

Thermal Insulation Panels with Bio-Based Adhesives

Valentina Vasilievna Strikun¹ Mikhail Andreevich Bajandin¹, Vladimir Nikolaevich Ermolin¹, and Zoltán Pásztor^{2}*

¹Reshetnev Siberian State University of Science and Technology, Krasnoyarsk Rabochy Av., Krasnoyarsk 660 037, Russian Federation

²University of Sopron, 4. Bajcsy Zs., Sopron 9400, Hungary, *E-mail address: pasztory.zoltan@uni-sopron.hu

Abstract. Environment friendly insulation panels were manufactured and tested made of pine wood fibers and glued with a bio-based adhesive, called dextran. The aim of this work was to determine relevant technical properties of panels fabricated with this new glue type. In the panels, the ratio of dry glue content was varied from 30% to 60% with 10% steps which runs served as comparison basis. The density of the insulation panels was set to 70 kg/m³, 100 kg/m³ and 125 kg/m³. Beside their thermal conductivity, the compression strength, the bonding strength, and the wetting angle of the adhesive were measured. With the variation of panel densities, the thermal conductivity is in a narrow range of 0.039 to 0.042 W/mK. The resistance to compression at 10% strain was measured to be 0.3 MPa, 0.35 MPa and 0.4 MPa in the panels with 70 kg/m³, 100 kg/m³ and 125 kg/m³, respectively. The wetting angle of the adhesives seemed to correlate only weakly with the bonding strength, and the glue's wetting ability diminished with the increase of the glue content. The results seem to be competitive if compared with the traditionally used glass and rockwool, and foam insulation materials.

1 Introduction

Generally, municipal and residential buildings are responsible for a high consumption of energy [1]. Their individual energy consumption depends strongly on both the climatic conditions and the insulation characteristics of the buildings. The most effective way to improve energy efficiency of buildings is to increase the thermal insulation of their cladding. In some geographic areas, the greenhouse gas reduction and lower energy dependence - and thus, lower building operation costs - are the driving forces for stricter building codes [2]. In order to increase the energy efficiency of edifices more insulation material needs to be used resulting lower operational cost and reduced carbon-dioxide emission. Also, the production, the transportation and the elimination of surplus insulation material emit significant amount of greenhouse gases. When environment protection is a determining factor then the issue of insulation material is of high relevance. Although foam as well as rock and fiberglass based materials have a good thermal performance – usually, at low market prices, their environment impairing effects are considerable during manufacturing and at final disposal. The recycling

of traditionally used foam, rock or glass wool insulation materials is still problematic, and often hazardous.

In the last decades, the research and the production of insulation panels made of natural raw materials, specifically lignin and cellulose, have come into the focus [3, 4]. Yet, the production of raw materials and of the adhesives pollute the environment. Because significant volume of adhesives is being used one of the most important goals was to lower formaldehyde emission [5]. More comprehensive works were published in the recent years concerning bio-based adhesives [6, 7,]. One of the interesting investigations deals with the activation of lignin within the constituting particles of the panels as an adhesive [8, 9]. Modified lignin is a suitable source material for adhesives with enhanced bonding properties [10, 11, 12, 13]. Norström et al. [14] have investigated a gum dispersion while Santoni and Pizzo studied vegetable protein as a possible wood adhesive [15]. A hydrophilic, eco-friendly bio-based adhesive was produced by Yuan et al. [16], and a mimosa tannin-based adhesive by Zhank et al. [17]. Although, these research endeavors have been conducted for longer than a decade no break-through could be realized on the market.

