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CHEM-E2200: Polymer blends and composites

Reinforcement processes

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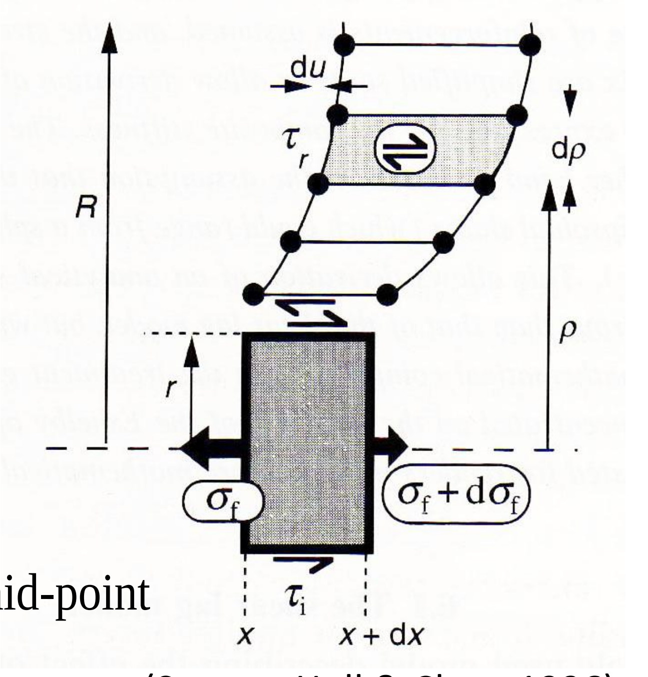
Today's topics

- A little more about stress transfer
- Reinforcement by slip
- Conditions for fibre fracture
- The effect of fibre behaviour on composite micromechanics

Elastic stress transfer: interfacial shear stresses

- Interfacial shear stress (τ_i) operates at the interface, parallel to the fibre surface
- Using a force-balance approach it can be shown that:

$$\frac{d\sigma_f}{dx} = -\frac{2\tau_i}{r}$$



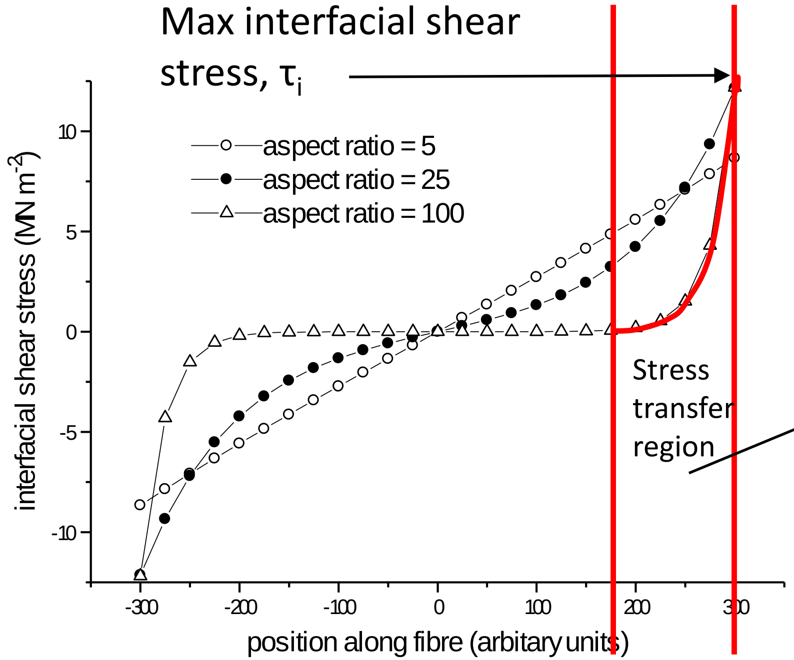
(Source: Hull & Clyne 1996)

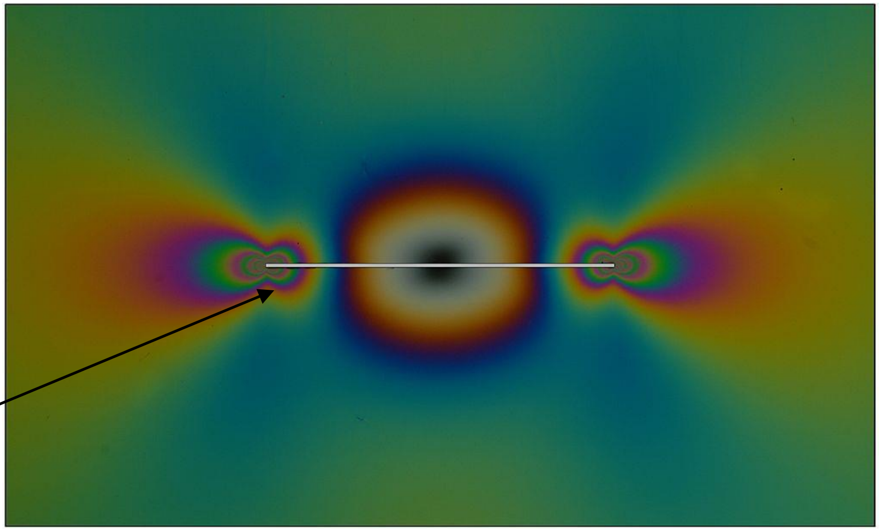
Where: x is the axial distance from the fibre mid-point

r is the fibre radius

- **The model assumes that there is no slippage at the interface, i.e. the system behaves elastically**

Shear stress distribution



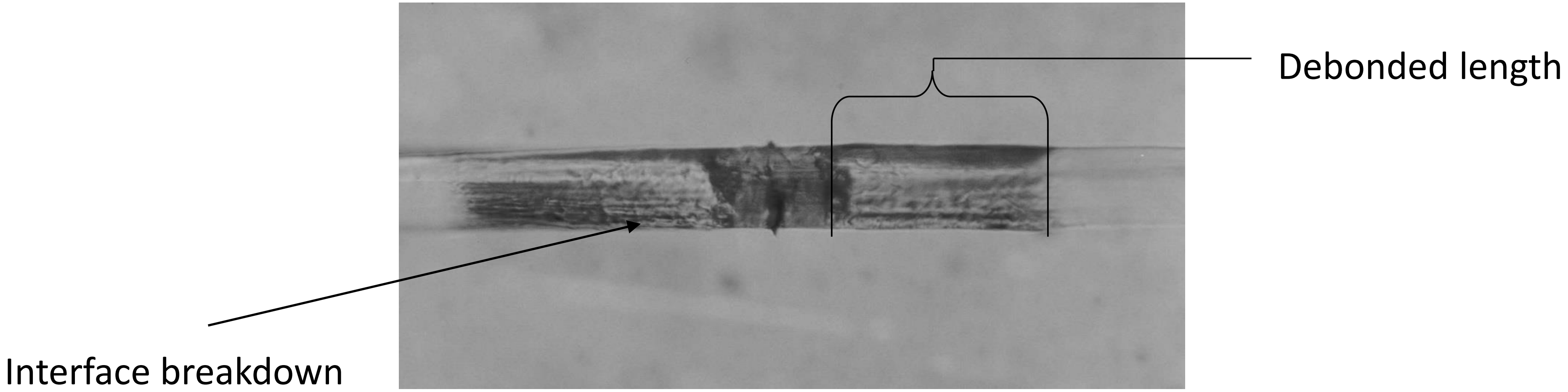


SSMG-ITALY - Laboratory for Physical Modelling of Structures and Photoelasticity (University of Trento, Italy)

