10th International Conference on Composite Science and Technology
ICCST/10
A.L. Araújo, J.R. Correia, C.M. Mota Soares, et al. (Editors)

SANDWICH BOARDS MADE FROM BIO-POLYURETHANE FOAM AND NATURAL FIBRE COVER LAYERS: NEW APPROACH FOR SUSTAINABLE LIGHTWEIGHT CONSTRUCTION.

Holger Fischer[*†], Nils Emde[*], Mansour Mirzaghavam[*]

[*]Faserinstitut Bremen e.V. -FIBRE-
Am Biologischen Garten 2; D-28359 Bremen

[†]corresponding author
fischer@faserinstitut.de

Key words: natural fibre; polyurethane; mechanical characteristics; bio-based composite; sandwich construction.

Summary: *a sustainable sandwich construction was designed consisting of bio-based PU equipped with natural fibre semi-finished products as cover layers. This new approach enables the production of materials, which are lighter and less expensive than the currently used conventional materials.*

This paper reports the initial assessment of fibre-matrix interaction, the influence of different cover layer constructions (linen fabrics, felts, laid scrim etc.) and/or core reinforcement by hemp fleeces. Finally it was possible to produce the sandwich boards in a one-shot process. Using bio-PU PUR 900 and linen fabrics, it was possible to increase e.g. the flexural strength of sandwich boards from 11.12 ±0.47 MPa to 30.80 ±1.39 MPa, i.e. an increase of 176%. The flexural modulus increased from 385.6 ±27.1 MPa to 1294.7 ±58.5 MPa, i.e. by 235%. The densities of the new sandwich boards is in the range of 300 – 490 kg/m³, which is 40% to >50% less than particle boards or OSB boards. The flexural strength is approx. twice as much as that of particle boards and comparable to OSB boards, while the flexural modulus is approx. 50% of particle boards and 30% of OSB boards. In addition, the new sandwich materials come with a low and completely reversible water uptake.

Summing up, it was possible to generate a new sustainable lightweight construction material with outstanding properties, making them suitable for markets, where weight saving or improved insulation are requested. This is valid for vehicle construction in general (esp. refrigerated vehicles), ship- and boat building (interior work), but as well furniture industry or booth construction.

1 INTRODUCTION

During the last decade, natural fibre reinforced plastics became a common material in industrial applications. In the German automotive industry the annual consumption of natural fibres (new & recycled) was 90,000 tons (16 kg fibres per vehicle), which is equal to 150,000 tons of composites [1]. If e.g. glass fibre reinforcement is substituted by natural fibres the density of the composite decreases by approx. one third from 1.8 g/cm³ to approx. 1.2 g/cm³ [2]. The usage of natural fibre reinforced synthetic PU is industrial standard for car parts like door-trim panels [3] or instrument boards [4].

In contradiction to the wide use of natural fibre reinforced fossil-based polyurethanes (PU), there is only few scientific work about the material development or mechanical properties. From the side of chemistry it is obvious, that the reaction of polyurethane formation promotes a good incorporation and fibre-matrix interaction of cellulose fibres in the resulting composite [5]. The isocyanate component can not only react with the polyol component, but as well with the hydroxyl groups at the fibre surface resulting in a covalent coupling.

For production of polyurethane foams water is added to the mixture, which reacts under CO_2 formation with the isocyanate component [6]. Thus, the natural fibres used in the actual automotive parts are dried prior to processing to prevent foam formation.

Concerning bio-polyurethanes, first commercial products have been established on the market consisting of a fossil-based isocyanate component and a bio-based polyol component [5]. This enables a share of 30% – 50% bio-based carbon atoms in the polymer. In addition, during the last years a large number of natural resources have been examined for the suitability as natural fibre-reinforced bio-polyurethane, based on e.g. palm-oil [7] or sugar cane [8]. A natural fibre reinforced bio-polyurethane containing polyols from soya, rapeseed or sunflower oil has been examined [9]. It contained up to 81% of bio-based resources and fulfilled all requirements for the use as car-interior trim panel. At that time the bio-based polyurethanes could not compete with the price of fossil-based ones, but in between the price gap has decreased to 10% – 15%, which can be compensated by advantages in disposal or recycling costs [2].

Sandwich constructions using a core made of natural fibre-reinforced bio-polyurethane could be a lightweight construction material for the future. From natural fibre-reinforced fossil-based polyurethane it is known, that the fibre reinforcement can improve the foam’s compression properties [7] and the insulation properties [10]. In general, natural fibre reinforced plastics are poorly used in sectors like refrigerated vehicles, ship- and boat building, interior work, booth construction etc., although the standard materials used instead (MDF, OSB, plywood or particle boards) would be easily to replace by sandwich boards made of natural fibre-reinforced PU.

Consequently a sustainable sandwich construction was designed consisting of bio-based PU equipped with natural fibre semi-finished products as cover layers. This new approach enables the production of materials, which are lighter and less expensive than the currently used conventional materials.

2 MATERIALS AND METHODS

2.1 Materials

Polyurethane systems, containing approx. 50% bio-based constituents, were obtained from Rühl Puromer GmbH (Friedrichsdorf/Ts., Germany): an Isocyanate ‘MDI’ (diphenylmethane-4,4'-diisocyanate, *puronate 900*, #07551) in combination with a bio-based polyol (*ep3272*, #EP 3272) as standard mixture, or with a modified lot containing more hydroxyl groups to increase the cross-linking (*ep3272*, #EP 3272-4). Both offer a sufficient starting time for manual processing (180 sec and 90 sec) and react at moderate temperature.

