



## International Conference on Wood Adhesives

May 11-13, 2022  
Portland, OR, USA

### Oral Presentation Abstracts

**Wednesday, May 11, 2022**

#### Progress in Established Adhesives I

**Paschalis Tsirogiannis**

Chimar Hellas S.A.

##### ***High Performance Formaldehyde-Based Adhesives: How Was the Progress Achieved?***

Thermosetting formaldehyde-based resins are the main binders used in today's industrial production of wood-based panels. They are derived by the condensation polymerization of formaldehyde with various monomers, and they are produced and applied in the form of aqueous suspensions of oligomers. The co-reactant or combination of co-reactants used with formaldehyde is selected depending on the cost, production conditions, and expected performance of the target panel product.

These types of adhesives were used in the industry since mid of 20th century due to their cost effectiveness and satisfactory performance. Cost is actually a critical issue on the choice of the adhesive, given the weight percentage of the adhesive in the wood panel product (2-10%).

Over the years, formaldehyde adhesives for wood panels have undergone through stages of development, evolving into high performance systems, with the ability to meet even the most stringent performance standards for composite wood panels at no sacrifice of production cost.

This presentation sheds light on the evolution of formaldehyde-based adhesive systems, comprising formaldehyde resins and their additives, which is a result of various parameters of the wood panel industry, e.g. the continuous pressure to reduce costs and withstand the strong market competition, the drive to develop products for new, more demanding applications and the need to protect the health of consumers from potential threats like the exposure to formaldehyde.

**Byung-Dae Park**

Kyungpook National University

##### ***An Old Adhesive, But New Story on Urea-Formaldehyde Resins in Wood Bonding***

Current low formaldehyde/urea (F/U) molar ratio (LMR) urea-formaldehyde (UF) resins for wood bonding are quite different from those used in thirty years ago. In general, LMR UF resins emit less formaldehyde at the expense of the resin reactivity and adhesion performance in wood-based composites. One of the main reasons of a poor adhesion of the LMR resins is the formation of crystalline

domains in liquid and cured UF resins via hydrogen bonds between linear molecules, which hinders their cross-linking in building three-dimensional networks. This presentation provides an overview of UF resins, and reports new innovative approaches of transforming the crystalline LMR resins into amorphous ones with modified-nanoclay, multi-reactive melamine, or dialdehyde cellulose (DAC), which improves the formaldehyde emission and adhesion at the same time. X-ray synchrotron showed that the crystalline domains isolated from LMR resins are isotropic crystals with random orientation. FTIR, <sup>1</sup>H-, and <sup>13</sup>C-NMR spectra confirmed the formation of hydrogen bonds between linear molecules in LMR resins. All LMR resins modified with nano-clay, melamine, or DAC become amorphous resins with an increased cross-linking density. These results suggested that the modification of UF resins with the nanoclay, melamine and DAC converted the crystalline domains to amorphous ones, leading to an improved cohesion strength and consequent adhesion.

### **Minjeong Kim**

Department of Wood and Paper Science, Kyungpook National University, Daegu, 41566, Republic of Korea

#### ***Effects of Molecular Weight of Urea-Formaldehyde Resins on Wettability and Adhesion of Wood Surface, Interphase, and Plywood***

This paper present effects of the molecular weight (MW) of urea-formaldehyde (UF) resins on wettability and adhesion of the resins for wood surface, interphase and plywood adhesion. Two formaldehyde/urea (F/U) molar ratios (1.6 and 1.0) of UF resins are synthesized to have three MW levels such as low MW (LMW) with the 'D-E' viscosity scale, medium MW (MMW) with the 'J-K' viscosity scale, and high MW (HMW) with the 'S-T' viscosity scale. These resins' surface tension, contact angle, surface adhesion, interphase adhesion and plywood adhesion are compared for early-wood and late-wood from either sapwood or heartwood before and after the removal of extractives. As expected, the molecular weights and curing peak temperatures of these resins increase and decrease as the viscosity increases while the 1.6 molar ratio resins have greater molecular weights and low curing temperatures than those of 1.0 molar ratio resins. The early-wood shows greater surface roughness than that of late-wood, regardless of sapwood and heartwood. The extractive removal resulted in an increase in the early-wood surface roughness, leading to lower contact angle and surface adhesion, regardless of the molar ratio, sapwood, or heartwood. However, the extractive removal causes a slight decrease in the surface roughness, resulting in an increase in the contact angle and decrease in the surface adhesion for most of sapwood and heartwood. As an indicator of interphase adhesion, the maximum storage modulus (E<sub>max</sub>) is highest at the HMW of 1.0 resins while the MMW of 1.6 resins show the highest. Tensile shear strength of plywood increases as the MW of two molar ratios of UF resins increases. These results suggest that the effects of MW of UF resins are not directly correlated with the wettability, surface adhesion, interphase adhesion, and plywood adhesion.

### **Luisa Hora de Carvalho**

Polytechnic Institute of Viseu

#### ***Impact of Urea-Formaldehyde Synthesis Parameters on Resin and Particleboard Properties***

This paper presents an overview about the studies carried out by our research group about the impact

of urea-formaldehyde synthesis parameters on resin and particleboard properties (PBs). In a first study, the chemical reactions and the most promising procedures for the preparation of UF resins, according to the alkaline-acid process, were analyzed. The main differences between the two procedures were the number of U loads and the methylation temperatures. The resins were characterized during the synthesis procedure by GPC/SEC, HPLC, <sup>13</sup>C-NMR and at the end by XRD. As usual, PBs were pressed and analyzed according to standard mechanical tests and Formaldehyde content. This study allowed to optimize the process and the properties of PBS produced. Using the best procedure, two crucial synthesis parameters were studied: condensation F/U molar ratio and U feed rate. The differences in the polymeric structures and the performance of new resins on the particleboards (PBs) properties were analyzed and compared with a standard resin. The molecular weight distribution of the resins was monitored by GPC/SEC and the unreacted oligomers by HPLC. The PBs produced were analyzed following European standards for physico-mechanical tests and formaldehyde emission. The data obtained were also submitted to a statistical analysis. Additionally, the effect of resin ageing was also studied and the PBs prepared using the best resin upon fifteen days of its production presented similar IB values and even lower F content in relation to those obtained using fresh resin.

#### Acknowledgements

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#### **Simon Aicher**

University of Stuttgart, Materials Testing Institute, Dept. Timber Constructions

#### ***Strength and Duration of Load Behavior of Butt Bonded Spruce Boards***

Strength and duration of load behavior of butt bonded spruce boards

It states a widely prevailing opinion that butt bonding of wooden boards, i.e. adhesive-based end grain joints represent a senseless, inefficient and unreliable timber jointing technology as opposed to bonding of wooden components parallel or slightly inclined to fiber direction. Hence, up to today world wide no structural wooden element / construction relying on adhesively butt jointed boards is known. Except hereof are a few CLT plates butt bonded at the narrow edges using the so-called TS3 technology which has been reported previously in rather generalized manner, e.g. WCTE 2016. In order to introduce this paradigm shift in timber bonding technology into construction reality, then accepted by building authorities and design codes, TS3 company has commissioned MPA University of Stuttgart to propose, initiate and conduct experimental and theoretical investigations altogether with expertises deemed necessary to obtain a German approval and a European Technical Assessment (ETA).

In the context of this demanding task numerous tests were performed and on-going. The investigations address the pure adhesive and the adhesive-primer-wood interface, jointed boards and large scale butt bonded CLT plates. Hereby different mechanical loading modes, varying climates, extraordinary moisture and load impacts and extensive duration of load (DOL) tests are performed. To enable the experimental campaign two new large test sheds had to be built at the University Campus.

This paper reports firstly on several aspects of the adhesive and adhesive/primer interface tests, highlighting bond line features of the specifically employed 2component Polyurethane adhesive used in-service with bond line thicknesses of 4 to 20 mm. Secondly, the experimental campaign conducted at the board level is revealed. Short term and DOL results for tension, bending and shear loading are presented altogether with proposals for the design relevant characteristic strength values and respective DOL modification factors.

## **Novel Resins I**

**Konrad Kwiecien**

AGH University of Science and Technology

### ***PUFJ and FRPU Innovative Polyurethane Bonding Systems for Wood Envelopes in Nearly Zero***

#### ***Energy Buildings***

Polyurethane Flexible Joints (PUFJ) and Fiber Reinforced PolyUrethanes (FRPU) have been

manifested their high efficiency in bonding and strengthening of structural elements for last years, assuring simultaneous transfer of high loads and high deformations. They can be used in structural and non structural bonding of various structural materials as concrete, masonry and wood. Many research and in situ applications confirmed they innovative and unique characteristics in civil engineering. PUFJ and FRPU systems are considering now as testing products in frame of a MEZeroEproject (MEZeroE - measuring envelope products and systems contributing to the next generation of healthy nearly-zero-energy buildings) funded by European Commission in Horizon 2020 program.