- **Maximum interfacial shear stress at the fibre ends**

Fibre-matrix debonding

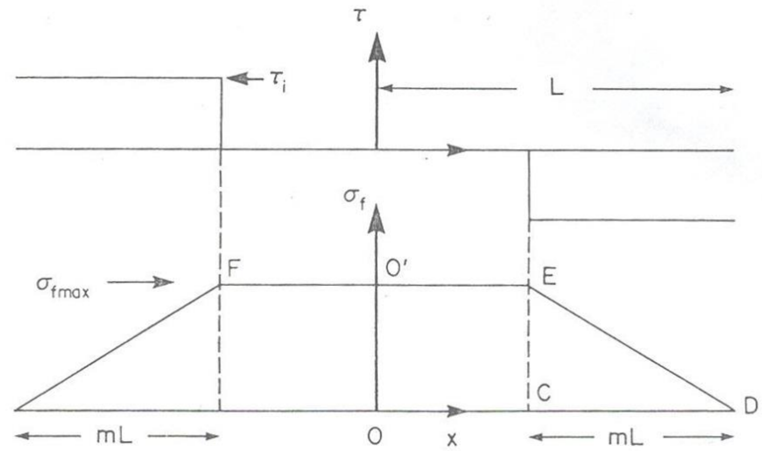
- Highest interfacial shear stresses are at the fibre ends
- Debonding is clearly visible due to a change in the refractive properties of the interface
- This is particularly visible in fragmented fibres



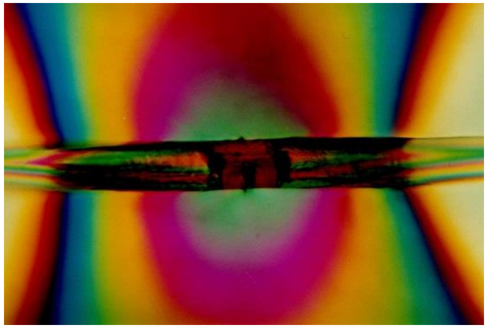
Debonded fibre ends in fragmented fibre (flax-epoxy SFC)

Reinforcement by slip

- If the interfacial shear stress exceeds some critical value (the interfacial shear strength), then the interface will breakdown leading to a loss in adhesion
- Reinforcement may still take place through frictional forces at the interface and a process analogous to the Cox shear-lag mechanism will operate
  - (Note: $F_{max} = \mu_s F_n$ for static friction)
- Assuming that the frictional interfacial shear stress is constant at the ends of the fibre, a model of the axial fibre stress is as shown opposite



Model of stress transfer by slip (Piggott, 1980)



Photoelastic response in an epoxy matrix at fibre ends

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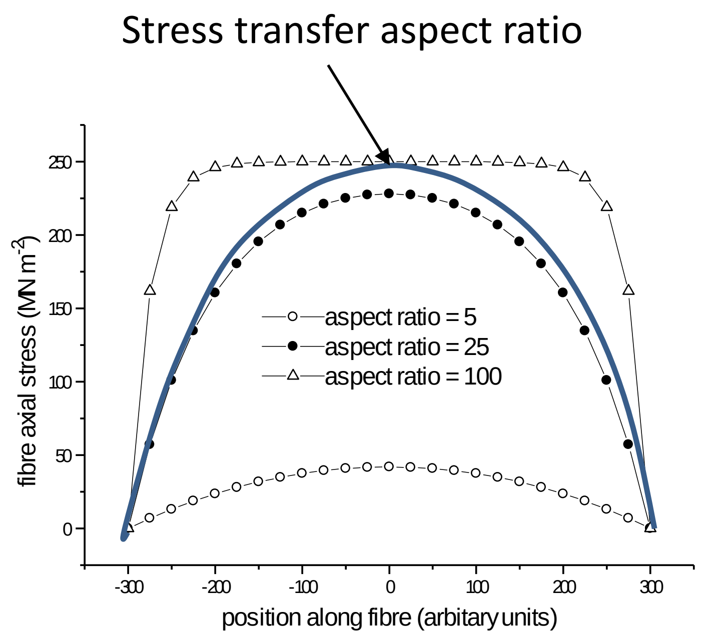
Stress transfer aspect ratio

- Whether by elastic stress transfer, or by friction, the axial fibre stress increases from the ends of the fibre towards the mid-point
- Over the stress transfer length, the fibre is not being fully effective as reinforcement
- If the aspect ratio, *s*, is too small, then the fibre axial stress will not reach a maximum and thus is not acting efficiently



- There will therefore be an aspect ratio, where the fibre axial stress just reaches a maximum value, for the applied composite strain. This is known as the **stress transfer aspect ratio**

Conditions for fibre failure

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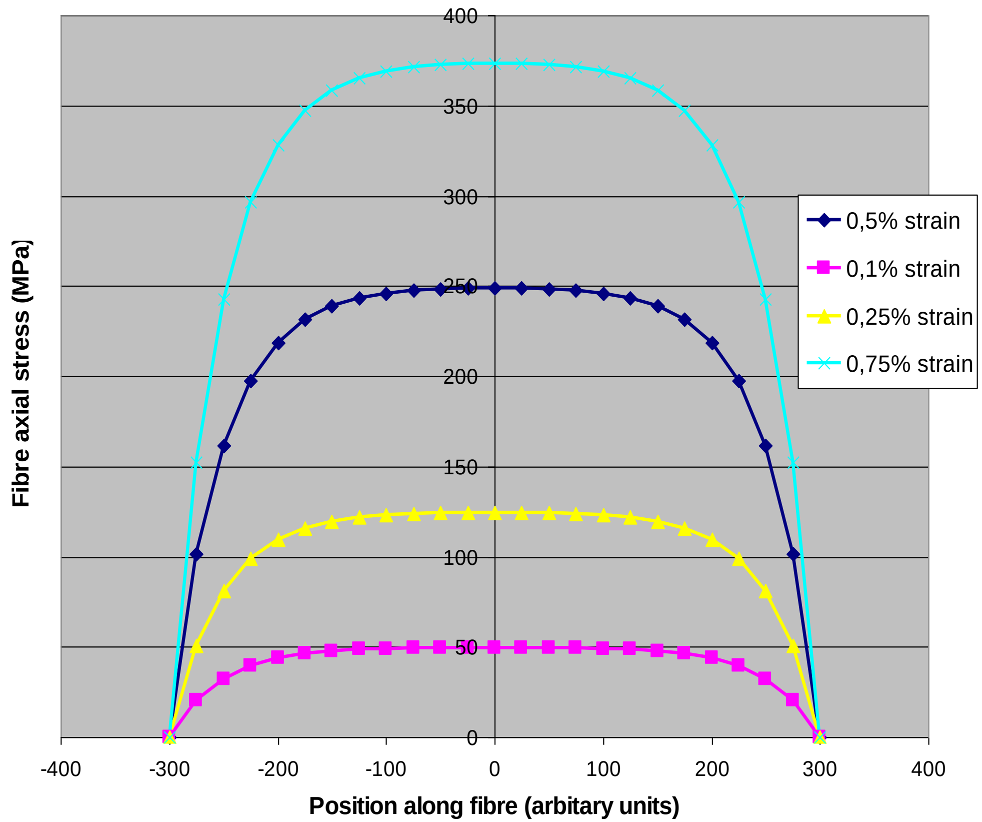
Whether by elastic stress transfer, or by friction, the axial fibre stress increases from the ends of the fibre towards the mid-point

Over the stress transfer length, the fibre is not being fully effective as reinforcement

If the aspect ratio, s, is too small, then the fibre axial

Fibre axial stress

- As the composite is deformed, the maximum value of axial fibre stress will increase (although the stress transfer profile will be the same)
- When the fibre axial stress reaches the tensile strength of the fibre, it will fail
- The fibre aspect ratio will decrease
- As the strain on the composite increases, the axial stress in the remaining section will continue to increase, leading to further fibre failure



Theoretical build up of fibre axial stress following a Cox type shear lag mechanism (E_f = 50 GPa; E_m = 3.5 GPa; s = 50)

