A CONTINUOUS PROCESS FOR UD FLAX FIBRE REINFORCED COMPOSITES

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ABSTRACT

Natural fibres like flax have specific mechanical properties competitive to glass fibres. However, the variability in fibre properties is large, despite thorough fibre selection, mixing of harvests, … When incorporated into composites, more variability is introduced through fibre handling, resin impregnation and process consolidation parameters. Hence, it becomes difficult to assess the effects that e.g. chemical treatments have on the properties of a natural fibre and its composites. To limit this additional variability, a continuous lab-scale production line for unidirectional natural fibre reinforced composites was set up.
The major advantage of the continuous way of production is a better control of the process in comparison with the manual placement of fibre reinforcement. This will result in improved mechanical properties. Also, it offers a better simulation of industrial production processes, which are mostly continuous. The production set-up has been extended with an additional fibre pre-treatment bath and a subsequent rinsing and drying system. A stepwise analysis of the effect of alkali pre-treatment on the composite properties has also been performed.

1. INTRODUCTION

Research on natural fibre reinforced composites experiences a growing interest in both academic and industrial world. These light weight and environment friendly fibres fit well in the ecology-conscious world of today. Several other ecological and/or economic advantages can be enumerated: their CO_2 neutral life cycle, the renewability, their low cost and the easy incineration at the end of their life cycle. Furthermore, some plant fibres exhibit fairly good mechanical properties, especially on weight basis, making them competitive to glass fibres as a reinforcement in composite applications [1-5]. However, mechanical, physical and even chemical properties of these plant fibres are strongly harvest-dependent, influenced by climate, location, soil characteristics, weather circumstances, etc. In addition, the properties are affected by fibre processing (retting, scutching, bleaching, spinning, etc.) and by their incorporation into composites: handling, impregnation and consolidation will introduce supplementary changes. Obviously, this variability complicates the prediction and evaluation of the composite properties and hence, should be kept under control.

A better parameter control is therefore essential, and switching the production method into a continuous line is an important step. Natural factors, such as soil characteristics or climate conditions, cannot be changed, but misalignments introduced during manual fibre placement are also far from negligible. Certainly the handling of natural fibres requires extra care because of their somewhat twisted structure which leads easily to inaccuracies (fibre damage, misalignments, …). However, by the use of a drumwinder equipment parallel fibre orientation can be smoothly achieved, and a constant composite quality is guaranteed as much as possible.

Another known problem in natural fibre reinforced composites is the poor interface quality between the fibres and the polymer matrix: to enhance the adhesion between both components, chemical pre-treatments are often applied [1, 6-11]. Simple and cheap methods, e.g. alkalisation, are proven to be effective. Also during treatment, it is important to keep possible variable factors under control. Therefore, a treatment section was integrated in the continuous prepreg manufacture route, so that fibres can be successively treated, rinsed, dried, impregnated with resin and aligned into UD prepreg material, all in one run.

2. MATERIALS AND METHODS

The drumwinder machine, shown in Fig. 1, was used to manufacture unidirectional (UD) flax fibre reinforced epoxy composites in a continuous way. The complete set-up is aimed at lab-scale production. Fibres are rolled off the bobbin and led through the resin bath, after passing through an oven. Via flattening pins and a guide roller, the impregnated fibres are finally placed parallel on the drum. The resulting material is a semi-finished unidirectional reinforced product (prepreg) which needs further curing in the autoclave, according to resin requirements. In these experiments an autoclave pressure of 3 bar is used during 1 hour at 125 °C. Also vacuum (-0.8 bar) is applied to remove air entrapments and excessive resin out of the laminates.

