Hot-Air/Hot-Steam Process for the Production of Laccase-Mediator-System Bound Wood Fiber Insulation Boards

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In this study, a new technical process for hardening wood fiber insulation boards is introduced. During the dry-process, the fibers are usually glued with polymeric-diphenylmethane-diisocyanate (pMDI) and hardened to wood fiber insulation boards using a steam-air mixture. However, the maximum temperature reached in the steam-air process was 100 °C, and it was impossible to use an alternative binding agent for the gluing of the wood fiber insulation boards other than pMDI. When incubated with laccase-mediator-system (LMS) as a naturally based bonding system, temperatures of over 120 °C are required because of the chemical wood composition, especially the lignin. In this case, the hot-air/hot-steam process offers new technical opportunities for realizing temperatures above 100 °C. In this study, wood fiber insulation boards were glued with LMS, *vs.* reference boards with inactivated LMS, laccase alone, and 4% pMDI. Then, the boards were hardened using one of three processes: with steam-air mixture, with hot-air, and with hot-air/hot-steam. Through the hot-air/hot-steam process, temperatures of well over 120 °C were attainable. All the insulation boards hardened using the hot-air/hot-steam process showed better physical and technical properties than those hardened with steam-air mixture or hot-air alone. The reason for this is a sudden increase of temperature after the adding of steam because high temperatures insure that the LMS activated wood fiber surface lignins are completely plasticized. As a result the physical-technological properties such as internal bond strength, compression strength, and short term water absorption of insulation boards treated with LMS were comparable to those boards treated with 4% pMDI.

Keywords: Hot-steam/hot-air process; Wood insulation boards; Laccase-mediator-system; pMDI

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INTRODUCTION

In the production of wood based panels, large quantities of petrochemical binding agents, such as urea formaldehyde or phenol formaldehyde resins, are required (González-Garciá *et al.* 2011). Besides the dependency on crude oil, harmful formaldehyde is released during the production process as well as out of the products, and this adversely affects ecosystem quality (Imam *et al.* 1999; US. EPA 2002). Possible solutions are seen in the reduction of those binder systems as well as the replacement by more environmentally-friendly, natural binders (González-Garciá *et al.* 2011). Equally, the recycling process of naturally bonded wood products is more efficient (Euring 2008).

In the market of insulation materials, there has been a recent increase in the use of renewable raw materials as insulation in house walls, ceilings, flooring, and roofs due to the Energy Saving Ordinance 2012 in Europe (Brandhorst 2012). In Germany, Switzerland, and Austria the total production volume of all insulation materials is estimated around 35 Mio m³ per year (Michel *et al.* 2013). The German market for wood fiber insulation materials has risen continuously for 11.9% each year during the last decade (Brombacher 2014). Nonetheless, the proportion of renewable resources on the market is quite low (4%), and most insulation materials are based on petrochemical or mineral components (Greiff 2009, Michel *et al.* 2013). An important advantage of insulation materials from renewable resources lies in the conservation of finite resources, such as raw oil and natural gas (Kleinhempel 2005). Along with the insulation factor, a certain level of sound insulation should be taken into account as well.

Wood as a renewable resource can be used to manufacture wood fiber insulation boards, which are finished traditionally using the wet-process, or (as in more recent years) using the dry-process. Current market developments and trends show a clear tendency towards the dry-process (Makas 2012). The advantages of the dry-process, compared to the wet process, lie in the possibility of producing a lower raw density range of 80 to 200 kg/m³, with thicknesses ranging from 20 to 300 mm. Furthermore, the dry-process results in more favorable energy consumption. Because of the technical and economic restrictions involved in using the wet-process, the boards are restricted to a thickness of 20 mm (Wagenführ and Scholz 2012). The disadvantages of the wet-process include high water consumption and long drying time per boards (6 min per mm). Compared to the dry-process, the energy expenditure for the wet-process is approximately 30% higher per ton of wood fiber insulation board (Lempfer 2004).