This polyurethane system is a so-called integral foam with harder surface structure, enabling the production of sandwich boards as one-shot process. The polyurethane system PUR 900 / EP3272 is recommended for isolation-foam packages as well as for production of medium hard foams, integral foams and coatings. Basic physical data of the isocyanate component at 25 °C are density 1.23 g/cm³, viscosity 220 mPa·s and processing temperature 20 – 25 °C [11].

Cover layers were obtained from different suppliers: flax fabric ('linen', #00 1818 00, 175 g/m²) was obtained from Leineweberei Hoffmann, Neukirch/Lausitz, Germany. Hemp laid scrim, rectangular, 2 threads per cm, 60 g/m² was obtained from Sachsenleinen GmbH, Waldenburg, Germany ('*Fadengelege*'). Polyester fabric ('*high-performance PE fabric*' #3493-100-B0-136 2) was obtained from Hänsel Tec GmbH, Iserlohn, Germany.

Hemp needle felt 800 g/m² was obtained from NAFGO GmbH (now Hempflax Deutschland GmbH, Wippingen, Germany). The felt is produced from hemp fibres type GDE02 (variety Fedora) with collective strength 44.9 ±4.7 cN/tex (674 ±20 MPa), single element strength 55.0 ±10.8 cN/tex (827 ±162 MPa), single element Young´s modulus 12984 ±2970 MPa, fineness 37.6 ± 1.5 µm and moisture content in standard climate 7.39 ±0.21% by weight. Details of the analysis pathway and results have been published elsewhere [12, 13].

2.2 Sandwich production

Sandwich boards are produced in a heatable moulding tool with internal dimensions of 560 x 400 x 10.4 mm³ in a one-shot process. The tool's surface is lined with Teflon foil to prevent glueing of the polyurethane. To avoid over-pressure, the tool is equipped with four exhaust channels (approx 1 x 5 mm²) at the corners. Depending on the type of polyol, different parameters are applied as listed in table 1. First the lower cover layer (and, if present the core reinforcement) is introduced into the tool, then the PU components are mixed in adequate proportions and poured in the tool. Finally the upper cover layer is laid over the PU and the tool is closed. To ensure complete curing of the boards, the reaction time was 15 minutes.

Polyol used	**EP 3272**	**EP 3272-4**
Start time in sec	180	90
Processing temperature in °C	68	68
Curing time in min.	15	15

Table 1: parameters for production of sandwich boards.

2.3 Methods of analysis

Sample specimens were cut from the composite boards in length direction of the reinforcing fibres. All test specimens were conditioned according to DIN EN ISO 291 [14] at 23°C and 50% relative humidity for at least 16 h prior to testing.

Flexural characteristics: sample specimens were prepared according to DIN EN ISO 14125 [15] (method A / class I / Appendix A: 200 x 25 mm²). The tests were conducted in a Zwick Z 250 universal tensile tester (manufactured by Zwick GmbH & Co. KG, Ulm, Germany) equipped with a 250 N load cell according to DIN EN ISO 14125 [15]. At least five test specimen were tested. Tensile strength and Young's modulus were calculated by the Zwick TestXpert software (version 070403, Zwick/Roell, Ulm, DE).

Moisture uptake was measured in a 'worst case scenario' using small specimen of 20 x 10 x 10.4 mm^3 (*l x b x h*). The specimen were pre-conditioned as mentioned above and then stored under demineralised water for two months. The weight and dimensions of each specimen were measured initially, after 1, 3, 7, 30 and 60 days. Finally the samples were allowed to dry at room temperature without conditioning for two weeks to assess the reversibility of the moisture uptake.

The fibre-PU interface was visualised using scanning electron microscopy (SEM): prior to the measurement the fibres are sputtered 2 min. at 50 mA with gold dust (Edwards Sputter Coater S150B; Crawley, West Sussex, GB). SEM images of the fibres are recorded using a Cam Scan CS24 device (EO Elektronen-Optik-Service GmbH, Dortmund, Germany; acceleration voltage 20 kV; Software: analySIS 3.2, SIS Soft Imaging System GmbH, Münster, Germany).

3 RESULTS

To reveal the intensity of fibre-PU interaction, a sample board was prepared containing 18% hemp fibres GDE02 (same lot of fibres as used in the hemp needle felt). A specimen from this board was broken in a tensile tester. Figure 1(a) shows exemplarily the fracture area of the sample. It can be seen easily that the high fibre share in weight occupies only a small share of the volume. Between the fibres are large areas of non-reinforced polyurethane. The detailed view on the pull-out-area of a fibre bundle in figure 1(b) depicts the good fibre-matrix interaction.