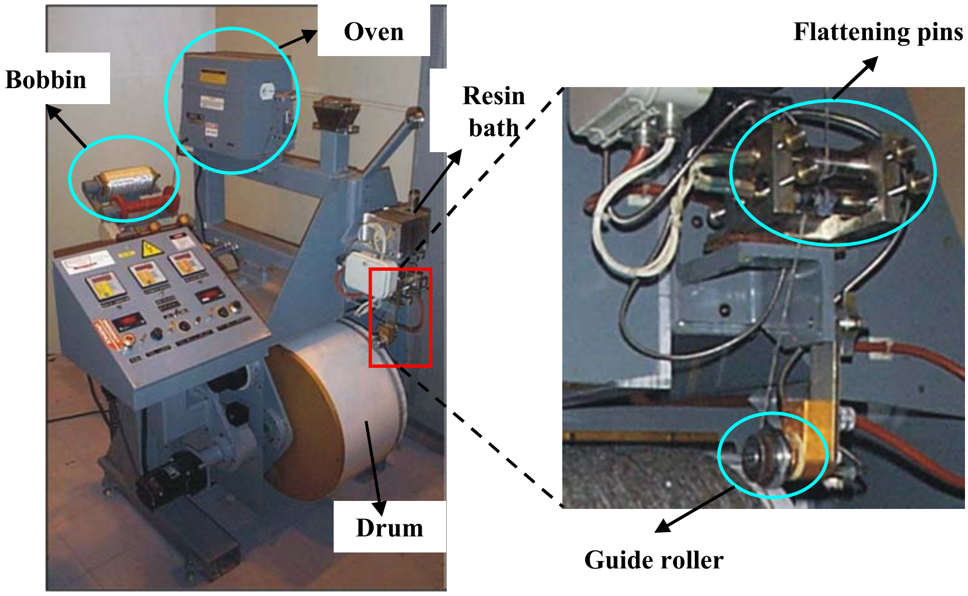

Fig. 1. Drumwinder**.**

Some technical details on the drumwinder machine, which was manufactured by the Research Tool Corporation (Ovid, Michigan, U.S.A.), are provided in Table 1.

Table 1. Technical details on the drumwinder machine.

Circumference drum (cm)	193
Width drum (cm)	31.12
Max. rotational drumspeed (RPM)	9
Max. carriage speed (cm/min)	2.54
Max. temperature of resin bath, guide roller, flattening pins (°C)	400
Max. temperature of oven (°C)	1100

The fibrous material used for the experiments was kindly offered by Tex-Dem N.V., Alveringem, Belgium. Fine flax roving, i.e. slightly twisted fibre slivers, were washed prior to winding on a bobbin. The linear density of the roving was determined to be approximately 600 tex.

The applied resin component was a two-part epoxy system, resin LMB 6305/ hardener HY 5021 BD, suitable for hot-melt prepreg production and manufactured by Vantico N.V. (Groot-Bijgaarden, Belgium). The hardener was added in a ratio of 100/24 on weight base.

For the alkaline fibre pre-treatments, high purity NaOH pellets (Acros Organics, Geel, Belgium) are dissolved in distilled water to get the appropriate concentrations. Two routes were followed to apply the treatment: in a first set of experiments, a semi-continous process was used. This comprises a batchwise treatment of the fibres on the bobbin, starting with a two minute immersion in an alkaline bath and followed by thorough rinsing of the fibres (including a washing with slightly acidic water of 10 drops HCl 0.1 M in 500 ml distilled water). Finally the bobbin with the fibres is dried in an oven at 80 °C during 8 hours before it is mounted on the drumwinder equipment to be impregnated with the resin and processed into UD prepregs.

The second route is a fully continuous processing method, in which the fibres are unrolled from the bobbin to be treated, rinsed, dried, impregnated and placed unidirectionally, all in one run. A sketch of the complete treatment section is drawn in Fig. 2. Within this set of experiments, the fibres are only rinsed with running distilled water and no acidic neutralisation is applied. Subsequently, the fibres pass through three drying stages before they are resin impregnated. Further details on this fully continuous processing route are described in paragraph 3 "Production and Testing".

