IOP Conf. Series: Materials Science and Engineering **123** (2016) 012054 doi:10.1088/1757-899X/123/1/012054

Improving the connection between wood and cement using LBL nanocoating to create a lightweight, eco-friendly structural material

L Bejo, B Major, L Csoka, Z Hantos, Zs Karacsonyi

University of West-Hungary, Simony Karoly Faculty of Engineering, Wood Sciences and Applied Arts

E-mail: laszlo.bejo@skk.nyme.hu

Abstract. Structural elements made out of cement bonded wood may be an excellent alternative to flammable organic bonded composite beams, and CO2 intensive, heavy and nonrenewable reinforced concrete. Unfortunately, preliminary studies showed that a sufficient load-bearing performance is difficult to achieve. Improving the compatibility of cement and wood by LbL nanocoating may be a significant step towards creating viable cement bonded wood load bearing elements.

The study involved creating multi layer nanocoating on the surface of poplar veneer using various polyelectrolyte combinations and numbers of treatment cycles, and testing the withdrawal resistance of the samples from a cement matrix. PDDA-PSS treatment was found to form increasingly uniform coating on the surface of wood, while the results were less straightforward for PAH-PSS. Both types and all levels of treatment caused dramatic improvement in load withdrawal resistance. The best result – a more than tenfold improvement – was achieved by at least 10 cycles of PDDA-PSS treatment. PAH-PSS treatment yielded a somewhat more modest improvement, which was already evident after five treatment cycles. The results point to the excellent potential of LbL nanocoating for creating cement bonded structural wood based composite materials.

1. Introduction

In many ways, wood based materials are ideal: they are lightweight, renewable and recyclable, and can be tailored to suit many applications. Among its many uses, wood based panels and beams have been used for structural purposes. Structural wood based beams, like LVL, PSL and LSL (Laminated Veneer Lumber, Parallel Strand Lumber, and Laminated Strand Lumber) in particular, are becoming increasingly widespread in the USA, and are also gaining popularity in Europe [1].

One significant drawback of these products is their limited fire performance. Along with solid wood, they are generally classified as flammable, with normal combustibility. For this reason, some regions enforce strict building codes that severely limit their application, despite their excellent specific strength and environment friendly production technology.

Steel reinforced concrete construction elements are perhaps the most prevalent in Europe, especially for large residential and commercial construction. While these materials are „tried and true”, they are very heavy, made of non-renewable materials, require much energy and emit much carbon dioxide during their production.

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd

A combination of the concepts of wood based structural beams and reinforced concrete could be the ideal solution for this problem. However, existing experience with cement bonded wood based materials, as well as some preliminary experiments showed that high-strength cement bonded wood based materials are difficult to produce. Their strength is much lower than required for structural applications [2]. One of the reasons identified was the chemical incompatibility of wood and cement. LbL nanotechnology is a relatively new technique developed in the 1990s. The idea behind this process is changing the surface chemistry of materials by the alternate application of cationic and anionic polyelectrolytes. The materials in the polyelectrolyte solutions bond to the surface, and form a nano-layer, to which the next layer of nanomaterial can bond. After the first two layers, the process can be repeated several times to form a multi-layer coating. Under ideal conditions, the layers become increasingly uniform, and the surface charge becomes homogeneous [3].

LbL nanotechnology is a very simple, cheap and efficient process. Several different kinds of polyelectrolytes or even other materials (e.g. nanomineral colloids) can be applied to a range of different substrates. de Villiers et al. [4] wrote a comprehensive review of the different materials methods and applications used in various studies. LVL nanocoatings are especially helpful in medical technology, where change of surface chemistry can bring dramatic improvements, e.g. in the delivery of certain drugs. It is also useful in many industrial applications, e.g. for creating special composites, improving paints and surface treatment, or for better adhesion, etc.

The earliest studies to create LBL nanocoating for wood surfaces were carried out around the turn of the century [5]. At first, these experiments typically concentrated on creating nanocoating on individual wood fibres, mainly to improve the characteristics of paper, or improved bonding in composite materials ([6],[7],[8],[9],[10],[11]). Lingström et al. [12] provided a thorough overview of many studies, and gives a systematic overview of the application of polyelectrolyte nanocoatings on lignocellulosic fibres.