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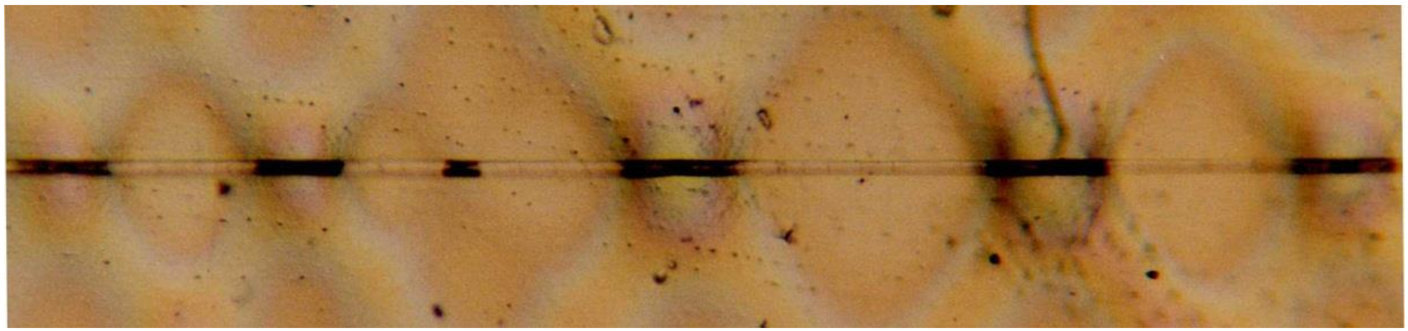
Fibre fracture

- As longer fibres are progressively shortened, the maximum axial tensile stress in the fibre that can be generated will reduce, to a point below which further fracture cannot occur. I.e. the interface fails before the fibre
- This is known as the critical fibre length



Fragmented flax fibre in an epoxy matrix SFC

Critical fibre aspect ratio

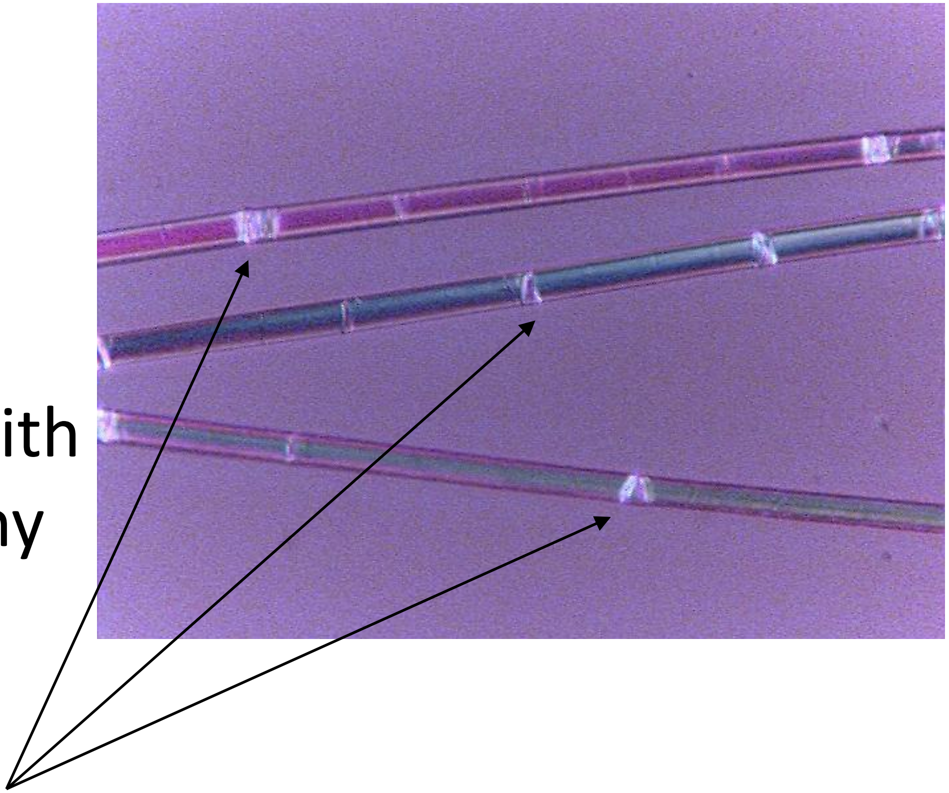
- A critical aspect ratio (S_c) can thus be identified. This is the point where the central axial stress in the fibre equals the ultimate tensile strength of the fibre (σ_{fu}). It can be shown that if stress transfer by slip (friction) is considered then:

$$S_c = \frac{\sigma_{fu}}{2\tau_{i^*}}$$

Where τ_{i*} is in the interfacial shear stress

Fibre strength

- Is the strength of the fibre going to be the same along its entire length?
- Unlikely! Particularly with natural fibres with many defects…

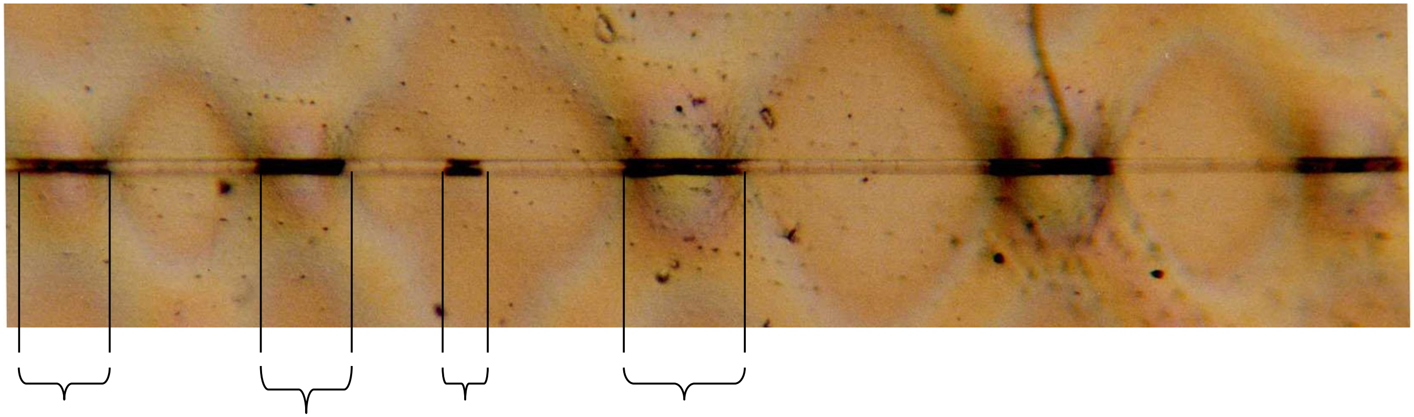


Fibre defects in flax fibre; points of weakness, seen under polarised light

Composite behaviour & relationship to micromechanics

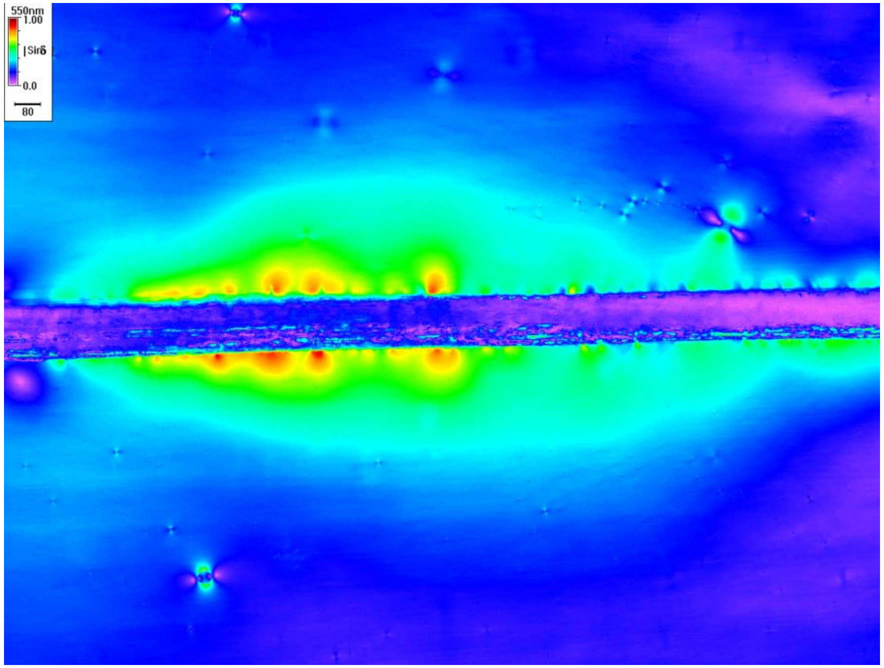
- Lower fibre aspect ratio, lower composite stiffness
- Theory predicts that the stress-strain relationship will be linear
- Non-linear behaviour will occur when microstructural damage commences. For example, if there is matrix yielding, or if interfacial failure occurs, leading to a reduction in ‘efficiency’ of stress transfer. If the fibre fractures, then it is unable to provide as effective reinforcement and therefore the composite stiffness will reduce

