Characterization of foam properties of 2C-polyurethane for wood bonding

Master Thesis

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Eidesstattliche Erklärung

Ich erkläre eidesstattlich, dass ich die Arbeit selbständig angefertigt habe. Es wurden keine anderen als die angegebenen Hilfsmittel benutzt. Die aus fremden Quellen direkt oder indirekt übernommenen Formulierungen und Gedanken sind als solche kenntlich gemacht. Diese schriftliche Arbeit wurde noch an keiner Stelle vorgelegt.

Wien, am

Bliem Peter

Abstract

In this master thesis, the mechanical foam properties of a two-component polyurethane were systematically investigated with the aim of using it for wood bonding with gap filling properties. The mechanical properties of the wood adhesive bonds are strongly dependent on the strength, stiffness and homogeneity of the adhesive foam induced, which in turn is influenced by various factors. These influencing factors are the ratio of isocyanate to polyol, the amount of the additional water, water available from the wood substrate (wood moisture content), the press temperature and the thickness of the foam. The longitudinal tensile shear strength and the shear modulus were measured using lap-joint specimens with defined bond-line thickness allowing the adhesive to foam between the adherents. A “Design of Experiments – DOE” was used in order to investigate the various conditions and potentially influencing factors. The data were evaluated with statistical analysis software, whereby possible correlations and/or interrelationships of influencing factors with the response values (strength and stiffness) were investigated more closely. Subsequently, constellations in which the best adhesive-foam properties could be achieved, were derived.

In principle, for low foam thickness (0.5 mm) the highest strength and stiffness values were achieved with those foams which were produced at a low press temperature (20 °C), a high wood moisture content (21 %), an average additional water content (5.5 %) and a high content of isocyanate (>85 %). An increase in the press temperature or the foam thickness leads to a major drop in the mechanical foam properties, whereby both lower wood moisture content as well as lower amount of isocyanate leads also to a reduction in performance. It has also been observed that with a low foam thickness (0.5 mm), a relatively high wood failure occurs, especially in the case of the samples using a low press temperature (20 °C). A very low wood moisture content (3 %) and/or a very low amount of additional water (1 %) leads to an insufficient foaming of the PUR adhesive, especially at higher foam thicknesses.

Keywords: mechanical properties of PUR-foam, 2-component PUR, longitudinal tensile shear strength, shear modulus, general full factorial design

Zusammenfassung

In dieser Masterarbeit wurden die mechanischen Schaumeigenschaften eines Zwei-Komponenten Polyurethans systematisch untersucht mit dem Ziel, es für die Holzverklebung mit Lückenfüllungseigenschaften zu verwenden. Die mechanischen Eigenschaften der Holzklebebindung sind sehr stark von der Festigkeit, Steifigkeit und Homogenität des Schaumes abhängig, wobei dieser wiederum von verschiedenen Faktoren beeinflusst wird. Diese Einflussfaktoren sind das Mischungsverhältnis Isocyanat zu Polyol, Menge an zusätzlichem Wasser, verfügbares Wasser aus dem Holz (Holzfeuchte), Presstemperatur und die Dicke des Schaumes. Die Längszugscherfestigkeit und der Schubmodul wurden unter Verwendung von Überlappungsproben mit definierten Klebefugendicken gemessen, was es dem Klebstoff ermöglicht dazwischen zu schäumen. Zur Untersuchung der verschiedenen Zustände und potenziell beeinflussenden Faktoren wurde ein "Design of Experiments - DOE" verwendet. Die Daten wurden mit statistischer Analysesoftware ausgewertet, wobei die Einflussgröße und mögliche Zusammenhänge bzw. Wechselbeziehungen dieser Faktoren auf die Zielgrößen (Festigkeit und Steifigkeit) genauer untersucht wurden. In weiterer Folge wurden jene Konstellationen daraus abgeleitet, bei welcher die besten Klebstoff-Schaumeigenschaften erreicht werden können.

Grundsätzlich wurden bei geringer Schaumdicke (0,5 mm) die höchsten Festigkeits- und Steifigkeitswerte mit jenen Schäumen erreicht, die bei einer niedrigen Presstemperatur (20 °C), hoher Holzfeuchte (21 %), mittlerem zusätzlichen Wassergehalt (5,5 %) und hohem Isocyanatgehalt (>85 %) hergestellt wurden. Durch Erhöhung der Presstemperatur oder der Schaumdicke kommt es zu einem starken Abfall der mechanischen Schaumeigenschaften, wobei sowohl eine niedrigere Holzfeuchte als auch eine geringere Menge an Isocyanat zu einer Leistungsminderung führt. Es wurde auch beobachtet, dass bei geringer Schaumdicke (0,5 mm) es noch zu einem relativ hohen Anteil an Holzbruch kommt, speziell bei den verklebten Proben bei geringer Presstemperatur (20 °C). Eine sehr geringe Holzfeuchte (3 %) und/oder eine sehr geringe Menge an zusätzlichem Wasser (1 %) führte bei Zunahme der Schaumdicke zu einer unzureichenden Aufschäumung des PUR-Klebstoffes.

Schlüsselwörter: mechanische Eigenschaften von PUR-Schaum, 2-K PUR, Längszugscherfestigkeit, Schubmodul, vollfaktorieller Versuchsplan

Contents

1 Introduction

Polyurethane adhesives were invented in the year 1937 by Otto Bayer, a German chemist. For wood bonding PUR adhesive was used for aircraft construction for the first time in the 1940s (Lohmann, 2010). Later, polyurethanes became more and more important for the wood industry, especially for producing engineered wood products. Polyurethanes are also used in form of polyurethane foams, which can be rigid or flexible. As it is shown in Figure 1, around 72 % of the world consumption of polyurethanes are foams (Ionescu, 2005).

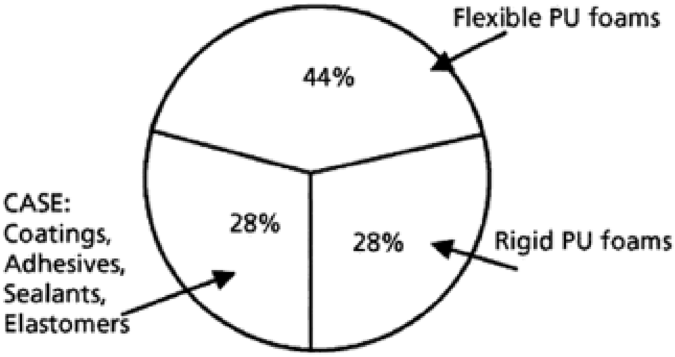


Figure 1: Worldwide consumption of polyurethane from the year 2000 to 2002 (Ionescu, 2005)

Due to the high flexibility in designing polyurethane properties applications may be found in almost every sector of daily life (Figure 2). In the year 2004 the amount of produced polyurethanes were about 10.6 million tonnes worldwide, in 2014 the production was almost 20 million tonnes per year (Ionescu, 2016).

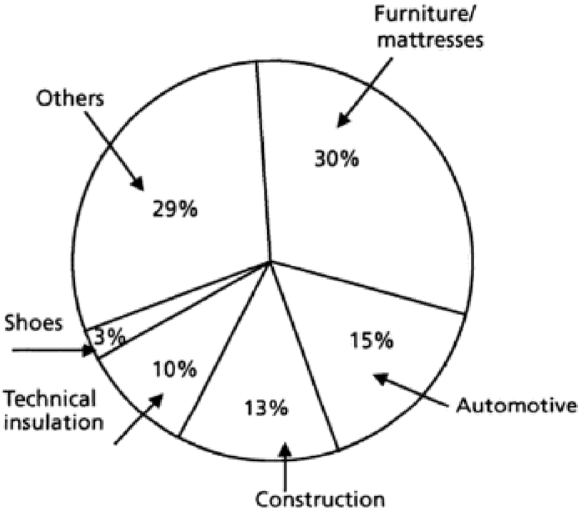


Figure 2: Main applications of polyurethanes (Ionescu, 2005)

In order to produce polyurethane foams, various types of isocyanates and polyols are required, which react with each other by polyaddition (Elsner et al., 2008).

1.1 Isocyanate

Isocyanates are very reactive (Van Langenberg et al., 2010) and characteristic for them is the functional group R–N=C=O (NCO). As reviewed in Elsner et al. (2008) the two most important isocyanates are toluene diisocyanate (TDI) and methylene diphenyl diisocyanate (MDI). Both of them have an aromatic character, but there are two essential differences between TDI and MDI (Elsner et al., 2008):

1. TDI is more toxic and special safety precautions are required during processing
2. TDI has a higher NCO-value, that means that there are more NCO groups available per mass, which means that for a necessary amount of NCO groups, less mass of isocyanate is required

1.1.1 Toluene diisocyanate (TDI)

According to Leimenstoll & Stepanksi (2016) there are two different types of isomers (Figure 3), depending on where the second NCO group is located. Normally the TDI mixtures consist of a higher amount of 2,4 TDI and a lower amount of 2,6 TDI. Table 1 shows common TDI products with the different amounts of the isomers. For the pure TDI, the TDI 100 (100 % 2,4 TDI) has the biggest importance (Elsner et al., 2008). Due to the high volatility and toxicity, TDI must be labelled as dangerous goods.

Table 1: Overview of common TDI products (Elsner et al., 2008)

	amount of 2,4-TDI [%]	**amount of 2,6-TDI [%]**
TDI 80	80	20
TDI 65	65	35
TDI 100	100	-

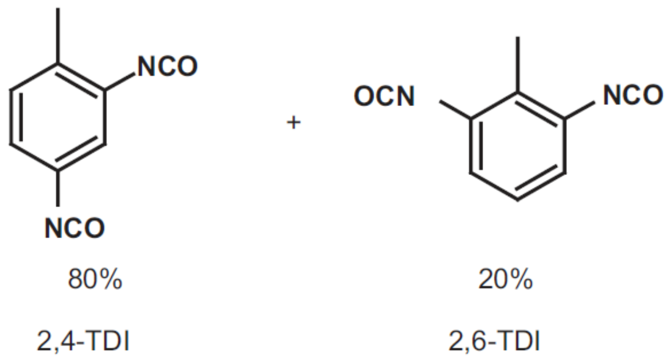


Figure 3: Two different TDI isomers (Leimenstoll & Stepanksi, 2016)

1.1.2 *Methylene diphenyl diisocyanate (MDI)*

For polyurethane adhesives, MDI is the most used isocyanate type (Leimenstoll & Stepanksi, 2016). It can be distinguished in pMDI (polymeric-MDI) and mMDI (monomeric-MDI), which may occur in the form of various isomers (Figure 4). 4,4-MDI and 2,4-MDI are the most used isomers in the mMDI subgroup. Polymeric-MDI has a quantitatively stronger presence on the market (Elsner et al., 2008). According to Leimenstoll & Stepanksi (2016) this is attributed to a better reactivity and the possibility to get 3-dimensional cross-linked duromeric structures.

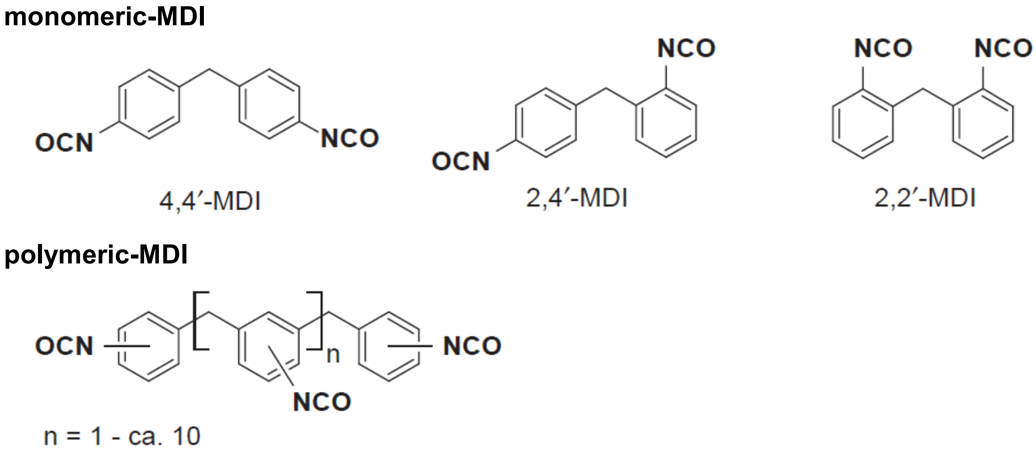


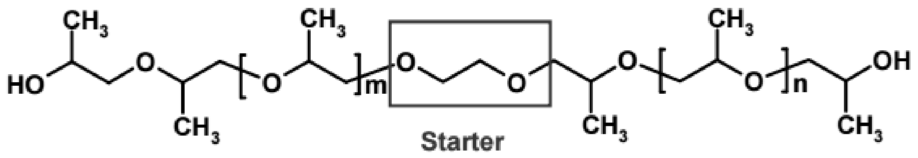
Figure 4: Various types of methylene diphenyl diisocyanate (Leimenstoll & Stepanksi, 2016)

1.2 Polyol

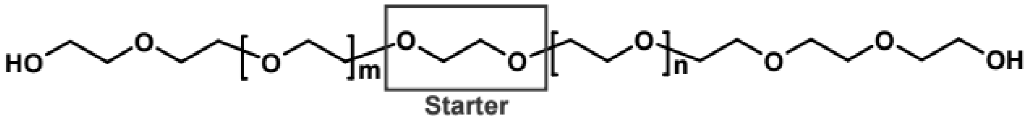
The second important constituent for the polyurethane chemistry is the polyol. This is an organic compound from the group of alcohols with a higher amount of hydroxyl groups (Leimenstoll & Stepanksi, 2016). Most of the polyols are obtained from the petroleum based industry, but there are also polyols available which are produced from renewable resources such as palm oil (Chian & Gan, 1998). Leimenstoll & Stepanksi (2016) describe that in contrast to isocyanates, polyols are with regard to danger classification less or uncritical and they are chemically stable. There is a high amount of different types of polyols, which differ among themselves by their functionality, their molecular weights, the molecular weights distribution, the reactivity and their chemical structure. For the last one, polyols can be divided into two sub-categories, the polyether and the polyester polyols.

1.2.1 Polyether polyols

Polyether polyols are available in different chain lengths and functionality for PUR applications. The functionality means the average amount of reactive hydroxyl groups per each molecule. For the production of polyether polyols different starters can be used, e.g. ethylene oxide (EO) and/or propylene oxide (PO). With these it is possible to achieve molecular weights ranging from 200 up to 6000 g/mol and also a functionality may be designed from 2 up to 6 hydroxyl groups per molecule (Elsner et al., 2008). In Figure 5 polyol molecules derived from two different starters are shown.



PO Ether (Polypropylenoxid)



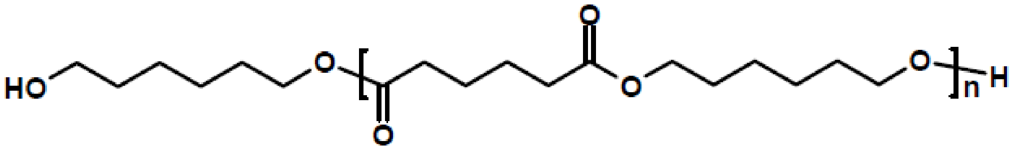
EO Ether (Polyethylenoxid)

Figure 5: Different types of polyether polyols (Leimenstoll, 2011)

1.2.2 Polyester polyols

Polyester polyols have along the molecular chain ester groups, which make the molecule to a highly polar substance. This means it has an excellent adhesion on other polar substances or surfaces (Leimenstoll & Stepanksi, 2016).

The polyester polyols (Figure 6) are produced by a polycondensation with polyols and polycarboxylic acids. The advantage over polyether polyols is, that polyester polyols have better mechanical properties, but they also have a lower stability against hydrolysis with water, especially air humidity (Elsner et al., 2008).



Hexandioladipat

Figure 6: Example for a polyester polyol (Leimenstoll, 2011)

1.3 Chemical reactions for producing polyurethanes

Starting from the isocyanate component, a variety of possible reactions may take place as summarized by Leimenstoll (2011) and shown in Figure 7. In this master thesis the reactions of isocyanate forming a urethane or a urea are considered as the most important ones.

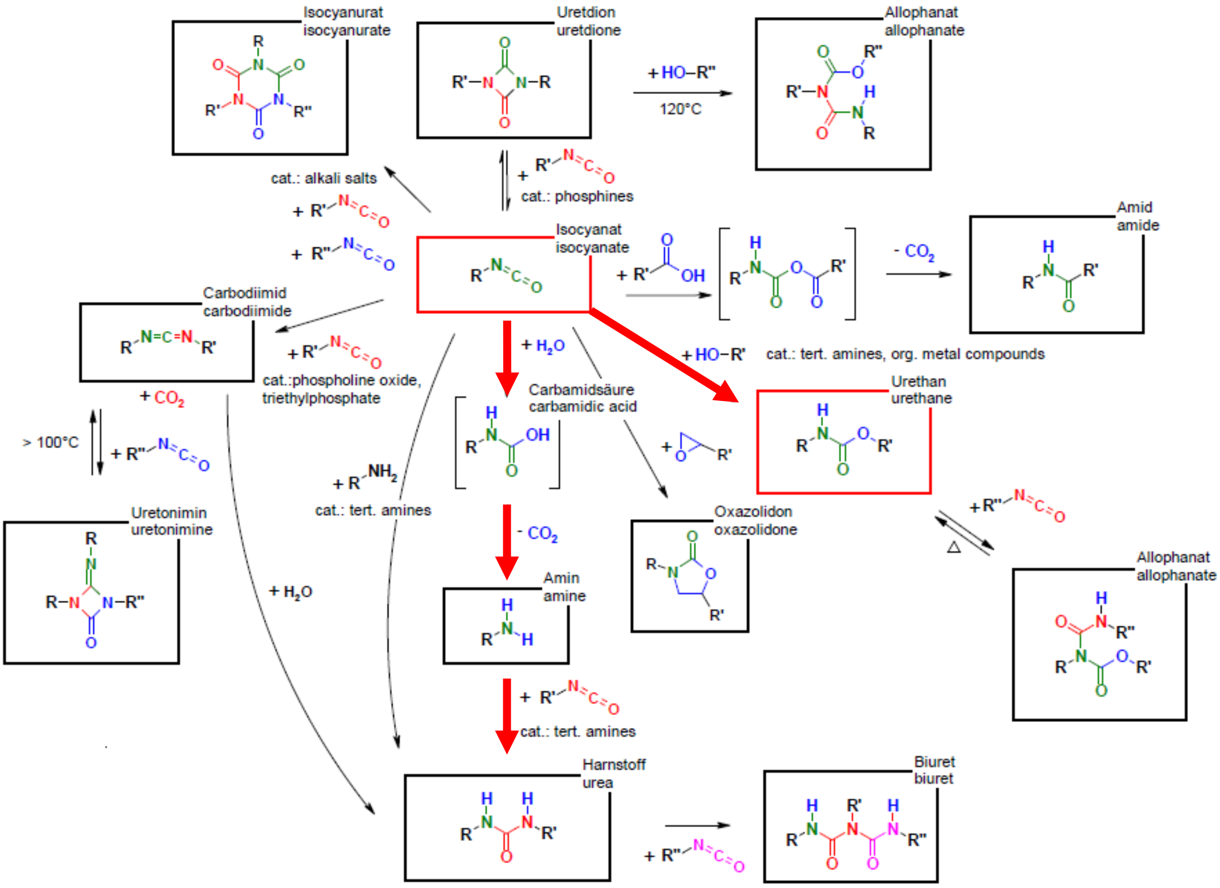


Figure 7: Possible chemical reactions of isocyanate (Leimenstoll, 2011)

According to Hepburn (1992) the reaction rate of diisocyanate with a hydroxyl group is higher than with water. This means at lower temperature the isocyanate reacts faster to a urethane than to a urea. A higher reaction temperature leads to a reduction in the difference in the reaction rate between the faster and slower reaction by favouring the latter.

1.3.1 *Urethane formation*

For the formation of a urethane molecule, isocyanate and an alcohol are required (Figure 7). A urethane is created directly through an addition of these two components. By using multifunctional isocyanates and polyols it is possible to get long molecular chains, which are the skeletal structure for the polymer (Elsner et al., 2008). During the exothermic reaction of isocyanate and polyol, heat will be released (Leimenstoll & Stepanksi, 2016). As reviewed in Elsner et al. (2008) this reaction is also called gel-reaction. With this reaction only it would not be possible to create a polyurethane foam, it is necessary that there is also a urea formation involved, creating CO_2 gas which is involved in the foam expansion. At higher temperatures isocyanate can also reacts to other products with undesirable consequences as described elsewhere (Pizzi & Mittal, 2003).

As described in Wicks et al. (2007) the urethane bonds have an oxygen linkage and thus leads to a formation of cyclic hydrogen bonds, which can absorb higher amounts of energy. Urea bonds can also form cyclic hydrogen bonds, but in a much smaller extent.

1.3.2 Urea formation

For the expansion of the polyurethane, the urea formation is important. In this reaction, isocyanate reacts with water as highlighted in Figure 7.
In the first step isocyanate reacts with water to an amine under release of carbon dioxide (CO_2). For the case of a wood adhesive bond this water may be present in the wood structure known as moisture content, in the adhesive mixture or from the moisture of the air. Due to this release of CO_2 it comes to an expansion of the polyurethane mixture and thus polyurethane foam is formed. The second step is, that the amine reacts with an isocyanate to a urea, which is a cross-linker between the isocyanates groups (Andersson, 2011).

Elsner et al. (2008) defines this reaction as “driving reaction” Because of this high reaction probability between isocyanate and water, it is very important to avoid contact with any kind of water or vapour during storage or when a reaction is undesired (Leimenstoll & Stepanksi, 2016).

By addressing these two reactions (urea and urethane) and through the selection and/or modification of the two main raw materials isocyanates and polyols, it is possible to design polyurethanes with various different macroscopic and mechanical properties (Elsner et al., 2008).

According to Efstathiou (2011) the urethane and urea groups have new hydrogen atoms. In side reactions (Figure 7) these can react with isocyanate to other products (biuret and allophanate), which may influence the mechanical properties of the polyurethane due to branching and cross-linking effect. At lower temperature the biuret – isocyanate reaction occurred much faster than the allophanate – isocyanate reaction.

With regard to the bonding of wood using PUR adhesive it is known that this is basically very suitable (Wagenführ & Scholz, 2012). At 2C-polyurethane systems isocyanate reacts with polyol which leads to the formation of urethane compounds. For 1C-PUR systems a minimum wood moisture content of 8 % is necessary for the reaction with isocyanate. It is clear that besides the typical polymers such as cellulose, hemicellulose, lignin and extractives, wood also contains a certain proportion of water (bonded or available).

It may be assumed that the wood substrate has a certain influence on the different reaction possibilities (Figure 7) due to its wood moisture content and therefore on the mechanical properties of the bond line.

1.4 Hypothesis and Goals

Isocyanates can interact with the surface hydroxyl groups of the wood fibres, whereby excellent adhesion properties can be achieved (Mosiewicki et al., 2009). Conventional 2C-PUR systems do not take into account the moisture content of the wood in its composition of isocyanate, polyol and water. Due to the different reactions described in chapter 1.3 and the resulting reaction products, the assumption is that the following factors:

- ratio of the components isocyanate ↔ polyol
- amount of additional water
- wood moisture content
- press temperature
- foam thickness

have a substantial influence on the foam properties. Therefore, the aim of this master thesis is to systematically examine the influence of the individual factors on the strength and stiffness of the polyurethane foam.

Based on a general full factorial experimental design, these factors will be systematically varied and furthermore tensile lap shear samples will be produced, tested and evaluated regarding longitudinal tensile shear strength and the shear modulus. The results will be analysed using a statistical software. Thus, possible correlations and/or interrelationships of the influencing factors with the response values (strength and stiffness), should be shown.

The hypothesis for this master thesis is that the systematic evaluation of these factors and their relationships can lead to significant improvements in the foaming and bonding properties of a two component polyurethane adhesive.

2 Materials and Methods

2.1 Chemicals

For this master thesis an experimental water free 2C-PUR system LOCTITE VN 3152 PURBOND from the company Henkel AG & Co. KGaA, Düsseldorf, Germany was used. Component B is isocyanate and component A is the polyol including some amount of not further defined additives. In Table 2 selected physical properties of the two components are shown.

Table 2: Overview of the physical properties of the used isocyanate and polyol (Henkel, 2016)

	Component A (polyol)	Component B (isocyanate)
form	liquid	liquid
colour	white-transparent	brown
density (20 °C)	1.01 g/cm³	1.15 g/cm³
viscosity (20 °C)	~ 600 mPas	~ 6000 mPas
equivalent weight	178.38 g/eq	211 g/eq
other	purity 99.9 % polyol	-

According to Huntsman (2001) the equivalent weights in Table 2 represent the amount of grams for each reactive group (NCO or OH) or the mass of a reactive group to have an equivalent reactivity. It is calculated with the molecular weight divided by the OH value (polyol) or weight percent NCO (isocyanate).

2.2 Pretests

In order to find suitable factorial points for the experimental design, two different preliminary tests had to be carried out beforehand. These two tests were first, a beaker test where the polyurethane components were mixed in a beaker and the foaming behaviour was simply observed. As second test a gap filling test was used, where the polyurethane adhesive mixture was applied on the substrate wood. In addition, the handling of the used materials was trained, which is also very important for the fast, accurate and safe work during sample preparation.

2.2.1 Beaker test

The aim of this preliminary experiment was to find the extreme values for the factors amount of isocyanate, amount of additional water in the polyol and temperature. As fast and easy way to get these results, a beaker test was used.

2.2.1.1 Experimental design and calculations

The first step was to create a small experimental design with a wide range of values for the factors, which are shown in Table 3 displayed in percentage of the individual constituents and the corresponding mass. The sum of isocyanate and polyol always results to 100 % or 14 grams, respectively. For the additional water the amount was 0.2 % or 10 % in relation to 100 % polyol. That means if there is a lower polyol content, the additional water content is also lower. For example 0.2 % of water from 0.7 grams polyol (which are 100 %) are 0.0014 grams or 1.4 µl water.

Table 3: Experimental design for the beaker test with the values in percentage and gram for each factor

isocyanate		**polyol**		**additional water**	
[%]	[g]	[%]	[g]	[%]	[g]
95	13.3	5	0.7	0.2	0.0014
95	13.3	5	0.7	10	0.007
90	12.6	10	1.4	0.2	0.0028
90	12.6	10	1.4	10	0.14
85	11.9	15	2.1	0.2	0.0042
85	11.9	15	2.1	10	0.21
70	9.8	30	4.2	0.2	0.0084
70	9.8	30	4.2	10	0.42
60	8.4	40	5.6	0.2	0.0112
60	8.4	40	5.6	10	0.56

The beaker test was done at a temperature of 20 °C using amounts displayed in Table 3. For the temperatures 70 and 120 °C no beaker tests were performed for the 90 % isocyanate values.

2.2.1.2 Procedure

For the beaker tests three different types of beakers were used each with a filling capacity of 200 ml. The first set of transparent beakers was made out of polystyrene (PS) and was used for experiment performed at 20 °C and 70 °C. For tests at 120 °C the high temperature melted the beakers in the oven. As a replacement white paper cups were used with the disadvantage, that these the foaming behaviour could not be directly observed as they were not transparent. The third type was a beaker made out of polypropylene (PP-beaker, item number L322.220, Rausch Packaging a division of MEDEWO GmbH, Wallern, Austria), which was able to withstand the high temperature.

First, all beakers were labelled with 4 different numbers indicating the mixture used, for example 95-5-10-20. This means, that the mixture had an amount of 95 % isocyanate, 5 % polyol, 10 % water (related and additional to 100 % polyol) and reacted at a temperature of 20 °C.

Then the amount of polyol (Table 3) was filled slowly into the beaker, controlled by a balance (EWJ 3000-2, Kern & Sohn GmbH, Balingen, Germany) with an accuracy of 0.01 g. After this step the required water (de-ionised) was added to the polyol with a micropipette (accuracy of 1 µl, ALV-500, Variable Volume Pipette 20-200 µl, Advantage-Lab). The polyol-water components were mixed together in the beaker with a stirring rod for approximately 30 seconds. The beaker was placed back on the balance and the amount of isocyanate was filled into it. The next step was the mixing of the polyol-water mixture with the isocyanate. This was done with the same stirring rod like before and also about 30 seconds. The reaction of the components with each other could already be observed by first bubble formation. On the beaker, the filling level of the entire mixture was marked.

The stirring rod was removed and depending on the temperature the beaker was placed into an oven with a glass window (for 70 °C and 120 °C samples) or under a extractor hood (20 °C). After reaching the highest level of the polyurethane foam, the beaker was removed from the oven. If the highest level of the PUR foam didn’t reach the top of the beaker for the non transparent paper beakers, a second mark indicating the maximum level of foam expansion was made.

After these steps the beakers were evaluated visually and pictures were taken, the results are shown under chapter 3.1.

2.2.2 Gap filling test

Another factor for the experimental design is the thickness of the bond line to be filled by the foam, for which extreme values had to be determined. Furthermore an appropriate spread rate i.e. the quantity of the PUR adhesive applied for foam formation had to be selected. This value was also experimentally determined by the gap filling test. Therefore two boards out of spruce wood (picea abies), were glued together, one board had milled grooves with different depths and the opposite board had a smooth surface, as described in detail below. The wood was conditioned at standard atmosphere (20 °C and 65 % relative humidity) and the arrangement of the year rings of the boards were in the range of 30-90°.
The idea was to find out which PUR adhesive application quantity would result in a sufficient foam formation in predefined cavities. These cavities should simulate the thickness of the foam, which is important for the experimental design to obtain enough valid samples.