There are few studies available that deal with treating larger particles or even large wood surfaces. Renaccer and Zhou [13] established that nanoscale films on wood can be deposited without changing the microscopic and macroscopic texture. Zhou [14] used various polyelectrolytes and coating techniques to create nanocoatings on pine veneer that can be used as adhesives.

LbL nanotechnology may also be used to improve the compatibility of wood and cement. Applying just one layer of montmorillionite nanomineral and PDDA to hybrid poplar wood wool, Alpar et al [15] managed to improve the bending strength of cement bonded wood-wool by 20%. These encouraging results will hopefully inspire further research in this area. The purpose of this study is to examine the potential of polyelectrolyte treatment to improve the connection between wood and cement. The objectives include:

- Treating wood particles with various types of nano-polyelectrolytes and using different numbers of treatment to change its surface charge,
- Monitoring the uptake of the nanomaterials in poplar wood, and
- Examining the effect of the treatments on the wood-cement interface by mechanical testing.

2. Materials and methods

The basic substrate chosen for the treatment was 2.5 mm thick peeled veneer made of hybrid poplar (Populus × euramericana cv. ‘I-214’). This poplar hybrid is very popular in Hungary, due to its relatively high strength and durability. It is also advantageous because it has a relatively low sugar content that would inhibit cement curing. The veneer was cut into 25 mm wide strips, at least 250 mm in length, and stored under normal conditions (21 °C, 65 % RH) for several days to reach a moisture content of approx. 12 %. The specimens were chosen for treatments in a random manner (CRD). 0.1 concentration (m/m) aqueous solutions were prepared of the following polyelectrolytes, using distilled water:

- Poly(Diallyl Dymethil Ammonium Chloride (PDDA) (Mw: 400-500 kDa)
- Poly(Allylamin Hydrochloride) (PAH) (Mw: 58 kDa)
- Poly(Sodium 4-Styrenesulfonate) (PSS) (Mw: 70 kDa)

The above materials are frequently used to create LbL nanocoatings. PDDA and PAH are strong polycations, and PSS is a polyanion. The following treatments were applied to the veneer strips:

- PDDA + PSS, repeated 5, 10, and 15 times,
- PAH + PSS, repeated 5 and 10 times,
- distilled water only (repeated 30 times), as control.

In each case, three veneer specimens were treated by dipping an 80 mm section at the end of the veneer strips into the solution for 10 minutes. The amount of solution was determined based on the mass of the treated sample. After each treatment, a sample of the solution was analysed using a WPA lightwave, Diode-array, S2000 UV/Vis spectrophotometer. Difference absorbance spectra were created using the corresponding measurements on the control samples, to verify how much material remained in solution after the treatment.

After completing the LbL treatments, the specimens – including the control – were encased in a cement matrix to a depth of 80 mm (see figure 1). Commercial Portland cement, type CEM I 42.5 based on EN 197-1, was used to create the paste, with a water to cement ratio of 7:20. The encased samples were left to cure for 28 days before testing.

After curing, pullout tests were performed on the specimens, as indicated in figure 2. The tests were carried out under displacement control, at a rate of 0.5 mm/min. After testing, the adhesion of cement to the surface of the extracted veneer strips was evaluated visually.

Figure 1. LbL treated poplar veneer samples encased in cement matrix

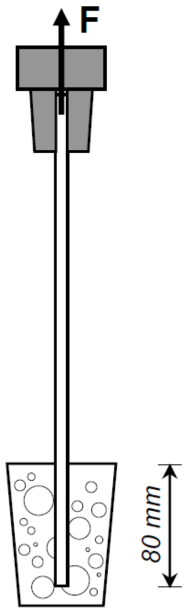

Figure 2. Schematic of the pullout test

3. Results and discussion

PDDA-PSS treatment proceeded as expected. Figures 3a and 3b show the difference absorbance spectra of PDDA and PSS, respectively (15 repetition samples). These diagrams display more-or-less decreasing values, i.e. decreasing residual concentrations with increasing number of treatments. This is consistent with our expectations; as the number of nanolayers increases, more and more uniform coverage is achieved, which progressively facilitates the bonding of the next layer. After about 5 PDDA-PSS treatment cycles, the residual concentration approaches zero, i.e. there is practically no PDDA or PSS remaining in the solution. (N.B.: slightly negative values calculated after several treatment cycles reflect experimental error, rather than physical reality.)

