

**EUROPEAN CO-OPERATION IN WOOD RESEARCH
FROM NATIVE WOOD TO ENGINEERED MATERIALS.
PART 2: DENSIFICATION MODIFICATION IN PRODUCT DEVELOPMENT**

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Abstract:

Wood is a renewable, biological material used in numerous applications and it is growing in importance due to sustainable development efforts. Wood also suffers from a number of disadvantages, were low hardness and abrasive resistance are characteristic for low-density species. This paper presents state of the art on different wood densification processes as one emerging process technology for increased use of low-density species. The presentation is based on work by different European research groups in wood science, collaborating in the field mainly through different COST Actions. The main principles for processes are discussed, such as bulk and surface densification, as well as methods for reducing the shape memory effect of densified wood. The main challenges for the future are in the field of finding fast and environmental friendly method for elimination of the set-recovery and scaling up to profitable industrial applications. To provide a better understanding with this regard, some relevant applications of densified wood are presented.

Key words: *compression; flooring; plasticization; thermal-hydro-mechanical processing.*

INTRODUCTION

Densification, i.e. a viscoelastic thermal transverse compression to achieve a permanent deformation of wood cells and thereby an increased in density of a piece of wood or of a part of it, is one of the approaches to improve the properties of wood that has been the subject of many studies during the recent decades (Sandberg et al. 2013). The main goal of densifying wood is to increase its hardness and surface abrasion resistance, but also in some cases to increase its strength.

For many decades, humans have been consuming more resources than the world has to offer in the long run. Increasing the use of renewable materials, such as wood, is essential if we are to achieve a sustainable use of the resources available to us. Densification has the potential to improve the properties of widely available low-density wood species, opening up new fields of application, and fostering the use of wood products in general. Fig. 1 shows an example of how low-density poplar from the Central Czech Republic (clones Max 4) was used in a Czech-Slovenian-US project to improve the strength properties by heating and compression. The density was increased up to three times compared to undensified poplar with the result that modulus of elasticity (MOE) also increased considerably (Hornicek et al. 2015; Rademacher et al. 2016).

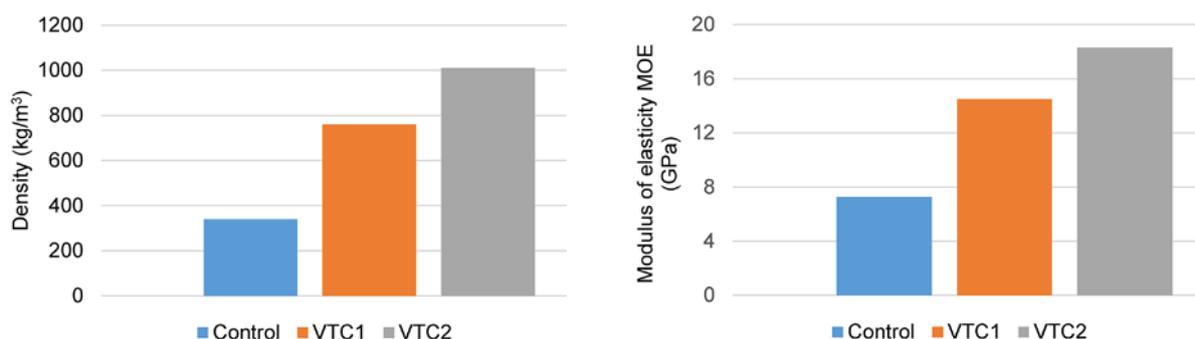


Fig. 1.

Densification of light-weight poplar wood by the VTC-process, i.e. heating and steaming in a closed-system at 170-200°C for about 7 min and compression. The compression ratio was 1:2.3 for control to VTC1 samples and 1:2.9 for the CTRL to VTC2 samples.

OBJECTIVE

The main objective of the present paper is to give a concise state-of-the-art presentation of the most recent developments in the field of densification of wood in product development.

THE DENSIFICATION PROCESS

The degree of densification of a piece of wood can be varied depending on what it is desired to achieve. To increase the gross density and thereby improve the overall mechanical properties of the wood material, all the cells throughout the thickness must be compressed and deformed (through-thickness densification). The gross density can theoretically be increased to a value close to that of the cell wall, i.e. 1500kg/m³. The energy consumption for this densification is very high, especially in the final phase of compression when the denser cells in the annual rings are to be deformed.

If the purpose is just to achieve a hard and abrasive-resistant surface, only the wood cells close to the surface need to be compressed and deformed (surface densification). Compared to through-thickness densification, surface densification offers several advantages. From a structural perspective, surface-densified wood has a higher material usage efficiency – imagine an I-beam versus a solid rectangular beam. For some products, better dampening characteristics are an asset. In addition, the compression and treatment to avoid the moisture-induced recovery of the densified wood cells back to their original shape need only affect the densified cells close to the surface and not the whole piece of wood. This may allow a faster, less energy-consuming, and thereby a less costly treatment process.

