NATURAL COMPOSITES BASED ON CELLULOSIC FIBRES AND POLYPROPYLENE MATRIX – THEIR PROCESSING AND CHARACTERISATION

H. Lilholt [1], H. Toftegaard [1], A.B. Thomsen [2], and A.S. Schmidt [2].

[1] *Materials Research Department*
[2] *Plantbiology and Biogeochemistry Department*
Risø National Laboratory, DK-4000 Roskilde, Denmark

SUMMARY: Natural fibre composites are fabricated based on jute, flax and wheat straw fibres, respectively, and a polypropylene matrix. The fibres are characterised chemically and have a high cellulose content. The treatment by wet oxidation of wheat straw may give fibres with improved surface characteristics for composite fabrication. The composites are characterised structurally and have fibre *weight* fractions up to 60 percent, while the fibre *volume* fractions show a maximum at about 30 percent; the porosity content is 2 to 30 percent. The mechanical properties of the composites show efficient reinforcement by the fibres; the composites have better strength than pure polypropylene at realistic working strains. The estimated fibre stiffness and strength values are acceptable in comparison to other estimates.

KEYWORDS: Natural composites, cellulosic fibres, jute/polypropylene, flax/polypropylene, straw/polypropylene, physical characterisation, mechanical properties.

INTRODUCTION

Natural fibres based on cellulose are finding increasing use in materials and industrial products. In composites the fibres are combined with a matrix, often a polymer, into a composite with full density, i.e. no porosity. In this form the natural composites are widening the spectrum of available load-bearing materials and sometimes competing with synthetic composites based on glass, carbon and other inorganic fibres in a polymeric matrix.

In order to evaluate the natural fibres and their composites it is necessary to characterise and test them in comparison to existing materials, especially (synthetic) composites. For the natural fibres to give efficient composites it is necessary that the fibres have good stiffness and strength and that the matrix is well bonded to the fibres so that loads can be transferred to the fibres. By this mechanism the good stiffness and strength of fibres are exploited to give load bearing composites of good performance. The potentially useful fibres in this context are flax, hemp, ramie, and jute, they have stiffness values comparable to those of glass fibres, and strength values somewhat lower than inorganic fibres.

The basis for the structure and thus stiffness and strength is the cellulose molecules, which form a sometimes crystalline structure, where the cellulose chains are stabilised by hydrogen-

bonds between hydroxyl groups and oxygen atoms. The potential stiffness is about 150 GPa, which should be compared to the practical values at about half of this value.

Polypropylene is a thermoplastic polymer with moderate stiffness and strength and large ductility. It is a representative of a large group of potentially useful polymers for matrices; some of these may be based on natural sources, e.g. starch and polylactate, and thus be renewable and possibly biodegradable.

The densities of cellulose fibres and of polymers are close to 1 g/cm^3; therefore the density of natural composites are low and compare favourably with other (synthetic) composites.

MATERIALS AND PROCESSING

To evaluate the potential for natural composites as load bearing materials a series of cellulosic fibres are tested. The jute fibres are used as mats of nearly random fibre orientation. The flax fibres are used as mats of random fibre orientation. These two types of fibres are obtained from natural sources and treated industrially before mat-forming. The wheat straw fibres are treated by a laboratory scale wet oxidation process which increases the cellulosic content of the fibres; after this the wheat straw fibres are made into mats (lab.scale process). All three types of mats have area densities of 300 – 600 g/m^2.

The polypropylene matrix (PP) is used in the form of thin foils. The composites are manufactured by alternating stacking of fibre mats and PP-foils. The individual mats are all placed with the same directional orientation, to allow this parameter to be evaluated from the composite characterisation. The PP-foils are used in different numbers between the fibre mats to allow a variation in volume fractions of fibres and PP-matrix, respectively. The total stack is sealed with a vacuum bag and hot pressed in an autoclave.

STRUCTURAL CHARACTERISATION

Composition of fibres

The densities of the three types of fibres are listed in Table 1 and compared to glass (fibres). The densities of natural fibres in the table are the cell wall densities; some fibres, e.g. jute fibres, have a lumen, which reduces the average fibre density, e.g. for jute the lumen is about 12% and the density is thus 1.29 g/cm^3 [1].

The chemical composition of the natural fibres shows the relatively high cellulose content of jute and flax fibres. The cellulose molecule is the basis for the potentially high values of mechanical stiffness and strength of these fibres. Jute fibres have potential stiffness values of 50-70 GPa, and flax fibres have stiffness values of up to 100 GPa. The wheat straw have relatively low cellulose content as raw fibres; the wet-oxidation treatment reduces the lignin and hemi cellulose content and thus leads to a high cellulose content in the treated fibres. This is the basis for potentially good stiffness and strength properties of wheat straw fibres; this will be evaluated at the end of the paper.