PUFJ and FRPU are proposed as new structural and non structural bonding systems or joints in wood constructions, mainly in envelopes of nearly-zero-energy buildings. PUFJ system can be applied as injection or prefabricated laminates creating wood-to-wood, wood-to-concrete or wood-to-masonry bonds. They can form thin or thick flexible adhesive bondlines with adjusted properties. FRPU system is dedicated quick repair intervention or strengthening as it uses high strength fibers and flexible polyurethane matrix for externally bonded composites. Special polyurethanes using in PUFJ and FRPU can work safely under various loads (strong winds, earthquakes) and manifest stable properties in elevated temperatures. They are dedicated to bonding of CLT panels, where they can replace steel connectors, providing thermal and humid problems and pure resistance to cyclic loads.

In the paper, properties of PUFJ and FRPU systems are presented with examples of connections, planned to be tested at Cracow University of Technology in a Pilot Measurement and Validation Line (PM&VL) of the MEZeroE project. Presented connection solutions bond wood elements with wood, concrete and masonry elements, providing advantageous hydro-thermal properties.

**Ning Yan**

University of Toronto

### ***Novel Flame-Retardant Formaldehyde-Free Wood Adhesive Platform Technology Using Vanillin as the Starting Material***

A novel vanillin-based phosphorus-containing building block was developed as a sustainable platform biomolecule for synthesizing flame-retardant formaldehyde-free wood adhesives based on epoxy and polyurethane chemistries. Structural characterizations confirmed the successful functionalization of the organophosphorus moiety within the molecular structure. Both epoxy and polyurethane adhesives had shown excellent dry, wet, and boiling bond strengths for wood. In particular, the blend of polyurethane and epoxy resins demonstrated superior flame-retarding performances with a self-extinguishing flame response. The optimum blend also achieved the highest bonding strength, the maximum Limited Oxygen Index value, and the lowest heat release rate in cone calorimetry tests due to the strong synergistic interpenetrating networks formed between the epoxy and PU macromolecules in the cured adhesive network. The GC-MS analysis of the char residues indicated that the mechanisms for flame retardancy were a combination of the quenching effect from the phosphorus-containing free radicals and the diluting effect of the nonflammable gases in the gas phase, plus the formation of phosphorus-rich char layers in the condensed phase. This research showcased a highly promising approach to develop environmentally friendly high-performance flame-retardant wood adhesives using nontoxic vanillin as the starting material.

#### **William Chirdon**

University of Louisiana at Lafayette

### ***Wood Adhesives from Algal Proteins***

Algal proteins are a common waste product from various bioprocesses, including the production of “green” fuels such as biodiesel, and as such, one of the major hurdles to the commercialization of bioprocessing technologies is the need for a technological solution for the utilization for this potentially massive volume of waste protein. To this end, algae glues have been developed, which have been found to serve as excellent binders for sugarcane bagasse and forest products. Through an alkaline denaturation, algal protein was converted to glue for wood and bagasse composites. The mechanical properties were assessed by a 3-point bending tests, and the flexural properties were found to exceed the minimum specifications for a standard grade 130 MDF (ASTM D 1037-06A) board. This technology has the potential to simultaneously address the need for waste protein co-product development that is absolutely critical for the emerging bioprocessing industry, while providing sustainably-sourced and formaldehyde-free adhesives for the wood composite industry.

#### **Charles R. Frihart**

Retired

### ***Ovalbumin; A Protein with Very Desirable Characteristics of a Wood Adhesive***

Egg proteins have not been investigated much as adhesives. Surprisingly, they have all the desirable properties for a very good wood bonding abilities, which a wide variety of plant proteins do have not. Although the whole egg has decent properties as an adhesive, the egg whites are the best because have the highest concentration of protein with few lipids and carbohydrates that are mainly in the egg yolks.

Much of our work has been done using the ovalbumin (OV) that can be readily isolated from the egg whites; the latter are also good wood adhesives. The most important is that is that the OV gives very good wet bond strength without any co-reactants or other additives, surpassing what is obtainable with the soy or other plant proteins. Literature indicates that OV reorganizes upon heating to create a hydrophobic surface causing aggregation of the proteins. The other important benefit is that the OV forms low viscosity dispersions, probably because there is low association of the individual proteins. The obvious drawbacks are cost, availability, and competition as human food source.

## **Progress in Established Adhesives II**

**Tunga Salthammer**

Fraunhofer WKI

### ***The Impact of Aldehydes and Organic Acids on Indoor Air Quality***

Data concerning the indoor air concentrations of formaldehyde, acetaldehyde, formic acid and acetic acid as well as data on air exchange were collected for the geographic regions of Europe and the United States of America. In Europe and especially Germany, these compounds are currently subject to new legislation and indoor air guidelines. The evaluation of current literature resulted in concentrations of formaldehyde within the range of 20  $\mu\text{g}/\text{m}^3$  to 30  $\mu\text{g}/\text{m}^3$  for European and US buildings under residential-typical conditions. A multitude of different permanent and temporary emission sources was identified. In addition to the typical building products, these also include chemical reactions occurring in indoor spaces, degradation of hemicelluloses, infiltrated outdoor air, combustion processes, the operation of equipment such as air purifiers and emissions from human activities such as cooking and cleaning.

For formaldehyde, the various emission sources were compared with one another under standardized conditions. It could be demonstrated that coatings and coverings drastically reduce the release of formaldehyde and other compounds into the room air. Overall, there was a distinction between permanent and temporary sources. It became clear here that formaldehyde peak concentrations are often caused by temporary sources, whose release potential for volatile organic compounds is not subject to any regulations whatsoever.

In view of the discussed aspects, the potential problem of exposure to formaldehyde in indoor areas can therefore not be solved through the further tightening of already existing regulations, in particular because peak concentrations and therefore high exposures would remain largely uninfluenced. This aspect is of considerable importance, as formaldehyde is a substance with a threshold effect. For this specific compound, the most appropriate risk management option would be to address the peak concentrations originating from temporary sources.

**Ryan A Gray**

Virginia Tech

### ***Rheology and Biopolymer Interactions in Phenolic Resin Formulations-Shear History Effects***

Structural wood-based biocomposites are crucial in constructing many homes in the United States. The

biocomposites are made using adhesive technologies based on the first known synthetic polymer system, phenol-formaldehyde thermosetting resins. These resins are formulated into adhesives with wheat flour and biomass fillers, such as corn-cob residue and walnut shell flour. The biomass fillers release polysaccharides, lignin, and various nonpolymeric phytochemicals. Consequently, these formulations are rheologically complex colloidal suspensions, and our group has previously shown that high shear treatments ( $\sim 4000 \text{ s}^{-1}$ ) stimulate the formation of liquid-state network structures. In addition, we found that these liquid-state network structures are unique to adhesives formulated with a corn-cob residue filler. Furthermore, the adhesive gel-state generated from these shear-induced network structures persist for periods equal to or greater than composite production times. Currently, the goal is to observe the effects of high shear spraying on the liquid structure of PF adhesives. Ultimately, we wish to determine if shear history represents a simple way to improve adhesive properties.

### **Jesse Paris**

Willamette Valley Company

#### ***Dynamic Rheology to Quantify Pre-Press Green-Strength (Tack)***

Adhesive pre-press tack is one of the key parameters that is controlled for in the manufacture of multiple wood-based composite products, including plywood and particleboard. Prepress tack effects process efficiency, raw material use and ultimate product quality. Although a wealth of applied knowledge, generated over the last several decades, has led to several effective tack control strategies, there are limited analytical tools available to characterize and quantify the subtle chemical and morphological changes that affect adhesive tack. Building on the state-of-the-art of adhesive tack measurement techniques, we at The Willamette Valley Company have developed a novel testing protocol that accurately measures real-time green strength development during the pre-press step. The evolution of various dynamic viscoelastic properties indicate how changes in the adhesive or glue-mix ingredients affect molecular scale interactions between wood and adhesives. This in turn provides an accurate predictor of the pre-press veneer and/or mat consolidation. In this work, we will present comparative analyses of glue-mixes with different extenders and fillers, and their effects on viscoelastic evolution of green strength and resulting tack.

### **Johannes Konnerth**

Wood K plus - Competence Center for Wood Composites & Wood Chemistry, Kompetenzzentrum Holz GmbH, Altenberger Straße 69, A-4040 Linz; Institute of Wood Technology and Renewable Materials, Department of Material Sciences and Process Engineering, BOKU University of Natural Resources and Applied Life Sciences-Vienna, Konard Lorenz Straße 24, 3430 Tulln, Austria

#### ***Influence of Moisture on Mechanical Properties of Flexible Melamine-Urea-Formaldehyde Adhesives for Structural Applications***

Melamine-urea-formaldehyde (MUF) offers a high degree of cross-linking, resulting in resonance stabilized structures with very limited flexibility. The rigidity in the backbone, as well as the high number of cross links, provide adhesive bonds with limited creep required for structural wood bonds. However,

lack of flexibility of the adhesive limits its ability to respond to dimensional changes in wood caused by swelling and shrinkage processes due to moisture changes.