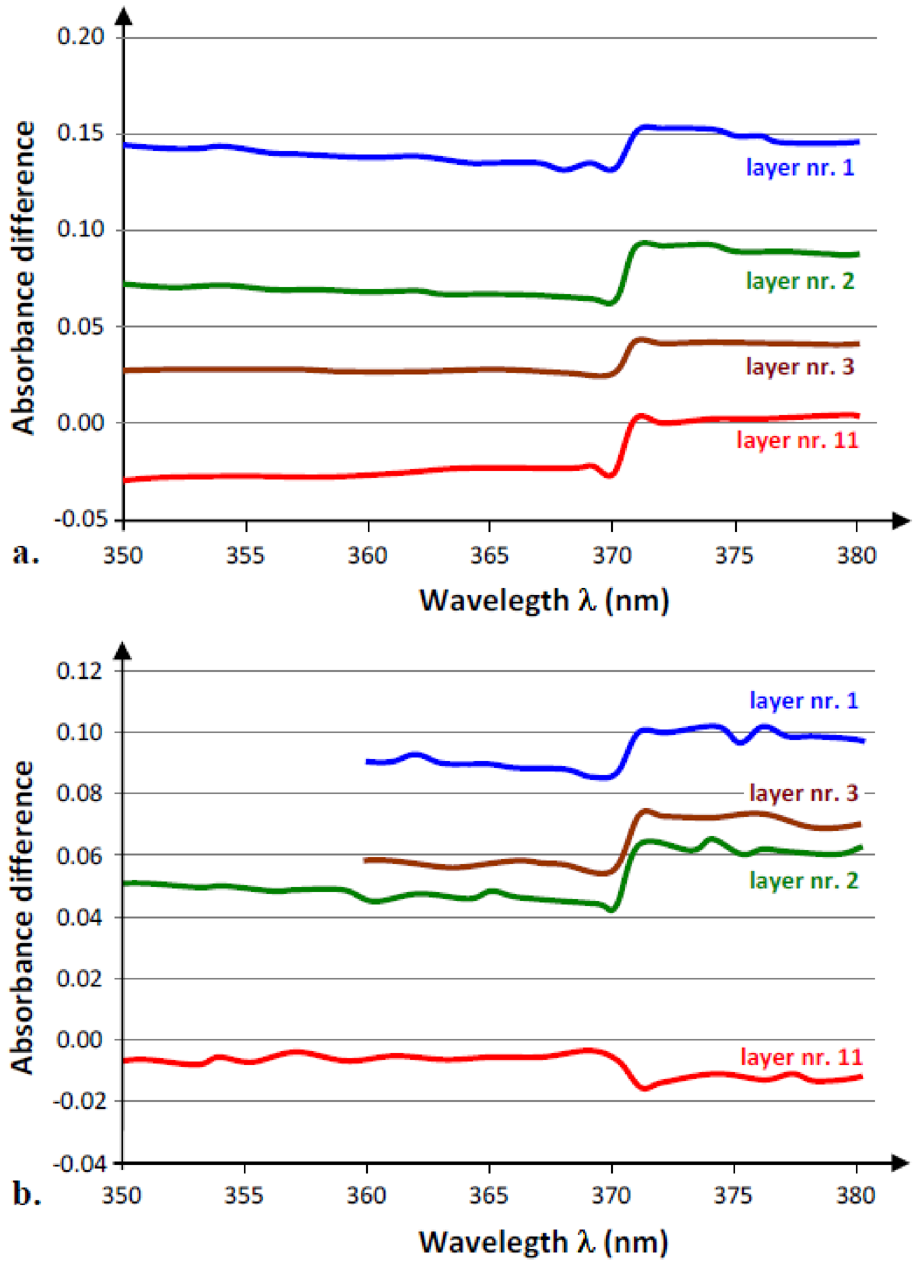

Figure 3. Difference absorbance spectra of the PDDA (a) and PSS (b) solutions after the adsorption of the polyelectrolytes on the surface, measured in various stages of the LbL treatment

PAH-PSS treatment yielded less straightforward results. Even though there was a general decreasing trend, residual concentrations changed somewhat more erratically. One possible explanation lies in the molecular weight of PAH, which is lower than that of PDDA. The small molecular dimensions may have prevented the formation of continuous nanolayers in some cases, and deteriorated the next layer's ability to bond. Nevertheless, by the end of the 5th cycle, fairly continuous nanolayer formation likely occurred. The experiences show that higher concentrations of PAH solution may be beneficial in similar future experiments.