2.2.2.1 Experimental design and calculations

The first step was to determine the different depths of the grooves, which will be later the bond line thickness filled by the PUR foam. Therefore the values were set to 0.5 mm, and from 1 mm up to 5 mm, in steps of 1 mm. For selecting the applied quantity of the PUR adhesive, six different spread rates were used: 25, 50, 75, 100, 150 and 200 g/m². For the latter the amount of isocyanate was set to 85 %, polyol 15 % and 10 % (related to 100 % polyol) of additional water at a press temperature of 70 °C for all boards. For each quantity applied the calculated amounts in grams of isocyanate, polyol and water are displayed in Table 4.
The following example is for the applied quantity of PUR adhesive of 25 g/m². It was necessary to calculate the area to be bonded between the two boards, which was 0.0653 m² (0.128 m x 0.510 m). Selecting a spread rate of PUR adhesive of 25 g/m², the applied mass of the two components (isocyanate and polyol) was 1.63 g. 85 % of this value was isocyanate corresponding to 1.39 g and 15 % polyol (0.24 g). The amount of the additional water was 10 % from the 0.24 grams polyol, which was 0.024 g or 24 µl.

Table 4: Calculated amounts of isocyanate, polyol and additional water for each application quantity

applied quantity [g/m²]	**25**	**50**	**75**	**100**	**150**	**200**
length [m]	0.510	0.494	0.510	0.510	0.510	0.497
width [m]	0.128	0.129	0.128	0.129	0.129	0.129
surface area of the board [m²]	0.0653	0.0638	0.0653	0.0657	0.0656	0.0642
applied quantity on board without water [g]	1.63	3.19	4.90	6.57	9.85	12.83
isocyanate amount [g]	1.39	2.71	4.16	5.58	8.37	10.91
polyol amount [g]	0.24	0.48	0.73	0.99	1.48	1.92
amount water [g]	0.024	0.048	0.073	0.099	0.148	0.192
applied quantity on board including water [g]	**1.66**	**3.24**	**4.97**	**6.67**	**9.99**	**13.03**

The pressing time was calculated with the equation (1). In addition to the pressing time at 70 °C, which was 60 minutes, a heating-up factor for the boards was added. The boards had a thickness of 7.5 mm and with a heating-up time of 1 min/mm the whole pressing time was 67 minutes and 30 seconds.

$$pressing\ time = pressing\ time_{70°C} + 1\ min/mm\ [min] \tag{1}$$

2.2.2.2 Procedure

The boards were planed on both sides to a thickness of 7.5 mm, whereby six of them were used for the adhesive application and the other ones for milling the grooves into them. On each of these six boards, six grooves were milled into them with a depth of 0.5, 1, 2, 3, 4 and 5 mm (Figure 8), respectively. This was done with a computer numerically controlled machine (CNC) (HOMAG GmbH, Schopfloch, Germany) with a 10 mm milling cutter. The width of the grooves was 14 ± 1 mm according to the DIN EN 302-1 (2013) and the length was 110 mm. For programming, the software Woodwop (HOMAG GmbH, Schopfloch, Germany) was used.

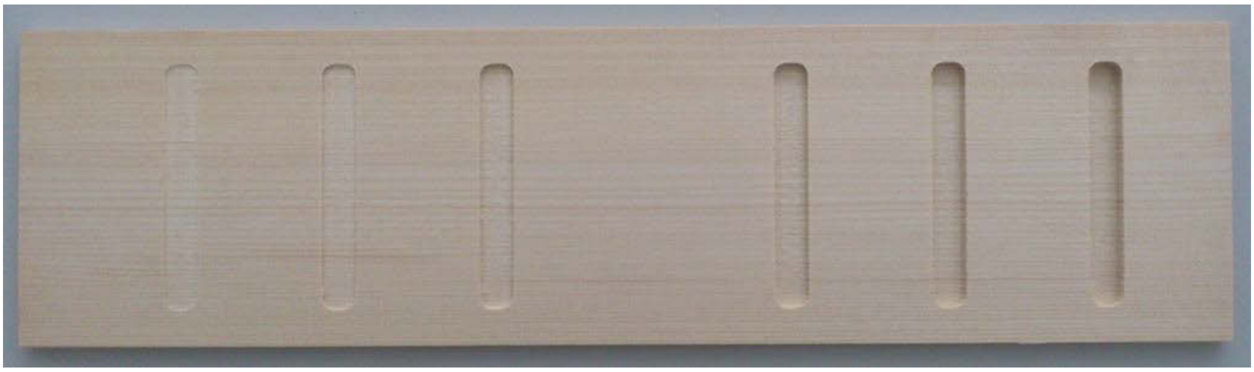


Figure 8: Board out of spruce with six grooves with a depth of 0.5, 1, 2, 3, 4 and 5 mm (from left to right) as used for the gap filling test

The application of the adhesive on the boards was done with a pneumatic applicator (Figure 9) (Sulzer Mixpac Ltd, Newbury, England).

Figure 9: Pneumatic applicator (Sulzer Mixpac, 2016)

Figure 10: 2 Components cartridges (Sulzer Mixpac, 2016)

This applicator had two cartridges with different filling capacities. In one of them the polyol-water mixture (which was mixed before in a beaker) was filled and in the other one the isocyanate. After closing the cartridges and putting them into the pneumatic applicator the maximum operating pressure was set to 10 bar. On the top of the cartridges an injector was mounted. Precise adhesive amounts as indicated in Table 4 were applied homogeneously onto the surface. After adhesive application joints were assembled within less than 5 min for all cases. The boards were pressed in two charges with a hydraulic hot press (Langzauner GmbH, Lambrechten, Austria) using a specific pressure of 0.8 MPa.

Before further evaluation all boards were stored for a minimum of four days at standard atmosphere (20 °C and 65 % relative humidity). All six boards were cut lengthwise, the cross sections were evaluated visually and pictures were taken, the results are shown in chapter 3.2.

2.3 General full factorial design

After evaluating the preliminary experiments, it was possible to create a general full factorial experimental design with five influencing factors, which lead through a cause and effect relations to the response values as shown in Figure 11 (Siebertz et al., 2010). Disturbance variables also affect these relationships, but it is very difficult to control these variables. For the later evaluation, it is important to know that these disturbance values were present, but they were not directly considered in the statistical analysis.

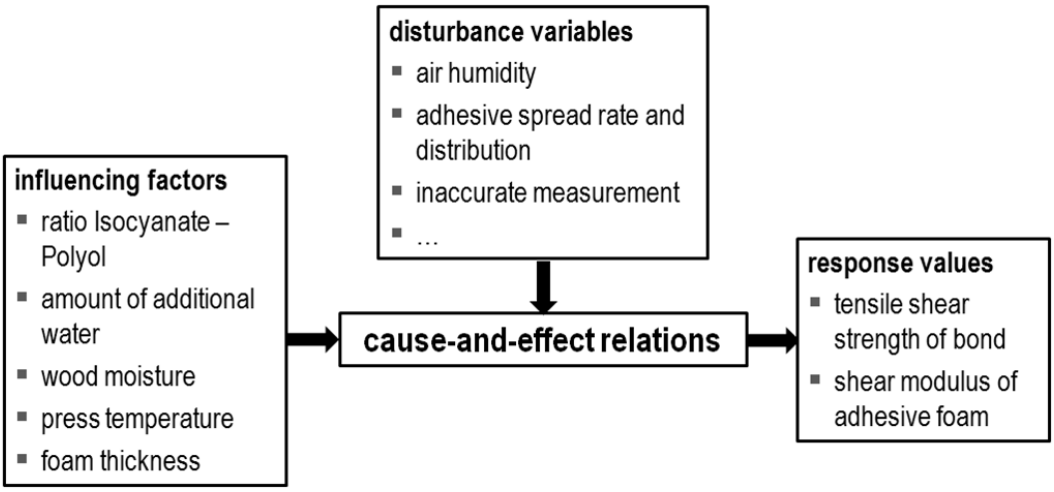


Figure 11: Cause and effect relations with influencing factors, disturbance variables and response values (own contribution)

2.3.1 Influencing factors

In this master thesis five influencing factors on the response values were investigated. Through the pretests, extreme values were identified for each factor. In Table 5, chosen values and the number of levels are shown for each factor. As the number of levels is not equal for all factors, a general full factorial design was used. For the ratio isocyanate-polyol only the component isocyanate is listed, because the sum of both is 100 %. Furthermore, the factor levels are symmetrical, to enhance the interpretation of the results (Lindner, 2013).

Table 5: Overview of the influencing factors with the number of levels and defined values

influencing factors	**number of levels**	**low**				**high**
amount of isocyanate [%]	5	60	68.75	77.5	86.25	95
amount of additional water [%]	3	1		5.5		10
wood moisture content [%]	3	3		12		21
press temperature [°C]	3	20		70		120
foam thickness [mm]	3	0.5		2.0		3.5

2.3.2 Response values

There are two response values, which are measured on the same lap shear sample, the tensile shear strength and the shear modulus. The calculations for these are shown in the next few points.

2.3.2.1 Calculation of the tensile shear strength

The tensile shear strength was measured according to the standard DIN EN 302-1 (2013) and is shown in equation (2).

$$tensile\ shear\ strength = \frac{maximum\ force}{overlapping\ area}\ [N/mm^2] \tag{2}$$

2.3.2.2 Calculation of the shear modulus

The determination of the shear modulus was carried out for the samples with a foam thickness of 0.5, 2 and 3.5 mm only. For the samples with no grooves an estimation of the shear modulus of the adhesive bond line was not performed as thickness of the pure adhesive bond line could not be determined in the required precision, in addition the contribution of the adherents to the total deformation was considered to be relatively high for this case. The shear modulus of the foam is obtained by the ratio of the shear stress ($\Delta\tau$) to the angular change (φ), as shown in equation (3). It is a measure of the resistance to a shear deformation and describes the elastic behaviour (Bargel & Schulze, 2008).

$$shear\ modulus\ of\ foam = \frac{\Delta\tau}{\varphi}\ [N/mm^2] \tag{3}$$

The shear stress ($\Delta\tau$) was calculated at a deformation of 0.25 % (τ_{high}) minus 0.05 % (τ_{low}) of the sample as shown in equation (4) (Abts, 2016). These values were read out by the testing software automatically and are used for calculation of mechanical parameters for synthetic materials.

$$\tau = \tau_{high} - \tau_{\text{low}}\ [N/mm^2] \tag{4}$$

For the angular change (φ) in equation (5), the change in position (Δl) and the foam thickness were needed as shown in the schematic Figure 12. The change in position was always 0.06 mm, because the shear stress ($\Delta\tau$) was determined at the same deformation values of 0.05 % and 0.25 %. The gauge length between the two sensor arms of the extensometer was 30 mm. By multiplying this value with 0.05 % (0.015 mm) and 0.25 % (0.075 mm) it is possible to calculate the difference between these two values, which is the change in position (Δl) with the value of 0.06 mm. Therefore, the angular change (φ) is the arcus tangent of the ratio change in length (Δl), which is 0.06 mm, to the foam thickness, which was measured before the testing with a vernier caliper with an accuracy of 0.01 mm.

$$\varphi = \tan^{-1}\left(\frac{0.06}{foam\ thickness}\right)\ [rad] \tag{5}$$

The change in position (Δl) value above includes not only the deformation of the PUR foam, but also the deformation of the two wooden parts within the 30 mm due to the tensile stress. For the samples with a foam thickness of 2 mm a maximum tensile shear strength of 1 N/mm² occurred. Based on the Hooke's law the strain of the wood can be calculated which is 0.003 mm (with the assumption the modulus of elasticity of wood is 10 GPa). Furthermore, by dividing this value with the total strain (2 %) within the 30 mm (0.6 mm), the error in measurement can be calculated, which is 0.5 %. At higher foam thickness this value gets lower due to the lower strength and the higher total strain value and for the lower foam thickness the error in measurements gets higher.

It is known that this error was included in the subsequent evaluation, but due to the low error values and a strong correlation between strength and shear modulus (Figure 37), this error was not considered further.

Figure 12 shows the schematic principle of the shear situation as assumed for the lap joint samples used. Through the application of a shear stress (τ) it comes to an angular change (φ) within the PUR foam and thus also to a change in length (Δl).

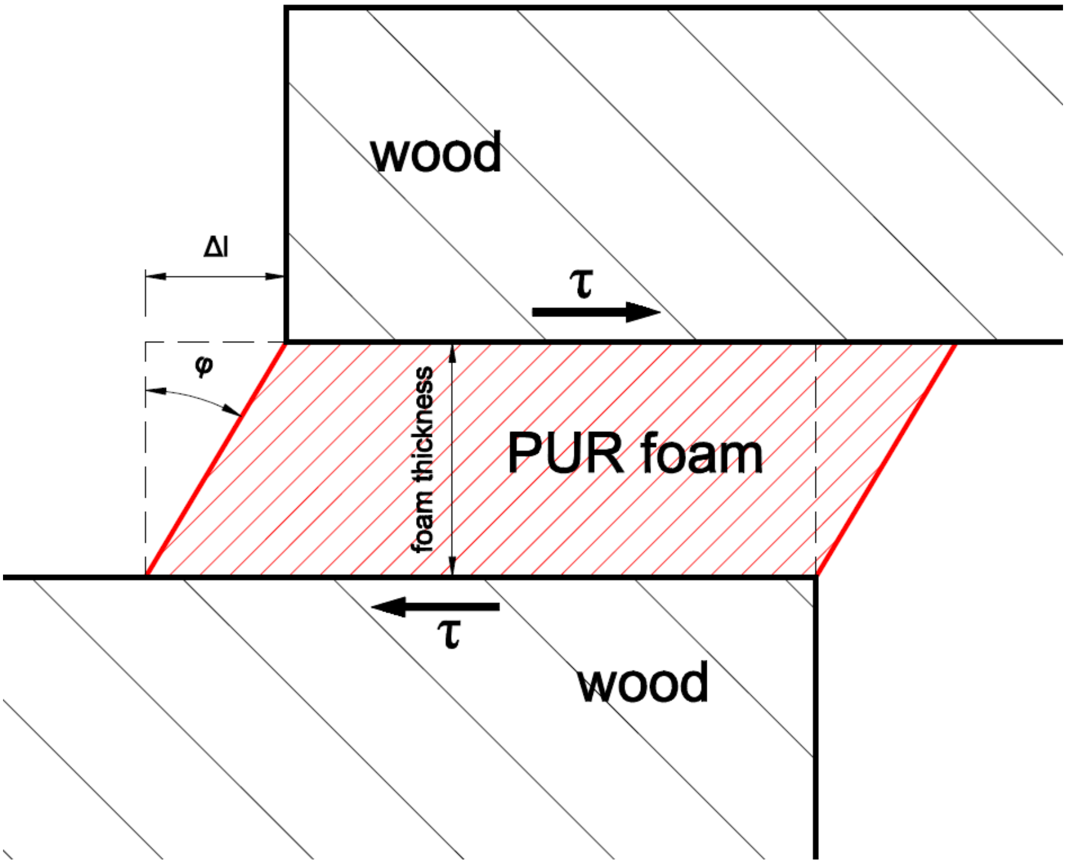


Figure 12: Schematic representation of the shear stress of PUR foam (own contribution)

2.3.3 Number of samples

By multiplying the number of levels for each factor in Table 5, the amount of possible combinations for this experimental design can be calculated and resulted in 540 different combinations. For each of them 8 samples were produced and tested. This means, that for the general full factorial experimental design, 4320 tensile lap shear samples were produced.

2.4 Preparation of lap shear samples

After the experimental design was defined, the sample preparation was started. The samples were based on the standard DIN EN 302-1 (2013) with some essential modifications, which will be described in the following points. Due to the high number of samples, it was important to work fast and accurate.

2.4.1 Wood species and density

According to the standard DIN EN 302-1 (2013) the samples should be made out of beech, but in this master thesis it was decided to produce all samples out of spruce wood (picea abies), which was supplied by the company Frischeis GmbH. Overall 16 poles with a thickness of 50 mm and a length of about four meters were used. These poles had a moisture content of 9 to 10 % which was analysed with a moisture meter Hydromette M 4050 (GANN Mess- und Regeltechnik GmbH, Gerlingen, Germany), which is based on the principle of the electrical resistance. The density of the wood was determined at a wood moisture content of 12 ± 1 %. Therefore the widths of 12 boards were measured using a vernier calliper with an accuracy of 0.01 mm and the length using a measuring tape with an accuracy of 1 mm. The length of the boards were about 500 mm and the width 120 mm. The thickness of the boards varied from 7 mm up to 9 mm. After that, the mass of the boards were determined with a balance, which had an accuracy of 0.01 g. By dividing the mass by volume the density of each board was calculated. The arithmetic mean of these 12 values was 0.38 g/cm³ with a standard deviation of 0.026 g/cm³, which is lower than the density for spruce wood according to Sell (1997) with the value of 0.45 g/cm³.

2.4.2 Preparation of the boards

First step was to cut the long poles in two pieces, which was done with a motor saw. Out of these, small boards with the dimensions of 500 x 120 x 50 mm were cut. This was done with a circular saw. In the next step these pieces of wood were cut with a band saw in boards with a thickness of 8 up to 12 mm. Depending on the target thickness of the grooves for the foam, either thinner or thicker boards had to be cut.

For each glued board 12 lap shear samples can be cut out, based on the cutting pattern in Figure 13. For this, two boards were necessary, the first one where the PUR adhesive was applied, which always had a thickness of 5 mm. The opposite board which had to include the grooves had various thicknesses depending on the depth of the grooves (Table 6) plus 5 mm remaining thickness in order to achieve a symmetric bond which is shown in Figure 24.

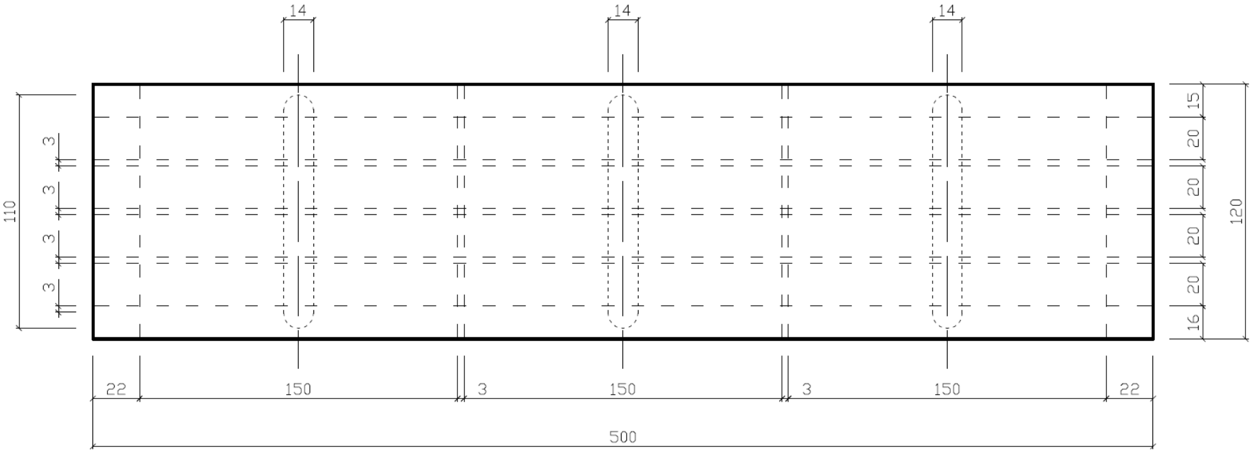


Figure 13: Cutting pattern for the tensile lap shear samples (dimensions in mm)

In chapter 2.3.3, the total number of samples has already been calculated. By dividing this value with the 12 samples for each board, 360 glued boards had to be produced. This means that 360 boards with a thickness of 5 mm were needed, on which the PUR adhesive were applied. For each of the opposite board with the different groove depths of 0, 0.5, 2 and 3.5 mm 90 boards were required. Through the variability of wood it was necessary to produce more boards than calculated. Table 6 lists the calculated and actually prepared number of boards, which are divided into the use and the final thicknesses of the boards.

Table 6: Overview of the calculated and prepared boards for the lap shear sample preparation, classify into the use and the final thicknesses of the boards

use of the board	**thickness of the board [mm]**	**calculated number of boards**	**prepared number of boards**
adhesive application	5.0	360	500
opposite board (groove depth 0 mm)	5.0	90	125
opposite board (groove depth 0.5 mm)	5.5	90	125
opposite board (groove depth 2.0 mm)	7.0	90	125
opposite board (groove depth 3.5 mm)	8.5	90	125

2.4.3 Conditioning of the boards

After cutting the boards, all of them were stacked to small packages. Between the boards, thin sticks (4 to 5 mm thickness) were used to improve the air circulation in order to achieve a homogeneous distribution of the desired wood moisture content. On the upper and lower side of the packages a 19 mm thick particleboard was placed, which was fixed with two lashing straps to reduce warping (Figure 15).

For this master thesis three different wood moisture contents were used. For each case one third of the boards were stored in the corresponding climate in order to reach the desired wood moisture content using a TIRA (Umweltsimulation GmbH, Schalkau, Germany) and a WEISS (Weiss Umwelttechnik GmbH, Reiskirchen-Lindenstruth, Germany) climate chamber.

In Table 7 the temperature and relative air humidity for the respective wood moisture content are shown. These values were determined with the help of a so called Keylwerth-diagram (Keylwerth & Noack, 1964).

Table 7: Values for temperature and relative air humidity for reaching a specific moisture content of the boards

Equilibrium wood moisture content [%]	**3**	**12**	**21**
temperature [°C]	40	20	30
relative air humidity [%]	14	65	92
description of storage	climate chamber TIRA	standard climate room	climate chamber WEISS

Figure 14: Conditioning of the boards with a moisture content of 3 %

Figure 14 shows the conditioning of the boards, which had to be dried to a moisture content of 3 %. Due to the small climate chamber it was necessary to dry the boards in three loads. When the moisture content of one load was reached, the gluing of these boards was started. Along with the drying it comes to a shrinking of the boards, therefore the lashing straps had to be tightened after some days.

To control the moisture content, the mass of one board for each package was measured over time. If the mass was almost constant, the real moisture content was measured using the oven-dry method according to the standard DIN EN 13183-1 (2002). Therefore a board was cut into 5 smaller pieces, which were weighted on a balance with an accuracy of 0.01 g (m_w). After that, the small wood pieces were given into an oven, which had a temperature of 103 ± 2 °C. The mass of the samples were measured over time and after reaching a constant mass they were placed in a desiccator. The samples cooled down at room temperature without the possibility of water absorption from the air humidity. After this, the mass of the samples was measured again (m_0). For the calculation of the moisture content of the boards the equation (6) was used.

$$\omega = \frac{m_w - m_0}{m_0} * 100\ [\%] \tag{6}$$

Only boards reaching a target moisture content with an accuracy of ± 1 % were used for further processing. In Figure 15 the conditioning of the boards with a moisture content of 21 % is shown. This climate chamber was big enough for all packages, which means only one load was needed. Due to the absorption of water it comes to a swelling of the boards, therefore the lashing straps were made a little looser after some days.

Figure 15: Conditioning of the boards with a moisture content of 21 %

To control the wood moisture the moisture meter Hydromette M 4050 was used, because this method provides instantaneous results instead of the oven-dry method. According to the standard DIN EN 13183-2 (2002) the electrodes were pulled into the wood transverse to the direction of the fibres. The temperature and the wood species were adjusted and after that, the moisture content of the wood could be read. After reaching a wood moisture of 21 ± 1% the boards were ready for the next steps. Also the boards with a 12 % moisture content were measured with the moisture meter.

2.4.4 Planing and milling the grooves in the boards

Before the boards were glued together, they had to be planed with a planing machine. First step was to plane 4 boards to a thickness of 5 mm, which were used for the adhesive application. Both sides of the boards were smoothened. After this, another 4 boards were planed to a thickness which depends on the depth of the grooves (Table 8).

Table 8: Thicknesses of the boards depending on the depth of the groove

use of the board	groove depth [mm]	thickness of the board [mm]
adhesive application	-	5.0
opposite board	0	5.0
opposite board	0.5	5.5
opposite board	2.0	7.0
opposite board	3.5	8.5

Next step was the milling of the grooves into the board, which was done using the same computer numerically controlled machine (CNC) with a 10 mm milling cutter as described in chapter 2.2.2.2. The width of 14 ± 1 mm of the grooves was machined according to DIN EN 302-1 (2013) and the length was 110 mm. For programming, the software Woodwop was used as described already in chapter 2.2.2.2. In order to guarantee that the boards are always on the same place on the CNC work station, a cover board (Figure 16) was used. Two planed spruce boards could be processed at the same time, so two passes were needed. Therefore the boards were placed onto the fibre board and a vacuum was generated, which held the boards during milling of the grooves according to the dimensions in Figure 13.

Figure 17 shows two finished milled boards, which are ready for the gluing and pressing. The grooves are not continuous to keep the adhesive between the two boards, which are glued together in the next step.

Figure 16: Cover board to protect the CNC work table

Figure 17: Finished milled boards for further processing

2.4.5 Gluing and pressing

Each surface of the boards for the adhesive application were divided into three areas. The reason for this was, that on each board three different mixtures of adhesive were applied. Every sector on the front- and the backside of each board got labelled with the combination number, which was stored in a database. With this number a subsequent assignment of the samples was possible.

Figure 18: 5 mm thick boards for the adhesive application

Due to the aim of providing eight samples for each combination, two different boards were labelled with the same numbers (Figure 18). This means that two replicates with each four samples (repeats) were produced.

For the preparing of the adhesive mixtures for one press cycle six beakers with a filling capacity of 200 ml were labelled with the same combination numbers as on the boards. By the combination number, the corresponding amounts of the components (Table 9) could be added to the beaker based on the same principle as in chapter 2.2.1.2. After mixing the components together, 3 g of the adhesive were applied on the first sector of the first board. Due to the preliminary gap filling test described in chapter 2.2.2 the applied quantity of the PUR adhesive of 150 g/m² was applied for all further tests. Each sector of a board had a dimension of 0.120 x 0.167 m. By multiplying this area with the 150 g/m² the amount of PUR adhesive for one sector can be calculated, which is 3 grams.

To prevent a premature reaction of the components they were mixed together immediately before the application. The same amount of adhesive was applied on the first and second board again. This process was done with all beakers.

Table 9: Required amounts of the components for the PUR adhesive preparation

isocyanate		polyol		additional water [g]		
[%]	**[g]**	**[%]**	**[g]**	**1 %**	**5.5 %**	**10 %**
60.00	7.20	**40.00**	4.80	0.048	0.264	0.480
68.75	8.25	**31.25**	3.75	0.038	0.206	0.375
77.50	9.30	**22.50**	2.70	0.027	0.149	0.270
86.25	10.35	**13.75**	1.65	0.017	0.091	0.165
95.00	11.40	**5.00**	0.60	0.006	0.033	0.060

In Table 9 the required amounts of the components for the PUR adhesive preparation are shown. For isocyanate and polyol there are 5 levels expressed as percentage and gram. The sums of these are always 100 % respectively 12 grams. The component additional water had 3 levels, which are listed also in percentage and grams wherein the amount of water was dependent on the amount of polyol. For example for a 77.5 % isocyanate – 22.5 % polyol – 5.5 % water – mixture, 9.3 g isocyanate, 2.7 g polyol and 0.149 g water were needed. From this mixture 3 g were applied on each sector of the board.

After this, the adhesive was distributed with a spatula so that the surface of each sector was covered with the adhesive. After each sector the spatula was cleaned with a paper towel to prevent a mixing of the adhesive from the individual sectors.

Figure 19: Distribution of the adhesive for each sector on the board

In Figure 19 the distribution of the adhesive is shown in which on the left side the adhesive is already distributed and on the right side not. A problem was here that the components react with each other within a few minutes, which means that a fast and accurate working was essential.

In the next step the opposite boards with the grooves were placed onto the boards with the adhesive. According to the standard DIN EN 302-1 (2013) the arrangement of the year rings of the two boards had to be in the range of 30-90°. Immediately thereafter the two or four pairs of boards were given into the press. Sticking of the samples towards the hotplates was prevented by using baking paper. The hotplates were heated up previously to the desired temperature of either 70 °C or 120 °C. The program for pressing was started and the press closed. After reaching the specific pressure of 0.8 MPa, the pressing time started. The press opened automatically when the time was elapsed.

After the first hot pressing cycles using a temperature of 120 °C combined with samples showing a wood moisture content of 21 % significant plastic deformation (compression) of the wood was observed. The reason for that was that through the high moisture content and the high temperature it comes to a reduction of the compressive strength of the wood.

As reviewed in Kollmann (1951) the compressive strength of the wood will decrease with a higher moisture content of the wood. Between a moisture content of ~8 and ~20 %, there is almost a linear relationship. For the compressive strength perpendicular to the fibre direction there is a 5 %

reduction in the compressive strength per a 1 % higher moisture content (Hoffmeyer, 1995).

The temperature also has a major impact on the compressive strength of the wood. With a higher temperature the compressive strength will decrease. As already reported by Glos & Henrici (1990) there is a loss of compressive strength of 5 % with a temperature increase of 10 °C. This is for softwood free from defects at a moisture content of 10 to 15 %. The compressive strength is reduced about 44 % (Blaß & Sandhaas, 2016) or 49 % (Glos & Henrici, 1990) at 100 °C compared to 20 °C.

Due to this plastic deformation of the wood, the specific pressure was reduced to 0.3 MPa for boards hot pressed at 120 °C.

In Table 10 the pressing times for each press temperature are shown. The total pressing time is calculated with the equation (1) shown in chapter 2.2.2.1 and depends on the press temperature and the thickness of the two boards which are glued together. For higher temperatures a heating-up time of 1 min/mm thickness was used.