This study aims to investigate the mechanical response of flexibilized MUF adhesive to altering environmental conditions. For this examination, thermoplastic component was used to provide certain flexibility to rigid MUF adhesive bonds with detectable creep and acceptable shear strength in the bondline, required for structural wood bonds. Therefore, increasing adhesive's ability to respond to dimensional changes in wood caused by the change in moisture.

To investigate this behavior, axially split spruce lamellas were planed and glued with MUF at two different melamine contents with various amounts of the thermoplastic flexibilizing component. The lap-shear samples were tested in dry and cooked conditions resulting in tensile shear strength similar to the shear strength of solid spruce wood under respective conditions. In addition, moisture-dependent elastic and viscoelastic properties of the adhesive were investigated using in-situ nano indentation. Apparent differences in elastic modulus, hardness, and indentation creep were observed at different amounts of thermoplastic components. The response of an adhesive bond line to moisture could be observed from indentation creep. With an increasing amount of thermoplastic components, the adhesive bond line became more flexible and showed higher percentages of creep observed in micro scale. Furthermore, MUF resin adhesive with higher melamine content showed more stable mechanical properties upon change in moisture with detectable creep.

### **Seongsu Park**

Kyungpook National University

### ***Converting Crystalline of Low-Molar-Ratio Urea-Formaldehyde Resins into Amorphous Ones with Bio-Based Dialdehyde Cellulose***

This study investigated to convert crystalline of low-molar-ratio urea-formaldehyde (UF) resins to improve their performance using bio-based dialdehyde cellulose (DAC). Serial resin samples during the synthesis were extracted to characterize changes in the molecular weight, chemical reaction, crystallinity, thermal curing behavior and cross-linking density using gel permease chromatography (GPC), Fourier transform infrared (FTIR), <sup>1</sup>H-NMR, and <sup>13</sup>C-NMR spectroscopy, X-ray diffraction (XRD), differential scanning calorimetry (DSC) and dynamic mechanical analysis (DMA). It is confirmed that there is evidence of the reaction between DAC and UF species. In addition, the addition of 20% DAC converted the crystalline domains to amorphous ones, which provided a very low formaldehyde emission from plywood and similar tensile shear strength of plywood. These results suggest that DAC has a potential of converting crystalline of low-molar-ratio UF resins into amorphous one to improve their performance.

### **Novel Resins II**

#### **Hendrikus (Erik) W.G. van Herwijnen**

Kompetenzzentrum Holz GmbH (Wood K plus); BOKU-University of Natural Resources and Life Sciences



### ***Strategies for Converting Renewable Resources for Implementation in Industrial Composite Board Production***

Several research groups have opened up pathways to future adhesives based on renewable resources in recent years. There are some obstacles to overcome before these solutions can be implemented on an industrial scale. Aside from the obvious economic criterion of renewable materials being available on a large scale at a reasonable price, there are also technical criteria that must be met for a timely implementation. The adhesives must be able to be manufactured and applied using standard or slightly modified industrial infrastructure. This necessitates the conversion of renewable materials to soluble or dispersed substances in order to create a "fluid" aqueous adhesive. Furthermore, the adhesive should be reactive enough to cure quickly enough to make the process economically viable. In this talk, technical methods are disclosed that provide new approaches aiming to bring renewable adhesives to the stage of industrial production.

#### **Wilfried Sailer-Kronlachner**

Kompetenzzentrum Holz GmbH (Wood K plus), Linz, Austria; BOKU University of Natural Resources and Life Sciences, Vienna, Austria

### ***Development of an In-Situ Approach Towards Sustainable, Carbohydrate-Based Adhesives***

5-Hydroxymethylfurfural (5-HMF) is a promising bio-based platform chemical for the development of new bio-based adhesive systems. Up to today, there is no large-scale industrial production of 5-HMF, which makes the pure compound expensive and not easily available. To tackle this issue, new in situ approaches for 5-HMF and subsequent adhesive production, are of high interest. It has been demonstrated that 5-HMF can improve properties of carbohydrate-amine adhesive systems.

This study describes the development of an in-situ process, in which a precursor comprising 5-HMF is produced from fructose syrup via acidic dehydratization. The formed intermediate reacts in the subsequent adhesive production with amine crosslinkers and additional fructose to the desired carbohydrate-based adhesives. Main focus is on the compatibility of 5-HMF precursor synthesis with follow-up adhesive production in terms of practical implementation, sustainability and scale-ability. The transition from a continuous reaction system to an improved batch reaction system in the upscaling process of the precursor and adhesive production as well as the influence of reaction conditions on side product formation, e.g. polymerization of 5-HMF to humins, are demonstrated.

The influence of 5-HMF rich precursors produced under different reaction conditions and with stabilizing additives like sodium dithionite ( $\text{Na}_2\text{S}_2\text{O}_4$ ) on the adhesive production and the adhesive properties as well as the effect of adhesive synthesis variables such as temperature, reaction time and reaction atmosphere on the adhesive performance, e.g. the tensile shear strength development, was studied.

#### **Catherine Rosenfield**

Kompetenzzentrum Holz GmbH (Wood K plus), Linz, Austria; Institute of Wood Technology, BOKU University, Vienna, Austria

### ***Effect of Hydroxymethylfurfural on the Cure Speed in Fructose-Based Adhesives for Interior Particleboards***

There is a push towards alternative adhesives in the particleboard manufacturing industry due to increasing regulations and public concern about emissions of hazardous substances and volatile organic compounds in chemical binders. A remaining barrier for the successful implementation of these alternative adhesives is a rapid cure behavior at lower temperatures.

Hydroxymethylfurfural (HMF) is a bio-based chemical and a promising key reactant in adhesives. The aim of this work was to evaluate the potential of fructose-HMF-amine adhesive for interior particleboard application. Special emphasis was on the temperature-dependent material behavior and on the investigation of composition-property relations. The suitability for the application was validated by testing the mechanical performance, e.g. tensile shear strength, for adhesives with different molar ratios. It was found that HMF significantly accelerates the cure speed of fructose-amine adhesives. In addition, the influence of production parameters, e.g. test and press temperature was investigated. The strength evaluation was complemented by adhesive characterization. The tensile shear strength development of HMF-containing adhesives with a molar ratio of 3.7:0.2:1 (fructose: HMF: amine) is comparable to conventional urea-formaldehyde adhesives. As a proof of concept, laboratory particle boards were produced and the performance was tested according to standards with quite promising results even for a non-optimized procedure. The press factor, an important process related parameter, is among the best of previously reported HMF- or carbohydrate-based adhesives.

**Charles R. Frihart**

Retired, USDA Forest Products Laboratory

### ***Naval Stores an Over Looked Raw Material as a Bio-Based Adhesive***

Naval stores originally described the natural products used for bonding and sealing wood boats. These mainly came from pine trees and the terminology now mainly refers to tall oil rosin and fatty acids, along with terpenes. These materials collected from the Kraft pulping of southern yellow pine continue to be used commercially for many applications, including in adhesive and ink formulations. These materials offer very interesting properties for wood bonding, but research on these bio-based materials has lagged behind other renewable sources. The structures of these materials and examples of their chemical reactivates are discussed. These bio-based rosin, fatty acids, and terpenes deserve more research as new wood adhesives.

**Thursday, May 12, 2022**

**Adhesives in Mass Timber Manufacturing I**

**Lech Muszynski**

Oregon State University

***Global CLT industry in 2021: Binder selections.***

The cross-laminated timber (CLT) industry seems to continue its incredible exponential growth across the globe. The total output of the industry in 2020 has been projected to reach 2 million cubic meters (or nearly 850 MMbf). The only continent where no new CLT plants are put in line or at least planned in 2020 is Antarctica. And yet, more than 25 years in development of the CLT technology the industry still feels young and no less intriguing. The goal of this presentation is to provide updated insights into the global CLT industry structure, output potential, production profile, internal diversity, competitiveness, innovativeness, and perceived barriers to further expansion. The presentation is based on 3 global CLT industry surveys 46 plant tours and supplemented with information obtained from other sources. This is the third iteration of a comprehensive review of the global CLT industry intended to provide insights for potential entrants and stakeholders into the CLT manufacturing sector including businesses along its extensive supply chain.

**Jinlan Ju**

RM Bilrite

**CLT Research and Market Development in China**

CLT is a new generation green and low-carbon building material suited for prefabricated buildings, which has attracted widespread attention around the world. Developing innovative CLT and then promoting CLT or hybrid CLT buildings can help reduce carbon emission and save energy consumption, thus it has important economic, social and ecological benefits in China. During the past 4-5 years, Ningbo Sino-Canada Low-Carbon Technology Research Institute has successfully built the first CLT pilot plant in China. A number of innovative CLT products have been developed, which include hemlock CLT, mixed species CLT, hollow CLT and bamboo-wood composite CLT, and so on. The patented bamboo-wood composite CLT can utilize local abundant bamboo resource in combination with softwood dimension lumber imported from NA, and also exhibit high strength performance and good feasibility for commercialization. Following the extensive R&D in CLT and the erection of a number of CLT demonstration buildings, the China first public CLT residential building has been recently built in Ningbo. By taking advantage of both structural and decorative features of the CLT, this public CLT building has successfully showcased the great potential of using prefabricated CLT to substitute steel and concrete for residential housing market in China.

