

Fakultät Wissenschaftszentrum Weihenstephan für Ernährung, Landnutzung und Umwelt Lehrstuhl für Holzwissenschaft

## Design of Tree Bark Insulation Boards: Analysis of Material, Structure and Property Relationships

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(Marcus Tullius Cicero)

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## Summary

Efforts to reduce the heating and cooling energy demand of buildings have led to an increased use of insulation materials. Insulation materials come up for a significant ratio of embodied energy in buildings and are therefore important for ecological considerations. Building materials based on renewable resources are beneficial to sustainable constructions because their use often has a favorable CO<sub>2</sub>-balance from a life-cycle perspective. Ligno-cellulosic plants are an important source for building materials, and especially timber has a long tradition as a construction material.

Currently, the wood-processing industry is facing an increasing shortage of resources caused by the promotion of biomass-based energy. Therefore, a more efficient use of available forest resources has become increasingly important.

So far, the tree component bark has barely been used. It is a byproduct of the sawmill industry and of wood chip production in pulp, paper, and wood composite industries. It is predominantly used as a source for internal process-energy generation. It has been neglected that bark exhibits very interesting properties as a material, most notably a relatively low density, low thermal conductivity, good fire resistance, a high amount of extractives, and accessory components preventing the attack of microorganisms.

The present thesis focuses on the thermal potentials of tree bark, which suggest its application as a thermal insulation material. This dissertation strives to evaluate the feasibility of this concept. More specifically, the aim of this dissertation is to test whether light particle-based bark insulation boards can be produced, and to evaluate the specific mechanical and physical properties relevant to construction use. Additionally, alternative resin systems based on renewable resources shall be analyzed in terms of their suitability for bark board production. The aim, in this respect, is to replace commonly used condensation resins, which are petrol-based. Finally, the structural composition of bark insulation boards is to be analyzed and potential optimization sources shall be determined. Furthermore, it shall be discussed whether a theoretical, structure-based model can be used for the prediction of a bark panel's thermal conductivity, which would allow to study the effects of structure variations before production.

To evaluate the suitability of tree bark for thermal insulation, bark panels were produced on a laboratory scale and were characterized focusing on physical-mechanical properties (compressive resistance, modulus of elasticity, modulus of rupture, internal bond, tensile strength, thickness swelling, water absorption, thermal conductivity, thermal diffusivity). Additionally, the insulation performance of bark insulation layers under transient temperature conditions was studied on a real-size wall element. Time-dependent temperature profiles were discussed. In each case, board properties were contrasted with those of insulation materials available on the market, to evaluate

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their suitability for specific applications. Furthermore, the bark insulation panels were optimized by replacing the widely used urea-formaldehyde resin with a more ecological tannin-hexamine resin. In addition, the panel production was improved with regard to tannin type, resin content, pressing time, and amount of hardener. Bark insulation panels were characterized in terms of their structural composition using X-ray computed tomography. Optimization potentials in panel structure were evaluated by contrasting the thermal conductivity of panels with horizontal and vertical particles. Finally, the insights were abstracted in order to allow for theoretical optimization. A discrete model based on finite differences was developed to predict a bark panel's thermal conductivity based on its structural composition.

Results showed that bark is indeed suitable as an insulation material as far as physical-mechanical properties of bark insulation panels are concerned. Their physical-mechanical characteristics seemed to be adequate compared with other commonly available insulation materials. Regarding the panel's thermal conductivity (0.05–0.08 W/(m\*K)), it was learned that it is not as low as with very light insulation materials (e.g., polystyrene, mineral wool), but that its thermal diffusivity is very low. The material can store a lot of heat energy and slowly conducts it. That makes bark insulation boards particularly suitable for heat storage-active insulation layers.

A tannin-hexamine resin could successfully replace a urea-formaldehyde condensation resin for particle binding. The use of mimosa tannin instead of quebracho tannin resulted in superior internal bond, lower thickness swelling, and water absorption. Hexamine remains the only petrol-based component in the panels produced. It could be shown that hexamine is necessary for the polymerization of the resin, but that it can be reduced to a very low amount without deteriorating panel properties.

Orienting bark particles parallel to the panel plane is likely to significantly lower the panels' thermal conductivity. These findings were substantiated using X-ray computed tomography to illustrate the panels' interior structure. Bark is a highly inhomogeneous material. Consequently, a special thresholding algorithm based on ANOVA had to be applied in the digital image analysis of X-ray tomograms to successfully distinguish between void, inner bark, and outer bark. Segmented images were used to study the pore size distribution in panels, discovering that especially the macro-void structure and distribution can be somewhat influenced in the pressing process. Finally, a model based on finite differences proved to be suitable for a theoretical description of the heat flow processes within a panel. Heat flow could be studied on a voxel-level and the global thermal conductivity of the panel could be determined. The model was found to have low deviations from real measurements and to constitute a valuable basis for the optimization and production of efficient bark insulation panels.

## Zusammenfassung

Die Anstrengungen, den Heiz- und Kühlenergiebedarf von Gebäuden zu reduzieren, haben zu einem gestiegenen Bedarf an Dämmmaterialien geführt. Dämmmaterialien beinhalten einen beträchtlichen Teil der in Gebäuden verbauten grauen Energie und sind darum auch für ökologische Betrachtungen bedeutsam. Baumaterialien aus erneuerbaren Ressourcen erweisen sich als vorteilhaft für nachhaltige Konstruktionen, da ihr Einsatz in einer Lebenszyklusbetrachtung oft durch eine vorteilhafte CO<sub>2</sub>-Bilanz gekennzeichnet ist. Lignocellulosäre Pflanzen sind eine wichtige Quelle für Baumaterialien und besonders Holz hat eine lange Nutzungstradition im Bauwesen.

Im heutigen Marktumfeld ist die Holzindustrie mit einem weit greifenden Rohstoffmangel konfrontiert — hervorgerufen durch die Ausweitung der Biomasse-Energieerzeugung aus Holz. Darum kommt der effizienten Nutzung der verfügbaren forstlichen Ressourcen steigende Bedeutung zu.

Baumrinde wird bis jetzt kaum stofflich veredelt eingesetzt. Sie ist ein Nebenprodukt der Schnittholzerzeugung in der Sägewerksindustrie und der Hackguterzeugung in der Papier- und Holzwerkstoffindustrie. Hier wird sie vor allem zur Erzeugung interner Prozessenergie eingesetzt. Dabei bleiben interessante Materialeigenschaften der Rinde ungenutzt. Beispielsweise besitzt Rinde eine relativ geringe Dichte, eine niedrige Wärmeleitfähigkeit, gute Feuerbeständigkeit, einen hohen Extraktstoffgehalt und Inhaltsstoffe, die dem Befall durch Mikroorganismen entgegenwirken.

Die vorliegende Arbeit thematisiert die Wärmedämmeigenschaften von Rinde und evaluiert deren Eignung als Wärmedämmstoff. Im Rahmen dieser Dissertation wird diese Nutzungsmöglichkeit diskutiert. Die spezifischen Ziele dieser Dissertation sind, leichte partikelbasierte Rindendämmplatten herzustellen und die für den etwaigen technischen Einsatz relevanten mechanischen und physikalischen Materialeigenschaften zu erheben. Darüber hinaus sollen alternative, auf erneuerbaren Ressourcen basierende, Bindemittel in Hinblick auf ihre Eignung für die Rindendämmplattenherstellung evaluiert werden. Das Ziel in diesem Zusammenhang ist der Ersatz der weitgehend verwendeten Kondensationsharze, da diese aus fossilen Rohstoffen gewonnen werden. Schließlich soll die Struktur von Rindenplatten analysiert und potenzielle Optimierungsmöglichkeiten abgeleitet werden. Zusätzlich soll erhoben werden, inwieweit ein theoretisches, struktur-basiertes Modell zur Vorhersage der Wärmeleitfähigkeit geeignet ist, um vorab die Effekte von Strukturveränderungen in Rindenplatten bewerten zu können.

Um die Eignung von Baumrinde als Wärmedämmmaterial zu bewerten, wurden Rindenplatten im Labormaßstab hergestellt. Deren physikalisch-mechanische Eigenschaften (Druckfestigkeit, Elastizitätsmodul, Biegefestigkeit, Querzugfestigkeit, Zugfestigkeit, Dickenquellung, Wasseraufnahme, Wärmeleitfähigkeit, Temperaturleitfähigkeit) wurden erhoben. Zusätzlich wurde das Dämmverhalten unter instationären Temperaturbedingungen an einem bauteilgroßen

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Referenzobjekt gemessen und zeitabhängige Temperaturprofile ermittelt. Die Platteneigenschaften wurden jenen von am Markt erhältlichen Dämmmaterialien gegenübergestellt, um die Eignung für spezifische Anwendungen abzuklären. Darüber hinaus wurden die Rindendämmplatten verbessert, indem der industriell häufig eingesetzte Harnstoff-Formaldehydharz Klebstoff durch einen, auf erneuerbaren Ressourcen basierenden, Tannin-Hexamin Klebstoff ersetzt wurde. Tanninart, Klebstoffgehalt, Presszeit und Härteranteil wurden variiert und die optimalen Rezepturparameter definiert. Die Rindenplatten wurden bezugnehmend auf ihren strukturellen Aufbau mittels Röntgen-Computertomographie charakterisiert. Dabei wurden Optimierungspotenziale in der Plattenstruktur erhoben, indem Platten mit horizontal und vertikal orientierten Partikeln untersucht wurden. Die gewonnenen Erkenntnisse wurden abstrahiert, um eine theoriegeleitete Optimierung zu ermöglichen. Dazu wurde ein diskretes, strukturbasiertes Modell zur Vorhersage der Wärmeleitfähigkeit abgeleitet.

Es zeigte sich, dass Baumrinde unter Berücksichtigung der mechanischen und thermischen Eigenschaften tatsächlich als Dämmmaterial geeignet ist. Die physikalisch-mechanischen Eigenschaften sind vergleichbar mit jenen von Standarddämmstoffen. Die Wärmeleitfähigkeit der Rindendämmplatten (0.05–0.08 W/(m\*K)) ist höher als jene der sehr leichten Dämmstoffe (z.B. Polystyrol, Mineralwolle), allerdings ist die Temperaturleitfähigkeit vergleichsweise sehr gering. Damit ist das Material geeignet, viel Wärmeenergie zu speichern und diese langsam zu leiten, was Baumrinde für wärmespeicheraktive Dämmschichten besonders geeignet macht.

Der eingesetzte Tannin-Hexamin Klebstoff ist geeignet, um den Kondensationsharz-Klebstoff zu ersetzten. Mimosa-Tannin erwies sich im Vergleich mit Quebracho-Tannin als vorteilhaft, da die jeweils produzierten Labor-Platten bessere Querzugfestigkeit und Feuchtebeständigkeit aufwiesen. Die einzige verbliebene, auf fossilen Rohstoffen basierende, Komponente in den Rindendämmplatten ist das Hexamin. Es konnte gezeigt werden, dass das Hexamin für die Polymerisation des Klebstoffs erforderlich ist, dass allerdings die eingesetzte Menge auf ein sehr geringes Maß reduziert werden kann, ohne die Platteneigenschaften zu verschlechtern.

Platten mit parallel zur Plattenebene ausgerichteten Partikeln weisen eine deutlich geringere Wärmeleitfähigkeit auf, als solche mit orthogonal zur Plattenebene orientierten Partikeln. Diese Erkenntnis wurde mittels Strukturuntersuchungen unter Einsatz von Computertomographie untermauert. Rinde ist ein sehr heterogenes Material und darum musste ein spezieller, auf der Varianzanalyse basierender, Segmentierungs-Algorithmus eingesetzt werden, um die Materialphasen (Luft, Innenrinde, Außenrinde) in den Tomographie-Aufnahmen abgrenzen zu können. Die segmentierten Bilder wurden verwendet, um die Porengrößenverteilung in den Platten zu evaluieren, und es wurde erkannt, dass vor allem die Struktur und Verteilung der größeren Poren in einem gewissen Rahmen im Pressprozess beeinflusst werden kann.

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Schließlich zeigte sich, dass das entwickelte, diskrete Modell geeignet ist, um die Wärmeflüsse in der Platte theoretisch zu beschreiben. Dazu wurden die Wärmeflüsse in jedem Voxel berechnet und die globale Wärmeleitfähigkeit abgeleitet. Die modellierten Ergebnisse zeigten geringe Abweichungen von den realen Messungen und das Modell erwies sich als geeignete Basis für die weitere Optimierung und Produktion von effizienten Rindendämmplatten.

## 1 Introduction

#### 1.1 Motivation

Wood is an important source of bio-fiber which has been used by humankind since ancient times. It is an important raw material for many industries and sectors (e.g., energy, pulp and paper, civil engineering, furniture design) and is vital for a globally growing society (Cardarelli 2008). The material has a long history of specialized use in various fields of human life, which has also resulted in highly developed craftsmanship (Teischinger 2010, Paulitsch and Barbu 2015). The potential for the use of bio-composites in industry is enormous because of their total or partial derivation from renewable resources (John and Thomas 2008). Knowledge of the advantages of different species and the effects of growing regions, climate, age, etc. was first gained by trial and error, but was later substantially driven by research (Young 2007).

Wood resource availability is decreasing. Reasons might be a growing world population, a rising standard of living accompanied by a higher demand for resources, reduced product life spans and, most importantly, a rise in wood use for energy production (Barbu 2011). Forest area has globally declined in the last three decades and has forced considerations about sustainable resource use (Whiteman 2014). As the limits of sustainable wood use have been partially reached, alternative use scenarios have to be discussed (Abolins and Gravitis 2015). Moreover, the sustainability of building materials is getting increasingly important (Pargana et al. 2014), which forces wood composite producers to rethink their resources. Additionally, international contracts are forcing industry and public institutions to reduce CO<sub>2</sub> emissions in order to limit the greenhouse effect. Enhanced use of wood from sustainably managed forests could help to mitigate climate change (especially global warming), as those forests function as carbon sinks (Gustavsson et al. 2006). Critics warn that a higher standard of living could result in irreparable damage for humans, their society and natural habitat (Wegener 1994). To avoid a lack of wood in future, its use has to be optimized by promoting cascade usage (Gärtner et al. 2012, Höglmeier et al. 2015) and exploiting alternative material sources (Teischinger 2007, Paulitsch and Barbu 2015). While knowledge in wood science and technology was expanded substantially, the development and industrial use of bark was neglected (Ogunwusi 2013).

Nature is inherently complex and systems are optimized to cater for various needs. Also, resources are used highly efficiently in natural processes, and therefore material development can be "bio-inspired" by analyzing natural approaches to problem solving (Paris et al. 2010, Ugolev 2014). In natural materials the understanding of relationships between structure and properties at different scales might be a valuable input to material engineering (Cranford and Buehler 2012). Looking at tree bark in more detail, its smart design becomes obvious.

Globally, roughly 1.6 billion m<sup>3</sup> of wood are harvested for industrial purposes every year. Considering that roughly 10 % of a tree is bark, a bark volume of 160 million m<sup>3</sup> per year (Xing et al. 2007b) is available. Bark is a highly optimized material by nature. On a tree it protects the sensitive vascular cambium from heat, sharp frost, mechanical damage, and attacks from microorganisms (Nicolai 1986, Vaucher 1997, Naundorf et al. 2004, Bauer et al. 2010). Whilst bark was an important raw material until the middle of the 20<sup>th</sup> century (e.g., roofing [Mooslechner 1999], resource for leather tanning [Mavlyanov et al. 2001]), it has become a classic byproduct in timber processing. Today bark is primarily burned and rarely used for value-added products (Harkin and Rowe 1971, Naundorf et al. 2004).

Especially in civil engineering, a lot of energy consumption is attributed to embodied energy in building materials and energy for building operation. The less energy is lost with highly insulated modern constructions, the higher is the proportion of the total energy input related to gray energy within materials (Zeumer et al. 2009, Pargana et al. 2014). Consequently, an ongoing trend is to choose materials strategically with regard to "gray energy" and to find constructive solutions to minimize a building's total energy consumption. These considerations favor bio-based, renewable materials, whose CO<sub>2</sub> balance is beneficial compared to fossil materials. Their cell walls are built during photosynthesis, where CO<sub>2</sub> and water are converted into glucose and oxygen, powered by the energy of sunlight (Werner and Richter 2007).

The idea of this dissertation is to use tree bark according to its inherent, natural purpose, by exploiting its protective and insulating properties in using it for the production of thermal building insulation materials. Several considerations ensure the adequacy of this approach:

- A huge amount of energy is attributed to the operation of buildings. 24 % of the global energy used comes from the energy demand of buildings, in the EU this share is 37 %. Key energy end uses are heating, ventilation, air-conditioning (HVAC), lighting, and appliances, coming up for 85 % of the total energy use in a building. As HVAC accounts for roughly 50 % of the energy consumption, insulation efforts are likely to be effective in reducing the total energy consumption (Pérez-Lombard et al. 2008).
- The energy demand of buildings has to be decreased by legal requirements (European Union 2010), which will potentially increase insulation material use.
- Natural resource-based materials are to be preferred because of their beneficial CO<sub>2</sub>balance (Lippke et al. 2004).
- Bark is a naturally optimized insulation material (Holdheide and Huber 1952, Vaucher 1997, Rosell et al. 2014).
- Bark is available in large quantities and is a byproduct with no existing substantial technical exploitation in large quantities (BMLFUW 2014b). However, first attempts have been made

in bark biorefineries to upgrade different constituents of bark to value-added bio-based products (Le Normand et al. 2014, Moncada et al. 2016).

- Historical examples showed that tree bark has good properties for specific purposes (e.g., tanning [Mavlyanov et al. 2001], roofing [Mooslechner 1999], paper making [Peters et al. 1987], nutritional purposes [Rautio et al. 2014]).
- Contemporary bark use for niche-applications is promising (e.g., bark textiles, leather substitutes [Heintz 2015] or decorative wall claddings [Egger 2014]).

This thesis is structured as a cumulative dissertation. First, a general introduction to the topic will be given in Chapter 2, followed by a brief state of the art regarding insulation materials based on renewable resources and the potentials of tree bark in Chapter 3. Six publications in Chapter 4 (references can be found in the appendix) represent the core piece of this dissertation. They deal with the following topics:

- Substantial bark use as insulation material (Publication 1).
- Using bark as a heat insulation material (Publication 2).
- Density-related properties of bark insulation boards bonded with tannin hexamine resin (Publication 3).
- Analyzing wood bark insulation board structure using X-ray computed tomography and modeling its thermal conductivity by means of finite difference method (Publication 4)
- Effects of different flavonoid extracts in optimizing tannin-glued bark insulation boards (Publication 5).
- Evaluation of relationships between particle orientation and thermal conductivity in bark insulation board by means of CT and discrete modeling (Publication 6).

Finally, the synthesis in Chapter 5 will summarize the core findings of this thesis, propose discussion of the results against the background of the state of the art, and it will point out suggestions for further research.

### 1.2 Main research objectives

With a rising awareness of the need for highly insulated exterior walls and roofs in building constructions, the importance of insulation materials is rising (Pfundstein et al. 2007). Natural (based on renewable resources) insulation materials play an important role in this regard and a huge variety of different resources, including wood fiber, flax, straw, cellulose, and others (Fachagentur Nachwachsende Rohstoffe e. V. 2012, Barbu et al. 2014), have been used in building insulation within the last years.

The main aim of this work is to evaluate whether tree bark is also a suitable resource for the production of insulation materials. In this dissertation, the basis for the production and application of insulation panels made from tree bark shall be laid. Generally, bark is directly burned where it occurs. Exceptions are limited to niche applications without significant market penetration so far. Thus, it is a material that is rarely used for high value products, even though its natural properties are very promising. It serves as a tree's natural protective layer and is therefore a highly optimized material by nature.

In order to use a resource technically, it has to be processed into a product. Nowadays bark is available at production sites in particles of different sizes, contamination, moisture content, and sometimes of different tree species. Although wood particles have been used for the production of particleboard for a long time, and it has been proposed many times to also use bark as a resource for particleboard (e.g., Schneider and Engelhardt 1977, Gupta et al. 2011), it is not clear whether pure and light bark particleboard can be produced. Moreover, the stability of particleboard increases with rising density. Keeping in mind that the aim of this work is to produce insulation materials, whose thermal conductivity is low when material density is low, the discrepancy between mechanical stability and insulation properties has to be discussed. Therefore, it is the first sub-aim of this work to discuss the technical, production-oriented aspects of bark insulation board, focusing especially on the raw material bark and its properties when accumulated in the wood industry. Moreover special interest is given to the process of board production out of particles.

When developing new materials, these need to be characterized with regard to many aspects, namely end-user requirements, regulatory requirements, corporate requirements, and technical requirements (Rounds and Cooper 2002). The technical use of a natural resource requires its characterization in terms of physical-mechanical properties, to allow for industrial applications. The physical-mechanical board properties shall be analyzed in order to characterize the material and to highlight advantages and disadvantages. Most important in this context is to measure characteristic insulation panel properties like panel stability, moisture resistance, heat insulation properties, and behavior under stationary and transient temperature conditions. The findings have to be discussed in comparison with commonly available insulation materials in order to address the suitability of a

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bark composite for insulation purposes. To enhance the credibility of the results obtained, the findings on laboratory scale are to be verified on a real-size wall element.

Thirdly, the issue of processing bark boards, with a special focus on bonding, is covered in this work. Condensation resins are the predominant resin system used in the wood-based composite industry (Paulitsch and Barbu 2015). Since the focus of the presented bark material is on a bio-based, renewable, and ecological material, these petrol-based resins have to be replaced by a bio-based resin. Consequently, panel properties and production parameters have to be verified and optimized with regard to the specific board constituents. Additionally, the tannin-based resins that are used in this work require a petrol-based hardener (hexamine). In order to limit the amount of this hardener in the composite, the polymerization processes are studied and the effect of the hardener content on the panel properties is discussed.

Finally, the structure of a material greatly influences its properties and is therefore a parameter when specifically designing a material (Lakes 1993). In this work, the interior structure of bark-based insulation boards has to be revealed using non-destructive evaluation, which is why X-ray computed tomography is applied. As the obtained images do not show the material phases clearly, due to high density inhomogeneity, a special thresholding procedure is discussed in this work. Furthermore, the relationships between panel structure and thermal conductivity are analyzed. To understand heat flow on a micro level, a discrete model for heat flow in bark composites is developed in this thesis and used to theoretically discuss structure-based optimization potentials for the bark material.

Based on the described knowledge gaps, the following subordinated research questions are in the primary focus of this work. Detailed research questions can be found in the publications in the appendix.

- Is it possible to produce lightweight (< 500 kg/m<sup>3</sup>) bark particleboard, and which production parameters are necessary in the process?
- 2) Which physical-mechanical properties relevant to insulation materials do such panels have, and how good are they compared with commonly available insulation materials?
- 3) Are tannin-hexamine resins suitable for the bonding of light bark particleboards? Which production parameters are necessary in this respect, and which panel properties can be achieved?
- 4) Which influence does the panel structure have on its thermal conductivity? Can the interior board structure be revealed using X-ray computed tomography? And is it possible to model the panel's thermal conductivity when its structural composition is known?

This work stretches from the development of a new product, its production, characterization and optimization, to the theoretical thermal modeling of the material with the main focus being on the latter. Thus, it lays the basis for the substantial use of bark as a thermal insulation material for various applications, focusing on the specific research questions listed above. Additionally, future research requirements shall be revealed and potentials for bark insulation optimization shall be identified.