Table 1: Density and chemical composition of natural fibres and of glass fibres

Fibre	Density g/cm^3	Chemical composition (w%) Cellulose	Hemi-cellulose	Lignin	Rest
Jute	1.52	60	12	18	10
Flax	1.54	67	11	2	20
Wheat Straw					
raw	1.51	39	30	9	22
wet oxidised	1.51	~74	~11	~7	~8
Glass	2.62	-	-	-	-

Composition of composites

The natural fibre composites are fabricated in the form of laminates of about 2 mm thickness. The total number of laminates fabricated are for each type of natural fibres: jute fibre composites: 23 plates; flax fibre composites: 30 plates, and wheat straw fibre composites: 10 plates.

From these plates samples are cut for characterisation of composition and mechanical properties. The composition of each laminate plate is described by the composite density, the weight fraction of fibres, the volume fraction of fibres, the volume fraction of matrix and the volume fraction of porosity. For each plate three samples are measured.

The density of composites is determined by the immersion method (Archimedes principle) with a balance with a beaker of water. The content of fibres, matrix and porosity in the composites is determined by gravimetric measurements; the composite sample is weighed, the PP-matrix is dissolved in xylene, and the jute-fibres are weighed; with the density of jute and of PP, the weight fractions and volume fractions are calculated, [1].

The data for the composition of the natural fibre composites are presented in Table 2; the values are given as a range rather than as (a large number of) individual values for each laminate. The *weight* fractions of fibres is generally moderate to high and can approach 60 weight per cent of fibres in the composites. The corresponding *volume* fractions of fibres depend on the (inherent or induced) porosity content of the composites. The *volume* fraction of fibres appear to have a maximum value of about 35 volume percent; this value is controlled by the compaction characteristies of the fibre mats [2]. With insufficient amounts of matrix, and thus correspondingly high (weight) fractions of fibres, this leads to rather high porosity contents (up to 35 percent). Such high porosity contents can be interpreted as a sum of structurally governed porosity and process controlled porosity [2].

Table 2: Density and composition of natural fibre composites

Composite	Density g/cm^3	Fibre weight fraction %	Fibre volume fraction %	Matrix volume fraction %	Porosity volume fraction %
Jute/PP	0.82-1.03	34-64	23-34	26-74	2-29
Flax/PP	0.81-1.05	22-65	14-37	22-84	2-33
Wheat straw/PP	0.68-0.96	51-59	27-33	32-52	15-38

MECHANICAL CHARACTERISATION

The mechanical properties are determined from samples of (gauge) length 60 mm or 100 mm and width 20 mm. The mechanical loading is simple tensile loading at a displacement rate of 1 mm/min, equivalent to an initial strain rate of 2.8 x 10^4 s^{-1}, the tests are performed at room temperature (25°C)

Stress-strain curves

The composities have stress-strain curves in tensile loading as illustrated for jute/PP composites in Fig. 1, for flax/PP composites in Fig. 2, and for wheat straw/PP composites in Fig. 3. The initial relatively steep part of the curve represents elastic behaviour and the slope of the curve defines the elastic modulus. The stress-strain curves for jute/PP and flax/PP show a moderately significant change of slope at about 0.2-0.4% strain. The "point" of change of slope is estimated by fitting two straight lines to the data of the curves between 0 and 0.5% strain; the intersection of these two lines defines a "knee point" for the stress-strain curve. This point is a formal transition from elastic behavior to visco-elastic deformation, which governs the remaining part of the stress-strain curve.

The strain value of the "knee point" is remarkably constant for all jute/PP composites and for all flax/PP composites; the value is close to 0.25% strain.