**Hui Li**

Washington State University

### ***Adhesive Cure Kinetics for CLT Manufacturing Part I: Employment of DMA and DSC Techniques to Characterize Cure Behavior of 1-Component Polyurethane (1C-PUR) Adhesives***

Cross-laminated timber (CLT) has gained increasing attention in North America due to its economic and environmental benefits in mid to high-rise timber buildings. The bonding quality of CLT is the focal point toward premium quality of CLT. Understanding cure kinetics in wood/adhesive systems is important for assessing adhesive performance and optimizing process parameters. The objective of the current investigation is to assess the cure behavior of 1C-PUR adhesive commonly used in the CLT industry. A combination of dynamic mechanical analysis (DMA) and differential scanning calorimetry (DSC) methodology will be employed mechanically and chemically to assess the cure behavior of a commercial 1C-PUR adhesive with varied assembly time. The development of mechanical properties in the storage modulus thermogram from DMA is investigated and used to determine the inception and completion of mechanical cure behavior. This will verify the assembly and curing time assigned by adhesive provider. Cure kinetics of 1C-PUR with varied assembly time was further investigated using DSC at various heating regimes to assess activation energy and to predict the cure evolution in isothermal condition. The current work establishes a correlation between chemical and mechanical cure in 1C-PURs that will provide theoretical and practical guideline in determining process conditions in the production of CLT.

**Jinlan Ju**

RM Biltrite

### ***Adhesive Curing Kinetics for CLT Manufacturing Part II: Determination of Process Parameters of 1C-PUR Adhesive for CLT Manufacturing***

The mechanical properties of manufactured cross-laminated timber (CLT), used for the building industry, must meet/surpass the requirements listed in the standard ANSI/APA PRG 320. The mechanical properties of CLT are dependent on the wood-adhesive bond, which is a reflection of the CLT manufacturing parameters. Based on adhesive manufacturer guidelines, a CLT panel should be placed under pressure within the adhesive assembly time for a recommended duration. However, the provided guidelines are designed based on certain ambient temperature and relative humidity. If manufacturing conditions deviate from the recommended guidelines, how does this affect the wood-adhesive bond development? DMA and DSC analysis techniques were used to develop methodologies to evaluate adhesive cure kinetics in the previous research. Those methodologies were used to determine the influence of moisture content and temperature on cure kinetics by evaluating adhesive cure in bonds formed between adherends of multiple moisture contents and temperatures. These findings have been applied in the process for CLT manufacturing.

**Ruben Jerves**

Washington State University

### ***Towards Durable Wood-Strand Composite Mass Timber Panels***

Mass timber construction is rapidly growing in North America, given its versatility and performance. In addition, wood has a high strength to weight ratio and low environmental impact in contrast with other construction materials. Nevertheless, durability is a significant concern that needs to be addressed for

the advancement of mass timber construction. Extreme care is required to protect members from exposure to high moisture environments while ensuring long-term durability. This project thus focuses on improvements of wood's longevity by combating two main issues: dimensional stability and decay resistance to biological organisms. At the same time, a sustainable approach is considered while utilizing small-diameter logs, which optimizes forest industry products. This is all achieved with the fabrication of Cross Laminated Strand-Veneer-Lumber (CLSVL) out of thermally modified wood strands. State-of-the-art concepts and technologies are implemented in the treatment process, fabrication, and assessment of the material. At first, wood strands are treated at different temperatures to understand its implications. A look at wettability, water sorption, and wood structure is used for this analysis. Furthermore, a series of macro-level mechanical benchmark tests in thin plies are conducted, such as internal bonding, bending, and tension. Later, accelerated decay tests are done to assess the improvement of wood's resistance to rot fungi. Finally, these analyses are used to systematically optimize the thermal treatment to obtain the best performing material to be used in the fabrication of mass timber as CLSVL. The presentation will discuss the effects of thermally modified wood-strand and wood-strand composite panels in terms of mechanical performance, bonding and durability.

## **Protein-Based Adhesives**

**Charles R. Frihart**

Retired

### ***Oilseed Protein Adhesives: Aggregation, Flocculation, Cohesion, and Coalescence***

For understanding protein performance as a wood adhesive, it is important to recognize the different interactions between protein molecules and their effects on the resulting properties. It is generally considered that the native state of a protein is the most compact version arrangement of the chains. Even under conditions that cause unfolding of the protein, the protein generally refolds to some very compact state as a result of overall protein hydrophobicity and energy minimization through formation of internal polar and hydrophobic associations. However, the normal internal energy minimization usually leads to many hydrophobic patches on the protein surface. These patches along with polar bonding and disulfide bridges lead to stable aggregates of the proteins, such as the glycinin and conglycinin for the soy. However in water dispersions these protein aggregates further associate by weaker flocculation forces that bring about high dispersion viscosities that are disrupted by higher shearing forces. Heating of these dispersions leads to stronger interactions of gel formation in aqueous environments and cohesive film formation in dry environments. Although these gels and films can have good mechanical strength, they are still susceptible to reversion. The water resistance of the films is improved by heating at higher temperatures or for longer times indicating that there is improved coalescence of the adhesive when sufficient energy is put into disrupting the aggregates and forming interpenetrating links. Without this interpenetration, the main route is to link these aggregates with external chemical cross-links.

**Christopher G. Hunt**

USDA Forest Products Laboratory

### ***Does Denaturation Really Expose Protein Reactive Groups?***

There is a common statement in soy adhesive literature that denatured proteins often have higher wet strength because denaturation exposes groups for reaction with the crosslinking agent. We tested this hypothesis by doing physical property characterization of soy protein with a range of denaturation treatments. If denaturation really did open up protein for reaction, we would expect a strong correlation between available primary amines (assayed by fluorescence emission after reaction with fluorescamine) in the protein and the wet strength when amine-reactive crosslinker (polyamido-amine epichlorohydrin, PAE) is used. When we did this, we found almost no correlation. Another hypothesis is that proteins with lots of hydrophobic surfaces would form bonds with better water resistance, because the hydrophobic interactions between adjacent proteins would retain strength while wet. Our data finds this hypothesis lacking as well.

These results prompt us to examine the structure – property relations in protein bondlines, with respect to wet bond strength. Our discussion will be supported by physical property and bond strength data generated from soy proteins modified through a variety of (mostly heat-based) processes. The most effective denaturation process we have encountered to date is jet cooking, where protein is exposed to temperatures 120-170°C for short times under high turbulence, followed by fast cooling and drying.

### **Osei Asibe Asafu-Adjaye**

Auburn University

### ***Soy Flour Substitution in pMDI Resin for Composite Panel Applications***

Partial substitution of pMDI resin by 10-15% soy flour for the manufacture of strand boards, improved board properties. For MDF the soy-substituted resin performed as well as the control pMDI. The reaction of soy flour with pMDI occurred over several hours as tracked by CO<sub>2</sub> evolution. Uniform mixing of soy flour with pMDI is critical because unreacted soy flour tends to retain water, which degrades the wet properties of the board. The mechanism of the soy amended resin in the panel appears to involve the increase in resin tack provided by the added soy flour, which increases the surface coverage and the relative bonded area at the glue line.

### **Almut Wiltner**

Institut fuer Holztechnologie Dresden

### ***Strategies to Use Protein-Based Material from Insects (*Hermetia illucens*) in Resin Formulations***

Proteins are known for their gluing potential since long time. Recently, various protein based glues found industrial application particularly in the wood industry. However, native protein glues suffer from low protein solubility as well as high viscosity at low solid content. Plant proteins (e.g. soy, wheat) are mainly used in resin formulations and these raw materials are processed in different manner to overcome the mentioned drawbacks. Among other potential protein sources exhibiting a higher solubility and an amino acid distribution offering possibilities for further modification strategies resulting in resin formulations, insects are of special interest. Larvae from different species show amino acid distributions comparable to legume (e.g. pea or soy) and are therefore rich in amino acids offering free

amino groups (e.g. lysine). The proposed paper presents strategies to use protein-containing raw material from insect larvae (*Hermetia illucens*). Main properties of the raw material, of dispersions based on it as well as modification strategies for resin applications are discussed.

**Jonathan J. Wilker**

Purdue University

### ***Biomimetic and Sustainably Sourced Adhesives for Bonding Wood***

Mussels, barnacles, and oysters are examples of the many creatures generating adhesives for affixing themselves to the sea floor. Our laboratory is characterizing these biological materials and creating synthetic mimics. For example, we can mimic the cross-linking of DOPA-containing mussel adhesive proteins by placing monomers with pendant catechols into various polymer backbones. Adhesion strengths of these new materials rival those of cyanoacrylate “super glues.” Underwater bonding is also appreciable.