The dry-process is characterized as the process in which drying and gluing with polymeric-diphenylmethane-diisocyanate (pMDI) immediately follows pulping. After the mat formation, the pMDI is hardened onto the wood fibers to produce insulation boards using the flow of a steam-air mixture. Because of physical restrictions, relatively low temperatures of 90 to 100 °C are involved. Further drying processes or post-conditioning are no longer required (Gutex 2014).

In the industrial production of wood fiber insulation boards using dry-process, only binders such as isocyanate glues in pMDI systems, prepolymers, and MDI are used (Wagenführ and Scholz 2012). Typically, the glue application is approximately 4 to 6%. During the production process, pMDI can enter the human body in the form of a dust or spray *via* the eyes or respiratory system, and may react with the mucus membranes or with human proteins. Polymeric-diphenylmethane-diisocyanate is therefore classified as a hazardous chemical agent, and requires high occupational safety measures (Dunky and Niemz 2002). In addition, the cost of pMDI is high, due to the complex synthesis process and the unstable crude-oil price (Dunky and Niemz 2002; Türk 2014).

Natural binders based on renewable resources, such as lignin, proteins, casein, glutens, starches, or tannins can be considered as possible substitutes (Niemz 1993; Kharazipour 1996). These renewable resources have increased in importance in recent years due to their manifold application possibilities. For example, the lignin on the surface of thermo-mechanical pulping (TMP) wood fibers can be activated through laccase-mediator-system (LMS) (Euring 2008, 2011, 2012, 2013). The reaction mechanism involves the laccase-mediator-system activating the surface lignin of the wood fibers through oxidation (Kües *et al.* 2007; Kudanga *et al.* 2008; Widsten and Kandelbauer 2008; Nyanhongo *et al.* 2010). As a result, free lignin fractions are produced, which, when the fibers are hot pressed, polymerize together to form wood fiber composites, such as medium density fiberboards (MDF) or high density fiberboards (HDF). During pressing, all fibers reach a temperature of at least 110 to 160 °C because within this temperature range lies the glass transition temperature of lignin (Nada *et al.* 2002; Bouajila *et al.* 2005). The surface lignin of the wood fiber plasticizes, and after cooling affects the fiber-to-fiber bonds which harden to produce wood fiber materials. The use of a catalyst mediator in LMS accelerates and intensifies the plasticization, causing a stronger fiber binding (Euring 2008; Euring *et al.* 2008; Müller *et al.* 2009; Kharazipour and Euring 2011; Euring *et al.* 2011; Kharazipour and Euring 2014).

With the development of LMS bound insulation materials, the hot-air/hot-steam process was developed (Kharazipour and Euring 2013). The most important innovation in this process is that the wood fiber mat, which is to be hardened into insulation boards, is not treated with a steam-air mix flow with a maximum temperature of 100 °C. Instead, it is first treated with hot-air, then with hot-steam, so that the temperatures of well over 100 °C, which are required in the hardening of lignin fragments, are available. This technology has made it possible, for the first time in the production of wood fiber insulation boards, to use binding agents other than pMDI.

Apart from the LMS discussed in this publication, many other binding agents, natural (*e.g.*, wheat proteins, starch) or petrochemical based (*e.g.*, urea-formaldehyde resin, phenol-formaldehyde resin) or both in combination can be used, which harden at temperatures over 100 °C and offer a more cost-effective alternative to pMDI. Using LMS offers not only the advantage of independence from fossil fuel resources, but also limits the production of any extra emissions (either formaldehyde or volatile organic compounds). Likewise, the production of toxic degradation products from synthetic bonding agents can be ruled out, so that the use and subsequent recycling of the product can be considered harmless (Euring 2008).

This publication will introduce the first investigations of LMS-bound wood fiber insulation boards using hot-air/hot-steam process.

EXPERIMENTAL

Materials

Wood fibers (20 kg absolutely dry fibers per batch)

Spruce lumber (containing 100% *Picea abies* wood) was defibrated into fibers using the thermo-mechanical-pulping process (TMP), by PAVATEX SA in Cham, Switzerland.