Table 10: Pressing time for each press temperature

press temperature [°C]	**20**	**70**	**120**
pressing time $_{\text{temperature}}$ [min]	180	60	30
heating-up time [min/mm]	-	1	1

For the determining of the pressing time at a press temperature of 120 °C a beaker test was done.

Therefore two beakers with a filling capacity of 200 ml with different adhesive mixtures were used. The first beaker had a mixing ratio of 85 % isocyanate, 15 % polyol and 10 % additional water, which were mixed together based on the same principle as in chapter 2.2.1.2. Then the beaker was put into an oven with a glass window which was preheated to 120 °C (Figure 20). There is also a wood stick with a one-centimetre scale on it for documentation the foam formation at 120 °C. After 6 minutes the foam reached the top of the beaker and after 7 minutes and 10 seconds it reached the maximum which was 1 cm above the beaker. After a time of 15 minutes the surface of the foam is elastic but not sticky. 20 minutes after putting the beaker into the oven, the surface of the foam is still elastic but it is harder than before. The beaker was given out of the oven and after a storage at room temperature for about 10 minutes the surface of the PUR foam got rigid. Also a shrinkage of the foam of 2 cm was observed.

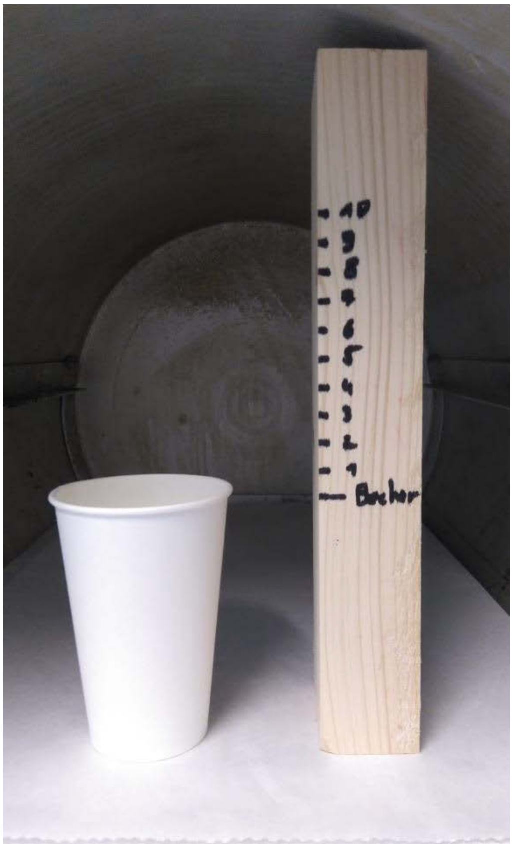


Figure 20: Observation of the foam formation at 120 °C

The same procedure was done with another mixture of the components. Therefore a mixture with 60 % (8.4 g) isocyanate, 40 % (5.6 g) polyol and 10 % (0.56 g) water was used. The reaction and the foam formation was quite faster than with the other mixture before. After 2 minutes and 20 seconds the foam reached the top of the beaker. With a total time of 3 minutes and 10 seconds the foam formation reached the maximum height of 5 cm above the beaker. After a time of 15 minutes the surface of the foam is elastic but not sticky anymore. The surface was slightly harder after 20 minutes but still elastic. After a storage of the beaker at room temperature the foam was still elastic. With these results a pressing time of 30 minutes at a press temperature of 120 °C was determined, as shown in Table 10.

After the pressing time the press opened automatically. For the boards which were pressed at 120 °C another problem occurred.

Due to the high temperature and therefore to a water vapour formation the bond line was not strong enough to hold the two boards together (Figure 21). Therefore after reaching the pressing time, the two hotplates were cooled down at a temperature of 70 °C. As soon as both hotplates reached this temperature, the press opened automatically. With this method separating of the boards from each other could be avoided.

Figure 21: Glued boards were separated from each other due to a water vapour formation

After pressing the boards together they were stored at standard climate (20 °C, 65 % relative air humidity) before further processing/testing as shown in Figure 22.

These boards were stacked by using 4 mm thick sticks between them to improve the air circulation and therefore to a faster conditioning of the boards. There were also given two poles on top of them to reduce warping.

After 3 weeks of storage the first glued boards were cut into tensile shear samples according to the standard DIN EN 302-1 (2013). Therefore the cutting pattern in Figure 13 was used.

Figure 22: Glued boards for conditioning in the standard climate

In Figure 23, a side view of a tensile shear sample is shown. The red line between the two boards is visualizing the bond line. In the middle of the sample there is the groove which should be filled out with the PUR foam. On the left and right side of the groove a cut with a circular saw was made. Thus the overlapping area consisted only of the PUR foam which assures that this material is accessed by the testing. The width of each sample was 20 mm according to DIN EN 302-1 (2013).

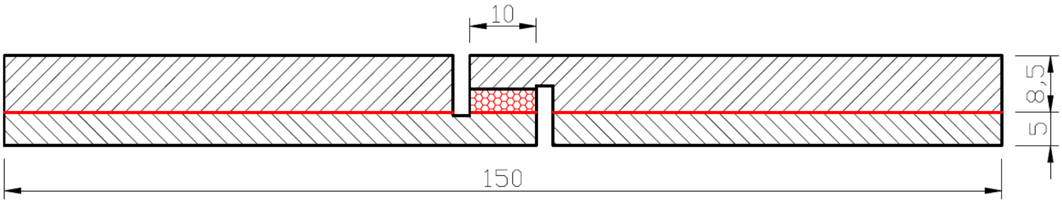


Figure 23: Side view of a tensile shear sample with a 3.5 mm adhesive foam thickness within the overlapping area (dimensions in mm)

All samples were labelled and stored in the standard climate again to make sure that all of them will be tested at the same wood moisture content.

2.5 Lap shear test

The tensile lap shear test was carried out using a universal testing machine Z100 (Zwick GmbH & Co. KG, Ulm, Germany) and was performed according to the standard DIN EN 302-1 (2013). The strain measurement was carried out with a makroXtens extensometer which was also provided by Zwick. For measuring the force, a load cell with a maximum load capacity of 5 kN was used.

The gauge length between the two sensor arms of the extensometer was 30 mm (Figure 24). For all samples the distance between the specimen grips was set to 70 mm. Due to the high variability of the strength and stiffness of the samples with different foam thicknesses the pre-force, the speed to reach the pre-force and the testing speed were varied, which is shown in Table 11. With the aim that the duration of the lap shear test is in the range of 30-90 seconds according to DIN EN 302-1 (2013), the testing speed had to be adapted.

Figure 24: Tensile shear sample during testing with a foam thickness of 3.5 mm

Table 11: Overview of the settings for the testing machine

foam thickness [mm]	**0**	**0.5**	**2.0**	**3.5**
pre-force [MPa]	0.05	0.05	0.025	0.01
pre-force speed [mm/min]	5	2	2	2
testing speed [mm/min]	1.5	1.0	1.5	1.5

The measured data from the testing machine were processed with the software Microsoft Excel 2007 and the response values were calculated according to the equations in chapter 2.3.2.

All data was further evaluated with two different software packages. For statistical analyses of tested data the software package SPSS (IBM Corp., IBM SPSS Statistics for Windows, Version 22, Armonk, New York) was used. As second software package Design-Expert (Stat-Ease Inc., Version 10 Free Trial, Minneapolis, Minnesota) was used for creating mathematical models based on a statistical analysis. For analysing the data of the samples with foam formation, the arithmetic mean of four samples (repeats), that were produced out of one glued board, were calculated first. This had to be done twice (replicates) to obtain two mean values out of eight tensile lap shear samples.

3 Results and Discussion

3.1 Beaker test

Figure 26 shows the cured polyurethane foam at various temperatures. At a temperature of 20 °C it is observed that with a higher amount of isocyanate (from right to left) a lower foam formation occurs. There is also a strong difference in the foam formation with the different amounts of additional water. With a higher content of additional water an increase in foam formation takes place. After a few days of storage the foam of the beakers with a low isocyanate (high polyol) content felt flexible, instead the foam types with higher isocyanate content felt stiffer. The foam bubble distribution was homogeneous and the diameter of the bubbles looked almost the same for the 20 °C beakers.

At a temperature of 70 °C and with a high content of isocyanate (95 %) there is a low foam formation and there is also an abundance of isocyanate which did not react with the polyol/water mixture. As before it comes to a better foam formation with a higher content of additional water. The distribution of the foam bubbles was homogeneous but there was a difference in the diameter of the bubbles. This may be due a faster chemical reaction as a result of the higher temperature and therefore more CO_2 is formed in shorter time. Moreland et al. (1994) report due to the increasing temperature, the gas pressure also increases and this leads to larger bubbles. After a few days of storage a shrinkage of the foam with a lower isocyanate content could be observed.

A reason for that could be that the CO_2 in the bubbles escapes and due to the lower stiffness of the foam it collapses. At a temperature of 120 °C in Figure 26, the most foam formation could be observed with a low isocyanate content, but this foam was very flexible. The distribution and diameters of the foam bubbles varied due to the high temperature as shown in Figure 25 with a mixture of 85 % isocyanate, 15 % polyol and 10 % additional water content.

Figure 25: Inhomogeneous polyurethane foam after the reaction at 120 °C (top view on beaker)

Figure 26: Results of the beaker test at various temperatures

With the results of the beaker test, it was possible to find the extreme values for the factors isocyanate, additional water and temperature for the general full factorial design (Table 5). For the amount of isocyanate as minimum value 60 % and for the highest amount 95 % were selected. At both of these levels a sufficient foam formation could be observed. The amount of the additional water was 0.2 % and 10 % for the beaker test. For the general full factorial design, the minimum was set to 1 % instead of 0.2 %, since a better dosage of the amount of additional water could be achieved. All three temperatures resulted in foam formation and therefore these values were taken for the general full factorial design.

3.2 Gap filling test

In Figure 27 the cross section of the glued boards for the gap filling test are shown. From left to the right the groove depth gets higher and from top downwards the applied quantity of PUR adhesive increases. With a higher amount of PUR adhesive it is possible to fill cavities with a higher volume.

From 25 up to 75 g/m² applied quantity of the adhesive only the grooves with a depth of 0.5 mm are filled with the PUR foam, whereas the other ones are empty. With 100 g/m² adhesive most of the cavities are not completely filled with the foam. From an application quantity of 150 g/m², a sufficient filling of the cavities with the PUR foam could be observed.

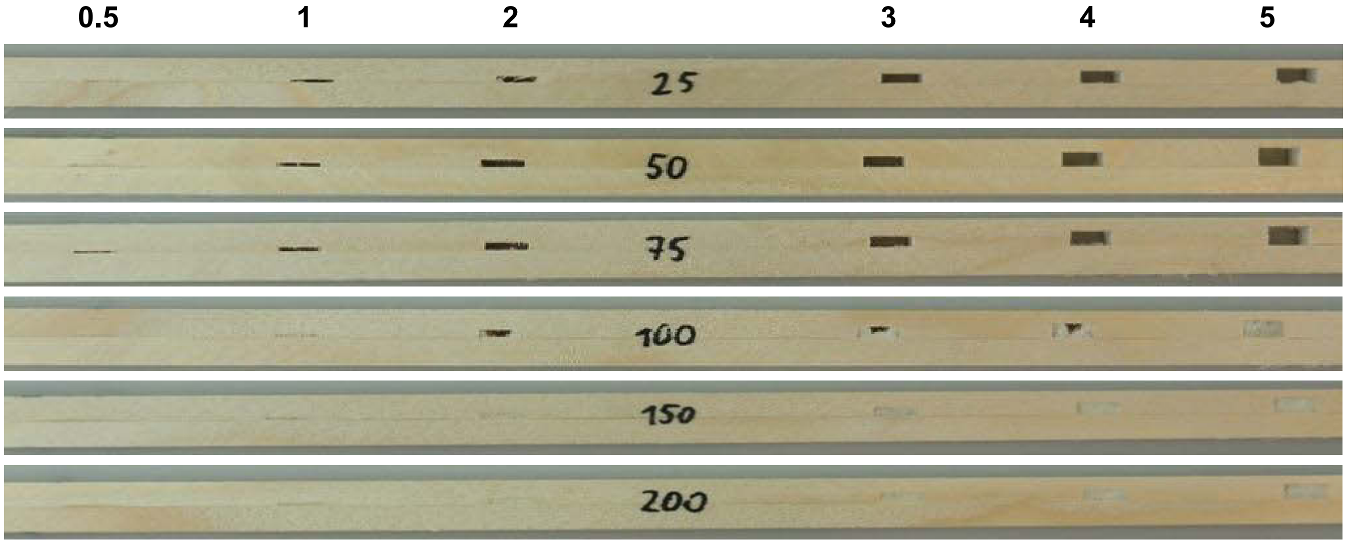


Figure 27: Results of the gap filling test (from left to right = groove depths [mm]; top down = applied quantity of PUR adhesive [g/m²])

Based on this pretest it was decided to apply 150 g/m² of the PUR adhesive for further tests on foam formation. At this value all cavities are filled with foam. Anyhow, it has to be considered that this test was done with a mixture of the components which reacted very good. To increase the probability of having sufficient valid samples for the general full factorial design testing, the maximum groove depth was limited to 3.5 mm whereas the minimum was kept at 0.5 mm.

3.3 Lap-joint tests using a foam-bonding

In the next few points the results of the tensile shear samples with a foam thickness of 0.5, 2 and 3.5 mm will be shown. For both of the two response values i.e. tensile shear strength as well as shear modulus, the results were displayed using mainly boxplot diagrams with SPSS followed by a statistical analysis and a model based description of the results using the software Design-Expert.

3.3.1 Tensile shear strength

3.3.1.1 Results of the measured values (SPSS)

Within the box from the boxplot there are 50 % of the measured samples which is called interquartile range (IQR). Between the 25 % percentile and the 75 % percentile there is the median which is the 50 % percentile. Below and above of the median there are 50 % of the measured samples. The whiskers have a maximum length of 1.5 times the IQR in which the end of the whiskers is always the last measured sample within this distance. This is the reason why the whiskers are different in length. Outside of the whiskers, possible outliers are displayed. There are two different types of outliers, the “mild” outlier (small circle) is between 1.5 to 3 times the IQR and the “extreme” outlier (small star) is any value more than 3 times the IQR.

In Figure 28 the tensile shear strength of all valid samples with a foam thickness of 0.5 mm are shown. A differentiation of the three wood moisture contents used is made (x-axis) as well as a subdivision of the three press temperatures with different colours. There is a wide scattering for each boxplot due to the inclusion of all types of mixtures.

As a general trend with increasing wood moisture content an increasing strength can be observed. There is also a considerable drop in strength as the press temperature increases.

Visible in Figure 28 the highest tensile shear strength has been reached with a press temperature of 20 °C (blue boxplots). The highest median is around 3.5 N/mm² at a wood moisture of 21 % and a press temperature of 20 °C, whereas the minimum median is approximately 0.5 N/mm² at a low wood moisture content (3 %) and a high press temperature (120 °C).

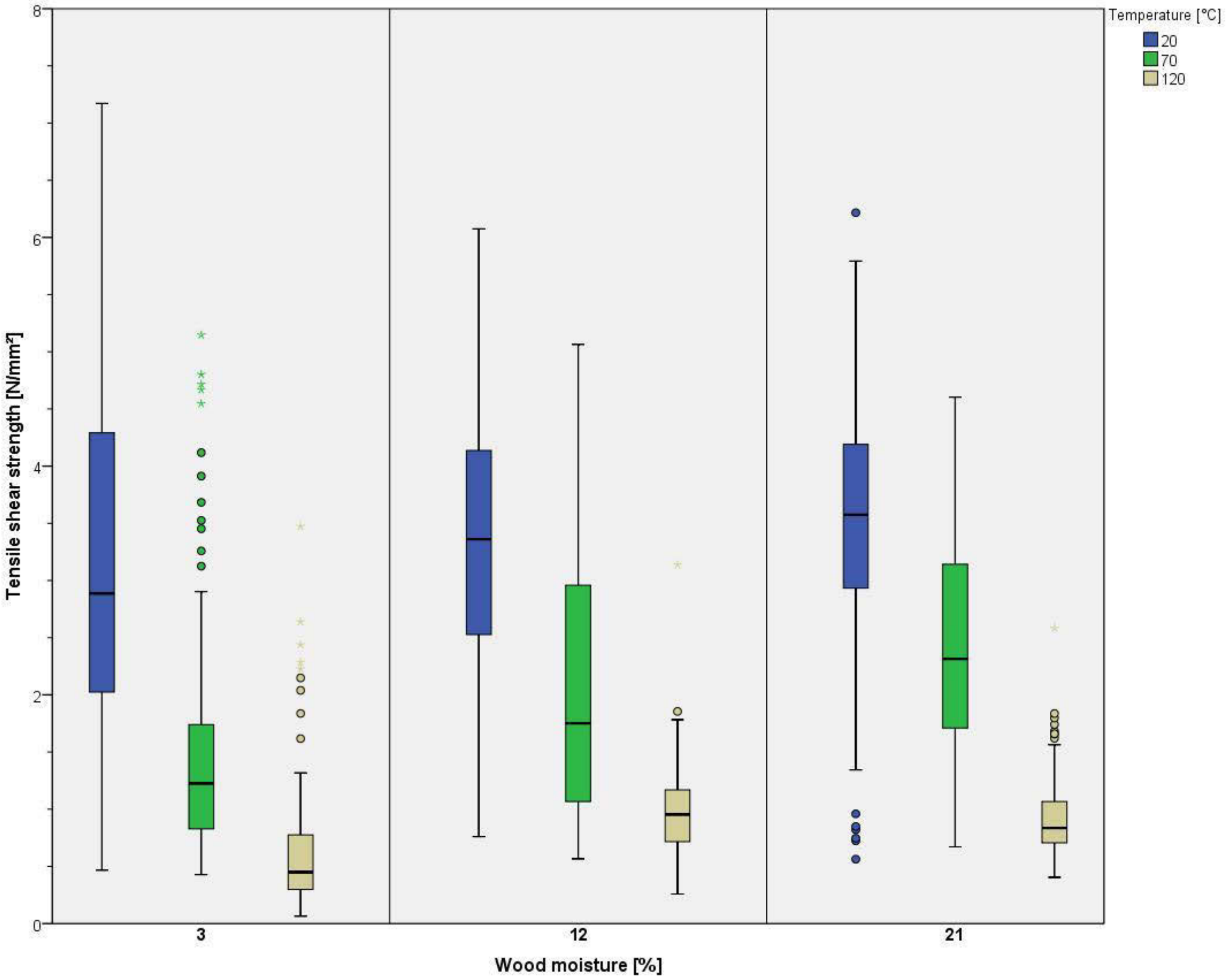


Figure 28: Tensile shear strength of samples with a foam thickness of 0.5 mm (max. n=120 samples per boxplot)

As referred in Thompson et al. (2003) the shear strength of a rigid PUR foam ranges between 0.7 N/mm² and 4.2 N/mm². Therefore it can be said that the samples with a foam thickness of 0.5 mm are in the range of a rigid or semi-rigid PUR foam. However, these reference values represent the strength of very homogeneous foams, whereas this was not always the case in the present experiment.

A reason for the lower strengths at higher temperatures could be, that the polyurethane bubbles burst due to the fast reaction triggered by the high temperature, which was observed already during the beaker tests. At lower temperature there is a slower reaction of the components and therefore a slower foam formation, which could be the reason for getting bubbles with smaller and more homogenous diameters. Due to the high temperature it also could be that the wood components like carbohydrates, amino acids, fats, resin or terpene migrate to the surface of the wood and influence the foam properties by reacting with the adhesive components. According to the statements of the adhesive manufacturer (Henkel, 2017) the isocyanate reacts faster with the available water than with the polyol in the case of higher temperatures. Therefore it is assumed that some amounts of polyol do not react with the isocyanate and remains in the bond line/foam. There is a difference of the speed of the reaction between primary OH groups and secondary OH groups, which could be present in the polyol, water or wood substance. At same conditions, primary OH groups react two to three times faster than secondary OH groups, whereby water is considered to consist of primary OH group (Pizzi & Mittal, 2003) whereas possible available OH groups of the polyol and the wood are considered to be of secondary type.

Another fact visible in Figure 28 is, that the scattering of the results visible in the IQR of the samples with a high temperature (120 °C) is very small in contrast to the one at lower temperatures.

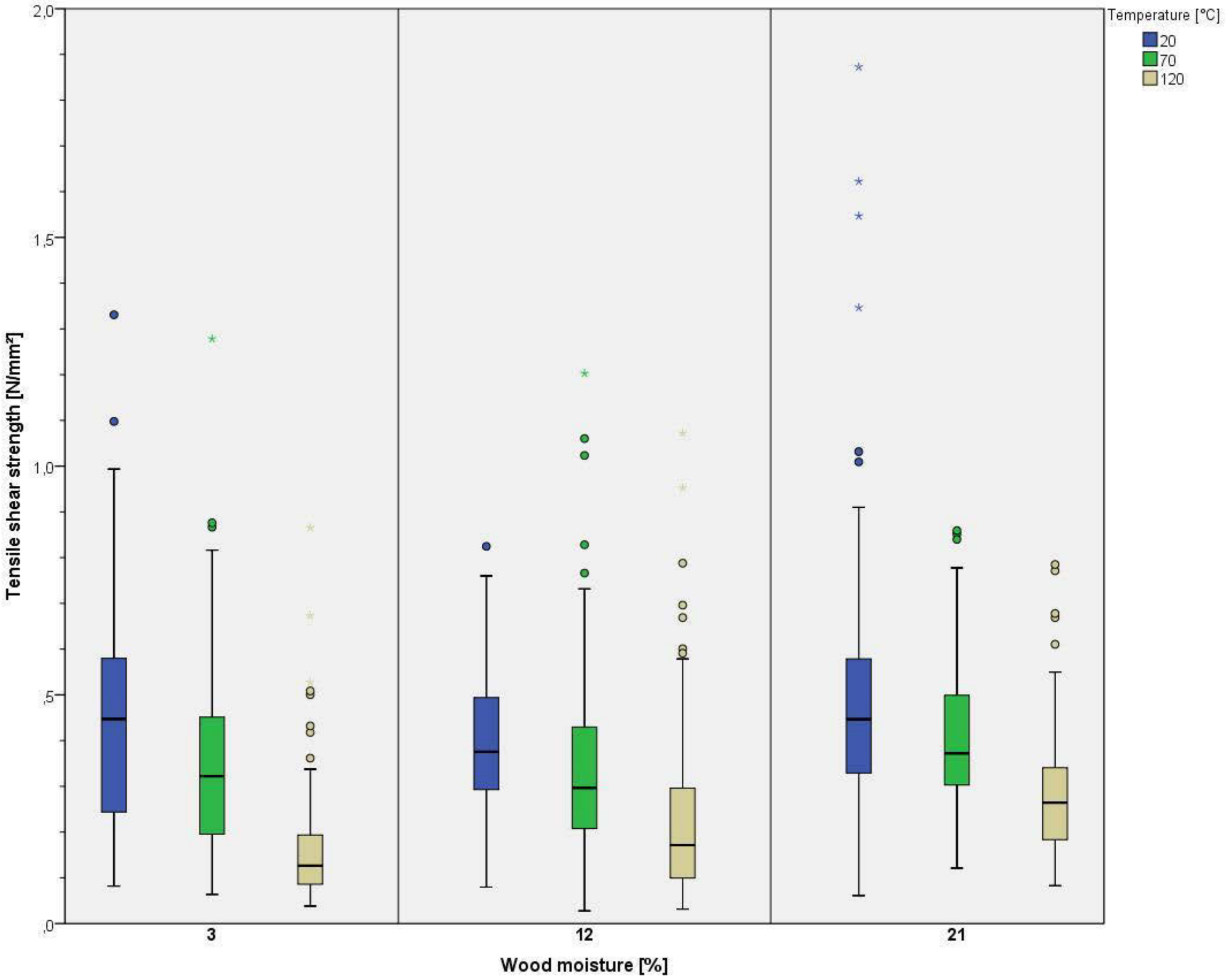


Figure 29: Tensile shear strength of samples with a foam thickness of 2 mm (max. n=120 samples per boxplot)

Figure 29 displays the tensile shear strength of the samples with a foam thickness of 2 mm. Compared to a foam thickness of 0.5 mm the observed strength values are on a significantly lower level, whereby the maximum median of the strength is about 0.45 N/mm² at a temperature of 20 °C and a wood moisture content of 3 % or rather 21 %. The lowest median is about 0.13 N/mm² at a temperature of 120 °C and a wood moisture content of 3 %. One reason for the significantly lower strengths could be that a larger volume should be filled by the same amount of applied PUR adhesive of 150 g/m². Similar to the observations made at 0.5 mm the tensile shear strength decreases with increasing temperature, but it seems that this influence is much lower compared to a foam thickness of 0.5 mm. A distinct increase in strength with increasing wood moisture content cannot be observed, nevertheless the medians of the boxplots with a wood moisture content of 21 % are the highest ones.

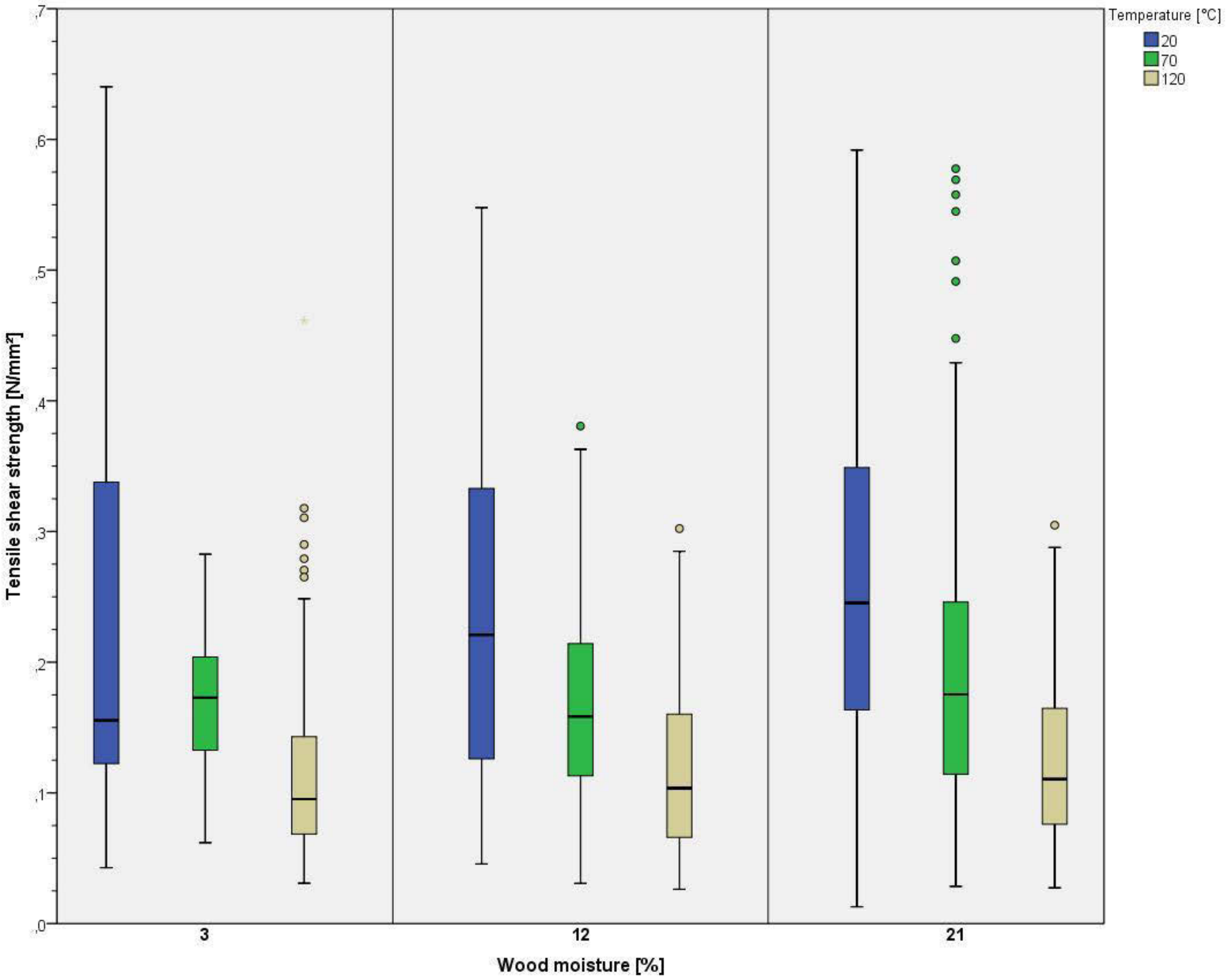
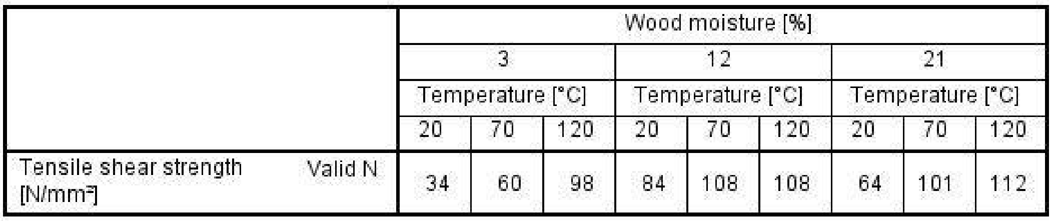


Figure 30: Tensile shear strength of samples with a foam thickness of 3.5 mm (max. n=120 samples per boxplot)

Figure 30 shows the results of the tensile shear strength on the samples with a foam thickness of 3.5 mm. The maximum number of valid samples for each boxplot is 120, but due to the higher foam thickness, a higher number of invalid samples occurred. These invalid samples are evaluated more detailed in chapter 3.3.4. In Table 12 the valid number of samples for each wood moisture content and temperature are listed.