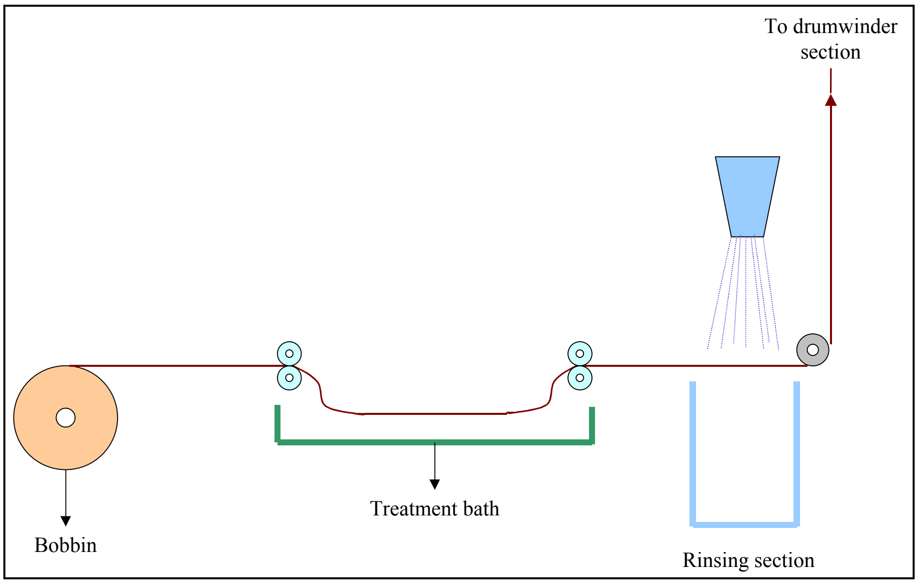

Fig. 2. Sketch of the treatment section.

3. PRODUCTION AND TESTING

Optimisation of the process parameters

Table 2 gives an overview of the applied settings in the performed fully continuous experiments. A part of the parameter optimisation is already presented in former work [12, 13]. For the parameters of the semi-continuous production process, the same settings are operative, but the "drying temperatures" and "alkali concentrations" do not apply in that case. Each of the listed parameters has its influence on the final quality of the prepreg: controlling the speeds and temperatures is essential when delicate flax fibres are to be used.

Table 2. Settings of the parameters.

Rotational drumspeed (cm/sec)	10.65
Carriage speed (cm/sec)	0.008
Temperature resin bath (°C)	85
Temperature flattening pins (°C)	80
Temperature guide roller (°C)	85
Drying temperatures (°C)	
step 1: hot air blower	210
step 2: oven	250
step 3: hot air blower	120
Alkali concentrations (%)	
control experiment	0
experiment 1	4
experiment 2	8
experiment 3	12

The process temperatures (i.e. temperatures of resin bath, flattening pins and guide roller) affect the viscosity of the resin: too low temperatures make the resin too sticky and will cause fibre damage during impregnation, possibly interrupting the continuity of the process. Moreover, the resin will not spread well between the fibres during flattening and will stick to the guide roller, creating rough and slovenly prepreg surfaces. On the other hand, the temperatures should be limited to prevent too fast curing of the resin and insufficient adhesion to the fibres. The temperature of the drum itself is non-adjustable and it is therefore kept at room temperature.

The rotational speed of the drum is taken at maximum to enable a fast production process, but should be limited to prevent splitting of the flax fibres. The carriage speed (i.e. the lateral movement of the drum) is set with relation to the rotational speed, to obtain the desired distance between the wound rovings.