Interfacial failure

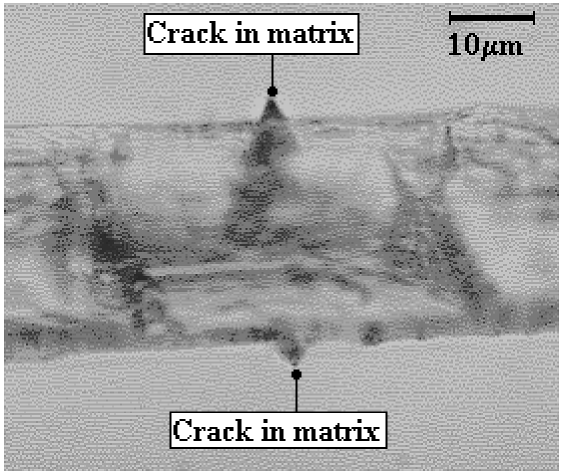


Interfacial debonding

Interfacial failure and matrix yielding

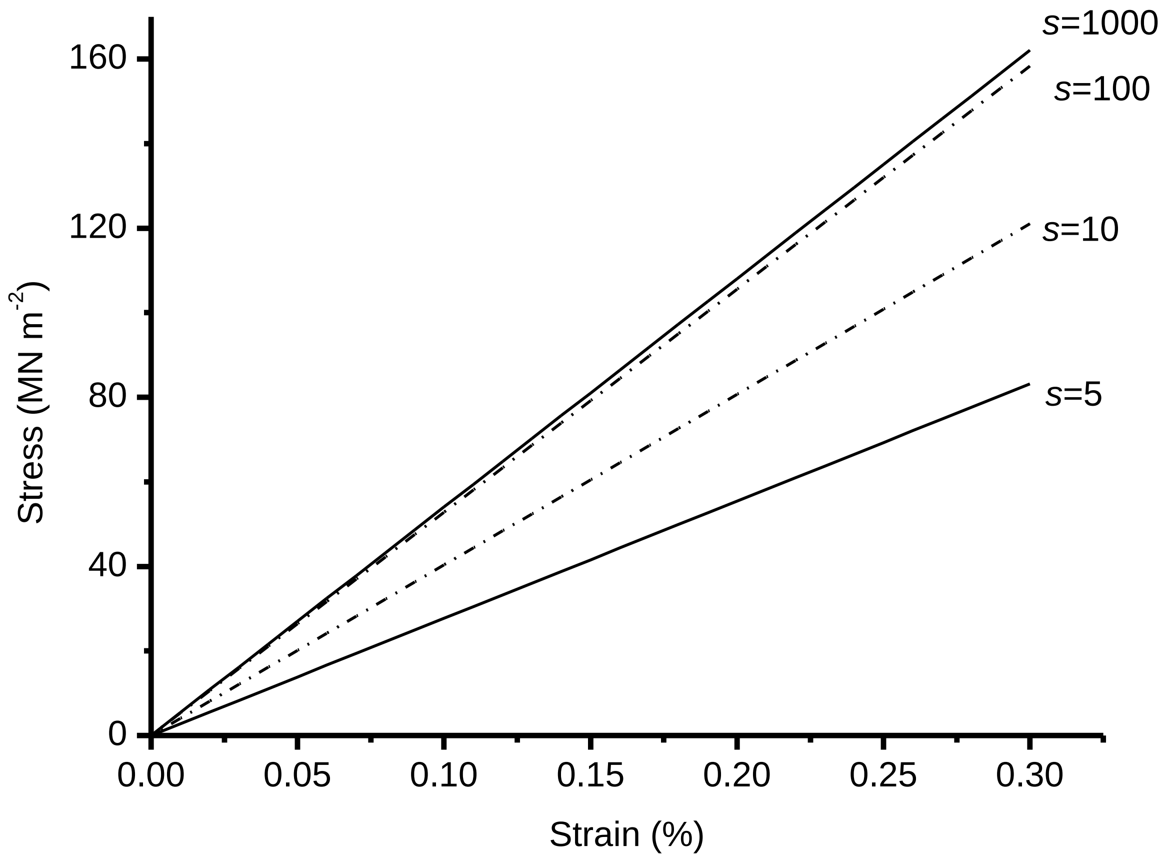


Matrix yielding



Matrix cracking
(Hughes et al 2000)

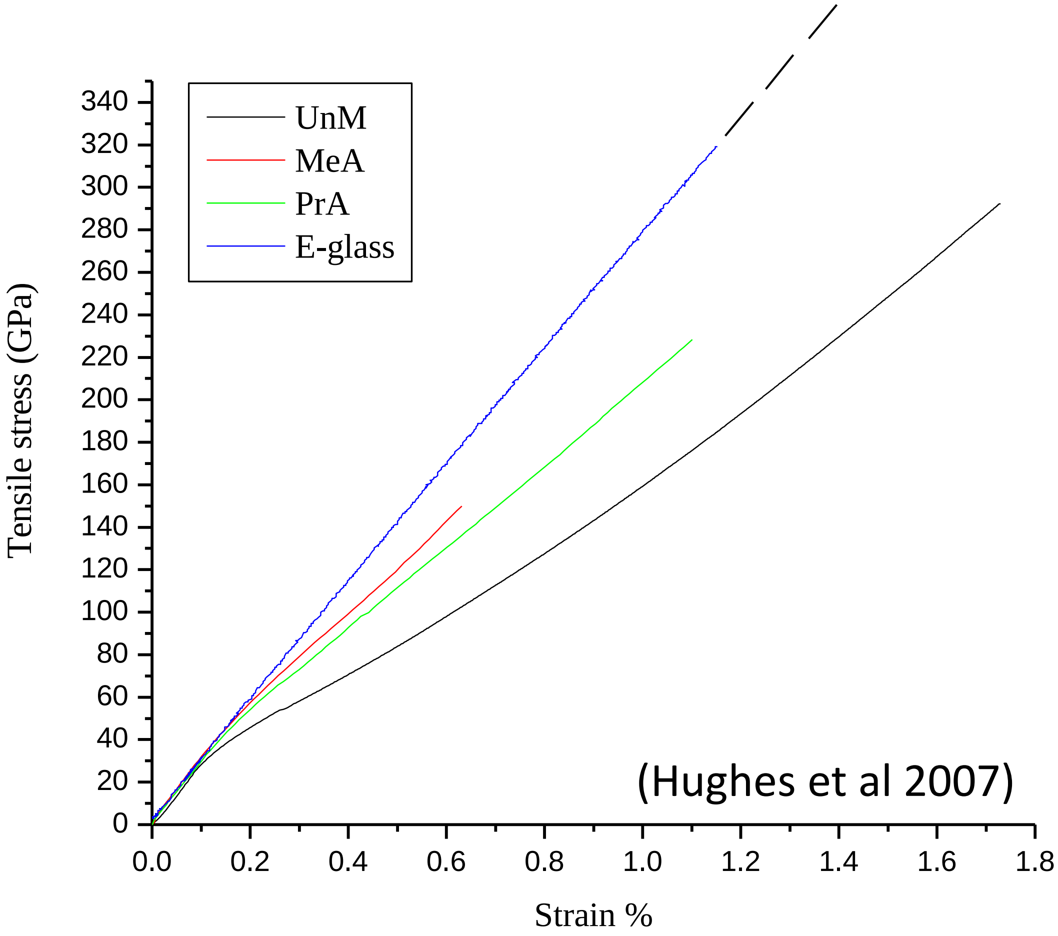
Effect of aspect ratio on composite stiffness