Table 12: Valid number of samples at a foam thickness of 3.5 mm



		Wood moisture [%]								
		3			12			21		
		Temperature [°C]			Temperature [°C]			Temperature [°C]		
		20	70	120	20	70	120	20	70	120
Tensile shear strength [N/mm²]	Valid N	34	60	98	84	108	108	64	101	112

The highest tensile shear strength is around 0.25 N/mm² at a wood moisture content of 21 % and a temperature of 20 °C.

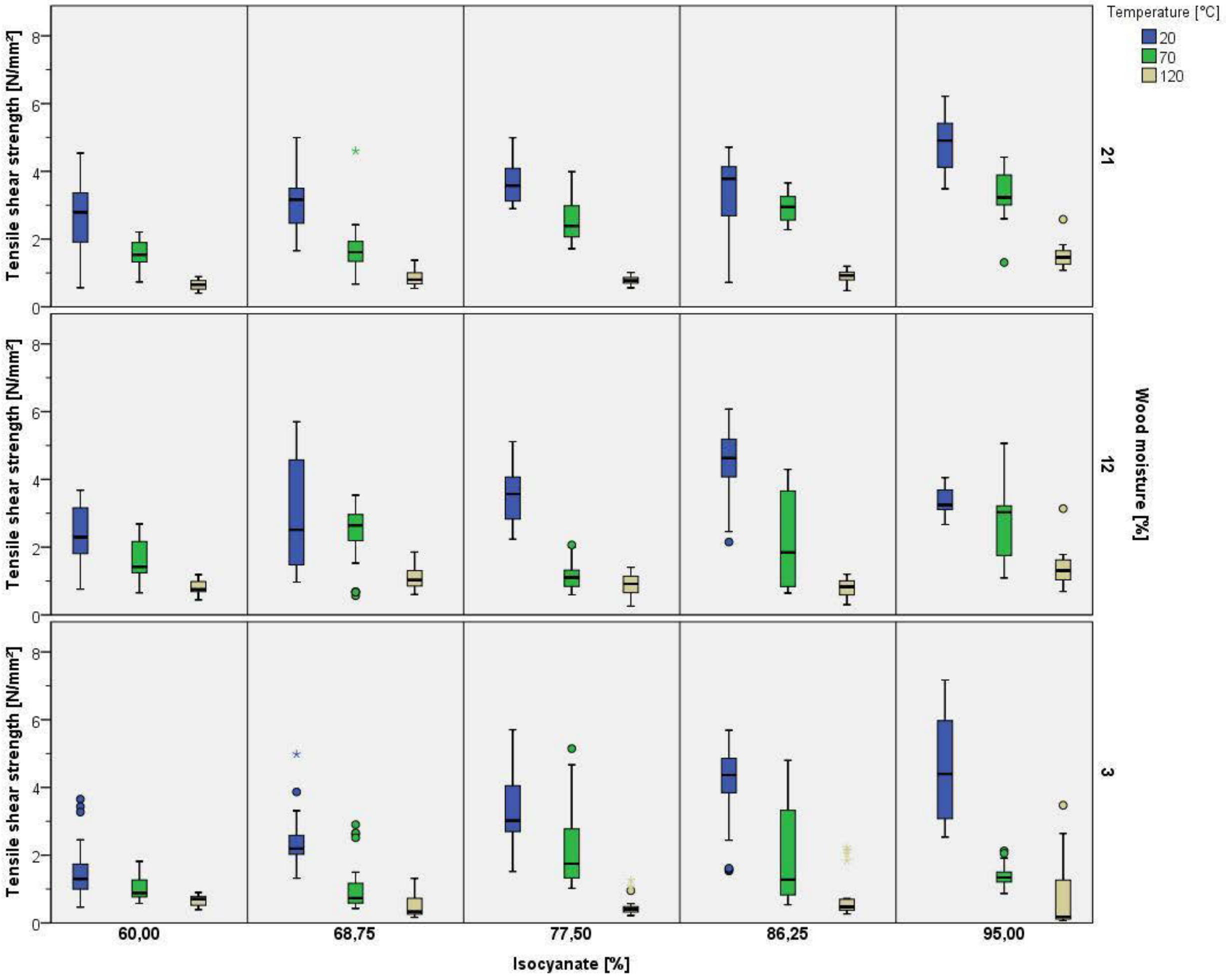


Figure 31: Tensile shear strength of samples with a foam thickness of 0.5 mm (max. n=24 samples per boxplot)

In Figure 31 again the results of the tensile shear strength of samples with a foam thickness of 0.5 mm are shown. This time more detailed with 5 levels of isocyanate content on the x-axis. As already shown in Figure 28, where the highest strength values were reached with a low press temperature, Figure 31 shows that there is also a strong influence on the strength as a result of the ratio of isocyanate to polyol. With a higher amount of isocyanate, the strength increases at a low press temperature (20 °C). This can also be partly observed at a temperature of 70 °C, whereas at a very high press temperature (120 °C) there is no evident influence of the amount of isocyanate on the tensile shear strength. The highest median can be observed at a wood moisture content of 21 %, 20 °C press temperature and an amount of isocyanate of 95 %. Similar detailed results of the samples with a foam thickness of 2 mm and 3.5 mm are listed in Figure 50 and Figure 51 in the appendix.

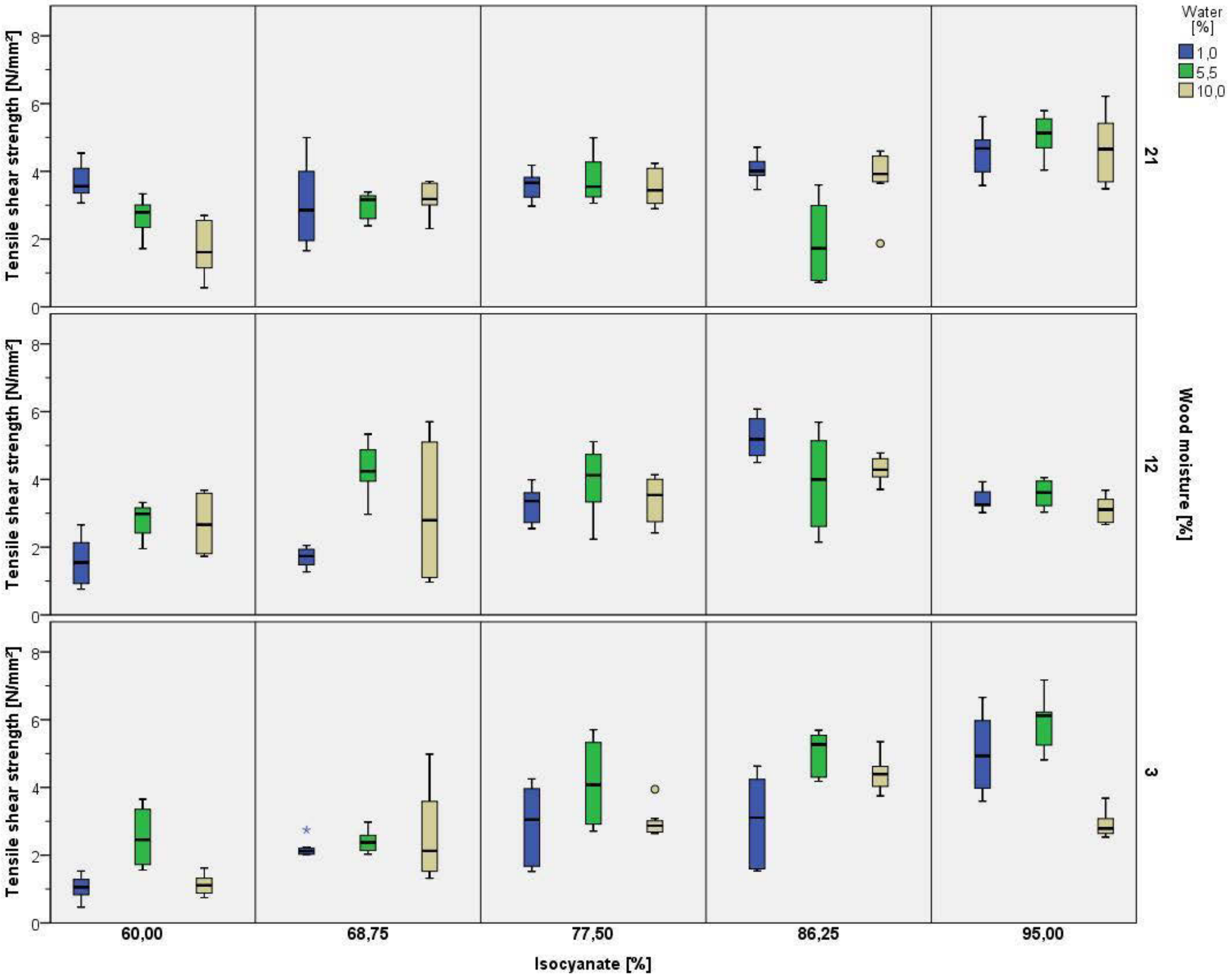


Figure 32: Tensile shear strength of samples with a foam thickness of 0.5 mm at a press temperature of 20 °C (max. n=8 samples per boxplot)

In Figure 32 the results of the tensile shear strength at a press temperature of 20 °C and a foam thickness of 0.5 mm are shown. Therefore the diagram is subdivided into the five isocyanate groups, three wood moisture contents and three amounts of additional water (colour). In most cases the medians of the samples with an additional water amount of 5.5 % tend to be highest, but there is no obvious correlation between the amounts of the additional water and the strength. The results for the 2 mm and 3.5 mm samples at a temperature of 20 °C are again shown in the appendix (Figure 52 and Figure 53). Additionally, the results of the tensile shear strength with all influencing factors for each foam thickness are displayed in the appendix (Figure 54 to Figure 56).

3.3.1.2 Results of mathematical models based on a statistical analysis (Design-Expert)

The statistical analysis was done with the software Design-Expert, therefore the data was evaluated by an ANOVA (analysis of variance) with a significance level of 1 % as well as by a multiple regression analysis. Due to the high ratio of the maximum tensile shear strength (6.434 N/mm² - red colour in the 3D-diagram) to the minimum (0.036 N/mm² - blue colour in the 3D-diagram), the software suggested a transformation of the data. Thus a natural logarithm transformation of the data was performed. In the next step the software suggested on the basis of the predicted coefficient of determination (R²) a quadratic model. Selecting of the factors for the model was done by the software automatically with a significance level of 1 % by a backward selection. After this the normal distribution of the residuals, the presence of a constant error and outliers were checked. Altogether there were 63 values (of maximum 810 values) which could not be included in the model due to missing values or outliers.

The coefficient of determination (R²) had a value of 0.8773 and the adjusted R² (which takes into account the high number of samples) had a value of 0.875. This value indicates the quality of the estimation of tensile shear strengths described by the different factors, whereby 1 means a perfect linear correlation of the independent variables with the dependent variable whereas 0 means that there is no correlation.

Table 13 shows the significance of all significant factors considered for the model, whereby values with a p-value lower than 0.01 are considered to describe the model well. Therefore linear terms, two-factorial interactions and quadratic terms are included in the model. The factor water added to the adhesive (B) did not show to have a significant linear influence for the model, but was significant as a quadratic term. Table 13 also displays the coded regression coefficients which are useful for identifying the relative impact of each factor on the response value (tensile shear strength). The higher the value (displayed as “coefficients (coded)” in Table 13), the greater the influence, whereby a plus means that the impact is positive and a minus indicates a negative influence. The highest negative impact on the tensile shear strength is evident for the factor foam thickness with a value of -1.18. Therefore, with a higher foam thickness it comes to a rapid decrease in strength. The highest positive linear impact on the tensile shear strength is apparent for the factor isocyanate with a value of 0.28. The highest negative impact on the tensile shear strength of the two-factorial interactions has isocyanate to temperature with a value of -0.21 (AD). This means that the difference in strength between low temperature and high temperature is lower at a lower isocyanate content compared to higher amounts of isocyanate.

The highest positive impact on the strength of the two-factorial factors can be seen by the temperature to the foam thickness (DE) with a value of 0.22. This indicates a higher difference in the tensile shear strength between lower foam thickness and higher foam thickness for lower temperatures. (Figure 33).

The highest positive quadratic impact in Table 13 has the foam thickness with a value of 0.50, whereby the highest negative impact have the factors isocyanate and additional water with a value of -0.28. With the inclusion of quadratic terms into the mathematical model, extreme values may occur which are important for the identification of possible optima for each factor.

Table 13: Overview of the significant factors and corresponding coded regression coefficients for identifying the relative impact of the factors on the tensile shear strength

Factors	**p-values**	**Coefficients (coded)**
Intercept	< 0.0001	-0.85
A-Isocyanate	< 0.0001	0.28
C-Wood moisture	< 0.0001	0.099
D-Temperature	< 0.0001	-0.46
E-Foam thickness	< 0.0001	-1.18
AC	0.0025	0.084
AD	< 0.0001	-0.21
AE	0.0005	0.10
CD	0.0015	0.079
CE	0.0008	-0.087
DE	< 0.0001	0.22
A^2	< 0.0001	-0.28
B^2	< 0.0001	-0.28
D^2	< 0.0001	-0.16
E^2	< 0.0001	0.50

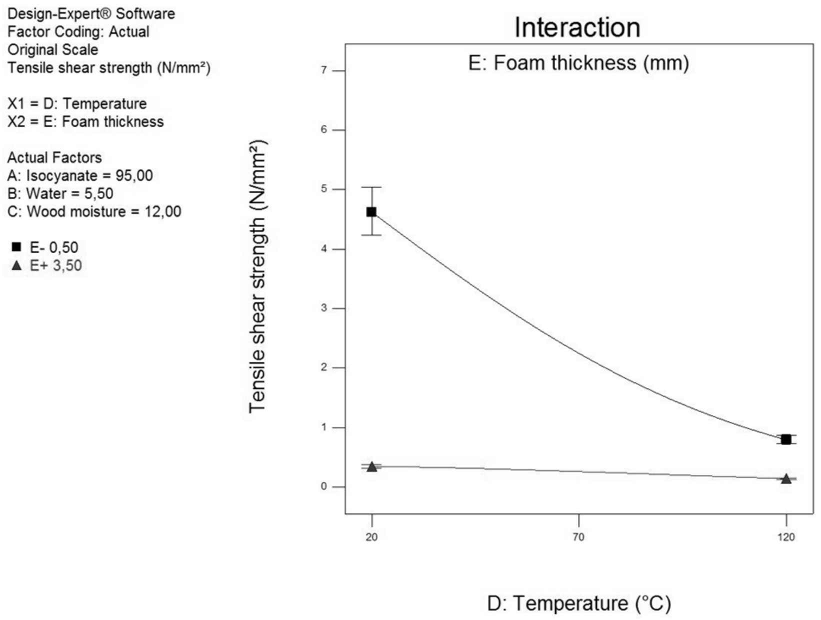


Figure 33: Interaction of temperature and foam thickness at an isocyanate content of 95 %, 5.5 % additional water content and 12 % wood moisture content

With the software Design-Expert it was also possible to create 3D diagrams, whereby in addition to one response value (tensile shear strength) two more factors can be displayed. Figure 34 shows the tensile shear strength depending on the amount of additional water and the amount of the isocyanate. These three diagrams show various wood moisture contents, all at a press temperature of 20 °C and a foam thickness of 0.5 mm. In all diagrams the highest tensile shear strength is indicated for an amount of 5.5 % water. Comparing the three diagrams displayed in Figure 34 an increase in wood moisture content is leading to an increase in strength, especially for higher isocyanate amounts above 77.5 %. At a lower amount of isocyanate (between 60 % and 68.75 %) this effect is less expressed.

According to the model shown in Figure 34 the highest strength is reached at a wood moisture content of 21 %, an amount of additional water of 5.5 % and an amount of isocyanate of about 92 %. This strength maximum shifts towards 86.25 % isocyanate content for decreasing wood moisture contents.

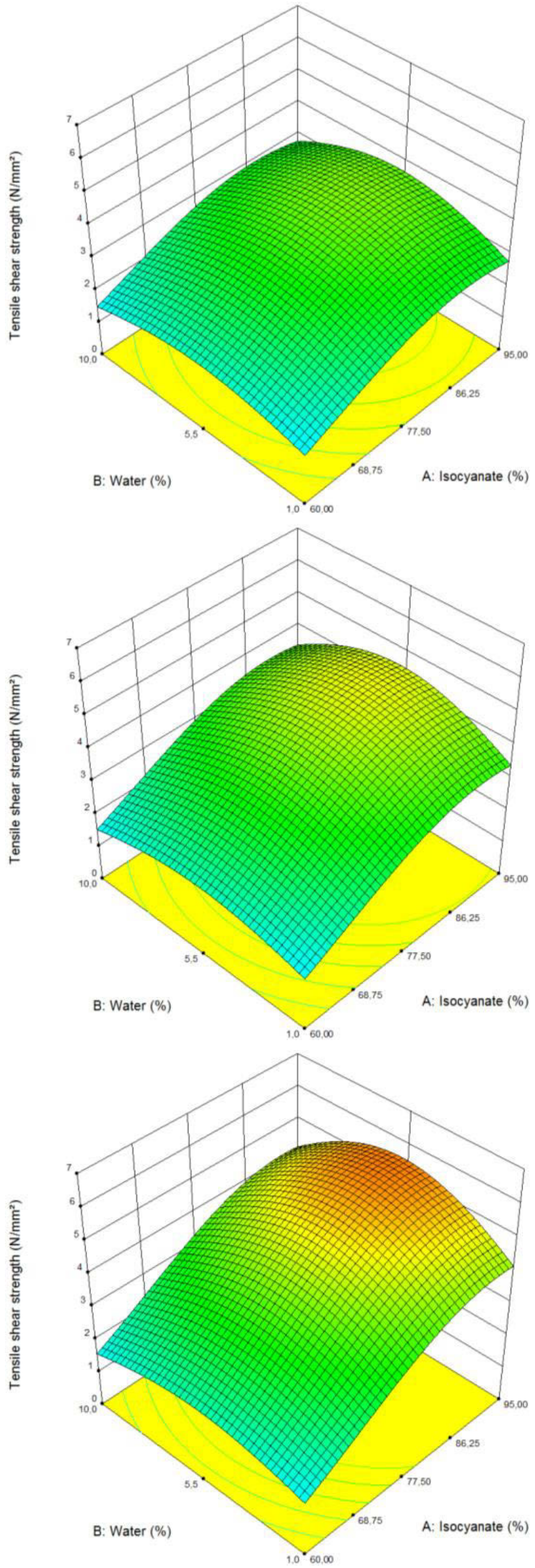


Figure 34: 3D model of the tensile shear strength at a temperature of 20 °C and a foam thickness of 0.5 mm at various wood moisture contents (top = 3 %, middle = 12 %, bottom = 21 %)

A plot of a comparable 3D model is shown in Figure 35 where tensile shear strength depending on the wood moisture content and the amount of the isocyanate at an additional water content of 5.5 % and a foam thickness of 0.5 mm at three different temperatures is shown. The highest tensile shear strengths are achieved by a press temperature of 20 °C (upper diagram) together with high wood moisture content and high isocyanate content. At low wood moisture content (3 %) and a temperature of 20 °C, the maximum of the strength is assigned around an isocyanate content of 86.25 % as already described in Figure 34.

From Figure 35 it is also evident that increasing press temperature (from top to bottom) dramatically decreases tensile shear strength. At a temperature of 70 °C there is a lower difference in strength between the lowest and the highest wood moisture content, whereby the highest strength is still achieved at high wood moisture content and an isocyanate content of 86.25 %. At a press temperature of 120 °C there are almost no differences in strength between the various wood moisture and isocyanate contents.

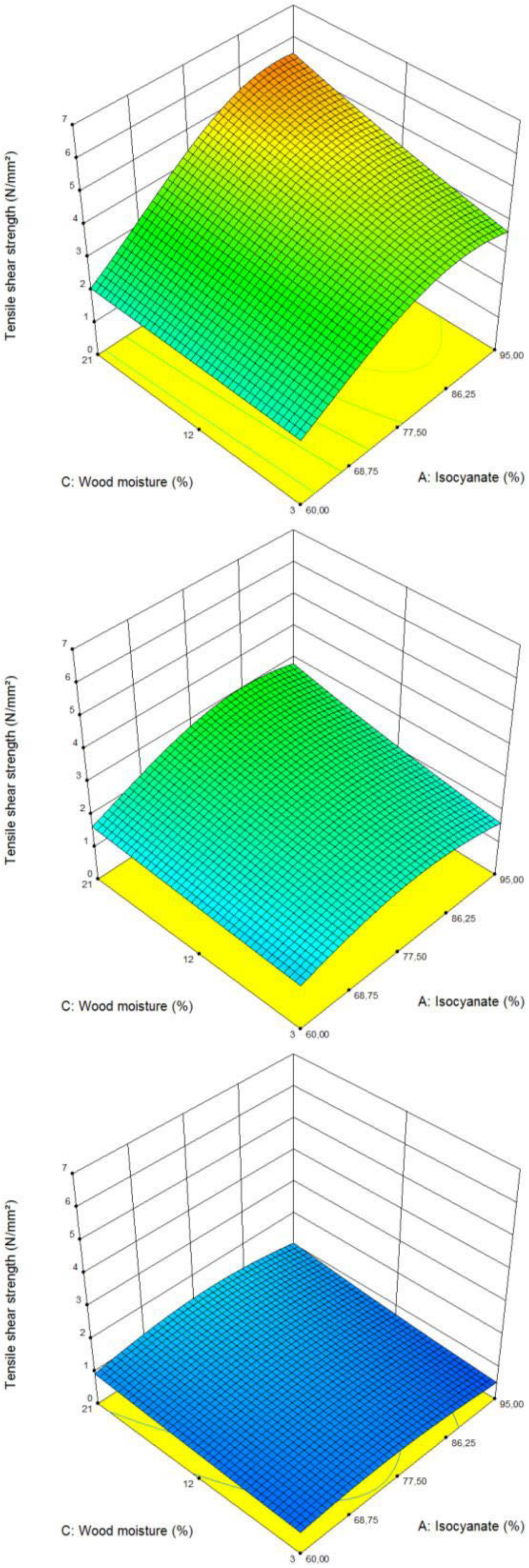


Figure 35: 3D model of the tensile shear strength at a water content of 5.5 % and a foam thickness of 0.5 mm at various temperatures (top = 20 °C, middle = 70 °C, bottom = 120 °C)

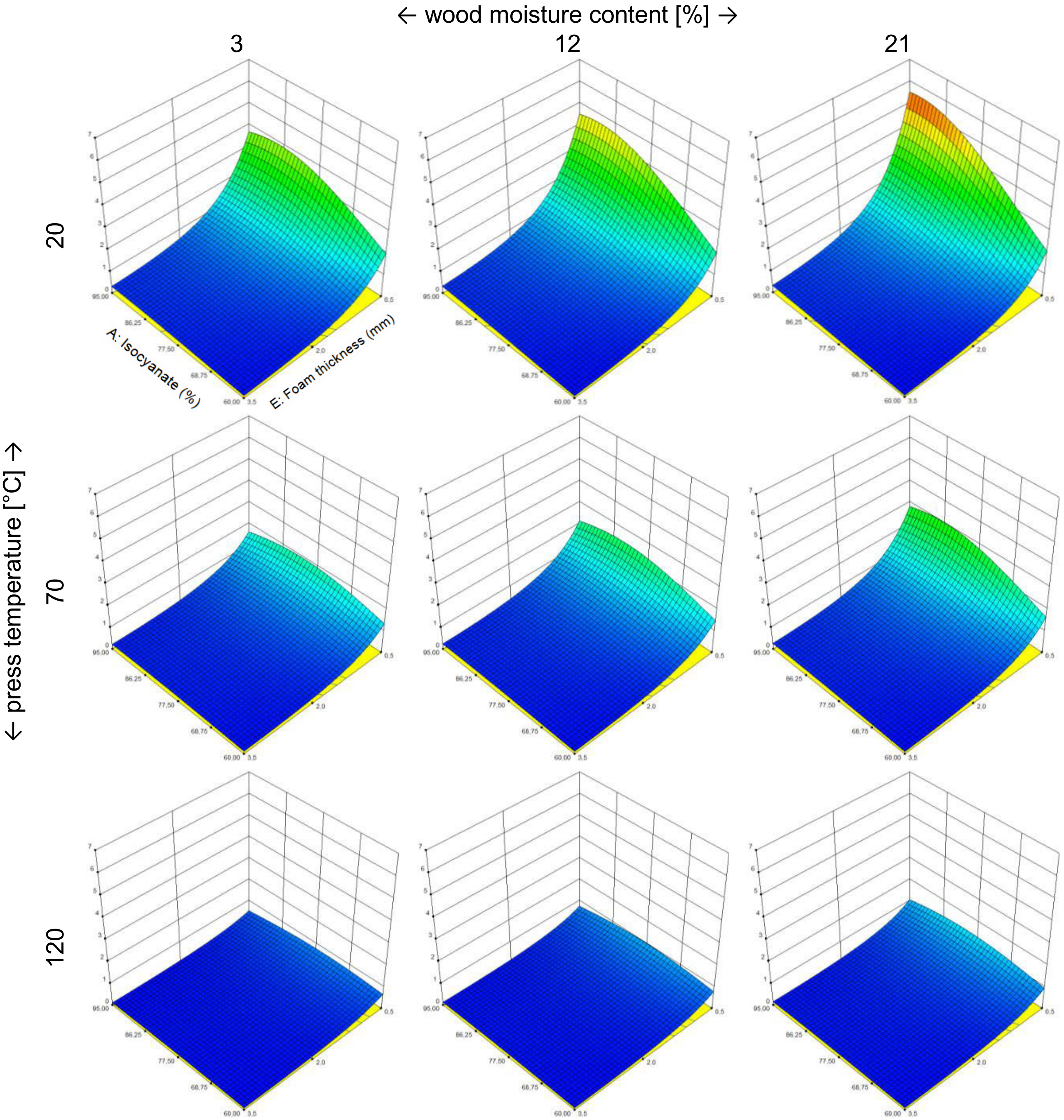


Figure 36: 3x3 matrix of the tensile shear strength depending on the wood moisture (from left to right) and the press temperature (from top to bottom) at a water content of 5.5 %

Figure 36 is a 3x3 matrix of the tensile shear strength model, where the wood moisture content is depicted on the x-axis and the press temperature on the y-axis. Each individual diagram shows the tensile shear strength depending on the amount of isocyanate and the foam thickness at an additional water level of 5.5 %. The highest strength is observed at a temperature of 20 °C, a wood moisture content of 21 %, a foam thickness of 0.5 mm and high levels of isocyanate content.

3.3.2 Shear modulus

The correlations between all measured tensile shear strength and shear modulus values, grouped by the foam thickness, are shown in the scatter plot of Figure 37. At all three foam thicknesses, a high value of R² = approx. 87 % is indicating a good correlation between the two response values. The slope of the correlation lines differ, whereby samples with a foam thickness of 0.5 mm show a steeper slope than the other two ones. Due to these high correlation values, especially obvious effects of influencing variables should be similar on both response variables and therefore less detailed diagrams will be shown for the shear modulus in section 3.3.2.1.

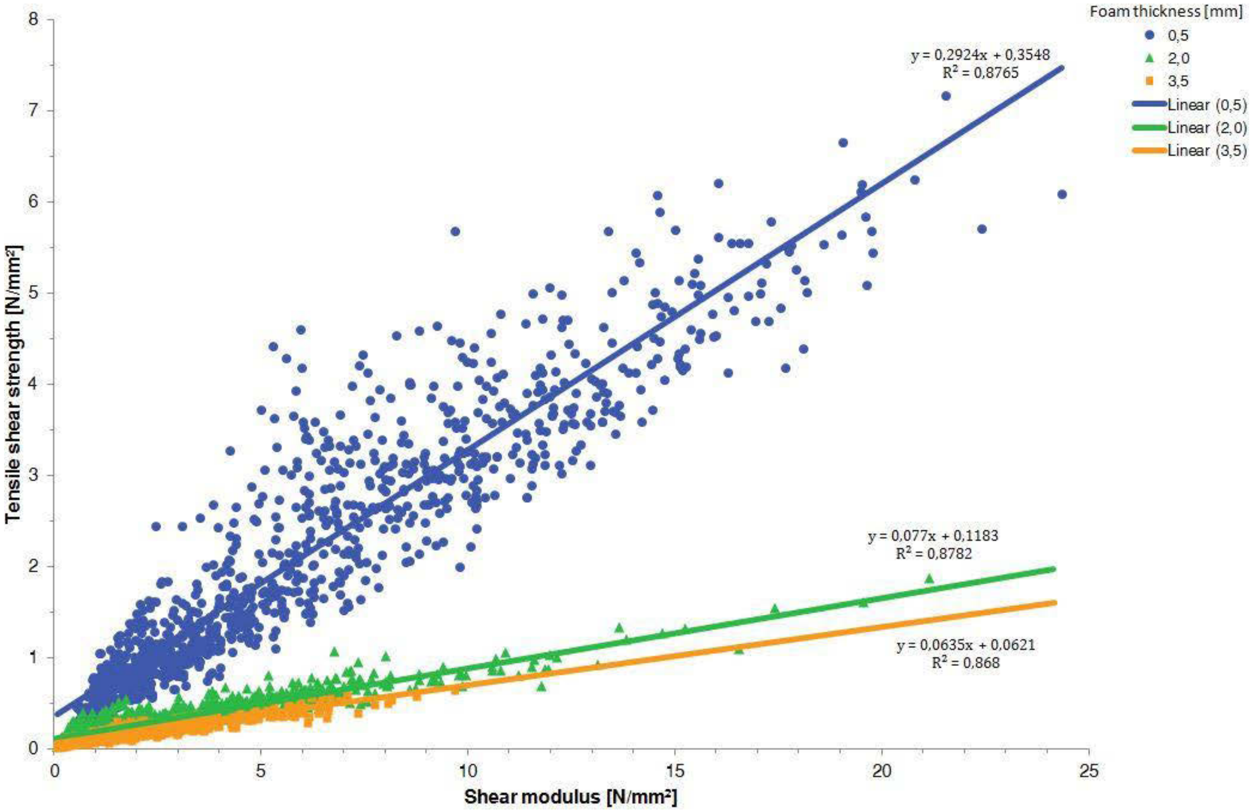


Figure 37: Correlation of tensile shear strength to shear modulus for different foam thicknesses

3.3.2.1 Results of the measured values (SPSS)

For the second response value, the shear modulus, the results were also evaluated with the software SPSS. Figure 38 shows the shear modulus for the samples with a foam thickness of 0.5 mm. The diagram is divided into the five levels of the amount of isocyanate, the three wood moisture contents and the three press temperatures (colours). The highest median of the shear modulus (~17 N/mm²) is at a wood moisture content of 21 %, a temperature of 20 °C and an isocyanate content of 95 %.

