WCTE 2016
World Conference on Timber Engineering
August 22-25, 2016 | Vienna, Austria

ENVIRONMENTALLY-FRIENDLY ADHESIVES FOR WOOD PRODUCTS USED IN CONSTRUCTION APPLICATIONS

Panagiotis Nakos[1], Christos Achelonoudis[2], Electra Papadopoulou[3], Eleftheria Athanassiadou[4], Emmanouil Karagiannidis[5]

ABSTRACT: The objective of this work was the development of environmentally-friendly, sustainable adhesive systems for the manufacture of engineered wood products (mainly glulam) to replace synthetic adhesives made from petrochemicals. Lignin-based adhesive systems were proven to provide industrial glulam products with performance comparable to the products produced with conventional gluing systems. The application of this innovative adhesives technology is expected to provide environmental and cost benefits and to lead to higher demand for glulam products when carbon neutral building solutions are sought.

KEYWORDS: cold-set wood adhesives, bio-based wood adhesives, lignin adhesives, glulam, laminated beams, structural panels, building construction

1 INTRODUCTION

Engineered wood products or panels are products dedicated for use in structural applications or else building construction. All of them are manufactured by combining wood strands, veneers, lumber or other wood elements with adhesive to form a larger composite structural unit [1]. They are designed and produced to maximize the natural strength and stiffness characteristics of wood by optimally orienting the wood veneers, strands or laminations and by combining wood with durable adhesives.

The evolution of engineered wood products has greatly expanded building options and methods in all forms of residential and commercial construction and offered environmentally-friendly, green, building products. According to CORRIM Life Cycle Assessment study, wood is better for the environment than steel or concrete in terms of embodied energy, global warming potential, air emissions, water emissions and solid waste production. Wood products require much less energy in manufacturing them compared to other industrial raw materials and act as carbon sinks.

Common engineered wood products represent plywood, oriented strand board, glulam, I-joists, structural composite lumber (SCL), a type of which is laminated veneer lumber (LVL). Specifically, glulam is one of the fastest growing structural materials of the last few decades. It is a type of structural engineered timber product comprising a number of layers of dimensioned timber bonded together with durable, moisture-resistant structural adhesives. The advantages of glulam towards convectional structural wooden elements may include the availability of standard components, versatility (can be manufactured in a wide range of shapes, sizes and configurations), no need for cladding, large spans, good strength-weight ratio, superior fire performances and corrosion resistance, while keeping the classic natural wood appearance. All these advantages come at a cost that is competitive with other structural systems [2].

Cross-laminated timber (CLT) is a new generation engineered wood product which has been gaining popularity in residential and non-residential applications since its introduction in the 1990s [3]. The superior attributes of the CLT panels such as good fire and seismic resistance, excellent sound and thermal insulation properties combined with ease of fabrication, quick assembly and many environmental advantages make CLT a preferred choice by designers and clients in some types of construction.

The adhesive is probably the most important parameter in the production of engineered wood products like the afore-mentioned described. The adhesives used today for the production of engineered wood products are still mostly based on synthetic resins, like phenolic, melamine, phenol-resorcinol, polyurethane and isocyanate adhesives. These resins are synthesised from petroleum and natural gas derived chemicals and therefore their prices are directly dependent on the fluctuation of the oil prices. Moreover, given the finite nature of the oil deposits, the long-term availability of petroleum-derived products is not guaranteed. Adhesives from renewable (non-petroleum) raw materials have a noteworthy role to play in relieving the reliance of wood products on petrochemicals and promoting the sector's sustainability.

[1] Panagiotis Nakos, CHIMAR HELLAS SA, office@ari.gr
[2] Christos Achelonoudis, CHIMAR HELLAS SA
[3] Electra Papadopoulou, CHIMAR HELLAS SA
[4] Eleftheria Athanassiadou, CHIMAR HELLAS SA
[5] Emmanouil Karagiannidis, CHIMAR HELLAS SA

Large quantities of renewable biomass materials and natural derivatives are available which can be converted into adhesives for wood panel products. The use of biomass as a source of chemicals and energy enables closed cycle materials fluxes and contributes to the efforts to reduce the atmospheric CO_2 emissions worldwide [4]. Several attempts have been made to replace or partially substitute the petrochemical adhesive components with bio-derived or naturally occurring materials in the preparation of adhesives for wood products [5]. The promising candidate bio-based adhesives should match the reactivity, applicability, bonding performance and cost requirements of the synthetic resins and outperform them in environmental acceptability and safety of use. Specifically, the new adhesives should satisfy the regulations concerning human health and safety and the environmental impact of construction materials, e.g. they should satisfy indoor air quality requirements by enabling the reduction of formaldehyde and other volatile organic compounds emissions at the level of natural wood and they should be less reliant on limited fossil chemicals.