Case study: the use of plant fibre as composite reinforcement

Composite behaviour

- What is the influence of changing the degree of interfacial bonding?
- Three forms of flax fibre-reinforced unsaturated polyester composite investigated:
  - No fibre treatment
  - Hydrophobic fibre surface (increased wetting)
  - Chemical bonding
- Stress-strain (and failure) characteristics altered

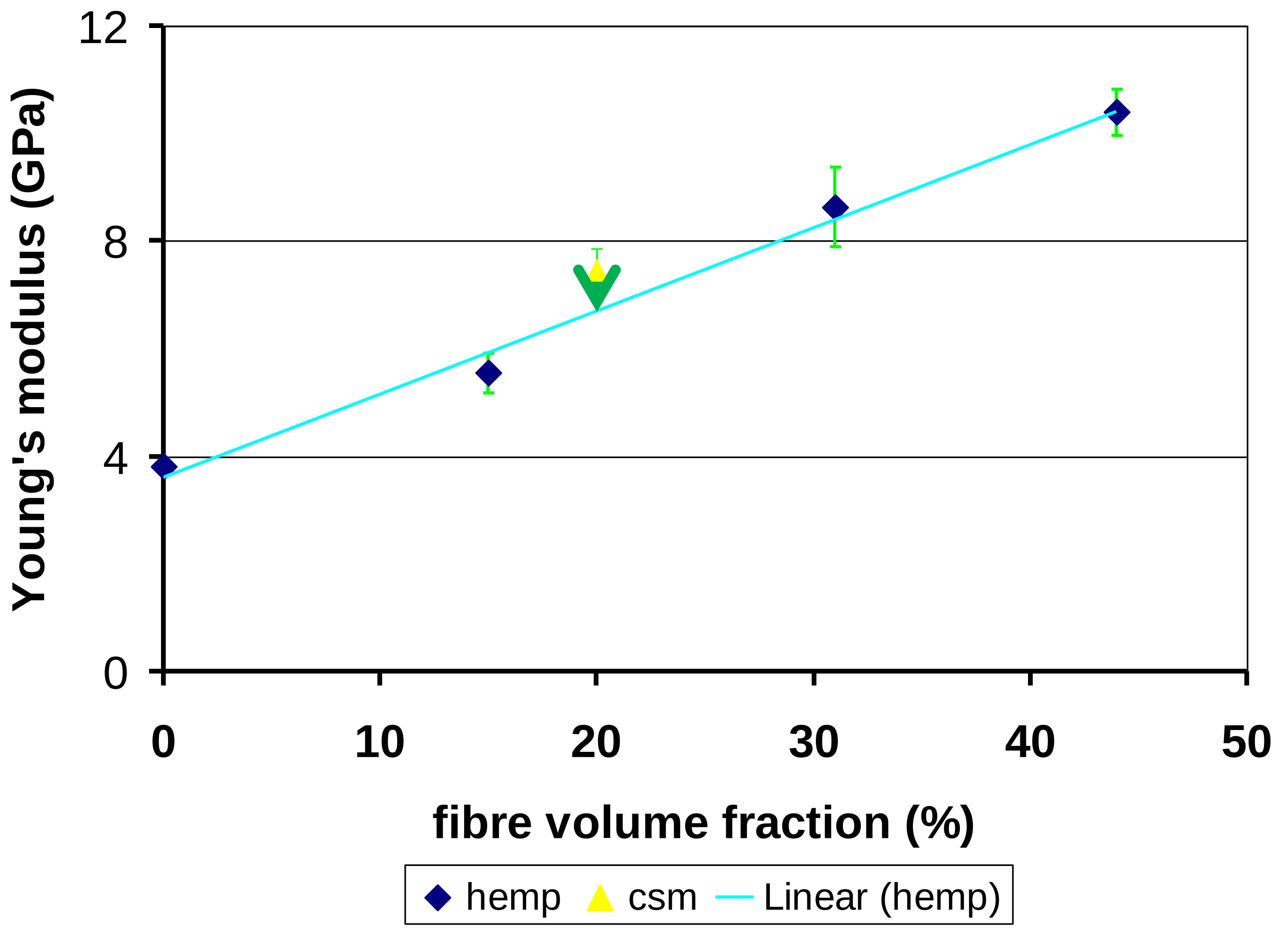


(Hughes et al 2007)

Properties of polymer-matrix composites based on natural fibres (hemp)

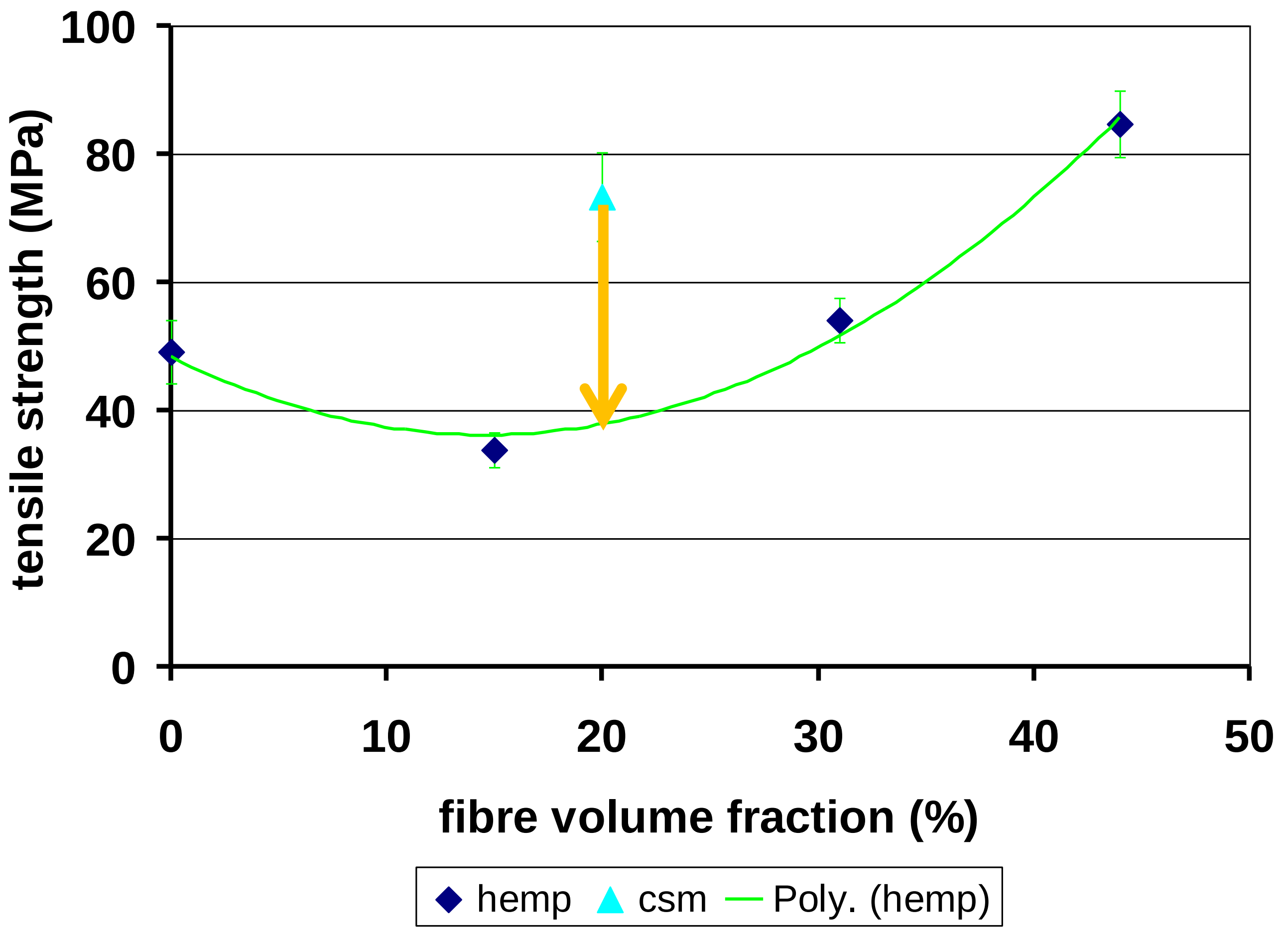
- Generally good stiffness - similar to glass-fibre reinforced material, especially on a specific basis
- Adequate strength for many commonplace applications, if not too demanding
- Poor toughness - order of magnitude lower?
- Yielding at low stress levels, making the working range of loading quite limited