As explained above, the influence of an alkaline fibre pre-treatment was studied in two parts: the first experiments were carried out in the "semi-continuous" way, using alkali concentrations of 15% and 30% [12, 13]. The immersion time in the alkaline bath was kept short at two minutes, to simulate the short immersion time during continuous processes. In a second set of experiments the set-up was further elaborated, and used as described in Fig. 2 (fully continuous treatment and impregnation of the fibres). Through the gradual increase of the NaOH concentrations, an optimum in the composite properties was tried to be found: solutions of 0% (control sample), 4%, 8% and 12% are applied to the flax reinforcement. In both methods, the resulting prepregs are further processed into flat composite plates by layering several prepregs to reach a sufficient composite thickness (approximately 3 mm). In total six different plates, based on different NaOH concentrations are produced.

Mechanical properties

The mechanical properties of the UD composites are determined through three point bending (3PB) and tensile tests. The dimensions of the test specimens can be found in Table 3.

Table 3. Specimen dimensions for 3PB and tensile tests, both longitudinal and transverse. (* except for the untreated, the 15% and 30% treated samples: width 10 mm, span 60 mm in longitudinal and 30 mm in transverse direction).

Test		Longitudinal & Transverse
3 PB	Thickness (mm)	3
	Width (mm)	25*
	Span (mm)	48*
Tensile	Thickness (mm)	3
	Width (mm)	20
	Gauge length (mm)	130

Unidirectional untreated flax epoxy composites

To estimate the influence of the production method on the composite properties, flat composite plates were made by manual placement (batch process) and by means of the drumwinder (continuous process). The properties of both were compared and sorted in Table 4.

Table 4. Mechanical properties of UD flax epoxy composites, measured by 3PB and tensile tests.

Production Method		Three Point Bending		Tensile	
		σ^*_{3PB} (MPa)	E (GPa)	σ^*_{tens} (MPa)	E (GPa)
Drumwinder ($V_f = 48$ %)	Long.	282 ± 11	23 ± 1	268 ± 26	32 ± 1
	Trans.	27 ± 3	1.3 ± 0.2	18 ± 1	4.0 ± 0.3
Manual ($V_f = 40$ %)	Long.	218 ± 18	18 ± 3	190 ± 10	26 ± 1
	Trans.	8 ± 4	0.4 ± 0.2	10 ± 1	4.0 ± 0.5

Both longitudinal and transverse composite properties are positively influenced by the change of production process from the manual placement method to the continuous drumwinder production method, even when the difference in fibre volume fraction is taken into account. The effective fibre strength σ^*_f can be calculated from the results in Table 4, using Eq. 1.

$$\sigma^*_{composite} = V_f \, \sigma^*_f + V_m \, \sigma'_m \tag{1}$$

With $\sigma^*_{composite}$ the composite **tensile** strength

V_f and V_m the fibre and matrix volume fraction

σ'_m the matrix stress at fibre failure (by approximation taken at 50 MPa)

So, the drumwinder produced plates result obviously in higher effective fibre strengths (σ_f^* = 505 MPa) compared to the manual produced plates (σ_f^* = 400 MPa). However, the increase will be partially due to the difference in fibrous material: the prepreg material was reinforced with flax roving, while flax slivers were used for the manual production method.

Influence of an alkaline pre-treatment on the composite properties

The alkali treatment used, involved an immersion in sodium hydroxide solution (NaOH) since earlier experiments [7] proved that a low concentration of NaOH (1, 2 or 3 %) acting upon the flax in a batch-wise manner for 20 minutes enhanced the final composite properties. Within this study the total immersion time of the fibres is lowered to enable a continuous process. In the first set of semi-continuous experiments, the concentration was simultaneously raised to 15% and also 30%, to compensate for the low treatment time (which was taken at two minutes). Results on the mechanical properties are summarised in Table 5 and can be found into detail in [12].

Table 5. Mechanical properties of treated UD flax epoxy composites, measured by 3PB tests [12].