Organic fluids have been studied as a possible alternative for adhesives, having obtained from cultivated molasses. They contain the macro-molecular substances eventually yielding dextran. Dextran is a polymer – with a mass more than 15 kilodaltons - having highly branching molecules which explains its gluing ability. The number and the positions of the branch connections and the length of the side chains significantly determine the adhesive's strength. Besides the strong adhesive capacity, it is non-toxic, thus completely safe for humans, and entirely biodegradable. Earlier studies on particle boards using organic liquids as a binder indicate the potential use of this type of glue in chipboard production [18]. Hosseinpourpia [19] reached at similar conclusions when studying fiberboards manufactured with the adhesive dextran. Presently, the use of carbohydrates-based adhesives (modified starch, dextrin) in the production of particle boards is being widely studied and seem to harbor promising prospects [20].

The overall objectives of the present research are hence 1) to investigate the wetting angle in dependence of the dry content of dextran, 2) to determine the correlation between dry content of the bio-based dextran adhesives and their bonding strength, 3) and to measure the resistance to compressive forces at 10% strain with different densities of the panels, 4) to determine the thermal conductivity of the insulation panels as manufactured with different densities.

2 Materials and methods

2.1 Sample preparation

Dark brown, organic aqueous solution of the glue was used for this investigation. In the initial stage, the physico-chemical characteristics of the adhesive's interaction with wood fibers have been studied. Subsequently, the efforts were concentrated on the manner of fabrication of the thermal insulating boards to arrive at the necessary technical characteristics. The dextran was produced by cultivating the bacterium *Leuconostoc mesenteroides*. Eventually, the cultivation stock was dried and ground to a fine powder. From this powder, aqueous solutions were prepared containing 30%, 40%, 50%, 60% dry glue powder by weight ratio. This liquid was used as adhesive and added to the fiber in 30% weight related to the dry fiber and were mixed. The matt were formed in a tray having a rectangular shape panel with constant thickness and parallel surfaces. In this manner, a series of insulation panels with differing densities were produced. Eight samples were manufactured for each type of dry glue content The thermal insulation panels were produced from fibers of Siberian pine (*Pinus*

sibirica) with a moisture content of 2-4%, donated by the MDF Factory in Krasnoyarsk region of Russia. The glue was sprinkled onto the rotating fiber mass by a pneumatic mixer. The mat was formed manually, and by using a laboratory press panels with a surface size of 400 mm by 400 mm and a thickness of 25 mm were produced (Fig. 1). The mat was subjected to a pressure of 0.2 MPa for five minutes without heating. Because of the cold pressing, no vapor overpressure had built up inside the panel, and thus the press could be opened without pressure reduction steps. Thereafter the panels were placed into a drying chamber with air circulation and exposed to 105°C for 24 hours.



Fig. 1. Insulation panel with dextran adhesive

2.2 Experimental setup for wetting angle measurement in dependence of dry glue content

Wetting behavior of the glue can significantly influence the strength of the bond. For this reason, the wetting angle of glue was studied with different dry contents. For the measurements, a glue was dropped with a glass rod onto the planed surface of a solid pine board near its edge. A reconstructed MIS-11 microscope was used to determine the wetting angle. The diameter and the height of the glue drop were determined with an ocular micrometer. The angle of the wetting edge was calculated by the formula 1:

$$\cos X = (r^2 - h^2) / (r^2 + h^2) \quad (1)$$

where, X is the wetting angle in degree, r is the radius and h the height of the droplet, both in μm . 50 tests were carried out for every dry content ratio of the glue, and the usual statistics calculated.

2.3 Compression strength

Compression strength tests were carried out on the panels as an indication of the adhesive's bonding strength. To this procedure, the 25 mm thick samples were cut to a surface size of 100 mm x 100 mm. 20 repetitions were prepared for each glue content variation. The results

were calculated as suggested by the standard GOST 17177-94 [21] according to the equation 2:

$$\sigma_{10} = P/lb \quad (2)$$

where σ_{10} is the compression stress level in MPa at the linear strain of 10%, P is the load in Newton; l sample length and b sample width, both in mm.