Regardless of how the densification itself is performed, the process basically always consists of three main phases: (1) softening of the wood material to be densified, (2) densification of the cells, and (3) elimination of the set-recovery. These phases interact with each other, but are here for simplicity described as separate processes. Other methods such as a modified wood welding technique (Vasiri et al. 2014; 2015) or self-bonding compression techniques (Cristescu et al. 2015a,b) can, however, be used for surface densification, but is not discussed further here.

Softening

Wood is a material that can be made plastic (or semi-plastic because it has areas with a crystalline molecular structure that are hard to plasticize), which means that wood can be softened and shaped so that it keeps its new shape after the plasticization is finished. The observed glass transition of wood occurs over a temperature range of ca. 50°C, and it is dependent on the moisture content and the time domain of the observed mechanical response. An increase in temperature or moisture content decreases the compressive modulus of wood. A combination of moisture and heat (hydro-thermal treatment) is therefore an effective way of softening wood, but chemical methods can also be used with excellent results, cf. Fig. 2. Nilsson et al. (2011) described a simple densification technique based on compressing Scots pine sawn timber in the radial direction, which shows no or little cell-wall fracture even if the densification is performed at 20°C at a moisture content of about 8%, i.e. un-plasticized. The densified wood was used as the surface layer of a multi-layer composite with a light-weight core. In the through-thickness densification of wood, however, plasticization treatment is an extremely important part of the process to soften the wood sufficiently in the specific region where densification is desired, and so that can without fracture withstand the compressive deformation.

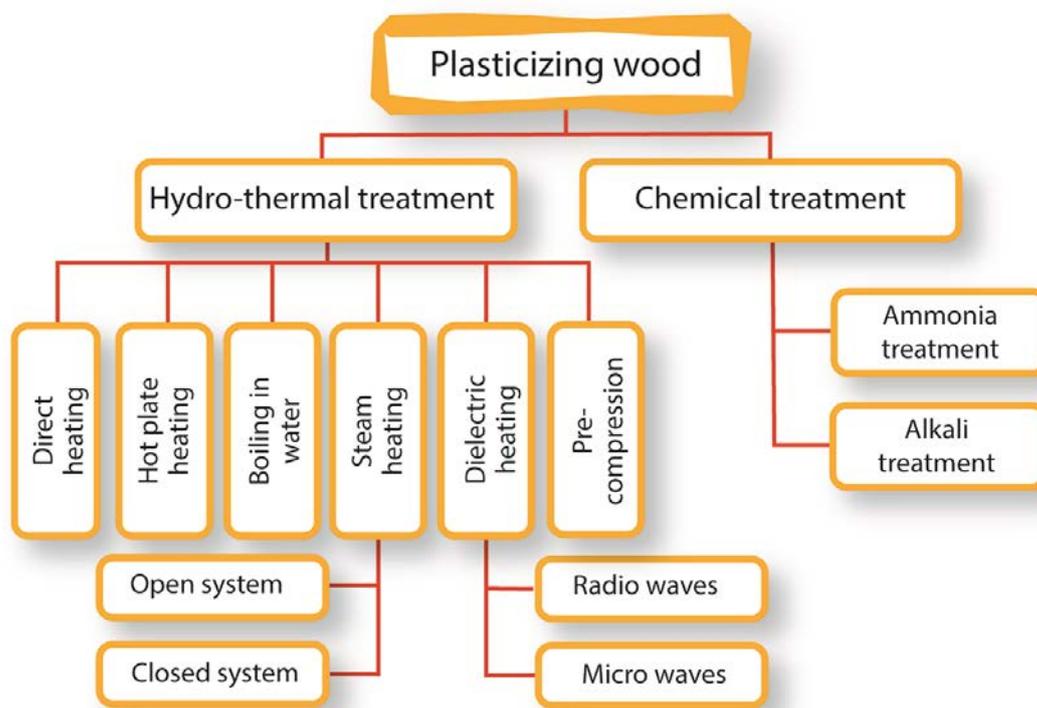


Fig. 2.
Classification of methods for making wood more flexible for shaping (Navi and Sandberg 2012).

Chemical treatment for plasticization can be as effective as hydro-thermal processes (Stojčev 1979). In one study (Rousek et al. 2015), plasticization by vacuum impregnation (0.2MPa) with ammonia gas at room temperature was compared to plasticization treatment with saturated steam at atmospheric pressure and a temperature of 100°C. Ammonia plasticization reduced the compression forces significantly and was sufficient for densification, but was slightly less efficient than saturated steam, Fig. 3. The ammonia plasticization effect could be improved by applying a higher gas pressure. The best plasticization effect was reached by combining these two methods.