## 2 State of the art

#### 2.1 Insulation materials based on renewable resources

Due to their heat-insulating properties, insulation materials are used to reduce the heating and cooling energy required for building operation and, accordingly, to reduce CO<sub>2</sub>-emissions (Pfundstein et al. 2007). In sufficiently insulated buildings, also the comfort for residents rises due to higher interior wall surface temperatures (Dear et al. 2013). Additionally, structural damage caused by water vapor condensation in structural elements or on surfaces can be prevented by adequately insulating constructions. Apart from that, some insulation materials can be used to reduce noise disturbance because of their sound-insulating effect (Riccabona and Bednar 2008). Sound insulation might be an increasingly important future market. It is estimated that 65 % of Europeans are exposed to unhealthy noise levels (D'Alessandro and Schiavoni 2015).

Approximately 40 % of the entire European energy consumption is caused by buildings, which has forced the European Union to pass a directive concerning the total energy efficiency of buildings. The energy consumption of new public buildings and private housing (regarding primary energy efficiency) will have to be significantly reduced from 2018 and 2020, respectively (European Union 2010). Embodied energy calculations and life-cycle analyses are key elements in the energy assessment of buildings, although these elements are ignored in most existing regulations on building energy consumption (Casals 2006). There are two design strategies to minimize the CO<sub>2</sub>-emissions caused by building operation: minimizing heating and cooling energy through energy efficient measures, and enhancing the use of renewable energy (Li et al. 2013a). The first strategy addresses the building envelope, aiming at limiting the summer heat gain and the winter heat loss. The average insulation thickness applied in walls and roofs of European buildings almost doubled between 1982 and 2000, especially in northern Europe (Papadopoulos 2005).

#### 2.1.1 Overview of bio-based insulation materials

The European insulation material market has grown by 50 % between 1991 and 2005. German insulation material sales come up for 31 % of the European volume, making Germany the biggest national market (Carus et al. 2008).

The most widely used insulation materials in Europe at present are inorganic fiber-materials (glass wool, stone wool) and organic foams (expanded and extruded polystyrene), which cover 90 % of the market. The rest is made up mainly of wood wool and, to a lesser extent, of insulation materials based on renewable resources and high performance insulation materials like vacuum panels or aerogels (Papadopoulos and Giama 2007).

The list of insulation materials based on renewable resources is long and steadily growing. Exemplary flax, hemp, wood shavings, wood fibers, wood wool, coco fibers, cork, sheep wool, reed, straw, cellulose flocs, and sea weed can be assigned to the group of insulation materials based on renewable resources (Fachagentur Nachwachsende Rohstoffe e. V. 2012; overview in Table 1). A review of unconventional bio-based insulation materials can be found in Asdrubali et al. (2015; overview in Table 2). Continuously, new inventions are made in the field of insulation materials based on renewable resources. An example would be wood foams – light wood composites with a porous structure made from fine wood particles. They do not contain synthetic resins as cohesion is created by wood components activated in the production process. The density lies between 40 and 200 kg/m<sup>3</sup> and the thermal conductivity ranges from 0.03 to 0.05 W/(m\*K) (BMLFUW 2014a). Another very promising approach is to obtain tannin foams by polycondensation of polyflavonoid tannins and furfuryl alcohol. Densities range from 50 to 120 kg/m<sup>3</sup>, thermal conductivities from 0.035 to 0.055 W/(m\*K) (Tondi and Pizzi 2009, Merle et al. 2016).

The main differences between different insulation materials based on renewable resources can be determined by thermal conductivity, density, moisture resistance, water vapor diffusion resistance, specific heat storage capacity, and flammability. Materials also differ in form. There are, for instance, panels, flexible mats, or loose bulks. Finally, also the field of application (wall, ceiling, roof, perimeter) depends on the material (Fachagentur Nachwachsende Rohstoffe e. V. 2012).

Material	Form offered	Density in kg/m <sup>3</sup>	Thermal conductivity in W/(m*K)
flax	panel/mat, loose	~ 30	0.036-0.040
hemp	panel/mat, loose	20–80	0.048
wood fiber	panel/mat, fill material	30–270	0.040-0.052
wood wool	panel/mat, aggregate	330–500	0.090
cork	panel/mat, fill material	75–120	0.040-0.050
sheep wool	panel/mat, loose	16–70	0.032-0.040
reed	panel/mat, aggregate	190	0.055
straw	panel/mat, loose, aggregate	90–110	0.052-0.080
cellulose	panel/mat, fill material,	30–70	0.040
	loose, aggregate		

Table 1. Bio-based insulation materials by comparison (Fachagentur Nachwachsende Rohstoffe e. V. 2012, BMLFUW 2014a).

Plant	Form offered	Density in kg/m <sup>3</sup>	Thermal conductivity in W/(m*K)
reed	panel	130–190	0.045–0.056
bagasse	particleboard	70–120	~ 0.046
cattail	particleboard	200–400	0.044-0.06
corn cob	particleboard	-	~ 0.101
cotton stalks	particleboard	-	0.059-0.082
date palm	bunches	-	0.072
durian	particleboard	428	0.064
oil palm fiber	bunches	100	0.055
pineapple leaves	panel	178–232	0.035-0.043
rice hulls	particleboard	154	0.046
sunflower piths	particleboard	50–180	0.040-0.050
straw	bales	60	0.067

Table 2. Insulation materials made from unconventional resources (Asdrubali et al. 2015).

### 2.1.2 Critical discussion of insulation material choice

Environmental awareness is not limited to the sufficient insulation of buildings, but also involves ecologic constructions, optimized with regard to minimum energy input, resource consumption, and pollution over the life cycle (Berge 2009). There is a huge variety of insulation materials available on the market. The material choice can be influenced by physical, economical, ecological, or health criteria (BMLFUW 2014a). Decisions with regard to material choice in civil engineering show that an increasing emphasis is placed upon non-toxic, recyclable materials (Pargana et al. 2014). Energy-demanding materials are progressively replaced by materials based on renewable resources, which tend to meet the requirements of sustainable constructions (Korjenic et al. 2011).

Biomass gained from agricultural crops and residues, forest resources and residues, and animal and municipal waste is the most important source for cellulose in the world (Mohanty et al. 2000). These resources are renewable, available in large quantities, and most of the time low-cost (Reddy and Yang 2005).

Insulation measures on buildings are usually discussed with regard to minimizing heat loss and costs. From this point of view, the most important design parameters are insulation thickness and material choice based on price and thermal conductivity. Such considerations either favor relatively cheap petrol-based insulation materials or high-performance insulation materials, such as aerogels and vacuum insulation panels, which have a thermal conductivity below that of still air (0.026 W/(m\*K)). These are called superinsulators (Ebert 2013).

Proper insulation material choice nonetheless requires a multiparameter analysis which is far from being trivial (Papadopoulos 2005, Roberts et al. 2015). The right choice depends on the specific

requirements, including the evaluation of a variety of material parameters, such as thermal conductivity, specific heat storage capacity, fire resistance, steam diffusion resistance, water absorption, resistance to direct sunlight, maximum service temperature, durability, sound absorption, cost (related to insulation efficiency), and potential health risks (Al-Homoud 2005). Another relevant parameter is optimum insulation thickness with regard to costs, energy consumption, and CO<sub>2</sub>-emissions over the life-cycle (Masoso and Grobler 2008). Insulation materials based on renewable resources are on average twice as expensive as classical insulation materials (Barbu 2011). Cost-effectiveness does not necessarily go hand in hand with environmental effectiveness. Another issue raised is the thermal mass applied in buildings, which significantly helps to avoid summer overheating and reduces cooling requirements (Henze et al. 2007).

In a holistic consideration of insulation materials, renewable ones have their advantages (e.g., renewable material source, regional availability, heat and moisture storage capacity etc.; Kleinhempel 2005) for specific applications, but cannot entirely replace conventional insulation materials such as mineral wool or polystyrene (Korjenic et al. 2011). Insulation materials should also be evaluated with regard to their availability, because the use of locally available materials leads to a reduction of economic and environmental impacts. The primary aim should be to use residues and byproducts from the forestry and agricultural sector and to avoid conflicts with the planting and harvesting of food crops (Asdrubali et al. 2015).

Another issue concerning insulation materials based on renewable resources was raised by Tran Le et al. (2010), who showed that the use of hemp concrete leads to a reduction of the indoor relative humidity variation over a day. Moreover, natural building materials can be beneficial to the human psyche because of their characteristic smell (Korjenic et al. 2011). A test house supplied with straw-bales as a wall material proved to have excellent properties for a healthy living environment (Ashour et al. 2011). Emissions from building materials can cause the sick-building-syndrome — a risk which can be reduced by using natural building materials (Bauer et al. 2013).

Up to now, insulation materials based on renewable resources have only made up a low percentage of the total European market (Papadopoulos 2005), partly due to disadvantages such as potentially high hygroscopicity, combustibility, and fungal growth (Hurtado et al. 2016). Some of these disadvantageous properties can be improved by material modification (Zach et al. 2013). Future potentials for bio-based insulation materials might be found in the fields of fiber insulations with bio-based phase change materials (Kosny et al. 2012), bioaerogels, bioplastics, bio-based foams, and, in a wider context, bio-inspired lightweight structural materials and photobioreactors (Torgal 2016).

### 2.2 Potential of tree bark as insulation material

#### 2.2.1 Anatomy

Tree bark is organic cellular tissue, which is built by higher plants (trees, shrubs) on the outer side of a tree's cambium as a shell of the tree's xylem. Bark consists of the outer bark or rhytidome and the inner bark or phloem. Knowledge of the structure and properties of bark is important for the determination of tree species, assessment of tree physiology, analysis of pathological processes in a tree, and for potential utilization (Vaucher 1997).

Bark has an important function for the tree, because it protects it from physical and chemical influences from its environment. It prevents the access of precipitation in form of rain, snow and hail, heat, frost, UV-radiation, and gases to the sensitive cambium. Moreover, bark is a physical-chemical barrier for bacteria, fungi, parasitic plants, and animals like insects, birds, and mammals (Sakai 2001). Apart from its protective function, bark is also a storage place. A tree bins substances which are poisonous for its metabolism, such as crystals, tanning agents, slimes and resins, to the bark (Holdheide and Huber 1952). Additionally, significant nutrient transport takes place in the living tissue of the phloem. At the beginning of the growth season, reserve substances are transported from the roots to the crown. Later on assimilates, which are produced in photosynthesis processes, circulate in the whole tree to nourish the existing tissue, and new tissue is created. Towards the end of the growth season some of these substances are stored again in the bark as a reserve (Lohmann 1982).

The inner vascular cambium produces xylem towards the inner side and phloem towards the outer side. Within the primary bark, the cork cambium (or phellogen) is built over time. The outer cork cambium produces secondary cortex cells (phelloderm) towards the inner, and cork cells (phellem) towards the outer side. Phelloderm, phellogen and phellem make up the periderm (Evert and Eichhorn 2006; Figure 1). The first periderm can be long-lasting (as with fir, birch, or beech), or short-lived, as is the case with most European trees. In that case, it dies off after some time and is replaced by a new one on its inner side. The new periderm surrounds the tree ring-like (ring periderm) or is imbricative (concave or flake periderm; Figure 2). The collection of periderms is called rhytidome (Fengel and Wegener 2003). Mature bark is mainly made up of secondary phloem and periderm (Sakai 2001). The sequence of periderms gives the bark its characteristic pattern on the outside and protects the tree from manifold damage (Walker 2006). Phloem and periderm cannot be easily separated with most tree species (Feng et al. 2013), apart from the cork oak (Vaucher 1997). Most trees have two bark zones — the inner bark, containing some living cells, and the outer bark, without any living cells, and are sometimes compared with sapwood and heartwood (Holdheide and Huber 1952).

The phloem contains conducting, sclerenchymatic, and parenchymatic cells and contains the conducting system of a tree. Here, assimilates in form of carbohydrates and amino acids are transported in horizontal and vertical direction. In the process, the living, dividable cellular tissue is supplied with photosynthesis products from leaves and needles (Sakai 2001). For this reason, the phloem cells are predominantly made up of sieve elements. In coniferous trees, the conducting elements are called sieve cells. They are perforated by small pores forming sieve fields. In deciduous trees the conducting elements are called sieve tubes, the larger pores in their cross walls are called sieve plates. Both types are interconnected by thin plasma strands (plasmodesmata; Holdheide and Huber 1952).

Sclerenchymatous cells are found in thick-walled, elongated bast fibers and in stone cells with polygonal shape, originating from parenchyma cells, whose cell walls have been thickened and lignified. Bast fibers are also used in technology, due to their toughness and durability (Vaucher 1997).

Parenchyma cells in the phloem form longitudinal strands or bands, which are dispersed among sieve cells, or form radially-oriented parenchymatic rays. Often, regular structures are found in the phloem, originating from a periodic sequence of conducting and consolidating tissue (Holdheide and Huber 1952, Fengel and Wegener 2003).

The outer bark contains primarily dead tissue with various depositions in its cells, which prevent the attack of microorganisms and the loss of water. It is formed because the underlying initial cork cambium stops its activities after some time, and in the subjacent phloem a new cork cambium is built. This process is iterated and the outer bark gains thickness. Outlying the new periderm, suberin is placed into the cells, which makes them impervious to water (Vaucher 1997).

The living cells in the sapwood of a tree's stem have to be supplied with oxygen, either by the transpiration stream in vertical direction or by radial diffusion through bark and xylem. Oxygen is required for oxidative respiration, which provides energy for the cells (Sorz and Hietz 2006). Especially with tree species being adapted to waterlogged soil, the cambium has intercellular spaces allowing oxygen supply through the bark (Buchel and Grosse 1990). Gas exchange between outside air and the inner, living tissues of the bark underneath the rhytidome is ensured by lenticels in the periderm. They are formed when numerous filling cells are created in the cork cambium. These break through the underlying tissue over time and form inter-cellular rooms, which enable an unhindered gas exchange (Vaucher 1997).

Bark morphology varies greatly within species and plant communities. The underlying reasons for bark diversity are not understood in detail, but the high morphological diversity suggests that differences in bark could be important for a plant's ecological strategy. A recent investigation has shown that tradeoffs and coordination within and beyond the bark determine the bark's morphology,

which is determined, among others, by thickness, stiffness, water content, and density (Rosell et al. 2014). Bark thickness varies greatly between tree species. The bark thickness of juniper (*Juniperus* spec.), for example, ranges from 2 to 6 mm, whilst the one of poplar (*Populus* spec.) ranges from 5 to 80 mm. It has been suggested that bark thickness can be explained by different fire regimes in fire-prone ecosystems. Some fire regimes account for thick barks on the whole tree (grass-fueled crown fires), others only on the base of a trunk (understorey fires). It has been shown that fire regime can explain a high ratio of the variability in bark thickness on a global scale (Pausas 2015). Recent work has shown for angiosperms that the global variation of the total bark thickness (TBT, considering inner bark and outer bark) is mainly explained by stem size. The environment of the tree is of less significance in this regard (Rosell 2016). The huge thickness variations in tree bark could also refer to other bark functions, such as photosynthesis, maintenance of water relations, and retention of non-structural carbohydrates (Paine et al. 2010).



Figure 1. Schematic illustration of coniferous bark (modified from Kraft 2007).



Figure 2. Cross-section larch bark flake extending from the wood to the outside (Kain et al. 2016a).

#### 2.2.2 Physical and chemical properties

#### **Physical properties**

The physical properties of bark are very relevant when it comes to its application. The following properties are of technical interest. The heat value of dry bark amounts to 17,000–22,000 kJ/kg (4.7–6.1 kWh/kg) and is comparable to that of dry wood (Vaucher 1997). The density by volume varies greatly, but most of the time it is significantly lower than that of the respective wood. It can, however, also be significantly higher. Miles and Smith (2009), for instance, provided an overview of wood and bark density of the most important tree species in North America, stating that bark is 36 % heavier (average of fir species) to 35 % lighter (average of various larch species) than the respective wood (oven-dry conditions, green volume basis). The bark density (air-dried samples) was found to vary between 0.71 g/cm<sup>3</sup> (*Fagus sylvatica*) and 0.23 g/cm<sup>3</sup> (*Sequoia giganteum*; Bauer et al. 2010). Finally, bark contains a large amount of ingredients which are important for the specific applications (Warnecke 2008).

The swelling and shrinking of bark is stronger than that of wood (volumetric expansion in softwood barks amounts to 10.9–16.6 %, in hardwood barks it is 9.5–18.5 % measured from fiber saturation point to oven-dry condition). The longitudinal shrinkage of bark is 4 to 20 times higher than that of wood, whereas transverse shrinkage lies in the range of wood (Martin and Crist 1968).

Bark of American hard- and softwoods has been shown to have considerably poorer mechanical properties than wood. In addition, bark displays a lower anisotropy between longitudinal and transverse directions when compared with wood (Martin and Crist 1968).

The thermal properties of bark were first systematically evaluated by Martin (1963). Bark is slightly less anisotropic than wood, and its thermal conductivity is a little lower. Oven-dry bark showed an approximately 20 % lower thermal conductivity than wood in radial direction. The TC of wood (12 % moisture content; *Quercus sp., Picea abies, Fagus sylvatica*) is 2.3–3.0 times higher in longitudinal direction than orthogonal to fiber direction. Oak wood has a longitudinal TC of 0.368 W/(m\*K), 2.4 times higher than the tangential TC of 0.152 W/(m\*K). The radial TC of oak wood is 17 % higher than the tangential TC. The longitudinal TC of beech and spruce wood is 2.9 times higher than the tangential TC was measured to be 14 % higher with beech, and 12 % lower with spruce, compared with the tangential TC (Vay et al. 2015). In contrast to that, the investigations of Martin (1963) regarding the TC of eight North American deciduous and coniferous barks showed the tangential TC in that respect. The radial TC. The longitudinal TC in bark ranges from 1.14 to 1.30 and is therefore minimal compared to that of wood. Consequently, the orientation of particles in bark panels is considered to have a low effect on the panels' TC (Martin 1963, Schneider and Engelhardt 1977).

Bark has proved to have on average a 50 % higher TC than commonly used insulation materials, but Martin (1963) emphasized that a well-considered production of insulation boards might reduce the global panel TC significantly.

Gupta et al. (2003) calculated linear regression models for the dependency of the specific heat storage capacity of wood and bark from balsam fir (*Abies balsamea*), white spruce (*Picea glauca*), and black spruce (*Picea mariana*) on temperature based on oven-dry conditions. In the early investigation of Martin (1963) the specific heat storage capacity of various barks was measured, yielding on average 1,383 J/(kg\*K) at oven-dry conditions, which is in the range of what Gupta et al. (2003) determined and is comparable with the heat storage capacity of wood. Nonetheless, the influence of the moisture content (MC) is very important, because water has a very high heat storage capacity (4,185 J/(kg\*K)) and also because energy is absorbed during sorption processes in the bark. In his investigation Martin (1963) presented a model for the calculation of the specific heat storage capacity of a bark-water mixture, where he takes this effect into account.

The equilibrium moisture content of bark is slightly higher than that of the corresponding wood under constant climatic conditions (Niemz 1993). Sorption isotherms were determined for the phloem and periderm of spruce (Picea abies), pine (Pinus sylvestris), horse chestnut (Aesculus hippocastanum), poplar (Populus spec.), and birch (Betula spec.). It could be shown that the periderm of poplar and birch is less hygroscopic than the phloem, probably due to a high suberin content, whereas the periderm of spruce, pine, and horse chestnut has a higher equilibrium moisture content than the corresponding phloem, when the relative air humidity (RH) is below 90 %. The moisture content of the inner bark of spruce, pine, and poplar rises strongly above 90 % RH and reaches an MC of 101 to 105 %. These extreme values can be referred to water-soluble sugars in the phloem. It was reported that for all the examined species the variation of the equilibrium moisture content of bark is approximately twice as high as for the corresponding wood. When the relative air humidity is 97.6 %, the equilibrium MC of the barks (various species) ranged from 24.1 to 38.3 %, that of wood between 22.9 and 30.4 %. The measurements of the bark moisture content were conducted in evacuated desiccators at room temperature, as gravimetric moisture determination does not apply due to the volatileness of extractives (Standke and Schneider 1984). It was also shown that the sorption behavior of tropical and central European tree barks is similar (Schneider and Parameswaran 1983).

Bark differs significantly with regard to porosity, density, and anatomy. Physical properties were quantified by Bauer et al. (2010), investigating the thermal insulation capability of different tree barks in a forest fire. The bark structure (variation in bark thickness) was found to be high with *Quercus suber*, *Pinus sylvestris*, and *Larix decidua*, compared to species with a low bark structure like *Fagus sylvatica* or *Tilia cordata*. Ignition time for oven-dry bark at 300 °C was found to range from 21 (*Fraxinus americana*) to 69 seconds (*Pinus strobus*). The amount of volatile material in the barks

does not correlate with time until ignition (Hengst and Dawson 1994). In terms of fire resistance, bark thickness has the strongest influence, whilst bark density and bark structure only contribute to a lesser extent (Bauer et al. 2010).

#### **Chemical properties**

Chemically, bark, just as wood, consists of the primary components cellulose, hemicelluloses, lignin, and extractives (Sakai 2001, Fengel and Wegener 2003). The most important difference in the chemical composition of bark is the presence of polyphenols and suberin, fewer polysaccharides and a higher amount of extractives (Sakai 2001). An analysis regarding the composition for various tree species can be found by Fengel and Wegener (2003), but comparison is restricted due to different extraction methods (a summary is given in Table 3). In addition, phloem and rhytidome have different chemical compositions. The amount of extractives and polysaccharides decreases from inner to outer bark, the amount of lignin and polyphenolic compounds increases (Fengel and Wegener 2003).

The most important sugar in wood is glucose, amounting to 16–41 %. Bark cellulose has a significantly lower degree of polymerization than wood cellulose. Moreover, in the outer bark less glucose is present than in the inner one. Regarding polyoses, the most important one in coniferous tree bark is galactoglucomannan and in deciduous tree bark it is arabino-4-O-methyl-glucuronoxylan (Sakai 2001).

The cell walls of fibers and sklereids are lignified, also the cells of periderm and rhytidome were shown to contain lignin. Structurally, wood and bark lignin are similar, although there are differences in constituting components (Fengel and Wegener 2003).

Polyphenols in bark are primary flavane derivates, which can be classified according to their molecular weight and solubility. Generally speaking, procyanidins, condensed tannins, and polyphenolic acids can be distinguished (Sakai 2001).

Suberin is an insoluble substance present in the rhytidome, and there especially in cork cells. It has a polyester-like structure (composed of long-chained fatty and hydroxyl fatty acids) whose composition depends on the tree species (Sakai 2001, Fengel and Wegener 2003).

The amount of extractives is up to ten times higher in bark than in wood and it is influenced by species, environmental conditions, seasonal influences, and genetic disposition. The extractive amount and components depend on the species and the solvent used and are primarily polymer flavonoids, fats, waxes, terpenes, free and combined acids, fatty alcohols, sterols, resin acids, glycosides, and stilbenes (Fengel and Wegener 2003).

The content of minerals in bark is up to ten times higher than in wood. It varies significantly. With some deciduous tree barks, an ash content of more than 10 % was measured. For example, the burning of *Salix alba* bark resulted in approximately 5 % ash (Klasnja et al. 2002), that of *Tectona grandis* bark in approximately 19 % (Baptista et al. 2013). Additionally, the frequency of elements is different. The most frequent element in bark is calcium (82–95 %) and, in significantly lower concentration, potassium and magnesium (Sakai 2001).