Figure 4 shows the results of the pullout tests. In general, all of the treatments led to dramatic improvement in pullout force. 5 cycles and 10 cycles of PDDA-PSS treatment increased the load withdrawal resistance more than fivefold and tenfold, respectively. After 10 cycles, further improvement was not observed. Similarly, 5 cycles of PAH-PSS treatment caused an almost tenfold improvement, after which no significant further increase occurred. Visual observation confirmed the excellent cement adhesion achieved after the LBL treatment, as opposed to the untreated samples.

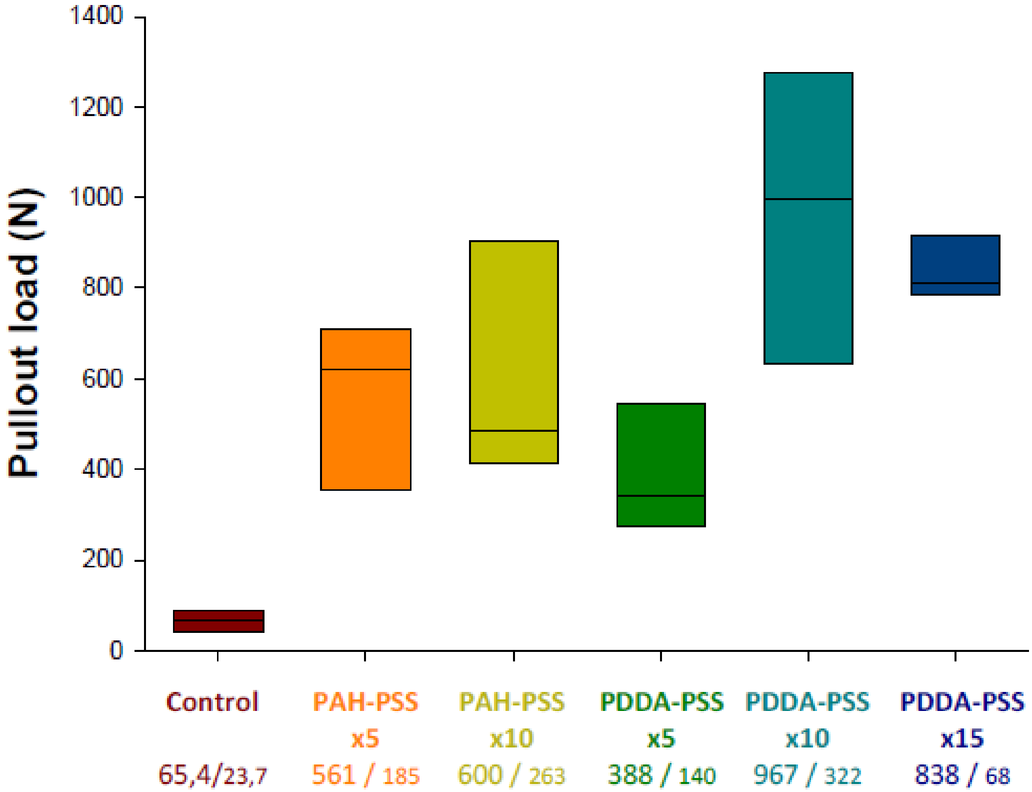

Figure 4. Pullout force measurement results of veneer samples treated with various numbers of polyelectrolyte layers. The average and standard deviation values are indicated on the horizontal axis

4. Summary and conclusions

The research described in this paper, concerning the use of LbL polyelectrolyte treatment of hybrid poplar veneer to improve its compatibility with cement, yielded the following conclusions:

- PDDA-PSS treatment was found to form nanolayers of increasing uniformity, as evidenced by UV-VIS difference spectroscopy results. After 5 treatment cycles, the residual concentration in the solutions was close to zero.
- PAH-PSS treatment performed reasonably well, but the results were less straightforward.
- Higher PAH concentrations may be required for uniform layer formation.
- All of the treatments caused dramatic improvement in load withdrawal resistance, with a more than tenfold ultimate increase in withdrawal load in case of PDDA-PSS treatment. No further improvement was observed in withdrawal resistance after 5 and 10 treatment cycles, in case of PDDA-PSS and PAH-PSS treatment, respectively.