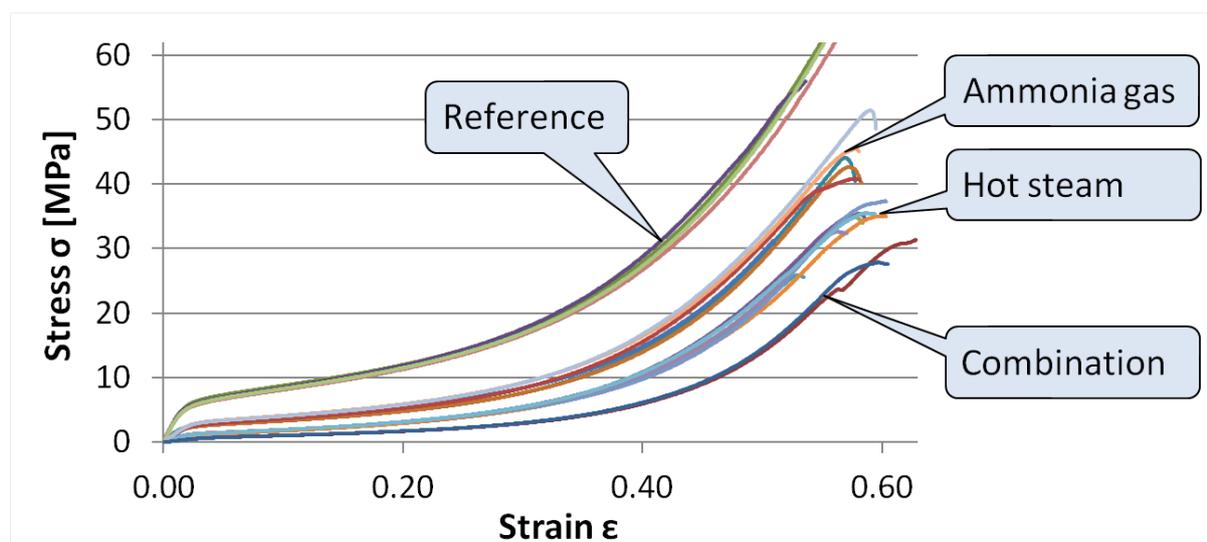


Fig. 3.
Stress-strain relationship for tangential compression of untreated beech (reference) and beech softened by ammonia gas, saturated steam or both in combination (Rousek et al. 2015).

Microwave heating is a time-reducing method of plasticization that has been studied by e.g. Norimoto and Gril (1989) and Dömény *et al.* (2014; 2017). Plasticization by MW heating has several advantages over

the conventional water vapour method, such as lower energy consumption, rapid heating of wood over the whole volume and application in continuous processes. In contrast, structural changes can occur during MW treatment and this may lead to changes in wood strength (Oloyed and Groombridge 2000). When microwave radiation is applied to the wood with a certain moisture content (MC), the water in the cell cavity absorbs the energy and is vaporized. The high internal steam pressure can cause the cell to rupture and generate micro cracks in the wood structure.

Densification

Densification can be achieved in one or more directions, but the process is mostly performed along one of the orthotropic axes, and the efforts are more successful for diffuse porous hardwoods than for softwoods. Densification can optimally be achieved mainly in the radial direction for softwoods and in the tangential direction for hardwoods with large aggregated rays. If e.g. Scots pine is densified along the tangential direction, the latewood (LW) of the annual rings spreads into the earlywood and forms waves or a zigzag pattern and the whole piece of wood may be crushed (Sandberg 1998a,b), Fig. 4. Densification in the radial direction, on the other hand, flattens the cells without any noticeable damage at the micro- or macroscopic level. In hardwoods with large aggregated rays, the same type of crushing phenomenon happens with the rays, Fig. 4 (Rousek et al. 2015).