The bark of teak (*Tectona grandis*), for instance, was investigated in detail regarding its chemical composition (based on the mass of oven-dry bark). It showed to contain a total of 10.7 % of extractives, 1.9 % of suberin, 29 % of lignin and 47 % of polysaccharides. Ash made up 18.5 %, primarily consisting of calcium, potassium and magnesium (Baptista et al. 2013).

Because of its chemical composition, bark has a lower pH-value than wood. Using hot water extraction, the pH-value of a bark extract of spruce, pine, beech and oak had a pH-value between 3.5 and 5.0, whereas the according wood extract had a pH-value between 4.2 and 5.3. The rhytidome is slightly more acid than the phloem, and with increasing age the pH-value of bark slightly decreases (Fengel and Wegener 2003).

Bark is also a passive sampler for pollutants from the surrounding air because of its high lipid content and large surface area. The highest bark pollutant concentrations were observed at urban sites. Good correlations between bark and atmospheric and precipitation concentrations of organic pollutants were determined (Salamova and Hites 2010).

Component	Share in bark in %	
Cellulose	23 – 54	
Polyoses	9 – 23	
Lignin	38 – 45	
Extractives	11 – 21	
Suberin	2 – 4	
Ash	1 – 7	

Table 3. Chemical composition of the bark of European trees (Fengel and Wegener 2003).

#### 2.2.3 Technical applications

#### Historic bark use

Tree bark use has a long tradition as a construction material for roofing, heat and sound insulation, and other specialized purposes. In various parts of Mexico, an over 1,400 year-old tradition of paper making from tree bark (Ficus species and Morus celtidifolia) exists. In the process, bark is harvested in long stripes when the cambium activity starts in spring. Then the phloem fibers are separated from the outer bark. These fibers are then boiled for several hours to make them softer. Subsequently the fibers are arranged in a grid-like pattern and beaten together with a stone. When the fibers are sufficiently interlaced, the bark sheets are dried in the sun. This paper production method reached dimensions partly causing a shortage of appropriate trees (Peters et al. 1987). Sami people in northern Sweden used the inner bark of Scots pines (Pinus sylvestris L.), harvested in early summer, as food. It is a source of nutrition due its carbohydrate content of up to 16 %. Proof of this habit are surviving trees which exhibit bark-peeling scars. The bark use of the Sami people can at least be confirmed for the time between 1700 and 1900. Bark was probably a healthy supplementary food in spring time when game meat naturally became scarce (Rautio et al. 2014). Bark (birch) was also used by Indians for canoe-building (Adney and Chapelle 2014) and for tapa-clothes in the South Pacific (Neich and Pendergrast 1997). Other traditional uses were flavors, anti-malaria drugs, antibiotics and other medicines in old societies (Ogunwusi 2013).

Logs were debarked in the forest to ease logging in central Europe. The bark produced in the process was used for the production of tanning agents. Bark extracts contain high amounts of tannins, which are polyphenolic compounds. They show a high reactivity with protein in animal skins (Feng et al. 2013). Lumberjacks also used large bark pieces to build primitive dwellings in the forests. They built tent-like frameworks out of wood poles and planked them with large bark pieces. Similar huts were in use for simple food stores and stables. Such bark roofs achieved a life time of up to 50 years (Mooslechner 1999).

#### Contemporary bark use as solid material

Today bark is more of an inconvenient byproduct, because it can be severely soiled and reduces the service life of tools when processed. Bark is highly inhomogeneous regarding its structure and chemical composition, which makes its processing much more difficult than that of wood (Baptista et al. 2013). Today, the economic interest in bark is low, because bark tends to be accumulated in relatively small pieces and its mechanical properties are inferior to those of wood (Martin and Crist 1968). Usually, bark is directly burned in roundwood-processing industries to gain process energy (Ogunwusi 2013). Oven dry bark has a heating value between 17,000 and 22,000 kJ/kg (Vaucher 1997) and exceeds the heating value of wood due to a higher carbon content (Feng et al. 2013).

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Nosek et al. (2016) reported, on measuring the impact of bark content on biofuel properties, that a larger amount of bark in softwoods results in a lower calorific value than with pure wood. In hardwoods, increases in combustion heat and calorific value were measured, and in both cases the ash content increased. They concluded that due to the abundance of bark and its relatively high calorific value, bark combustion is reasonable. Problems concerning bark combustion are caused by a moisture content that varies strongly over the seasons. It ranges from 55 to 185 % (Grammel 1989). A research note of the U.S Department of Agriculture (Miles and Smith 2009) listed the average bark MC of over 150 North American tree species with values ranging from 55 % (birch) to 97 % (hemlock). In Finland, birch bark is a major energy source for heat and electricity. Due to a high initial MC of bark (50-60 %) after debarking, the efficiency of power plants could be increased by reducing the MC before burning. Drying bark in fixed beds requires specific drying curves because of heterogeneous particle size distribution and particle shape of bark beds. Both drying temperature and bed height have to be considered when estimating drying times (Holmberg et al. 2011). Moreover, the burning of bark partly causes technical problems, because more ash is produced and, due to the high amount of extractives, unwanted emissions occur (Naundorf et al. 2004). Another problem is that, due to the high ash content of bark and its low sintering point, bark incineration can lead to fouling, which can damage combustors (Feng et al. 2013, Nosek et al. 2016). It was reported that resinous bark pellets showed better cohesion than wooden ones, making them more durable (Filbakk et al. 2011). The extent to which bark (and other forest residues) is used in energy production is also strongly influenced by the market situation regarding raw material prices (Mantau 2015).

More recently bark has been used as ground cover, soil improvement, bark mulch, and raw material for composting, humus production, culture media, biofilters, and bio-absorption materials (Vaucher 1997, Dalahmeh et al. 2012). Bark did not prove to be suitable for the production of fermentable sugars, because it contains a high amount of lipophilic extractives and aromatic compounds such as tannin and lignin (Kim et al. 2005).

There is a long tradition of using bark as a resource for wood composites. Mimosa bark was used in the core layer of particleboard. Evaluation of panel properties showed that a bark content of more than 6 % leads to significantly poorer mechanical board properties; formaldehyde emissions and thickness swelling were lower with a higher bark content (Nemli and Colakoglu 2005). Bark was also used as feedstock in the production of medium density fiberboard, with no negative effect on thickness swelling. However, mechanical properties were also significantly poorer when the bark content was increased (Xing et al. 2006). Yemele et al. (2008b) produced particleboards made out of hot water-extracted bark of black spruce, noting that the bark extracts could be used in pharmacology or for the production of adhesives. Bark particle geometry has a strong influence on the internal bond of bark particleboard. Coarse particles result in higher MOE, MOR, and IB values

(Yemele et al. 2008a). A more recent study has proved that particleboard with a density of more than 800 kg/m<sup>3</sup> can be produced from lodgepole pine (*Pinus contorta*) bark without adding any additional resin. The press temperature has a significant influence on the mechanical board properties in this respect (Gupta et al. 2011). Plasticization, physical consolidation, and polymerization of bark extractives contribute to the self-bonding of bark particles. Pressing temperatures below 200 °C mainly result in bark softening, whilst with higher temperatures polymerization and partial degradation of bark components contribute to bonding (Marashdeh et al. 2011). Additionally, bark fiber panels were produced without adding additional resin, and it was found that the panels meet the standard requirements regarding IB, MOR, MOE, and TS. The best properties were achieved with a pressing temperature of 260 °C (Gao et al. 2011). Pure bark panels are not commercially available at present, because of their partly inferior appearance and performance characteristics (high TS), high pressing temperature, and long pressing time resulting from the low thermal conductivity of bark (Blanchet et al. 2000). Possibilities to overcome the last problem are the pre-treatment of bark by steaming (Xing et al. 2007a) or alkali treatment (Geng et al. 2006).

Pine (*Pinus sylvestris*) bark was used to produce pallet blocks with a density of between 600 and 750 kg/m<sup>3</sup>. It was bonded with urea formaldehyde (UF) and melamine-reinforced urea formaldehyde (MUF) resin, meeting the requirements of the relevant standards regarding internal bond, nail withdrawal and moisture resistance (Kain et al. 2013).

Apart from bark composites, also bark plastic composites were investigated. Black spruce and aspen bark fibers (50 and 60 % based on the oven dry weight) and polyethylene were used to produce bark-plastic composites in an extrusion process. The evaluation of flexural and tensile properties showed that the strength is lower than that of wood-plastic composites (Yemele et al. 2010).

The bark of the American chestnut (*Castanea dentata*) and of the American tulip tree (*Liriodendron tulipifera*) can be used for exterior cladding. In spring, the bark is peeled off freshly felled trees, cut, and stacked for drying. Drying is necessary to ensure dimensional stability and to prevent biological degradation (Mühlbacher and Taylor 2009).

Furthermore, sophisticated application opportunities reach from flame-retardant particleboards (Mengraw 1976) to heat insulation for intercontinental ballistic missiles (Hovey 1965). A recent unconventional bark use has been that of a bio-composite designed for interior automotive panels made of bark cloth and a green epoxy polymer. Bark cloth was harvested from natal fig (*Ficus natalensis*) trees. The outer bark can be peeled off without harming the inner bark (similar to the cork oak), which is additionally wrapped with banana leaves after peeling to avoid the dehydration of the tree. The bark gained was then subjected to heat in order to soften it, pummeled and sundried. Afterwards, the bark cloth was treated in alkali solution, flushed with water, and oven dried. The composite was produced by stacking layers of bark cloth impregnated with a biodegradable

expoxy resin at angles of 90, 0, -45, and 45 ° in a mold and curing the composite in a hot air oven (Rwawiire et al. 2015).

A very special traditional but also contemporary bark use is the one of cork. The cork oak (Quercus suber) is a unique tree growing in the Mediterranean and south Atlantic region. Portugal comes up for 50 % of the world's cork production. At an age of 20-25 years, the cork oak is debarked for the first time. The resulting cork is called "male cork" and is of inferior quality and used for granulates. 8-10 years later the cork has grown again to a thickness of 3-5 cm and the high guality "female cork" can be peeled off. This process is iterated every 8-10 years. The specialty of the cork oak is that the rhytidome can be easily separated from the phloem, which prevents damage to the living bark components while peeling (Vaucher 1997). Cork has valuable properties, such as low density (150–250 kg/m<sup>3</sup>), good sound and heat insulation, low flammability, low capillarity, and an attractive appearance. Applications range from bottle corks, over insulation granulates, mats, and panels to design elements in interior design (Gil 2015). Recent work on the potentials of cork bark has additionally suggested the use of low-quality cork granulate in timber-concrete composites for structural applications in floors (Martins et al. 2016), and the use of cork powder and granules for the absorption of pollutants (Pintor et al. 2012). Another suggestion was to liquefy cork dust or granulates, while simultaneously yielding components for the production of adhesives (Santos et al. 2016). Moreover, SiC ceramics were produced from cork as a precursor, characterized by open pores, low weight, high permeability, low thermal conductivity, high corrosion resistance, and high temperature stability. Potential applications are porous heaters, catalyst supports, filters for molten metals, or thermal shields (Yukhymchuk et al. 2016).

#### Biorefinery of bark and use of bark components

In biorefining, biomass feedstock is converted to different classes of biofuels and biochemicals using jointly applied conversion technologies. The aim is to produce sustainable biofuels and high-value chemicals applying green chemistry and low environmental impact production technologies (Cherubini 2010). At present chemicals are predominantly based on fossil fuels, whose availability and ecological impact is unsure or negative. Biomass, consisting of cellulose, hemicelluloses, lignin, extractives and ash, could be converted into value-added chemicals using thermomechanical and biochemical processes. Bark accumulates in the wood-processing industry in large quantities and at low prizes, which makes it an interesting source for biorefinery (Feng et al. 2013). Moreover, integrated biorefinery could be an additional source for adding value in the forest industry (Mohan et al. 2006, Devappa et al. 2015). Difficulties are being faced in defining standard processes because bark is highly complex and inhomogeneous.

Nowadays the use of bark tannins is worth mentioning. Tannins can be divided into hydrolysable tannins and condensed tannins. Condensed tannins are made up of complex structures of polyphenolic compounds and have a high reactivity to chemicals like formaldehyde. 90 % of the global tannin production refers to condensed tannins. Condensed tannins are mainly extracted from quebracho (*Schinopsis balancae*) wood, wattle (*Acacia mearnsii*) bark, and chestnut (*Castanea sativa*) bark (Khanbabaee and Ree 2001). Tannins are predominantly extracted with water, but also organic solvents or alkaline solutions are used. A detailed summary of different extraction methods and chemicals used can be found in a review by Feng et al. (2013).

Adhesives for wood composites can be made out of polyphenolic fractions extracted from tree bark (Harking and Rowe 1969). Bonding strength is achieved through the formation of methylene bridges between the reactive positions in the flavonoid units of tannin with formaldehyde or other hardeners, such as hexamethylenetetramine, tris(hydroxymethyl) nitromethane, glyoxal, or polymeric 4,4' diphenylmethane diisocyanate (pMDI). The tannin-hardener mixture is adjusted to alkaline conditions, which are needed for the hardeners to decompose to formaldehyde (Feng et al. 2013). Pizzi (1982), for example, used tannins from the bark of pine to bind particleboard and Roffael et al. (2000) used spruce tannin as a binder in particleboard and MDF. Tannin adhesives can be fortified with synthetic resins (UF, PF, RF, pMDI) and vice versa. Moreover, phenol can be partly replaced by tannin in PF resins. Finally, polyflavonoid tannin can be hardened without hardener. Also, formaldehyde-free tannin adhesives, such as tannin/furfuryl alcohol, cornstarch/tannin, or tannin/lignin resins, can be used. They are particularly suitable for interior applications (Feng et al. 2013).

Lignin can be substituted for phenol in PF resins by mixing lignin, phenol, and formaldehyde. This approach was discussed especially in the context of delignification processes in pulp production. Eucalyptus bark, for example, was boiled in NaOH, lignin was recovered by precipitation and used to partially replace phenol in a PF resin. The authors found that up to 50 % of the phenol can be replaced (Khan et al. 2004). A recent study has proved that lignin foams can be produced from the waste liquor of magnefite pulping with characteristics comparable to tannin foams (Tondi et al. 2016).

Suberin was extracted from the outer bark of birch (*Betula verrucosa*) and polymerized via lipase, resulting in an epoxy-activated polyester, which was crosslinked with tartaric and oxalic acid. Finally, polyester-impregnated cellulose sheets were compression-molded and found to be hydrophobic (shown by a water contact angle of over 100 °; Li et al. 2015).

Polyflavonoids gained from bark extracts can be processed to foams. Tannin can be used as a precursor for polyurethane (PU) foam production. Additionally, bark tannin can be mixed with furfuryl, formaldehyde, acid, water, and diethyl ether. Water is the solvent, diethyl ether the blowing agent. The tannins are crosslinked by the formaldehyde and the furfuryl alcohol functions as a heat generator and strengthener because of autopolymerization and condensation reactions with tannin

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in acid environment (Celzard et al. 2010). Rigid tannin foam structures result from polycondensation of polyflavonoid tannins and furfuryl alcolhol (Tondi et al. 2009b). Such acid- or alkali-catalyzed polyflavonoid tannin-based rigid foams have similar mechanical and physical properties as phenolic foams. Quebracho and mimosa tannin is widely used as a basis for these foams (Meikleham and Pizzi 1994). Rigid foams from tannins reveal excellent properties similar to their synthetic counterparts. Foams made from spruce tannins were produced with a density below 50 kg/m<sup>3</sup> and a thermal conductivity below 0.045 W/(m\*K) (Lacoste et al. 2015). Tannin foams can, for example, be applied for cavity insulation in doors and wall elements. Besides having comparable building-physical properties as, for example, polyurethanes, they do not burn or emit toxic gases (Tondi et al. 2008). A recent study has shown that foams primarily made from hydrolysable and condensed tannins, and lignosulfonate from the pulp industry, can be produced with a structure similar to that of bio-based polyurethane (Merle et al. 2016).

Bark liquefaction is a process where bark is liquefied in alcohols and other solvents using bark phenolysis and solvolytic liquefaction. Bark pyrolysis, a thermal decomposition of bark under inert conditions, yields bark pyrolysis oil, water, gaseous products, and charcoal (Feng et al. 2013). Bark pyrolysis is likely to yield more than 50 % of bio-oil. It was shown that bark pyrolysis oil can be considered CO<sub>2</sub>-neutral and it can be burned in conventional burners (Bridgwater et al. 2002). Since bark pyrolysis oil has disadvantages, such as high contents of oxygen, water, solids and ash, high viscosity and surface tension, a low pH-value, and thermal instability, further upgrading is necessary (Qiang et al. 2009).Therefore it would be more efficient to use it in the production of chemicals. Bark pyrolysis oil can be used as a basis for phenolic compounds used in the production of adhesives (Feng et al. 2013).

Ethanol and furfural can be produced out of pine bark (*Pinus patula*). Most promising in this respect is an alkali (NaOH) pre-treatment of bark and further enzymatic saccharification of cellulose, yielding fuel ethanol and furfural out of xylose- and lignin-rich hydrolysate from pretreatment. From a technoeconomic point of view, the biorefinery concept requires energy integration to achieve a positive performance. Moreover, a biomass-fired cogeneration system would increase the economic income and lower the environmental impacts, as fossil emissions can be reduced (Moncada et al. 2016).

In smaller amounts, bark is also used in the production of tanning chemicals, stains, dispersing- and flocking agents (Naundorf et al. 2004).

Another promising approach is to use lodgepole pine (*Pinus contorta*) bark as a source for submicron- and nano-sized fibers. For this purpose, bark is ground, extracted, and then treated with chlorite to reduce the lignin content. Then the fibers are mechanically fibrillated, resulting in more than 80 % of the fibers being less than 0.4 mm in length. Fiber diameter is strongly influenced by the number of passes through the grinder but can be reduced to nano-sized (less than 100 nm) fibers with 5 passes. These fibers also show the best specific strength (Nair and Yan 2015).

Extracted and unextracted bark of black alder (*Alnus glutinosa*) was destructed using thermocatalytic destruction and the resulting particles were impregnated with HCl, dried, and dispersed in a ball mill. Then gel-like dispersions containing micro- and macroparticles (average size = 300 nm) were formed and applied on paper sheets as coating. Paper coated with bark gels containing chitosan and sodium carboxymethylcellulose showed good strength, air resistance, and vapor sorption properties (Laka et al. 2015).

Compared to leaves, roots and herbs, bark is of low significance to phytotherapy. Nevertheless, there are some barks which have been of great importance in the history of drugs. Salicin, originating from the bark of *Salicaceae*, was the basis for the development of the analgesic acetylsalicylic acid. Cinchona bark, gained from *Chinchona pubescens*, was used as an antifebrile and drug against malaria. Cinchona bark contains alkaloids, of which chinin is used against malaria in special cases, and chinidin, which is used for the treatment of heart arrhythmia (Vaucher 1997). The use of taxol from yew (*Taxus sp.*) bark is a promising cancer-chemotherapeutic agent (Sakai 2001, Weaver 2014). Bark components also have antifungal properties as shown, for example, by the use of a sumach bark extract in the fermentation of Philippine sugarcane wine to prevent the growth of lactic acid bacteria (Sakai 2001). A recent investigation has focused on the potentials of phytochemicals as value-added co-products in the bark of poplar (*Populus tremuloides*). Bark extracts were collected and analyzed regarding their composition. The most important components yielded were furfural, 5-hydroxymethylfurfural, benzoic acid, salicylic acid, catechol, and sakuranin. The authors noted that there is potential to use these chemicals in agro-pharmaceutical industries, but also emphasized the necessity for further end-use studies (Devappa et al. 2015).

All in all, the extreme variations in chemical and physical properties among barks impede high-grade utilization in biorefinery, or high pre-processing is required (Harkin and Rowe 1971). Important for all approaches to using bark as a biorefinery feedstock is that bark has to be properly characterized regarding its anatomy and chemical composition, as it is highly inhomogeneous not only between species, but also within a tree (Baptista et al. 2013). Nonetheless, the use of alternative feedstocks in biorefinery could be pushed by economic incentives or climate change regulations — especially as chemical engineering knowledge is far advanced and technical advances could occur quickly if the economic situation changes (Mohan et al. 2006).

#### 2.2.4 Availability

Bark is the second important tissue of a trunk. Its ratio in a trunk varies between 8 and 20 %, because the bark thickness depends on the tree species. The Austrian timber trade rules, for example, suggest a bark content for the most important central European species between 8 % for beech and 15 % for oak (Kooperationsplattform Forst Holz Papier 2006). The national forest inventory in the

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United States uses a bark content range from 11 % for maple to 19 % for oak (Miles and Smith 2009). Looking at a whole tree, the bark content is the highest in the crown, with 20–35 %. Also in the roots the bark content is higher than in the stem (Fengel and Wegener 2003). In line with logging activities in Austria, annually 2.8 million m<sup>3</sup> of bark are available, most of it is used in energy production (BMLFUW 2014b). This is backed up by information provided by the Trade Association of the Timber Industry Austria (2010), according to which between 2000 and 2009 on average 2.03 million m<sup>3</sup> of bark (calculated from roundwood) were accumulated in the timber industry each year. 1.38 million m<sup>3</sup> were consumed in various facilities. Most of it was used to produce energy. In 2009 a total of 392 million m<sup>3</sup> of roundwood under bark were removed in the EU (Eurostat 2011). Approximately 24 % are used as fuelwood (Paulitsch and Barbu 2015). Assuming that the rest is industrially used and the average bark content is 10 %, the result is an annual bark volume of approximately 30 million m<sup>3</sup>. Between 2003 and 2007, 3.28 billion m<sup>3</sup> of roundwood under bark were removed (Paulitsch and Barbu 2015). Calculating the bark volume for the remaining 43 % results in approximately 141 million m<sup>3</sup> of bark globally available per year.
# 3 Materials and methods

# 3.1 **Production of bark insulation boards (Publications 1, 3, 5)**

Spruce (*Picea abies*), pine (*Pinus sylvestris*), and larch (*Larix decidua*) bark was collected in small sawmills in Upper Austria and Salzburg. The bark chips were taken from bark piles generated in log debarking processes. In order to avoid changing effects at the boundary layer, the chips were retrieved at several spots and at an approximate depth of 30 cm (Kooperationsplattform Forst Holz Papier 2006). The bark was industrially dried from an initial moisture content (MC) of about 100 % to a final MC of about 6 % using a vacuum dryer (Brunner High VAC-S/HV-S1).

Bark particles were chipped using a four-shaft shredding system (RS 40, Untha Shredding Technology) and fractionated to a defined particle size using alternately hand sieves and a laboratory sieving machine. Particles of different sizes (between 6 and 45 mm) and different size distributions were used (details can be found in the publications).

The bulk density and densification properties of the loose bulks were determined. This is important, as the bulk density of a loose fill material is the lower bound of a potential composite made from that material. Bark particles of three fractions (0 mm  $\leq x_1 < 8$  mm, 8 mm  $\leq x_2 < 13$  mm, 13 mm  $\leq x_3 < 45$  mm) were put inside a prismatic box and the density was determined (DIN EN 15103 2010). The densification properties were estimated by concussing the box filled with particles and measuring the density again afterwards.

The bark particles were mixed with different adhesives in a laboratory blender. Then comparatively light panels with a density between 200 and 500 kg/m<sup>3</sup> were manufactured at laboratory scale by hand-forming a mat in a mold and hot-pressing it in a laboratory press (Höfer HL OP 280). Barkbased insulation panels of varying species, particle size, resin type, resin content, press factor (press time per mm panel thickness), and target density were produced (an overview is given in Table 4). A detailed design of experiments and the number of test specimens can be found in the publications in the appendix.

