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# Dynamic mechanical properties of decorative papers impregnated with melamine formaldehyde resin

## Dynamisch mechanische Eigenschaften von mit Melamin-Formaldehyd-Harz imprägnierten Dekorpapieren

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### Abstract

Papers impregnated with melamine formaldehyde based resins are widely used in decorative surface finishing of engineered wood based panels for indoor and outdoor applications. For cost-effective production of high-quality impregnated papers it is of great importance to understand the complex interplay between manufacturing conditions and technological property profile. In the present study, three raw papers from different suppliers were impregnated with melamine formaldehyde resin in an industrial scale experiment to study the influence of some important manufacturing variables on the processability of impregnated papers. As numerical factors the resin loading, the final moisture content and the amount of curing catalyst were systematically varied according to a statistical central composite design. The model papers were analyzed for their rheological and thermal properties using the dynamic mechanical method developed by Golombek. As target values flow time, cure time, curing rate and flexibility were used to calculate quantitative models for the processability of the impregnated papers using response surface methodology. It is shown that the relevant rheological and thermal paper parameters are significantly influenced by the supplier of the raw paper as well as the manufacturing variables.

### Zusammenfassung

Melaminharz imprägnierte Dekorpapier werden häufig in der Oberflächenveredelung von plattenförmigen Holzwerkstoffen für Innen- und Außenanwendungen verwendet. Für die kosteneffiziente Produktion von qualitativ hochwertigen imprägnierten Papieren ist es nötig, das komplexe Zusammenspiel zwischen den industriellen Herstellungsbedingungen und dem technologischen Eigenschaftsprofil der Materialien zu verstehen. In der vorliegenden Studie wurden drei vergleichbare Rohpapiere von unterschiedlichen Herstellern im Rahmen eines industriellen Experiments unter verschiedenen Bedingungen mit Melaminharz imprägniert, um den Einfluss einiger

wichtiger Prozessvariablen auf die Herstellung von imprägniertem Papier zu analysieren. Die Modellpapiere wurden auf ihre rheologischen und dynamisch-mechanischen Eigenschaften nach der Methode von Golombek untersucht. Als Zielgrößen wurden Fließzeit, Härtingszeit, Härtinggeschwindigkeit und Flexibilität bestimmt und der Einfluss der Produktionsvariablen wurde nach Response Surface-Methoden quantifiziert. Es wird gezeigt, dass die relevanten rheologischen und dynamisch-mechanischen Papierparameter signifikant vom Rohpapierhersteller und den Produktionsbedingungen abhängen.

## 1. Introduction

Paper sheets impregnated with an aminoplastic thermosetting resin are frequently used for the surface protection and decoration of medium density fibreboards (MDF) and particleboards in the furniture (Kandelbauer et al. 2008, Nemli and Colakoglu 2005, Soiné 1995) and the laminate flooring (Kalaycioglu and Hiziroglu 2006, Bauer and Kandelbauer 2004) industries. Such paper sheets are typically core impregnated with urea formaldehyde (UF) or melamine formaldehyde (MF) resin in a first step and surface coated with MF resin in a second step (Bader et al. 2000, Ruhdorfer 1980) using a paper impregnation machine such as the one designed by Vits Systems GmbH (Vits 2007, 2001), (Figure 1). After impregnation, the paper is dried to a final moisture content of 6 – 9 % and subsequently pressed onto the carrier board at temperatures around 180 °C. Since the impregnation resin is not completely cured during paper impregnation there is no additional adhesive required for the surface finishing of the boards.

Figure 1: Schematic representation of the impregnation process of papers for decorative laminates

The technological properties of impregnated papers must not only fulfil numerous requirements with respect to the final product such as durable surface films, hardness, temperature and chemical resistance etc (cf. for example EN 13 329). They must also fit the demands of the laminates manufacturer in terms of processability such as sufficiently long shelf-life, complete curing during rapid pressing, no blocking when stored in a stack, homogenous film formation etc. The properties of the finished panel product as well as of the intermediate paper sheet are governed by the manufacturing conditions of impregnated papers. Important process parameters that need to be carefully adjusted during the manufacture of such papers are the composition of the resin solution, the numerous roller parameters, the drier conditions and the type of paper used as raw material.

One of the most important components of the impregnation solution is the curing catalyst (Becker and Braun 1988). The reactivity of the catalyst governs the speed at which impregnated papers can be processed in the laminating step (Barash 2008). Very reactive systems may cause the resin to pre-cure during impregnated paper manufacture, leading to bad gluability. The reactivity of impregnated paper needs therefore to be carefully adjusted. The drier conditions are important since they govern the cross-linking of the impregnation resin. If set too harsh the required self-gluing property of the paper may be lost rendering the product unusable.