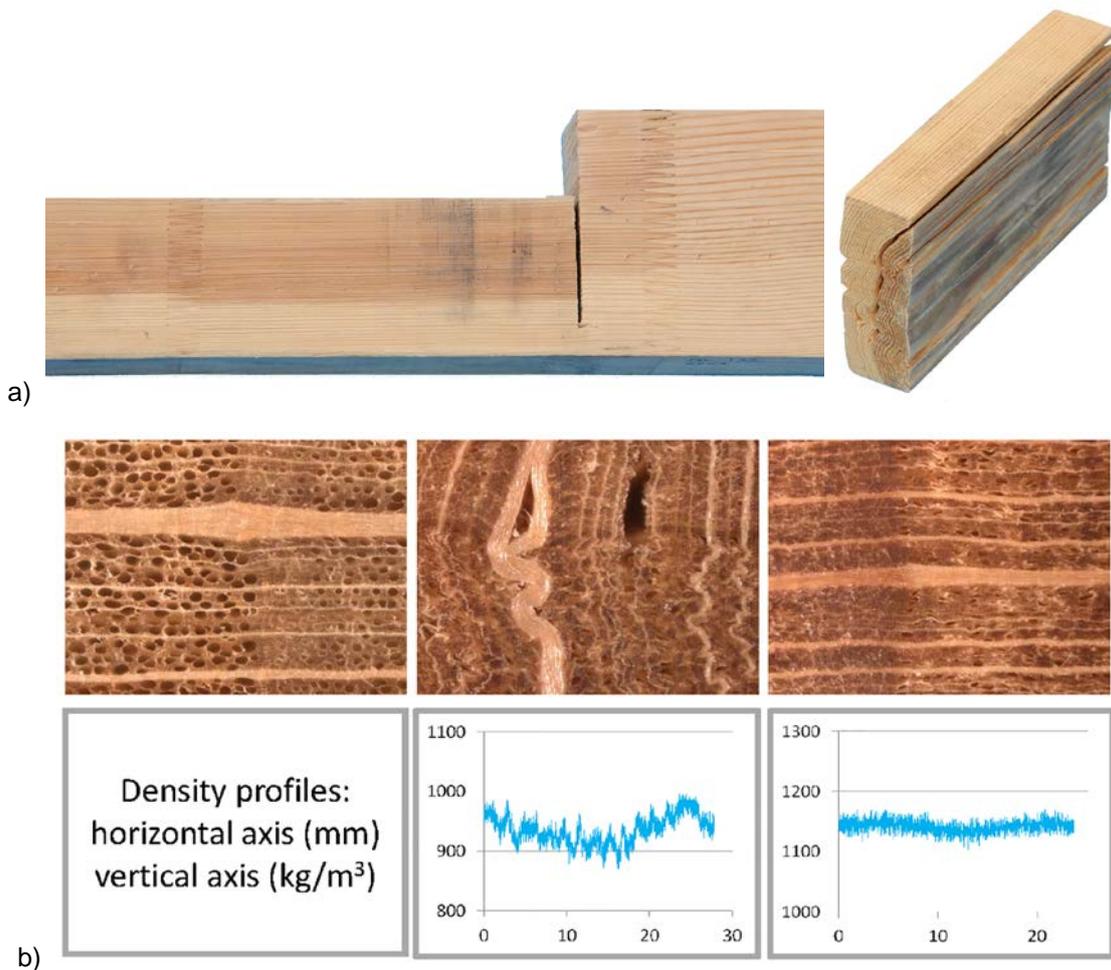


Fig. 4.

Densification of wood: a) Macro-views of densified Scots pine; densification in radial direction of a finger-jointed 50x100 mm board (left) and damage in a tangentially densified 22x75 mm board (right). b) Cross-section microscopic views of un-densified beech (left), and of beech densified in the radial (middle) and tangential (right) directions. The diagrams show the density profile through the cross-section after densification.

During transverse compressive loading, the typical stress-strain curve of wood has three distinct regions corresponding to three different types of cell deformation: 1) A linear elastic part, 2) a 'collapse' region where the stress is almost constant even at high strain, 3) a sharp increase in stresses due to contact

between the inner cell walls (cf. Fig. 3). Cellular collapse occurs by elastic buckling, plastic yielding or brittle crushing, depending on the test conditions and on the nature of the cell wall. It is also known that wood responds differently to radial and tangential compression. Rousek (2014) has described how the strain varies within beech under densification according to the annual ring orientation in its cross-section, Fig. 5.

In radial compression, earlywood primarily controls the elastic and plastic parts of the stress-strain response, while the final compaction stage is dominated by the elastic deformation of latewood. In the tangential direction, the final compression stage begins after buckling of the latewood layers. The compressibility of different wood tissues affects the distribution of void areas, and thus also the density distribution and mechanical properties of compressed wood (Kamke and Casey 1988). The location of the buckling of both the cell walls and the annual rings during transverse densification cannot be predicted precisely, as it depends on the morphology of the microstructure and degree of plasticisation.

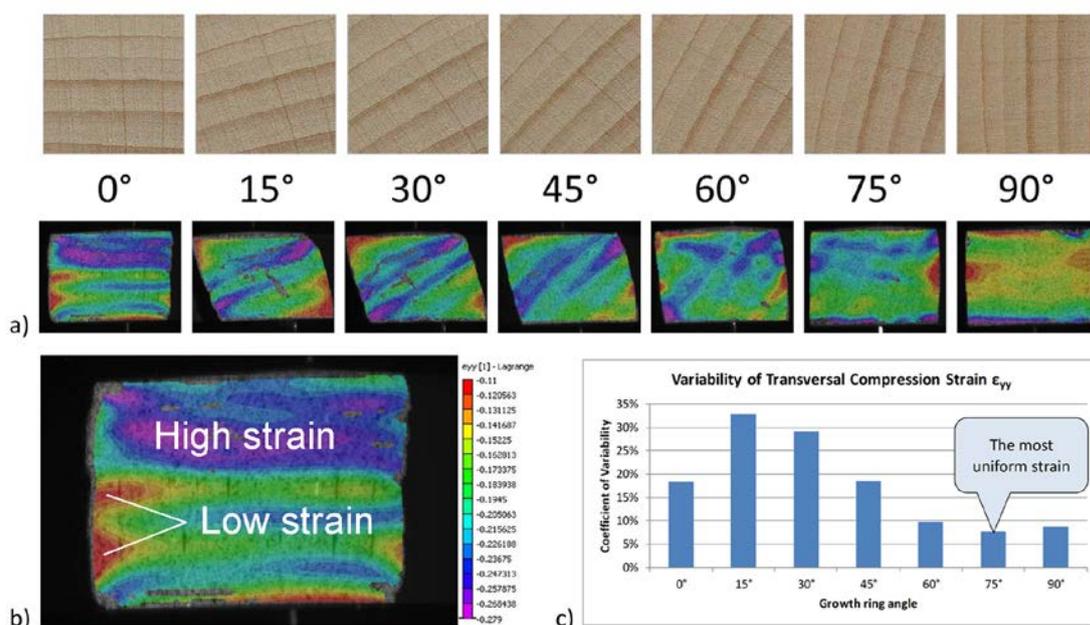


Fig. 5.