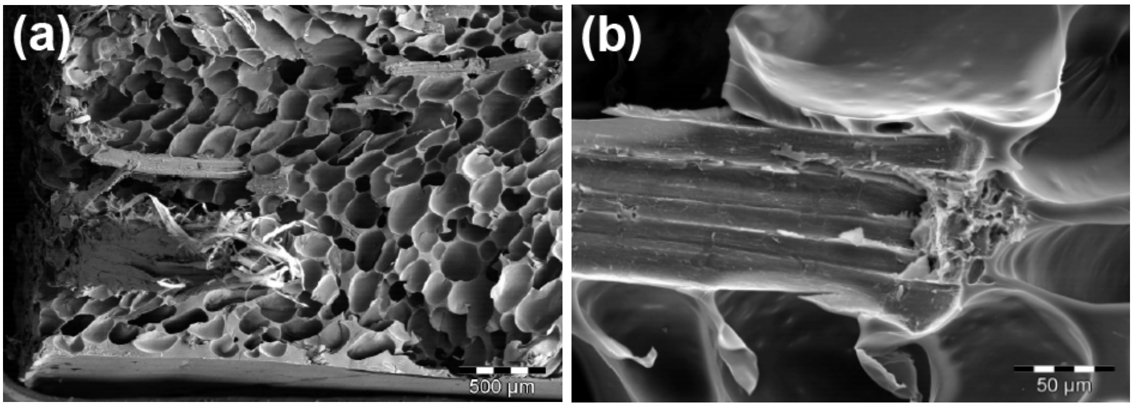

Figure 1: SEM image of the fracture area of a hemp fibre reinforced PU sample, (a) overview and (b) detailed area of pull-out.

The flexural strength and flexural modulus of the neat PU are displayed in figure 2. Neat PU boards were produced in different densities by variation of the amount used. Smaller amounts of PU can fill the tool as well, but give larger internal pores and thus a board of lower density. The displayed variants with densities of 0.22 and 0.31 g/cm^3 represent the minimal possible density and an average density with only low material loss through the exhaust channels. For comparison, the standard amount of the higher cross-linked PU with a density of 0.34 g/cm³ is shown.

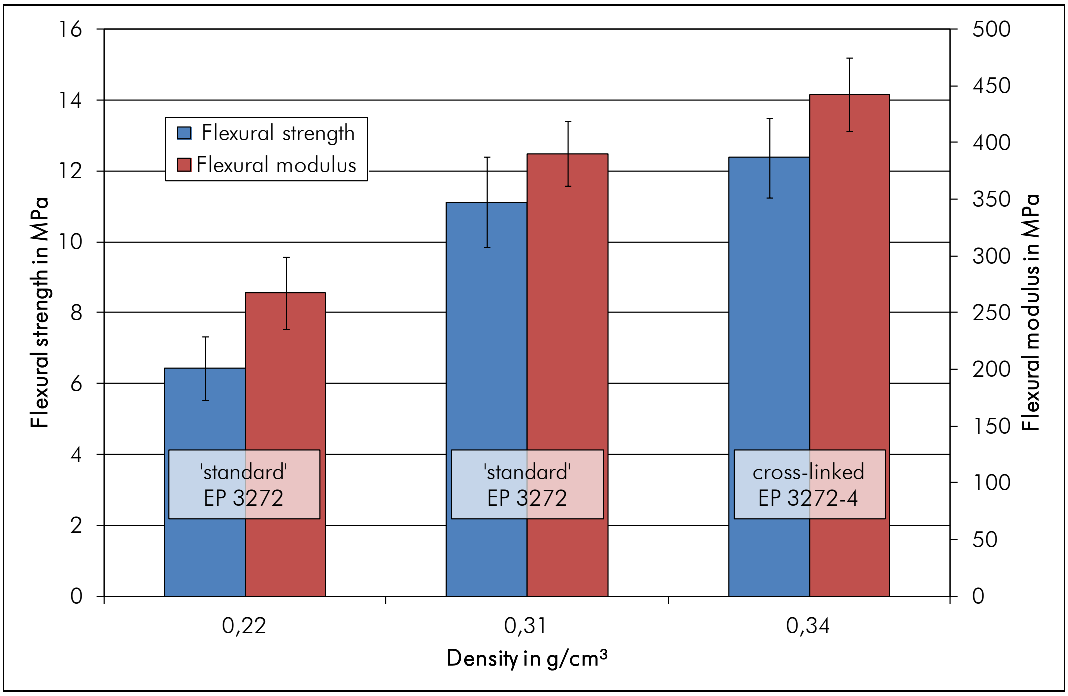

Figure 2: mechanical properties of the neat PU resins vs. density.