At a low temperature (20 °C) the shear modulus increases with an increasing amount of isocyanate. For the wood moisture contents of 3 % and 21 % this maximum is at an amount of 95 % isocyanate, whereby the maximum of the median for the wood moisture content of 12 % is at a 86.25 % isocyanate content.

For the samples at a press temperature of 70 °C the maximum is between an isocyanate content of 77.5 % and 86.25 %. At a temperature of 120 °C there is no significant difference between the levels of the isocyanate contents. At a low isocyanate content (60 %) the difference of the shear modulus between the different press temperatures is much lower than with a higher content of isocyanate.

Konnerth et al. (2007) identified for an 1-component polyurethane film a shear modulus of 0.18 GPa. Due to the foam formation, there is an increase in the volume around the 10 to 20 times. By dividing this value with the shear modulus value above, the shear modulus for the PUR foam should be between 9 N/mm² and 18 N/mm². Whether the shear modulus of the foam can be calculated with this method is questionable, however, the measured values are in this range.

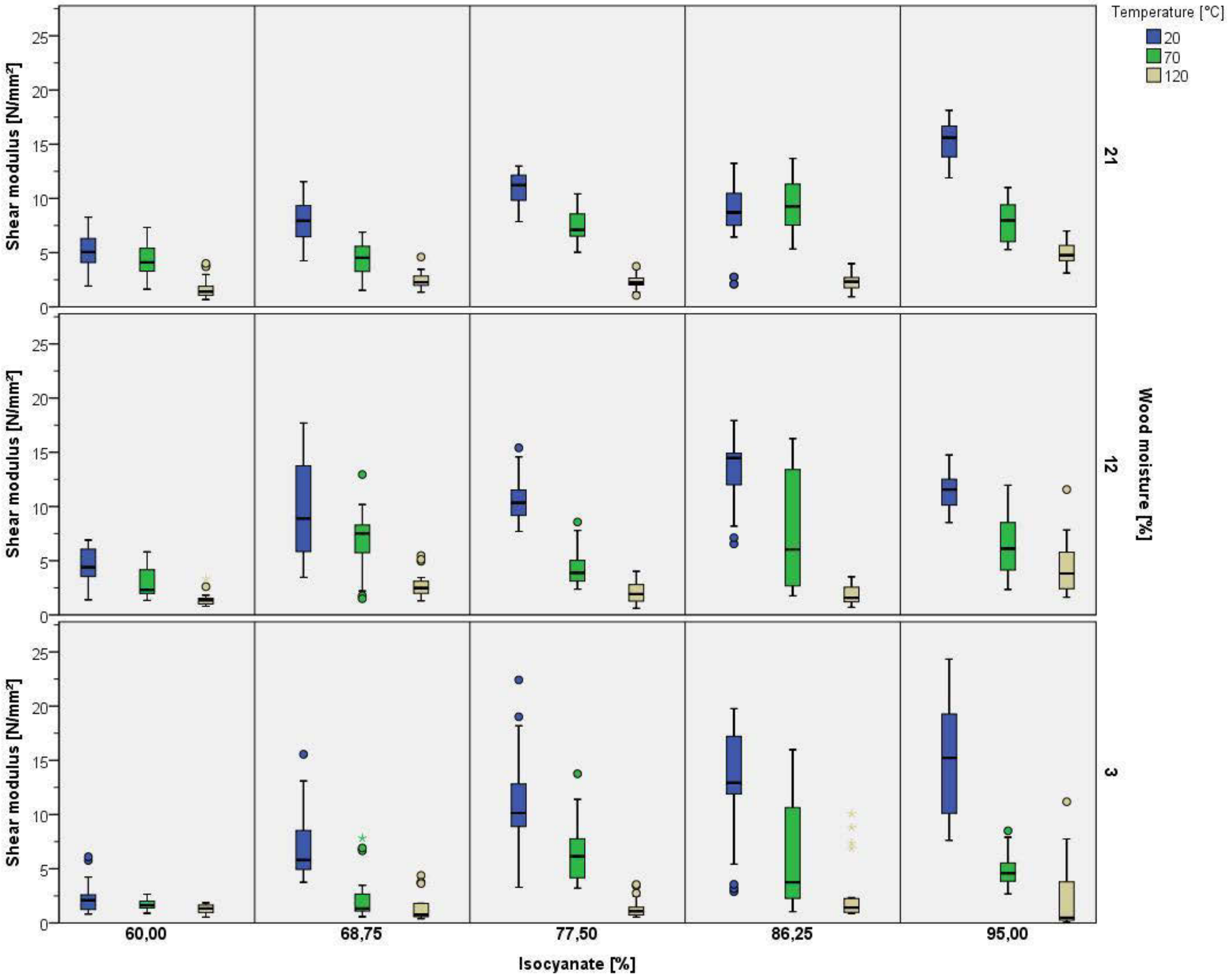


Figure 38: Shear modulus of the samples with a foam thickness of 0.5 mm (max. n=24 samples per boxplot)

In Figure 57 and Figure 58 in the appendix the shear modulus for the samples with a foam thickness of 2 mm and 3.5 mm are shown. For the 3.5 mm samples there are not all box plots in the diagram due to an insufficient foam formation. The shear modulus decreases with an increasing foam thickness, whereby the highest value for the 2 mm samples is 10 N/mm² and for the 3.5 mm samples it is 7 N/mm².

3.3.2.2 Results of mathematical models based on a statistical analysis (Design-Expert)

For the second response value, the shear modulus, again a statistical analysis was made with the software Design-Expert. The data were evaluated with an ANOVA with a significance level of 1 %. Due to the high ratio of maximum (20.328 N/mm² - red colour in the 3D-diagram) and minimum (0.075 N/mm² - blue colour in the 3D-diagram) modulus values, the software suggested again a data transformation similar as described for the case of tensile shear strength. Based on the predicted R^2 the software suggested a quadratic model. For the ANOVA the linear, the two-factorial interactions and the quadratic terms were selected from the software automatically with a backward selection and a significance level of 1 %. The residuals, the presence of a constant error and outliers were then checked again. Due to missing values or outliers, 62 values (of maximum 810) could not be included into the model. After this revision of the data, the ANOVA was carried out and the model was ready for the evaluation of the results.

The corresponding coefficient of determination (R^2) had a value of 0.7482 and the adjusted R^2, which takes into account the high number of samples, had a value of 0.7437. These values are lower compared to the values for the tensile shear strength, which means that the model is describing shear modulus somewhat comparably less good as it was the case for the tensile shear strength model. Nevertheless the values are considered to be at an acceptable level for sufficiently describing the influence of the different factors on the estimated shear modulus.

In Table 14 all significant factors with the p-values and the coded coefficients are listed. All linear terms, two-factorial interactions and the quadratic terms with a p-value lower than 0.01 describe the model. The factor water (B) is again not significant for the model, only as a quadratic term. As before the coded regression coefficients are useful for identifying the relative impact of each factor on the shear modulus. The highest linear factor is isocyanate with a positive value of 0.69, which means with a higher amount of isocyanate the estimated shear modulus would be also higher. The highest negative linear impact on the shear modulus is resulting from the foam thickness with -0.65. Also the temperature shows a very high negative impact on the shear modulus.

Table 14: Overview of the significant factors and corresponding coded regression coefficients for identifying the relative impact of the factors on the shear modulus

Factors	**p-values**	**Coefficients (coded)**
Intercept	< 0.0001	1.29
A-Isocyanate	< 0.0001	0.69
C-Wood moisture	< 0.0001	0.12
D-Temperature	< 0.0001	-0.64
E-Foam thickness	< 0.0001	-0.65
AD	< 0.0001	-0.27
AE	< 0.0001	0.25
CD	0.0005	0.11
CE	< 0.0001	-0.17
DE	0.0015	0.11
A^2	< 0.0001	-0.68
B^2	< 0.0001	-0.31
D^2	< 0.0001	-0.20
E^2	0.0042	0.13

For the two-factorial interactions the isocyanate to the temperature (AD) has the highest negative value (-0.27), as before for the tensile shear strength. This means that the difference in the shear modulus between the low temperature and high temperature is at a lower isocyanate content much lower than at a high amount of isocyanate (Figure 39). The highest positive impact on the shear modulus is again caused by isocyanate to the foam thickness (AE) with a value of 0.25. The highest positive quadratic impact in Table 14 has the foam thickness with a value of 0.13, whereby the highest negative impact has the factor isocyanate (-0.68).

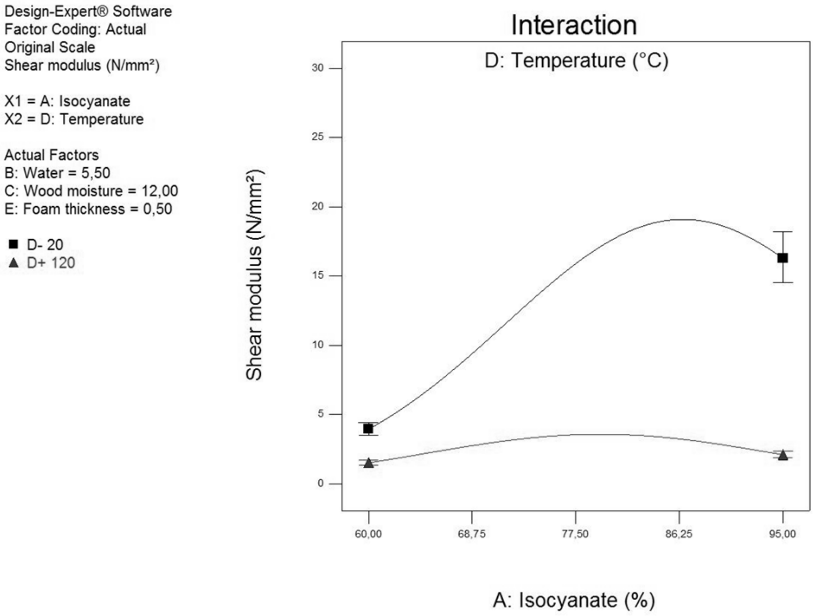


Figure 39: Interaction between the isocyanate content and the temperature at an amount of 5.5 % of additional water, a wood moisture of 12 % and a foam thickness of 0.5 mm

Figure 40 shows the 3D model of the shear modulus at a temperature of 20 °C and a foam thickness of 0.5 mm at various wood moisture contents. Each diagram displays the shear modulus depending on the amount of additional water and the amount of isocyanate. As already shown for the tensile shear strength, there is an optimum for the additional water content which is at a level of 5.5 %. This optimum is not particularly pronounced for lower isocyanate contents compared to higher ones.

There is also an increase in the shear modulus with a higher amount of isocyanate, whereby the highest values are reached at an isocyanate content of 86.25 %. With a higher content of isocyanate the shear modulus will decrease again. Figure 40 also shows a significant increase in the shear modulus as a result of increasing wood moisture content. This increase in the shear modulus is observed particularly for higher amounts of isocyanate.

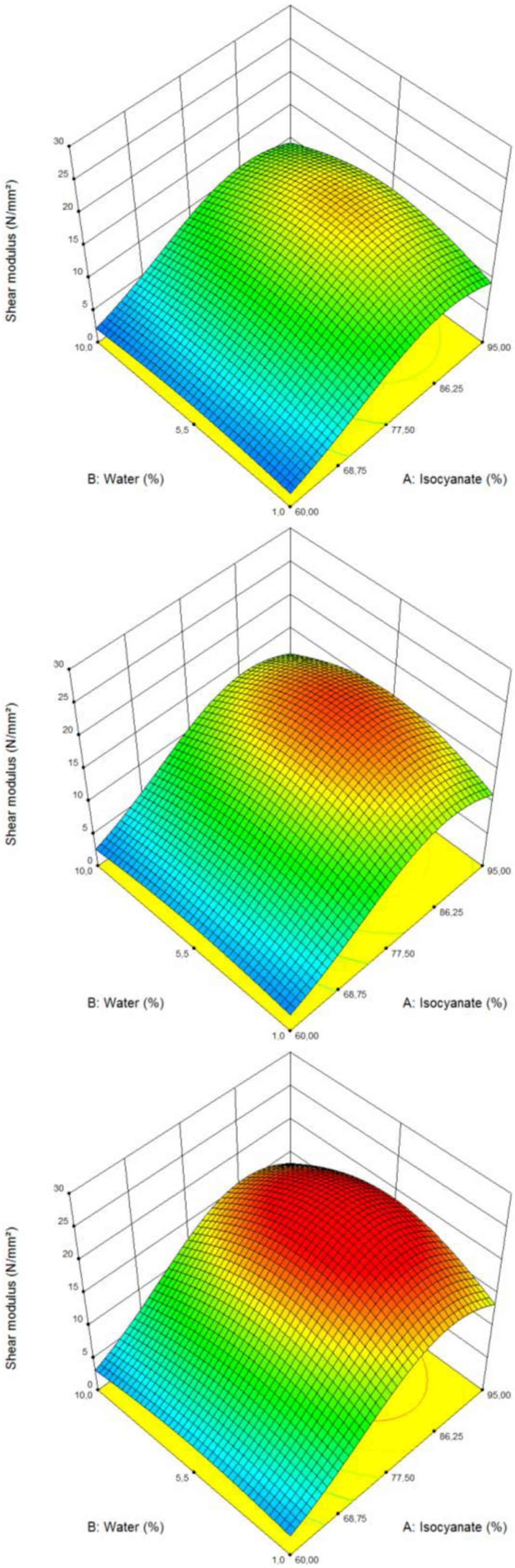


Figure 40: 3D model of the shear modulus at a temperature of 20 °C and a foam thickness of 0.5 mm at various wood moisture contents (top = 3 %, middle = 12 %, bottom = 21 %)

Figure 41 displays the 3D model of the shear modulus at a water content of 5.5 % and a foam thickness of 0.5 mm at different wood moisture contents. Each diagram shows the shear modulus depending on the press temperature and the amount of isocyanate. The highest shear modulus can be observed at an isocyanate content of 86.25 %, at a temperature of 20 °C and a wood moisture content of 21 %. With increasing temperature this maximum moves towards 77.5 % isocyanate content. Figure 41 also shows a significant influence of the temperature on the shear modulus, whereby higher temperature result in lower shear moduli.

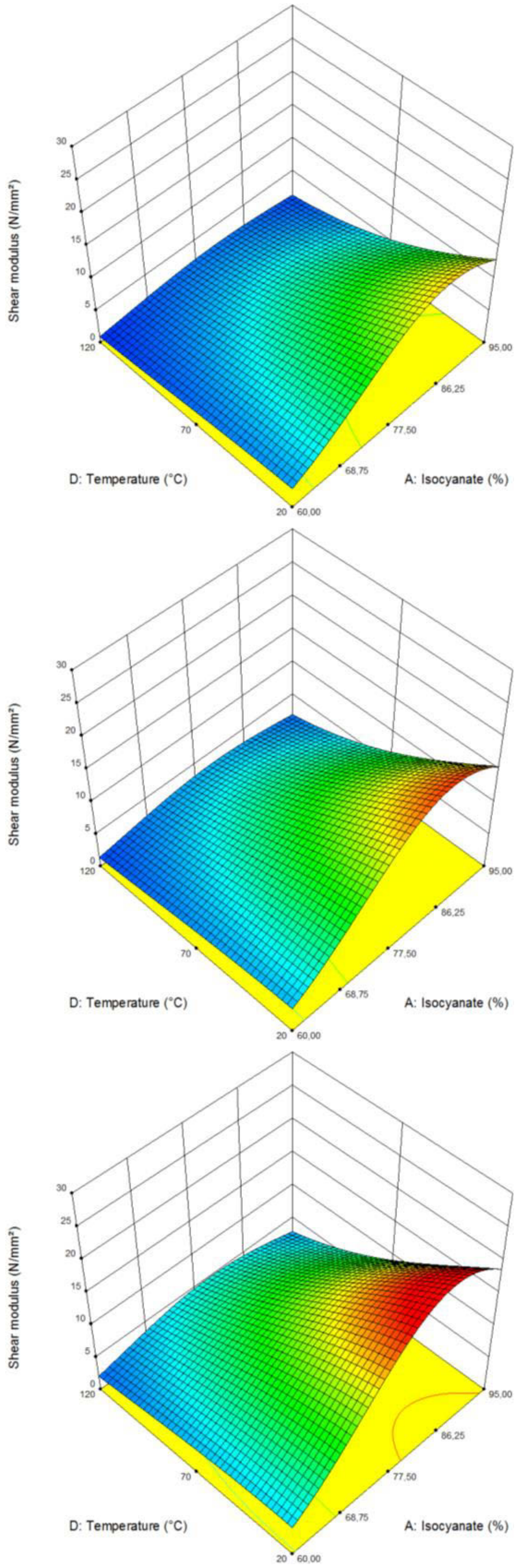


Figure 41: 3D model of the shear modulus at a water content of 5.5 % and a foam thickness of 0.5 mm at various wood moisture contents (top = 3 %, middle = 12 %, bottom = 21 %)

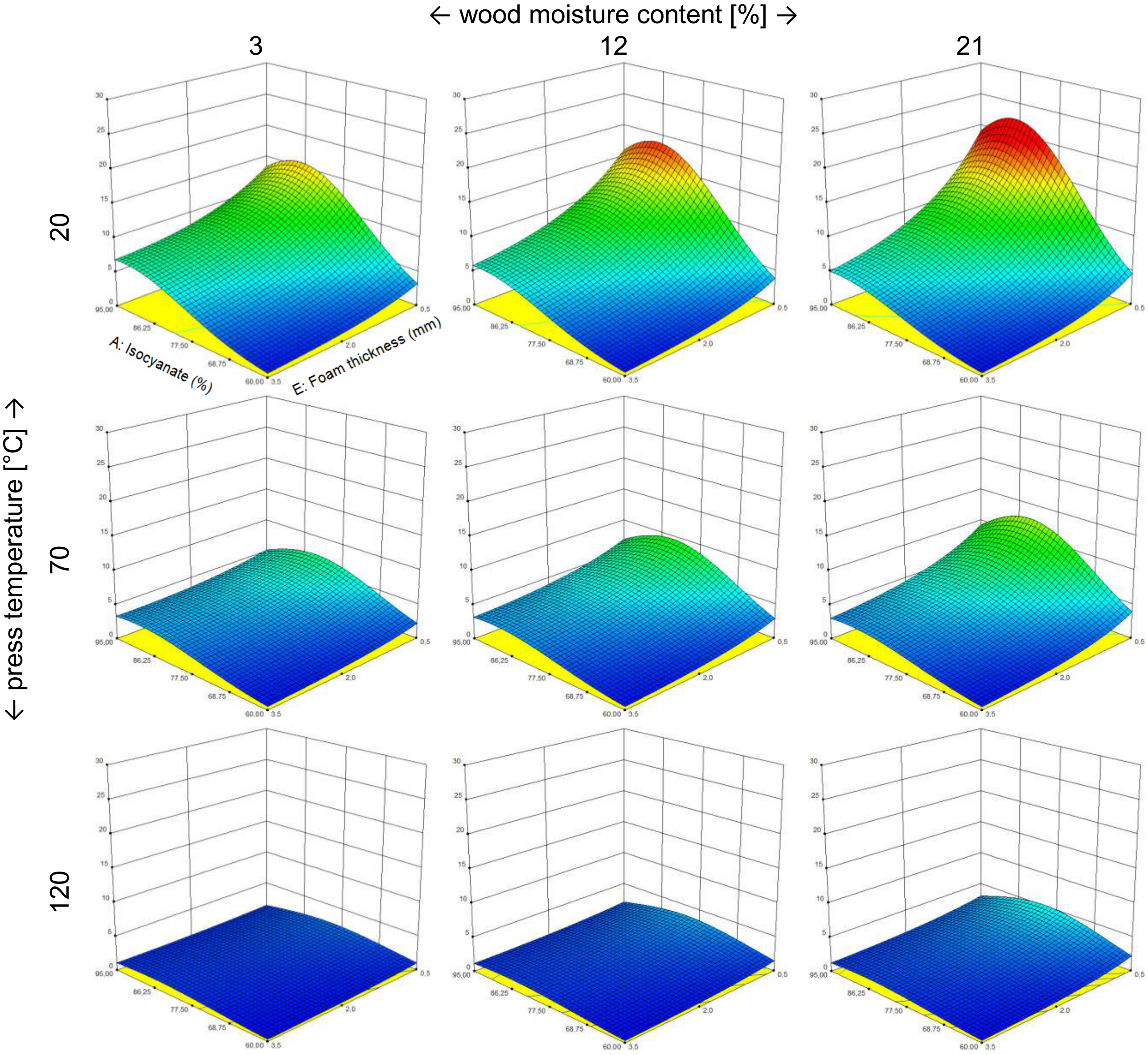


Figure 42: 3x3 matrix of the shear modulus depending on wood moisture content (from left to right) and press temperature (from top to bottom) at a water content of 5.5 %

Figure 42 shows a 3x3 matrix of the shear modulus model. Each diagram shows the shear modulus depending on the amount of isocyanate and the foam thickness at an additional water level of 5.5 %. The highest shear modulus is observed at a temperature of 20 °C, a wood moisture content of 21 %, a foam thickness of 0.5 mm and an isocyanate content of 86.25 %.

3.3.3 *Wood failure amount*

After mechanical testing the fracture area of each sample was evaluated visually and differentiated for fracture taking place in the adhesive foam or fracture of the wood substrate. At a foam thickness of 0.5 mm there were some samples where the fracture was not only in the bond line but also in the wood. According to DIN EN 302-1 (2013) wood failure was estimated in 10 % increments within the overlapping area of the sample. The value was noted and for each combination (8 samples) the arithmetic mean was calculated.

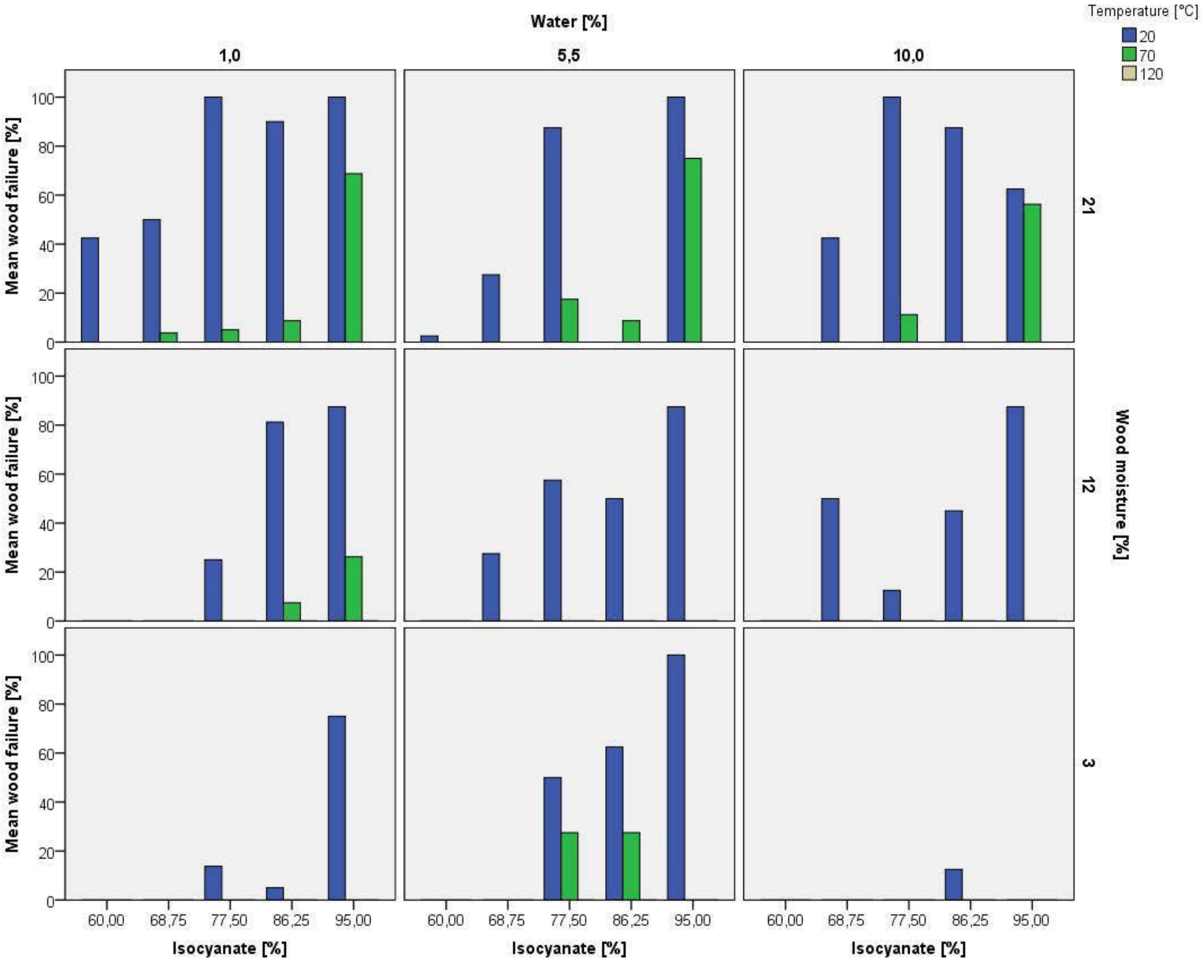


Figure 43: Mean wood failure of the samples with a foam thickness of 0.5 mm (max. n=8 samples per bar)

From Figure 43 a higher occurrence of wood failure can be found at a press temperature of 20 °C, which means the adhesion of the PUR foam seems to be better than at a higher press temperature. For higher wood moisture contents wood failure also increases. A reason for that could be that the isocyanate reacts at a lower temperature preferably with the water in the wood and results in higher adhesion to the adherent. There is also a trend that the mean wood failure increases with a higher content of isocyanate. Within the levels of the additional water, no significant difference in the wood failure can be observed. At a press temperature of 70 °C there is still some wood failure, especially for the samples with high wood moisture content (21 %) and a high amount of isocyanate (95 %). At a temperature of 120 °C, the adhesion seems to be too weak to cause wood failure.

3.3.4 Invalid samples due to an insufficient foam formation

As an additional result the amount of samples showing insufficient foam formation were evaluated. Out of the entire set of 4320 samples, 348 samples could not be mechanically tested due to insufficient foam formation. For these samples no tensile shear strength and shear modulus could be calculated and therefore they could not be included in the evaluation of the data shown above, especially to the statistical analysis with the software Design-Expert. Of the 348 invalid samples are 49 samples with a target foam thickness of 2 mm, which represents 4.5 % invalid samples of all samples with a foam thickness of 2 mm. The remaining 299 invalid samples have a target foam thickness of 3.5 mm (27.7 % of all samples with a foam thickness of 3.5 mm).