Densification of beech with different annual ring orientations in the cross section (upper): a) Digital Image Correlation (DIC) showing the strain at 26% compression, b) DIC showing the vertical strain ϵ_{yy} distribution in the radial compressed sample (0°), and c) the variability of the vertical strain (Lagrange) calculation.

Set Recovery

Already after early studies in this field, it was clear that the unmodified and densified wood cells would recover once they were exposed to moisture. This phenomenon is related to the shape memory effect of wood called "set-recovery" and is related to the release of internal stresses within the densified wood cells. In contrast to the slow set-recovery, elastic spring-back occurs immediately when the pressure is released after the densification process. In general, the elastic spring-back can be attributed to elastic strains within the chemical bonds of the wood material. Fig. 6 shows the different stages of the densification process on the cell level and when elastic spring-back and set-recovery occur (Neyses 2016).

Five general approaches to achieve a long-term fixation of the densified wood cells have been identified: (1) mechanical fixation by gluing or impregnation with adhesives, such as epoxy resin, or nailing, screwing, etc., (2) the formation of cross-links between molecules of the wood matrix by chemical modification: deactivation of the OH-sites, for example by BELMADUR process with DMDHEU, or (3) formaldehydation (fixing of H_2CO between two hydroxyls to obtain a strong chemical bond), (4) relaxation of internal stresses within the wood matrix during densification, for example by thermo-hydro-mechanical treatment, (5) reducing the accessibility of the cell wall to water.

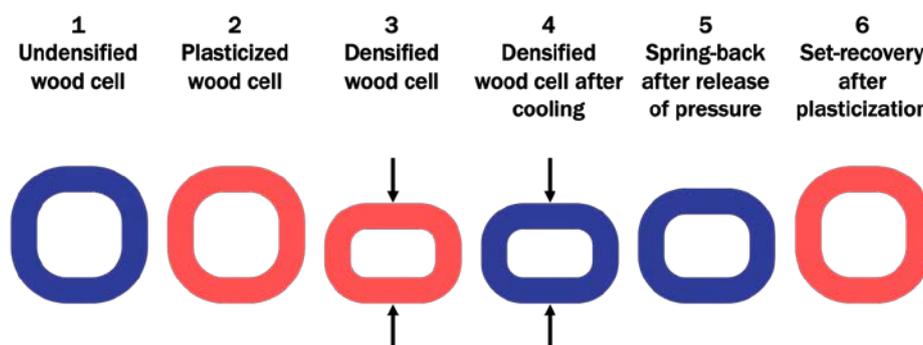


Fig. 6.

Cross-sectional view of a wood cell showing the nature of set-recovery and spring-back.

CASE STUDIES

Only a few wood densification applications have been industrialized. There are many reasons for this relatively low transfer of research to a fully up-scaled industrial production. Some of them are related to unsolved problems at the laboratory level on small-sized samples and others are related to the scaling-up processes in industry and the market. One particular reason was the lack of adequate consideration of the plasticization or stability of the products. The latter issue was solved by the development of resin-impregnated laminated and densified products, which are today being commercially produced, such as Delignit®, Dehonit®, Permawood® (also known as Lignostone®), Permali®, Insulam®, and Ranprex® (Kutnar et al. 2015).

Lignamon® is ammonified and densified beech wood. The treatment of beech by gasification with ammonia and simultaneous steaming plastifies the wood and allows densification of the material up to a density of about 1100kg/m³ with an appropriate increase in the mechanical properties, Fig. 7 (Pařil et al. 2014).

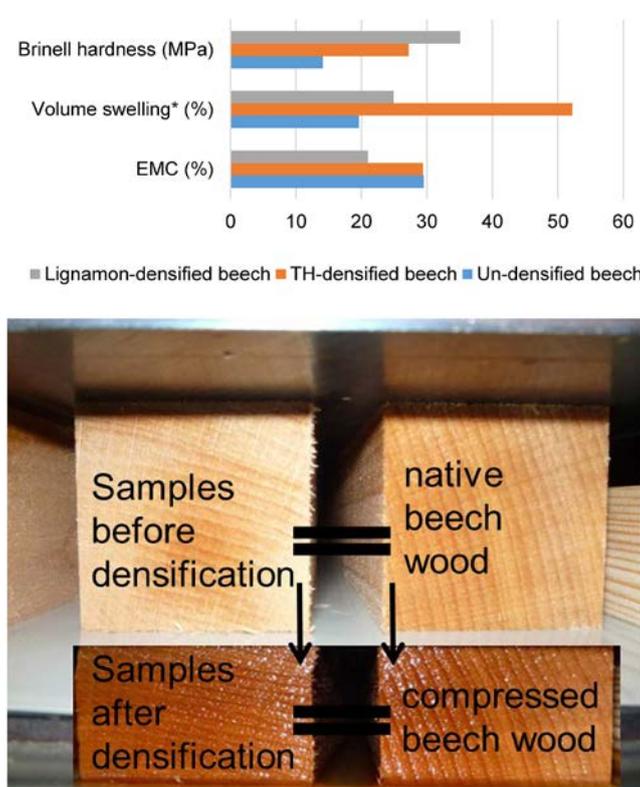


Fig. 7.