2 EXPERIMENTAL

The R&D team of CHIMAR HELLAS has investigated the use of various biomass products like tannin, lignin, cellullose, starch, plant proteins, extraction-liquefaction-thermolysis products of forest and agriculture wastes and biomass treatment by-products in the production of adhesives for wood-based panels [5-10] with **the aims to:**

- reduce the demand on fossil fuels and promote sustainable development by using renewable raw materials for adhesives
- develop adhesives with the same or even enhanced bonding quality over conventional ones
- provide safe, emission-free wood products
- offer cost-effective solutions to the adhesive and wood panel industry.

Driven by the increasing need for the use of bio-based resins as natural and sustainable alternatives to traditional petrochemical adhesive materials, further R&D efforts were focused on bio-derived adhesives suitable for the production of engineered wood products like glulam, LVL and CLT.

The present paper discusses the development of lignin-based adhesive systems which can be effectively used in the production of engineered wood products with performance comparable to the products produced with conventional gluing systems. The successful performance of such adhesive systems was proven in pilot and industrial scale tests.

2.1 MATERIALS AND METHODS

The objective was to develop an adhesive with a green character capable of setting in cold pressing conditions in order to be used as binder for engineered wood products building materials. CHIMAR HELLAS R&D team developed and tested various binders based on natural raw materials so as to find the most promising ones for such an application. The various adhesive systems were evaluated at lab and pilot scale, in accordance with the prevailing European standards. The best performing adhesive system was further tested at industrial scale.

2.2 PLYWOOD PRODUCTION

Promising bio-based resin systems produced with the use of tannin or lignin, were prepared at CHIMAR lab. Adhesive formulations containing the said resins and suitable hardeners, fillers and occasionally crosslinkers were evaluated in the production of plywood panels under cold curing conditions. In particular, a phenol–formaldehyde-lignin (PFL) resin was prepared by replacing 50% of the phenol needed with lignin. Quebracho wood tannin was used in combination with either a hardener material or a hardener and a crosslinker. A conventional phenol–formaldehyde (PF) resin of the resole type was applied too as a reference.

All the above adhesive systems were applied in the production of 3-layer plywood panels of dimensions 50cm x 50cm at a glue mixture level of 150g/m^2. The plywood panels were prepared using spruce veneers of dimensions 50cm x 50cm x0.26cm and by following the conventional industrial practice for plywood production with the exception of application of cold pressing for about 48 hours.

Samples of the plywood panels produced were cut and prepared according to the EN314.1 standard and they were subsequently tested dry for their mechanical strength (shear strength and wood failure). The testing results are reported in the following Table 1 and represent average values from various series of tests and replications performed.

Table 1: *Lab plywood testing results*

Adhesive system	PF-hardener	Tannin-hardener	Tannin-crosslinker-hardener	PFL-hardener
Shear strength, N/mm^2	1.85	0.89	0.67	1.44
Wood failure, %	46	11	4	32

The results of Table 1 indicate that the most promising bio-based adhesive system is the combination of PFL resin and hardener. This system was selected for further application in pilot laminated timber production tests after modification of the hardener to enhance the performance of the resin.

2.3 PILOT LAMINATED TIMBER PRODUCTION

The adhesive system comprising PFL resin produced by 50% replacement of the phenol with lignin and a properly formulated resin hardener (of CHIMAR proprietary technology) was applied in the production of laminated timber structures at CHIMAR pilot facility. For this purpose, spruce lumber pieces of dimensions 50cm x 10cm x 0.9cm, 50cm x 4cm x 1.8cm and 50cm x 4cm x 2.7cm were used to prepare 2- and 3-lumber specimens. Gluing mixtures containing either PFL or reference PF resin and corresponding fillers were prepared. The glue mixtures containing the resins and the fillers were not mixed together with the hardener as done in the case of plywood panels. Instead, the glue mixtures were applied on one side of a timber piece via a brush, while the hardener was applied on one side of the next piece ("honeymoon" gluing). The average loading of the glue mixture used in these tests was 150-200g/m^2, while the hardener average loading was 80-100g/m^2.

After the application of both the glue mixture and the hardener, the timber pieces were attached to each other and pressed either with the help of hand clamps or in a cold press so as the treated timber surfaces to come in contact and the resin curing/hardening to be effected. The laminated timber structures that were formed were kept under continuous pressure at ambient temperature for a period of time of 16-20 hours. The pressure time depends on the season of the year and in summer season this time is reduced since the adhesive system would set/cure at a faster rate due to the higher prevailing ambient temperature. The use of the press instead of the hand clamps helped to control the specific pressure applied.