All the samples were stored at standard conditions (20 °C/65 % RH) to achieve a comparable equilibrium moisture content and were then cut according to the relevant standard (EN 326-1 1994) to create test specimens. Then the samples' density was determined. The MC after storing the samples at standard conditions ranged between 12.2 and 15.6 % in the various experiments conducted.

Specimens with controlled particle orientation (horizontal and vertical to panel plane) were produced in order to study the effect of particle orientation on thermal conductivity in Publication 6. Therefore, a special press mold was applied to manufacture panels with a defined particle orientation. Boards

with particles oriented orthogonally to panel plane were manufactured using two steel plates with a wooden rim, which defined their distance (30 mm) and panel size (240 × 350 mm<sup>2</sup>). This press mold was mounted vertically with a narrow side open. Particles were then added to the mold and their orientation was corrected with an aluminum stick. Afterwards, the particles were cold pre-pressed by applying pressure only in the direction of the panel plane. Then the press mold was closed on all sides and put into the hotpress, where the panel-mat was cured. Panels with plane-parallel particle orientation could be manufactured by leaving the press form open at the surface and putting it in a horizontal position. Resinated particles were strewed into it and the particles were oriented in longitudinal direction with a scraper. Finally, these panel mats were also cured in a final hot press process.

Paper	Bark type	Particle size in mm	Resin type	Resin content in %	Press factor in s/mm (temp. in °C)	Target den- sity in kg/m³
1	Pine	8 ≤ x <sub>1</sub> < 13 13 ≤ x <sub>2</sub> < 30	UF	8 12	18 (180)	350 400 500
3, 4	Larch	6 ≤ x < 10	Tannin-hexamine (quebracho)	5 8 10 15 20	24 (180)	250 300 350 400 500
5	Larch	6 ≤ x < 10	Tannin-hexamine (quebracho, mi- mosa)	10	15 (180) 20 (180) 24 (180)	350
6	Larch	6 ≤ x < 10	UF	10	16 (180)	200 283 366 450 500

Table 4. Parameters for bark panel production at laboratory scale.

# 3.2 Determination of physical-mechanical bark insulation board properties (Publications 1, 2, 3, 5)

# 3.2.1 Measurements conducted

The bark insulation panels were to be characterized with regard to physical-mechanical properties. In order to do so, compressive resistance (CR), modulus of rupture (MOR), modulus of elasticity (MOE), tensile strength (T), and internal bond (IB) were determined using a universal testing machine (Z 250, Zwick Roell). In addition, thickness swelling (TS) after 2 and 24 hours of storage time in water absorption (WA) after 2 and 24 hours of storage time in water were measured. Moreover, the thermal conductivity of bark insulation boards was determined, partly at different measuring temperatures, using a lambda-meter (EP500, Lambda Measurements Technologies Corporation). For single samples in Publications 3 and 5, also the density profiles were determined using a laboratory density profile analyzer (Dense-Lab X, EWS), and they were analyzed for maximum, minimum and average density.

#### 3.2.2 Statistical data analysis

Data analysis was performed using the software PASW Statistics 18 (IBM). Different statistical methods had to be applied due to varying scales of the variables. In Publication 1, the influence of the density on the physical-mechanical board properties was quantified using coefficients of correlation (Pearson). Furthermore, a multivariate analysis of variance, including interaction effects, was conducted in order to quantify the influence of particle size, resin content, and target density on the dependent variables (physical-mechanical board properties).

In Publication 3, a linear bivariate regression analysis was performed in order to study the influence of density and resin content on the panel properties. The explanatory power of the regressors was assessed by means of standardized regression coefficients (Beta-values), because a direct assessment was not possible due to different scales (Backhaus et al. 2011).

In Publication 5, again a unidimensional multivariate analysis of variance was used to analyze the influence of the independent variables (tannin type, amount of hexamine hardener, and press time) on the material properties. The strength of the influence was quantified by applying partial eta-squared values. The dependancy of TC on panel density was studied using a linear regression analysis.

Finally, important panel properties such as internal bond — a main characteristic of particle cohesion — and thermal conductivity were compared to commonly available insulation materials. For this purpose the Ashby-chart-technique (Ashby 2011) was used in order to evaluate the suitability of the panels for practical application.

#### 3.2.3 Transient heat flow in bark insulation layers

Spruce (*Picea abies*) bark particles were fractionated ( $8 < x_1 < 13 \text{ mm}$ ), dried, and poured into a wall construction element (1,300 × 1,250 × 330 mm<sup>3</sup>). Within this wall element, eight temperature sensors were placed at regular distance. Then the wall element was positioned between two climate

chambers and was subjected to a winter temperature difference (20 °C/67 % relative air humidity (RH) and -15 °C/50 % RH). The moisture content of the bark particles was measured using gravimetric moisture determination. Temperature measurements were taken at an interval of five minutes over 552 hours. Then the time-dependent temperature distribution in the wall element was studied.

A modeling function for the time- and position-dependent temperature in the wall element was determined using Fourier's second law (Meschede 2015), and the results observed were discussed with values for other insulation materials known from literature.

Finally, condensation zones within the wall element were determined by analyzing bark moisture content change over time and it was tried to retrace the observed values applying the Glaser method (ÖNORM B 8110-2 2003).

# **3.3** Bonding of bark insulation panels (Publications 3, 5)

On the one hand, the influence of the resin amount and the resin type on the physical-mechanical board properties was studied using ANOVA and regression analysis (3.2.2). On the other hand, tannin resin at different polymerization steps was examined by means of Fourier transform infrared spectroscopy (FTIR) in order to better understand the chemical processes within the resin system dealt with in Publication 5.

Samples (2 g) of each tannin extract (mimosa — *Acacia mearnsii*, quebracho — *Schinopsis balancae* spp.) powder were mixed with water (10 g). The pH-value of the mixture was adjusted to 9 by adding NaOH solution (32 %). Then, 0, 10, and 20 weight-% of a hexamine solution (33 %) were added. The resins were dried at 180 °C for 16 h and then stored at reference climate (20 °C/65 % RH) for 2 days. The hardened resins were grounded to fine powder and analyzed with an attenuated total reflectance (ATR)-equipped PerkinElmer Frontier FTIR spectrometer. Spectra were obtained in the range of 4000 – 600 cm<sup>-1</sup> at a resolution of 4 cm<sup>-1</sup> from powder samples of a few milligrams by accumulating 32 scans. Each sample was scanned three times. Before averaging, the spectra were baseline corrected and vector normalized, using the software Unscrambler (Camo Software). They were then discussed by comparing the obtained spectra with literature values.

# 3.4 Structure-property relationships in bark insulation boards (Publications 4, 6)

# 3.4.1 CT-based structure analysis

## Reasons for applying computer tomography

Since a composite material consists of two or more material phases, its interior structure has to be considered to adequately address its thermal conductivity (Hale 1976). Measurement technologies for non-destructive evaluation of physical wood properties were developed because an object often has to be inspected without being destroyed. For this purpose a huge variety of different non-destructive testing techniques (NDT) are available. They can be classified regarding the physical properties of interest or by the wavelength of radiation interacting with the specimen (X-ray, infrared, microwave, ultrasonic, nuclear magnetic resonance [NMR]). The method used depends on the resolution needed for the specific material component studied. X-ray, for example, is suitable for measuring the density variation in wood for analyzing macrovoids in particleboards, whereas NMR spectroscopy is more suitable for detecting the chemical structure of the specimens (Bucur 2003).

Tomographic images are created by ionizing radiation, whereby the source or detectors are moved around the object studied (Figure 3). The attenuation of X-rays, which depends on the density and chemical composition of the sample, is measured. The slices produced are used to calculate 3D objects, using sophisticated algorithms (Bucur 2003). Depending on an object's volume and the scale of the heterogeneity which is to be detected, different tomographic methods are available (Figure 4): macro-XCT or standard-XCT (> 10  $\mu$ m), micro-focus X-ray tomography  $\mu$ -XCT (> 3  $\mu$ m), sub- $\mu$ m XCT (> 0.4  $\mu$ m) and synchrotron tomography s-XCT (currently > 0.2  $\mu$ m; Kastner and Heinzl 2015).



Figure 3. Functioning of X-ray computed tomography (Kastner and Heinzl 2015).



Figure 4. Resolutions and object sizes accessible to X-ray computed tomography (Kastner and Heinzl 2015).

Various work has been carried out applying computed tomography (CT) for similar questions as the ones dealt with in the present study, which confirms the adequacy of the method chosen. X-ray computed tomography (XCT) proved to be a promising technique to study the inner structure and microstructure of wood and wood-based composites (Kaick and Delorme 2005). This is due to its non-destructiveness, the possibility to simultaneously investigate material structure, three-dimensional geometry, and, to a certain extent, physical and mechanical properties. Moreover, improvements in computer hardware have reduced the efforts to apply tomographic methods (Kastner and Heinzl 2015).

Computed tomography was used to study the 3D-density distribution of particleboard, OSB and MDF (Standfest et al. 2009b). Fibrous networks of a light insulation fiberboard were studied by means of CT. 3D images were obtained and porosity, fiber and pore size distributions were determined by means of mathematical morphology in digital image analysis. Individual fibers could be defined during image segmentation and the real fiber network was modeled, which could be used as a basis for numerical analysis (Lux et al. 2006b). X-ray tomography, combined with morphological image analysis tools, was used to describe a fiber network and to model the thermal conductivity of wood-fiber-boards (Lux et al. 2006a).

X-ray micro-tomography also proved to be a suitable tool for studying tannin-derived foams. The restrictive parameter determined hereby was the limited resolution of 4 microns ( $\mu$ m). Nonetheless CT and coupled image analysis were found to be very useful, as they allowed for a non-destructive sample analysis (Tondi et al. 2009a). A wood composite made of wood fibers and leather residues was assessed with X-ray computed tomography, finding that the material phases could be

distinguished, and pore size distributions were determined (Wieland et al. 2013). Moreover, sub-µm-CT was successfully applied to characterize pores within OSB and particleboard. Good correlations were found between mean pore diameter and board density (Standfest et al. 2012). Finally, the resin distribution in particleboard was studied applying X-ray micro-computed tomography (Evans et al. 2010). Moreover, the resin distribution on the fibers in MDF was studied using synchrotron microtomography (Walther and Thoemen 2009).

#### Structural evaluation of bark insulation panels

Larch (*Larix decidua*) bark insulation panels of varying density (200–500 kg/m<sup>3</sup>) and a size of 50 × 50 × 20 (30) mm<sup>3</sup> were scanned with an industrial computed tomograph.

The scans were conducted at the Upper Austrian University of Applied Sciences in Wels using a Nanotom 180 NF desktop sub-micro CT device (GE Measurement & Control) with a 180-kV nanofocus tube with an exchangeable transmission target and a flat panel detector with 2,304 × 2,304 pixels. The X-ray tube allows a maximum voltage of 180 kV, but scans were conducted with 60 kV. The measurement current was 410  $\mu$ A and the integration time at the detector was 1000 ms. Molybdenum was used as a target material. The source-to-detector distance in the machine is 0.5 m and samples with a maximum diameter of 68 mm can be scanned.

16-bit tiff-image stacks with a voxel size of 27-31 microns<sup>3</sup> were exported for further data processing. The image stacks were linearly scaled from a minimum of 0 to a maximum of 255 grayscale (8-bit). A panel cross section area of 1.5 mm from the surface and 1.5 mm from the edge was not taken into account to avoid inhomogeneities caused by edge effects. For each sample, 50 images at regular distance were considered in the evaluation, and the frequencies for gray values were determined (Equation 1, equations can be found in the mathematic appendix). The number of images was reduced (it corresponds to a reduction of resolution in *z*-direction) in order to reduce calculation time when applying the modeling algorithm.

An algorithm based on ANOVA was developed for the segmentation of CT-tomograms in order to distinguish material phases (void, inner bark, and outer bark). The optimization criterion for the definition of borders between material phases is based on the idea that the variation of gray values within classes has to be as low and, between classes, as high as possible (Figure 5, Equation 2; Otsu 1979, Standfest et al. 2009a, Petutschnigg et al. 2009) to obtain a good threshold. Therefore, the gray value variance in the groups was minimized according to Equation 3, using the Microsoft Excel Solver tool. The quality of the threshold obtained was quantified by the amount of between-group variance and corresponding p-values of ANOVA. Porosity, component ratios, and pore size distribution were determined, based on the structural information gained from CT, applying morphological operators (Standfest et al. 2012). For this purpose, the open source software ImageJ

was used. Correlations between structural characteristics (percentage of void, inner bark, and outer bark ratio), panel density, and thermal conductivity were investigated.

3D-matrizes describing the bark board samples (visualizations can be seen in Figure 6) were exported to further model thermal conductivity.



Figure 5. CT-tomogram of a bark insulation panel and its gray value histogram with optimized class boundaries and summarized theoretical normal distribution (Kain et al. 2016b).



Figure 6. Rendering of samples with horizontal and vertical particles (size =  $50 \times 50 \times 30$  mm<sup>3</sup>, density = 382 and 206 kg/m<sup>3</sup>, porosity = 0.20 and 0.53).

#### 3.4.2 Modeling of heat flow

An algorithm based on finite differences was used to model the panel's thermal conductivity. The structural characterization was used as a basis for a 2D (Publication 4) and 3D (Publication 6) numerical thermal conductivity model applying the finite difference method, thus enabling the theoretical study of steady-state heat flow in the boards.

In addition to Publication 4, the influence of the particle orientation on the thermal conductivity was evaluated in Publication 6. CT was used to gain information about the interior structure of the panel. Then the modeling algorithm was extended to a 3D set-up and the thermal conductivity of the panels with defined particle orientation was modeled. The influence of density and particle orientation on thermal conductivity was evaluated applying an analysis of covariance (Equation 4) with partial eta-squared-values (Equation 5). Heat flow density in spatial directions was evaluated. Furthermore, the deviation of the heat flow from the direction of the global temperature gradient was modeled and overlaid with the structural situation. Therefore, structure-property relationships in bark-based insulation boards were discussed in order to enable theoretical ex-ante studies.

In the following, a short introduction to heat flow modeling will be given in order to enable understanding of the functionality of the model applied (considerations are restricted to the 3D-version of the algorithm; the 2D-algorithm is presented in Publication 4).

#### Modeling heat flow applying finite differences

Heat energy is an orderless molecule movement. It can be transported by radiation, conduction or convection. Conduction only takes place in solid matter through molecule pushes. The energy theorem states that heat always flows along a temperature gradient; the steeper the gradient, the stronger the flow (Meschede 2015). Temperature can vary within space and is therefore a function of the room position. For this function, the temperature gradient gradT can be defined, which is a vector field with the partial derivatives of the partial differentiable scalar field (temperature field T; Equation 6).

For a small volume dV the heat energy content Q changes as shown in Equation 7. The divergence is a differential operator that assigns a scalar field to a vector field. The divergence in a point is the sum of the directional derivatives of the components in a point. The divergence  $div(\dot{q})$  describes the change of the heat content per volume and time unit. The heat capacity of a volume is  $\rho * c_p * dV$ and considering Fourier's first law in Equation 6, the general model for heat conduction is derived (Equation 8). It can be used for transient heat flow problems (Neunzert et al. 1998). Including also internal heat sources, the general Fourier law for heat conduction is established (Equation 9; Marek and Nitsche 2012). The thermal diffusivity  $\lambda/(\rho * c_p)$  is therefore the transport coefficient for the transient heat transport. Using the gradient (grad) and divergence (div), the Fourier-law can be displayed without coordinates. Equation 9 is a partial differential equation of second order. Thereby the definition of the initial condition for the temperature distribution at time  $t_0$  is important for transient heat flow problems (Equation 10). Boundary conditions are obligatory for the solution of partial differential equations for both the stationary and the transient heat flow. First type boundary conditions (Dirichlet boundary condition) define the temperature at the particular border ( $T(x = x_0, y, z, t) =$   $T_0(y, z, t)$ ). Second type boundary conditions (Neumann boundary condition) define the heat flow density and consequently, the temperature gradient at a particular border ( $\dot{q}(x = x_0, y, z, t) = -\lambda * dT(x, y, z, t)/dx | x = x_0$ ). Finally, third type boundary conditions (Newton boundary condition) characterize heat transfer from a solid surface to a fluid, and forth type boundary conditions (Stefan boundary condition) are used to describe simultaneous conduction and radiation on a surface (Marek and Nitsche 2012). In this dissertation, stationary heat flow processes with a one-dimensional temperature gradient were considered. The surface temperatures next to the lambda-meter platen were defined to be  $T_1$  and  $T_2$ , which corresponds to a first type or Dirichlet boundary condition. The sample edges were insulated in the experiment and, in theory, no heat flow interaction from the sample with surrounding space exists (adiabatic). Therefore, on the edges, second type boundary conditions were applied in the specification of adiabatic surfaces ( $\dot{q}(x = x_0, x_X, z = z_0, z_Z) = 0$ ).

For most standard geometries, analytical solutions for partial differential equations related to heat transfer exist. For inhomogeneous, multi-phase objects or complicated geometries, numerical solving-procedures are used. One attempt is the application of grid methods. They allow to replace differentials with difference quotients (Strikwerda 2004).

Numerical solving primarily requires the discretization of a partial differential equation. The differential of a function in a point  $x_0$  is defined according to Equation 11. If not the limit for  $h \rightarrow 0$  is considered, but replaced by the difference quotient, the differential can be approximated (Equation 12). The error that is made with this approximation (symbolically shown with the Landau-symbol *O*) depends on the increment of  $\Delta x$  (Strikwerda 2004). The spatial room can thus be overlaid by a grid (Figure 7). The derivation of a function *f* at position (*x*, *y*, *z*) can be expressed with 7 points ( $h = \Delta x = \Delta y = \Delta z$  [equidistant]) following Equation 13 (Thomas 1995).

There are many possibilities to solve partial differential equations numerically (e.g., finite differences, finite elements, spectral methods, collocation methods, etc. [Le Dret and Lucquin 2016]). For this thesis finite differences are particularly relevant, because CT-scanning results in pixel- (or voxel- in the 3-dimensional case) images. Those pixels (voxels) can be directly used as volume elements for discretization as shown in the following. Therefore, the description of methods concentrates on finite differences. In this work, the finite difference method is used as a grid method with volume elements (Figure 7).

Provided that temperature conditions are stationary, finite differences can be used to describe the heat flow balance (Equation 14). The sum of heat flows around a volume element is zero in the stationary case (Equation 15). Finally, boundary conditions have to be considered as outlined at the beginning of the chapter.



Figure 7. Volume element and its neighboring elements with entering and leaving heat flows. Dimensions are  $\Delta x \times \Delta y \times \Delta z$ . *Z* goes into paper plane.

#### Modeling the bark panels' thermal conductivity

The CT tomogram stacks were segmented into void, inner bark, and outer bark. For numeric modeling, the thermal conductivity values for the three compartments were separately determined for the panels with horizontal and vertical particles (Equation 16). The constraint of the thermal conductivity of still air (void) having to be at least 0.025 W/(m\*K) was included (Ebert 2013). For the thermal model, the stack's resolution was linearly reduced to  $200 \times 111 \times 50$  pixels to reduce calculation time. The resulting gray value matrix comprised  $1.1125 \times 10^6$  voxel elements (Figure 8). Further steps were conducted using the software Matlab R 2009 (MathWorks).

Each gray value corresponds to a thermal conductivity, which is represented in a 3D thermal conductivity matrix. For numeric modeling, a matrix T(x, y, z) was created where every matrix element represents a pixel's temperature, according to Equation 17. Since the voxels are prismatic and not cubic (Figure 8), the directional (x, y, z) heat flows had to be weighted by the factors  $F_x$ ,  $F_y$ , and  $F_z$  based on Equation 18.



Figure 8. Visualization of voxel element (y-direction is orthogonal to panel plane).

The heat flow around an element was defined as 0 (i.e., a stationary temperature condition). Due to three spatial dimensions, various cases had to be considered: (i) heat flow around internal elements (Equation 19), (ii) heat flow around exterior corner elements (Equation 20), and (iii) heat flow around exterior edge elements (Equation 21). In doing so, a system of equations for every point of the grid was defined. Afterwards the coefficients were arranged in a coefficient matrix L, the boundary conditions were summarized in a matrix R and the unknown temperatures were given by matrix Y. Solutions to the equation system were determined based on Equation 22. The heat flow density (Figure 9) orthogonal to panel plane (y-direction) was determined following Equation 23, and the average sample thermal conductivity was calculated using Equation 24.

Specific heat flows depending on spatial directions (Figure 10) could be determined with Equation 25 and the deviation of heat flow vectors from the plane that is orthogonal to the panel was calculated with Equation 26.



Figure 9. Heat flow density in W/m<sup>2</sup> in a bark board sample (density 382 kg/m<sup>3</sup>, average heat flow density 38.94 W/m<sup>2</sup>, thermal conductivity 0.072 W/(m\*K), temperature gradient 0.56 K/mm).



Figure 10. Calculation of heat flow angle (*y*-direction is orthogonal to panel plane).

# 4 Main investigations

#### 4.1 Publication 1: Substantial bark use as insulation material

Günther Kain, Marius-Catalin Barbu, Alfred Teischinger, Maurizio Musso, Alexander Petutschnigg 2012, Forest Products Journal, Volume 62(6):480–487

#### Abstract

Tree bark is a resource which has not been used much for products with a higher value added. This is indeed surprising. Given that tree bark has many interesting natural properties, such as a relatively low density, low thermal conductivity, low flammability, and a high degree of extractives, the aim of the present study is to evaluate whether particleboard made out of tree bark is suitable as insulation material.

In this study, the bulk density and densification properties of bark particles were evaluated. Moreover, bark-based insulation panels made out of pine (*Pinus sylvestris*) bark were produced at three different density levels (350, 400, 500 kg/m<sup>3</sup>), with two different particle sizes ( $8 < x_1 < 13$  mm,  $13 < x_2 < 30$  mm), and two urea-formaldehyde (UF) resin amounts (8 % and 12 %). Furthermore, the physical-mechanical properties (modulus of elasticity [MOE], modulus of rupture [MOR], compressive resistance [CR], tensile strength [T], internal bond [IB], thickness swelling [TS], water absorption [WA], and the thermal conductivity [TC]) of the boards were evaluated.

It was found that the bulk density of loose bark bulks (densified by concussion) depends on particle size and ranges from 190 to 255 kg/m<sup>3</sup>, and that concussion of particles leads to a densification between 13 and 20 %. The panels produced proved to be sufficiently stable compared with commonly available insulation materials such as mineral wool, PUR-foam or wood wool ( $0.1 \le IB \le 0.2 \text{ N/mm}^2$ ). The thermal conductivity of the investigated boards ranges from 0.06 to 0.09 W/(m\*K). Results showed that the bark boards' properties seem to be promising with regard to thermal conductivity, heat storage capacity, and mechanical characteristics. The obtained values were compared with those of commonly available insulation materials, showing that the limiting factors are moisture resistance with low resin content, mechanical stability with low densities, and a high thermal conductivity compared with very light insulation materials. On the other hand, it could be shown that the panels' thermal diffusivity is superior to commonly known insulation materials. For this reason, bark-based insulation panels could probably be used for efficient heat-storage-active insulation layers in civil engineering.

# 4.2 Publication 2: Using bark as heat insulation material

Günther Kain, Marius-Catalin Barbu, Stefan Hinterreiter, Klaus Richter, Alexander Petutschnigg 2013, Bioresources, Volume 8(3):3718–3731

#### Abstract

As shown in Publication 1, bark insulation panels have a thermal conductivity which is higher than 0.05 W/(m\*K), but have a low thermal diffusivity. It is a material parameter describing the rate at which a heat wave moves through a material and is therefore very relevant to transient heat flow processes. In the presented work, this transient behavior of bark insulation was studied on a real-size wall element.