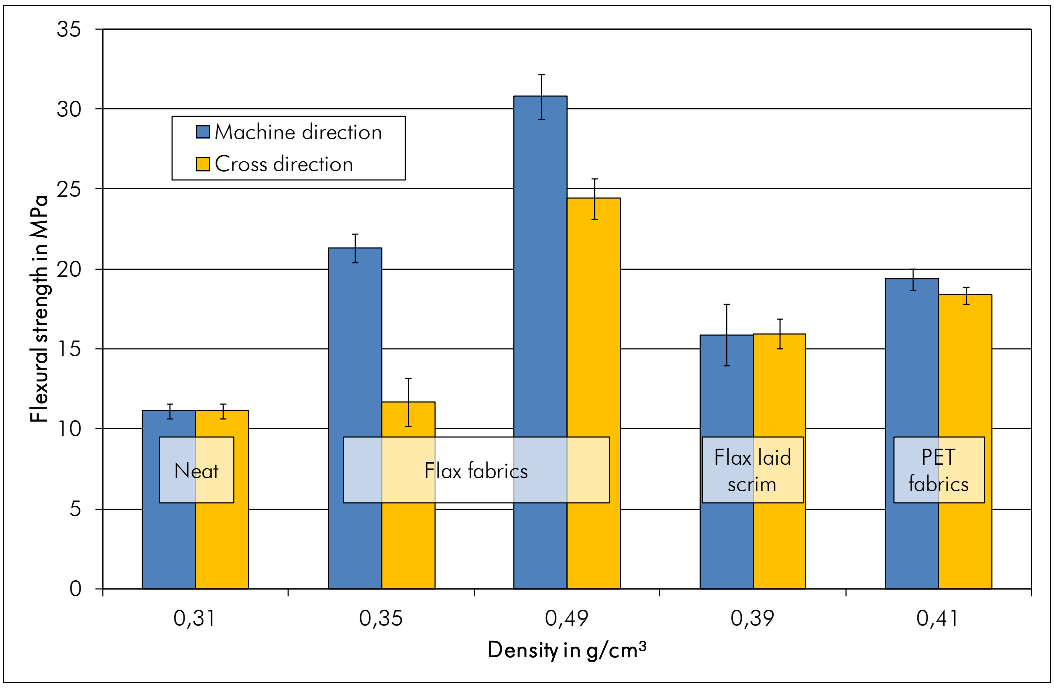

Figure 3: flexural strength of PU sandwich boards with different cover layers vs. density.

Obviously the flexural strength and flexural modulus increase with increasing PU density. This effect has to be considered when comparing results of different experiments. The flexural strength of the neat PU increases from 6.4 ±0.9 MPa (0.22 g/cm³) to 11.1 ±0.5 MPa (0.31 g/cm³), and the value for the PU EP3272-4 is 12.4 ±1.1 MPa (0.34 g/cm³). The flexural modulus follows the same tendency: it is 267.3 ±32.1 MPa (0.22 g/cm³), 385.6 ±27.1 MPa (0.31 g/cm³), and 442.5 ±32.6 (cross-linked, 0.34 g/cm³).

If this material is reinforced as sandwich structure with cover layers like flax fabrics (linen), flax laid scrim, or high-performance PET fabrics, the flexural strength and modulus increase dramatically. As displayed in figure 3, the addition of flax fabrics in combination with minimal and maximal possible amount of PU increases the flexural strength by a factor of up to three to 21.3 MPa (0.35 g/cm³) or 30.8 ±1.4 MPa (0.49 g/cm³). One has to mention, that due to the structure of the fabrics the reinforcing effect in cross-direction is approx 20% lower with 24.4 ±1.3 MPa. If flax laid scrim or PES fabrics are used as reinforcement, the properties are identical in machine- and cross-direction with 15.9 ±1.9 MPa for the laid scrim and 19.4 ±0.7 MPa for the PET fabrics.

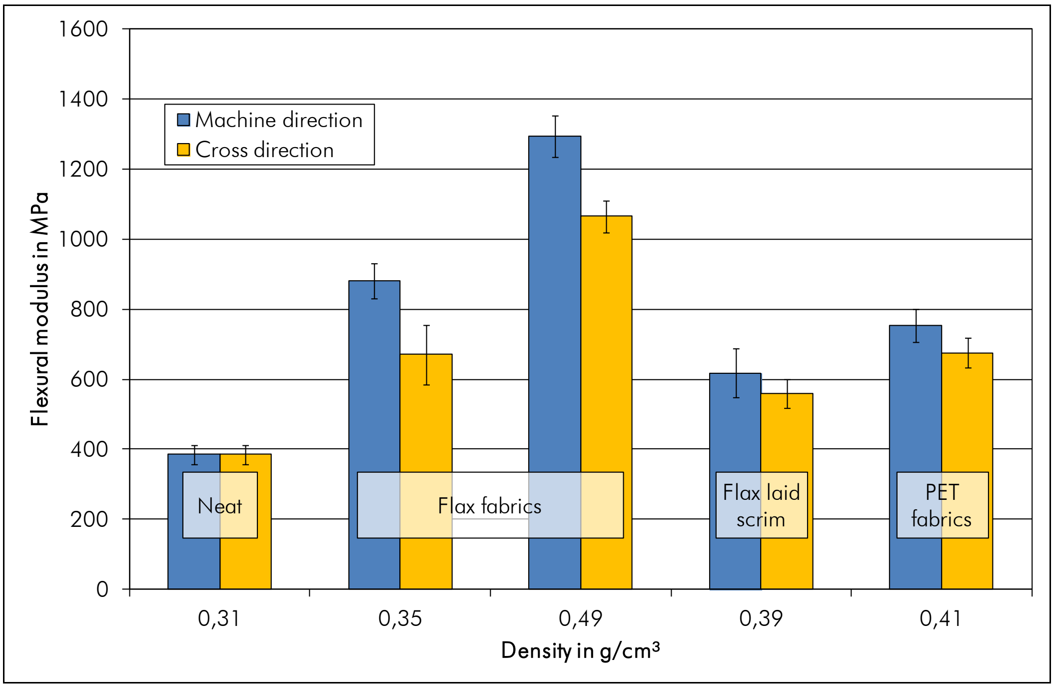

Figure 4: flexural modulus of PU sandwich boards with different cover layers vs. density.