The amount of resin is another relevant factor. To reduce the costs, low resin loads are desired. However, if too low amounts of resin are applied the paper is not saturated during core impregnation, the pores are not completely filled and subsequently applied coating resin may sink in leaving a defective surface film (Roberts and Evans 2004). Finally, the paper properties of the raw paper such as density, wet strength or ash content may strongly influence the production process. For instance, paper density affects resin penetration and indirectly determines production speed. Although testing methods have been devised to determine the penetration behaviour of paper/resin systems (see for example <http://www.emtec-papertest.com/deutsch/index.html>), little is known about the interrelation of raw paper and the properties of impregnated paper. The rheological and thermal properties of impregnated paper such as paper flexibility, resin flow and curing characteristics of MF are very important for evaluating its quality and

processability. For instance, the resin flow at the surface of impregnated paper governs film formation and the final surface properties of the coated board. Too low resin flow causes an inhomogeneous film leading to optical defects like low gloss, stains, flow marks or bad cleanability. Resin flow, paper flexibility as well as reactivity of impregnated papers can be quantitatively determined using dynamic mechanical measurements, and thermal analysis of impregnated papers may serve as a rich source of information on both the processability and the final film properties of impregnated paper. In the present contribution the rheological and dynamic mechanical properties of impregnated papers and their relevance for the impregnation process are investigated and discussed. It is aimed at a quantitative mathematical model for the prediction of the processability of impregnated paper sheets under varying manufacturing conditions. In a large-scale industrial experiment at an Austrian impregnation factory the four major process variables raw paper type, resin loading, catalyst level and final moisture content were varied. As response the dynamic mechanical behaviour of the impregnated papers was studied and the effects of process variables on several rheological and thermal paper parameters were quantified using response surface methodology. The used dynamic mechanical descriptors are routinely used for evaluation of the processability of impregnated papers. Hence, the current study gives insights in the predictive modelling of the processability of impregnated papers in dependence of the process conditions.

## 2. Materials and methods

### Chemicals

For paper impregnation, industrial melamine formaldehyde (MF) resin containing various levels of a commercial curing catalyst was used. The impregnation resin was freshly prepared at the Impress Décor Austria GmbH impregnation plant in St. Veit/Glan, Austria and used directly in the impregnation experiments at the industrial machinery.

### Paper

The raw papers used for the present study were all kraft papers of the same type but obtained from three different suppliers, namely Hoffsümmmer (raw paper 1), Smurfit (raw paper 2) and Technocell (raw paper 3). They are typical for the use in decorative laminates and are often interchangeably applied for the same type of product. Table 1 summarizes their technological properties.

### Preparation of impregnated paper samples

All impregnation experiments were performed with the industrial paper treatment plant at Impress Décor Austria GmbH in St. Veit/Glan, Austria. As factors, the amount of curing catalyst in the impregnation resin mixture, the final moisture content of impregnated paper, the raw paper type and the resin loading were systematically varied. The amount of curing catalyst and the remaining moisture content each were varied on 5 levels according to a regular central composite design for all three papers at an intermediate resin loading of  $135 \text{ gm}^{-2}$ . The limits for catalyst concentration were set to 0.5 and 1.5 % (w/w) with respect to the resin dry mass. The drier conditions were adjusted such that the final moisture content of the impregnated paper was regularly varied between the limits of 6 and 9 %. Additionally, supplemental papers were impregnated at centre and vertices points under variation of the resin load between 230 and  $255 \text{ gm}^{-2}$  final paper weights to account for effects of resin loading. Samples of  $30 \times 30 \text{ cm}^2$  were cut from the industrial formats, wrapped in plastic and stored at room temperature and 50 % humidity until further use.

## Dynamic mechanical measurements

Dynamic mechanic parameters of the impregnated papers were determined using the paper sheet tester developed by Golombek (Golombek 1991; Figure 2a). This tester is widely used in the laminate industry and consists of an oil bath and a vertically adjustable sample holder coupled to a reversible rotary drive. The motor is connected to a galvanometer which allows continuous monitoring of the motor current required to move the paper sample.

Figure 2a: Schematic representation of the measurement principle for dynamic mechanical properties of impregnated paper. *a ... Thermostat; b ... Inert silicone oil; c ... Paper sample; d ... Sample holder; e ... Rotary drive; f ... Data processor*

Small sample sheets (96 x 60 mm<sup>2</sup>) of impregnated paper were cut from the stored sheets and immersed in a hot silicone oil bath at 140 °C and the paper was cured. Oscillatory movements were performed at a frequency of 1 rpm and the consumed electrical current for performing the oscillations was recorded in dependence of time. While the impregnated paper was exposed to the hot silicone oil, resin curing took place and the stiffness of the paper increased due to cross-linking of MF resin. This increase was recorded during the test based on electrical current measurement. Figure 2b shows the time course of motor current versus time during a typical Golombek experiment.

Figure 2b: Typical response curve of dynamic mechanical measurement using the Golombek-apparatus. IP ... Inflection point;  $t_{flow}$  ... Flow time;  $t_{cure}^{max}$  ... Time point of maximum curing rate;  $t_{cure}$  ... Time for 95 % cross-linking of MF resin;  $r_{cure}$  ... Maximum curing rate

The initial current value reflects the force required to move the not yet fully warmed, rather stiff paper sample. As the resin liquefies after being exposed to 140 °C the paper sample softens and its resistance against circular motion declines and correspondingly the energy consumption of the motor decreases. After passing through a minimum the current consumption increases since curing of the impregnation resin causes the paper stiffness to rise. At the inflection point (IP) the curing rate reaches its maximum and slows down again until the resin is completely cross-linked. The tests were finished when the impregnation resin was completely cured as indicated by no further increase in electric current. This typically was the case after approx. 5 minutes.