2.4 Measurement of the panel's stress level at 10% strain in dependence of dry glue content

Panels have been tested to study the effect of the dry glue content on the stress level at 10% strain. To this goal, eight pieces of five-layer plywood samples were prepared according to the standard EN 314-1:2004 [22] for all four dry content ratios of the glue. Prior to testing, the samples have been stored in the climate chamber until they have reached the required moisture content of 12%. Altogether, thirty-two panel samples were tested. Compression strength tests were carried out by using a universal testing machine USN-30 with a measurement accuracy of 0.01 Newton, and 0.01 mm.

2.5 Measurement of thermal conductivity of the panels

For the determination of the thermal conductivity coefficient, the 25 mm thick panels samples were cut to the dimensions of the measuring device's working surface of 150 mm by 150 mm. The tests were carried out according to the requirements of the standard GOST 7076-99 [23] at steady state condition and an ambient temperature of 22°C and 65% relative humidity, respectively. 20 samples were cut out from the pressed panels for each variation of density and dry content of the glue. The results were calculated as the average of the 20 individual results.

3 Results and discussion

3.1 Wetting angle measurement in dependence of dry glue content

In Table1, the data for the equilibrium wetting angles represent mean values of 50 measurements. The values in parentheses (standard deviations) indicate that the angles significantly differ from each other, that the dry glue content alters indeed the interaction with the wood fibres.

Table 1. Equilibrium wetting angle of bio-based adhesive with different dry contents

Type of substrate	Dry glue content, [%]	Radius of the droplet, [µm]	Height of droplet, [µm]	Equilibrium wetting angle, [°]
Coniferous wood (Siberian pine fibres)	30	158 (±2.162)	75 (±1.969)	50.785 (±1.266)
	40	175 (±1.871)	91 (±0.700)	52.345 (±0.531)
	50	211 (±2.462)	114 (±1.256)	55.089 (±0.772)
	60	241 (±2.370)	131 (±2.118)	57.054 (±0.817)

As the dry content of the glue increases the wetting angle rises too which means the wetting ability is decreasing. According to Fig. 2, the interdependence seems to be linear. The wetting ability of the glue with 30% dry content is the highest as indicated by the wetting angle (when 180° stands for water-repellence and 0° for full solubility). Yet, when the dry content is doubled from 30% to 60% the wetting angle increases only slightly, by about 6 + degrees. Thus, the increasing dry glue content makes the panels only marginally more hydrophilic.

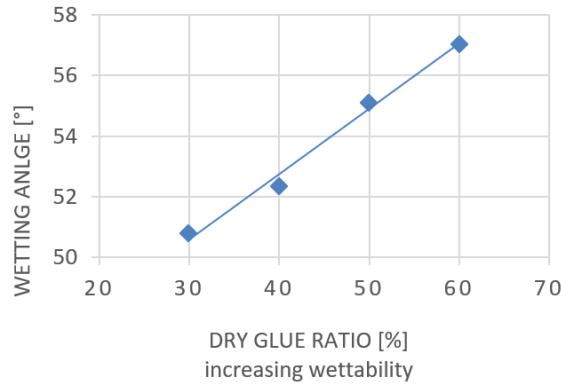


Fig 2. Wetting angle [deg] in function of dry glue content [%]

3.2 The effect of dry content of glue on the adhesive bond strength

The data in Fig. 3 shows the average data of the 20 samples from the 100 kg/m³ density panel according to the dry content of the adhesive.