Fibre Treatment	Longitudinal		Transverse	
	σ* (MPa)	E (GPa)	σ* (MPa)	E (GPa)
15 % NaOH (V_f= 49 %)	284 ± 8	26 ± 1	21 ± 4	1.2 ± 0.2
30 % NaOH (V_f= 47 %)	240 ± 11	22 ± 1	19 ± 5	1.1 ± 0.2
Untreated (V_f= 48 %)	282 ± 11	23 ± 1	27 ± 3	1.3 ± 0.2

The alkali treatment will remove (partially or completely) several non-cellulose constituents of the fibre, such as hemicellulose and pectin, leading to a rougher and purified fibre surface (dewaxing effect) [10, 14]. This will enhance the fibre-matrix adhesion, giving better composite properties. However, as can be seen in Table 5, the properties decrease when high NaOH concentrations are used, due to a weakening of the fibre. In this case, the damaging effect of alkali surpasses the positive effect of removing impurities and waxy substances, even for this short immersion time. Probably, an optimal alkali concentration will be reached before 15%. Therefore, additional experiments were carried out to provide more data in the range up to 15%: fibres were treated with 4%, 8% and 12% NaOH solution for ¾ minute using the adapted and fully continuous set-up (Fig. 2). A control sample existed of fibres run through a demineralised water bath (= 0% NaOH). The preliminary results on the flexural properties are reported in Table 6*.

* The full analysis of the influence of alkali treatments on the mechanical properties of flax epoxy composite properties will be shown during the presentation at ECCM-11.

Table 6. Mechanical properties of treated UD flax epoxy composites, measured by 3PB tests.

Fibre Treatment	Longitudinal		Transverse	
	σ* (MPa)	E (GPa)	σ* (MPa)	E (GPa)
Untreated (V_f = 48 %)	282 ± 11	23 ± 1	27 ± 3	1.3 ± 0.2
Control sample (V_f = 42 %)	256 ± 5	22 ± 2	36 ± 1	2.9 ± 0.6
4 % NaOH (V_f = 53%)	283 ± 10	25 ± 1	35 ± 2	3.6 ± 0.3
8 % NaOH	*To be presented at ECCM-11 conference*			
12 % NaOH	*To be presented at ECCM-11 conference*			

The composite samples treated with 8% NaOH and 12% NaOH are still in production phase and are not yet tested. Therefore, these results will be presented at the conference.
As far as the finished results are considered (i.e. untreated, 0% and 4% NaOH solution), it is obvious that the longitudinal composite properties do not seem to vary significantly when raising the NaOH concentration to 4% (taking into account the fibre volume fraction). However, the transverse composite properties show an increase in both strength and stiffness when the fibres are treated with pure water (0% NaOH). The difference between the control sample and the 4% treated sample is yet again none.

The mild treatments carried out on the flax fibres do not affect the fibre properties in such an extent that the longitudinal composite properties are altered. This explains the lack of variation between the longitudinal flexural properties of the processed samples. On the other hand, the transverse properties prove clearly the positive effect of even mild treatments. Washing the fibres with demineralised water (0% NaOH) removes a part of the impurities on the fibre surface, thus promoting the adhesion between the flax fibres and the matrix, and hence resulting in a better transverse performance. Raising the alkali concentration is expected to have a positive influence on the composite properties, but this experiment demonstrates that a 4% NaOH solution during ¾ minute does not result in any surplus value compared to the control sample. The on-going experiments comprise tests with stronger alkali concentrations and will therefore be necessary to complete this discussion and to draw clear conclusions out of the gathered results. A complete discussion on the effect of alkali treatments on the properties of flax fibre reinforced composites will be presented at the conference.

4. CONCLUSION

A continuous processing method has been presented, suitable for the production of unidirectional reinforced natural fibre composites. In one run, roving material can be treated, impregnated and aligned in a UD prepreg. This method seems promising to make highly reproducible natural fibre composite plates with a good alignment.
Secondly, this set-up allowed for a stepwise analysis of the effect of an alkaline fibre treatment. Changing the concentration of the NaOH solution between 0% and 12% in 4% steps, and processing these treated fibres into composites, gives insight in the evolution of the composite properties as a function of alkali concentration.

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