Spruce (*Picea abies*) bark particles were used as an insulation fill material for the thermal insulation of a timber frame wall ( $1300 \times 1250 \times 330 \text{ mm}^3$ ), which was subjected to a simulated winter temperature difference between indoor (20 °C/67 % RH) and outdoor climate (-15 °C/50 % RH). The temperature profile development of the wall's cross section was measured over time. In addition, the moisture content of the bark was determined at the beginning and end of the experiment. Moreover, the results obtained were used to test an analytical model for the time-dependent temperature distribution in a wall based on Fourier's heat equation.

It was shown that bark layers conducted heat more slowly than commonly known blow-in insulation materials due to their low thermal diffusivity. The observed time-dependent temperature profile was compared with the modeled results. The model applied describes the real thermal material behavior quite well and can be used to study time-dependent heat flow processes in bark insulation layers. Moreover, material moisture development due to water vapor streams caused by vapor pressure differences between the inside and outside climate was studied, and it confirmed general timber construction rules. The widely used Glaser-method can be used effectively to predict water vapor transport and potential condensation zones within bark insulation layers.

These important findings prove that laboratory measurements focusing on thermal characteristics (thermal conductivity and thermal diffusivity) can be extended to real-size wall elements. Summing up, the study has effectively shown the languid thermal behavior of a bark insulation layer, which is the basis for specific applications of the material.

# 4.3 Publication 3: Density-related properties of bark insulation boards bonded with tannin hexamine resin

Günther Kain, Viola Güttler, Marius-Catalin Barbu, Alexander Petutschnigg, Klaus Richter, Gianluca Tondi

2014, European Journal of Wood and Wood Products, Volume 72(4):417-424

#### Abstract

Building owners are increasingly interested in a healthy and sustainable living environment, which is a trend favoring ecological building materials with outstanding structural physical parameters. To meet this specific demand, the research question of the present paper is whether a tannin-hexamine resin based on renewable resources can replace the widely used petrol-based urea-formaldehyde (UF) resin.

Insulation boards made from particles of larch bark (*Larix decidua* Mill.) bonded with a formaldehydefree tannin (quebracho) resin were pressed at different levels of density (250, 300, 350, 400, and 500 kg/m<sup>3</sup>) and resin content (5, 8, 10, 15, 20 %). Then these panels were evaluated for their physical-mechanical properties (MOR, MOE, IB, TS, WA, TC), applying a regression model with standardized regression coefficients in order to test the influence of the production parameters.

It could be shown that light boards (target density 250 kg/m<sup>3</sup>) can be pressed and that their thermal conductivity is low (0.065–0.090 W/(m\*K)). With regard to physical-mechanical characteristics, the influence of panel density and resin content was studied, and it was found that density and resin content are positively correlated with MOR and IB. Both TS and WA are lower when the resin content is higher. A higher panel density also reduces TS and WA.

Discussion of the panel properties showed that a certain compaction ( $\rho \ge 350 \text{ kg/m}^3$ ) is necessary to meet the requirements for IB of the relevant standard. Interestingly, the resin amount did not influence the mechanical board properties as strongly as expected, which made panel density the most important variable in this regard. In terms of TS, a resin content of at least 15 % is necessary to limit the swell to 15 % after 24 hours of storage in water.

In conclusion, the study has proved that tree bark cannot only be used for substantially upgraded insulation panels, but can also be bonded with a formaldehyde free tannin resin, in which case the technical panel characteristics are comparable to standard insulation materials.

# 4.4 Publication 4: Analyzing wood bark insulation board structure using X-ray computed tomography and modeling its thermal conductivity by means of finite difference method

Günther Kain, Johann Charwat-Pessler, Marius-Catalin Barbu, Bernhard Plank, Klaus Richter, Alexander Petutschnigg

2016, Journal of Composite Materials, Volume 50(6):795-806

#### Abstract

As shown in Publications 1, 2, and 3 the use of the natural "tree insulation material" bark for building insulations is somehow limited by a relatively high thermal conductivity of 0.05–0.08 W/(m\*K). In the present study, the interior structure of bark insulation boards was analyzed and it was evaluated whether the knowledge of the structural composition enables thermal modeling of the panels in order to assess optimization potentials.

Insulation boards made out of larch (*Larix decidua*) bark with a density range between 250 and 500 kg/m<sup>3</sup> were scanned with an industrial computed tomograph in order to study the structure of the boards. The CT images were segmented using a thresholding algorithm based on ANOVA. Digital image analysis was performed to evaluate the panels' porosity. Based on the board's microstructure, a numerical model based on the finite difference method for thermal conductivity was applied.

The segmentation method proved to be suitable for distinguishing three material phases: void, inner bark, and outer bark. That could be demonstrated by a high between-class variance and highly significant F-values of the model. Studying the pore size distribution within the panels showed that, on the one hand, it can be influenced in the pressing process, and, on the other hand, the main source for optimization is the distribution of macro voids. The thermal model proved to predict the panels' thermal conductivity quite precisely, confirmed by a comparison with measured thermal conductivity values. The limitations of the model were also demonstrated by finding that the model precision decreases with decreasing panel density. Especially with the light boards (~ 250 kg/m<sup>3</sup>) the deviation of the model is higher, probably due to the larger voids.

These findings pave the way for further developments of efficient bark insulation panels with welldefined pore structure. Finally, the model proposed can be used to study effects of structure variations prior to production.

# 4.5 Publication 5: Effects of different flavonoid extracts in the optimization of tannin-glued bark insulation boards

Günther Kain, Viola Güttler, Bernhard Lienbacher, Marius-Catalin Barbu, Alexander Petutschnigg, Klaus Richter, Gianluca Tondi

2015: Wood and Fiber Science, Volume 47(3):1-12

#### Abstract

Tannin-based resin was shown to be suitable for replacing commonly available condensation resins in bark insulation boards in Publication 3. Nevertheless, using a tannin-hexamine resin based on quebracho (*Schinopsis balancae* spp.) tannin resulted in partly weak IB and TS properties. Therefore, it is the aim of this study to evaluate whether the use of mimosa (*Acacia mearnsii*) tannin for the production of bark insulation panels has an advantageous effect on panel properties.

Thus, bark insulation panels were produced from larch (*Larix decidua*) bark and bonded with a formaldehyde-free tannin-hexamine resin. Quebracho and mimosa tannin, containing different levels of hexamine, were mixed with bark particles at different ratios and cured in a hot press at varying press time. Finally, physical-mechanical board properties were studied and the polymerization of different resin formulations was assessed using FTIR spectroscopy.

Mechanical (MOR, MOE, IB) and water-related (TS, WA) board properties of mimosa bound panels proved to be superior. This was confirmed by MOR and IB being higher (69 and 31 % respectively) and TS and WA being lower (5 and 6 % respectively) when mimosa tannin instead of quebracho tannin was used. This was supported by Fourier transformed infrared (FTIR) spectroscopic analysis of tannin polymers, which gave insight into the chemical activation by the hardener and possible reasons for the better performance of mimosa resin. Most important in this respect proved to be the lower initial viscosity of mimosa tannin (due to a higher degree of initial in-plane bending of the polymer before polymerization), which resulted in better penetration of particles and, hence, greater adhesive performance. The crosslinking effect of the hexamine was confirmed by an evaluation of absorption spectra obtained by FTIR-spectroscopy. Analysis of the physical-mechanical properties suggested that a hexamine amount of more than 6 % is not beneficial. Evaluating the thermal conductivity of the panels showed that its dependence on temperature is comparable to commonly available insulation materials.

Finally, optimal press time and amount of hexamine hardener were derived, which is important because of the influence on the profitability of potential industrial production.

# 4.6 Publication 6: Evaluation of relationships between particle orientation and thermal conductivity in bark insulation board by means of CT and discrete modeling

Günther Kain, Bernhard Lienbacher, Marius-Catalin Barbu, Bernhard Plank, Klaus Richter, Alexander Petutschnigg

2016: Case Studies in Nondestructive Testing and Evaluation, Volume 6:21-29

#### Abstract

As shown in Publication 4, the targeted design of a bark-insulation-panel structure could be a means to reduce thermal conductivity. It was the aim of this study to evaluate the effects of particle orientation on the thermal conductivity in bark panels. Moreover, the model for thermal conductivity used in Publication 4 should be extended to a 3-dimensional version in order to enable the modeling of spatial effects.

Insulation boards made out of larch (*Larix decidua*) bark were pressed with a defined particle orientation (horizontal and vertical to panel plane) and scanned with an industrial X-ray computed tomograph (CT) in order to study the structure of the boards and to allow for structure-based thermal modeling. The CT images were segmented using a categorization algorithm based on ANOVA. A numerical model for thermal conductivity using finite differences was applied based on the board's microstructure gained from CT.

Panels with horizontal particles (oriented parallel to the panel plane) proved to have a significantly (13 % on average) lower thermal conductivity than panels with vertical particles (oriented orthogonally to the panel plane). This trend could be confirmed by means of the presented modeling approach, with a mean deviation of 5.7 % from measured values. The model of local heat flow in the panel could be used to study the direction of heat flow vectors on a voxel level. The average heat flow density in void is significantly higher in panels with horizontal particles compared to those with vertical particles. Findings showed that the average deviation of the heat flow from the direction of the global temperature gradient is significantly higher with vertical particles. This is due to bridges built by particles between panel surfaces, which are followed by the heat flow.

The findings point the way towards developments of efficient bark insulation panels with a welldefined microstructure. Contrary to simple cut-ups, the application of CT and subsequent modeling enables the evaluation of the effects of particle orientation on a panel's thermal conductivity, which is the basis for a theoretical ex ante optimization in the production process.

## 4.7 Own contribution

#### **Publication 1**

Günther Kain developed the idea to use tree bark as insulation material, organized the material, pressed the panels, measured their mechanical and physical properties, conducted statistical evaluation, and composed the article. Alfred Teischinger and Maurizio Musso gave advice on content and data evaluation. Klaus Richter contributed with scientific information to the state of the art. Marius-Catalin Barbu and Alexander Petutschnigg guided the research project and revised the article.

#### Publication 2

Günther Kain developed the measuring concept, evaluated the data and wrote the article. Stefan Hinterreiter and his students manufactured the test wall and helped with logistic aspects. Marius-Catalin Barbu, Klaus Richter, and Alexander Petutschnigg contributed with critical discussions to the refinement of the research questions and revised the article.

#### **Publication 3**

Günther Kain defined the research question, developed the design of experiment for the test samples together with Gianluca Tondi, carried out statistical data evaluation, and wrote the article. Viola Güttler produced the panels and conducted the measurements. Marius-Catalin Barbu, Alexander Petutschnigg, Klaus Richter, and Gianluca Tondi discussed the results with the main authors, shaped and critically revised the manuscript before submission.

#### **Publication 4**

Günther Kain prepared the test samples, carried out digital image analysis, developed the model, wrote the code for modeling, performed the data evaluation, and wrote the article. Bernhard Plank made the CT-scans. Johann Charwat-Pessler and Alexander Petutschnigg contributed to image analysis. Marius-Catalin Barbu, Klaus Richter, and Alexander Petutschnigg guided the research progress with recommendations and assisted and advised the manuscript editing.

#### **Publication 5**

Günther Kain developed the design of experiment for test specimens, carried out statistical data processing and wrote the article. Viola Güttler and Bernhard Lienbacher prepared the test samples

and performed the physical-mechanical measurements. Günther Kain and Viola Güttler conducted the FTIR-spectroscopy, Gianluca Tondi contributed to the discussion of absorption spectra. Marius-Catalin Barbu, Alexander Petutschnigg, Klaus Richter, and Gianluca Tondi gave advice on content and structure of the article and proofread it.

#### **Publication 6**

Klaus Richter and Alexander Petutschnigg raised the question of the influence of the particle orientation on thermal properties of the bark boards. Günther Kain developed the design of experiment for test specimens, developed the model, wrote the code for modeling, performed the image analysis and data processing, and wrote the article. Bernhard Lienbacher prepared the test samples and Bernhard Plank conducted the CT-scans. Alexander Petutschnigg and Klaus Richter gave critical input on the model applied. Marius-Catalin Barbu and Klaus Richter contributed to structure and content of the article and revised it.

# 5 Synthesis

### 5.1 Introduction

In the course of this thesis, the potentials of bark as bio-based feedstock for thermal insulation applications have been investigated. The production process for bark-based particle insulation boards was developed at a laboratory scale and material characteristics were determined. Furthermore, the insulation capacity of bark was tested on a real-size wall element. The standard, petrol-based resins of the wood-panel industry could be replaced at laboratory scale by a flavonoid-resin, based on renewable resources, and potentials for optimization of the pressing parameters were proposed. Finally, the structure of the bark-based insulation panels was studied by means of CT and a discrete modeling algorithm for the panels' thermal conductivity was developed, which allows for the theoretical analysis of heat flow processes in the panel. In this chapter, the main findings of this thesis will be critically discussed and an outlook for potential further research will be presented. Finally, closing words will conclude this dissertation.

## 5.2 Discussion of main findings

The research conducted can be subdivided into four main topics:

- Production of bark panels
- Physical-mechanical characteristics of bark insulation panels
- Optimization of the resin system used
- Study of bark panel structure and abstract modeling of thermal conductivity

Eventually, the aim of any research activity is to add to existing knowledge. Hence, the four main issues of this dissertation will be given further consideration in the following paragraphs. Nonetheless, the main findings cannot be seen isolated as they influence each other (Figure 11) and justify their integrated presentation as a dissertation thesis. Efforts in panel production are the basis for substantial bark use as insulation material. Physical-mechanical panel characteristics had to be evaluated in order to clarify the potentials of lightweight bark boards for thermal insulation applications. Most importantly in this respect was that the real thermal performance of bark insulation was evaluated in a real-size test wall element, confirming the material properties determined with small samples. In order to create a panel mainly based on renewable resources, petrol-based components (UF resin) were replaced by a tannin-hexamine resin, whose influence on physical-mechanical board properties had to be assessed. Finally, the structure studies and the subsequent

abstraction in modeling approaches laid the theoretical basis for an ex-ante estimation of the effects of changes to the bark panels produced.

Secondary questions concerning the effects of two different tannin-hexamine resins (as a substitute for the UF-resin) on physical-mechanical board properties were discussed. Additionally, optimal production parameters focusing on the use of tannin adhesives were evaluated and the gained insights regarding the influence of the panel structure on its thermal conductivity were discussed with consideration of board production, bonding, and material characterization (Figure 11).



Figure 11. Main investigation issues of the dissertation and their interactions.

## 5.2.1 Bark panel production

To start with, the idea of using bark as a resource for particleboard is not new. Many attempts have been made to partly or totally replace wood particles with bark particles (e.g., Volz 1973, Nemli and Colakoglu 2005, Yemele et al. 2008b, Gupta et al. 2011) or bark fibers (Xing et al. 2006). The primary aim of those investigations was to use a cheaper resource to increase profit margins in the wood composite production. Other investigations proposed to use bark as fill-in insulation material in

granulate form (Naundorf et al. 2004), as raw material for pallet blocks (Kain et al. 2013), bark-plastic composites (Yemele et al. 2010), outside cladding (Mühlbacher and Taylor 2009), bark textiles (Heintz 2015), decorative panels for interior design applications (Egger 2014), or interior automotive panels (Rwawiire et al. 2015). These examples are recent approaches highlighting the potential of bark as a highly optimized material. Another approach is to see bark as a feedstock for biorefinery (an overview is given by Feng et al. 2013). First systematic research on bark properties proposing its use as insulation material was conducted by Martin (1963) and Schneider and Engelhardt (1977).

#### Bark bulk density

In the course of this dissertation it could be shown that bark particleboards of low density (< 500 kg/m<sup>3</sup>) can be produced at laboratory scale (Figure 12). Measurements in the course of Publication 1 on the density of loose pine bark bulks showed their density at a moisture content of 12 % to range between 169 (13 mm  $\le x_1 < 45$  mm) and 213 (0 mm  $\le x_2 < 8$  mm) kg/m<sup>3</sup> depending on particle size. Literature values on bark bulk density show a high variation (Gupta et al. 2002) due to non-uniform species, varying moisture content, particle geometry, densification, and particle size. The volume-based conversion factor between solid bark and loose bark particles is on average 0.301 in industrial debarking processes (Kooperationsplattform Forst Holz Papier 2006). Gupta et al. (2002) determined the bark density of solid particles for a mixture of balsam fir, white spruce, and black spruce bark with 540 kg/m<sup>3</sup> (corrected to a moisture content of 12 %). Using a volume conversion factor of 0.301, the bulk density would be 163 kg/m<sup>3</sup>, which is in line with the measurements for the coarse fraction. Miranda et al. (2012) determined the bulk density of air-dried Scots pine bark with 202 kg/m<sup>3</sup> on average for several fractions.

The densification by concussion was determined to be 13 % with the coarse particles (13 mm  $\leq$  x<sub>1</sub> < 45 mm) and 20 % with the fine particles (0 mm  $\leq$  x<sub>2</sub> < 8 mm), which coincides with measurements of Böhm and Hartmann (2005), who reported the densification of bark bulks to range between 12 and 23 %.

These findings are important, because they define a lower density bound for bark insulation panels, and the initial density of the material defines the compression ratio in the pressing process. Larger particles have a lower initial bulk density due to geometrical difficulty in compaction (Miranda et al. 2012). The higher the bark bulk density, the lower the compression ratio for constant panel density. The compression ratio in bark particleboard production is positively correlated with MOE and MOR (Yemele et al. 2008a). Bulk density is also an issue when using loose bark bulks as a blow-in insulation material, as suggested in Publication 2, because it is positively correlated with TC. Low-density particleboard (300–500 kg/m<sup>3</sup>) was produced from *Shorea* ssp. using an isocyanate resin to bind the particles. The lower limit of the compression ratio (board density divided by solid particle

density) was found to lie between 0.7 and 0.8 (Kawai and Sasaki 1993). Considering that larch bark has a particle density of 352 kg/m<sup>3</sup> at 12 % MC (Miles and Smith 2009), the compression ratio for a board with 220 kg/m<sup>3</sup> is 0.63, following the calculation method of Kawai and Sasaki (1993), and is therefore significantly lower, although the mechanical properties of such boards were poor (Publication 3).



Figure 12. Larch bark-based insulation panel (density 400 kg/m<sup>3</sup>, thickness 20 mm, tannin-hexamine resin, 6 mm  $\leq x < 10$  mm; Kain et al. 2014).

#### **Production parameters**

Pure softwood bark-based lightweight (<500 kg/m<sup>3</sup>) insulation panels have not been discussed in detail before, which was confirmed by the successful patenting of the idea (Kain et al. 2012a). The idea to use bark in heat insulating particleboards was already suggested by Martin (1963) and Schneider and Engelhardt (1977), who focused, however, on panels with a density of 700 kg/m<sup>3</sup>.

Bark particles used for the production of insulation panels had a size between 6 and 30 mm (precise fractions are given in the publications in the appendix), and are therefore coarser than in other work focusing on the production of bark particleboard (e.g., 0.02–6 mm — Blanchet et al. 2000, 0.02–7 mm — Yemele et al. 2008a, 1–5 mm — Gupta et al. 2011). The reason for choosing coarse particles was the lower achievable panel density.

A standard UF-resin proved to be suitable for binding bark particles. Resin content (8–10 %), press temperature (180 °C) and press factor (16–18 s/mm panel thickness) are comparable to other laboratory-scale particleboards produced from renewable resources (Ashori and Nourbakhsh 2008). UF-resin contents of 9 to 11 %, a press temperature of 160 °C, and press factors between 12 and 18 s/mm for particleboard made from date palm, eucalypt, mesquite and saltcedar were reported. In another study, 8 % UF-resin was used in the core layer and 12 to 16 % in the surface layer of pure bark particleboard (750 kg/m<sup>3</sup>). In that case, the press temperature was 200 °C and the press factor

21 s/mm (Blanchet et al. 2000). The press parameters used in the present study are therefore in the range of results achieved in similar laboratory experiments conducted.

For potential industrial production, resin content and especially press factor should be reduced to lower production costs (Dunky and Niemz 2002, Paulitsch and Barbu 2015).

Tannin-hexamine resin was used as a substitute for UF-resin, applying a partly higher resin content of between 5 and 20 % and a press factor between 15 and 24 s/mm. Otherwise production parameters could be kept constant. Experience from industry (Valenzuela et al. 2012) shows that industrial particleboard using solely tannin-hexamine as adhesive can be produced meeting the requirements of the relevant standards. The press factor in doing so is 13 s/mm (19 mm panel thickness), the press temperature 186 °C, the surface resin content is 12 %, the core resin content 10 %, the hexamine amount is 6 %, proving that the laboratory particleboard production for this thesis is close to industrial requirements regarding production parameters, and that there is potential to lower the press factor.

Panels with vertical and horizontal particles were produced to study the effects of particle orientation on thermal conductivity. The orientation of particles in the pressing process was conducted fully manually, which would have to be altered in a potential industrial surrounding. Nonetheless, production facilities which can influence particle orientation in particleboard production are available (Paulitsch and Barbu 2015). A systematic analysis of material structures on a meta-level has been conducted by Lakes (1993), highlighting the opportunity for optimized new materials with designed properties.

## 5.2.2 Physical-mechanical bark particleboard properties

#### **Mechanical properties**

Physical-mechanical properties of bark insulation panels are relevant regarding their technical applicability (Pfundstein et al. 2007).

It could be confirmed for lightweight bark panels that their mechanical properties are weaker than with wood particles (Yemele et al. 2008a). Whilst low-density wood particleboard (10 % isocyanate resin) with a density between 250 and 500 kg/m<sup>3</sup> had a MOE between 1000 and 2500 N/mm<sup>2</sup>, a MOR between 2.5 and 15 N/mm<sup>2</sup>, and an IB between 0.2 and 0.6 N/mm<sup>2</sup> (Kawai et al. 1986), bark particleboard (5–20 % tannin resin) with the same density range showed a MOE between 50 and 500 N/mm<sup>2</sup>, a MOR ranging from 0.3 to 3.0 N/mm<sup>2</sup>, and an IB between 0.1 and 0.3 N/mm<sup>2</sup> (some difference is due to the different resin). On the one hand, the lower mechanical properties of bark board could be explained by the lower cellulose content of bark itself (Sakai 2001), resulting in significantly lower strength properties than wood. On the other hand, the thin-walled phellogen layers

between periderms function as a separative layer (Martin and Crist 1968), leading to a lower overall strength of the composite. Another reason for the low mechanical strength of bark particleboard is that bark is a porous material absorbing the resin, and therefore adhesion levels between particles are low (Nemli and Colakoglu 2005). Strength requirements are often lower for insulation materials than they are for structurally engineered wood products (Pfundstein et al. 2007) and, consequently, a lower mechanical strength of the bark insulation panels might not be a problem. Other work focusing on low-density particleboard for insulation purposes reported panels with similar properties (e.g., binderless particleboard from kenaf core, 100–300 kg/m<sup>3</sup>, MOR 0–2 N/mm<sup>2</sup>, IB 0.02–0.17 N/mm<sup>2</sup> [Xu et al. 2004]; bagasse particleboard bonded with citric acid and sucrose, 300–500 kg/m<sup>3</sup>, MOR 1–7 N/mm<sup>2</sup> [Liao et al. 2016]). The mechanical stability (MOR, IB) of bark insulation panels was shown to be higher than that of most standard insulation materials, also due to a significantly higher density (~ 200–550 kg/m<sup>3</sup>; Figure 13).