Laccase (100 U laccase/g wood fibers)

Commercial Novozym 51003 *Trametes villosa* laccase, recombinantly produced in Aspergillus oryzae by Novozymes (Bagsveard, Denmark), was used. Its specific activity was routinely measured by monitoring the oxidation of di-ammonium salt of 2,2´-azinobis-3-ethylbenzothiazoline-6-sulfonic acid (ABTS) (Matsumura *et al.* 1986). The activity of the laccase stock was about 1100 U/g.

Mediator (10 mM/g absolutely dry wood fibers)

4-hydroxy-benzoic-acid (HBA) with a purity of 99% was purchased from Alfa Aesar (Ward Hill, USA).

Buffer (pH 6)

As reaction buffer for the LMS, McIlvain-buffer (pH 6.0) made up of 0.2 M dipotassium hydrogen phosphate (K_2HPO_4) and 0.1 M citric acid ($C_6H_8O_7$) (AppliChem; Darmstadt, Germany) was used in all experiments.

pMDI (4% pMDI per 20 kg absolutely dry wood fibers)

Commercial pMDI I-Bond WFI 4370 from Huntsman (Rotterdamm, Netherlands), with a minimum MDI of 60% (methylene diphenyl diisocyanate) and a minimum standard of 13% polyole, was used for reference insulation boards.

Methods

Temperature gradient measurements

Temperature measurements were taken over a 110-s time span during the production of the wood fiber insulation boards using the hot-air or the hot-air/hot-steam process. Measurements were taken on both the top and bottom surfaces, as well as from the middle of the board. In the process of steam-air mix alone, the temperatures were only measured over a 60-s time period because it was not possible for the boards to be heated to temperatures of more than 100 °C using this method. Also, the boards were be increasingly moistened, which can lead to quality reduction. The measurements were taken using a Greisinger GMH 3250 precision quick response thermometer (Germany).

Production of the wood fiber insulation boards

The wood fiber isolation boards were produced at the Institute's own pilot plant for fiberboards (BINOS; Springe, Germany), and the hot-air/hot-steam unit was self-constructed with parts from Leister Technologies (Aachen, Germany) and MG Dampftechnik (Bedburg, Germany). The fiberboard plant consists of the following units: a horizontal blender system with three injectors for spraying binders onto fibers, a tube dryer (into which the blended fibers are conveyed), a cyclone from which the fibers are strewed to a conveyer, a bunker into which the fibers are moved, and a form conveyer onto which the fibers are pre-shaped into a thick fiber mat. The pre-shaped mat is finally moved into the hot-air/hot-steam unit.

As this unit represents a new technology, the scheme is shown in Fig. 1.

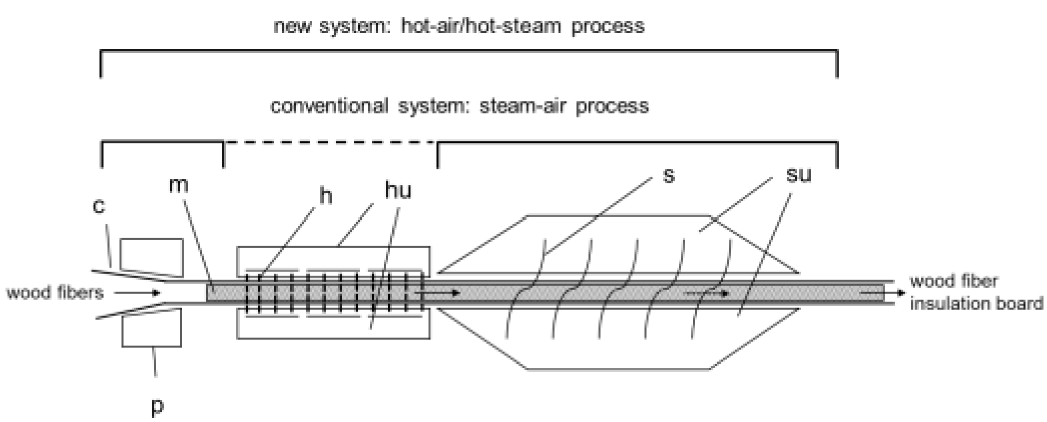

Fig. 1. Scheme of the new hot-air/hot-steam process. Wood fibers are treated with LMS, laccase, or pMDI, then transferred into the hot-air/hot-steam unit. Components or materials: c = conveyor, p = pre-press, m = fibermat, h = hot-air, hu = hot-air unit, s = hot-steam, su = steam unit