From the curve shown in Figure 2b several rheological and thermal parameters were obtained such as the flow time  $t_{flow}$ , the curing rate  $r_{cure}$ , the time of maximum cure rate  $t_{cure}^{max}$ , the curing time  $t_{cure}$  and the flexibility  $f$ . Flow time,  $t_{flow}$ , is the time at 140 °C until the curing of the liquefied resin in the impregnated paper starts to significantly increase the current response. It is defined in relation to the minimum response as illustrated in Figure 2b. The curing rate,  $r_{cure}$ , is defined as the slope of the curve at the inflection point (IP, Figure 2b). Correspondingly,  $t_{cure}^{max}$ , represents the time where the curing process proceeds at the highest rate and is defined as the intercept with the x-axis at the inflection point. The curing time,  $t_{cure}$ , is defined as the time at which 95 % of the cross-linking has occurred. Flexibility,  $f$ , is the percentage of cross-linking that can be reached with the liquefied resin. For its definition, also refer to Figure 2b. All parameters describe impregnated papers well and allow drawing conclusions on their technological performance.

All dynamic mechanical measurements were repeated twice for each sample. The average values of each response were used for response surface analysis (Myers and Montgomery 2002). In the current paper, the statistical analysis was focussed on  $t_{flow}$ ,  $t_{cure}$ ,  $r_{cure}$  and  $f$ . Since the remaining parameter  $t_{cure}^{max}$  yields no additional information it was considered redundant for the presented data material.

For the statistical evaluation and response surface modelling of the results the computer software Design Expert (Stat-Ease Inc., Minneapolis, MN, USA) was used. For all three categorical levels studied a quantitative mathematical model was calculated and validated with randomly chosen experimental conditions within the limits of the design which were produced subsequently to the industrial experiment at the paper impregnation plant.

### 3. Results and discussion

#### Raw paper properties

Prior to analyzing the dynamic mechanical properties of impregnated papers, the three investigated raw papers were characterized. Although their average paper weight was always the same, the papers supplied from different manufacturers varied significantly in some of their technological properties. Some important characteristics of the various raw papers are summarized in Table 1.

Wet strength (WS) is one of the most important parameters of raw papers intended for impregnation with aqueous melamine resins for decorative laminates. The wet strength strongly influences the production speed at which a certain paper may be processed without paper breaks. During impregnation, the papers are wetted and soaked with aqueous dispersions of resin. In a typical impregnation plant, papers are processed with speeds between 50 and 100  $\text{mm}^{-1}$  and they must withstand the forces of numerous rapidly spinning roller coating cylinders in a wet state. Thus, in principle a higher wet strength especially in production direction,  $WS_{\parallel}$ , allows for safer production at higher impregnation speeds.

In the present case, RP 1 had clearly the highest values for wet strength whereas both  $WS_{\parallel}$  and  $WS_{\perp}$  of RP 2 and 3 were about 50 % smaller. With respect to paper anisotropy, all papers displayed a higher wet strength in the production direction than perpendicular to it. RP 3 showed the highest ratio of the values for paper strength in parallel and perpendicular to the direction of paper production for both the wet and the dry strengths of the raw paper. RP 1 was the most anisotropic paper with the same ratio of 1.36 for both the dry and the wet strengths. The other papers were more symmetric in dry strength and showed higher ratios of  $WS_{\parallel}$  and  $WS_{\perp}$ .

The ash content of a paper is also very important for decorative papers. Since papers with high filler content contain comparatively lower amounts of swellable fibre material at the same paper weights, ash content may influence the speed of resin penetration during impregnation and the total amount of resin load a paper can take up. RP3 showed the highest content of inorganic fillers. The ash content is obviously not directly related to the mechanical strength since although the ash content of RP 1 was not significantly lower than that of RP 3, its absolute values in wet and dry strength were much higher. The pH of all papers was neutral.

#### Flow time

The flow time  $t_{flow}$  is a very important descriptor of impregnated paper since it indicates the time frame for the MF film formation on the paper surface during the curing in the hot pressing of the laminates. Papers with low  $t_{flow}$  are already pre-cured to a high degree. They might not allow the formation of a continuous and homogenous surface and hence yield laminates of poor surface quality. Thus,  $t_{flow}$  is desired to be as high as possible. Figure 3 summarizes the influences of the amount of curing catalyst and resin load on the flow time of impregnated paper as measured with the Golombek method for the raw papers from three different suppliers studied (Figure 3 a–c). Table 2 summarizes the statistical parameters of the response surface analysis. For all three papers, the flow time responded linearly to changes in catalyst amount and was also significantly dependent on the paper supplier. Resin load and final moisture content were not significant. The linear equations describing the response surfaces for  $t_{flow}$  constitute three parallel planes as the linear combination of catalyst amount ( $-9.37113[A]$ ), final moisture content ( $0.028597[B]$ ) and resin loading ( $0.048257[D]$ ) and differed only in the off-set of the planes for the three papers (9.71344 for RP1, 11.45718 for RP2 and 10.31303 for RP3, respectively; see also Figure 3abc). The accuracy of the linear model was  $R^2=0.7700$ .