The Lignamon process delivered material with improved properties (upper) compared to control and steamed-densified beech (lower), Photo: Martin Brabec.

The production process combines vacuum-pressure impregnation of beech with ammonia vapour at a temperature of 90°C and densification. The process continues with drying, stabilization (180°C) and acclimatization (Stojčev 1979). The heating also increases the dimensional stability of the wood under moist conditions and can improve the durability compared with that of untreated beech (DC 5) up to durability class DC 1. Lignamon was presented as a material with considerably improved properties, but despite this the factory closed and Lignamon is no longer being produced.

Densification throughout the cross section may be a drawback in situations where, for example, it is desirable to maintain the low bulk density of wood, e.g. flooring applications. In such situations, an alternative approach is to densify only the first few millimetres beneath the surface of the wood, which places a great demand on the plasticisation and compression process stages to achieve the desired density profile within the wood. In one study, a high-speed continuous surface densification process was introduced, where the surface of solid Scots pine boards could be densified at speeds of up to 80 m/min by a roller pressing technique (Neyses et al. 2015). The present focus is to make the process more industrially adapted by integrating the roller pressing technique with various pre- and post-treatment methods to reduce negative effects such as set-recovery, colouring, or embossment of the surface, Fig. 8. A continuous process with steel belts instead of rollers has been studied by Sadatnezhad et al. (2017).

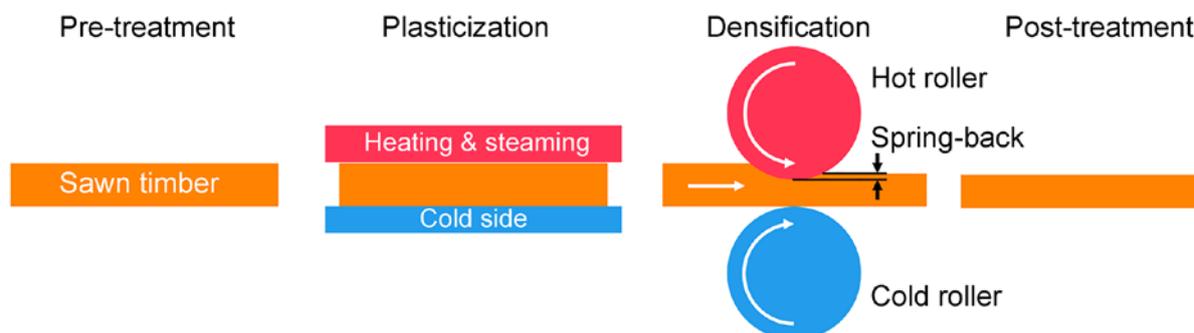


Fig. 8.
Schematic illustration of a continuous surface densification process.

Dömeny et al. (2017) used microwaves for plasticisation before and stabilization after the densification of beech. The densified beech was used as surface layer for a two-layered flooring panel where one of the layers was of densified wood and the other was particleboard, Fig 9.

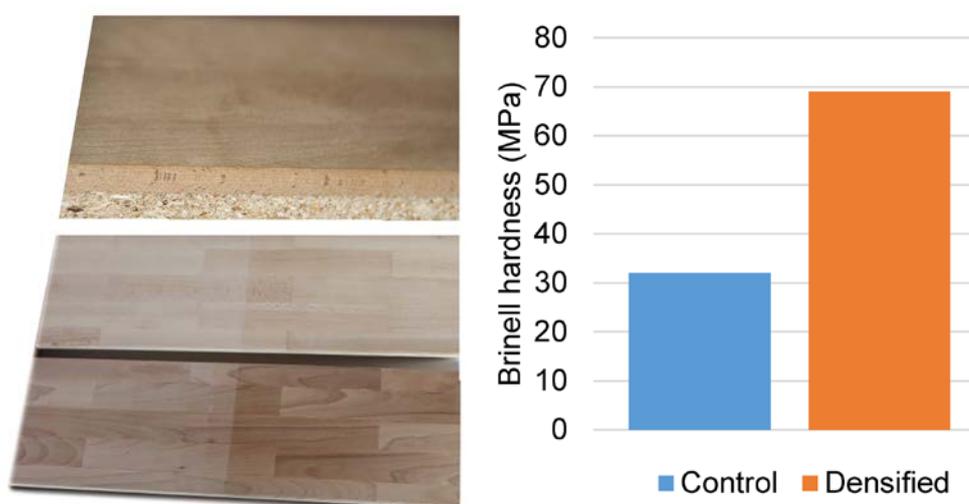


Fig. 9.
A layered flooring panel (left top) with densified beech as top surface (left bottom). The Brinell hardness value (right) of the densified beech is more than double that of the un-densified beech (left middle).