Once the pressing period ended, the press was opened and suitable samples were prepared from the produced laminated timber structures and tested for shear strength and percentage of wood failure or bending strength (modulus of rupture, MOR) and breaking point examination. The formaldehyde release of selected samples was determined too via the gas analysis method (EN 717-2). The testing results are reported in Table 2 and represent average values of the various tests and replications performed, while the laminated timber assembling, pressing and testing is shown in Pictures 1-4.

Table 2: *Pilot laminated timber testing results (average values)*

Adhesive system	PF-hardener	PFL-hardener
Shear strength, N/mm^2	4.2	4.0
Wood failure, %	100	88
MOR, N/mm^2	85	78
Breaking point	wood	wood
Formaldehyde release, mg/m^{2}h	0.80	0.33

Picture 1: *Laminated timber assembling*

Picture 2: *Laminated timber cold pressing*

Picture 3: *Testing of 3-lumber specimens (50 cm x 4cm x 2.7cm), 100% wood failure*

The results of Table 2 indicate that the bio-based PFL honeymoon adhesive system is a promising binder for the production of engineered wood products such as glulam, LVL and the like. The formaldehyde release of the laminated timber specimens was found to be in the E1 class, i.e. the lowest emission class as per EN 13986: 2015. The PFL adhesive system was hence promoted for application in industrial scale glulam tests.

Picture 4: *Laminated timber specimens after testing showing 0% and 100% wood failure*

2.4 INDUSTRIAL SCALE LAMINATED TIMBER PRODUCTION

Glued laminated timber (glulam) beams were produced at TECNIFUSTA plant, in Spain, using the developed PFL adhesive system. For this purpose, spruce timber beams of dimensions 220cm x 12cm x 5cm were used and the bio-based gluing mixture was prepared from the developed PFL resin and corresponding filler. The loading of the glue mixture used in these tests was 200g/m^2, while the hardener loading was 80g/m^2. The glue mixture and hardener were applied as a honeymoon system on the wood surfaces.

After the application of the glue mixture and the hardener, the beams were attached to each other and pressure was applied via hand clamps at ambient temperature for a period of time of 22 hours. The production tests were carried out in cold winter time, i.e. the site temperature was -10^{o}C.

Samples of the 3-layer glulam thus produced were tested on-site for bending strength (modulus of rupture, MOR) and percentage of wood failure. The testing results are presented in Table 3 representing average values of various test replications, while on-site glulam testing is shown in Picture 5.

It should be noted that the reference adhesive system in this case was a commercial melamine-urea-formaldehyde (MUF) resin and corresponding hardener, applied as honeymoon system too.

Table 3: *Industrial glulam testing results-winter time*

Adhesive system	MUF-hardener	PFL-hardener
MOR, N/mm^2	66	71
Wood failure, %	100	100

Further production tests were performed during early summer period when the ambient temperature was around 25^{o}C. The curing time of the PFL adhesive system as well as that of the reference MUF system were shorter and therefore pressing times of only 3 hours were realized. Samples of the glulam produced were tested for their bending strength performance and the corresponding testing results are given in Table 4, representing average values of the various replicate tests performed.

Picture 5: *PFL-bonded glulam testing at TECNIFUSTA, Spain*

Table 4: *Industrial glulam testing results-summer time, Spain*

Adhesive system	MUF-hardener	PFL-hardener
MOR, N/mm^2	50	61

At the same period of time, similar glulam production tests were performed at INWOOD plant, UK. The reference gluing system was a commercial polyurethane (PUR) adhesive system. A number of glulam beams were cured via pressing in a vertical structurally graded press, while timber clamps were used for some other test beams with the sole purpose of holding the lamella in place during curing. It was found that the same bonding effect was possible in either case and therefore the pressing way was not of any significance. The results from the testing of the bending strength of the glulam samples produced in these tests are given in Table 5 and represent average values of the tests performed.

Table 5: *Industrial glulam testing results-summer time, UK*

Adhesive system	PUR	PFL-hardener
MOR, N/mm^2	51	60

The results of the Tables 3-5 indicate that the developed bio-based adhesive system has acceptable performance in the production of glulam beams performing at the same or even at a higher level than conventional MUF or PUR bonding systems. Moreover, the pressure needed to effect the PFL resin curing is of low level, the application of this bio-based adhesive can be realised via a simple roller device and curing time is similar to commercially available MUF and PUR glues. Therefore, no capital investment is needed for the introduction of the bio-based PFL adhesive in the glulam conventional production facilities and gluing lines. A disadvantage traced by the industrial manufacturers, however, is the dark colour of the glulam glue lines produced with the PFL adhesive as opposed to the MUF or PUR glue lines which are clear.

Further pilot trials have recently been realized in a glulam manufacturer in Northern Greece with the objective to test the performance of the developed PFL adhesive according to the Delamination test of glue lines as per EN 391, which actually constitutes an accelerated ageing test method.