Going into detail, the physical-mechanical board properties (i.e., CR, MOR, MOE, T, IB, TS) are positively correlated with the panel density, which has been demonstrated for wood composites in which the final density of the product is higher than the density of the bulk material (Thoemen 2010).

The resin content proved to be positively correlated with all mechanical and physical board properties. The self-bonding of particles did not seem to be an issue with low-density bark particleboard. This is in line with the findings of Gupta et al. (2011) and Marashdeh et al. (2011), who showed that self-bonding in bark particleboard is only feasible with higher panel density and especially with press temperatures above 200 °C.

The particle size proved to have a significant influence on the MOR of low-density particleboard (demonstrated with UF resin bound panels), which is on average 13 % lower when using coarser particles. Yemele et al. (2008a) found coarse particles to lead to a slightly higher MOR in bark particleboard. The authors emphasize that interaction effects with wood species and wood content of bark particles have to be taken into consideration. Similarly, it has been shown for wood particleboard that an increasing particle size has a positive effect on MOE and MOR (Arabi et al. 2011a). The reason for not finding this coherence in low-density particleboard might be a low bonding strength between particles due to the low compaction ratio and a small bonding area.

Contrary to standard particleboard (EN 13986 2015), IB was shown to be on average 33 % higher with coarse bark particles. This finding was confirmed by Yemele et al. (2008a) for 100 % bark panels. A reason might be that the coarse particles build solid bridges between panel surfaces without being interrupted by a bonding, which has a positive influence on IB. This consideration is backed by the fact that the trend is stronger with lower resin content, where consequently the negative effect of adhesive joints is stronger. In addition, the amount of fines absorbing the available resin is low when using coarse particles. In general, particle size and packing of particles significantly affect board properties, whereby fine or mixed particles can be packed more tightly. The closer

contact between particles and a greater particle surface area contribute to mechanical bark board properties (Miranda et al. 2012) — an effect studied for bark panels with a density greater than 800 kg/m<sup>3</sup> (Gupta et al. 2011). The panels in the present study are significantly more lightweight and the advantages of fine particles cannot be exploited because the compression ratio would decrease as well (Yemele et al. 2008a). By trend, low-density bark panels show better mechanical characteristics with coarse particles, which is in line with the findings of Sackey and Smith (2010) investigating the effect of different particle-sizes on the mechanical properties of industrial particleboard. The mechanical properties of bark-based insulation boards are at least comparable to commonly available insulation materials focusing on IB (Figure 13) and MOR.



Figure 13. Internal bond of bark insulation panels by comparison (Kain et al. 2014).

#### Properties focusing on moisture resistance

TS and WA both proved to be significantly lower using coarse particles, confirming what Yemele et al. (2008a) found out. Schwemmer (2010) limited the thickness swell after 24 hours of water storage to 15 % when developing an insulation material out of reed mace. In the present study, the TS of panels with 18 % UF-resin after 24 hours of water storage is significantly higher. Consequently, TS should be limited by wax additives or other resin systems (Paulitsch and Barbu 2015). It has been reported that bark reduces TS in particleboard due to phenolic compounds reacting with formaldehyde in the adhesive (Nemli and Colakoglu 2005) — an effect which could not be confirmed in the present study. TS after 24 hours of water storage for boards bonded with Mimosa tannin could be kept below 15 %, although no wax or other water repellants were used in laboratory trials.

#### **Thermal properties**

Measurements of the thermal conductivity showed that it is positively correlated with the density ( $R^2 = 0.9$ ). Investigations of the bark particles' bulk density specified that the minimum board density is about 200 kg/m<sup>3</sup>, with the thermal conductivity being 0.055 W/(m\*K). Wood shavings and kenaf fibers were used to press insulation boards with a density of 70 kg/m<sup>3</sup>, showing a thermal conductivity of 0.061 W/(m\*K) (Nakaya et al. 2016). Bark has good insulation properties, because the bark panels in the present study have a comparable TC with a significantly higher density. Boards with a density of 500 kg/m<sup>3</sup> showed a thermal conductivity of approximately 0.09 W/(m\*K). Sonderegger and Niemz (2009) investigated particleboard with constant density, finding that smaller particles lead to reduced thermal conductivity — a relationship that could not be confirmed for the presented lightweight barkbased particleboards. A reason might be that both studied particle fractions were relatively coarse (8 mm  $\le x_1 < 13$  mm, 13 mm  $\le x_2 < 30$  mm), and due to the thinness (20 mm) of the studied specimens the structures of void and solid matter were similar.

Spruce (*Picea abies*), pine (*Pinus sylvestris*), and larch (*Larix decidua*) bark particles were used to produce low-density particleboards in the course of this dissertation. Their TC is comparable and mainly influenced by panel density, although there might be advantages to larch bark (Figure 14), as it has the lowest density compared with spruce and pine bark (Miles and Smith 2009) and can therefore be used to produce lighter panels with, consequently, a lower TC.

With regard to temperature dependence of TC across the investigated range (10–40 °C), the thermal conductivity of the panels was shown to be significantly correlated with the panel temperature. The gradient of the linear regression function for temperature-dependent TC was 0.32. Wood wool (0.31, 348 kg/m<sup>3</sup>) has a comparable, mineral wool (0.13, 145 kg/m<sup>3</sup>) a significantly lower gradient in this respect (Abdou 2005). This finding is relevant because the effectiveness of insulation layers applied to limit the thermal load of a building is also a function of the ambient temperature. From this viewpoint, the studied bark panels showed a satisfactory rather than an excellent performance.



Figure 14. Thermal conductivity of bark boards by comparison (modified from Kain et al. 2012b).

An experiment with a larger scale wall element (1300 × 1250 × 330 mm<sup>3</sup>), filled with spruce (Picea abies) bark particles showed that the transient heat flow can be well described by Fourier's second law (Meschede 2015). Bark particles were loosely poured into the wall cavity, resulting in a bulk density of 258 kg/m<sup>3</sup>. The thermal conductivity of that bulk was recursively determined and ranges between 0.057 and 0.062 W/(m\*K). The specific heat storage capacity of the bark was estimated according to the results of Martin (1963) and ranges between 1475 and 2139 J/(kg\*K). This results in a thermal diffusivity of between 1.027\*10<sup>-7</sup> and 1.983\*10<sup>-7</sup> m<sup>2</sup>/s (Figure 15). This level is definitely much lower than with the established insulation materials (e.g., mineral wool, polystyrene; Figure 15). The analysis showed clearly where the potential of bark insulation lies for practical application. It is not so much situations where only the heat flux according to Fourier's first law is to be limited, but rather applications where insulation has to be combined with a high heat storage capacity (which for example is the south wall of a building, optimized for passive solar heating or roof insulations preventing overheating in summer; Martin 1963, Ashby 2011). In future, the energy demand for cooling buildings will increase significantly. One approach to reduce the cooling energy demand is to apply building insulation with a high heat storage capacity, so that solar radiation does not result in a quick temperature increase in the interior rooms on sunny days (Bettgenhäuser et al. 2011). Bouguerra et al. (2001) showed for wood concrete mixtures (up to 50 weight-% wood particles with a size between 3 and 8 mm) that wood composite materials possess thermal inertia and heat storage capacity together with good thermal insulation. The same applies to the developed bark insulation boards, demonstrated by the measurements on the wall element.



Figure 15. Thermal conductivity and thermal diffusivity of insulation materials by comparison (figure modified from Kain et al. 2012b).

At this stage, it also needs to be discussed, whether particles are the best basis for bark insulation composite production. There is work suggesting the use of bark fibers (Moncada et al. 2016). Bark fibers were used for the production of MDF (740 kg/m<sup>3</sup>), finding that the obtainable fiber size distribution varies strongly between tree species, and refining parameters have to be adapted specifically (high wear and tear in refining machinery when using bark [Paulitsch and Barbu 2015]). It has also been highlighted that bark fibers show higher bulk densities than wood fibers, due to the abundance of short fibers. The resulting mechanical properties proved to be inferior to those of pure wood fiber panels (Xing et al. 2006). Submicron and nano-sized cellulose fibers (CNFs) were produced from lodgepole pine (Pinus contorta) bark with 80 % of the fibers having a length below 0.4 mm. CNFs are considered to be a good raw material for packaging applications due to their ability to form hydrogen bonds and the resulting high barrier properties (Nair and Yan 2015). Further research will have to clarify whether bark fibers can be the basis for insulation materials, especially with regard to fiber length and energy efficiency, as a lot of energy is required for refining. Finally, the cellulose content is significantly lower in bark than it is in wood (Fengel and Wegener 2003). The benefit of particle-based bark boards is the low effort (investment and tool wear) in material preparation. Consequently, a comparison of the amount of energy used in the process of refining bark with insulation efficiency gains caused by this refinement has to be suggested.

### 5.2.3 Bonding of bark insulation panels

Considering the segmentation of the insulation material market in Europe (Papadopoulos 2005, Carus et al. 2008, Jelle 2011), which is dominated by petrol-based products, the potential market

position of tree bark insulation can only be that of a niche among insulation materials based on renewable resources. Considering the origin of its resources, the petrol-based resin system (ureaformaldehyde) used in the first trials of producing bark insulation panels, has to be replaced. Many researchers have proposed to crosslink tannins in an alkaline environment (Pizzi 1982, Pichelin et al. 1999, Tondi and Pizzi 2009, Mansouri et al. 2011). Therefore a tannin (quebracho)-hexamine resin was used to produce bark-based insulation panels at a laboratory scale with a density between 250 and 500 kg/m<sup>3</sup>. Their mechanical and moisture-related properties were evaluated and it was found that they are comparable to condensation resin bound panels. This assessment is shared by Pizzi (2016), stating that synthetic resins in the wood composite production can be largely replaced by bio-sourced adhesives. Only with regard to moisture performance slight disadvantages have been detected. Summing up, the performance of a standard wood fiber insulation panel (EN 622-4 2010) regarding MOR was only achieved with a density of 350 kg/m<sup>3</sup>, and in order to limit the TS after 24 h of water storage to 15 %, a resin content of 15 % was required. These findings are similar to those of Kim et al. (2003), determining the TS of wood particleboard with a tannin resin content of 14 % (mimosa with paraformaldehyde as a hardener) to be between 10 and 17 %.

Furthermore, the production parameters hardener (hexamine) content and pressing time were optimized to meet industry requirements. It was found that the use of mimosa tannin leads to superior panel performance compared with quebracho tannin, in terms of mechanical (also shown by Pizzi et al. 1995) and water-related properties. This is due to a lower initial viscosity and therefore better particle wetting. The only chemical difference between the two tannin species was observed with regard to aromatic in-plane bending of flavonoids (Tondi and Petutschnigg 2015) based on the conducted FTIR spectroscopy of tannin without hexamine. The mimosa showed a higher absorbance in the respective wavenumber zone. Macroscopically, that results in lower viscosity of mimosa tannin, backing the physical measurements. It has been reported that mimosa tannin has a lower degree of polymerization than quebracho tannin (Pizzi and Stephanou 1993), confirming the present macroscopic observations. Moreover, the linear quebracho tannin molecule might produce a higher amount of secondary forces (Pasch et al. 2001), resulting in a higher initial viscosity.

Concerning the press factor, it was shown that 15 s/mm panel thickness (studied with 20 mm thick panels) is enough, as mechanical properties did not improve with longer hot-pressing. This is in contrast with findings of Kim et al. (2003), which showed that mechanical properties gain from increasing press time when they produced particleboard from wood particles and mimosa tannin. A potential reason might be that the authors used paraformaldehyde as a hardener, whose reactivity towards the tannin was shown to be higher than that of hexamine. Finally and most importantly, it could be shown for mimosa tannin resin that 6 % of hexamine (whe/wt) is sufficient and that an increased hardener amount does not lead to a better panel performance. Also Kim et al. (2003) found that more than 6 % of hexamine in a tannin resin did not significantly increase MOR and IB of

particleboard. This experimental finding was confirmed by an FTIR-spectroscopy of tannin polymers with a varying hexamine amount. It was shown that with the addition of hexamine, amino-methylene activation occurs, which is an indicator of a higher number of crosslinks. Further signals at specific frequencies indicate the networking of the tannin molecules when hexamine as a hardener is added. Spectroscopic studies did not clearly show a positive effect of a higher hardener amount and therefore supported the experimental findings. In an industrial application of a pine tannin-hexamine adhesive, also a hardener amount of 6 % is used (Valenzuela et al. 2012). In another industrial application of a tannin-hexamine adhesive it was shown for thick wood panels (up to 120 mm) that press steam injection can help to overcome aggregation problems of tannin-hexamine resins (Pichelin et al. 2006). This technology might be suitable for reducing press time in bark particleboard production.

#### 5.2.4 Panel structure and thermal modeling

Whilst the first three research directions were very application-oriented, the investigation of the panel structure and the abstraction of heat flow processes within bark panels can be seen as theoretical considerations, with the potential to inspire the other areas (Figure 11). Examples for manifold approaches to measuring and modeling thermal conduction processes in wood were given by Deliiski (1977) and Weres et al. (2000). Olek et al. (2003) evaluated various literature focusing on heat flow modeling in wood, finding that specific heat storage capacity, moisture content, and the temperature of specimens have to be considered to obtain good modeling results. The best models for thermal conductivity are more complex, including physical and morphological wood information. Heat flow processes in bark have not been considered in models so far due to a low economic interest in its use.

#### Structural characterization

Bark panels were scanned by means of CT. As bark is a highly heterogeneous material (Holdheide and Huber 1952), the segmentation of the gray value images was a complex task requiring the application of an algorithm based on ANOVA (Otsu 1979, Petutschnigg et al. 2009, Wieland et al. 2013). Similarly, the method could be applied for segmenting CT-images of OSB and particleboard (Standfest et al. 2009a) as well as a wood-leather composite (Wieland et al. 2013). Segmented images were used as a basis for structural evaluation and thermal conductivity modeling. An algorithm based on finite differences was applied in a 2D and 3D set up, explaining the heat flow in the panels quite well. The input parameters (properties of the single material phases) were derived from data and discussed by the relevant literature (Martin 1963, Hale 1976, Ebert 2013).

SYNTHESIS

The porosity of bark insulation boards was studied, finding that it is low (10–49 %) compared with high-performance insulation materials (93 %; Li et al. 2013b), leading to a relatively high thermal conductivity with a minimum of approximately 0.05 W/(m\*K). Porosity (inter-particle voids) of wood particle mats before pressing ranges between 57 and 73 % (higher with coarse particles), that of a particleboard (670 kg/m<sup>3</sup>) between 6 and 12 % (Sackey and Smith 2010). In this respect, the low-density bark boards were produced with a porosity near that of the loose bulk. Further work will have to clarify whether an optimization of particle shape and especially panel-to-particle thickness ratio might be a means to reduce board density whilst also creating a sufficiently stable panel. It has been shown for particleboard that density and resin content can be reduced without losing mechanical stability by choosing the optimal particle shape (Arabi et al. 2011a), and that thicker particles increase void in particleboard (Sackey and Smith 2010).

As discussed, the lower density of bark insulation panels is limited to approximately 200 kg/m<sup>3</sup> for stability reasons; a source of optimization is to find the ideal interior structure of bark composites. It has been shown that spherical inter-spaces result in the lowest thermal conductivity values (Ordonez and Alvarado 2012). Heat conduction variation in wood was found to be primarily influenced by voids, whereas variation in bulk structure and chemical composition has negligible effects (Suleiman et al. 1999). Smaller pores (predominantly intra-particle voids) in the investigated bark panels (<1 mm<sup>2</sup> — in a 2D consideration) are rather spherical, whilst the bigger pores (inter-particle voids, >1 mm<sup>2</sup>) are more compact. Especially the bigger voids can be actively influenced in the production process (shown in Publication 4) and might be an interesting source of optimization.

Heat flow in wood has been thoroughly investigated. Knowledge of heat flow processes in wood is important for processing (e.g., timber drying and wood modification, cooking and steaming of logs for veneering, composite production in hot presses, etc.), but also for wood use (e.g., heat transfer in constructions; Niemz 1993, Vay et al. 2015). Less work has been carried out concerning heat flow in bark and the existing work mainly focuses on TC of particleboard with a density >500 kg/m<sup>3</sup> (Schneider and Engelhardt 1977) or insulation capability of bark in case of forest fires (Bauer et al. 2010).

The TC of bark board phases was indirectly estimated based on the phase ratio in two independent experiments (Publications 4 and 6). The results in this discussion are therefore referred to as a range of values. The TC of still air in void was shown to range between 0.025 and 0.030 W/(m\*K) — values which are confirmed by Ebert (2013) with 0.025 W/(m\*K), Thoemen et al. (2008) with 0.026 W/(m\*K), and Hale (1976), mentioning that with fibrous insulation materials the conduction through air in the voids accounts for 0.027 W/(m\*K). The range of thermal conductivity of void in the present investigation might be attributed to slightly differing void sizes. The inner bark in the present insulation panels was shown to have a TC between 0.076 and 0.094 W/(m\*K), the outer bark between 0.108 and 0.153 W/(m\*K). The findings indicate slightly higher conductivities when using

vertical particles. A possible reason is that particles are more strongly compacted when they are oriented vertically to panel plane, and therefore a slightly higher density of inner and outer bark results in the slight increase of TC. A direct measurement of the minimal local density differences in CT images is not possible, because contrary to medical CT, no calibrated gray values were used. The assumption is backed up by density profiles measured in Publication 5, partially showing heterogeneous panel densities. In the case of particleboard porosity, Sackey and Smith (2010) also experienced that coarse particles (>2 mm) show high compression resistance, leading to cell collapse in the pressing process. A detailed study of relevant material properties is of great importance to numeric modeling (Troppová et al. 2014). Therefore, the efforts in material characterization of bark insulation boards can be justified.

#### Structure-property relationships

Profound investigations on the effects of particle orientation showed that the thermal conductivity of the bark boards can be decreased by 13 % when orienting the particles parallel to the panel plane (Figure 16). Bark was shown to have a significantly lower thermal anisotropy than wood (Martin 1963, Vay et al. 2015) and chip orientation is less important in panel production from this point of view. Nonetheless, if the aspect ratio of particles is greater than one, their orientation becomes relevant for a composite's thermal conductivity, because the composite is then considered to be inherently anisotropic. The more the particle orientation deviates from the direction of the global temperature gradient, the lower the resulting thermal conductivity, which has been shown for WPC (Couturier et al. 1996). This relationship has been confirmed with the present study and also by Tiedje and Guo (2014), modeling the thermal conductivity of multiple particle composites. Schneider and Engelhardt (1977) determined the TC (orthogonal to panel plane) of bark (spruce, pine, beech) particleboard with a density of 700 kg/m<sup>3</sup>, produced in a flat press process with 0.122–0.158 W/(m\*K) at 12 % MC. The authors suggested that panels produced with particles lying orthogonally to plane have a significantly lower thermal conductivity, because the TC of panels with horizontal particles showed a 30 % lower TC in plane than orthogonal to plane. The opposite fact was found in the present study - a potential reason is that with the coarse particles used, the void size and distribution is of higher importance for the global panel TC than the slight TC-difference of bark in its anatomical directions. This was confirmed by Joščák et al. (2012) stating that particles and fibers in wood composites with a low TC should be oriented orthogonally to heat flow direction. Another study from Brombacher et al. (2012) on the TC of various insulation materials revealed that differences are not only due to density differences, but also resin system, resin content, particle geometry and orientation proved to have an influence. The authors noted that, for example, fiberboards produced in a wet process have fibers predominantly oriented parallel to panel plane and therefore have a lower TC than fiberboards produced in a dry process with 3D fiber orientation. That effect can be

explained by the TC of wood being 2.25 to 2.75 times higher parallel to grain (Sonderegger and Niemz 2012). Multilayered wood lamellae panels with horizontal grain proved to have a slightly lower TC than those with vertical grain because of the lower tangential TC of wood (Bader et al. 2007). That effect should be lower with bark boards due to a lower thermal anisotropy (Martin 1963), and therefore the TC-differences of the present study might refer to the distribution of voids.

The results obtained also confirmed indirectly that the tangential orientation of flakes in the bark of larch (Figure 2) enhances the bark layers' thermal insulation properties on a tree. From that point of view the present discussion of particle orientation in a bark particleboard is the transfer of a biologically-inspired architecture (Studart 2016) to a technical material.

Wood composites consist of wood elements bonded with synthetic resin in various combinations and configurations (Gillespie 1981, Paulitsch and Barbu 2015). Consequently, panel structure evaluations are an interesting source of optimization, shown by Thoemen et al. (2008) for wood fiber networks. Considering that bark particleboard can have a structure determined by particle orientation and bark particles have a structure themselves (Lakes 1993), especially the orientation of particles seems to be relevant to the panels' global TC.



Figure 16. Influence of particle orientation on panel thermal conductivity and modeled results (modified from Kain et al. 2016b).

#### Modeling the thermal conductivity

Flow in inhomogeneous media, to which wood composites belong, is the topic of the mathematical discipline called flow theory, which deals among others with the conductivity and permeability of space filled with particles with differing properties (Wang et al. 2006). The structure of the composite
is inherently important for conductivity properties, which is especially relevant as that structure can be influenced to some extent in the production (mat forming and pressing; Thoemen 2010). Efficient modeling can be used to engineer the design of building products to serve for special applications (steam diffusion barrier, thermal-insulating structural elements, etc.).

The thermal conductivity of samples was determined by means of a 2D (Publication 4) and a 3D (Publication 6) model for TC. For panels with vertical and horizontal particles, the 3D calculations led to slightly better results (5.7 % average deviation from measured values compared with 8.6 %). As with measured values, also with the model panel density ( $\eta^2 = 0.95$ ) and particle orientation ( $\eta^2$ = 0.85) have a highly significant (p < 0.001) influence on the thermal conductivity. The thermal model captures the trend of the real measurements, based on the examination of the slopes of the regression functions, quite well (Figure 16). For both particle orientations the real values were undervalued by the model (i.e., 6 % for the vertical and 5 % for the horizontal particles). A possible reason might be attributed to the fact that heat transfer due to radiation and convection in larger voids is of higher importance (Hale 1976, Joščák et al. 2012); an effect which is not considered in the model working with effective thermal conductivities, which combines the effects of different heat transfer mechanisms (Ebert 2013). This simplification was also proposed by Fan et al. (2006) when predicting effective TC of wood samples obtained precise results. At ambient temperatures, the effects of radiation are minor. Convection within voids less than 3-4 mm in diameter is low (Hale 1976, Joščák et al. 2012); however, especially in panels with a density below 300 kg/m<sup>3</sup>, the pores are partly larger and as a result the model precision decreases.

Analysis of the heat flow density in the different spatial directions showed that the heat flow in xand z-direction (in-plane) is 100 to 1000 times lower than in y-direction. This observation is logical for the given adiabatic edge definition, where heat flux always follows the direction of the temperature gradient (Meschede 2015).

Although the model slightly undervalues the thermal conductivity, the trend is recognizable and the thermal processes on a voxel level were used to learn about the reasons for the lower thermal conductivity of the panels with horizontal particles. The average heat flow density in void is on average 20 % higher (p < 0.001) in boards with horizontal particles than in those with vertical particle orientation. The heat flow density in inner and outer bark, however, is on average 9 and 28 % lower (p < 0.001) in boards with horizontal particles (Figure 17). In other words, more heat energy is forced through area with high heat flow resistance, which results in a lower global TC. Moreover, the average heat flow density is significantly higher in the outer bark with vertical particles than with horizontal particles, and thus the outer bark's higher TC has a more disadvantageous influence (Figure 17, Figure 19). This issue is dealt with in the theory of optimal heat conduction pathways, in which the heat flow avoids the dispersed phase (in the present case void) if the TC of the continuous phase (bark) is higher than that of the dispersed phase (Carson et al. 2005).

