
3.	Defatted soy flour + 25 mL 1% NaOH	Bond failed	Bond failed	Bond failed
4.	60% casein + 1 mL ethylene diamine + 25 mL water	24.9	40.5	21.1
5.	60% casein + 1 mL ethylene diamine + 25 mL 1% NaOH	10.2	19.5	12.4
6.	75% casein + 1 mL ethylene diamine + 25 mL water	43.1	56.7	38.3
7.	75% casein + 1 mL ethylene diamine + 25 mL 1% NaOH	14.2	25.5	16.1
8.	90% casein + 1 mL ethylene diamine + 25 mL water	65.2	83.4	60.8
9.	90% casein + 1 mL ethylene diamine + 25 mL 1% NaOH	20.4	30.1	23.9

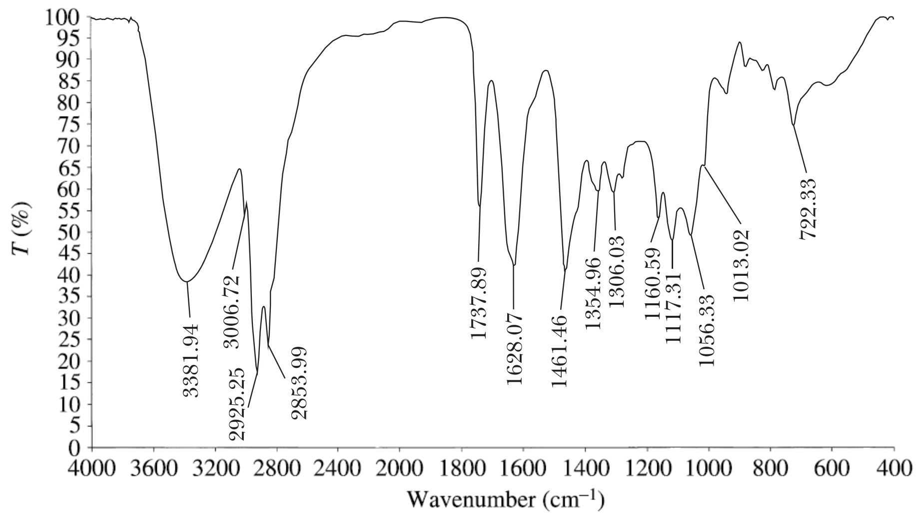

Figure 4 IR spectra of casein and ethylene diamine-based wood adhesive

Therefore, the formulation prepared from 90% casein and ethylene diamine was selected as the best ecofriendly wood adhesive for wood joints.

REFERENCES

Allen AJ, Marcinko JJ, Wagler TA & Sosnowick AJ. 2010. Investigations of the molecular interactions of soy-based adhesives. *Forest Products Journal* 60: 534–540. https://doi.org/10.13073/0015-7473-60.6.534.

Alma MH & Kelley SS. 2000. Thermal stability of novolak-type thermosettings made by the condensation of bark and phenol. *Polymer Degradation and Stability* 68: 413–418. https://doi.org/10.1016/S0141-3910(00)00029-X.

Andrews BAK, Meyer C & Reinhardt RM. 1986. *Formaldehyde Release from Wood Products.* American Chemical Society, Washington, D.C.

ASTM D897-08. 2016. *Standard Test Method for Tensile Properties of Adhesive Bonds.* American Society for Testing and Materials, ASTM International, West Conshohocken, Pennsylvania.

Hunt C, Wescott J & Lorenz L. 2009. Soy adhesive–moisture interactions. Pp 270–274 in Frihart CR, Hunt CG & Moon RJ (eds) *Proceedings of the International Conference on Wood Adhesives* 2009. 28–30 September 2009, Lake Tahoe, Nevada.

Indian Standards Is 548: Part I. 1964. *Methods of Sampling and Test for Oils and Fats.* Bureau of Indian Standards, New Delhi. https://standardsbis.bsbedge.com.

John N & Joseph R. 1998. Rubber solution adhesives for wood-to-wood bonding. *Journal of Applied Polymer Science* 68: 1185–1189. https://doi.org/10.1002/(SICI)1097-4628(19980516)68:7<1185::AID-APP15>3.0.CO;2-X.

Kim S. 2009. Environment-friendly adhesives for surface bonding of wood-based flooring using natural tannin to reduce formaldehyde and TVOC emission. *Bioresource Technology* 100: 744–748. https://doi.org/10.1016/j.biortech.2008.06.062.

Milota M & Wilson JB. 1985. Isocyanate-polyol resin as a binder for particleboard. *Forest Products Journal* 35: 44–48.

Nasir M, Gupta A, Beg MDH, Chua GK & Kumar A. 2014. Physical and mechanical properties of medium-density fiberboards using soy-lignin adhesives. *Journal of Tropical Forest Science* 26: 41–49. https://www.jstor.org/stable/23617012.

Patel DS, Toliwal SD & Patel JV. 2012. Eco-friendly adhesives based on tannin and N,N-bis(2-hydroxy-ethyl) fatty amides (HEFAs) from non-traditional oils for wood bonding. *Journal of Adhesion Science and Technology* 26: 2217–2227. https://doi.org/10.1163/156856111X610144.

Papadopoulou E, Nakos P, Tsiantzi S & Athanassiadou E. 2008. The challenge of bio-adhesives for the wood composite industries. In *Proceedings of the 9th Pacific Rim Bio-Based Composites Symposium.* 5–8 November 2008, Rotorua, New Zealand.

Petrie EM. 2007. *Handbook of Adhesives and Sealants.* Second edition. McGraw-Hill Co, New York.

Pizzi A & Mittal KL. 1994. *Handbook of Adhesive Technology.* Marcel Dekker, New York. ISBN 0-8247-0986-1.

Pizzi A & Mittal KL. 2011. *Wood Adhesives.* CRC Press, London.