4-layer glulam was produced in this case using beams of dimensions 200cm x 15 cm x 2cm. The curing was effected via the use of hand clamps. The PFL adhesive was compared with a standard PF resin and a commercial MUF resin used in the plant's production. However, the EN 391 testing results of the glulam samples produced were not available at the time of writing of the present paper (Pictures 6-7).

Picture 6: *Spread PFL glue mixture on beam*

Picture 7: *Spread PF glue mixture on beam*

2.5 RESULTS AND DISCUSSION

It was found that a lignin-based adhesive system comprising resin and hardener was the most satisfactory one, fulfilling the standard requirements for glulam application. This adhesive resin is of the phenol–formaldehyde type where 50% of phenol has been replaced by lignin. This new lignin-based gluing system cures at room temperature within only a few hours and can be effectively used in the production of glulam beams and columns with a performance comparable to the products produced with conventional gluing systems. The resin and the hardener are used in a so called "honeymoon" gluing system. The process for the preparation of a gluing system comprises:

(a) a bio-based phenol-formaldehyde polymer where up to 50% of phenol can be replaced by lignin;
(b) a hardener to promote the cold-setting of this polymer.

This system displayed higher bonding performance compared with commercial melamine-urea-formaldehyde and polyurethane adhesives and is a more environmentally-friendly solution as it has half of the petrochemical phenol needed, having replaced it with lignin, which is a renewable biomass product. It should also be cheaper given that lignin is available as a residue from the paper manufacturing process and therefore could be cheaper than the oil-derived phenol.

The developed lignin-based adhesive is easy to apply without any modifications of the existing glulam production facilities and gluing lines. It should also be appropriate for application in the production of other engineered wood types like LVL, CLT.

Compared to other commercially available binders, the only restriction of the lignin-based cold curing adhesive system would be its darker colour. This could still represent a limitation factor in wider use in wood construction and in particular in glulam products, due to established architectural trends and aesthetic requirements set up by the sector professionals and customers. However, this bio-based binder can be suitable for outdoor construction applications and situations where colour effect does not jeopardize the design ideas.

3 CONCLUSIONS

A binder system comprising a cold setting PF resin modified with renewable lignin and a suitably formulated resin hardener was developed for application in the production of glulam beams with performance comparable to the products produced with conventional gluing systems. This bio-based binder should also be appropriate for application in the production of other engineered wood types like LVL, CLT.

ACKNOWLEDGEMENTS

Part of this work has been supported by the European Commission under the CIP Eco-innovation Framework Programme, Contract no: ECO/10/277331/SI2.598233 [11].

The authors would also like to acknowledge the important contribution of Mr. Marc Figueras of

TECNIFUSTA, Spain and of Mr. Edward Stenhouse of INWOOD, UK.

REFERENCES

[1] Engineered Wood Construction Guide. Form No. E30V, APA, 2011. www.apawood.org
[2] Glulam Product Guide. APA, 2008. www.apawood.org
[3] Mohammad M., Gagnon S., Douglas B.-K., Podesto P.E. L.: Introduction to Cross Laminated Timber. Wood Design Focus, V. 22, N. 2, 2012. http://www.forestprod.org/buy_publications/resources/untitled/summer2012/Volume%2022,%20Issue%202%20Mohammad.pdf
[4] Danner H., Braun R.: Biotechnology for the production of commodity chemicals from biomass: Chem soc Rev 28: 395-405, 1999.
[5] Borges J.C., Athanassiadou E., Tsiantzi S.: Bio-based resins for wood composites. In: ECOWOOD 2006-2nd International Conference on Environmentally-compatible Forest Products, Fernando Pessoa University, Oporto, Portugal, 2006.
[6] Markessini E., Tsiantzi S.: Bonding resins. International Patent Publication WO 00/23490, 1999.
[7] Papadopoulou E., Nakos P., Tsiantzi S., Athanassiadou E.: The Challenge of Bio-Adhesives for the Wood Composite Industries. In: 9th Pacific Rim Bio-Based Composites Symposium, Rotorua, N. Zealand, 2008.
[8] Papadopoulou E., Kountouras S., Sevastiadis T., Nikolaidou Z., Roumeli E., Chrissafis K.: Lignin utilisation as bonding material for plywood panels. COST Action FP1105 Workshop, Trabzon, Turkey, 2013.
[9] Nakos P.: Development of Lignin based glue, Final Workshop of European project CELLUWOOD, Catalan Institute of Wood, Lleida, Spain, 2014.
[10] Tseitlin A., Anderson C., Athanassiadou E., Markessini C., Salehpour S., Ireland D.: Advancement in Sustainable Resin Binders for Wood Composites using Engineered Bio-polymers, Forest Products Society 68th International Convention, Québec City, Canada, 2014.
[11] http://www.celluwood.com/Home/tabid/410/Default.aspx