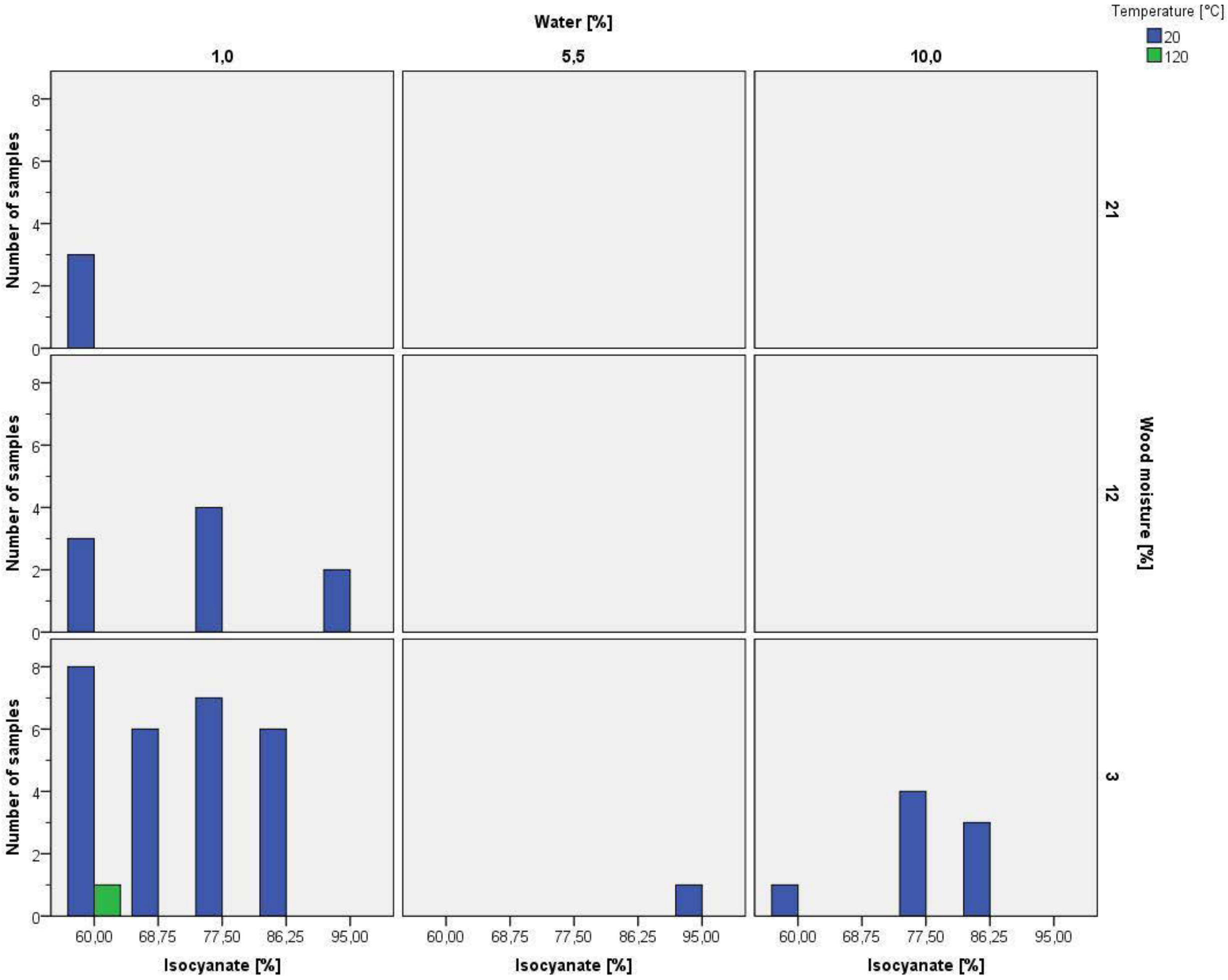


Figure 44: Number of invalid samples due to an insufficient foam formation at a target foam thickness of 2 mm (max. n=8 samples per bar)

From Figure 44 it can be seen, that most invalid samples resulted from a combination of low wood moisture content (3 %), together with a low amount of additional water (1 %) and a low press temperature (20 °C). As already described in chapter 1.3.2, water is important for the foam formation due to the chemical reaction and the formation of CO_2. Figure 44 also shows that it has a negative effect on the formation of the PUR foam when either very little additional water or low wood moisture content is available, and even worse when both cases are occurring at the same time. At a press temperature of 70 °C there were no invalid samples and for the temperature at 120 °C there was only one sample with an insufficient foam formation, which indicates a higher sensitivity of foam formation at lower temperature.

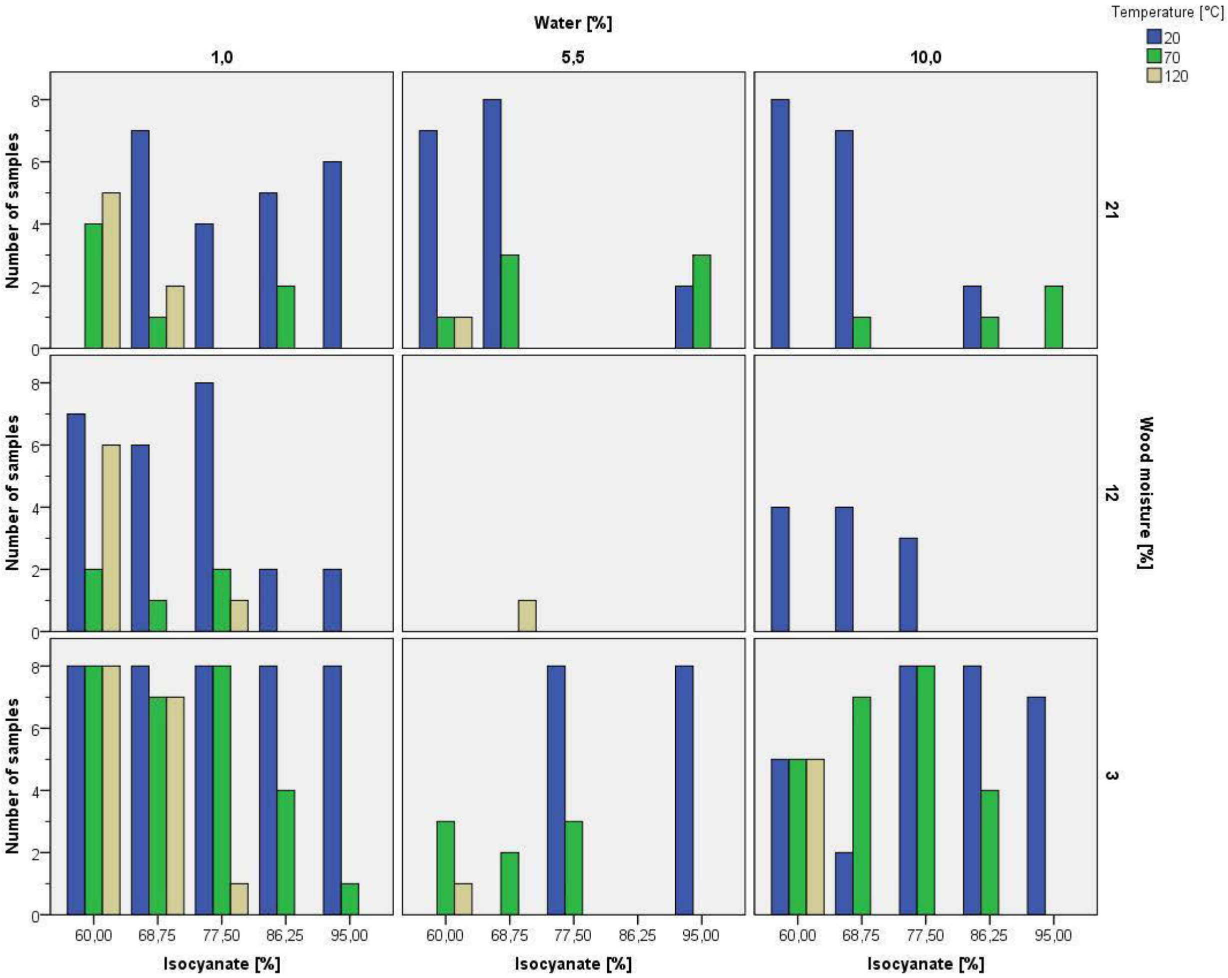


Figure 45: Number of invalid samples due to an insufficient foam formation at a target foam thickness of 3.5 mm (max. n=8 samples per bar)

As before most invalid samples are counted at the combination of low wood moisture content (3 %) together with a low amount of additional water (1 %) (Figure 45). In this case all of the samples with a press temperature of 20 °C were invalid. In general, the most invalid samples occurred at a temperature of 20 °C as shown in Figure 46.

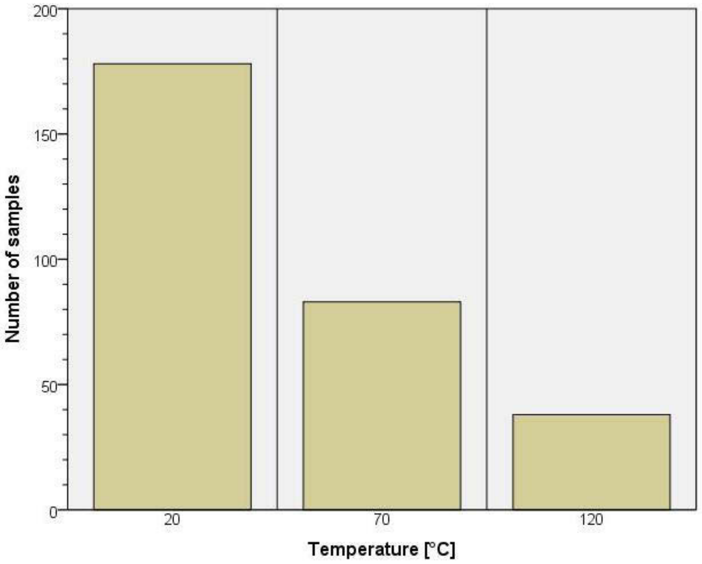


Figure 46: Number of invalid samples for each press temperature for the samples with a target foam thickness of 3.5 mm

A reason for that could be, that due to higher temperature the additional water reacts faster with the isocyanate - polyol mixture and therefore this leads to a better foam formation due to a higher release of CO_2. For the wood moisture it also could be that with a higher temperature more water from the cell wall is available in the bond line (possibly in the form of vapour, which is assumed to be higher in reactivity).

The least number of invalid samples was observed for the combination of a medium additional water content (5.5 %) and a medium wood moisture content (12 %). Only one sample at a press temperature of 120 °C was invalid.

Another interesting reason for the high number of invalid samples could be due to an insufficient mixing of the components isocyanate, polyol and water. According to Sonnenschein (2015) the mixing is essential for the foam formation, because after this no new bubbles are formed anymore, whereby the produced CO_2 diffuses to these existing bubbles and further the PUR foam is formed.

3.3.5 Prediction of the optimum parameters for adhesive foam bonding

Based on the models which were created with the help of Design-Expert software, it was possible to predict an optimal composition of the individual factors. This was also done with this software by a numerical optimization. The first step was to set the target, the lower and upper limit and the importance for each factor. For the factors isocyanate, additional water, wood moisture content and the press temperature the optimal value were set to be in the range of the investigated minimum and maximum of each factor. The importance was set to 3 for each of these values, which is moderately important (1 = less important; 5 = very important). The target for each foam thickness was set to the respective thickness with an importance of 5. For the two response values, the tensile shear strength and the shear modulus, the aim was to maximize these values with three different types of importance. As a first one the tensile shear strength was set to an importance of 1 (less important) and the shear modulus to an importance of 5 (very important). The second type was exactly the opposite as before and the third one was to set the levels at an intermediate range. Based on the models for each type and for each foam thickness 100 solutions were calculated by the software automatically.

The differences in the various solutions are minimal and do not behave contrary, whereby this trend is shown in Table 15.

The desirability in Table 15 and Table 16 indicates how good the results fit to the settings which were defined before. This value is calculated automatically by the software and values close to 1 indicate a very high agreement of the settings.

Table 15: First 10 solutions for a target foam thickness of 0.5 mm to maximize the tensile shear strength (importance = 1) and shear modulus (importance = 5)

Solutions	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**
Isocyanate [%]	91,86	90,75	90,21	92,70	93,20	91,54	93,00	89,99	92,14	90,00
Water [%]	5,40	5,14	5,72	5,24	4,59	5,39	4,84	6,15	4,29	5,97
Wood moisture [%]	21,00	20,98	21,00	20,83	21,00	20,67	20,56	20,60	21,00	20,28
Temperature [°C]	20,00	20,00	20,00	20,00	20,00	20,00	20,03	20,00	20,00	20,00
Foam thickness [mm]	0,50	0,50	0,50	0,50	0,50	0,50	0,50	0,50	0,50	0,50
Tensile shear strength [N/mm²]	5,64	5,62	5,62	5,61	5,57	5,60	5,55	5,54	5,53	5,53
Shear modulus [N/mm²]	21,37	21,81	22,04	20,85	20,38	21,39	20,45	21,82	20,75	21,76
Desirability	0,9880	0,9877	0,9877	0,9876	0,9869	0,9869	0,9866	0,9864	0,9862	0,9862

After this, the arithmetic mean (x̄) and the standard deviation (σ) of the first 50 solutions were calculated for each factor, response value and desirability value with the software Microsoft Excel 2007.

Table 16 shows the results of the optimization at the example of a target foam thickness of 0.5 mm for different importance levels to optimize either preferably tensile shear strength or shear modulus. All of these three types indicate a favourable high isocyanate content of about 89 % at a low standard deviation. Also the additional water content is almost at a same level of 5.5 % for all scenarios. For achieving a maximum in tensile shear strength and shear modulus, a high wood moisture content (~19.5 %) together with a low temperature (~21 °C) are needed.

Table 16: Optimization of the factors to achieve highest tensile shear strength and shear modulus at different importance levels and a target foam thickness of 0.5 mm

		target foam thickness = 0.5 mm					
Tensile shear strength **Shear modulus**	**Importance**	**1** **5**		**5** **1**		**3** **3**	
		x̄	σ	**x̄**	σ	**x̄**	σ
Isocyanate [%]		**88.7**	3.0	**89.3**	3.9	**88.5**	3.5
Water [%]		**5.6**	1.1	**5.4**	1.2	**5.5**	1.2
Wood moisture [%]		**19.3**	2.2	**19.7**	1.8	**19.5**	2.0
Temperature [°C]		**20.8**	2.3	**21.3**	3.2	**20.6**	1.5
Foam thickness [mm]		**0.5**	0.001	**0.5**	0.007	**0.5**	0.003
Tensile shear strength [N/mm²]		**5.24**	0.25	**5.21**	0.27	**5.24**	0.25
Shear modulus [N/mm²]		**20.76**	0.77	**20.36**	1.20	**20.74**	0.98
Desirability		**0.979**	0.008	**0.903**	0.024	**0.941**	0.016

In Table 17 a similar analysis was performed for a target foam thickness of 2 mm is shown. To maximize the tensile shear strength and the shear modulus an isocyanate content above 90 % is required for each type of optimization, whereas the additional water content is again at a level of about 5.5 %. For the wood moisture content the optimal values are in the range of 15.5 and 19.6 % for the various types. For cases where a maximization of the shear modulus is more important, the average wood moisture content should be at lower levels of 15.5 %, for other cases it should again be on the higher levels of around 19 %. Same as for the case of a 0.5 mm thick bond line the proposed press temperature should be at 20 °C. The total desirability in Table 17 is much lower compared to the foam thickness of 0.5 mm. The reason for that is, that the simulated values for the tensile shear strength and the shear modulus for the case of a 2 mm thick bond line is in general on a much lower level than the maximum values of the entire model. The highest input values for the model of 6.434 N/mm² for the tensile shear strength and 20.328 N/mm² for the shear modulus can only be reached by 0.5 mm thick bond lines, whereas the values reach by 2 mm thick bond line are a magnitude lower.

Table 17: Optimization of the factors to achieve highest tensile shear strength and shear modulus at different importance levels and a target foam thickness of 2 mm

		target foam thickness = 2 mm					
Tensile shear strength **Shear modulus**	**Importance**	**1** **5**		**5** **1**		**3** **3**	
		x̄	σ	**x̄**	σ	**x̄**	σ
Isocyanate [%]		**90.1**	0.6	**93.2**	1.7	**90.9**	0.9
Water [%]		**5.4**	0.4	**5.4**	0.6	**5.5**	0.4
Wood moisture [%]		**15.5**	4.5	**19.6**	1.8	**18.5**	3.1
Temperature [°C]		**20.0**	0.02	**20.5**	1.5	**20.1**	0.3
Foam thickness [mm]		**2.0**	0.0003	**2.0**	0.001	**2.0**	0.001
Tensile shear strength [N/mm²]		**0.79**	0.03	**0.83**	0.02	**0.82**	0.03
Shear modulus [N/mm²]		**9.17**	0.06	**8.87**	0.21	**9.17**	0.06
Desirability		**0.572**	0.003	**0.359**	0.004	**0.453**	0.004

Table 18 shows the optimal values for a target foam thickness of 3.5 mm. As before a very high isocyanate content at average additional water content is required. The preferable wood moisture content is much lower than before with values ranging from 4.5 % to 9.7 % depending on the importance of the maximization of the two response values. The temperature is about 20 °C again.

As these results of the optimization are based on modelling valid samples for the tensile shear strength and the shear modulus only, it has to be considered that at this combination of the individual factors a higher probability of insufficient foam formation could occur as already shown in Figure 45.

Table 18: Optimization of the factors to achieve highest tensile shear strength and shear modulus at different importance levels and a target foam thickness of 3.5 mm

		target foam thickness = 3.5 mm					
Tensile shear strength **Shear modulus**	**Importance**	**1** **5**		**5** **1**		**3** **3**	
		x̄	σ	**x̄**	σ	**x̄**	σ
Isocyanate [%]		**92.8**	1.6	**94.0**	1.0	**92.7**	2.0
Water [%]		**5.3**	0.9	**5.4**	0.4	**5.5**	0.8
Wood moisture [%]		**4.5**	2.1	**9.7**	6.2	**7.1**	4.4
Temperature [°C]		**20.1**	0.4	**20.2**	0.6	**20.6**	1.7
Foam thickness [mm]		**3.5**	0.003	**3.5**	0.001	**3.5**	0.002
Tensile shear strength [N/mm²]		**0.34**	0.01	**0.34**	0.00	**0.34**	0.01
Shear modulus [N/mm²]		**6.70**	0.25	**6.26**	0.63	**6.41**	0.42
Desirability		**0.455**	0.008	**0.226**	0.001	**0.316**	0.005

3.4 Solid wood bonding

In the next sections the results of the samples with thin bond lines, which did not have a groove for foam formation, are shown. The measured data of the samples were evaluated only with the software SPSS. For the solid wood bonding there is only one response value, the tensile shear strength. According to the standard DIN EN 302-1 (2013) also the wood failure amount was evaluated visually.

3.4.1 Tensile shear strength

Figure 47 shows the tensile shear strength for solid wood bonding which is divided into the three wood moisture contents and the three press temperatures.

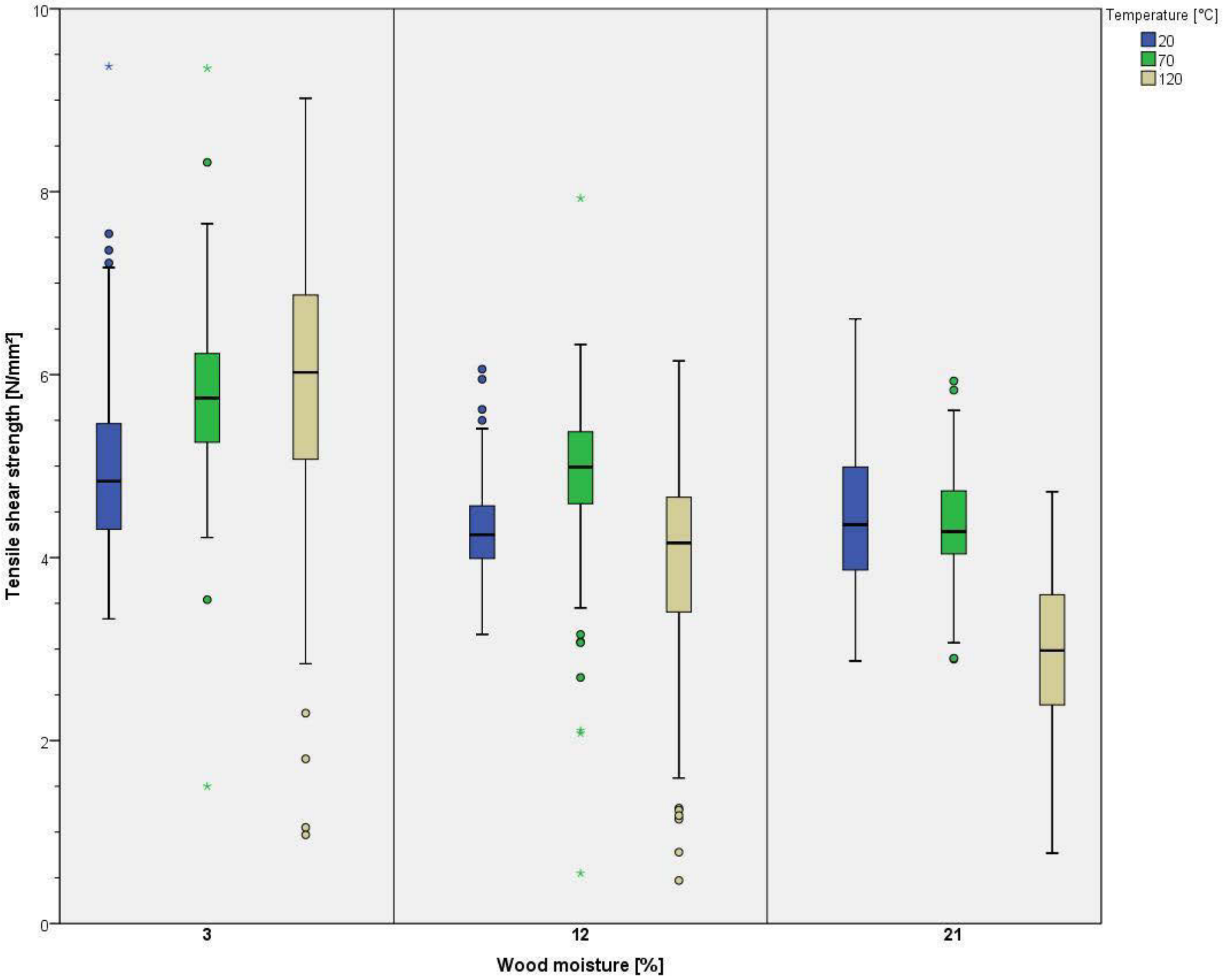


Figure 47: Tensile shear strength of the solid wood bonding divided into the wood moisture contents and the press temperatures (max. n=120 samples per boxplot)

In contrast to the bonds consisting of a foam filled bond line as described in chapter 3.3.1.1, for the thin bond lines the highest median (~6 N/mm²) was measured at low wood moisture content (3 %) and a press temperature of 120 °C. There is an overall tendency of decreasing tensile strength for increasing wood moisture content. For the case of low wood moisture content (3 %) an overall increase in strength with a higher press temperature is visible. At a wood moisture content of 12 % the highest median is located at a temperature of 70 °C with 5 N/mm², whereas at a temperature of 20 °C and 120 °C, the median is almost on the same level. For the highest wood moisture content (21 %), the samples which were glued together at a temperature of 20 °C have the highest median of 4.4 N/mm², whereas higher temperature resulted in lower strength of the samples (3 N/mm²).

Figure 48 displays the tensile shear strength again in more detail including the additional levels of the isocyanate contents. Differences depending on the amount of isocyanate seem to be less pronounced compared to the foamed bond lines as described in section 3.3.1.1. Anyhow, some additional differences can be derived e.g. at 12 % wood moisture content a slight increase in tensile shear strength with increasing isocyanate content can be observed for 120 °C, whereas other temperatures seem to be less effected.

In the appendix (Figure 59 and Figure 60) additional detailed results are presented for the tensile shear strength with the levels of additional water, therefore each boxplot represents a maximum number of 8 samples.

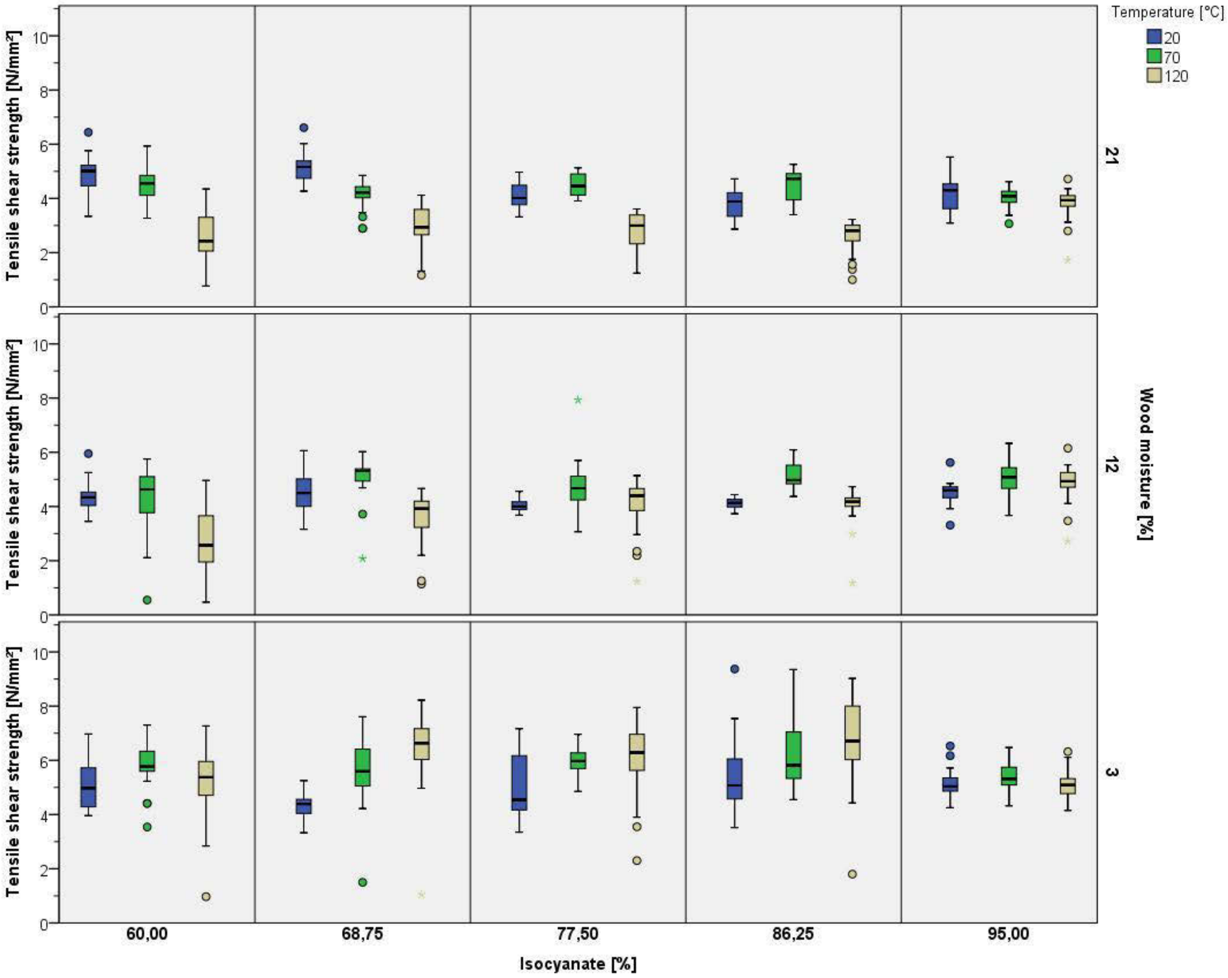


Figure 48: Tensile shear strength of the solid wood bonds divided into the wood moisture contents, the press temperatures and the isocyanate contents (max. n=24 samples per boxplot)

3.4.2 *Wood failure amount*

For each solid wood bonding type, the fracture area was evaluated visually according to the standard DIN EN 302-1 (2013) and the wood failure was estimated in 10 % increments of the overlapping area of each sample. The value was noted and for each combination (consisting of 8 specimens) the arithmetic mean was calculated.

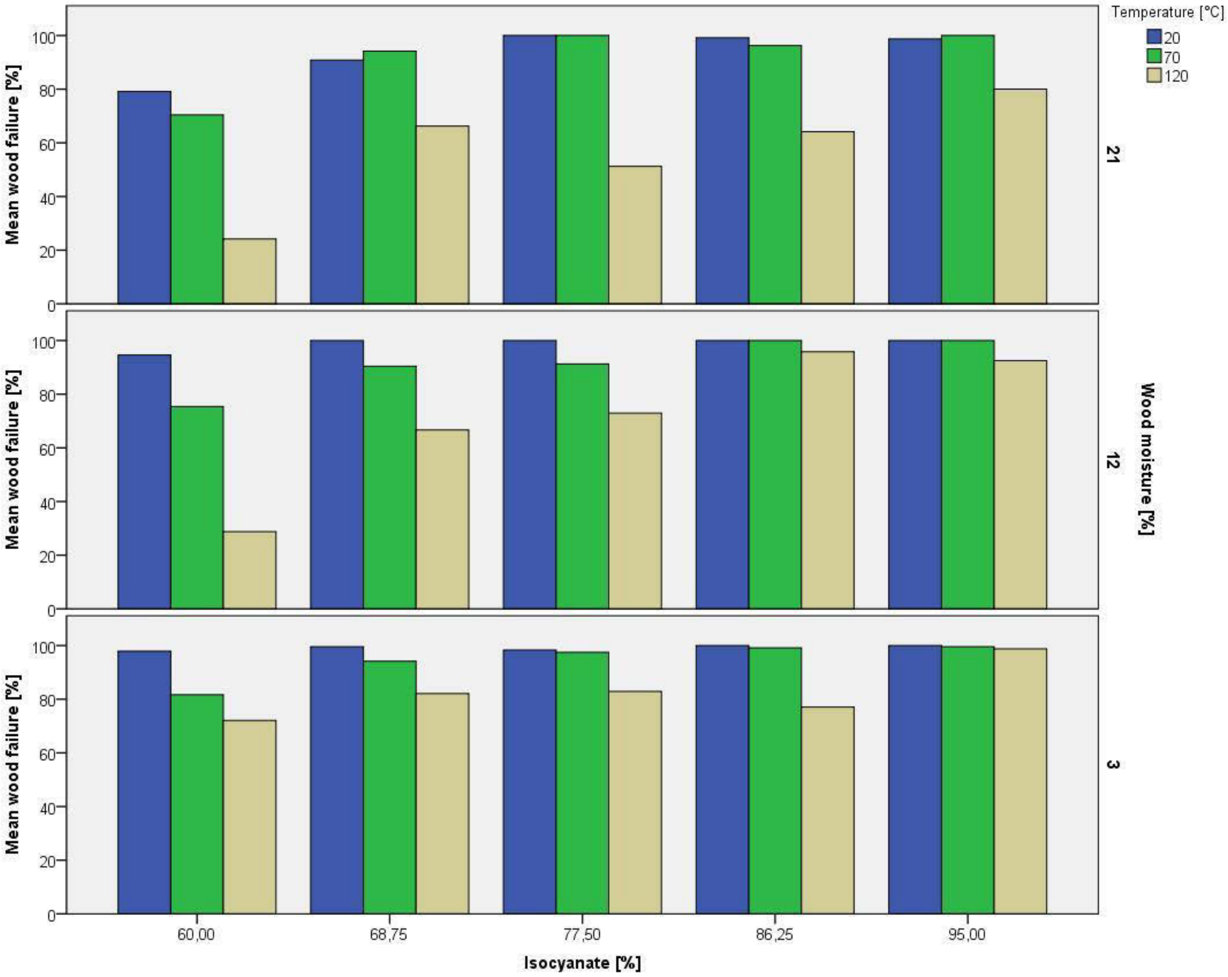


Figure 49: Mean wood failure of the solid wood bonding samples (max. n=24 samples per bar)

Figure 49 shows the mean wood failure of the solid wood bonding samples with the levels of the amount of isocyanate, the wood moisture contents and the press temperatures. With these same levels as in Figure 48, it is possible the compare the tensile shear strength and the corresponding mean wood failure. As shown in Figure 49 the wood failure is very high at a low press temperature (20 °C). With higher temperature the mean wood failure decreases, especially at a very high temperature (120 °C). Between the levels of the amount of isocyanate there is also a difference evident regarding wood failure. With a higher amount of isocyanate the mean wood failure tends to increase. In the appendix in Figure 61 the detailed mean wood failure for each combination is shown.