Stiffness



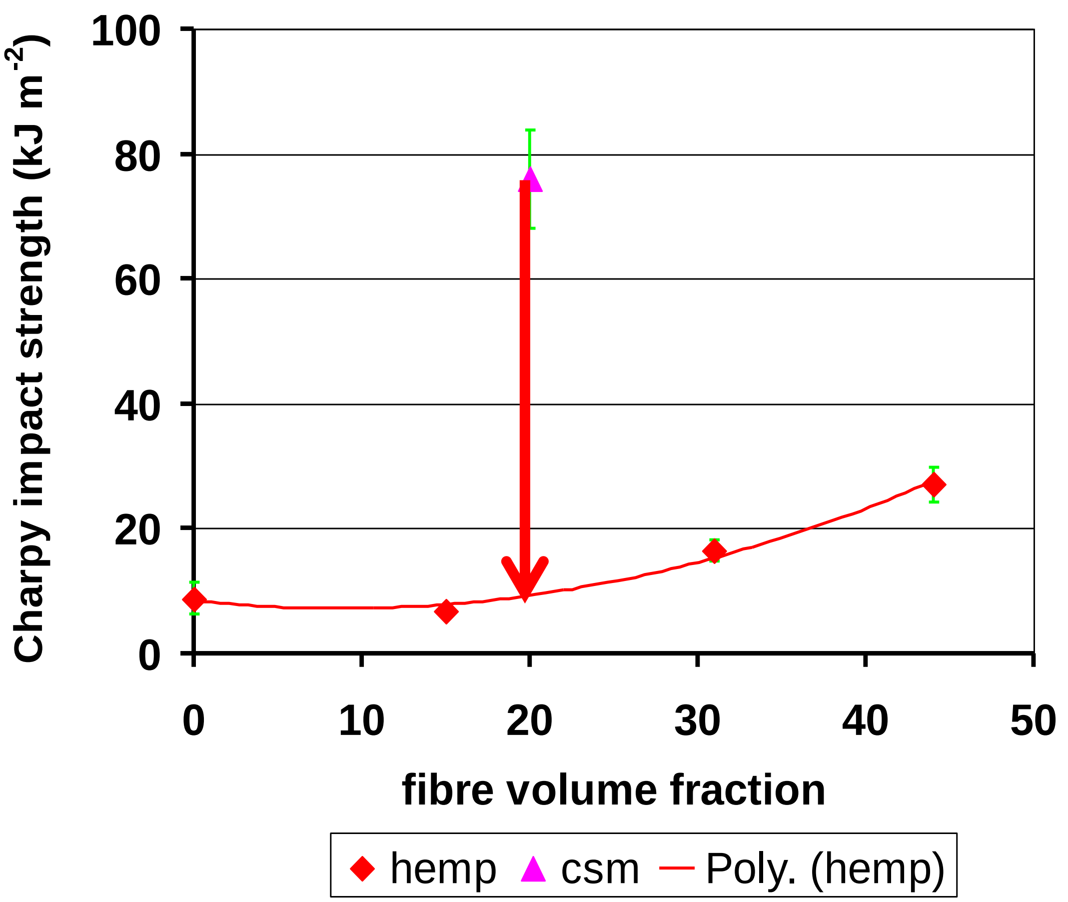
Young's modulus (GPa)
0
4
8
12
0
10
20
30
40
50
fibre volume fraction (%)
hemp
csm
Linear (hemp)

Strength



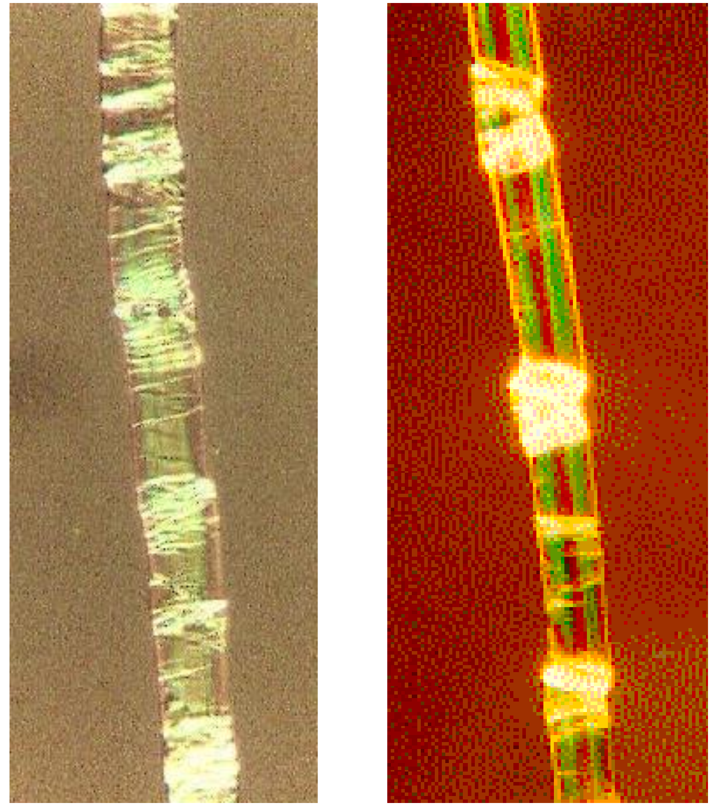
tensile strength (MPa)
100
80
60
40
20
0
0
10
20
30
40
50
fibre volume fraction (%)
hemp
csm
Poly. (hemp)

Toughness (Chapy impact strength)