The flow time was higher with lower concentrations of catalyst. Since small percentages of catalyst means a slower resin cure the resin stays in a liquefied state for a longer time and consequently the flow time increases. Higher resin loadings also caused a slight increase in flow time. This can be explained by the fact that an overall higher amount of resin will take more time to cure and thus will also display slightly longer flow times in the Golombek tester. However, within the experimental setup the variations in resin loadings

were obviously not wide enough to statistically significantly prove this expected observation.

The final moisture content of the paper after drying the impregnated paper in the second drier line does not have any significant effect on the flow time of the material (see Table 2).

On the other hand, it is interesting that the paper supplier has a significant influence on the flow time (Table 2). Although RP 1 and 3 have practically identical response surfaces, the flow times obtained with RP 2 are comparatively higher and the response surface is shifted for about two seconds towards higher values. Since raw papers 1 and 2 have the same pH of 6.9, this observation is most probably not caused by a direct catalytic effect of the paper on the curing reaction of the reaction through acid catalysis. It is more likely that the mechanical properties of the paper influence the measurement and the findings are related to differences in the stiffness of the papers (see below).

### Cure time and curing rate

The cure time  $t_{cure}$  and curing rate  $r_{cure}$  are important target values since they allow drawing conclusions on the possible manufacturing speed in laminate pressing and can be used to derive suitable press programs. Since cost efficient mass production relies on short cycle times,  $t_{cure}$  and  $r_{cure}$  should be as high as possible.

The results for both  $t_{cure}$  and  $r_{cure}$  are very similar and the amount of curing catalyst, raw paper supplier and final moisture content were the important factors. With  $r_{cure}$  the effect of paper supplier was less pronounced. Figure 4 (a, b, c) summarizes the influences of the amount of curing catalyst and the final moisture content on the cure time of impregnated paper as measured with the Golombek method for the raw papers from the three different suppliers studied. For the response surface analysis a quadratic model was applied. The accuracy of the quadratic model was  $R^2=0.9364$  ( $t_{cure}$ ) and  $R^2=0.9772$  ( $r_{cure}$ ). Table 2 summarizes the corresponding statistical parameters. For raw papers 1 to 3, the final equations in terms of actual factors for  $t_{cure}$  after impregnation are:

$$t_{cure, RP1} = -22.33365 + 0.52308[A] - 0.55043[B] + 0.20787[D] + 0.87688[A]^2 - 0.011021[B]^2 - 4.56582 \cdot 10^{-4}[D]^2 - 0.024994[A][B] - 0.010630[A][D] + 3.16365 \cdot 10^{-3}[B][D]$$

$$t_{cure, RP2} = -0.81333 + 0.53862[A] - 0.53591[B] + 0.20130[D] + 0.87688[A]^2 - 0.011021[B]^2 - 4.56582 \cdot 10^{-4}[D]^2 - 0.024994[A][B] - 0.010630[A][D] + 3.16365 \cdot 10^{-3}[B][D]$$

$$t_{cure, RP3} = -20.33254 + 0.33982[A] - 0.53282[B] + 0.19965[D] + 0.87688[A]^2 - 0.011021[B]^2 - 4.56582 \cdot 10^{-4}[D]^2 - 0.024994[A][B] - 0.010630[A][D] + 3.16365 \cdot 10^{-3}[B][D]$$

The most significant factor for  $t_{cure}$  was the amount of curing catalyst which was expected since the catalyst concentration will directly affect the curing kinetics and high levels of catalyst speed up the condensation.

The paper supplier was also very significant which was less expected. It is difficult to understand how papers of different provenience should exhibit any catalytic effect on the curing behaviour of MF resin since they are all of the same pH and are generally considered as an inert resin carrier matrix. The most logic explanation again is that the measurement is influenced by the different mechanical properties of the various papers (see below).

Eventually also the final moisture content does effect the curing time. Higher moisture content was associated with lower curing times which can be explained by different pre-condensation degrees of differently dried papers. Higher final moisture content means that the paper is treated more mildly during drying than is paper with a low final moisture content. Hence MF in moist paper is less pre-condensed and will take longer for the cross-linking to complete during the Golombek test.

Resin loading was not considered a statistically significant factor although higher resin loads take slightly longer to cure since the total amount of resin present in the reaction system is higher. However, within the experimental error of the design the variations in final paper weights of resin impregnated papers obviously were not pronounced enough to see the effect as statistically significant.

## Flexibility

Flexibility is another important descriptor for impregnated papers and gives an indication of its cross-linking status. In highly flexible papers the degree of curing during lamination is high and hence a large portion of the MF remains yet uncured. On the other hand if  $f$  is too low, the remaining activity of MF may not be sufficient for proper lamination. Values of  $f < 80$  typically result in a deficient surface finish.