The average deviation of heat flow from the direction of the temperature gradient is significantly (p < 0.001) lower in boards with horizontal particles with board densities below 400 kg/m<sup>3</sup>. Consequently, the heat flow is forced to find its way through void with higher heat flow resistance. The difference of heat flow deviation is not significantly different statistically in the bark board compartments. Considering Fourier's first law, one can see that the heat flow through areas with low thermal resistance (inner and outer bark) is impeded and therefore the thermal conductivity on a sample's global level is lower (Figure 20). That influence is minor regarding boards with a density higher than 400 kg/m<sup>3</sup> (Figure 18).



Figure 17. Average heat flow density in bark board compartments (Kain et al. 2016b).



●void horiz. ●inner bark horiz. ●outer bark horiz. Ovoid vert. Oinner bark vert. Oouter bark vert.

Figure 18. Average deviation of heat flow from *y*-direction in different bark board compartments (Kain et al. 2016b).



Figure 19. Heat flow density in a bark panel with horizontally and vertically oriented particles (density 380 kg/m<sup>3</sup>, temperature gradient 0.56 K/mm; modified from Kain et al. 2016b).



Figure 20. Deviation of heat flow from *y*-direction in a bark panel with horizontally and vertically oriented particles (density 380 kg/m<sup>3</sup>, temperature gradient 0.56 K/mm; modified from Kain et al. 2016b).

Model fit was very good to satisfactory, showing in critical discussion that the numerical model proved to describe the basic trends, but slightly underestimated the real values in part (especially when particle orientation varies). It can be concluded on that issue that the approach presented is likely to quantify the overall sample conductivity, but does not consider secondary effects (such as convection processes within larger voids). This limitation to the approach chosen was mentioned by Gu (2001) studying transient heat conduction in wood. The author also applied small control volumes and defined heat flow balances to solve the equation systems. The results showed that to increase model precision a parallel consideration of mass transfer mechanisms is necessary. Discrete numerical procedures were also used by Zombori (2001) to study transient effects during hot pressing of wood-based composites. The underlying partial differential equations could not be solved analytically and were therefore also solved applying a finite difference scheme, confirming the adequacy of the approach chosen in the present thesis. Finally, Khattabi and Steinhagen (1993) used computerized finite difference solutions for the description of 3D transient heat flow in a piece of wood, confirming the method to be suitable and adequately precise.

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One limitation to the present results is the choice of the sample volume in thermal modeling. Representative volume elements (RVEs) refer to the smallest volume a measurement or model concentrates on, so that it becomes statistically representative for the entire material (Kanit et al. 2003). Therefore, RVEs are of particular importance in the theory of composite materials. The particles of the bark insulation panels are relatively coarse (Figure 5), and consequently the specimens ( $50 \times 50 \times 20$  [30] mm<sup>3</sup>) chosen for structural characterization and modeling are not representative for the whole bark panel. Nonetheless, a similar sample size was chosen to characterize OSB and PB and it was found that this sample size is suitable for providing information on structural elements (Charwat-Pessler et al. 2014). The results regarding structure and thermal modeling have to be restricted to the samples studied and further investigation will have to concentrate on larger samples. Nevertheless, the model applied fits the measurements of TC conducted with larger panels quite well.

It can be concluded that the present modeling method is interesting with regard to linking the microstructure of a material with its thermal properties. The importance of knowing more about the link between microstructure and material properties was also highlighted by Thoemen et al. (2008) using 3D-modeling to link structural composition and thermal properties of wood fiber mats for MDF production. The strength of the approach presented is that the method is structure-based and that it can be adapted to other problems and possibly other materials.

### 5.3 Potential for future research

Nature is inherently complex and the more it is investigated the more questions occur, which for a natural scientist is reassuring and motivating likewise. So with this dissertation on the suitability of softwood tree bark for thermal insulation applications and some considerations of theoretical concepts, research questions were adequately addressed, but also new questions were proposed, which shall be discussed at this point.

#### Optimization of the proposed bark-based insulation panels

When using bark-based panels as a thermal insulation material, their thermal conductivity should be reduced. Here four main approaches seem to be feasible.

First of all, the bark could be prepared differently (i.e., in fibrous form; Xing et al. 2006, Nair and Yan 2015). Consequently the initial bulk density could decrease, allowing also for a lower panel density.

Secondly, a foaming resin (i.e., tannin foam) system could be applied to fill the voids between the bark panels, which would limit the convective effects and therefore result in a lower thermal

conductivity. It was shown that expandable fillers (expandable polystyrene granulates) in particleboard can be used to lower the board density significantly (Shalbafan et al. 2016).

Thirdly, the addition of low-density components (as was tried with pop-corns) would reduce the global conductivity of the composite. In addition, the heavier bark fractions (outer bark) quite strongly contribute to heat flux in the panels (Figure 17). Therefore, sorting those compartments out is likely to reduce the panels' global thermal conductivity (also discussed in Publication 4 - replacing the outer bark with the inner bark resulted in a TC reduction of 2.5 % in the model; this effect could be enhanced by a more restrictive density-based sorting). It has been shown for straw (Silva et al. 2011) and bark (Miranda et al. 2012) that size fractions with a specific chemical composition can be separated, which should also apply to density. Wood is used in various forms (solid wood, particles, and fibers) as a feedstock for insulation materials (Paulitsch and Barbu 2015). It could therefore be interesting to evaluate the potential of wood-bark-mixtures as insulation material. The advantages of each material (wood – strength and better adhesion properties, bark – lightweight, low thermal conductivity) could be exploited forming a composite with advanced properties. This has already been proposed in an early work by Place and Maloney (1975) showing that particleboards with a core layer containing bark particles have a significantly lower TC than pure wood particleboard. Today this finding could be particularly interesting regarding structural engineered wood products with heat insulating layers.

Fourthly, bark origin and particle morphology have a significant effect on physical-mechanical bark particleboard properties (Yemele et al. 2008a). Consequently, a strategic choice of bark type and particle geometry could be a source for bark-based panel optimization. For example, it has been shown that the board density of particleboard can be decreased without adversely affecting mechanical properties by choosing the right particle size (Arabi et al. 2011a). Slenderness ratio of particles in wood particleboard strongly influences its mechanical properties (Arabi et al. 2011b) and particle geometry is influenced by the raw material and its processing (Juliana et al. 2012). Moreover, macro-voids in particleboard can be effectively influenced by targeted particle size mixtures (Sackey and Smith 2010). Consequently, it seems to be promising to study the effects of particle geometry on mechanical and thermal properties of bark insulation board, aiming at a further reduction of density without lowering the mechanical stability and creating a composite with a high porosity in order to lower the global thermal conductivity.

As the bark panels do not have a very low (<0.05 W/(m\*K)) thermal conductivity by nature, perhaps they could also be efficiently used for acoustic insulation. The surface of the panels — especially with coarse particles — is rough, which is ideal for the absorption of sound waves and their subsequent dissipation into heat energy (Hazrati-Behnagh et al. 2016). Moreover, panels with fine particles have a very smooth and colorful surface (Mazzitelli 2014, Tudor 2014), which is likely to serve as an aesthetic, decorative panel (a first attempt in this respect can be seen in the Austrian

contribution to the SolarDecathlon — an international competition for low energy houses; Solar Decathlon Team Austria 2013).

Finally, a process-based optimization will be necessary for industrial application. Although many production issues have been addressed in this thesis, some fine adjustments, especially with the tannin resin (for example industry requirements concerning pressing and curing time) will be necessary.

#### Further research regarding bark use as insulation material

The present dissertation focuses mainly on laboratory production and physical-mechanical board properties. For practical application, the durability of the panels is very relevant. Bark contains a high amount of sugar-containing components, especially in the phloem (Sakai 2001, Fengel and Wegener 2003), which are attractive for insects and fungi. On the other hand, bark is made up of a comparatively high amount of extractives and polyphenolic acids, which add to the durable character of a material (Barabash and Levin 1970, Sakai 2001). There is work suggesting a good resistance of spruce bark against soft rot fungi and termites (Morris et al. 1999). Moreover a study on sugi bark (*Cryptomeria japonica* D. Don) clearly showed that bark has a higher resistance against fungi than the respective wood (Doi and Kurimoto 1998). The decay resistance of particleboard could be improved by impregnating particles with a pine (*Pinus brutia*) bark extract (Nemli et al. 2006). Nonetheless, the durability of bark insulation board should be an issue of further research.

The same is true of flammability. Bark has a significantly higher amount of inorganic compounds than wood (Sakai 2001), and a trees' sensible cambium is protected by the bark in forest fires (Bauer et al. 2010, Odhiambo et al. 2014). A study on the flammability of spruce bark pellets with 50 % brown coal for wall insulation showed that bark is fire-retardant, because spruce board started to burn earlier than the bark pellets under the same experiment conditions (Naundorf et al. 2004). Nevertheless, the real flammability of light bark boards and smoldering in case of fibers has to be assessed in standardized industry tests (promising results were obtained in preliminary small flame tests [SFT]).

In the course of the present study, the moisture resistance of bark-based insulation boards was assessed measuring TS and WA after 2 and 24 hours of water storage. In addition, the moisture-related aspects of bark panels have to be studied regarding equilibrium moisture content at a given climate, sorption behavior, and steam diffusion properties. Fundamental work on natural bark has been carried out by Martin (1963), focusing on the thermal conductivity of bark and its relation to moisture content. Martin and Crist (1968) investigated the volumetric and linear dimensional expansion of bark from saturation point to ovendry conditions and Niemz (1993) and Holmberg et

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al. (2016) measured the sorptive behavior of bark. These properties should also be discussed with regard to bark composites.

Another aspect concerning bark ingredients is emissions. Wood and bark can emit terpene compounds, which can be oxidized to simple aldehydes like formaldehyde. In addition, natural solid wood releases a small but detectable amount of volatile organic compounds (VOCs) such as terpenes and organic acids (Que et al. 2013). Both formaldehyde and VOCs can have a negative health impact (World Health Organization 2006). The amount of emissions strongly depends on wood species, temperature, intensity of solar radiation, moisture content of wood, and storage conditions (Roffael 2006). Their negative effects are stronger in modern energy-efficient buildings, due to low air exchange rates leading to higher concentration levels in the air (Kim et al. 2003). Investigations have shown that bark contains a higher amount of extractives compared with wood (Prasetya and Roffael 1991, Fengel and Wegener 2003). This has positive implications like the fact that, for example, bark in particleboards acts as a formaldehyde scavenger (Prasetya and Roffael 1991, Takano et al. 2008, Costa et al. 2013). Orientation tests on bark insulation panels showed low formaldehyde emissions — lower than 4 (UF) and 0.5 (tannin) mg/100g atro-panel with the perforator method (EN 120 2011) and lower than 0.1 (UF) and 0.04 mg/l with the desiccator method (JIS A 1460 2001, Lohninger 2014). This is due to the reaction of phenolic bark compounds with the formaldehyde (Nemli and Colakoglu 2005). A natural function of bark is to emit volatiles in order to attract insects within plant communities (Szmigielski et al. 2012). Nevertheless, there could also be negative effects, such as the unwanted release of VOCs into interior rooms. In how far emissions from bark insulation panels could have a negative influence on the human health should be a topic of further investigations.

Apart from technical and application-oriented aspects, the economic situation of bark insulation panels should be given a detailed discussion. On the one hand, softwood bark appears in large quantities in the Alps, the Carpathian region, and Europe in general (calculated from forest cuts, Eurostat 2011, BMLFUW 2014b, Chapter 2.2.4). On the other hand, it is a source of process energy in the wood industry (Ogunwusi 2013, Nosek et al. 2016). It has to be clarified, whether a potential upgrading of high-quality bark to insulation materials is economically reasonable considering material costs, production costs, and potential prices achievable on the market for bark insulation panels. Moreover, the ecological benefits of using bark as insulation material should be quantified in a life-cycle analysis (Mantau 2015). The production costs of particleboard depend on many variables, of which the most important is the raw material price (Buehlmann et al. 2000). Bark particleboard production should be economical in this respect, because for the period of January 2005 to May 2012, softwood bark in Austria was on average 38 % (SD = 8 %) cheaper than softwood chips and 14 % (SD = 12 %) cheaper than sawdust referred to a loose cubic meter (Kain 2013).

All in all, it could be shown that bark has very specific advantages (e.g., resource basis, thermal diffusivity) whilst also having disadvantages (not a very low thermal conductivity, heterogeneous composition, etc.) which will result in specific applications. It is a trend in insulation material use to adjust the material choice to the particular structural situation (Jelle 2011) — an approach favoring the applicability of bark as an insulation material. Consequently, a more detailed characterization of the material will be necessary with regard to classical structural physical parameters. These are, besides density and thermal conductivity, steam diffusion resistance, moisture resistance, and fire resistance.

#### Questions arising from modeling efforts

The numeric model used works with effective thermal conductivities, which primarily consist of conduction, but also incorporate effects of convection and radiation. In order to improve the quality of the model, it could be tried to incorporate all heat transfer mechanisms. This will definitely lead to a much higher complexity of the model, because then, for example, also the inner surfaces of voids will be of importance. Modeling approaches based on this consideration might require a numeric model based on finite elements instead of finite differences (Rappaz et al. 2003).

Moreover, the study of the wall element filled with loose softwood bark has shown that due to water vapor diffusion, condensation water occurs where the dew point is undercut. Steam diffusion processes are described by similar partial differential equations as the heat flow (Meschede 2015). Therefore, it is likely that the model proposed can be transformed for steam diffusion modeling, which was also suggested by Thoemen et al. (2008) when they investigated permeability and thermal conductivity of fiber-based boards. As the density profile of the panels can be controlled within the pressing process, building materials with defined steam diffusion resistance could be produced. An extension of the numerical model could serve as a valuable basis for that development.

#### 5.4 Closing words

Bark as an interesting natural resource has been evaluated on a scientific level and the link to practical application has been established. At a time where humankind partly overuses the planet's resources (Meadows 2004), the industrial exploitation of the, so far, neglected resource bark is adequate. Today the focus sometimes lies too much on growth, whereas a higher efficiency of resource use would have additional and sustainable benefits (Sharp et al. 2015). It could be shown with this dissertation that softwood bark is a highly efficient material by nature, which can serve specific purposes. In the attempt to use this potential technically for thermal insulation, the theoretical

basis for understanding and further optimizing of bark insulation materials has been laid. Many application fields for bark insulation on an industrial scale are imaginable, for which this work might be a valuable basis.

### 6 References

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# 7 Nomenclature

# Latin Symbols

а	thermal diffusivity
Α	area
$c_p$	specific heat storage capacity
C <sub>W</sub>	specific heat storage capacity of water
d	thickness
df	degree of freedom
div	divergence
е	error
erf	Gauss error function
f	function
F	empirical F-value
$f_i$	relative frequency of object
ġ	mass flow density
grad	gradient
h <sub>i</sub>	absolute frequency of object
MS <sub>W</sub>	mean sum of squares within groups
ġ	heat flow density
Q	heat energy
Rs <sub>e</sub>	external heat transmission resistance
Rs <sub>i</sub>	internal heat transmission resistance
SS <sub>B</sub>	sum of squares between groups
$SS_T$	sum of squares total
SS <sub>W</sub>	sum of squares within groups
t	time
Т	temperature
V	volume
WC	water content
W <sub>he</sub>	weight hexamine
W <sub>t</sub>	weight tannin
x, y, z	spatial dimension

### **Greek symbols**

$\delta_p$	material permeability
Δ	difference
$\delta_{p0}$	permeability of water vapor in air
$\eta^2$	partial eta-squared value
θ	temperature
λ	thermal conductivity
μ	vapor diffusion resistance
$\mu_i$	expected value of distribution i
ν	wave length
ρ	density
σ	standard deviation

### Abbreviations

%	percent
°C	degree Celsius
2D	2-dimesional
3D	3-dimensional
ANCOVA	analysis of covariance
ANOVA	analysis of variance
AR	aspect ratio
ca.	circa
cdf	cumulated density function
cm	centimeter
CNFs	cellulose nano-sized fibers
CR	compressive resistance
СТ	computed tomography
DIN	German institute for norming
e.g.	exempli gratia
emc	equilibrium moisture content
EN	European norm
et al.	et alii
etc.	et cetera
FD	finite differences
FE	finite elements

FIBT	focused ion beam tomography
FSP	fiber saturation point
FTIR	Fourier transform infrared
g	gram
g/cm³	gram per cubic centimeter
h	hour
HDF	high density fiberboard
HVAC	heating, ventilation, air-conditioning
IB	internal bond
ISO	international organization for standardization
J	joule
К	Kelvin
KB-mirror	Kirkpatrick-Baez mirror
kg	kilogram
kJ	kilojoule
I	liter
lim	limes
m	meter
m³	cubic meter
MC	moisture content
MDF	medium density fiberboard
mg	milligramm
ml	milliliter
MOE	modulus of elasticity
MOR	modulus of rupture
ms	millisecond
n	number of test specimens
N/mm²	Newton per square millimeter
NDT	non-destructive testing
OSB	oriented strand board
р	p-value
PB	particleboard
PDE	partial differential equation
рН	potentia Hydrogenii, pH-value
pMDI	polymeric methylene diphenyl diisocyanates
PUR	polyurethane
R²	coefficient of determination

RC	resin content
RH	relative humidity of air
rpm	revolutions per minute
RVE	representative volume element
S	second
SC	solid content
SD	standard deviation
SFT	small flame test
SS	sum of squares
Т	tensile strength
TS	thickness swelling
UF	urea formaldehyde
UV	ultra violet
VOC	volatile organic compound
WA	water absorption
wt	weight
X-ray	X-radiation

# 8 Mathematic appendix

$$f_i = \frac{\sum_{j=1}^{50} h_{i,j}}{\sum_{i=0}^{255} \sum_{j=1}^{50} h_{i,j}}$$
(1)

$$\sum_{k=1}^{3} \sum_{l=l_{k}}^{L_{k}} (g_{kl} - \bar{g})^{2} * h_{kl} = \sum_{k=1}^{3} \sum_{l=l_{k}}^{L_{k}} (g_{kl} - \overline{g_{k}})^{2} * h_{kl} + \sum_{k=1}^{3} (\overline{g_{k}} - \bar{g})^{2} * h_{k}$$
(2)

 $SS_t = SS_w + SS_b$ 

$$MS_{w} = \frac{\sum_{k=1}^{3} \sum_{l=l_{k}}^{L_{k}} (g_{kl} - \overline{g_{k}})^{2} * h_{kl}}{\sum_{k=1}^{3} (h_{k} - 1)} \to min$$
(3)

$$\begin{array}{ll} f_i & \mbox{relative frequency for a gray value } i \mbox{ summed up for all cross section images } j \mbox{ of a sample} \\ h_{i,j} & \mbox{absolute frequency of a gray value } i \mbox{ (ranging from 0 to 255) and cross section image } j \\ SS_t & \mbox{total sum of squares} \\ SS_w & \mbox{sum of squares within classes} \\ SS_b & \mbox{sum of squares between classes} \\ g_{kl} & \mbox{gray value } l \mbox{ (ranging from } l_k \mbox{ to } L_k) \mbox{ in class } k \mbox{ (ranging from 1 to 3)} \\ \bar{g} & \mbox{gray value overall mean} \\ h_{kl} & \mbox{absolute frequency for a gray value } l \mbox{ in class } k \\ \hline g_k & \mbox{gray value mean of class } k \\ \hline g_k & \mbox{gray value class boundaries (for $k = 1: l_k = 0$, $L_k = L_1 - 1$; for $k = 2: l_k = L_1$, $L_k = L_2 - 1$; for $k = 3: l_k = L_2$, $L_k = 255$ \\ h_k & \mbox{absolute frequency of gray values in class $k$ \\ \hline \end{array}$$

 $MS_w$  mean sum of squares within classes

$$\lambda(po,\rho) = \mu + \alpha_{po} + f(\rho) + \varepsilon_{po,\rho} \tag{4}$$

$$\eta_i^2 = \frac{df_i * F_i}{df_i * F_i + df_{error}}$$
(5)

λ	thermal conductivity in W/(m*K)
μ	constant term
ро	particle orientation (vertical, horizontal)
ρ	panel density in kg/m³
$\alpha_{po}$	effect of particle orientation
f( ho)	effect of the covariate panel density
$\varepsilon_{po,\rho}$	random effects that are not controlled in the experiment
$\eta_i^2$	partial eta-squared value for factor i
df <sub>i</sub>	number of degrees of freedom of factor i
F <sub>i</sub>	empirical F-value of factor i
df <sub>error</sub>	number of degrees of freedom for unexplained residual variance

$$\dot{q} = -\lambda * grad(T) \tag{6}$$

$$-\dot{Q} = div(\dot{q}) * dV \tag{7}$$

$$\dot{T} = \frac{\lambda}{\rho * c_p} * div[grad(T)]$$
(8)

$$\frac{dT}{dt} = \frac{\lambda}{\rho * c_p} * \left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2}\right) + \frac{\dot{e}}{\rho * c_p}$$
(9)

$$T(x, y, z, t = t_0) = T_0(x, y, z)$$
(10)

heat flow density in W/m <sup>2</sup>
thermal conductivity in W/(m*K)
gradient of the temperature field T
heat energy in W
divergence of the vectorfield <i>q</i>
small volume in m <sup>3</sup>
temperature in K
density in kg/m³
specific heat storage capacity in J/(kg*K)
time in s
spatial directions
volume-related density of inner heat sources in W/m <sup>3</sup>

$$f'(x_0) = \lim_{h \to 0} \frac{f(x_0 + h) - f(x_0)}{h}$$
(11)

$$f'(x_0) \approx \frac{f(x_0 + \Delta x) - f(x_0)}{\Delta x}$$
(12)

$$\left(\frac{df}{dx} + \frac{df}{dy} + \frac{df}{dz}\right) = \frac{f(x+h, y, z) + f(x-h, y, z)}{h} + \frac{f(x, y+h, z) + f(x, y-h, z)}{h} + \frac{f(x, y, z+h) + f(x, y, z-h)}{h} - \frac{6*f(x, y, z)}{h} + O(h)$$
(13)

$$\begin{split} \dot{Q}_{x,in} &= \left(\frac{\Delta x}{\lambda_{x-1,y,z}} + \frac{\Delta x}{\lambda_{x,y,z}}\right)^{-1} * \Delta y * \Delta z * (T_{x-1,y,z} - T_{x,y,z}) \\ \dot{Q}_{x,out} &= \left(\frac{\Delta x}{2}}{\lambda_{x+1,y,z}} + \frac{\Delta x}{2}}{\lambda_{x,y,z}}\right)^{-1} * \Delta y * \Delta z * (T_{x+1,y,z} - T_{x,y,z}) \\ \dot{Q}_{y,in} &= \left(\frac{\Delta y}{2}}{\lambda_{x,y-1,z}} + \frac{\Delta y}{\lambda_{x,y,z}}\right)^{-1} * \Delta x * \Delta z * (T_{x,y-1,z} - T_{x,y,z}) \\ \dot{Q}_{y,out} &= \left(\frac{\Delta y}{\lambda_{x,y+1,z}} + \frac{\Delta y}{\lambda_{x,y,z}}\right)^{-1} * \Delta x * \Delta z * (T_{x,y+1,z} - T_{x,y,z}) \\ \dot{Q}_{z,in} &= \left(\frac{\Delta z}{\lambda_{x,y,z-1}} + \frac{\Delta z}{\lambda_{x,y,z}}\right)^{-1} * \Delta x * \Delta y * (T_{x,y,z-1} - T_{x,y,z}) \\ \dot{Q}_{z,out} &= \left(\frac{\Delta z}{\lambda_{x,y,z+1}} + \frac{\Delta z}{\lambda_{x,y,z}}\right)^{-1} * \Delta x * \Delta y * (T_{x,y,z+1} - T_{x,y,z}) \end{split}$$

$$\dot{Q}_{x,in} + \dot{Q}_{x,out} + \dot{Q}_{y,in} + \dot{Q}_{y,out} + \dot{Q}_{z,in} + \dot{Q}_{z,out} = 0$$
(15)

ffunction
$$\Delta x, \Delta y, \Delta z$$
increments in spatial directions $O(h)$ error made by the discretization $\dot{Q}$ heat flow in W $\lambda_{x,y,z}$ thermal conductivity at position  $x, y, z$  in W/(m\*K) $T_{x,y,z}$ temperature at position  $x, y, z$  in K

$$\sum_{j=1}^{J} e_j^2 = \sum_{j=1}^{J} [\lambda_j - (\lambda_{1h} * f_{1j} + \lambda_{2h} * f_{2j} + \lambda_{3h} * f_{3j})]^2 \to min$$

$$\sum_{k=1}^{K} e_k^2 = \sum_{k=1}^{K} [\lambda_k - (\lambda_{1v} * f_{1k} + \lambda_{2v} * f_{2k} + \lambda_{3v} * f_{3k})]^2 \to min$$
(16)