Polarised light

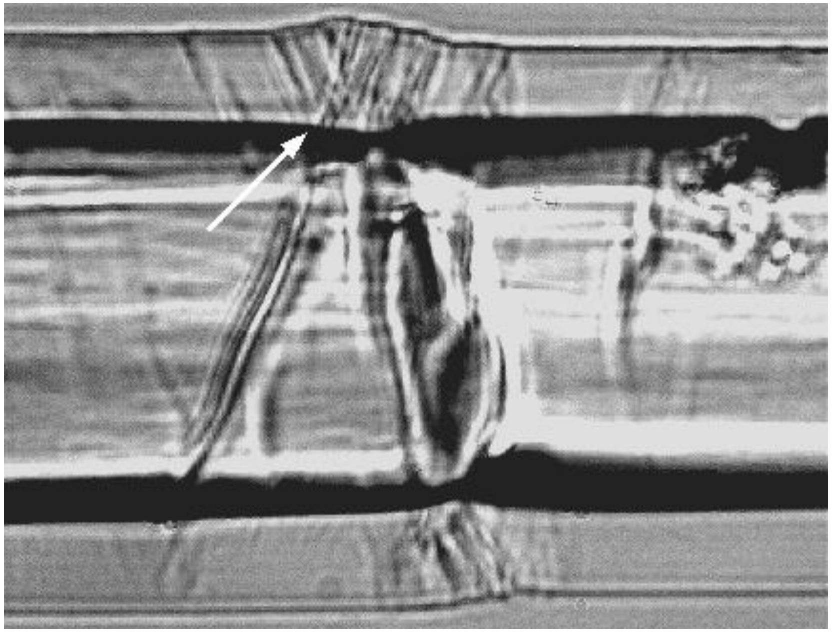
Unprocessed
hemp fibre

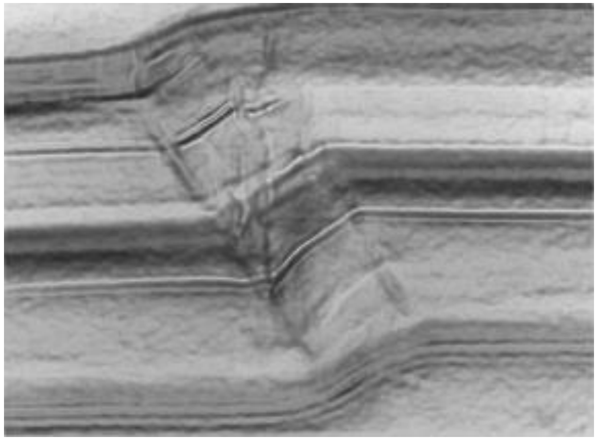


Mechanically processed
hemp fibre

Fibre structure

- Highly aligned structure leads to excellent tensile properties in fibres such as flax, hemp, jute, ramie, etc.
- But: prone to compressive failure through the formation of kink-bands, affecting either the cell wall or the entire fibre
- This affects the behaviour of the fibre





(Hughes et al. 2000)

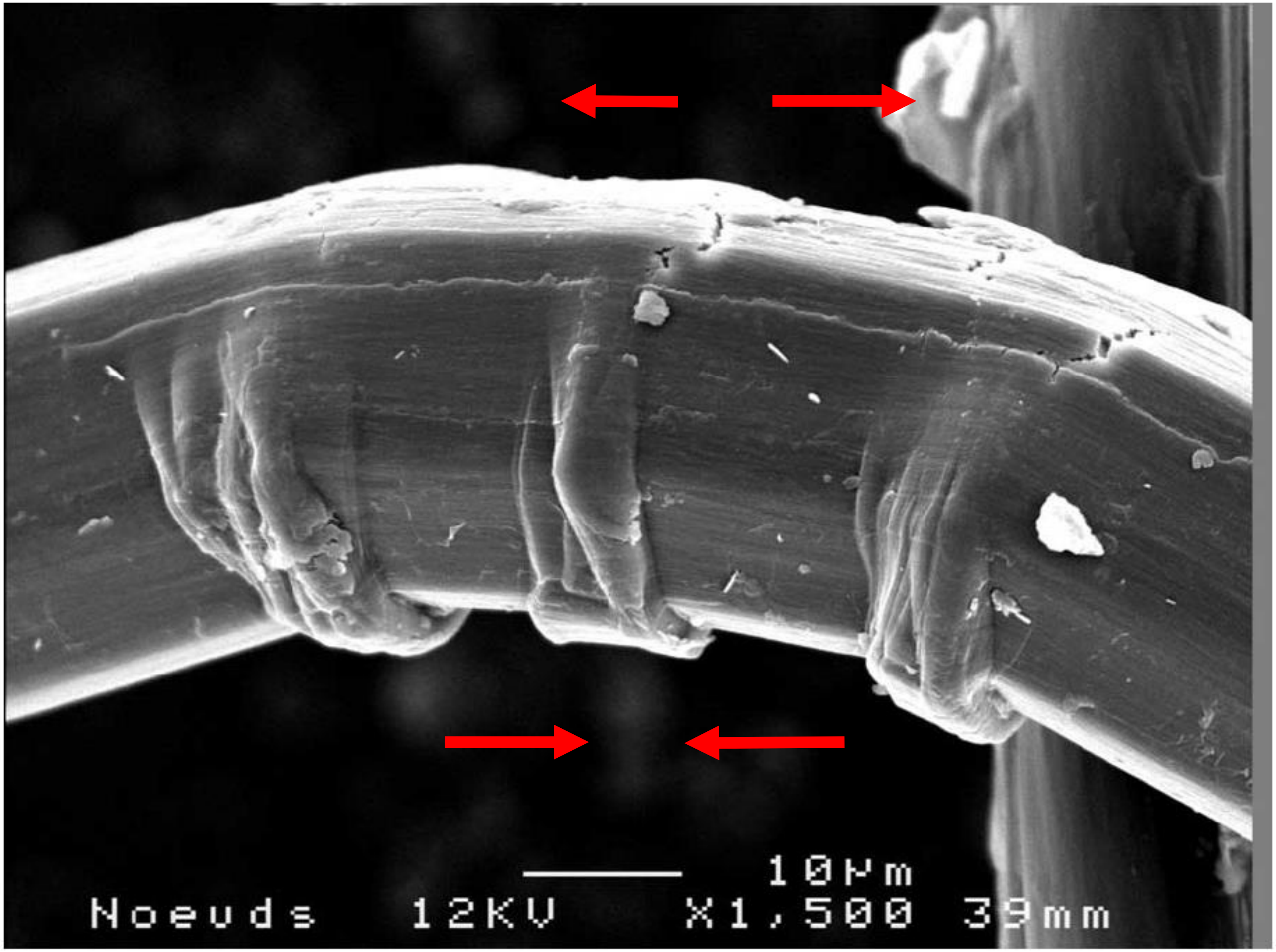


Figure 2 Bending of a flax fiber with buckling of cell walls.

(Baley 2004)