The results indicate the excellent potential of LbL nanotechnology to improve the strength of cement bonded wood based materials. Further experiments are required, however, to establish the applicability of this technique to create structural load bearing cement bonded wood composites.

Acknowledgements
This study was supported by the Environment conscious energy efficient building TAMOP-4.2.2.A-11/1/KONV-2012-0068 project sponsored by the EU and European Social Foundation.

References

[1] Forest Products Laboratory 2010 *Wood handbook* – Wood as an engineering material, General Technical Report FPL-GTR-190. 508
[2] Bejo, L., P. Takats, N. Vass. 2005 *Acta Silvatica et Lignaria Hungarica* **1** 111
[3] Decher, G., Y.M. Lvov, J. Schmitt 1994 *Thin Solid Films* **244** 771. DOI: 10.1016/0040-6090(94)90569-X
[4] de Villiers, M.M., D.P. Otto, S.J. Strydom, Y.M. Lvov 2011 *Advanced Drug Delivery Reviews* **63** 701 DOI: 10.1016/j.addr.2011.05.011.
[5] Forschberg, S, L. Wågberg 2000 Production of Particles or Fibres having a Coating of Polyelectrolytes Interacting with Each Other and Paper or Nonwove Products with Improved Opacity Therefrom. *SCA Hygene Products* AB Sweden, 19
[6] Agarwal, M., Y.M. Lvov, K. Varahramyan 2006 *Nanotechnology* **17** (21) 5319. doi:10.1088/0957-4484/17/21/006
[7] Zheng, Z., J. McDonald, R. Khillan, Y. Su, T. Shutava, G. Grozdits, Y.M. Lvov 2006 *Journal of Nanoscience and Nanotechnology* **6**(3) 624. DOI: 10.1166/jnn.2006.081
[8] Lu, Z., S. Eadula, Z. Zheng, K. Xu, G. Grozdits, Y. Lvov 2007. *Colloids and Surfaces* A: Physicochemical and Engineering Aspects **292** (1) 56
[9] Peng, C.Q., Y.S. Thio, R. A. Gerhardt. 2008. *Nanotechnology* **19** (50) 505603. doi: 10.1088/0957-4484/19/50/505603
[10] Lin, Z., S. Renneckar 2011. Composites Part A: *Applied Science and Manufacturing* **42** (1) 84. DOI: 10.1016/j.compositesa.2010.10.011
[11] Lee, J, J. Ryu, H.J. Youn. 2012. *Cellulose* **19** (6) 2153. DOI: 10.1007/s10570-012-9781-6
[12] Lingström, R., E. Johansson, L. Wågberg 2009 Polyelectrolyte Multilayers for Fibre Engineering. In: L.A. Lucia and O.J. Rojas eds. *The Nanoscience and Technology of Renewable Biomaterials* Wiley and Sons, Chichester, UK. 123.
[13] Renneckar, S., Y. Zhou 2009. *ACS Appl Mater Interfaces* **1**(3 559. doi: 10.1021/am800119q.
[14] Zhou, Y 2008 Nanoscale surface modification of wood veneers for adhesion. MS Thesis, Virginia Polytechnic Institute and State University, Blacksburg, VA, USA. 80 pp.
[15] Alpar, T.L., A.A. Pavlekovics, L. Csoka, L. Horvath 2011 Wood wool cement boards produced with nano minerals. In: *Proceedings 3rd Int. Scientific Conference on Hardwood Processing* (ISCHP3 2011) I: Peer reviewed contributions. October 16-18, 2011. Blacksburg, VA, USA. pp. 75-82. ISBN 978-0-9837700-0-8.