For each treatment, 20 kg of absolutely dry wood fibers were incubated at room temperature by spraying on 1.5 L/min of a total of 10-L buffer solution, which contained 100 U laccase/g wood fibers with or without 10 mM/g wood fibers of the mediator HBA onto 1.5 kg/min of wood fibers. After blending, the fibers were moved directly into the tube dryer, using warm air with a temperature of 100 °C, to reduce the fiber moisture content to approximately 8%. Conveyed through the fiber bunker, the dried fibers were directly strewed onto the form conveyer to be pre-pressed into a one-layer mat. The handling procedure of blending, drying, and mat generating lasted about 30 min.

Once the pre-shaped fiber mat was transferred through a pre-press (no heat) to form the required board thickness of 40 mm and raw density of 200 kg/m^3, the mat was conveyed into the hot-air unit, and treated for 80 s. In the hot-air unit, the controlled hot-air flow had an initial temperature of about 140 °C and a flow rate of 15 m/s. Afterwards, the fiber mat was conveyed into the hot-steam unit and vaporized for another 10 s with hot-steam (100 °C). The temperature was measured for another 30 s, without hot-air or hot-steam, for recalibration.

To compare the individual technical processes, insulation boards were prepared using the same formula. The boards were either treated with only steam-air mix (100 °C, 30 s flow and 30 s recalibration) or with only hot-air (140 °C, 15 m/s, 90 s flow, and 30 s recalibration).

For all treatments, wood fibers were parallel blended with heat-inactivated LMS (cooked for 10 min), 100 U laccase, and 4% pMDI. The pMDI I-bond WFI 4370 from Huntsman is a standard binding agent used for wood fiber insulation boards, and is usually dosed with a concentration of 4% to 6% on absolutely dry fiber. The 4% pMDI used in this study was sprayed through the same spray attachment used to distribute the laccase and LMS. Each treatment was replicated six times.

The testing of physical-technological properties was conducted for the internal bond strength of the boards, in accordance with EN 1607 (2013). The compression strength was studied according to EN 826 (2013), and for short time, water absorption after 4 h was in accordance with EN 1609 (2013). Thermal conductivity tests were carried out according to EN 12667 (2001).

RESULTS AND DISCUSSION

Temperature Gradient

For reasons of clarity, the chart (Fig. 2) shows only the temperature gradient from the middle of the boards treated with the varying processes and binding agents.

The temperature gradients for the measurements, taken from both the top and bottom surface of the boards, varied only marginally in comparison to the measurements taken from the middle of the board, by an average of ± 5 °C. The advantage of the wood fiber insulation boards over wood composites (*e.g.*, medium-density fiberboards) is that their lower density creates a more uniform material, and faster warming can be achieved. It can be assumed from the measured temperature gradients that the steam-air mixture process alone achieves only temperatures of 100 °C in the middle of the board. In this instance, after 10 s of the steam-air process, temperatures of almost 100 °C were reached in the fiber mat. However, the temperature measurements had to be halted after 60 s because the condensing steam created an increase in the moisture content of the wood fiber insulation boards. A decrease in temperature corresponded with the spike in moisture content as well. Because of the design of conventional steam-air technology, temperatures of over 100 °C could not be reached. However, the situation differs when using a hot-air warming process to harden insulation boards. To date, airflow press dryers have been used only for the production of flexible insulation materials comprised of bi-component fibers (natural fibers and polyethylene fibers). The binding component is the plastic based fiber, which melts at temperatures above 120 °C in the fiber mat, and binds with itself and with the natural fibers, to form an insulating material.