We are now developing biomimetic adhesives that are sourced from fully sustainable components. In order to provide market impact, further design constraints include having high structural bond strengths, degradability, containing no formaldehyde, requiring no chemical synthesis, having all components already available at industrial scales, and costs being low. Two such systems are under current development. One starting point is zein, a corn protein that is a waste product from ethanol production. Several natural phenolic compounds containing catechol groups can then be added to bring about the cross-linking and adhesion chemistry seen in mussel proteins. With some formulations the testing of bonded joints results in splitting of the wood substrates rather than failure of the adhesive. We also have a promising sustainable wood adhesive system that is based upon vegetable oils. Looking forward, we hope that development of such adhesives will help to create a sustainable materials infrastructure.

## **Adhesives in Mass Timber Manufacturing II**

**Trine Viljugrein**

Dynea AS

### ***Adhesive Systems Optimized for Cross Laminated Timber***

There is an increasing use of Cross Laminated Timber (CLT) in the building industry worldwide. CLT is more frequently used in large constructions and several story buildings, pushing the limits for building in wood. The future CLT products will have optimized properties. Can the adhesive systems play a role in this process?

The following chemistries are used for CLT; Melamine-Urea-Formaldehyde (MUF), Polyurethanes (PU) and Emulsion-Polymer-Isocyanates (EPI). Other systems are a possibility for the future, like further optimized Hybrid systems and Phenol-Resorcinol-Formaldehyde (PRF). Adhesive systems for CLT must meet regulatory requirements for load bearing constructions, such as requirements to fire resistance, heat resistance and long-term durability under referenced conditions. Important properties for producers are reactivity/pressing time, assembly time, VOC and price. For architects and end users’

environmental issues like indoor climate and CO2 footprint are in focus. The adhesive systems show different advantages in all these properties when used for CLT, and this will be discussed.

No adhesive system for CLT in the market today is superior in all the aspects mentioned above, and further product development is ongoing. Building in wood is a green, sustainable option, and it is important for the wood industry that the CLT market keeps on growing, and that CLT keeps the good reputation that makes this the preferred building method for the future.

### **John Nairn**

Oregon State University

#### ***Effect of Environmental Stresses on Cracking and Delamination in Cross Laminated Timber and Role of the Adhesives in Those Failure Modes***

Cross-laminated timber (CLT) is a mass-timber product comprised of layers of dimensioned lumber glued on the faces between layers. Despite some similarities in manufacturing processes, CLT is very distinct from other mass-timber products such as glue-laminated timber beams (Glulam) or laminated veneer lumber (LVL). Compared to Glulam and LVL, where grain directions of all layers are the same, CLT has a heterogeneous structure consisting of alternating layers with 0 degree and 90 degree grain directions. The heterogeneity of CLT has two consequences. First, it causes new failure modes where type of failure depends on whether it occurs within a 0 degree or 90 degree layer. Second, whenever CLT experiences changes in moisture or temperature, differential expansion properties between layers will cause internal residual stresses that can lead to cracks and delaminations. This talk will discuss cracking and delamination that occurs when panels dry out as well as delamination from notches that are promoted by increases in moisture content. The development of residual stresses also places a greater burden on adhesive properties, which now must be able to withstand those stresses. The role of the adhesive, as well as limitations to developing better adhesives will be discussed. Finally, some guidelines will be proposed for designing durable CLT structures including structures with notches. The most important recommendations are to use thinner layers and to control moisture content during fabrication.

### **Karl-Christian Mahnert**

Norwegian Institute of Wood Technology

#### ***Initial Study on Bonding Performance of Fire-Retardant Spruce Lamellae in Cross Laminated Timber (CLT) Elements***

The use of CLT in the construction sector has grown significantly in recent years. However, challenges with fire performance result in indoor surfaces in multi-storey wood constructions clad with plaster boards to obtain fire safety. Optimising reaction-to-fire performance of CLT could result in more exposed wooden surfaces in buildings, helping to unlock CLT's full potential for healthier indoor climate and natural looks. Using fire-retardant treated spruce in the visible outer lamellae of CLT could be a possible solution to this issue.



This research is looking into the influence of boron-based fire-retardant treatment on the bondability of spruce lamellae with three adhesive systems. The systems, one MUF- and two PUR adhesives, were chosen due to their relevance for CLT-production. The systems resistance to delamination was tested following EN 302-2 and EN 16351 Annex A, their compressive shear strength was tested following EN 14080:2013 Annex D.

The current study was designed to represent the most realistic production scenario. Thus, planing of the wood prior to bonding was kept to an absolute minimum, resulting in lower surface quality than required for standardised adhesive testing and at the same time disobeying the recommendation not to plane fire-retardant treated timber. Thus, the results do neither represent the full bonding potential of the investigated adhesives nor the full potential of the fire-retardant treatment to hamper bonding processes. Still, 22 out of 24 samples fulfilled the requirements for resistance to delamination of GLT according to EN 302-2/EN 301:2017. All 24 samples fulfilled the requirements for shear strength according to EN 14080:2013 Annex D. Seventy-four out of 79 CLT-samples fulfilled the requirements for resistance to delamination of CLT according to EN 16351 Annex A. A correlation between the fire-retardant treatment of the samples and the performance of the adhesive systems was not found.

**Simon Aicher**

MPA University of Stuttgart

### ***Testing and Classification of High Temperature and Fire Robustness of Structural Wood Adhesives***

Strength and duration of load behavior of butt bonded spruce boards

It states a widely prevailing opinion that butt bonding of wooden boards, i.e. adhesive-based end grain joints represent a senseless, inefficient and unreliable timber jointing technology as opposed to bonding of wooden components parallel or slightly inclined to fiber direction. Hence, up to today world wide no structural wooden element / construction relying on adhesively butt jointed boards is known. Except hereof are a few CLT plates butt bonded at the narrow edges using the so-called TS3 technology which has been reported previously in rather generalized manner, e.g. WCTE 2016. In order to introduce this paradigm shift in timber bonding technology into construction reality, then accepted by building authorities and design codes, TS3 company has commissioned MPA University of Stuttgart to propose, initiate and conduct experimental and theoretical investigations altogether with expertises deemed necessary to obtain a German approval and a European Technical Assessment (ETA).

In the context of this demanding task numerous tests were performed and on-going. The investigations address the pure adhesive and the adhesive-primer-wood interface, jointed boards and large scale butt bonded CLT plates. Hereby different mechanical loading modes, varying climates, extraordinary moisture and load impacts and extensive duration of load (DOL) tests are performed. To enable the experimental campaign two new large test sheds had to be built at the University Campus.

This paper reports firstly on several aspects of the adhesive and adhesive/primer interface tests, highlighting bond line features of the specifically employed 2component Polyurethane adhesive used in-service with bond line thicknesses of 4 to 20 mm. Secondly, the experimental campaign conducted at the board level is revealed. Short term and DOL results for tension, bending and shear loading are

presented altogether with proposals for the design relevant characteristic strength values and respective DOL modification factors.

### **Lech Muszynski**

Oregon State University

#### ***Integrity of Melamine Formaldehyde Bonds in Ponderosa Pine Cross-Laminated Timber - Isolating Adhesive Compatibility Effect***

Integrity of melamine formaldehyde bonds in prototype cross-laminated timber (CLT) specimens was tested as part of a project concerned with utilization of Ponderosa pine (PP) from forest restoration programs in Western US forests. Bond integrity tests, block shear and cyclic delamination, are prescribed by ANSI/APA PRG-320 performance standard for CLT to qualify new products. Of these, the cyclic delamination criterion (<5% in all specimens) is notoriously challenging for layups developed in research labs and pilot plants. The failures are often blamed on poor compatibility between the adhesive and the species, neglecting other potential factors. The criterion is crucial for approval of new CLT products, poor understanding of effects other than the adhesive compatibility may result in premature discounting of prototypes showing acceptable engineering characteristics. Therefore, one of the study objectives was to separate the effect of adhesive compatibility from other potential factors affecting bond integrity in CLT. The approach involved bonding integrity tests conducted on prototype specimens bonded with Melamine Formaldehyde (MF) adhesive fabricated three ways: 1) in pilot-plant scale line, 2) in full-scale industrial CLT plant; 3) small (76 mm x 76 mm) cross-laminated blocks with no bridging between neighboring laminations within a layer to eliminate the effect of thickness tolerance and other fabrication issues. The small blocks included groups with juvenile wood and blue-stain on bonded surfaces. Specimens from all groups passed the PRG-320 block shear criteria. All small scale blocks passed the delamination criterion, demonstrating compatibility of the MF adhesive system with PP regardless of heavy presence of blue-stain and juvenile wood. Specimens harvested from laboratory and industrial prototypes did not meet the delamination criterion. Some of the delaminations propagated from interlaminar gaps observed in laboratory and industrial prototype panels prior to test tracked back to thickness variation and inconsistent clamping pressure in the pilot plant.