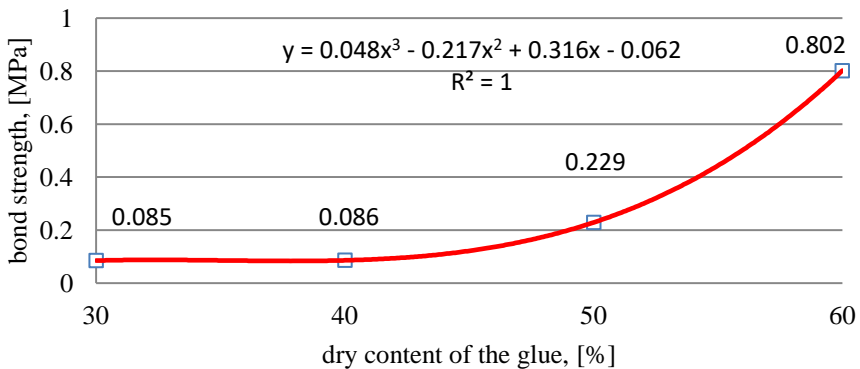


Fig. 3. Influence of the dry content of glue on tensile

The fig. 3 shows that the panel strength starts to increase as from 40% dry glue content. The steepness of the function line increases with higher dry glue content. The higher polymer content of the glue shows the increased adhesion ability of the solution. It should be noted that the high content of dry component increases not only its adhesive ability, but also simultaneously increases the viscosity of the glue, which made the process of wetting of wood fibers more difficult. Wood is a porous material full of capillaries, and when glue can intrude into these voids, the bonding strength increases. The amount of moisture absorbed by the wood depends on both the initial water content of the glue and the moisture content of the fibers. Accordingly, with low dry glue content more water will be absorbed by the panel.

That increases the concentration of dry substances on a surface of the fiber particles and contributes to reach strong bonding connection. Despite of this theory the results showed that the glue with more than 50% dry content shows a significant bonding strength.

3.3 Measurement of the resistance against compressibility

The mechanical compressibility of the panels is an important information for the builder. The resistance to mechanical compressibility of the panels were measured at 10% of the strain as derived from the data in [Table 2](#).

Table 2. Compression strength at 10% compression

	Panel density		
	70 kg/m ³	100 kg/m ³	125 kg/m ³
Stress at 10% strain [MPa]	0.030 (±0.002)	0.035 (±0.004)	0.040 (±0.003)

3.4 Thermal conductivity

Intuitively, a strong interdependence between thermal conductivity and resistance against mechanical compressibility of the panels is presumed. According to our results, however, the thermal conductivity of the panels seemed to increase significantly while the stress at 10% strain level rose only marginally from one density step to the next. When the panel density changed from 70 kg/m³ to 125 kg/m³ the thermal conductivity was enhanced by merely 0.003 W/mK (Table 3). In contrast, the compression stress at 10% strain increased by about one third while the panel density nearly doubled from 70 kg/m³ to 125 kg/m³. It is a significant increase indeed as is shown in Fig. 4.

Table 3. Properties of thermal-insulating panels using 30% dry content of glue

	Panels density		
	70 kg/m ³	100 kg/m ³	125 kg/m ³
Thermal conductivity [W/mK]	0.039 (±0.003)	0.040 (±0.004)	0.042 (±0.003)

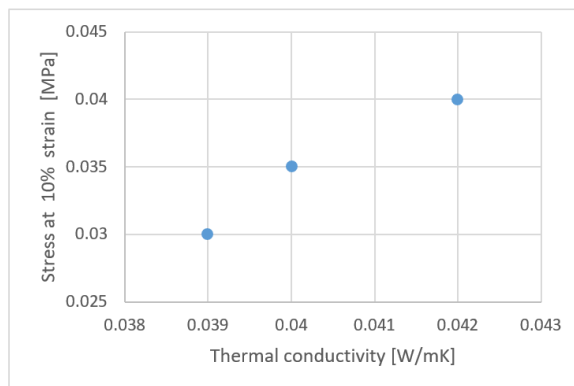


Fig. 4. Interdependence between thermal conductivity and 10% mechanical compressibility

4 Conclusion

The overall goal of this study was to investigate the different parameters of thermal insulation panels made of wood fiber and dextran as a bio-based glue, at varying dry glue contents. The results showed the following conclusions:

- 1) The dry content of bio-based adhesive significantly affects the bonding strength in the insulation panels. Generally, the higher the dry content the higher is the panel's bonding strength which seems non-linearly increase with the change of dry glue content from 30% to 60%.
- 2) By augmenting the dry glue content in the panel, the wetting angle rises only slightly. The angle increase is small, about 6°, while the glue dry content doubles from 30% to 60%.
- 3) Also, the thermal conductivity has increased by a small value while the panel density changed from 70 kg/m³ to 125 kg/m³.
- 4) Resistance against mechanical compressibility increased moderately with the increased density of the insulation panels.