Figure 5 summarizes the influences of resin loading and the final moisture content on the flexibility  $f$  of impregnated paper at low (Figures 5 a, b and c) and high (Figures 5 d, e and f) amounts of curing catalyst as measured with the Golombek method for the raw papers from the three different suppliers studied. For the response surface analysis a quadratic model was applied. The accuracy of the quadratic model was  $R^2=0.9658$ . Table 2 summarizes the corresponding statistical parameters. For raw papers 1 to 3, the final equations in terms of actual factors for  $f$  after impregnation are:

$$f_{RP1} = -1868.08355 + 110.92874[A] + 10.68553[B] + 15.27254[D] - 19.67073[A]^2 - 1.02168[B]^2 - 0.031080[D]^2 + 3.35986[A][B] - 0.42632[A][D] + 0.025223[B][D]$$

$$f_{RP2} = -1838.39170 + 106.23929[A] + 9.77641[B] + 15.20582[D] - 19.67073[A]^2 - 1.02168[B]^2 - 0.031080[D]^2 + 3.35986[A][B] - 0.42632[A][D] + 0.025223[B][D]$$

$$f_{RP3} = -1701.65691 + 102.69526[A] + 10.80965[B] + 14.61338[D] - 19.67073[A]^2 - 1.02168[B]^2 - 0.031080[D]^2 + 3.35986[A][B] - 0.42632[A][D] + 0.025223[B][D]$$

The most important process variable for flexibility of impregnated paper was the final moisture content with a  $P>F$  much lower than 0.0001. With increasing moisture content the flexibility increased. This is explained by the fact that water molecules act as flexibilizers in the three-dimensional resin network. Flexibility also varied among the tested papers and different shapes of response surfaces were calculated for RP1, RP2 and RP3. The effect of raw paper is illustrated by the significant  $P>F$ -value of factor [C] in Table 2. The values for flexibility were always lower at lower moisture content. While the shapes of the response surfaces for RP1 and RP2 (Figures 5a/5d and 5b/5e) were very similar, the behaviour of RP 3 (Figures 5c/5f) differed much. The most significant influence of the catalyst amount was observed with RP 3 (5c / 5f), where the slope of the response surface became steeper when the catalyst amount was increased. In contrast, with RP 1 and 2, flexibility was practically not influenced at all by the amount of curing catalyst while for RP 3 a moderate effect was observed. Although obvious in the response surfaces, the effect of resin load was statistically much less significant than that for the catalyst amount. When comparing the  $P>F$  values for factor [D] (=resin load) it is seen that the only term of real statistical significance is for the interaction term between [C] (=the raw paper) and [D]. Neither [D] ( $P>F = 0.3726$ ) nor [D<sup>2</sup>] ( $P>F = 0.1387$ ) contribute much to the overall model. In principle, [D] and [D<sup>2</sup>] could even be omitted from the model calculations without losing much information or changing the response surface much. However, it is difficult to meaningfully discuss effects of interactions without considering at least some contribution of the corresponding main effects of the factor.

## Influence of raw paper supplier

For all responses it was found that the characteristic shape of the response surface was the same for all three raw papers studied. The mathematical form of the response surface was equivalent and the numerical factors final moisture content, catalyst concentration and resin loading influenced the technological properties of all papers in a similar way. However, the papers varied significantly in the exact quantitative values and even more important in the gradient of susceptibility of a response in dependence of the factor level. All measurement results were strongly influenced by the raw paper (see also the significance level for raw paper in Table 2) and the factor "raw paper supplier" played an important role.

In general, if papers of different densities are measured with the Golombek tester, no comparable testing results are obtained since their mechanical stiffness is co-measured with the apparatus. Papers of a density of 105 gm<sup>-2</sup> evidently exhibit much greater resistance against torsion in the hot oil bath than papers of a density of 80 gm<sup>-2</sup> and thus

they greatly vary in their rheological response (data not shown). Eventually, papers of much lower grammage such as overlay papers of 20–30 gm<sup>-2</sup> do not give any useful response at all. Since their stiffness is much too low, they simply fold and agglutinate when they are subjected to rotation in the hot oil bath.

However, in the present case papers of the same grammage (~105 g) were compared to each other and the actual quantitative values for the target responses did still not match exactly, although the overall trends as reflected by the form of their response surface profiles were similar. To some extent this may be explained by differences in paper stiffness of the raw papers as reflected by the variety in mechanical strength parameters given in Table 1. Still, this finding is rather unexpected and interesting since in industrial practice papers of the same type (décor paper, balance sheets or overlays) and the same grammage are often used interchangeably for the same purpose. Papers are often exchanged from one supplier to another depending on the current cost structure but the corresponding recipes and procedures for their impregnation are treated as compatible. Typically, the Golombek tester is used for determining the impregnation properties of a new lot of raw paper based on laboratory impregnations in comparison to an earlier reference paper from another lot; if the measured values match, the tested impregnated paper is assumed to result in the expected technological profile. The results of the current study however imply that the absolute reference values used for comparison with new production lots of raw paper purchased from different suppliers do not necessarily match perfectly since the absolute values of rheological and thermal responses differ significantly. The findings indicate that models based on data obtained with one paper may not be simply transferred when the raw paper supplier is changed but will have to be adapted.

It is difficult to relate the raw paper properties from Table 1 to the results of the response factor analysis. It seems that values for flow time, cure time and flexibility are all especially low with the raw paper that displays the highest values for wet strength whereas the reason for this might not be simply deduced. Based on the results collected so far it is also not yet possible to quantify the relation between selected raw paper quality parameters and the technological property profiles of papers in their impregnated state. Such studies will be the focus of future work.