Concerning the flexural modulus, the same tendency is observed for the samples (cf. figure 4): with flax fabric the modulus is increased to 881.3 ±49.8 MPa (0.35 g/cm³) or 1294.7 ±58.5 MPa (0.49 g/cm³). For the modulus the anisotropy is similar with 1065.3 ±46.3 MPa in cross-direction. For laid scrim and PET fabrics the values are 618.2 ±70.6 MPa and 754.2 ±47.5 MPa, resp.

Compared to conventional boards these are good properties: as listed in table 2, the sandwich boards made from Bio-PU are approx. half as dense as standard particle boards and display approx. the double flexural strength, but are approx. 40% lower in their flexural modulus. Compared to OSB or MDF boards, the density is 30 – 40% lower, the flexural strength is on the same level, and the flexural modulus is approx 50% lower.

Material type / parameter	**Bio-PU sandwich board, this work**	**Particle board DIN EN 13986 [16]**	**Oriented strand board (OSB) DIN EN 13986 [16]**	**Medium-density fibreboard (MDF) DIN EN 622-5 [17]**
Density in kg/m³	350 – 490	650 – 850[1]	600 – 1000[1]	500 – 650[1]
Flexural strength in MPa	21 – 31	12 – 20[1]	20 – 30[1]	22 – 32[1]
Flexural modulus in MPa	881 – 1667	1800 – 3000[1]	2500 – 4800[1]	2700 – 3700[1]
Maximum water sorption in mass-%	3 – 8	88 – 119[2]	103 – 137[2]	126 – 132[2]

[1]standard values required by DIN EN 13986 [16] or DIN EN 622-5 [17], resp.
[2]values measured according to DIN 52103 in [18]

Table 2: mechanical characteristics and water sorption of Bio-PU sandwich boards compared to standard boards boards (values for 10 mm thickness).

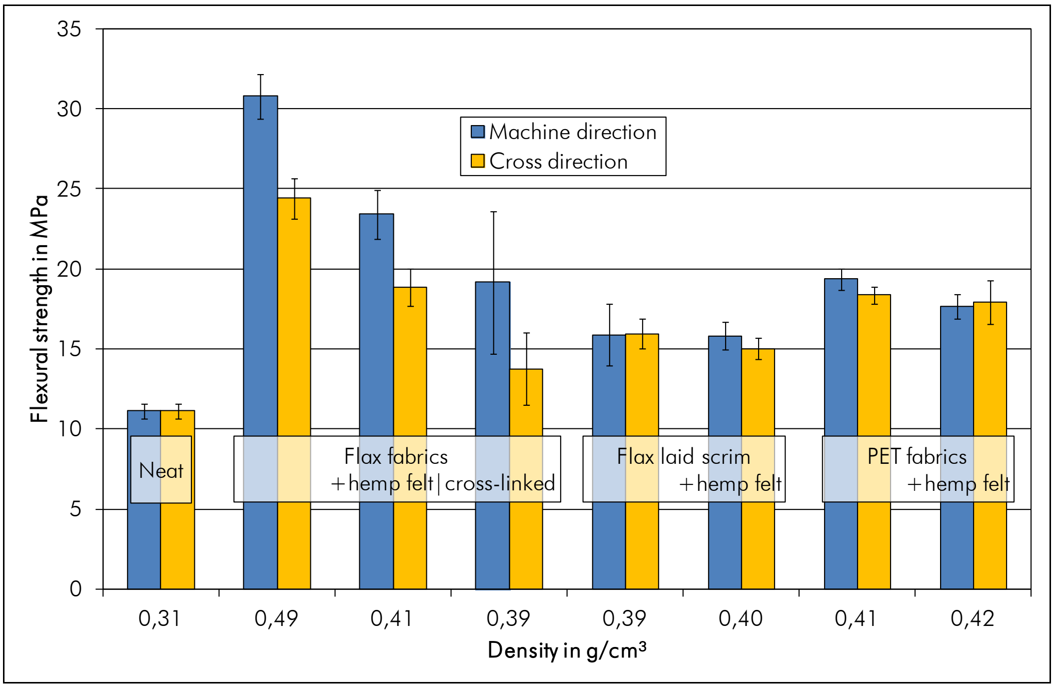

Figure 5: flexural strength of PU sandwich boards with and without core reinforcement vs. density.

The effects of additional core reinforcements or usage of higher cross-linked PU on the flexural strength are displayed in figure 5: compared to the reinforcement only by cover layers there are only negligible or even negative effects. For flax fabrics as cover layer, core reinforcement by hemp felt gives a flexural strength of 23.4 MPa (0.41 g/cm³), comparable to that of 0.35 g/cm³ density in figure 3. The usage of stronger cross-linking PU reduces the tenacity to 19.2 MPa (0.39 g/cm³), which is below that of 0.35 g/cm³ density in figure 3. In both cases the values in cross-direction are approx. 20 – 25% lower. For flax laid scrim and PET fabrics there is no significant effect of the core reinforcement observable — the differences are within the statistical uncertainty. Concerning the flexural modulus displayed in figure 6 the effects are similar. For flax fabric the moduli in machine direction are with core reinforcement on the same level as without, but in cross-direction they are lower and the difference increases. For laid scrim and PET fabrics there is again no significant effect of the core reinforcement observable.