Konnerth et al. (2016) report a tensile shear strength of about 6.5 N/mm² for 1-C PUR bonded samples with a mean wood failure of 100 %. The spruce wood had a higher mean density of 0.445 g/cm³ at a mean moisture content of 12.74 %. In this experiment, the density of the wood is considerably lower and therefore also the strength values are lower.

Due to the low density of spruce wood as mentioned in chapter 2.4.1, this would explain why there is a high mean wood failure at a maximum tensile shear strength of 6 N/mm².

A reason for the significant difference in strength for the samples which were hot pressed at 120 °C could be, that due to the lower specific pressure of 0.3 MPa small bubbles occurred within the bond line in the case at a high wood moisture content (21 %). This could explain why the strength is higher with lower wood moisture content (3 %) than with high wood moisture content (21 %) Compared with the mean wood failure in Figure 49, where lower wood failure values occurred at high wood moisture content, the bubble formation theory within the bond line also could explain this assumption.

4 Conclusion

According to the aim/hypothesis of the present master thesis to systematically examine the influence of the individual factors on the strength and stiffness of a 2C-PUR adhesive with foaming behaviour the following main conclusion may be drawn.

Basically, there is a clear difference in the mechanical properties between solid wood bonding and foam bonding. An important result is that the influences of the factors are different for the two types of bonding. In the case of foam bonding the main findings are as follows.

The **ratio of isocyanate to polyol** with a higher isocyanate content has a positive influence on the tensile shear strength and the shear modulus. A high isocyanate content means that the costs for producing a product made out of the PUR foam will increase due to the high price of isocyanate. For optimization this amount could be reduced to a certain level to have still a sufficient strength/stiffness of the foam. The factor of the **additional water amount** is not significant for the mathematical model. For the tensile shear strength and the shear modulus, an optimum of 5.5 % could be identified, whereby this water is very important for the foam formation. In general, a higher **wood moisture content** leads to an increase in the tensile shear strength and shear modulus. The advantages of the high wood moisture content are that there is a better foam formation, reducing costs for wood drying and the strength/stiffness will be higher. A disadvantage could be that the moisture stays in the wood, therefore the wood should have the same moisture content for which it is used later and only the surface of the wood should has a higher moisture content. The **press temperature** has a negative effect on the strength and stiffness of the PUR foam. A lower press temperature also means a longer pressing time but higher strength and stiffness. The low temperature could lead to an insufficient foam formation. The highest tensile shear strength and shear modulus were reached at a low **foam thickness**. With an increase in the thickness of the foam it comes to a strong decrease in strength and stiffness. The foam thickness has the greatest influence on the mechanical properties of the PUR foam, which is important to know for producing a material made out of the foam.

For further studies the foam distribution and the foam structure could be of central importance for the mechanical properties of the PUR foam.

5 Register of Illustrations

6 List of Tables

7 References

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8 Appendix

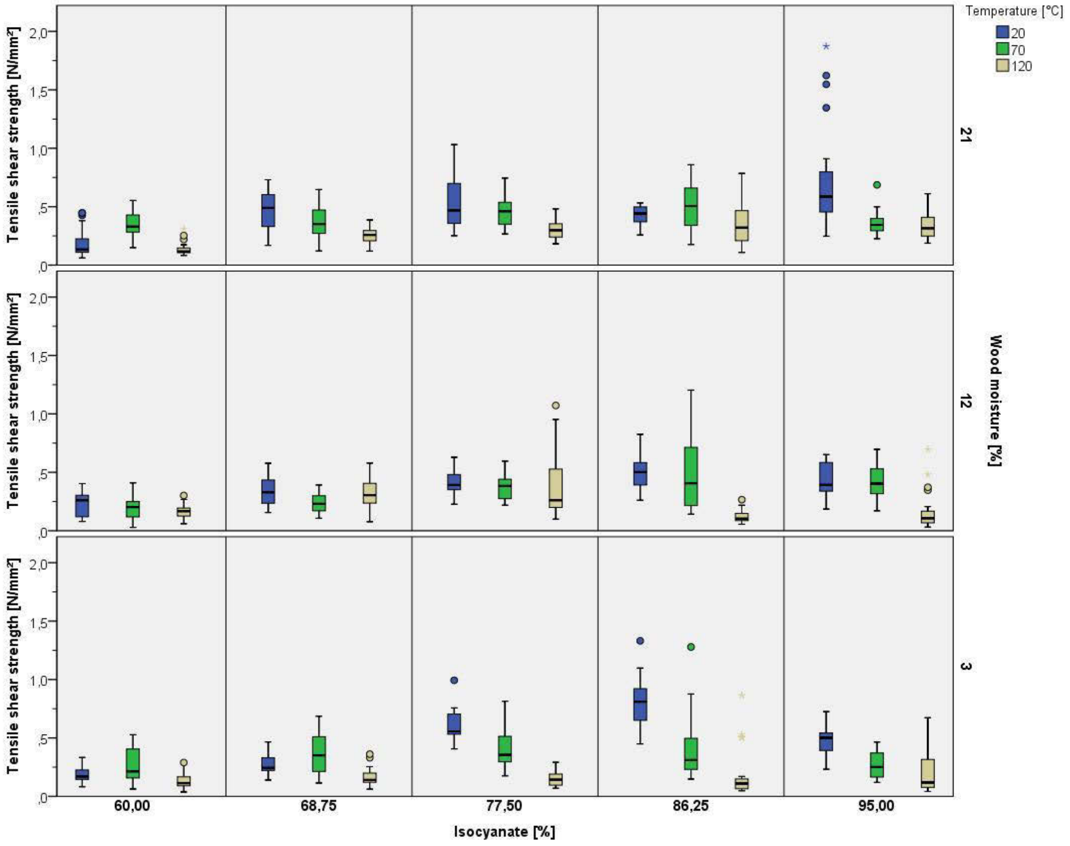


Figure 50: Tensile shear strength of samples with a foam thickness of 2 mm (max. n=24 samples per boxplot)

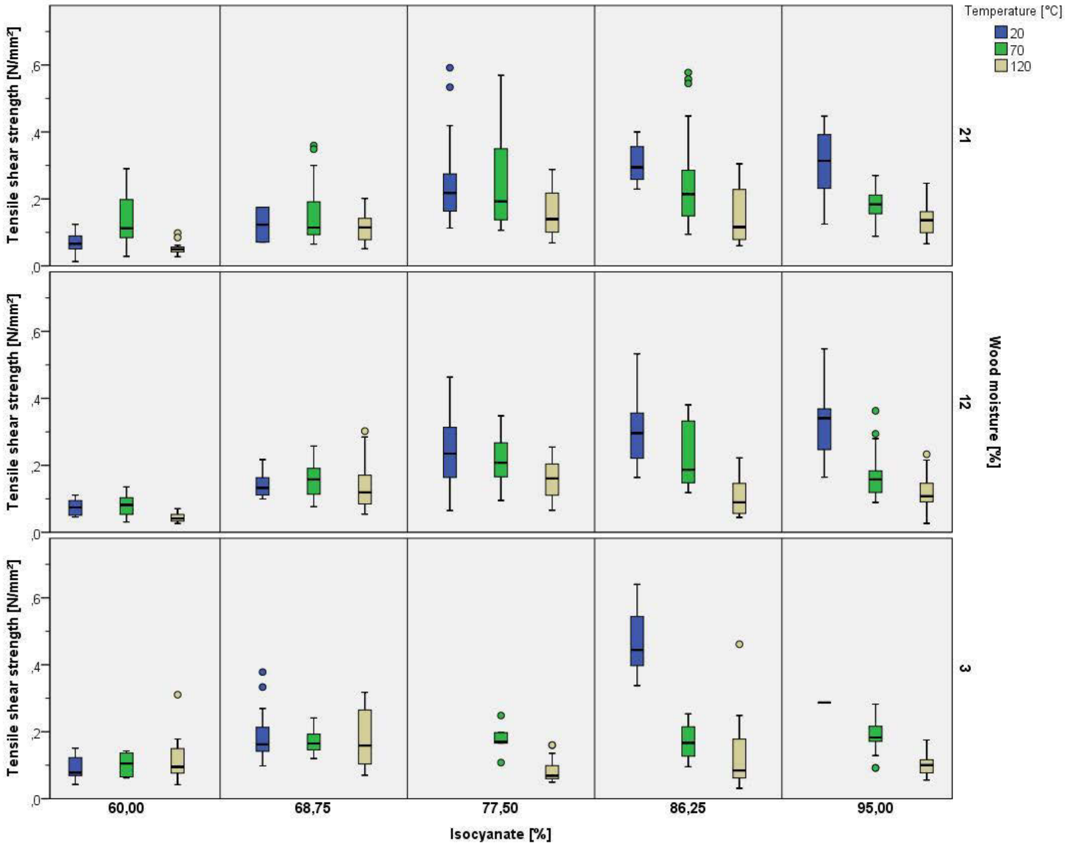


Figure 51: Tensile shear strength of samples with a foam thickness of 3.5 mm (max. n=24 samples per boxplot)

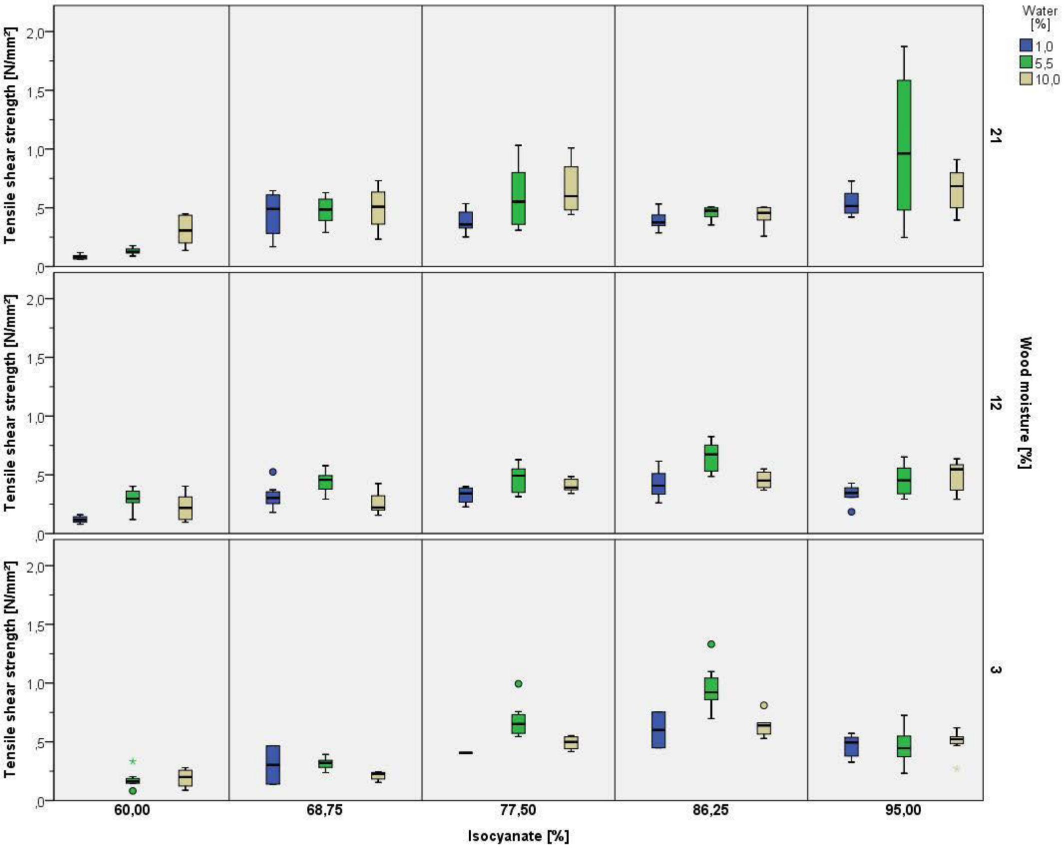
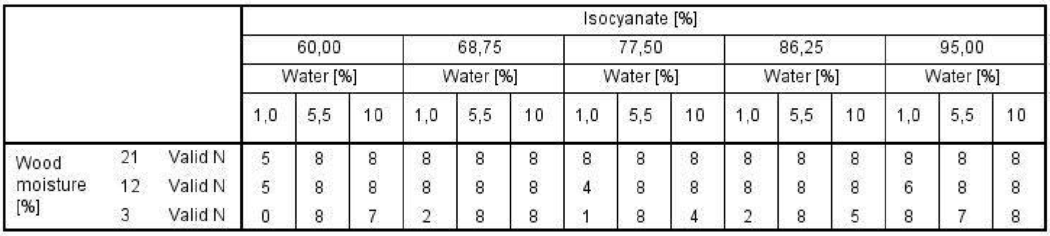


Figure 52: Tensile shear strength of samples with a foam thickness of 2 mm at a press temperature of 20 °C (valid number of samples in Table 19)

Table 19: Valid number of samples at a foam thickness of 2 mm and a press temperature of 20 °C



			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Water [%]			Water [%]			Water [%]			Water [%]			Water [%]		
			1,0	5,5	10	1,0	5,5	10	1,0	5,5	10	1,0	5,5	10	1,0	5,5	10
Wood moisture [%]	21	Valid N	5	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	12	Valid N	5	8	8	8	8	8	4	8	8	8	8	8	6	8	8
	3	Valid N	0	8	7	2	8	8	1	8	4	2	8	5	8	7	8

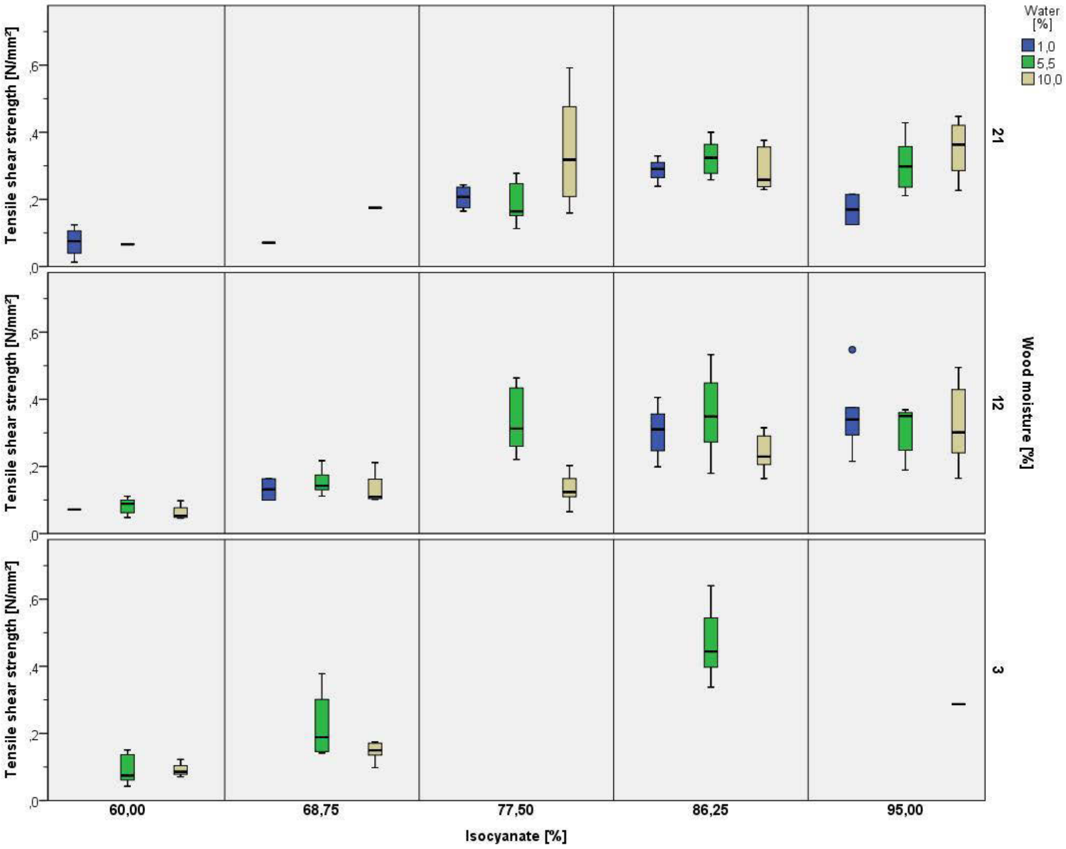


Figure 53: Tensile shear strength of samples with a foam thickness of 3.5 mm at a press temperature of 20 °C (valid number of samples in Table 20)

Table 20: Valid number of samples at a foam thickness of 3.5 mm and a press temperature of 20 °C

			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Water [%]			Water [%]			Water [%]			Water [%]			Water [%]		
			1,0	5,5	10	1,0	5,5	10	1,0	5,5	10	1,0	5,5	10	1,0	5,5	10
Wood moisture [%]	21	Valid N	8	1	0	1	0	1	4	8	8	3	8	6	2	6	8
	12	Valid N	1	8	4	2	8	4	0	8	5	6	8	8	6	8	8
	3	Valid N	0	8	3	0	8	6	0	0	0	0	8	0	0	0	1

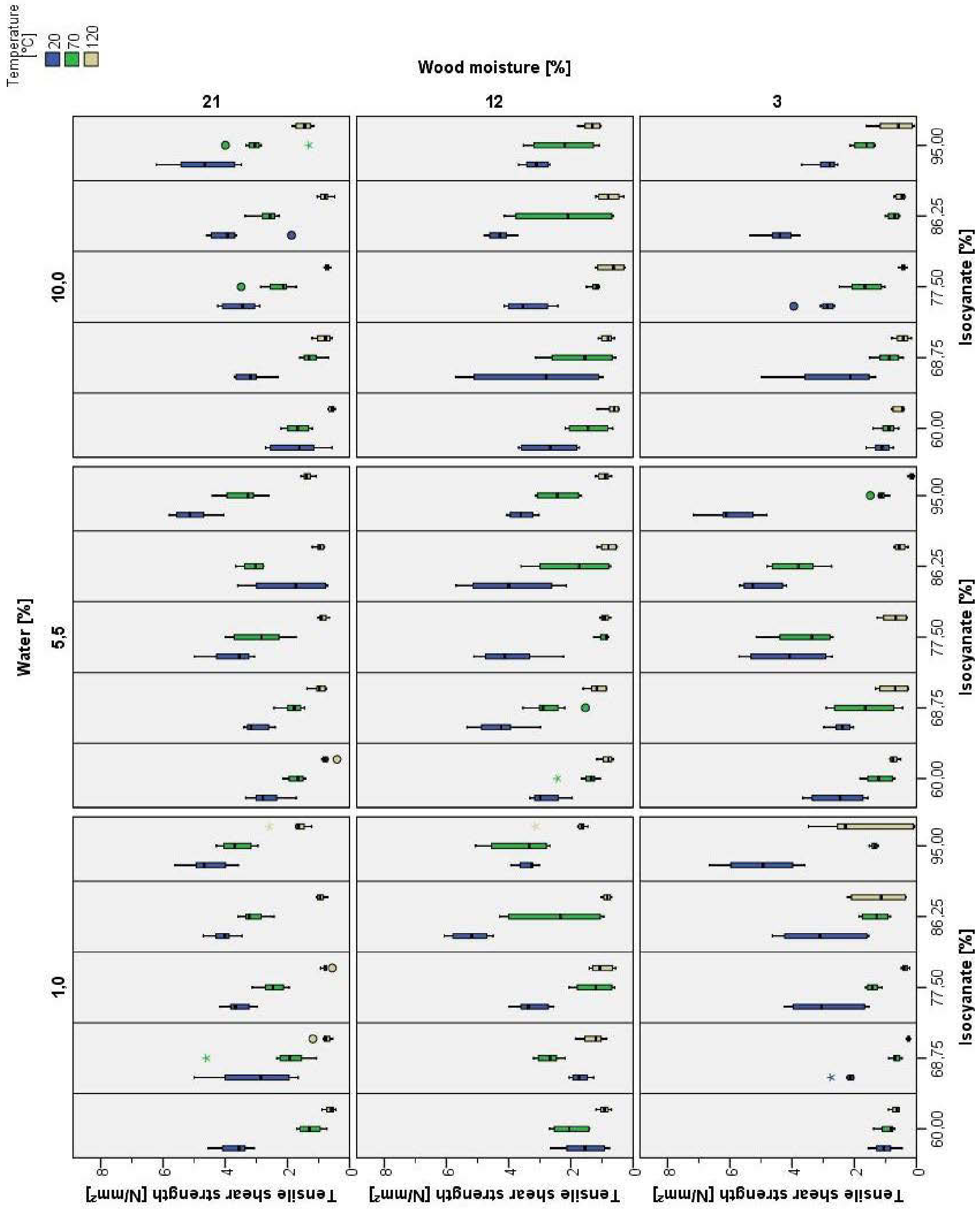
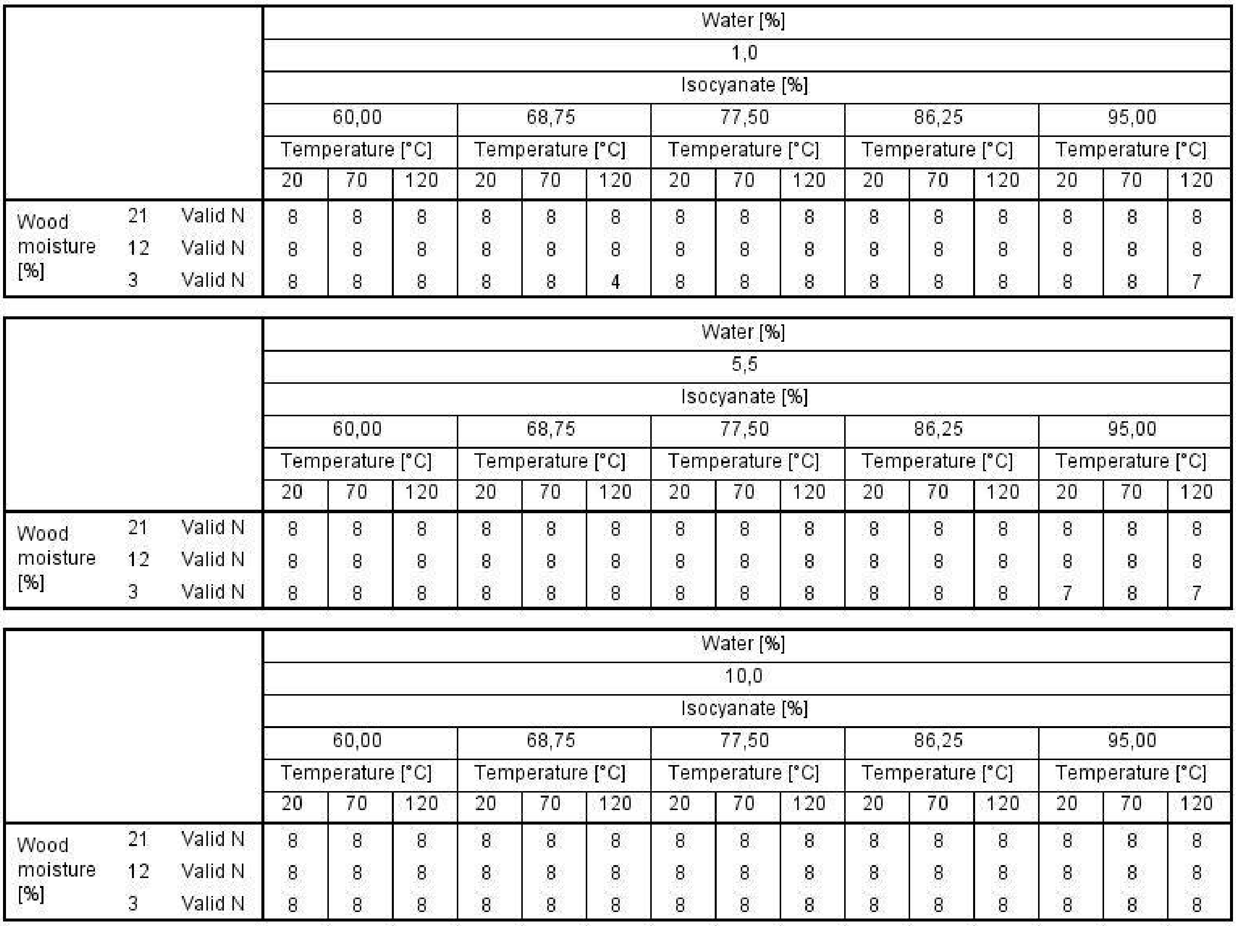


Figure 54: Tensile shear strength of samples with a foam thickness of 0.5 mm (valid number of samples in Table 21)

Table 21: Valid number of samples at a foam thickness of 0.5 mm



			Water [%]														
			1,0														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	12	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	3	Valid N	8	8	8	8	8	4	8	8	8	8	8	8	8	8	7

			Water [%]														
			5,5														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	12	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	3	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	7	8	7

			Water [%]														
			10,0														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	12	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	3	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8

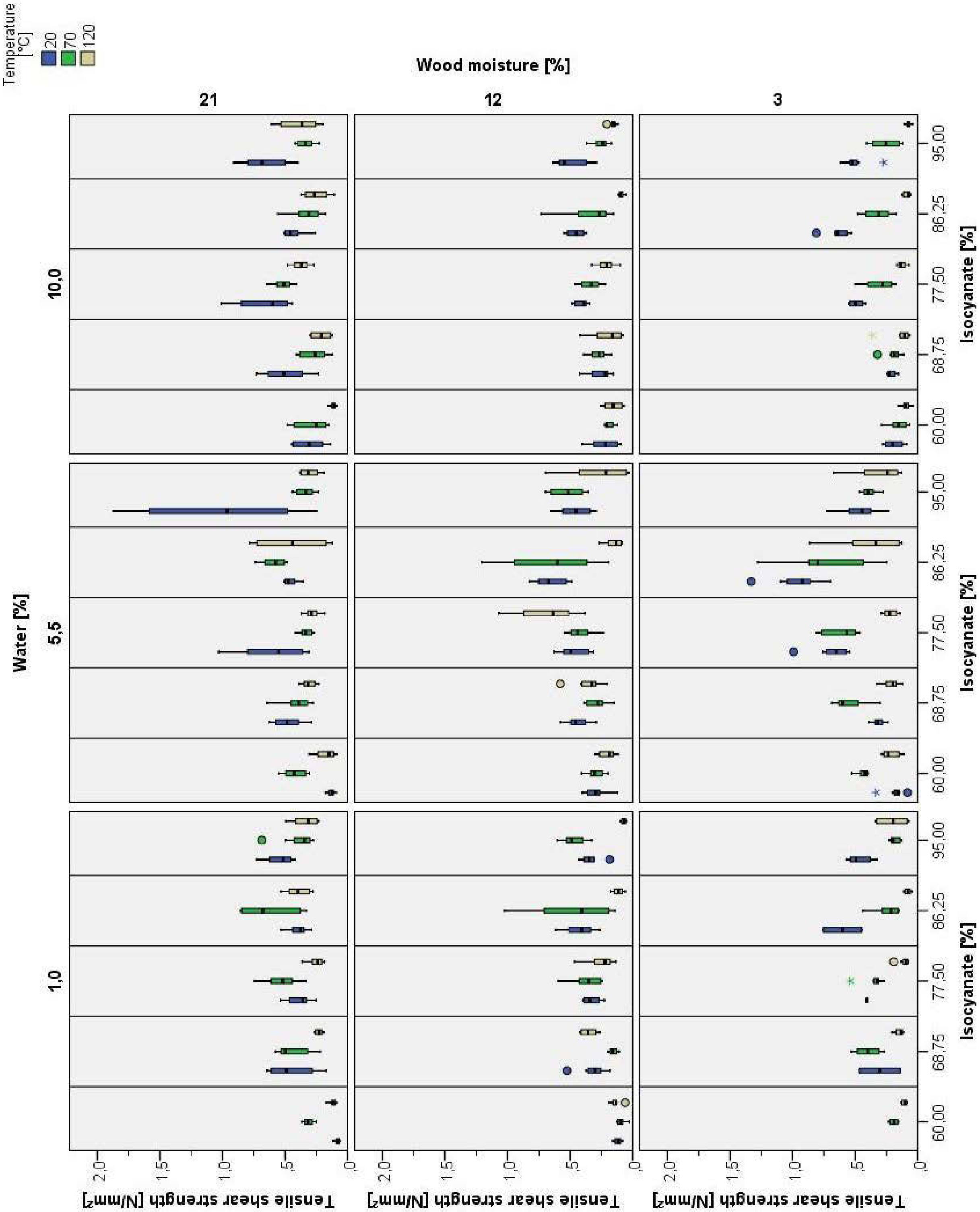
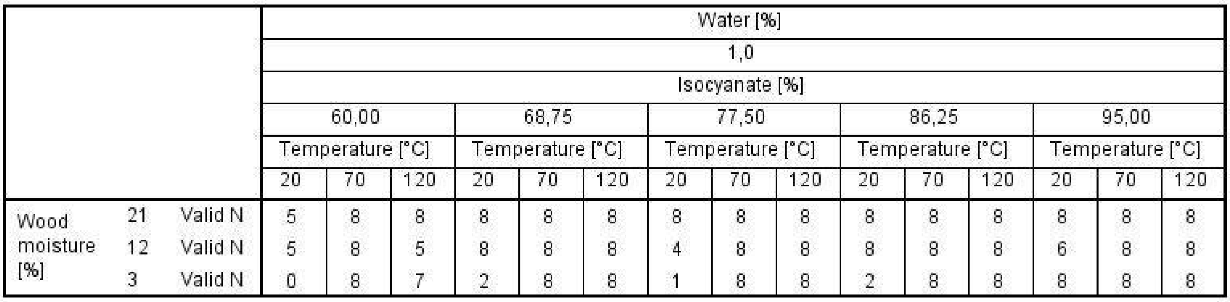
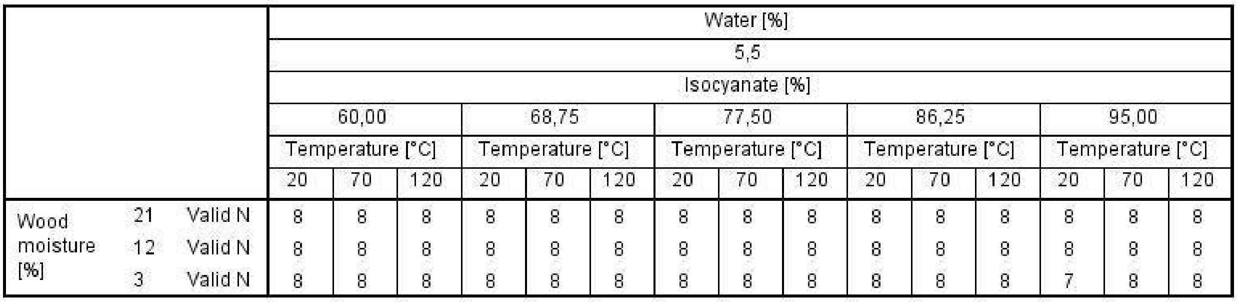