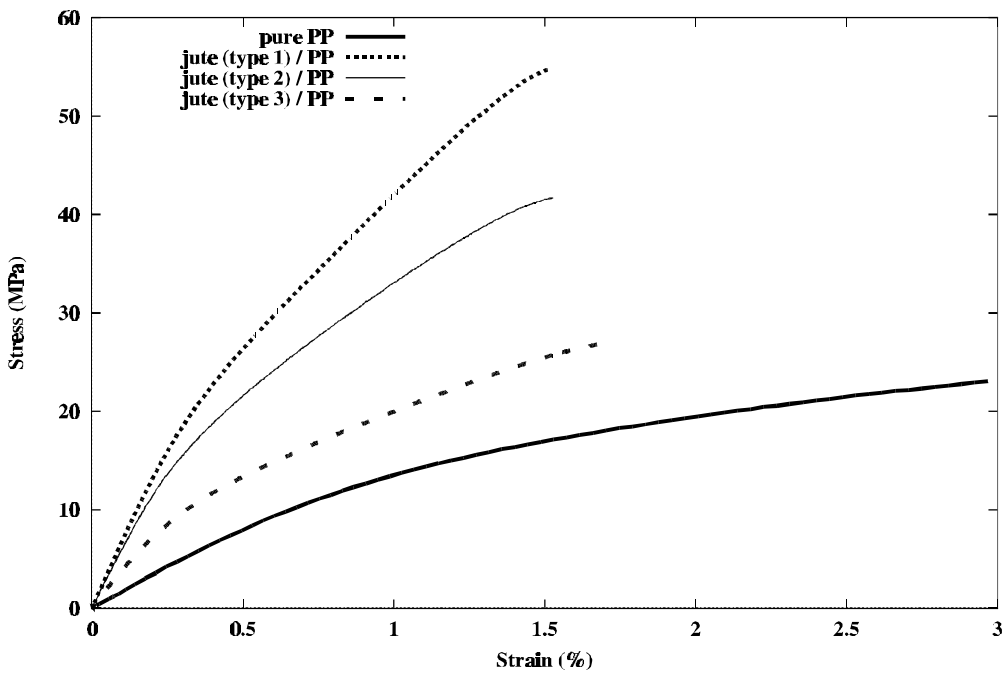

Fig.1: Stress-strain curves of jutefibre/PP composites; three different types of jute fibres and mats are used, while the amount of PP matrix is the same in all composites; the pure polypropylene stress-strain curve is shown for comparison.

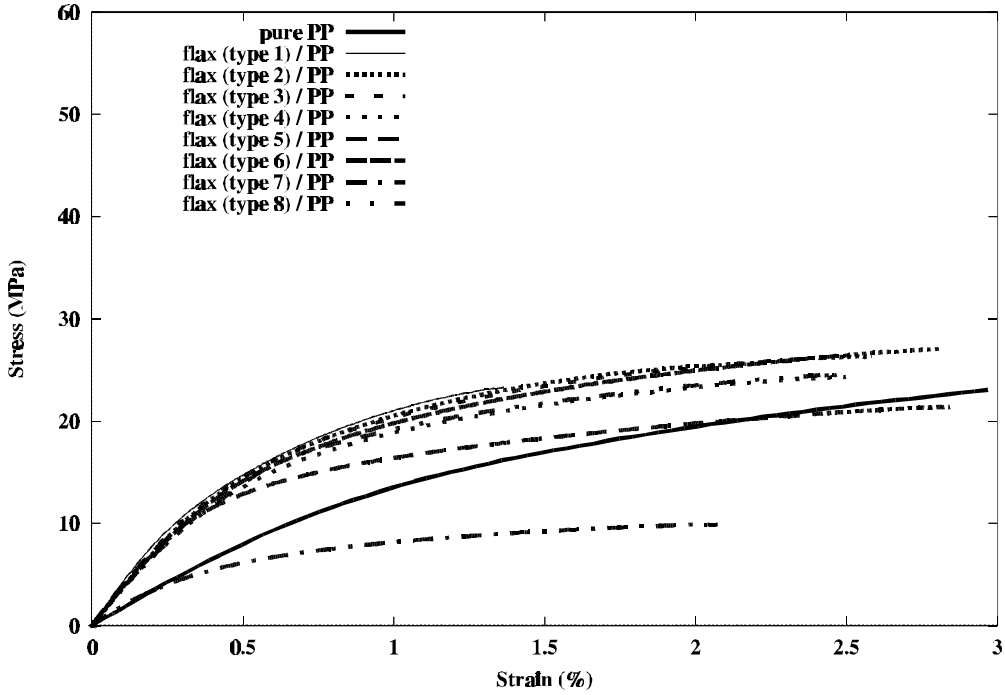

Fig.2: Stress-strain curves of flax fibre/PP composites; eight different types of flax fibre mats are used, while the amount of PP matrix is the same in all composites; the pure polypropylene stress-strain curve is shown for comparison.

Such a strain value may be used as a design parameter for load bearing components, in the sense that below this value the deformation will be essentially elastic, and above this value permanent deformation and damage of the composite will take place.