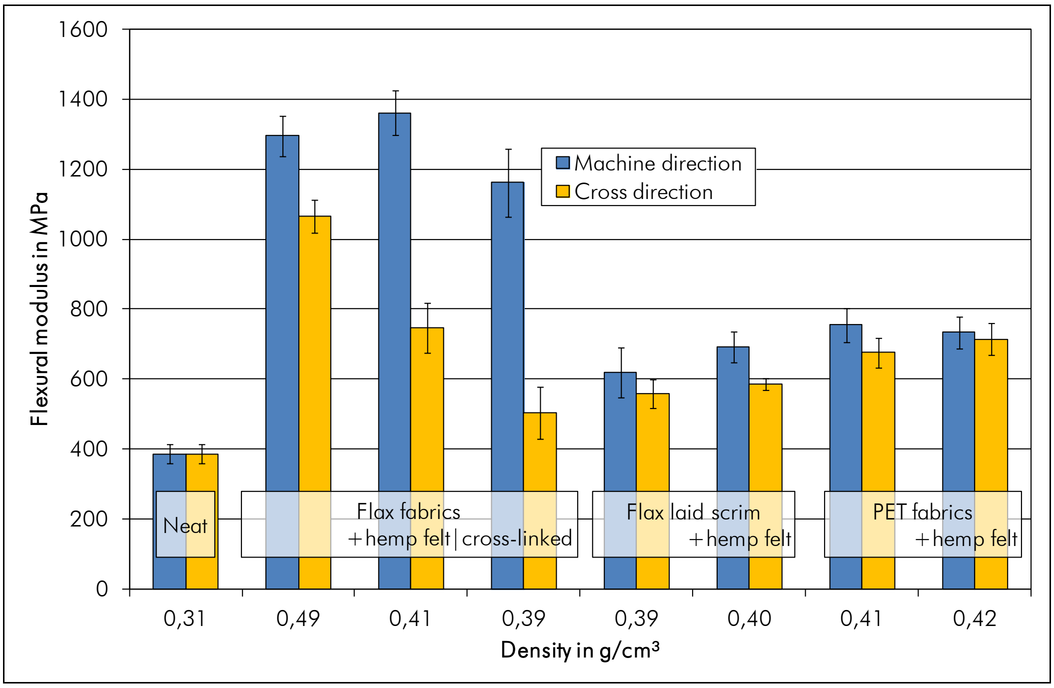

Figure 6: flexural modulus of PU sandwich boards with and without core reinforcement vs. density.

An additional advantage of the Bio-PU sandwich boards compared to standard boards is the low moisture absorption. As displayed in figure 7, the total uptake is even after two months under water as low as 3 – 4% of weight for the boards with cover layers of flax or PET. If additional core reinforcement by hemp felt is present, the water absorption can reach 8% after two months for the flax fabric covered sandwich board. If the cover layer is PET, the moisture absorption is limited. Interestingly the water absorption is totally reversible: 14 days storage at room temperature without conditioning is sufficient to dry the samples totally. Final values below 0% are caused by the effect, that the samples have initially been pre-conditioned at 50% rel. humidity. Compared to standard boards like MDF- OSB- or particle boards the water absorption is negligible. As listed in Table 2, those standard boards exhibit moisture absorption values of 88% – 136% (mass) [18], i.e. a factor of more than ten higher.

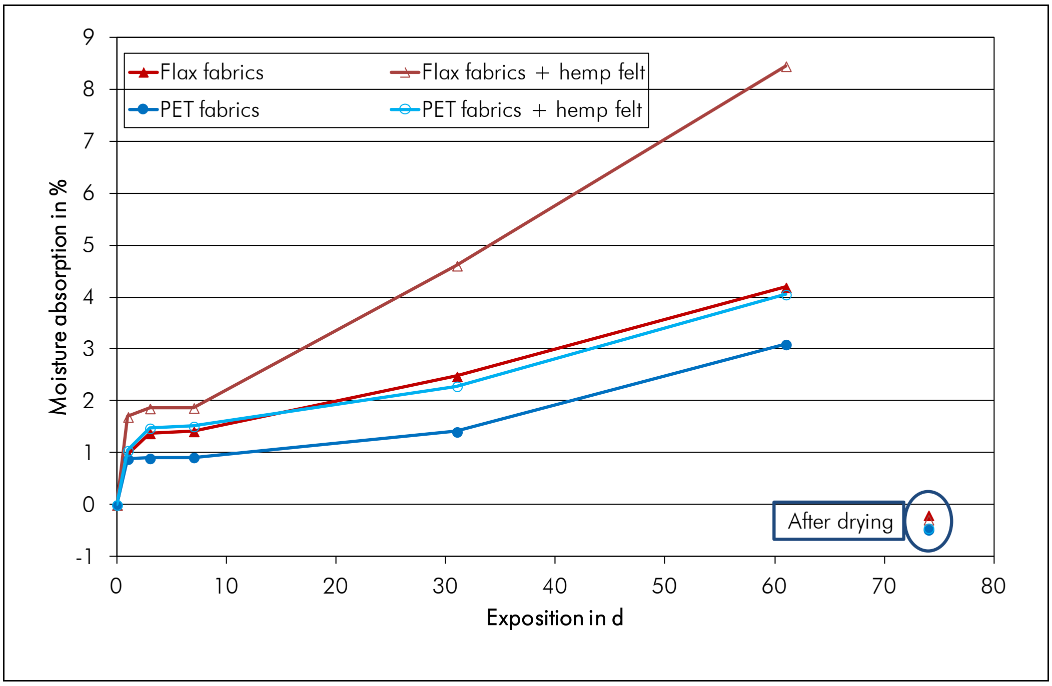

Figure 7: moisture absorption of PU sandwich boards with different cover layers and core reinforcement.