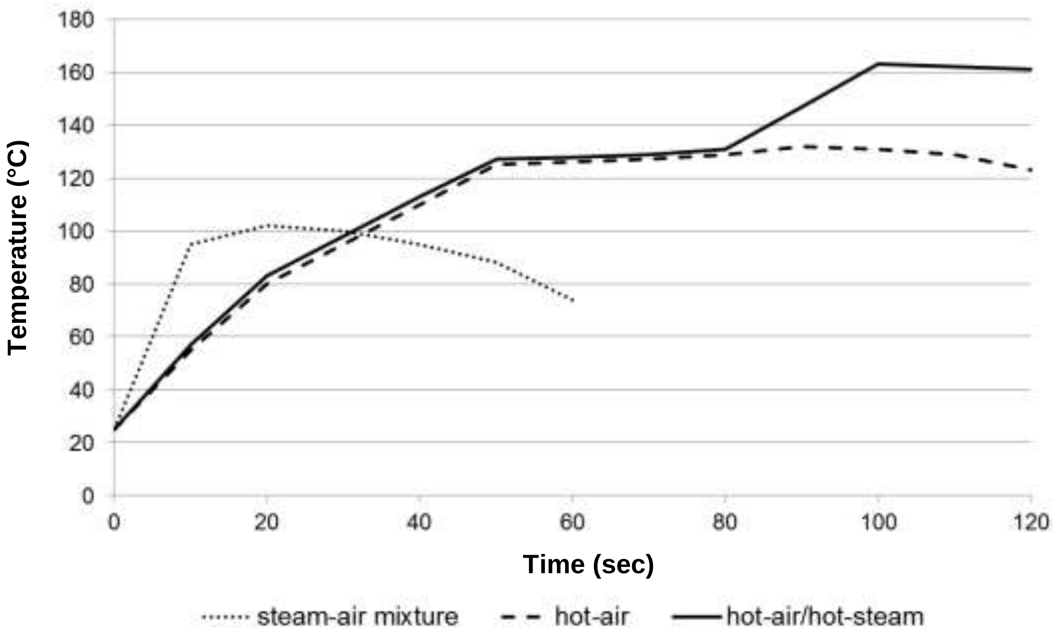

Fig. 2. Temperature gradient within the insulation boards during treatment with steam-air mixture, hot-air, or hot-air/hot-steam

Whereas it is possible to reach temperatures above 120 °C using the hot-air process to insulate material production, longer process times are required, which increases the danger of over drying and combustion of the fibers. The temperature gradient shows a temperature increase, with the peak temperature registered at over 120 °C after 50 s. After 120 s, the temperature was still under the chosen level of 140 °C, which can be attributed to the existing and evaporating moisture levels. The hot-air/hot-steam method, discussed in this paper, combines both processes, in that the insulation boards are first treated for a defined period of time, with hot-air process followed by a hot-steam treatment. As can be seen in the diagram above, after 80 s of hot-air exposure in the hot-air section and 10 s of hot-steam treatment in the hot-steam section, a sudden increase in the temperature can be observed. After a span of 100 s, the temperature reached 160 °C despite the interruption of the hot-steam flow.

This sudden increase can be interpreted as the decisive chemical-physical process for the ultimate hardening of the insulation boards after being treated with LMS and other natural binding agents (Kharazipour and Euring 2013). The high temperatures ensure that the LMS activated wood fiber surface lignins are completely plasticized, causing the insulation boards to harden completely (see Introduction).

Currently the use of the hot-air/hot-steam process takes twice the amount of time in comparison to using the conventional steam-air process. To reduce the time, the hot-air/hot-steam unit has to be optimized for a better performance.

Internal Bond Strength

It is apparent, when looking at Fig. 3 that the highest internal bond strength for all varieties of boards occurs after the insulation boards hardened were by the hot-air/hot-steam process.