A densification process to manufacture wooden tubes out of sawn timber for load-bearing and conveying applications such as wind turbines has been presented by Kutnar et al. (2015), Fig. 9. The tubes can be fibre reinforced at the outer face to increase strength and protect the structure. Haller et al. (2013) also investigated the use of moulded tubes in aggressive environments. Hot, highly concentrated brine was conveyed at temperatures of up to 60°C. Compared to steel tubes, the spruce tubes did not show any noticeable erosion after 4 weeks' exposure, whereas the steel tubes had been considerably affected.

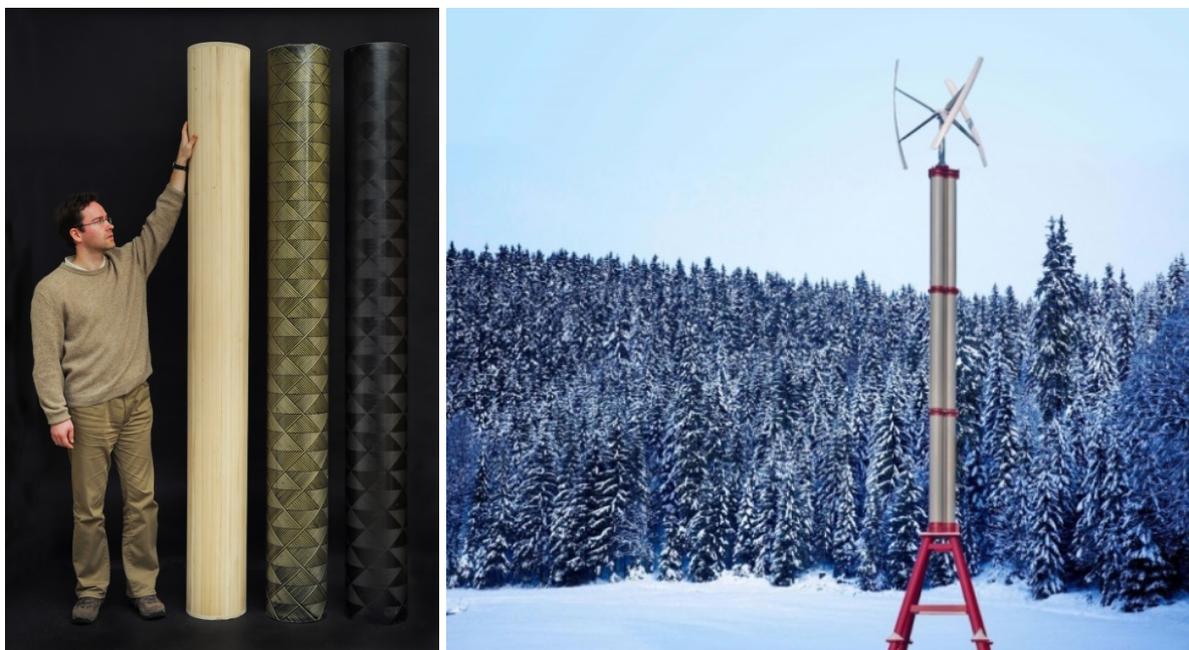


Fig. 9.

Moulded tubes from Norway spruce (left) with a length of 250 cm, a diameter of 28 cm, and a tube-wall thickness of 2 cm, and to the right a wind-power plant from fibre-reinforced moulded tube (Kutnar et al. 2015).

CONCLUSIONS

Research on wood densification is intensive in several research groups in Europe trying to understand the plasticization and densification of wood, and to develop processes and products that are environment-friendly. The focus has changed from through-thickness densification towards the densification of specific regions where the increase in density is needed in a specific product, e.g. surface densification for flooring. The main challenges for the future are finding a fast and environment-friendly method for the elimination of the set-recovery and scaling up to profitable industrial applications.

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REFERENCES

Cristescu C, Sandberg D, Ekevad M, Karlsson O (2015a) Influence of pressing parameters on mechanical and physical properties of self-bonded laminated beech boards. *Wood Material Science and Engineering* 10(2):205-214.