Figure 55: Tensile shear strength of samples with a foam thickness of 2 mm (valid number of samples in Table 22)

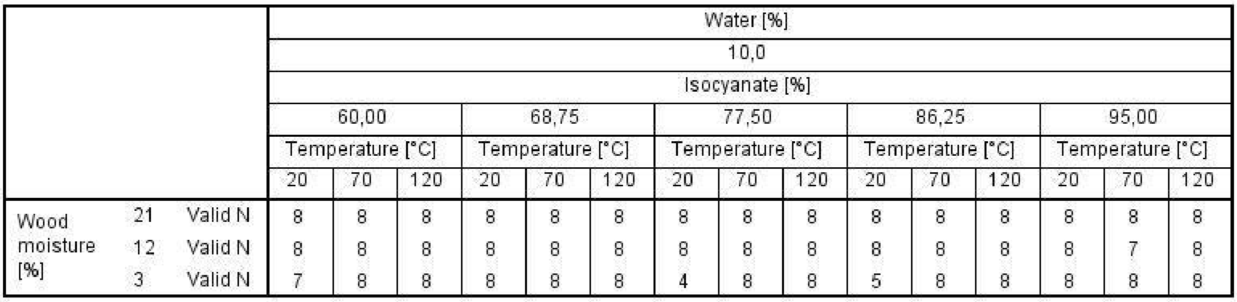
Table 22: Valid number of samples at a foam thickness of 2 mm



			Water [%]														
			1,0														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	5	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	12	Valid N	5	8	5	8	8	8	4	8	8	8	8	8	6	8	8
	3	Valid N	0	8	7	2	8	8	1	8	8	2	8	8	8	8	8



			Water [%]														
			5,5														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	12	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	3	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	7	8	8



			Water [%]														
			10,0														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	8	8
	12	Valid N	8	8	8	8	8	8	8	8	8	8	8	8	8	7	8
	3	Valid N	7	8	8	8	8	8	4	8	8	5	8	8	8	8	8

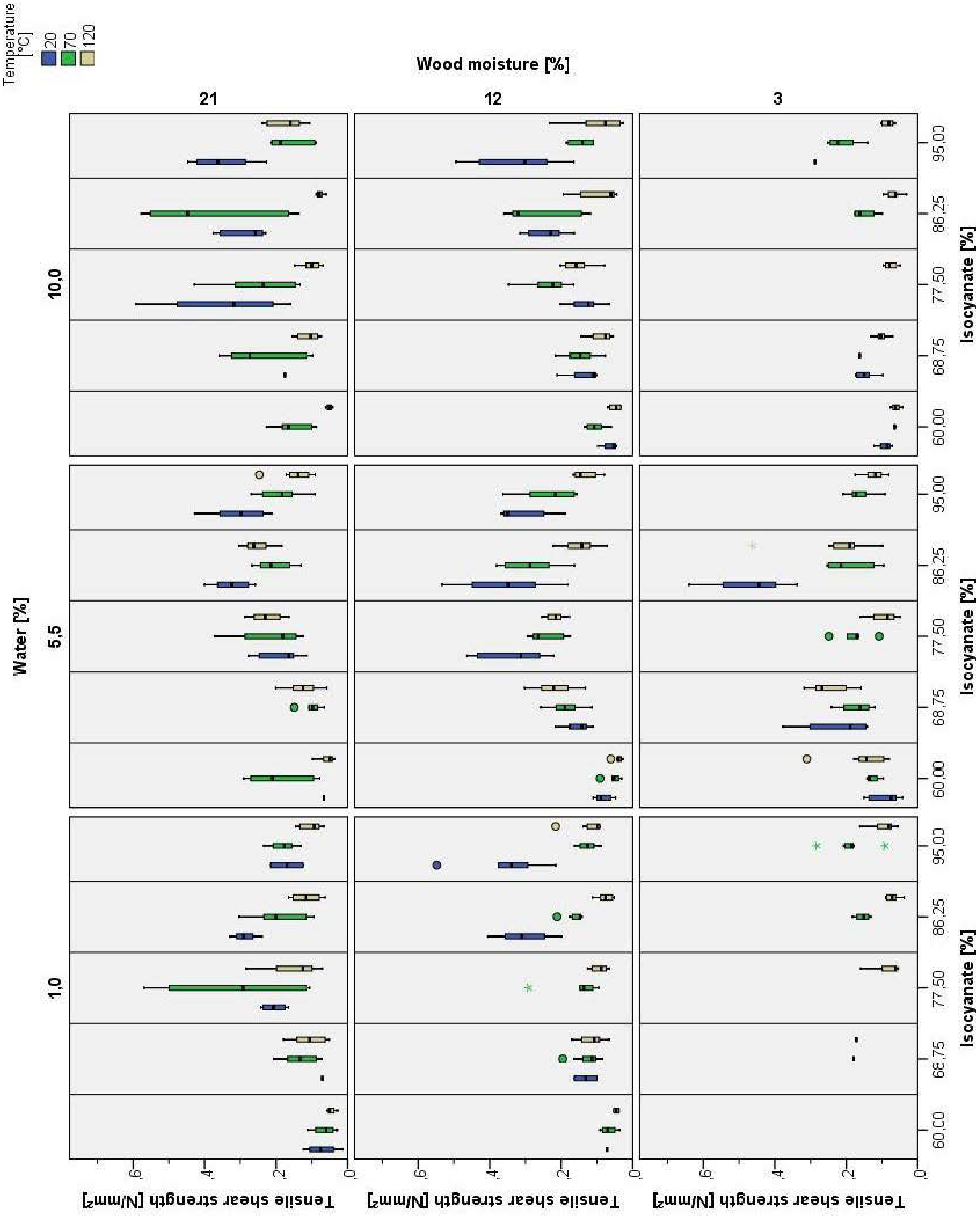
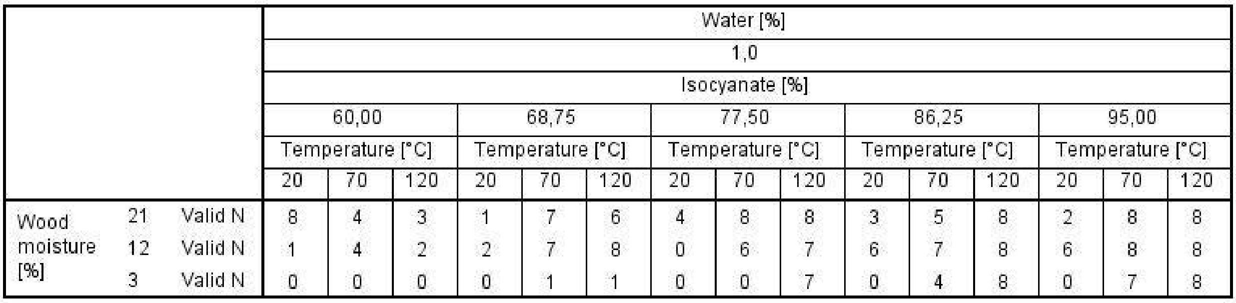
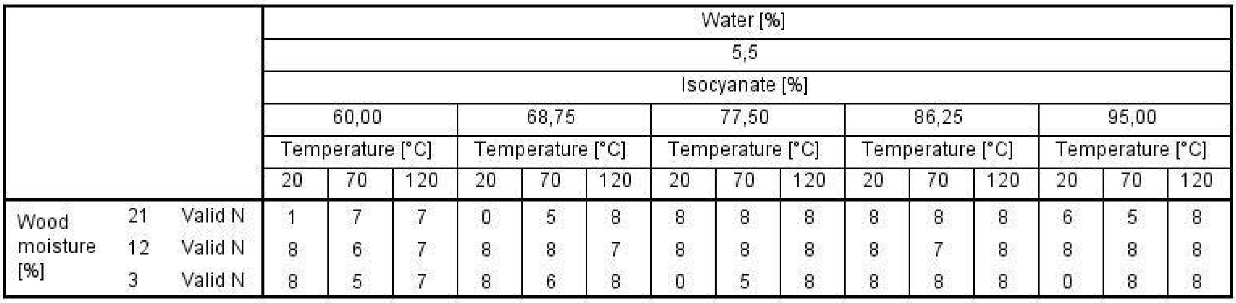


Figure 56: Tensile shear strength of samples with a foam thickness of 3.5 mm (valid number of samples in Table 23)

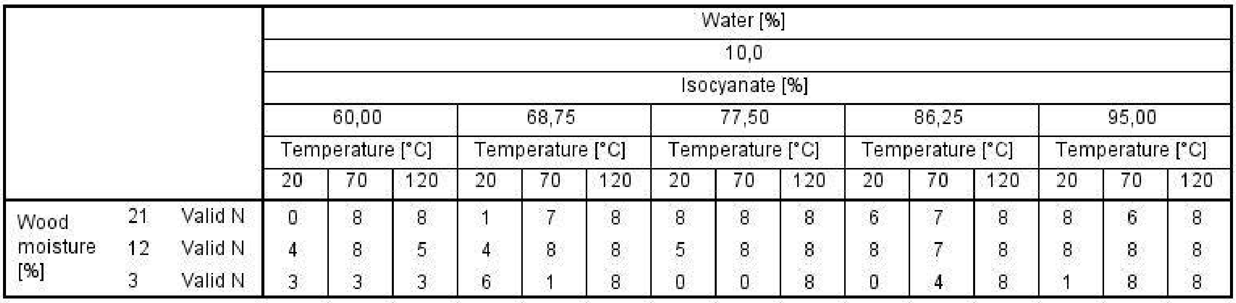
Table 23: Valid number of samples at a foam thickness of 3.5 mm



			Water [%]														
			1,0														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	8	4	3	1	7	6	4	8	8	3	5	8	2	8	8
	12	Valid N	1	4	2	2	7	8	0	6	7	6	7	8	6	8	8
	3	Valid N	0	0	0	0	1	1	0	0	7	0	4	8	0	7	8



			Water [%]														
			5,5														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	1	7	7	0	5	8	8	8	8	8	8	8	6	5	8
	12	Valid N	8	6	7	8	8	7	8	8	8	8	7	8	8	8	8
	3	Valid N	8	5	7	8	6	8	0	5	8	8	8	8	0	8	8



			Water [%]														
			10,0														
			Isocyanate [%]														
			60,00			68,75			77,50			86,25			95,00		
			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]			Temperature [°C]		
			20	70	120	20	70	120	20	70	120	20	70	120	20	70	120
Wood moisture [%]	21	Valid N	0	8	8	1	7	8	8	8	8	6	7	8	8	6	8
	12	Valid N	4	8	5	4	8	8	5	8	8	8	7	8	8	8	8
	3	Valid N	3	3	3	6	1	8	0	0	8	0	4	8	1	8	8

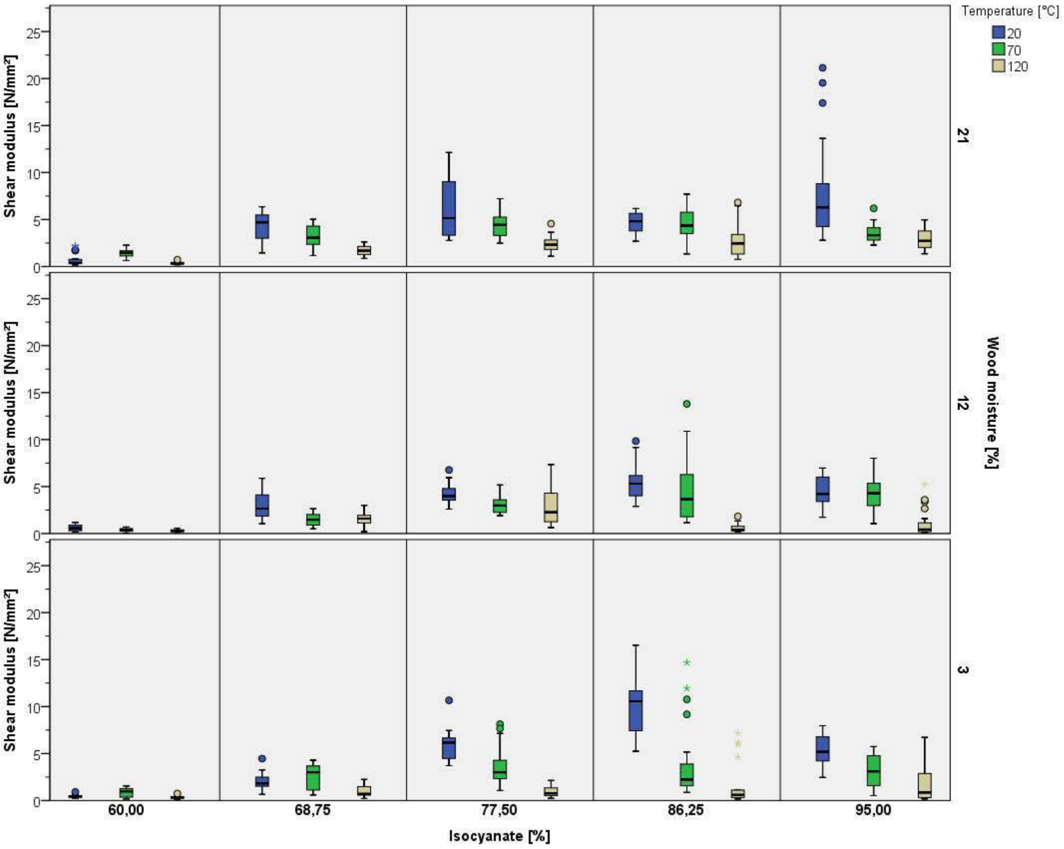


Figure 57: Shear modulus of samples with a foam thickness of 2 mm (max. n=24 samples per boxplot)

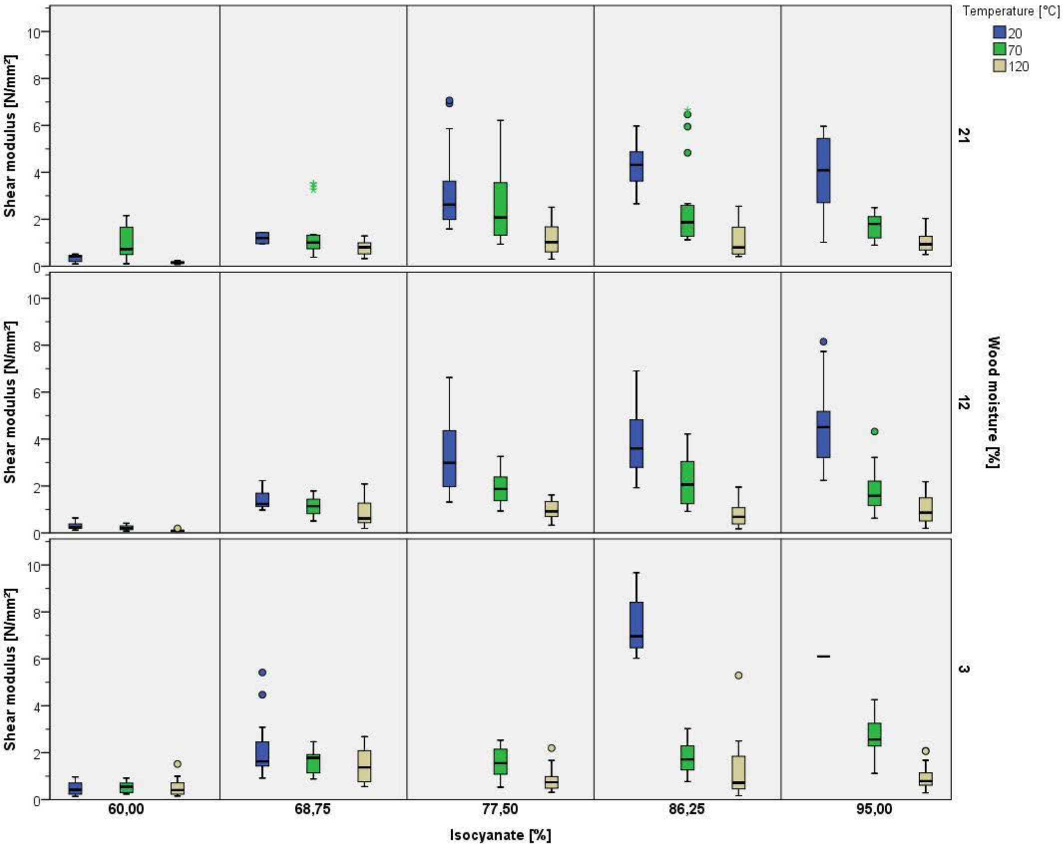


Figure 58: Shear modulus of samples with a foam thickness of 3.5 mm (max. n=24 samples per boxplot)

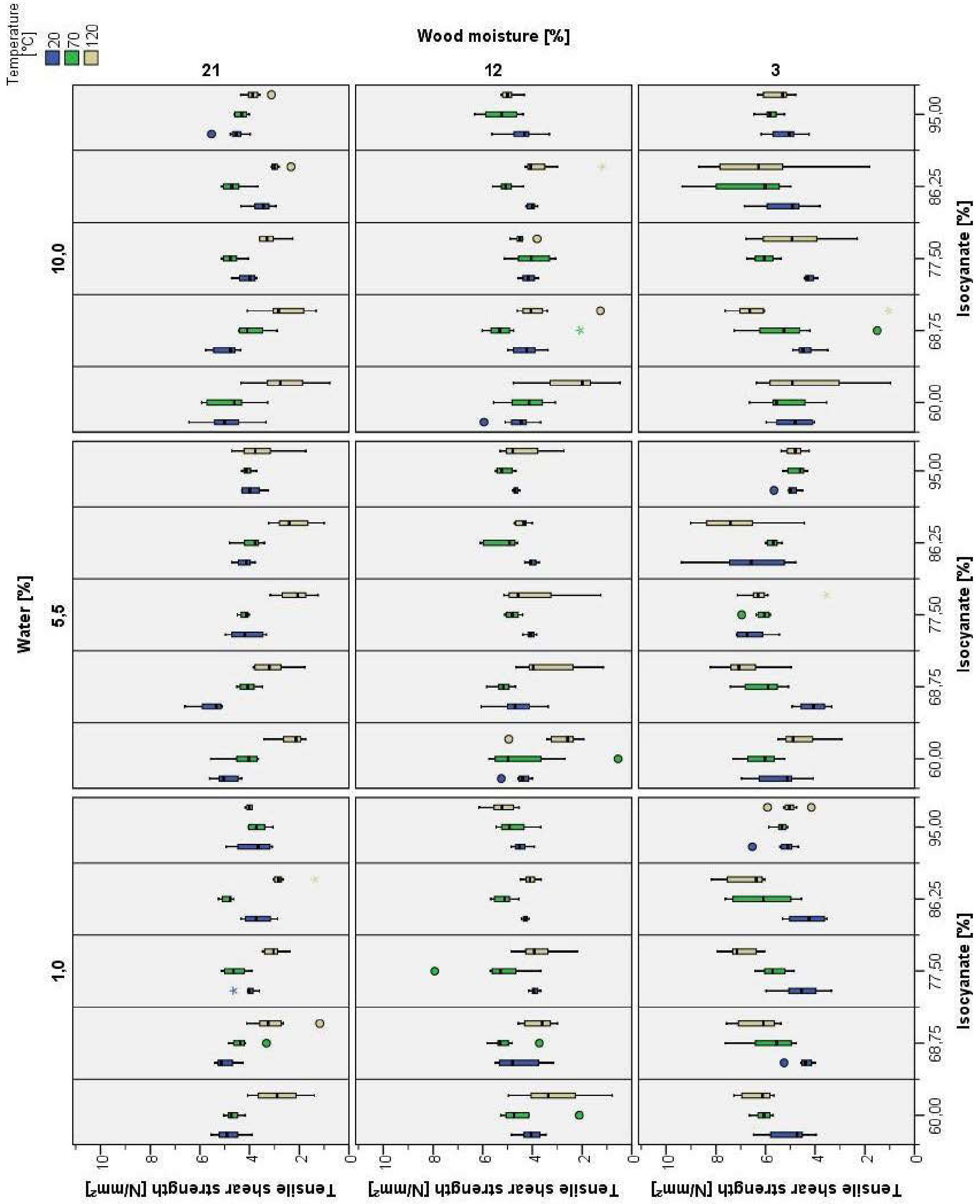


Figure 59: Tensile shear strength of the solid wood bonding (division by colour is the temperature; max. n=8 samples per boxplot)

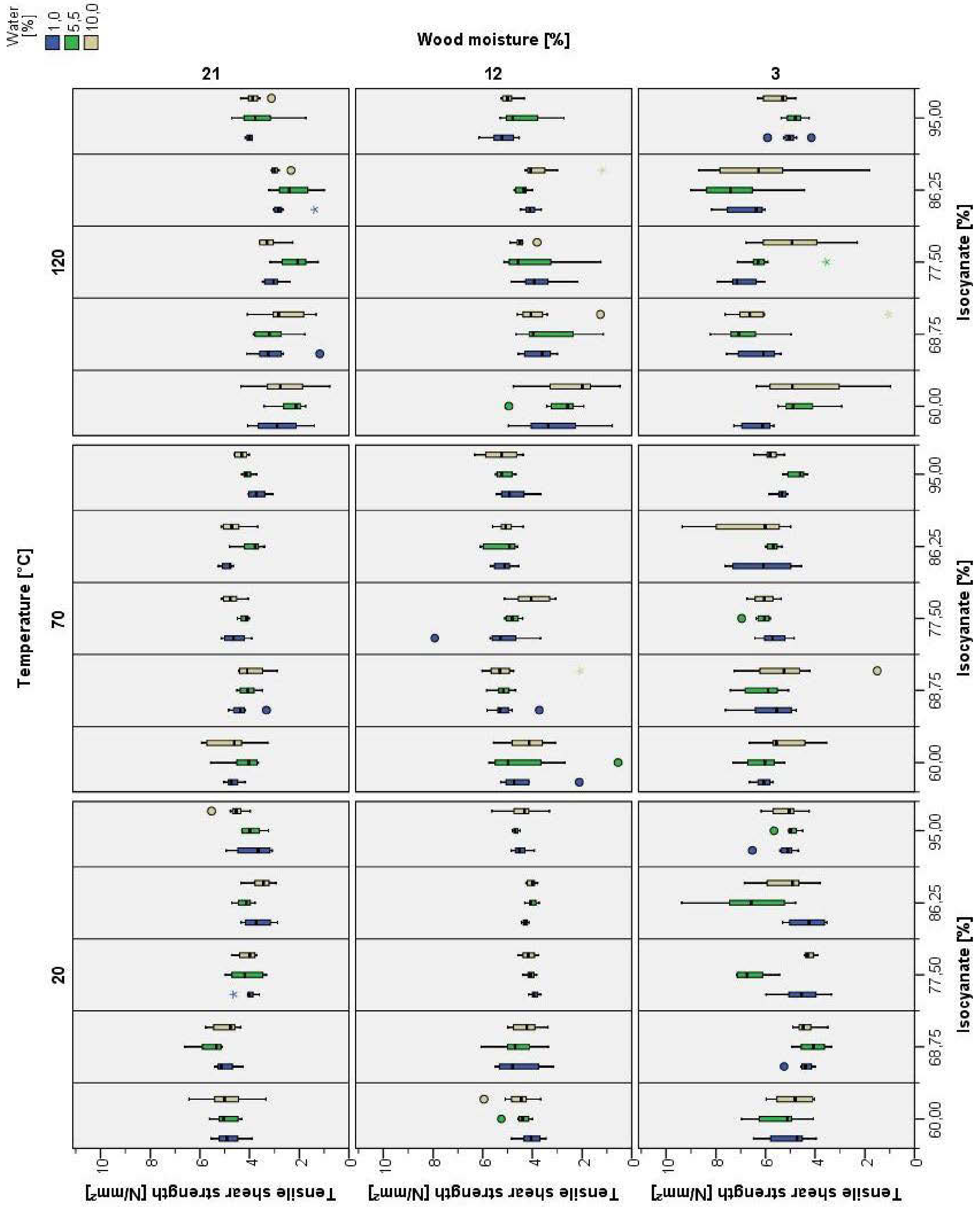


Figure 60: Tensile shear strength of the solid wood bonding (division by colour is the amount of the additional water; max. n=8 samples per boxplot)

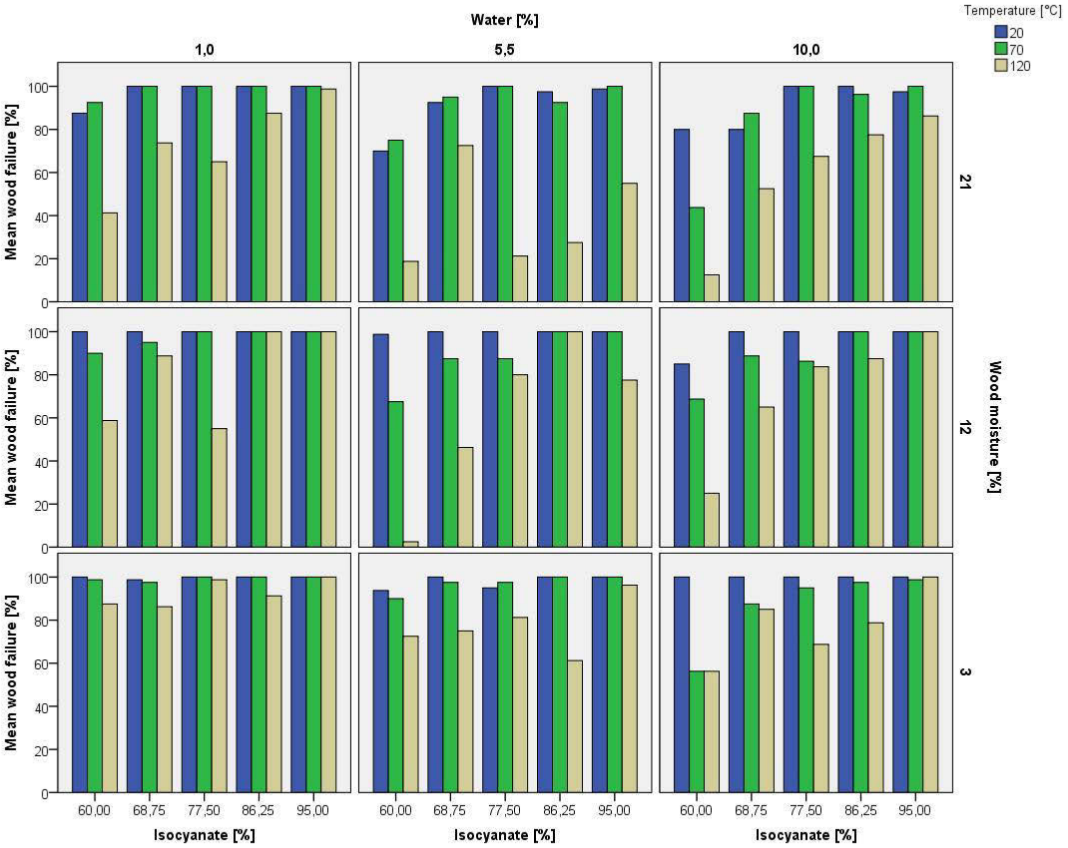


Figure 61: Mean wood failure of the solid wood bonding samples (max. n=8 samples per bar)