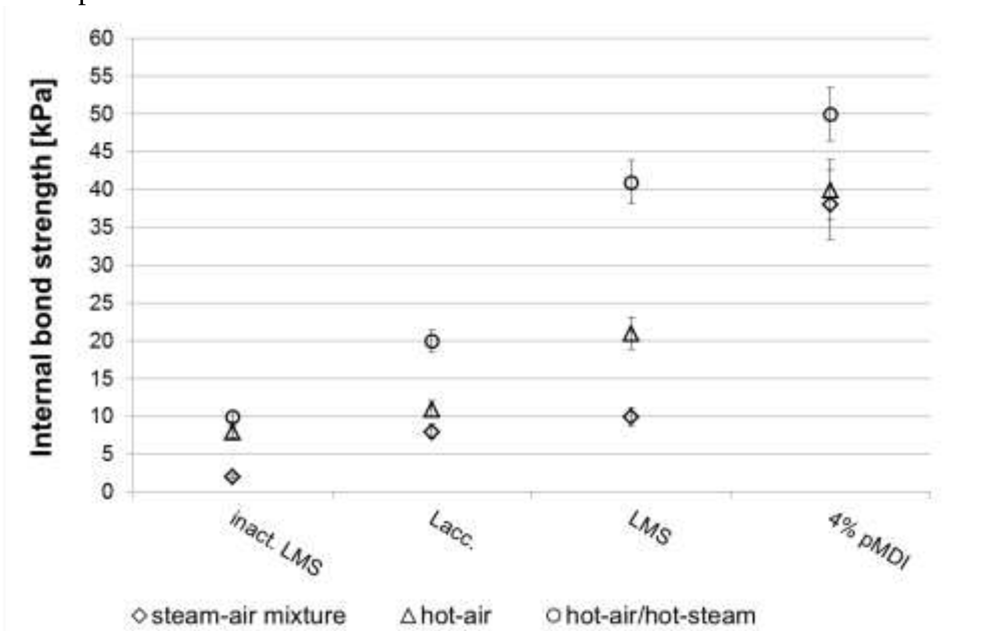

Fig. 3. Internal bond strengths of differently treated insulation boards. The types of wood fiber treatments were the steam-air mixture, hot-air, or hot-air/hot-steam: inact. LMS = inactivated laccase-mediator-system in buffer; Lacc. = laccase in buffer; LMS = laccase-mediator-system with mediator HBA in buffer; 4% pMDI = solely pMDI-glue (4%). Data are presented as mean ± SD

Worthy of emphasis are both the LMS insulation boards and the pMDI insulation boards, which achieved internal bond strength values ranging from 40 to 50 kPa. These values reflect those normally attained in 40-mm thick, industrially produced, wood fiber insulation boards, which typically possess a density of 200 kg/m^3. The difference in mechanical strength can be noteworthy depending on which hardening process is used on the standard 4% pMDI glued boards. The mean values for the mechanical strengths were 38 kPa when using steam-air hardening and 40 kPa when using the hot-air process. The internal bond strength for the hot-air/hot-steam treatment was 50 kPa.

Therefore, this new process provides an opportunity to improve the production of wood fiber insulation boards. The largest potential for the hot-air/hot-steam process lies in the use of natural binders, such as LMS. Different LMSs have already been tested successfully for the production of medium density fiberboards (MDF) in pilot scale and fulfilled the required EN-Norms for internal bond strength around 0.7 N/mm² (Euring *et al.* 2011, 2013). In the case of wood insulation boards, internal bond strengths of 10 kPa after hardening with steam-air mixture and 22 kPa with hot-air process are not adequate for this type of insulation boards in comparison to 4% pMDI bonded wood fibers. During the hot-air/hot-steam process, high enough temperatures in the middle of the board are reached, which allow the LMS-activated wood fiber surface lignin to react adequately (see Introduction). The internal bond strengths of the LMS variety are high, correspondingly. Although the air temperature during the hot-air process reached 140 °C, the temperature in the middle of the board merely reached 130 °C (see Results and Discussion, Temperature gradient). These conditions are not adequate to facilitate the binding of the wood fibers with one another. Possible reasons for the inadequate conditions could include over drying in the fibers and/or inadequate plasticizing of the wood fiber surface lignin. Only the insulation boards subjected to the laccase treatment hardened better with the hot-air/hot-steam process (20 kPa) than with hot-air (12 kPa) or steam-air mixture (8 kPa). However, the mediator, which supports the laccase, was lacking in these boards. The inactivated LMS hardly affects the bonding of the insulation boards, which is reflected in the low internal bond strengths.