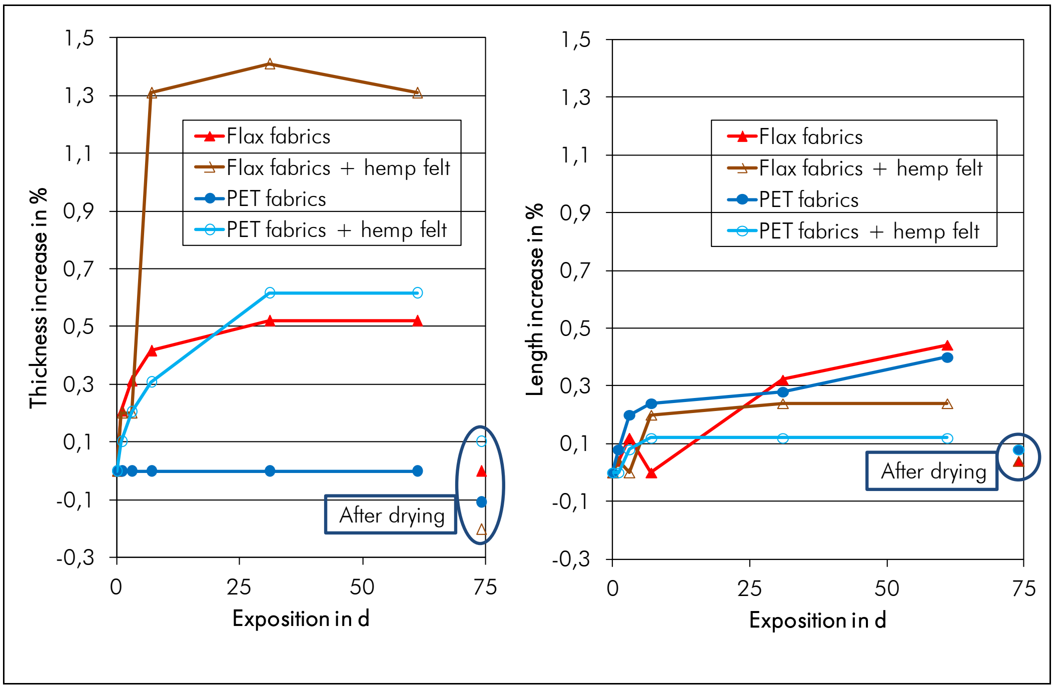

Figure 8: dimensional changes of PU sandwich boards caused by moisture absorption.

As displayed in figure 8, dimensional changes occur only to a small extent. The maximal effects have been observed in thickness increase, where the maximum is reached after 30 days with no effect for the PET covered and 0.5% swelling for the flax fabric covered sandwich. If additional core reinforcement by hemp felt is present, the swelling is 0.6% for PET and 1.4% for the flax fabric covered sandwich. In length direction the swelling is negligible low as 0.2 – 0.4%. It is as well as the thickness swelling completely reversible. The swelling in cross-direction is the same as in length and thus not displayed here.

4 CONCLUSIONS

It has been shown, that it is possible to produce boards of bio-based PU, which are 40 – 60% lower in density than conventional boards. The flexural strength is approx. twice as much as that of particle boards and comparable to OSB or MDF boards. Compared to those, the flexural modulus of the bio-PU boards is >50% lower. This is not necessarily a disadvantage, because high modulus stands as well for brittleness. Especially for materials used in vehicle or boat construction the materials should have a higher elasticity coming along with improved impact behaviour — and this is much easier with lower flexural modulus.

Another advantage of the bio-PU sandwiches is their low and completely reversible water absorption in combination with the excellent dimensional stability. The water absorption is as low as ten percent compared to standard MDF-, OSB- or particle boards [18].

5 ACKNOWLEDGEMENTS

We are much indebted to Thomas Mack (Rühl Puromer GmbH) and Michael Blumenstein (Hobum Oleochemicals) for supply of the PUR 900 system, to Hubert Mense (Hänsel Tec GmbH), Jürgen Steger (Sachsenleinen GmbH) and Mark Reinders (HempFlax Deutschland GmbH) for supply of the cover layers and core reinforcements. The authors wish to express their special thanks to Mrs. Manuela von Salzen and Mrs. Tanja Slootmaker (both FIBRE, Germany) for their support in composite analyses.

The IGF research project no. 17913 N/1 of the research union *Forschungskuratorium Textil e.V.*, Reinhardtstraße 12-14, 10117 Berlin, is funded via the AiF within the framework of the programme for funding the industrial co-operative research and development (IGF) by the German federal ministry of economics and technology, based on a resolution of the German Federal Parliament.