## 4. Conclusion

In the present study, the mathematical modelling of practically rheological and thermal parameters for impregnated paper characterization has been accomplished. The calculations were based on the response surface analysis of an industrial experiment where the four production parameters raw material (raw paper supplier), impregnation resin mixture (catalyst concentration), roller coater (resin loading) and dryer (final moisture content) were systematically varied according to a central composite statistical design. It was found that the numerical factors catalyst amount, moisture content and paper supplier were statistically significant and strongly influenced the further processability of impregnated papers which was quantitatively expressed in terms of flow time, cure time, curing rate and flexibility of the impregnated paper sheet. A quantitative model suitable for predicting the paper processability in dependence of chosen production conditions was established. Moreover, the provenience of raw paper influenced strongly the rheological and thermal properties of the impregnated papers as measured with the Golombek apparatus. Paper wet strength seems to be correlated with the Golombek parameters as it is associated with the mechanical stiffness of a paper.

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Tabelle 1: Technologische Eigenschaften der in der Studie getesteten Rohpapiere

Tabelle 2: ANOVA für die Response Surface Analyse der Zeitgrößen "Fließzeit", "Härtungszeit" und "Flexibilität" von imprägniertem Papier

Abb. 1: Schematische Darstellung des Imprägnierprozesses von Papieren für dekorative Schichtstoffe

Abb. 2a: Schematische Darstellung des Messprinzips der dynamisch-mechanischen Eigenschaften von imprägniertem Papier. a) Thermostat; b) inertes Silikonöl; c) Papierprobe; d) Probenhalterung; e) Motor mit Antriebswelle; f) Datenverarbeitung  
mit dem Golombek-Apparat. IP: Wendepunkt; t<sub>flow</sub>: Fließzeit; t<sub>curemax</sub>: Zeitpunkt der maximalen Aushärtungsgeschwindigkeit; t<sub>cure</sub>: Zeit bis zum Erreichen von 95 % Vernetzung von Melaminharz; r<sub>cure</sub>: Maximale Aushärtungsgeschwindigkeit

Abb. 3(a-c): Einfluss der Härtermenge und der Harzbeladung auf die Fließzeit von imprägniertem Papier für die drei getesteten Rohpapiere

Abb. 4 (a-c): Einfluss des Feuchtigkeitsgehaltes des imprägnierten Papiers sowie der Härtermenge im Imprägnierharz auf die Härtingszeit der imprägnierten Papiere 1 - 3 bei hohen Harzbeladungen

Abb. 5 (a-f): Einfluss des Feuchtigkeitsgehaltes des imprägnierten Papiers und der Harzbeladung auf die Flexibilität der imprägnierten Papiere 1 - 3 bei niedrigen (a-c) und hohen (d-f) Konzentrationen von Härtingkatalysator in der Imprägnierharzmischung

## Tables

Table 1: Technological properties of raw papers tested in the present study

RP	D [gm <sup>-2</sup> ]	WS <sub>  </sub> [g/15 mm]	/WS <sub>⊥</sub> [g/15 mm]	WS <sub>  </sub> /WS <sub>⊥</sub>	DS <sub>  </sub> [g/15 mm]	DS <sub>⊥</sub> [g/15 mm]	DS <sub>  </sub> /DS <sub>⊥</sub>	Ash [%]	pH
1	106	1700	1250	1.36	4140	3040	1.36	26.5	6.9
2	105	910	605	1.50	3300	2610	1.26	22.4	6.9
3	105	825	480	1.72	3100	2050	1.51	26.8	6.6

RP ... raw paper, D ... density, WS<sub>||</sub> ... wet strength parallel to the direction of paper production, WS<sub>⊥</sub> ... wet strength perpendicular to the direction of paper production, DS<sub>||</sub> ... dry strength parallel to the direction of paper production, DS<sub>⊥</sub> ... dry strength perpendicular to the direction of paper production

Table 2: ANOVA results (partial sum of squares) for the response surface analysis of the responses flow time, cure time and flexibility of impregnated paper for the three different raw papers 1 to 3

	<i>t<sub>flow</sub></i>		<i>r<sub>cure</sub></i>		<i>t<sub>cure</sub></i>		<i>f</i>	
	F-value	P > F	F-value	P > F	F-value	P > F	F-value	P > F
Model	22.09	< 0.0001	53.06	< 0.0001	18.20	< 0.0001	34.84	< 0.0001
[A]	86.27	< 0.0001	569.78	< 0.0001	168.96	< 0.0001	7.15	0.0142
[B]	0.068	0.7958	7.87	0.0106	11.17	0.0031	105.52	< 0.0001
[C]	12.32	0.0001	15.54	< 0.0001	10.84	0.0006	6.79	0.0053
[D]	2.10	0.1568	4.81	0.0397	2.49	0.1293	0.83	0.3726
[A] <sup>2</sup>			11.06	0.0032	6.38	0.0196	1.55	0.2262
[B] <sup>2</sup>			15.32	0.0008	8.67	0.0078	36.03	< 0.0001
[D] <sup>2</sup>			0.69	0.4165	1.06	0.3158	2.37	0.1387
[AB]			0.021	0.8850	0.14	0.7149	1.20	0.2859
[AC]			0.72	0.4993	1.11	0.3473	0.98	0.3904