Compression Strength

The determination of the compression strength is an important parameter for wood fiber insulation boards. It indicates the load bearing capacity of the material, and is important for building construction as well. Forty-millimeter-thick wood fiber insulation boards, with a density of 200 kg/m³ and 4% pMDI, usually have compression strengths of over 200 kPa.

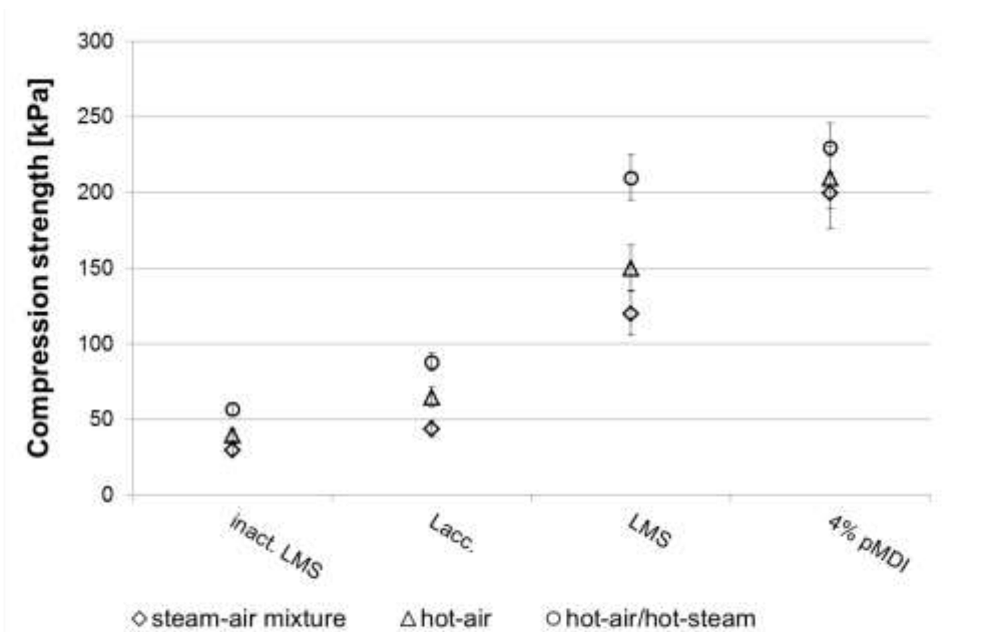

Fig. 4. Compression strengths of differently treated insulation boards. Treatment of wood fibers either with steam-air mixture, hot-air, or hot-air/hot-steam: inact. LMS = inactivated laccase-mediator-system in buffer; Lacc. = laccase in buffer; LMS = laccase-mediator-system with mediator HBA in buffer; 4% pMDI = solely pMDI-glue (4%). Data are presented as mean ± SD

As can be seen in Fig. 4, the insulation boards with the 4% pMDI binder achieved compressive strengths of 200 kPa and 230 kPa in both the steam-air mixture and hot-air/hot-steam process, respectively. This is the advantage of pMDI alone for the steam-air mixture as the values are representative for the range of products of the wood insulation board producers (Siempelkamp 2012; Gutex 2014). Furthermore, with hot air alone, the value is 210 kPa.

As with the internal bond strength, it becomes obvious that the insulation boards with the LMS benefitted from the hot-air/hot-steam process. They could compete favorably with the standard boards, with the compression strength values of 210 kPa. After hardening process with the hot-air (150 kPa) and steam-air mixture (120 kPa), the boards were lacking in strength. Compression strength values for boards with non-activated LMS or laccase were very low and did not meet current expectations for wood fiber insulation boards.

Short Term Water Absorption after 4 h

Analogous to the results of the internal bond strength tests and the compressive strength tests is also the advantage of the hot-air/hot-steam process, reflected in the short-term water absorption results.