### **Lignin In Adhesives I**

#### **Charles R. Frihart**

Retired

#### ***Use of Lignin and Tannin in Epoxy Adhesives***

Epoxies have the largest volume of any thermoset because of their use in adhesives, coatings, paints, embedding, etc. Although some the final products use significant quantities of bio-based materials, most of the final weight is from fossil-fuel sources. Most epoxies are cured by adding a hardener that reacts in near stoichiometric ratio to the epoxy. Given the aromatic structure of most epoxies and some hardeners, it would seem lignins and tannins can be considered as potential sources of these materials. The most common type of epoxy uses epichlorohydrin modification of bis-phenol A. Despite the wide use of this material, there has always been a lingering concern about the safety of the bis-phenol A. This

presentation covers the literature work on using lignin and tannin as the epoxy or hardener component for making adhesives.

### **Sandro Stucki**

Swiss Federal Institute of Technology (ETH)

#### ***Using biopolymers to improve the moisture stability of linear friction welded wood***

Engineered wood products are indispensable for wood-based construction. However, such products rely on the bonding of wood with synthetic and non-renewable adhesives. While achieving high quality and resistant wood bonds, using such adhesives leads to an increased environmental impact and impedes the degradability of the naturally grown wood. As an alternative binding process, linear friction welding shows potential for adhesive-free bonding of wood. In the linear friction welding process, wood is bonded by moving the adherends relative to each other in a linear reciprocal motion while applying pressure perpendicular to the movement direction. This uses the wood's natural polymeric components to create a bond through a combination of mechanical interlocking and entanglement of the wood-fibres as well as softening and flowing of the cell-interconnecting amorphous polymers (e.g. lignin) through the friction heat generated during the welding.

However, while the bonded wood shows good bond strength in dry state, a tremendous decrease in strength is observed upon changing moisture conditions. This moisture sensitivity impedes the use of such bonded products, especially in outdoor applications. We investigate wood-derived biopolymers, such as technical lignin and derivatives thereof, as bonding aid to decrease the moisture sensitivity and strengthen the bond of the friction welded wood products. By avoiding synthetic resins, friction welded wood offers the possibility for increased biodegradability and reduced environmental impact of engineered wood products.

### **Sara Yazdi**

Virginia Tech

#### ***Lignin Thermochemistry for Novel Composite Properties***

Emerging with vascular plants, lignin is the incredible aromatic polymer that evolved to physically bind and protect cellulose from fungi and bacteria. Lignin's protective mechanism includes a "self-healing" reaction, where abundant and reactive  $\beta$ -aryl ether linkages cleave and reform stronger, less-reactive carbon-carbon bonds. Known as lignin acidolysis, the reaction scheme has two pathways that are subject to catalytic selection, C2 and C3 cleavage. Ultimately, both pathways can lead to C-C crosslinking, but more so in C2 cleavage. Previous studies revealed that standard hot-pressing conditions promote a great deal of lignin reaction- our hypothesis is that this reaction can be controlled to introduce new functionality into advanced wood-based composites. Long-term goals include increasing resistance to water, fire, and biodeterioration. In the short-term, our objective is to evaluate the effect of C2/C3 ratio in terms of lignin structure and formaldehyde generation. We promote lignin acidolysis by acid-catalyzed thermal treatment of wood tissues using different mineral acid catalysts. To probe the thermochemistry of lignin, we have studied the effects of acid-catalyzed thermal treatments on lignin  $\beta$ -

O-4 content, chemical structure and functional groups, glass transition temperature, oxidative thermal stability, and molecular weight.

### **Milan Sernek**

University of Ljubljana, Biotechnical Faculty

#### ***Characterisation of Bio-based Adhesives by an Automated Bonding Evaluation System***

The characterization of the curing process by an automated bonding evaluation system (ABES), allows the determination of the optimal pressing parameters, which is essential for the economical bonding of wood and wood-based composites. In this study, ABES was used to determine the curing process of bio-based adhesives at different temperatures. ABES is a combination of a small hot press and tensile testing machine. Two 0.84 mm thick beech (*Fagus sylvatica*) veneers were bonded together to form an overlap specimen. The geometry of the bonding surface of the veneer lap joints was 5 mm x 20 mm. The bio-based adhesives studied were made from tannin and lignin, whereas the reference urea-formaldehyde adhesive was a commercial adhesive.

It was found that the curing process was fastest for UF adhesives, followed by tannin and then lignin adhesives. The curing was greatly accelerated by temperature for all adhesives. Both bio-based adhesives required higher press temperatures than UF adhesive for adequate cure. The development of the shear strength of the adhesive bond during curing consisted of three phases: initiation, growth, and termination. The results of adhesive bond strength development were fitted to a three parameter logistic model. It was found that the tannin adhesive exhibited significantly higher shear strength of the adhesive bonds than the bonds prepared with lignin adhesives.

### **Anti Rohumaa**

South-Eastern Finland University of Applied Sciences, FiberLaboratory, Finland

#### ***Eliminating the Sources of Variability in Small Scale Adhesive Bond Testing***

A significant barrier in developing new adhesives is the extensive time and cost required for testing. There is a long list of standard test methods for adhesive bond testing, many of them focusing on specific products and were not meant for the development of novel adhesives. Moreover, the performance of the final product depends not only on the cohesive strength of the adhesive, but on dozens of other interactions with the wood that need to be tested.

One reason for the high testing burden is that variability is typically large and not well understood. A significant source of this variability is the wood surface. While adhesives should not always be tested on ideal surfaces, standard tests should be on consistent substrate and generate consistent results.

This talk will focus on the influence of the wood surface on bonding performance in ASTM D7998 using the Automated Bonding Evaluation System (ABES). In addition, we will show the impact of other, typically operator-dependent variables such as spread rate, contact time with adhesive, and assembly time. These results provide guidance on how to reduce variance in routine testing.

## **Structure and Properties of Adhesives Systems I**

### **Gordian Stapf**

Henkel Engineered

#### ***New Heat and Fire Resistant 1c PUR Adhesives***

The two-dimensional building product Cross Laminated Timber (CLT) is subjected to the new phenomenon “fall-off of char layers” in the event of fire, which results in a faster charring rate of the element and can impair the self-extinguishment of CLT buildings. Another fire related situation that has not been covered by building design standards in the past is the heating of finger joints of I-joist or finger jointed lumber that are protected by plasterboards during a fire.

The first approach to introduce fire resistance was ASTM D7247, a block shear test at 220°C. Since this test did not correlate with the fall-off behavior of CLT layers in fire, additional large-scale (compartment) and small-scale Bunsen burner tests have been introduced in the North American CLT product standard PRG 320-2018. In Europe, a medium scale fire test has been developed in the meantime and will be published as an Annex to Eurocode 5 in the next years.

This presentation will point out the relevance of heat and fire performance of adhesives in buildings and how they relate to the different test standards. It will be shown that those new requirements can be passed by 1c PUR adhesives. A special highlight will be put on the (lack of) correlation between the different test approaches mentioned before.

### **Gustavo Leon**

BASF Corporation

#### ***Improve the pMDI Performance in Wood Composite Panel by Using Catalyst in the Adhesive Systems***

The use of Methylene diphenyl diisocyanate (MDI) and its polymeric MDI (pMDI) (CAS # 9016-87-9) derivative, is standard practice as an adhesive in the wood composite industry. Interest in MDI utility continues to increase as the performance of this adhesive demonstrates improved physical properties to wood panels that meet CARB ATCM Phase II / TSCA Title IV emission standards.

While polymeric MDI is recognized in the wood composite industry as a good performance adhesive, there are some misconceptions around its use that it requires high process optimization to maintain productivity unlike amino resins adhesives.

This paper demonstrates the use of synergistic additives with pMDI in wood composite panel applications that improve mill productivity during the panel pressing process.

### **Arkadiusz Kwiecien**

Cracow University of Technology, Poland

#### ***Rigid and Flexible Polyurethane Adhesives in Aspect of Their Application in Timber Structures - Experimental Tests and Numerical Modeling***

Cooperation of an university and a private company resulted in developing of composite skeleton made of spruce wood of C24 class and polymer flexible adhesive, being innovative solution investigated during a project in Poland. Structural elements were constructed using flexible adhesive layers made of Sikaflex-418 PW of 3 mm thickness and installed in prototype timber houses. Carried out research on this innovative solution indicated that flexible bonding of wood can be applied in civil engineering practice.