The stress-strain curves for wheat straw/PP composites in Fig. 3 are compared to the pure polypropylene matrix. The data for the composites show a clear reinforcing effect of the fibres through the increased stiffness of the composites compared to pure polypropylene. The composites fail at lower strain values than the pure polypropylene, but even at these strains the stress levels of the composites are still higher than the corresponding stress levels for the pure polypropylene. This implies that although the nominal ultimate tensile strength of the composites is lower than that of the pure polypropylene, the load bearing stress of the composites at realistic working strain values (e.g. 0.5% strain) is higher than that of the pure polypropylene.

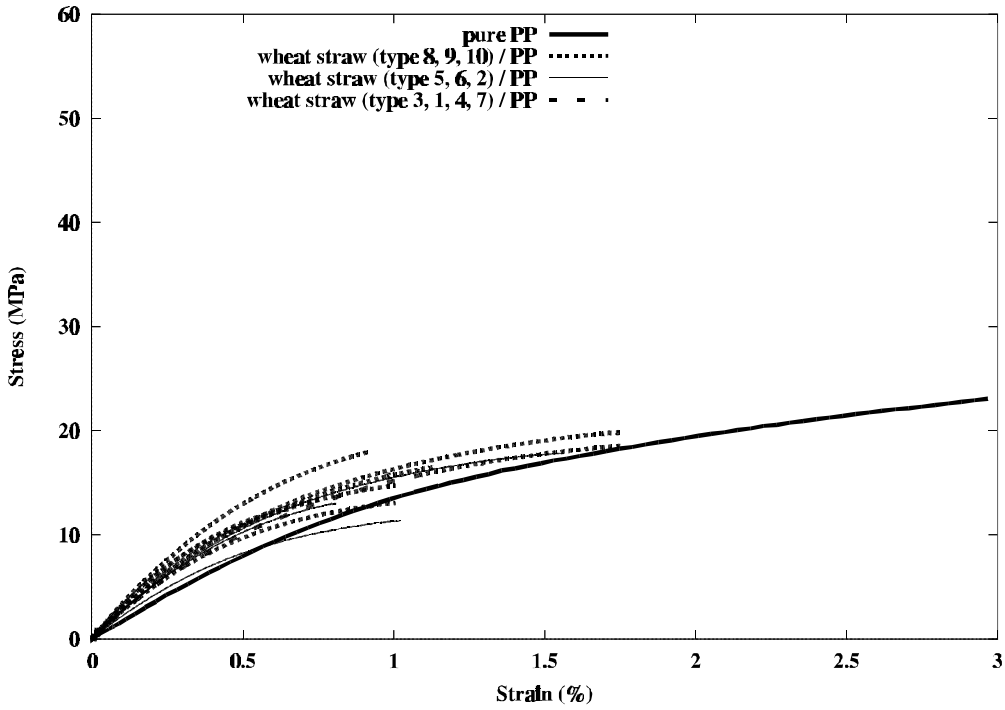

Fig. 3: Stress-strain curves of wheat straw /PP composites; ten different wet oxidation treatment are used for the wheat straw, the resulting composites are arranged in three groups; the amount of PP matrix is the same in all composites; the pure polypropylene stress-strain curve is shown for comparison.

Stiffness and strength

The results for the stiffness and the (ultimate) strength of the composites are presented in Table 3; the values are given as a range rather than as (a large number of) individual values for each laminate.

Table 3: Mechanical properties of natural fibre composites.

Composite	Stiffness GPa	Strength MPa	Failure strain %
Jute/PP			
strong direction	5-8	28-55	1.0-1.5
weak direction	3-5	19-32	1-2
Flax/PP	2-5	12-28	1.5-3.5
Wheat straw/PP	2-3	11-18	1-2
Polypropylene	1.7	29	5

The data show for all three composites a clear reinforcing effect of the natural fibres, based on the higher stiffness values for composites, in the range from 2-3 GPa to 8 GPa against the value of 1.7 GPa for pure polypropylene. The strength values at *failure* for the composites are comparable to that of the pure polypropylene, with a range from below to above the value for pure PP. This is because the (ultimate) strength is measured at failure, where the pure PP can be deformed to higher strain values and thus correspondingly higher stress (strength) values. At realistic working strains for materials and components (e.g. 0.5% strain) the composites can carry larger stresses than the pure polypropylene.

The large range for stiffness and strength values for the three composites, respectively, is caused by the relatively wide range of fibre volume fractions (Table 2), as well as the wide range for the porosity contents (Table 2). Generally the stiffness follows a linear ralationship with fibre volume fraction, but this is masked by the varying porosity content of the indiviual laminates.

For the jute/PP composites a more detailed and precise analysis has been made for a smaller number of laminates [1]. When the individual data for each laminate are corrected to zero porosity content, the linear relationships is confirmed for stiffness as well as for strength of the composites.