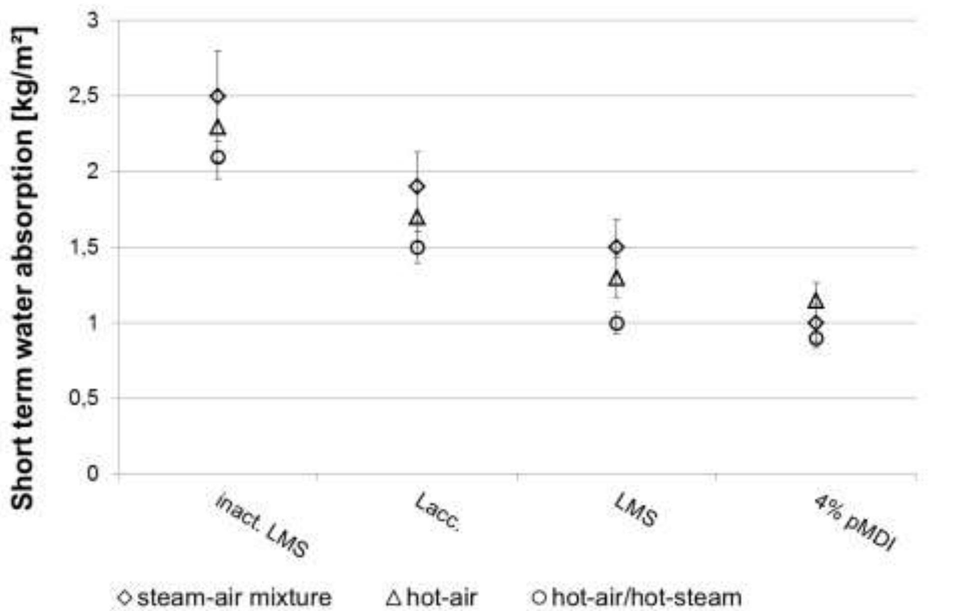

Fig. 5. Short term water absorptions of differently treated insulation boards. Treatment of wood fibers either with steam-air mixture, hot-arm, or hot-air/hot-steam: inact. LMS = inactivated laccase-mediator-system in buffer; Lacc. = laccase in buffer; LMS = laccase-mediator-system with mediator HBA in buffer; and 4% pMDI = solely pMDI-glue (4%). Data are presented as mean ± SD

In all samples, the water absorption level was favorably lower in comparison to using just the steam-air mix or hot-air hardening alone (Fig. 5). However, for the insulation boards treated with 4% pMDI, there was barely a difference between 0.9 and 1.1 kg/m², considering this is a dimension for standard insulation boards (Gutex 2014). Applying hot-air/hot-steam resulted in lower water absorption due to higher internal bond and compression strengths. With LMS, the water absorption for the hot-air/hot-steam process differs significantly for 1 kg/m², as well as in the other processes by 1.2 to 1.5 kg/m². Positive effects of LMS resulting in lower water absorptions and thickness swellings of MDF have been reported in Euring *et al.* (2008, 2011, 2013). The control samples with non-activated LMS and laccase absorbed too much water by comparison.

Thermal Conductivity

Measuring the thermal conductivity of all insulation boards revealed an average thermal conductivity value of 0.041 ± 0.001 λ (W/mK). The reason for this is that the thermal conductivity value is generally not influenced by the binder system but mainly by the raw density of the materials (Foglia *et al.* 2006). Due to the same raw density of 200 kg/m³, no difference of the thermal conductivity was observed within the tested insulation boards.

CONCLUSIONS

1. The hot-air/hot-steam process provides clear advantages in the production of wood fiber insulation boards. In comparison to the conventional production process of preparing insulation boards using steam-air mixture, pMDI, a binding agent, is no longer necessary in the new process.
2. The LMS treated fibers can be completely hardened through the hot-air/hot-steam process, and the resulting chemical-physical temperature rises. The mechanical and physical properties of these boards approximate those of conventionally manufactured wood fiber insulation boards.
3. Nevertheless, further research is required to optimize the hot-air/hot-steam process, especially reducing the processing time.

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