Dextran is a suitable adhesive for producing sustainable fiber-based insulation panels, with higher dry content of at least 50% or rather 60%. The wetting angle is less important than it was supposed. The higher the dry content, the higher the wetting angle, although the bonding strength will be higher also. The results indicate that this bio-based adhesive is suitable for producing environment friendlier insulation material than most of them on the market. It has more advantageous: harmless the environment during manufacturing process, provides healthier living conditions because no VOC emission, and after life cycle can be reuse or recycle without any environment damage. The expected life cycle of this new insulation material glued with bio-based adhesive can be the same than that of the normal glass wool or polystyrene insulations. Beside these facts the thermal conductivity is almost the same than that of the commercialized insulation materials and the 10% compression strength is high enough for using under floor. In the future studies are planned to determine the optimal size of wood fiber used as a filler. For the practical use of this thermal insulation material, there must be research on its fire resistance and durability. Secondly it would be important to estimate the production cost and the carbon dioxide equivalence and compare to the competing fiber-based insulation already in the market.

Acknowledgement

This article was made in frame of the project TKP2021-NKTA-43 which has been implemented with the support provided by the Ministry of Innovation and Technology of Hungary (successor: Ministry of Culture and Innovation of Hungary) from the National Research, Development and Innovation Fund, financed under the TKP2021-NKTA funding scheme.

References

1. Schiavoni, S., D'Alessandro, F., Bianchi, F., Asdrubali, F. Insulation materials for the building sector: A review and comparative analysis. *Renew. Sust. Energ.* 62, 988-1011. (2016)
2. DIRECTIVE 2010/31/EU of the European Parliament and of the Council of 19 May 2010 on the energy performance of buildings.