Following this idea, new research was carried out at Cracow University of Technology in Poland focusing on parametric analysis of flexible bonding using in structural connection of Douglas Fir wood (*Pseudotsuga Carriere*) and flexible polyurethane Sika PS. Tested specimens were constructed in form of beams with three various flexible adhesive thicknesses of 1 mm, 2 mm and 4 mm. The beam specimens were tested in bending under elevated temperatures of 20, 40, 60 and 80 Celsius degrees, resulting in load capacity and deflection measurements. Nonlinear behavior of tested flexible adhesives provided advantageous stress concentration reduction and even stress redistribution along the bond-line.

Basing on the obtained results, various specimens' parameters were determined and they were used next in numerical modeling of the innovative system. Numerical analysis allowed to visualize phenomenon of structural flexibility, which manifested promising properties in behavior of structural elements made of wood and flexible adhesives. Carried out research is aimed on developing of new wood-adhesive technology, more advantageous than classical systems using stiff and very thin layers of adhesives.

### **Karl-Christian Mahnert**

Norwegian Institute of Wood Technology

#### ***Long Time Durability of Adhesive Joints in Glulam Beams After Exposure to Nordic Climate***

Durability of adhesive joints in laminated wood products is crucial for these products service life. In the current study, the adhesive joint durability of 10 different adhesive systems was examined.

A total of 6 spruce glulam beams (400\*15\*24-28 cm, l\*w\*t) per adhesive system (2x EPI, 2x MF, 3x MUF, 2x PUR, 1x PRF) were produced in commercial plants. Two beams for each adhesive system were exposed to 2 different climates (average climate 1: 20°C, 68% RH, climate 2: 5°C, 85% RH). Delamination and shear strength of the adhesive joints were tested upon production of the beams and after 5 years of exposure.

The average overall delamination of all adhesive types increased from 1.9% upon production to 2.4% after 5 years. The average overall shear strength increased from 9.0 to 9.2 N/mm<sup>2</sup> during the same period.

Overall delamination and shear strength of the adhesive systems from the same type of adhesive upon production was not significantly different for EPI, MUF -1 and MUF-2. This had changed after 5 years of exposure.

The adhesive joint durability after 5 years storage at 20 °C and 65% RH was maintained. The shear strength of 3 adhesive systems increased significantly during this period.

Delamination of EPI-1 and MF-1 was significantly higher after exposure to climate 3 compared to delamination after exposure to climate 1. A significant decrease of shear strength of the PRF adhesive joints after exposure to climate 2 compared to climate 1 was observed. The same trend, but not significant, was observed for 8 of the 9 remaining adhesive systems.

Changes in wood failure during exposure to climate 2 were neither consistent nor significant. The same applies for changes in fibre failure. However, a decrease in wood failure was accompanied by an increase in fibre failure and vice versa.

### **Anita Tran**

Department of Material Science and Process Engineering, Institute of Wood Technology and Renewable Materials, University of Natural Resources and Life Sciences Vienna

#### ***Temperature and Moisture Dependent Rheological Cure Characterization of Wood Adhesive***

Curing kinetics of adhesives are important for the manufacture of engineered wood products. Various influencing factors such as temperature and humidity can affect the reactivity of the adhesive and thus also the curing speed and the resulting adhesive strength. This research investigates curing kinetics of adhesives for structural timber products where mechanically stable and durable adhesive bonds are crucial under conditions currently not foreseen by relevant standards.

The curing behavior of room-temperature cured melamine-urea-formaldehyde (MUF) and one-component polyurethane (1C PUR) was investigated at different temperatures and humidities. A rheometer set-up with conditioned poplar plates allowed to observe the interaction of adhesive and substrate in realistic conditions. The curing kinetics were monitored for temperatures from 0 °C to 20 °C and controlled wood moisture contents ranging from dry, standard to humid conditions. The resulting gel times and reaction rates slowed down for both adhesives with lower temperatures. MUF showed substantially prolonged curing reactions with decreasing temperature. In contrast, 1C PUR exhibited extended gel times for dry specimens as the moisture required for cure was not sufficiently available. Adhesive strength was verified by tensile shear strength testing of veneer lap joint samples with both adhesives. The obtained tensile shear strength supported the rheological results. The findings indicate an influence of temperature and humidity on the gel time. Additionally, PUR was least influenced by the temperature of the substrate at the time of adhesive application permitting bonding at low temperatures.

### **Lignin in Adhesives II**

#### **Robert Narron**

Borregaard

#### ***Technical Lignin Materials in Commercial Resins: A Review of Main Benefits and Technical Challenges***

Bio-based adhesives were used for centuries as wood binders until they were replaced by oil-based thermoset adhesives about 70 years ago. Adhesive industry interest on partial or total replacement of synthetic chemicals by natural polymers has been on and off over past decades, mainly linked to

potential cost savings when crude oil price rose. Other drivers like the growing concern on sustainability and health issues have also gained importance. Now, the adhesive industry interest in bio-based adhesives has grown significantly.

Technical Lignin Materials (TLM) are available in the volumes demanded by the wood adhesive industry. TLM are obtained from a renewable source, therefore they can have a significant contribution to reduce carbon footprint of synthetic resins as evidenced by LCA data. Moreover, TLM can also contribute to reduce formaldehyde emissions in wood panels when used in UF resins or in the formulation of NAF adhesives.

LTM are also highly variable and complex matrices that display different properties depending on the raw material source and the extraction methods used to isolate the lignin. Close cooperation among lignin manufacturers, resin producers & users, and research institutions will be key to learn how to deal with this complexity and to implement successful production at industrial scale of lignin-based resins with similar properties to existing synthetic resins: reactivity, adhesion strength & moisture resistance. Some case studies will be presented with new options for lignin introduction into resin systems.

## **Heidi Suomela-Uotila**

UPM Biochemicals

### ***Lignin Based Binders: Heading for Mainstream***

Lignin is an ideal bio-based substitute for various Petro-based products used today. Lignin has already found its way into a rapidly growing number of industrial applications; lignin-based phenol formaldehyde resins (LPF) for wood panels being a prominent example.

UPM is a pioneer in the utilization of lignin in phenolic resins for plywood, OSB, paper impregnation and mineral wool. Lignin has been used as part of the WISA BioBond gluing technology in UPM Plywood's production. As of today, lignin can replace approximately two thirds of the phenol typically used in the adhesive without compromising performance or productivity of the production line. UPM BioPiva™ lignin and the resin technology related to its use are supplied to many of the biggest resin producers worldwide.

The focus of further development work has been on achieving even higher phenol substitution levels, improving robustness towards varying plywood production conditions, and versatility regarding adhesive application methods, as well as broadening the range of wood species. Due to its nature, LPF resins are traditionally considered as slow curing and having limited water resistance compared to conventional PF resins. Based on the experiences we have gathered over the years in implementing LPF-resins industrially, in a global context, it can be concluded that reactivity of LPF resins can be adjusted to meet the requirements of short press cycle. In this presentation, a summary of the latest technological development steps and findings are included. We show a range of case studies, from pilot to industrial scale, focusing on the versatility of the lignin technology and critical aspects and challenges of industrial implementation. In addition, data on sustainability related features of the latest generations of LPF-resins are presented.

Finally, we give an overview on future developments towards making LPF-resins become the sustainable new binder generation in wood panel manufacturing industries.



Keywords: lignin, resin, adhesive, wood panel, curing

## **Mojgan Nejad**

Michigan State University

### ***Lignin-Based, Formaldehyde-Free Wood Adhesives***

This talk is focused on using suitable lignins to replace 100% of petroleum-based phenol and polyol in formulating phenolic and polyurethane adhesives for engineered wood products. In the first study, both phenol and formaldehyde were entirely replaced by biorefinery corn stover lignin and a biobased dialdehyde (glyoxal) to formulate phenolic adhesive. The developed lignin-glyoxal adhesive had high dry adhesion strength (4.2 MPa) with over 90% wood failure based on lap-shear analysis results but failed the boil water test. The formulated biobased, formaldehyde-free adhesive can be used for interior plywood applications, but more in-depth analyses are needed to investigate why it failed the boil water test. In the second study, kraft softwood lignin was used to replace 100% of petroleum-based polyol to formulate a zero-VOC polyurethane adhesive for cross-laminated timber (CLT) application. When cured at room temperature, the developed lignin-based PU adhesive had higher dry (6.7 MPa) and wet (4.6 MPa) adhesion strength than commercial PU adhesive. In addition, we will discuss our effort in leading a consortium of 6 industry partners (Roseburg, West Fraser, Fortum, Hexion, Henkel, and Element 5) in scaling up the production of lignin-based adhesives by Hexion and Henkel and the plan to run trials on the manufacturing lines of plywood and CLT (<https://cribe.ca/projects/roseburg-forest-products/>).