The effect of porosity on stiffness and strength can be evaluated from experimental results and simple composite relationships [1]; this shows the expected non-linear decrease of stiffness and strength with increasing porosity content. The mathematical relation is controlled by a term $(1-V_p)^n$, where V_p is porosity fraction and the exponent n has values of about 2 to 3 [3].

The jute mats used for the composites do not have a random fibre orientation distribution, but as the results in Table 3 show the “strong” direction has stiffness and strength values which are about double those of the “weak” direction. The analysis of this anisotropy of mats and composites is reported elsewhere [1].

The mechanical properties of the natural fibre composites investigated in this study compare favourably with other natural fibre composites and with glass fibre composites; a detailed comparative analysis is performed elsewhere [1].

In almost all composites the interface between (natural) fibres and matrix is of importance for the mechanical properties as well as for other characteristics related to the performance of the materials and components. Generally a mechanically strong interface is aimed at, to ensure efficient transfer of loads to the fibres. In the present study the jute fibres and the flax fibres are used without modification of their surface, such that only the “natural” adhesion between the fibres and the polypropylene is achieved in the composites. On the basis of efficient chemical surface modifications it can be expected that composites fabricated from such fibres will show better mechanical properties than those of the present composites. The wheat straw has been wet oxidised, which besides the increased cellulose content, may change/improve the surface characteristies, by removal of lignin and/or by retainment of some hemicellulose; these implications of wet oxidation treatments need further experiments for a detailed evaluation.

Fibre properties

From the experimental results for stiffness and strength of the composites, it is possible via composite theory to calculate stiffness and strength values for the fibres. The simplest calculation implies composites with zero porosity and has been attempted elsewhere [1]. A

more generalised model and calculation has been worked out [3], which includes the actual porosity content of the individual composite samples; the mathematical relation includes the term $(1-V_p)^n$, and uses a generalised simultaneous fitting of all relevant composite data. The fitting is most reliable when the spread/range of porosity fractions is relatively large.

With this method the mechanical properties of the natural fibres are estimated and presented in Table 4, where glass fibres are included for comparison.

Table 4: Mechanical properties of plant fibres and glass fibres
estimated from composite experimental data and composite theory

Fibre	Stiffness GPa	Strength MPa
Jute	53	360
Flax	44	290
Wheat straw	41	280
Glass	70	3000

These values represent the effective properties of the fibres in the composites. The scatter is relatively moderate, and it is of the order ± 3 GPa for stiffness and ± 20 MPa for strength. The values are first estimates based on many experimental results, but often with a limited spread of parameters, especially porosity content, for an efficient data fitting.

The stiffness values for jute, flax and wheat straw are comparable to, but somewhat lower than expected for these fibres.

CONCLUSIONS

Natural fibre composites are fabricated, based on jute, flax and wheat straw fibres, respectively, and a polypropylene matrix.

The fibres are characterised chemically and all have a high cellulose content, 60-70 percent.

The treatment by wet oxidation of the wheat straw fibres may give improved surface characteristics in relation to composites; the jute and flax fibre have untreated surfaces.

The composites are characterised structurally; the densities are in the range 0.8-1.0 g/cm^3; the fibre *weight* fractions are in the range 30-60 percent; the fibre *volume* fractions show a maximum at about 30 percent; and the porosity content is in the range 2-30 percent.

The fibre mat characteristics and the composite processing conditions are not optimised.

The composites are characterised mechanically; the increased composite stiffness over that of the pure matrix indicates on efficient reinforcement by the fibres; the composite (ultimate) strength values are comparable to those of the matrix; at realistic working strains the composites have higher strength values.

The fibre stiffness and strength values are estimated; these values are acceptable and somewhat lower than literature quoted values; the natural fibres are a potential supplement to glass fibres for composites.

REFERENCES

1. Lilholt, H., and Bjerre, A.B., “Composites based on jute-fibres and polypropylene matrix, their fabrication and characterisation”. Proceedings of the 18th Risø International Symposium on Materials Science: Polymeric Composites – Expanding the limits, (S.I.Andersen, P. Brøndsted, H. Lilholt, Aa. Lystrup, J.T. Rheinländer, B.F. Sørensen, H. Toftegaard (eds.)), Risø National laboratory, Denmark, 1997, pp. 411-423.

2. Andersen, T.L. and Lilholt, H., “Natural fibre composites: compaction of mats, press consolidation and material quality”, Proceedings of the Euro-Japan Symposium, Paris, France, July 1-2, 1999 pp.xx.

3. Toftegaard, H., [paper in preparation], 1999.