3. Mimini, V., Kabrelian, V., Fackler, K., Hettegger, H., Potthast, A., Rosenau, T. Lignin-based foams as insulation materials: a review. *Holzforschung*, **73**, 117-130. (2019) <https://doi.org/10.1515/hf-2018-0111>
4. Jones, D. COST FP1303 "performance of bio-based building materials" FOREWORD. *Wood Material Science & Engineering*, **14**, 1-2. Special Issue (2019) <https://doi.org/10.1080/17480272.2018.1528569>
5. Mantanis, G.I., Athanassiadou, E.T., Barbu, M.C., Wijnendaele, K. Adhesive systems used in the European particleboard, MDF and OSB industries. *Wood Material Science & Engineering*, **13**, 104-116. (2018) <https://doi.org/10.1080/17480272.2017.1396622>
6. Zhongqi, He. *Bio-based Wood Adhesives, Preparation, Characterization, and Testing*. Taylor & Francis Group; (2017)
7. Gadhav, R.V., Mahanwar, P.A., Gadekar, P.T. Starch-Based Adhesives for Wood/Wood Composite Bonding: Review. *Open Journal of Polymer Chemistry*, **7**, 19-32. (2017) <https://doi.org/10.4236/ojpcem.2017.72002>
8. Domínguez-Roblesa, J., Tarré, Q., Delgado-Aguilar, M., Rodríguez, A., Espinach, F. X., Mutjé, P. Approaching a new generation of fiberboards taking advantage of selflignin as green adhesive. *International Journal of Biological Macromolecules*, **108**, 927–935. (2018) <https://doi.org/10.1016/j.ijbiomac.2017.11.005>
9. Ghaffar, H.S., Fan, M. Lignin in straw and its applications as an adhesive. *International Journal of Adhesion & Adhesives*, **48**, 92–101. (2014) <http://dx.doi.org/10.1016/j.ijadhadh.2013.09.001>
10. Ibrahim, V., Mamo, G., Gustafsson, P.J., Hatti-Kaul, R. Production and properties of adhesives formulated from laccase modified Kraft lignin. *Industrial Crops and Products*, **45**, 343– 348. (2013) <http://dx.doi.org/10.1016/j.indcrop.2012.12.051>
11. Ji, X., Guo, M. Preparation and properties of a chitosan-lignin wood adhesive. *International Journal of Adhesion and Adhesives*, **82**, 8–13. (2018) <https://doi.org/10.1016/j.ijadhadh.2017.12.005>
12. Jin, Y., Cheng, X., Zheng, Z. Preparation and characterization of phenol–formaldehyde adhesives modified with enzymatic hydrolysis lignin. *Bioresource Technology*, **101**, 2046–2048. (2010) <https://doi.org/10.1016/j.biortech.2009.09.085>
13. Pradyawong, S., Qi, G., Li, N., Sun, X. S., Wang, D. Adhesion properties of soy protein adhesives enhanced by biomass lignin. *International Journal of Adhesion and Adhesives*, **75**: 66–73. (2017) <http://dx.doi.org/10.1016/j.ijadhadh.2017.02.017>
14. Norström, E., Fogelström, L., Nordqvist, P., Khabbaz, F., Malmström, E. Gum dispersions as environmentally friendly wood adhesives. *Industrial Crops and Products*, **52**, 736– 744. (2014). <http://dx.doi.org/10.1016/j.indcrop.2013.12.001>
15. Santoni, I., Pizzo, B. Evaluation of alternative vegetable proteins as wood adhesives. *Industrial Crops and Products*, **45**, 148– 154. (2013) <http://dx.doi.org/10.1016/j.indcrop.2012.12.016>
16. Yuan, C., Chen, M., Luo, J., Li, X., Gao, Q., Li, J. A novel water-based process produces eco-friendly bio-adhesive made from green cross-linked soybean soluble polysaccharide and soy protein. *Carbohydrate Polymers*, **169**, 417–425. (2017) <http://dx.doi.org/10.1016/j.carbpol.2017.04.058>
17. Zhang, J., Xiong, L., Zhou, X., Du, G. Development of Mimosa Tannin-Based Adhesive Cross-Linked By Furfuryl Alcohol-Formaldehyde and Epoxy Resins. *Wood Research*, **64**, 847-858. (2019)

18. Pashkin, S.V., Ivanova, M. A., Shchekolov, V. M. Innovative technologies for producing environmentally friendly glued materials based on water-soluble bio-glue from low-grade wood. *Lesnoj vestnik (Forest Bulletin)*, **8**, 59-63. (2012)
19. Hosseinpourpia, R., Adamopoulos, S., Mai, C., Taghiyari, H. R. Properties of Medium-Density Fibreboards Bonded With Dextrin-Based Wood Adhesive, *Wood Research*, **64**, 185-194. (2019)
20. Bajandin, M.A. Chipboard on a binder modified dextran. / M.A. Bajandin // *Aktual'nye napravleniy anauchny hissledovaniy XXI veka: teoriyaipraktika* (Current research areas of the XXI century: theory and practice). 13-1, 355-359. (2015)
21. GOST 17177-94 (1996): Материалы и изделия строительные теплоизоляционные. Методы испытаний (Thermal insulating materials and products for building application. Test methods)
22. EN 314-1:2004 Plywood. Bonding quality Test methods, the standard is in harmony with GOST 15613.1-84 (1984): ГОСТ 15613.1-84 Девесина клееная массивная. Методы определения предела прочности клеевого соединения при скалывании вдоль волокон, (Gluedmassivewood. Methods for determination of glued joint of ultimate strength while shearing along grain)
23. GOST 7076-99 (1999): ГОСТ 7076-99. Материалы и изделия строительные. Метод определения теплопроводности и термического сопротивления при стационарном тепловом режиме, (Buildingmaterialsandproducts. Method of determination of steady-state thermal conductivity and thermal resistance)