Bundesministerium für Wirtschaft und Technologie

REFERENCES

[1] M. Carus, J. Müssig and C. Gahle, *Naturfaserverstärkte Kunststoffe — Pflanzen, Rohstoffe, Produkte —*. Booklet, 40 pp, Fachagentur Nachwachsende Rohstoffe e.V. — FNR, Gülzow, Germany, 2nd revised ed. 2008. Online available http://www.nova-institut.de/pdf/pdf_227-brosch_nfk_2008.pdf. Accessed 2015-03-18.

[2] J. Müssig, M. Schmehl, H.B. von Buttlar, U. Schönfeld, K. Arndt, Exterior components based on renewable resources produced with SMC technology — considering a bus component as example. *Ind Crop Prod*, **24,2**, 132 – 145, 2006. ISSN 0926-6690.

[3] H. Müller and K.W. Fries, PURe Natur im Automobil — Mit Flachs und Sisal

verstärkte Polyurethane für Automobil-Innenraumteile. *Kunststoffe*, **88,4**, 544 – 546, 1998. ISSN 0023-5563.
[4] J. Müssig, *Fasern aus Nachwachsenden Rohstoffen als Konstruktionswerkstoffe*. In *Fachgespräch Konstruktionswerkstoffe* (organiser Fachagentur Nachwachsende Rohstoffe), Gülzow, Germany 2004-10-14). FNR, Gülzow, DE, 2004.
[5] H.J. Endres and A. Siebert-Raths, *Technische Biopolymere — Rahmenbedingungen, Marktsituation, Herstellung, Aufbau und Eigenschaften*. Hanser-Verlag, Munich, DE, 2009. ISBN 978-3-446-42104-2.
[6] E. Baur, S. Brinkmann, T.A. Osswald, E. Schmachtenberg, *Saechtling Kunststoff Taschenbuch*. 30th edn. Hanser-Verlag, Munich, DE, 2007. ISBN 978-3-446-40352-9.
[7] E.S. Ali and S. Ahmad, Bionanocomposite hybrid polyurethane foam reinforced with empty fruit bunch and nanoclay. *Composites: Part B*, **43,7**, 2813 – 2816, 2012. ISSN 1359-8368.
[8] J. Vega-Baudrit, K. Delgado-Montero and S. Madrigal-Carballo, Biodegradable Polyurethanes From Sugar Cane Biowastes. *Cellulose Chemistry and Technology*, **45,7-8**, 507 – 514, 2011. ISSN 0576-9787.
[9] B. Dahlke, H. Larbig, H.D. Scherzer, R. Poltrock, Natural fiber reinforced foams based on renewable resources for automotive interior applications. *Journal of Cellular Plastics*, **34,4**, 361 – 379, 1998. ISSN 0021-955X.
[10] S.A. Ibraheem, A. Ali and A. Khalina, Development of Green Insulation Boards from Kenaf Fibres and Polyurethane. *Polymer-Plastics Technology and Engineering*, **50,6**, 613 – 621, 2011. ISSN 0360-2559.
[11] Rühl Puromer GmbH, *Technisches Informationsblatt PUR900*, edition 11/2008. Rühl Puromer GmbH, Friedrichsdorf, DE.
[12] J. Müssig, H. Fischer, N. Graupner, A. Drieling, *Testing Methods for Measuring Physical and Mechanical Fibre Properties (Plant and Animal Fibres)*. Chapter 13 in J. Müssig (ed) *Industrial Applications of Natural Fibres — Structure, Properties and Technical Applications*. Wiley-VCH, Weinheim, DE, 2010, pp. 269 – 309. ISBN 978-0-470-69508-1.
[13] J. Müssig, G. Cescutti and H. Fischer, *Integrated quality management for bast fibres in technical applications*. Chapter 10 in P. Bouloc, S. Allegret and L. Arnaud (eds): *Hemp — Industrial Production and Uses*. CABI Press, Wallingford, Oxfordshire, UK, 2013. ISBN 978 1 845937 928.
[14] DIN EN ISO 291:2008. *Kunststoffe — Normalklimate für Konditionierung und Prüfung (ISO 291:2008)*; German version EN ISO 291:2008.
[15] DIN EN ISO 14125:2011. *Faserverstärkte Kunststoffe — Bestimmung der Biegeeigenschaften (ISO 14125:1998 + Cor.1:2001 + Amd.1:2011)*; German version EN ISO 14125:1998 + AC:2002 + A1:2011.
[16] DIN EN 13986:2004. *Holzwerkstoffe zur Verwendung im Bauwesen — Eigenschaften, Bewertung der Konformität und Kennzeichnung*; German version EN 13986:2004.
[17] DIN EN 622-5:2010. *Faserplatten - Anforderungen — Teil 5: Anforderungen an Platten nach dem Trockenverfahren (MDF)*; German version EN 622-5:2009.
[18] Fitz, C.; Krus, M. & Zillig, W., *Untersuchung zum Feuchteschutz von Leichtbaukonstruktionen mit Bauplatten aus Holzwerkstoffen*. Report no. HTB-10/2004. Fraunhofer Institut für Bauphysik, Stuttgart, DE, 2004. Online available http://www.hoki.ibp.fraunhofer.de/ibp/publikationen/berichte/HTB-10-04_DGFH%20_Endbericht02_2003.pdf. Accessed 2015-03-18.