- J number of samples with horizontal particles
- *K* number of samples with vertical particles
- $e_{j or k}$ values for residual quantity for sample j (ranging from 1 to J) or k (ranging rom 1 to K) in<br/>W/(m\*K) $\lambda_{j or k}$ thermal conductivity for sample j or k in W/(m\*K)
- $\lambda_{ah}$  thermal conductivity of material phase *a* (ranging from 1 to 3) with horizontal particles

 $f_{aj or k}$  relative frequency of material phase a of sample j or k

$$T(:,:,1) = \begin{pmatrix} T_{1,1,1} & T_{2,1,1} & T_{3,1,1} & \cdots & T_{X,1,1} \\ T_{1,2,1} & T_{2,2,1} & T_{3,2,1} & \cdots & T_{X,2,1} \\ T_{1,3,1} & T_{2,3,1} & T_{3,3,1} & \cdots & T_{X,3,1} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ T_{1,Y,1} & T_{2,Y,1} & T_{3,Y,1} & \cdots & T_{X,Y,1} \end{pmatrix}$$
(17)

for 
$$1 \le x \le X \land 1 \le z \le Z$$
:  $T_{x,1,z} = T_1 \land T_{x,Y,z} = T_2$ 

$$F_{x} = \frac{d_{y} * d_{z}}{d_{x}}, F_{y} = \frac{d_{x} * d_{z}}{d_{y}}, F_{z} = \frac{d_{x} * d_{y}}{d_{z}}$$
(18)

for 
$$1 < y < Y \land 1 < x < X \land 1 < z < Z$$

$$(\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_y + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_y + (\lambda_{x-1,y,z} + \lambda_{x,y,z}) * T_{x-1,y,z} * F_x + (\lambda_{x+1,y,z} + \lambda_{x,y,z}) * T_{x+1,y,z} * F_x + (\lambda_{x,y,z-1} + \lambda_{x,y,z}) * T_{x,y,z-1} * F_z + (\lambda_{x,y,z+1} + \lambda_{x,y,z}) * T_{x,y,z+1} * F_z - [\lambda_{x,y-1,z} * F_y + \lambda_{x,y+1,z} * F_y + \lambda_{x-1,y,z} * F_x + \lambda_{x+1,y,z} * F_x + \lambda_{x,y,z-1} * F_z + \lambda_{x,y,z+1} * F_z + (2 * F_x + 2 * F_y + 2 * F_z) * \lambda_{x,y,z}] * T_{x,y,z} = 0$$
(19)

for  $1 < y < Y \land x = 1 \land z = 1$ 

$$\begin{aligned} (\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_y + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_y + (\lambda_{x+1,y,z} + \lambda_{x,y,z}) * T_{x+1,y,z} * F_x \\ &+ (\lambda_{x,y,z+1} + \lambda_{x,y,z}) * T_{x,y,z+1} * F_z - [\lambda_{x,y-1,z} * F_y + \lambda_{x,y+1,z} * F_y + \lambda_{x+1,y,z} * F_x \\ &+ \lambda_{x,y,z+1} * F_z + (F_x + 2 * F_y + F_z) * \lambda_{x,y,z}] * T_{x,y,z} = 0 \end{aligned}$$

for 
$$1 < y < Y \land x = X \land z = 1$$

$$\begin{aligned} (\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_y + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_y + (\lambda_{x-1,y,z} + \lambda_{x,y,z}) * T_{x-1,y,z} * F_x \\ &+ (\lambda_{x,y,z+1} + \lambda_{x,y,z}) * T_{x,y,z+1} * F_z - [\lambda_{x,y-1,z} * F_y + \lambda_{x,y+1,z} * F_y + \lambda_{x-1,y,z} * F_x \\ &+ \lambda_{x,y,z+1} * F_z + (F_x + 2 * F_y + F_z) * \lambda_{x,y,z}] * T_{x,y,z} = 0 \end{aligned}$$

for  $1 < y < Y \land x = 1 \land z = Z$ 

(20)

 $\begin{aligned} (\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_y + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_y + (\lambda_{x+1,y,z} + \lambda_{x,y,z}) * T_{x+1,y,z} * F_x \\ &+ (\lambda_{x,y,z-1} + \lambda_{x,y,z}) * T_{x,y,z-1} * F_z - [\lambda_{x,y-1,z} * F_y + \lambda_{x,y+1,z} * F_y + \lambda_{x+1,y,z} * F_x \\ &+ \lambda_{x,y,z-1} * F_z + (F_x + 2 * F_y + F_z) * \lambda_{x,y,z}] * T_{x,y,z} = 0 \end{aligned}$ 

for 
$$1 < y < Y \land x = X \land z = Z$$

$$\begin{aligned} (\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_y + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_y + (\lambda_{x-1,y,z} + \lambda_{x,y,z}) * T_{x-1,y,z} * F_x \\ &+ (\lambda_{x,y,z-1} + \lambda_{x,y,z}) * T_{x,y,z-1} * F_z - [\lambda_{x,y-1,z} * F_y + \lambda_{x,y+1,z} * F_y + \lambda_{x-1,y,z} * F_x \\ &+ \lambda_{x,y,z-1} * F_z + (F_x + 2 * F_y + F_z) * \lambda_{x,y,z}] * T_{x,y,z} = 0 \end{aligned}$$

$$\begin{aligned} (\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_{y} + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_{y} + (\lambda_{x-1,y,z} + \lambda_{x,y,z}) * T_{x-1,y,z} * F_{x} \\ &+ (\lambda_{x+1,y,z} + \lambda_{x,y,z}) * T_{x+1,y,z} * F_{x} + (\lambda_{x,y,z+1} + \lambda_{x,y,z}) * T_{x,y,z+1} * F_{z} - [\lambda_{x,y-1,z} * F_{y} \\ &+ \lambda_{x,y+1,z} * F_{y} + \lambda_{x-1,y,z} * F_{x} + \lambda_{x+1,y,z} * F_{x} + \lambda_{x,y,z+1} * F_{z} + (2 * F_{x} + 2 * F_{y} + F_{z}) \\ &* \lambda_{x,y,z}] * T_{x,y,z} = 0 \\ for 1 < y < Y \land 1 < x < X \land z = Z \\ (\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_{y} + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_{y} + (\lambda_{x-1,y,z} + \lambda_{x,y,z}) * T_{x-1,y,z} * F_{x} \\ &+ (\lambda_{x+1,y,z} + \lambda_{x,y,z}) * T_{x+1,y,z} * F_{x} + (\lambda_{x,y,z-1} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_{y} \\ &+ \lambda_{x,y+1,z} * F_{y} + \lambda_{x-1,y,z} * F_{x} + \lambda_{x+1,y,z} * F_{x} + \lambda_{x,y,z-1} * F_{z} - [\lambda_{x,y-1,z} * F_{y} \\ &+ \lambda_{x,y,z}] * T_{x,y,z} = 0 \end{aligned}$$
(21)

for  $1 < y < Y \land 1 < x < X \land z = 1$ 

$$\begin{aligned} (\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_y + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_y + (\lambda_{x+1,y,z} + \lambda_{x,y,z}) * T_{x+1,y,z} * F_x \\ &+ (\lambda_{x,y,z-1} + \lambda_{x,y,z}) * T_{x,y,z-1} * F_z + (\lambda_{x,y,z+1} + \lambda_{x,y,z}) * T_{x,y,z+1} * F_z - [\lambda_{x,y-1,z} * F_y \\ &+ \lambda_{x,y+1,z} * F_y + \lambda_{x+1,y,z} * F_x + \lambda_{x,y,z-1} * F_z + \lambda_{x,y,z+1} * F_z + (F_x + 2 * F_y + 2 * F_z) \\ &+ \lambda_{x,y,z}] * T_{x,y,z} = 0 \end{aligned}$$

for 
$$1 < y < Y \land x = X \land 1 < z < Z$$

$$\begin{aligned} (\lambda_{x,y-1,z} + \lambda_{x,y,z}) * T_{x,y-1,z} * F_y + (\lambda_{x,y+1,z} + \lambda_{x,y,z}) * T_{x,y+1,z} * F_y + (\lambda_{x-1,y,z} + \lambda_{x,y,z}) * T_{x-1,y,z} * F_x \\ &+ (\lambda_{x,y,z-1} + \lambda_{x,y,z}) * T_{x,y,z-1} * F_z + (\lambda_{x,y,z+1} + \lambda_{x,y,z}) * T_{x,y,z+1} * F_z - [\lambda_{x,y-1,z} * F_y \\ &+ \lambda_{x,y+1,z} * F_y + \lambda_{x-1,y,z} * F_x + \lambda_{x,y,z-1} * F_z + \lambda_{x,y,z+1} * F_z + (F_x + 2 * F_y + 2 * F_z) \\ &+ \lambda_{x,y,z}] * T_{x,y,z} = 0 \end{aligned}$$

 $T_{x,y,z}$ temperature for voxel x, y, z in °C $\lambda_{x,y,z}$ thermal conductivity for voxel x, y, z in W/(m\*K)

$$L * Y = R$$

$$Y = L^{-1} * R$$
(22)

$$\dot{q}_{y(or x or z)}(x, y, z) = -\lambda(x, y, z) * \frac{dT}{dy(or \, dx \, or \, dz)}$$
(23)

$$\lambda_{Model} = \frac{\iiint_{0,0,0}^{X,Y,Z} \dot{q}_{y}(x, y, z) * dxdydz}{V} * \frac{d}{\Delta T}$$
(24)

*L* coefficient matrix

*Y* temperature matrix

- *R* matrix containing boundary conditions
- *x*, *y*, *z* index defining position (*X* = length, *Y* = thickness, *Z* = width of sample)
- $\lambda(x, y, z)$  thermal conductivity of the sample at position x, y, z in W/(m\*K)
- $\left(\frac{dT}{dx}, \frac{dT}{dy}, \frac{dT}{dz}\right)$  3D temperature gradient in K/m

$\dot{q}_y(x, y, z)$	heat flow density in y-direction at position $x, y, z$ in W/m <sup>2</sup>
V	volume in m <sup>3</sup>
d	panel thickness in m
$\Delta T$	temperature difference in K
$\lambda_{Model}$	modeled average thermal conductivity for sample in W/(m*K)

$$\dot{q}_{x}(x, y, z) = -\lambda(x, y, z) * \frac{dT}{dx}$$

$$\dot{q}_{y}(x, y, z) = -\lambda(x, y, z) * \frac{dT}{dy}$$

$$\dot{q}_{z}(x, y, z) = -\lambda(x, y, z) * \frac{dT}{dz}$$

$$\alpha_{x,z}(x, y, z) = \tan^{-1}\left(\frac{\sqrt{\dot{q}_{x}(x, y, z)^{2} + \dot{q}_{z}(x, y, z)^{2}}}{|\dot{q}_{y}(x, y, z)|}\right)$$
(26)

 $\dot{q}_x(x, y, z)$  heat flow density vector in x-direction for voxel x, y, z in W/m<sup>2</sup>

 $\dot{q}_y(x, y, z)$  heat flow density vector in y-direction for voxel x, y, z in W/m<sup>2</sup>

 $\dot{q}_z(x, y, z)$  heat flow density vector in *z*-direction for voxel *x*, *y*, *z* in W/m<sup>2</sup>

 $\alpha_{x,z}(x, y, z)$  deviation of heat flow from cross direction (x, z) for voxel x, y, z in degrees

## 9 List of publications

#### **Publications (reviewed)**

- Petutschnigg A, Pferschy U, Katz H, Kain G, Teischinger A (2009) Algorithms to define limits for wood property categorization. Forest Products Journal 59(7/8):75–83.
- Kain G, Barbu MC, Teischinger A, Musso M, Petutschnigg A (2012) Substantial bark use as insulation material. Forest Products Journal 62(6):480–487.
- Kain G, Barbu MC, Hinterreiter S, Richter K, Petutschnigg A (2013) Using bark as heat insulation material. Bioresources 8(3):3718–3731.
- Kain G, Güttler V, Barbu M, Petutschnigg A, Richter K, Tondi G (2014) Density related properties of bark insulation boards bonded with tannin hexamine resin. European Journal of Wood and Wood Products 72(4):417–424. doi: 10.1007/s00107-014-0798-4
- Kain G, Güttler V, Lienbacher B, Barbu MC, Petutschnigg A, Richter K, Tondi G (2015) Effects of different flavonoid extracts in optimizing tannin-glued bark insulation boards. Wood and Fiber Science 47(3):258–269.
- Kain G, Charwat-Pessler J, Barbu M, Plank B, Richter K, Petutschnigg A (2016) Analyzing wood bark insulation board structure using X-ray computed tomography and modeling its thermal conductivity by means of finite difference method. Journal of Composite Materials 50(6):795–806. doi: 10.1177/0021998315581511
- Kain G, Lienbacher B, Barbu MC, Plank B, Richter K, Petutschnigg A (2016b) Evaluation of relationships between particle orientation and thermal conductivity in bark insulation board by means of CT and discrete modeling. Case Studies in Nondestructive Testing and Evaluation 6:21–29.

### **Publications (non-reviewed)**

- Kain G, Teischinger A, Musso M, Barbu MC, Petutschnigg A (2012) Stoffliche Rindennutzung in Form von Dämmstoffen. Holztechnologie 53(4):31–37.
- Kain G, Heinzmann B, Barbu MC, Petutschnigg A (2013) Softwood bark for modern composites. Pro Ligno 9(4):460-468.
- Kain G (2013) Dämmstoffe aus Baumrinden. Akademiker Verlag, Saarbrücken, 138 p.
- Kain G, Barbu MC, Richter K, Plank B, Tondi G, Petutschnigg A (2015) Use of tree bark as insulation material. Forest Products Journal 65(3/4):S16-S25.
## **Conference presentations and others**

- Kain G, Barbu MC, Petutschnigg A (2012) Dämmplatten aus Baumrinden. Patent (Austria), 8/512707.
- Kain G, Barbu MC, Richter K, Petutschnigg A, Hinterreiter S, Charwat-Pessler J, Plank B (2013) Bark insulation – use of tree bark as insulation material. 10<sup>th</sup> Werkstoffkongress, Leoben/Styria, November 6-7 (oral presentation and conference paper).
- Kain G, Barbu MC, Richter K, Plank B, Tondi G, Petutschnigg A (2014) Use of tree bark as insulation material. 3<sup>rd</sup> International Conference on Processing Technologies for the Forest and Bio-based Products Industries, Kuchl/Salzburg, September 24-26 (oral presentation and conference paper).
- Kain G, Barbu MC, Richter K, Plank B, Tondi G, Petutschnigg A (2015) Suitability of tree barks for thermal insulation applications. 1<sup>st</sup> Holzwissenschaftliches Seminar SS 2015, TU Munich, April 24 (oral presentation).
- Kain G, Barbu MC, Richter K, Petutschnigg A (2015) Baustoffe mit Zukunft Innovative, regionale und nachwachsende Baustoffe am Beispiel der Baumrinde. Ö1 Hörsaal TU Graz, Graz/Styria, Mai 7 (oral presentation).
- Kain G (2015) Regionale Baumaterialien und ihre Bedeutung für nachhaltiges Bauen. Ö1 Hörsaal Universität Innsbruck, Innsbruck/Tyrol, Mai 28 (oral presentation).
- Kain G, Barbu MC, Petutschnigg A (2015) Insulation material made from tree bark. 14<sup>th</sup> World Forestry Congress, Durban, South Africa, September 7–11 (conference paper).
- Kain G, Plank B, Lienbacher B, Barbu MC, Richter K, Petutschnigg A (2016) Evaluation of relationships between particle orientation and thermal conductivity in bark insulation boards by means of CT and discrete modeling. 6<sup>th</sup> Conference on Industrial Computed Tomography, University of Applied Sciences Upper Austria, Wels, February 9-12 (oral presentation and conference paper).

# 10 Thesis-publications

The following publications represent the core piece of this dissertation.

### **Publication 1**

Published in the Forest Products Journal; Günther Kain, Marius-Catalin Barbu, Alfred Teischinger, Maurizio Musso, and Alexander Petutschnigg; Substantial bark use as insulation material, 2012, 62(6), pp 480–487.

The original can be accessed with doi: http://dx.doi.org/10.13073/FPJ-D-12-00052.1.

#### **Publication 2**

Published in Bioresources; Kain Günther, Marius-Catalin Barbu, Stefan Hinterreiter, Klaus Richter, and Alexander Petutschnigg; Using bark as heat insulation material, 2013, 8(3), pp 3718–3731.

The original can be accessed at http://www.ncsu.edu/bioresources/BioRes\_08/BioRes\_08\_3\_3718 \_Kain\_Bark\_Heat\_Insulation\_Material\_3807.pdf (open access).

#### **Publication 3**

Published in the European Journal of Wood and Wood Products; Günther Kain, Viola Güttler, Marius-Catalin Barbu, Alexander Petutschnigg, Klaus Richter, and Gianluca Tondi; Density related properties of bark insulation boards bonded with tannin hexamine resin, 2014, 72(4), pp 417–424.

The original can be accessed with doi: http://dx.doi.org/10.1007/s00107-014-0798-4.

#### **Publication 4**

Published in the Journal of Composite Materials; Günther Kain, Johann Charwat-Pessler, Marius-Catalin Barbu, Bernhard Plank, Klaus Richter, and Alexander Petutschnigg; Analyzing wood bark insulation board structure using X-ray computed tomography and modeling its thermal conductivity by means of finite difference method, 2016, 50(6), pp 795–806.

The original can be accessed with doi: http://dx.doi.org/10.1177/0021998315581511.

### **Publication 5**

Published in Wood and Fiber Science; Günther Kain, Viola Güttler, Bernhard Lienbacher, Marius-Catalin Barbu, Alexander Petutschnigg, Klaus Richter, and Gianluca Tondi; Effects of different flavonoid extracts in optimizing tannin-glued bark insulation boards, 2015, 47(3), pp 1–12.

The original can be accessed at http://www.swst.org/publications/wfs/preprints/47(3)/WFS1812.pdf.

### **Publication 6**

Published in Case Studies in Nondestructive Testing and Evaluation; Günther Kain, Bernhard Lienbacher, Marius-Catalin Barbu, Bernhard Plank, Klaus Richter, and Alexander Petutschnigg; Evaluation of relationships between particle orientation and thermal conductivity in bark insulation board by means of CT and discrete modeling, 2016, 6, pp 21–29.

The original can be accessed with doi: http://dx.doi.org/10.1016/j.csndt.2016.03.002 (open access).

# Eidesstattliche Erklärung

Ich erkläre an Eides statt, dass ich die bei der promotionsführenden Einrichtung Wissenschaftszentrum Weihenstephan für Ernährung, Landnutzung und Umwelt der Technischen Universität München zur Promotionsprüfung vorgelegte Arbeit mit dem Titel

## Design of Tree Bark Insulation Boards: Analysis of Material, Structure and Property Relationships

am Lehrstuhl für Holzwissenschaft unter der Anleitung und Betreuung durch Univ.-Prof. Dr. rer. nat. Klaus Richter ohne sonstige Hilfe erstellt und bei der Abfassung nur die gemäß § 6 Abs. 6 und 7 Satz 2 angegebenen Hilfsmittel benutzt habe.

Ich habe keine Organisation eingeschaltet, die gegen Entgelt Betreuerinnen und Betreuer für die Anfertigung von Dissertationen sucht, oder die mir obliegenden Pflichten hinsichtlich der Prüfungsleistungen für mich ganz oder teilweise erledigt.

Ich habe die Dissertation in dieser oder ähnlicher Form in keinem anderen Prüfungsverfahren als Prüfungsleistung vorgelegt.

Ich habe den angestrebten Doktorgrad noch nicht erworben und bin nicht in einem früheren Promotionsverfahren für den angestrebten Doktorgrad endgültig gescheitert.

Die öffentlich zugängliche Promotionsordnung der TUM ist mir bekannt, insbesondere habe ich die Bedeutung von § 28 (Nichtigkeit der Promotion) und § 29 (Entzug des Doktorgrades) zur Kenntnis genommen.

Ich bin mir der Konsequenzen einer falschen eidesstattlichen Erklärung bewusst.

Mit der Aufnahme meiner personenbezogenen Daten in die Alumni-Datei bei der TUM bin ich einverstanden.

Salzburg, den 14.04.2016

(Günther Kain)

## **Curriculum vitae**

## Günther Kain

Born September 6, 1984, in Bad Ischl, Austria

- 2012 2016 Doctorate studies at the Holzforschung München (Wood Research Munich), Technical University of Munich: "Design of tree bark insulation boards: Analysis of material, structure and property relationships", under the supervision of Professor Dr. Klaus Richter (Technical University of Munich), Professor Dr. Alexander Petutschnigg (Salzburg University of Applied Sciences, BOKU Vienna), and Professor Dr.-Ing. Dr. Marius Catalin Barbu (Salzburg University of Applied Sciences, University "Transilvania" Brasov).
- 2014 Examination for the master craftsman's diploma for carpentry (Guild of Carpentry, Salzburg).
- 2013 Qualifying examination for consulting engineers in the sectors interior design, wood technology, and timber industry (Economic Chamber of Trade, Commerce and Industry, Salzburg).
- 2011 up to now Lecturer at the Salzburg University of Applied Sciences with focus on wood technology and material development at Campus Kuchl.
- 2010 2012 Master studies in Forest Products Technology & Management with focus on product development, Salzburg University of Applied Sciences, Kuchl.
- 2009 up to now Technical teacher at the Higher Technical College Hallstatt in the field of wood technology and interior design.
- 2009 2010 Seminar for engineering pedagogics (College of Education, Linz).
- 2005 2009 Diploma studies in Forest Products Technology and Timber Construction, Salzburg University of Applied Sciences, Kuchl.
- 2004 2005 Civilian service (Old people's home, Bad Goisern).
- 1999 2004Studies and matriculation examination with focus on interior design and<br/>furniture building, Higher Technical College Hallstatt.