[AD]	0.024	0.8790	0.16	0.6887	0.13	0.7236
[BC]	0.44	0.6527	0.55	0.5826	1.78	0.1939
[BD]	1.39	0.2509	2.86	0.1053	0.088	0.7695
[CD]	1.08	0.3586	0.93	0.4092	3.80	0.0391

$t_{flow}$  ... flow time;  $r_{cure}$  ... curing rate;  $t_{cure}$  ... cure time;  $f$  ... flexibility; [A] ... amount of curing catalyst in the impregnation resin mixture; [B] ... final moisture content of impregnated paper; [C] ... raw paper supplier; [D] ... resin load of impregnated paper

<sup>a</sup>Values of “P>F” less than 0.05 (5 %) indicate that model terms are significant

Figure 1

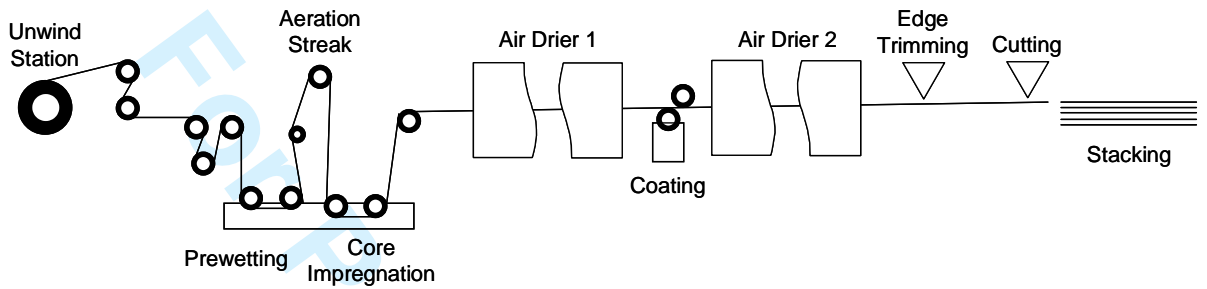


Figure 2 a

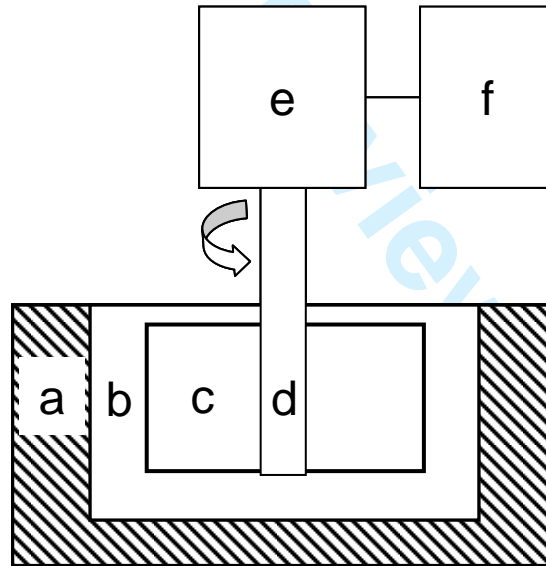


Figure 2 b

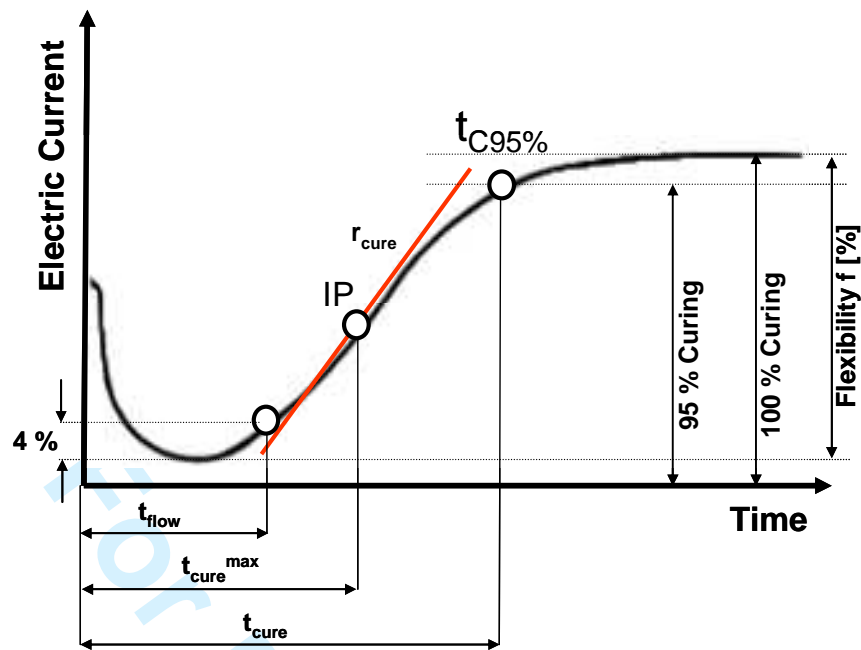


Fig. 3 abc

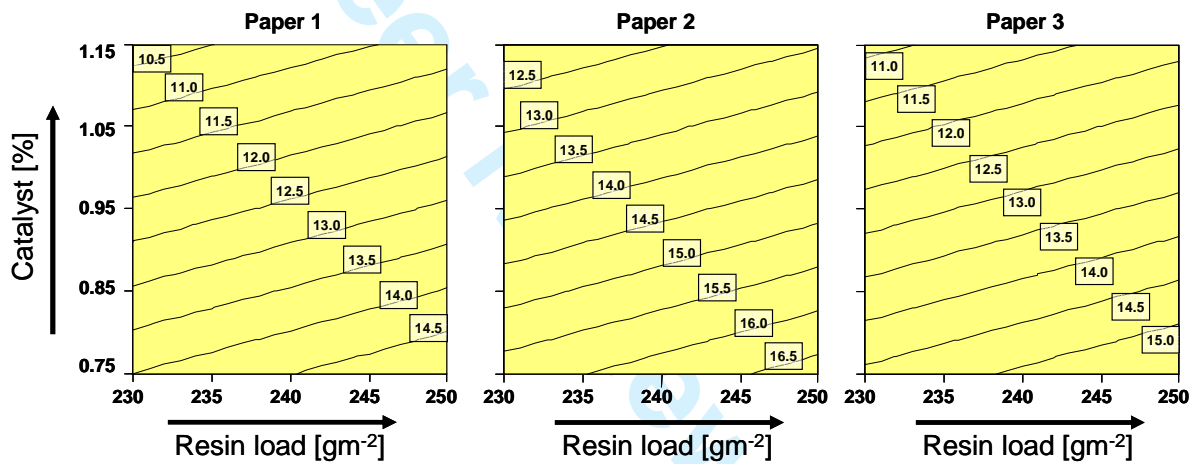


Fig. 4 abc

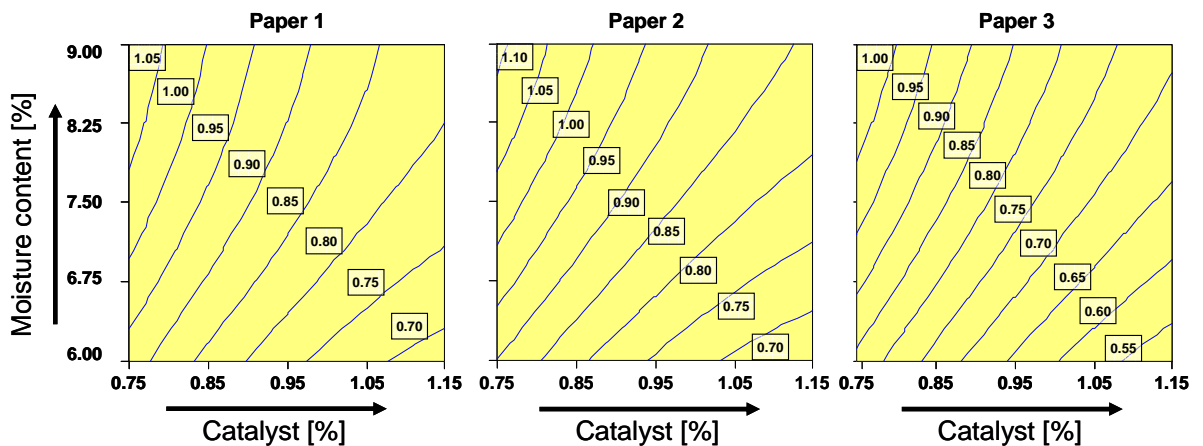


Fig. 5abc

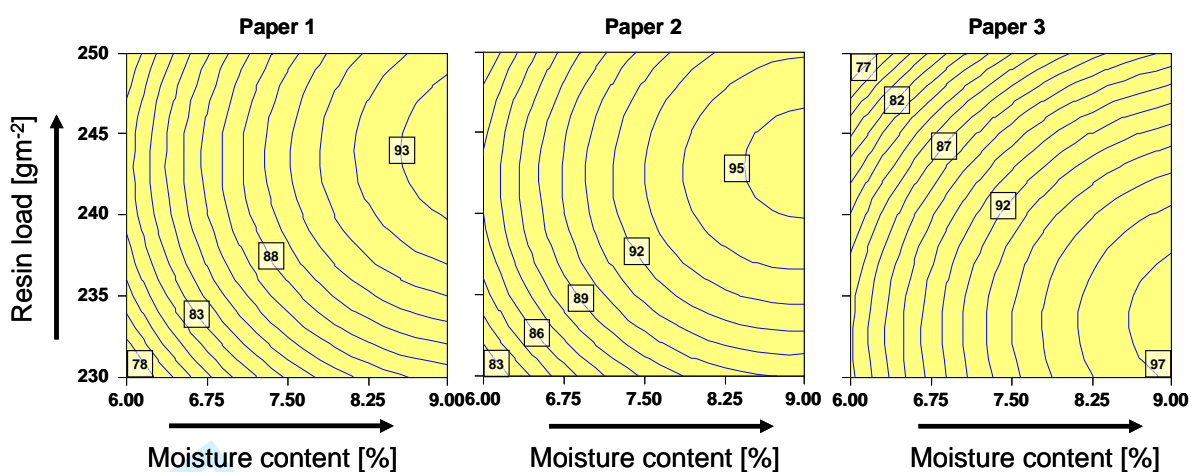


Fig. 5 def