- Cristescu C, Neyses B, Sandberg D, Söderström O (2015b). Modeling of the temperature distribution in self-bonded beech-veneer boards during hot pressing. *Pro Ligno* 11(4):97-103.
- Dömény J, Koiš V, Zapletal M (2014) Application of microwave treatment for the plasticization of beech wood (*Fagus sylvatica* L.) and its densification for flooring system purposes. *BioResources* (9)4:7519-7528.
- Dömény J, Čermák P, Koiš V, Tippner J, Rousek R (2017) Density profile and microstructural analysis of densified beech wood (*Fagus sylvatica* L.) plasticized by microwave treatment. *European Journal of Wood and Wood Products*. DOI: 10.1007/s00107-017-1173-z.
- Haller P, Putzger R, Wehsener J, Hartig J (2013) Formholzrohre – Stand der Forschung und Anwendungen (Molded wood pipes - state of research and applications). *Bautechnik* 90:34-41.
- Hornicek S, Rademacher P, Kutnar A, Kamke FA, Rousek R (2015) Selected physical properties of viscoelastic thermal compressed wood from fast growing poplar. *Proceedings InWood2015: Innovations in wood materials and processes, Brno, Czech Republic*, pp. 28-29.
- Kamke FA, Casey LJ (1988) Fundamentals of flakeboard manufacture: internal-mat conditions. *Forest Products Journal* 38(6):38-44.
- Kutnar A, Sandberg D, Haller P (2015) Compressed and moulded wood from processing to products. *Holzforschung* 69(7):885-897.
- Navi P, Sandberg D (2012) Thermo-hydro-mechanical processing of wood. Lausanne, Presses Polytechniques et Universitaires Romandes, pp. 376.
- Neyses B (2016) Surface-densified wood – From laboratory-scale towards a competitive product. Licentiate thesis, Wood Science and Engineering, Luleå University of Technology, Sweden.
- Neyses B, Sandberg D, Hagman O, Wålinder M (2015) Development of a continuous wood surface densification process with a reduced environmental impact. *Proceedings of the 1st COST Action FP1407 Conference, August 25-26, Koper, Slovenia*, pp. 30-31.
- Nilsson J, Johansson J, Kifetew G, Sandberg D (2011) Shape stability of modified engineering wood product subjected to moisture variation. *Wood Material Science & Engineering* 6(3):132-139.
- Norimoto M, Gril J (1989) Wood bending using microwave heating. *Journal of Microwave Power and Electromagnetic Energy*, 24:203-212.
- Norimoto M, Ota C, Akitsu H, Yamada T (1993) Permanent fixation of bending deformation in wood by heat treatment. *Wood Research* 79(1):23-33.
- Oloyede A, Groombridge P (2000) The influence of microwave rating on the mechanical properties of wood. *Journal of Material Processing Technology* 100(1):67-73.
- Pařil P, Brabec M, Maňák O, Rousek R, Rademacher P, Čermák P, Dejmal A (2014) Comparison of selected physical and mechanical properties of densified beech wood plasticized by ammonia and saturated steam. *European Journal of Wood and Wood Products* 72(5):583-591.
- Rademacher P, Nemeth R, Fodor F, Rousek R, Hornicek S, Pařil P, Baar J, Paschová Z, Kutnar A, Koch G (2016) Extended utilization of wood material – usage from native properties to wood modification. *Proceedings 27th International Conference on Wood Modification and Technology*.
- Rademacher P, Meier D, Hofmann T, Pařil P, Baar J, Sablík P, Čermák P, Paschova Z, Rousek R, Koch G, Schmitt U, Melcher E, Nemeth R (2016) Property improvement of wood from fast growing plantations due to wood modification with renewable modification agents. *Proceedings 25th International Scientific Conf.: New Materials and Technologies in the functions of Wood Products*.
- Rousek R (2014) Comparison of hydro-thermal and chemical method of wood plastification. 68th Forest Products Society Convention, 10-13 August, Qubec City, Canada, (Poster).
- Rousek R, Rademacher P, Brabec M, Dejmal A, Hornicek S, Baar J (2015) Beech wood modification with ammonia gas – improved properties. *Pro Ligno* 11(4):230-238.
- Sadatnezhad SH, Khazaeian A, Sandberg D, Tabarsa T (2017) Continuous surface densification of wood: A new concept for large-scale industrial processing. *BioResources* 12(2):3122-3132.
- Sandberg D (1998a) The influence of isostatic compression on the cellwall structure of wood. KTH – Royal Institute of Technology, Report TRITA-TRÅ 98-35, ISSN1104-2117, pp. 30.

Sandberg D (1998b) Value activation with vertical annual rings – material, production, products. PhD Thesis, KTH – Royal Institute of Technology, Report TRITA-TRÅ 98-36, ISSN1104-2117, pp. 168.

Sandberg D, Haller P, Navi P (2013) Thermo-hydro-mechanical (THM) wood treatments. Wood Material Science and Engineering 8(1):64-88.

Stojčev A (1979) Lignamon - zušlechtěné dřevo: Výroba, vlastnosti a použití. [Lignamon - treated wood: Production, properties and uses], 1st edition. Praha: SNTL - Nakladatelství technické literatury. ISBN 06-099-79.

Vaziri M, Berg S, Sandberg D, Tavakoli Gheinani I (2014) Three-dimensional finite element modelling of heat transfer for linear friction welding of Scots pine. Wood Material Science and Engineering 9(2):102-109.

Vaziri M, du Plessis A, Sandberg D, Berg S (2015) Nano X-ray tomography analysis of the cell-wall density of welded beech joints. Wood Material Science and Engineering 10(4):368-372.