## **Scott Rennekar**

University of British Columbia

### ***Uniform and Functionalized Softwood Kraft Lignin for Adhesive Technologies***

Land plants have been 'performance engineered' over time with certain phenylpropanoids polymerizing into lignin that supports towering trees. This key structural polymer is modified and removed in industrial delignification processes during pulp and paper manufacturing. As softwood kraft lignin is currently recovered in multiple operations around the world, utilization of this material is key in reducing our carbon footprints and ushering in a new era in biobased products. In part, utilization is hampered by the resulting variation of properties of the new products. Some of this variation in performance most likely arises from disparate structures and properties of the isolated lignin itself. With a focus on lignin fractionation and the refinement of tools to rapidly assess chemical structures of lignin that change drastically from their original state, enhanced understanding of lignin structure-property behavior is enabling material development. Our research group has focused on methodology to create lignin with defined functionality through the process of using a combination of carbonate reagents and organic acids. In this process, we developed a real-time control reaction to produce modified lignin with a variety of functionality with a two-step, one-pot process. Uniform lignin samples can be readily recovered with downward precipitation. The process results in technical lignin with uniform molar mass and chemical functionality ideal for new adhesive technologies. Some examples of uniform lignin chemical functionality include primary aliphatic hydroxyls or esters of organic acids (C1-C18). If the acid, like oleic acid, has vinyl groups these samples can be furthered epoxidized enhancing the number of

adhesion systems lignin can be utilized in. In this talk both chemical structure and thermal properties of modified lignin will be highlighted and related to the usefulness in adhesives.

## **Peter Lingenfelter**

Prefere Resins Finland Oy

### ***Significant Industrial Use of Lignin in Phenolic Resins***

Phenolic resins are the oldest synthetic polymers with availability in industrial scale since 1909. The three-dimensional network of the cured polymer is responsible for the high level of chemical and thermal resistance and good mechanical properties. Phenolics can be found in construction, insulation or industrial applications such as abrasives, friction, foams, foundry and others.

Lignin is the second most abundant renewable raw material available. It can be found in the cell walls of plants and can make up to 30% of biomass. Lignin has a phenolic structure with high hydrophobicity that makes it a promising bio-replacement for phenol in synthetic resins. Despite the tremendous amount of work done around the utilization or so-called "valorization" of lignin in phenolic resins, its high availability, and its similarity to synthetic phenol-formaldehyde resins, only a few lignin-based resins have ever been commercially available or successful.

Currently, well-defined lignin is available in sufficient quantities for industrial application, opening up new options for substantial utilization in phenolic resins. In 2017 Prefere Resins/Phenolics began industrial production of lignin-modified phenolic resins for panel applications with a high replacement rate of phenol. Year-on-year volumes have grown leading to over 7000 MT production of lignin-substituted resins in 2019. Initial good performance on roller spreaders for plywood was followed up by achieving equivalent performance on a variety of other spreader types commonly used in the plywood industry. 2021 has seen the launch of these resins in mills using extruder and foam extruder spreading, and we have reached as high as 80% substitution of phenol in another industrial application. Promising results have also been seen with resins for a variety of applications such as fiberboard, HPL and CPL, floral foam, abrasives and rock-wool insulation. Further investigations are constantly ongoing, and we are continuously pushing the envelope of where lignin substitution of phenol in phenolic resins can succeed.

**Friday, May 13, 2022**

## **Structure and Properties of Adhesives Systems II**

### **Tomas Pipiska**

Department of Wood Science and Technology, Faculty of Forestry and Wood Technology, Mendel University in Brno

#### ***Utilization of the Less Known Wood Species in the Strandboards***

The global climate change and consequent bark beetle calamity, together with the changes in forest management in Central Europe, have contributed to the decrease of the percentage of spruce raw material in the future. Due to these changes, it is very important that the production of wood-based composites will be able to reorient to other, economically important wood species in the future while maintaining the required parameters of the panels. A comprehensive analysis of the properties of less known wood species primarily Birch, Larch, Alder, and their comparison with spruce will be made for use in strandboard manufacturing. Laboratory manufacturing of the strands will be placed with analysis of the strands for the dimensions. The strands will be used for manufacturing the strandboards with different types of adhesives. The verification of the physical and mechanical properties of the boards will be made for different wood species.

### **Joseph Marcinko**

Polymer Synergies, LLC

#### ***Characterizing The Molecular Interactions of Latex Wood Adhesives Containing an Engineered Polysaccharide Polymer Additive***

International Flavors and Fragrances (formerly DuPont Industrial Biosciences) offers a family of engineered polysaccharides through the selective polymerization of sucrose to provide materials ranging in molecular weights, solubility, and polymer morphology. One specific example, the linear alpha-1,3-polyglucose has been assessed as a performance enhancing additive in polyvinyl alcohol stabilized polyvinyl acetate copolymer latexes. The glucan polymer was incorporated as a dry powder and the adhesive solids adjusted for comparative purposes. Adhesive performance was measured using ASTM Standard Method D-905 on Hard Maple (*Acer saccharum*) and ASTM D-906 on yellow poplar (*Liriodendron tulipifera*) plywood veneer. Dynamic Mechanical Analysis (DMA) and Nuclear Magnetic Resonance (NMR) spectroscopy reveal unique molecular interactions between the alpha-1,3-polyglucose and the latex polymers, which are shown to provide significant improvements in adhesive strength and wood failure performance.

### **Christopher G. Hunt**

USDA Forest Products Laboratory

#### ***Soy Adhesive Mechanical Properties Driving Wet Product Performance***

One issue preventing soy adhesives from making more significant gains in the wood products sector is ability to produce cost competitive wet strength. This project is focused on understanding what

mechanical properties of the adhesive are related to improved performance in the standard product tests of wet shear and 3 cycle soak. The effort used different soy sources, additives, and temperatures to generate plywood with a wide range of wet strengths. Furthermore, parallam samples were made under identical conditions to the plywood and were used to determine mechanical properties of the different adhesive formulations. Nanoindentation tests of gluelines were performed to identify adhesive hardnesses and elastic moduli. Adhesive fracture energy release rates were determined from a series of identical four point bending tests on notched laminates. All measurements are made on wet samples. The talk will briefly review the methods and discuss results.

## **Adhesive Characterization**

**Mark Cashman**

Virginia Tech

### ***Carbon Isotope Ratios: Providing Insight Towards Formaldehyde Emission Source***

Non-structural wood-based composites, like those used to manufacture kitchen cabinets, have faced continued regulation from the federal government regarding allowable formaldehyde emissions. These wood composites are composed of two main components, wood furnish and urea-formaldehyde resin, however, the regulations fail to take into consideration that wood naturally generates formaldehyde. Increasingly low limits complicate compliance with the new regulations. This work aims to advance awareness and scientific context in light of future regulatory restrictions by investigating the source of formaldehyde emissions. By capturing formaldehyde emissions it's possible to distinguish synthetic and biogenic formaldehyde through measurement of the carbon isotope ratio,  $^{13}\text{C}/^{12}\text{C}$ . The efficacy of different formaldehyde capture methods was explored and capture systems were designed for small scale model formaldehyde capture experiments and formaldehyde capture during particleboard manufacture and during post-press thermal equilibration.

**Mario Beyer**

Institut fuer Holztechnologie Dresden

### ***Melamine Quantification in Amino Resins***

Melamine-reinforced amino resins (MUF) can be analyzed by means of their free and total melamine content in different manner. The free melamine content can be estimated using short extraction duration in solvents prohibiting hydrolysis and applying high performance liquid chromatography (HPLC). Usually, the total melamine content is determined after resin hydrolysis and photometry. However, for the performance a special equipment to prevent material loss during hydrolysis at elevated temperatures is required (Soxhlett device). Since melamine shows characteristic infrared vibration bands originating from the triazine ring and the NH-bonds, respectively, one can use spectroscopy in middle (MIR) and near infrared (NIR) range to determine the total melamine content. MIR and NIR spectroscopy can be used for melamine determination in liquid and dried sample material. We will present possibilities and testing performance for the determination of free and total melamine content in liquid and dried amino resins. The results from MIR/NIR spectroscopy are compared to those resulting from hydrolysis and UV/VIS-spectroscopy. Free melamine determination in liquid resins is

presented by means of different solvents (H<sub>2</sub>O and DMSO), extraction duration as well as temperature. The resin hydrolysis is observed applying gel permeation chromatography (GPC).

**Lech Muszynski**

Oregon State University

***An Indirect Method for Detecting Adhesive Performance in CLT Subjected to Fire Performance Tests ABased on ASTM E119 Furnace Temperature Signal Analysis***

The most desired behavior for CLT panels in fire events is a gradual transformation of flaming combustion into smoldering combustion until the natural self-extinguishment. Such behavior is observed when the charred wood on the surface provides layer of insulation for the core material. However, a large proportion of CLT products is bonded with an adhesive system that at some point of fire event lets the char layer fall off creating additional source of heat in the interior and, exposing unburned surface of the next layer to fire, leading to second flashover. The severity of the effect of such event on the fire dynamic may depend on the precise timing compared to the progress of the char front through the panel section. The hypothesis of this study is that the char fall-off events may be detected with reasonable accuracy based on the analysis of the micro-disturbances of the furnace temperature signal. We will present the theoretical background and a proof of concept for a practical application of such method.