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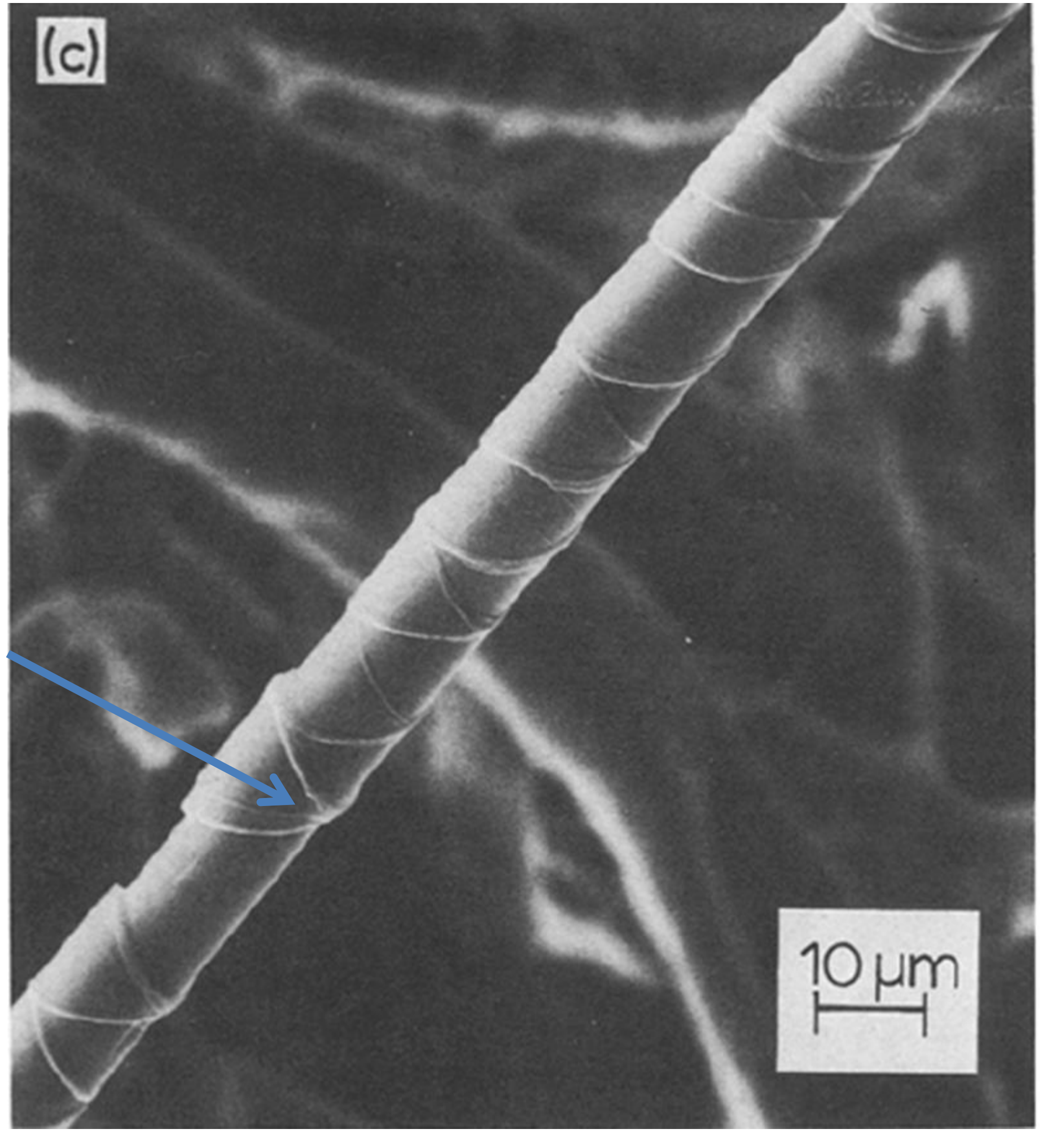
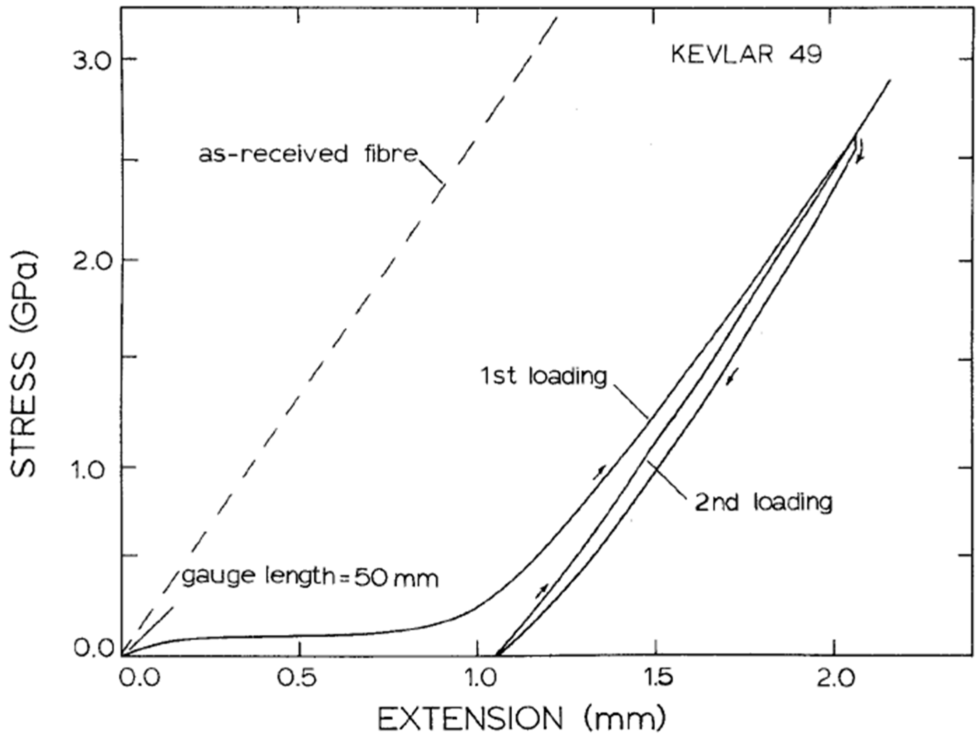
- Failure in wood and non-wood fibre in compression is analogous to the compression failure seen in polymer composites, or synthetic fibre such as Kevlar 49



(DeTeresa et al, 1984)



(DeTeresa et al, 1984)

Figure 7 Tensile stress–elongation curve of Kevlar 49 fibre previously compressed ~ 3% due to matrix shrinkage.

- Kevlar 49 fibre without defects exhibits Hookean behaviour
- Fibre containing kinks induced by compressive failure, exhibit significant non-linear behaviour initially, before strain hardening
- In subsequent cycles the fibre exhibits nearly linear behaviour

Kink bands in flax fibre

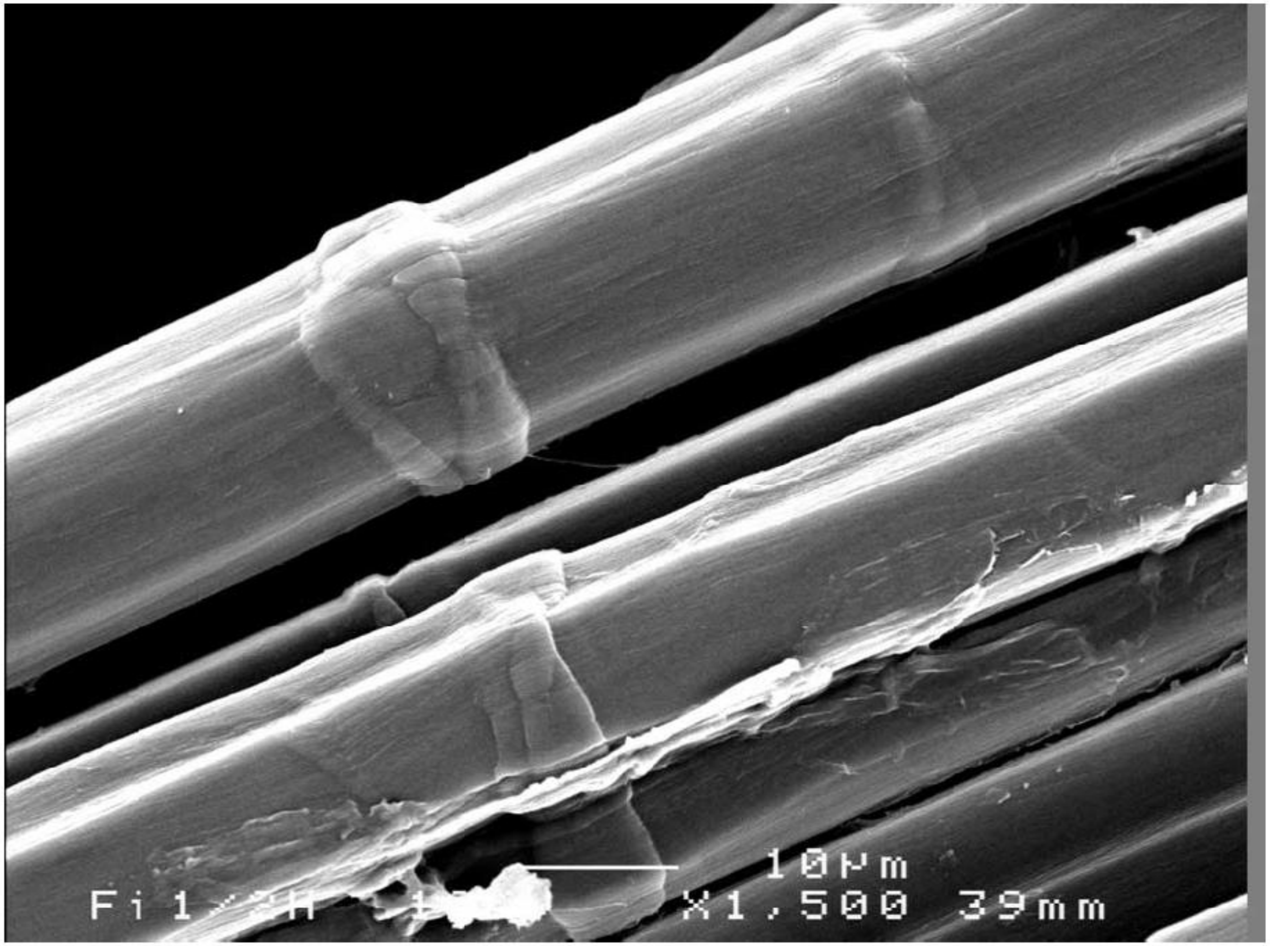
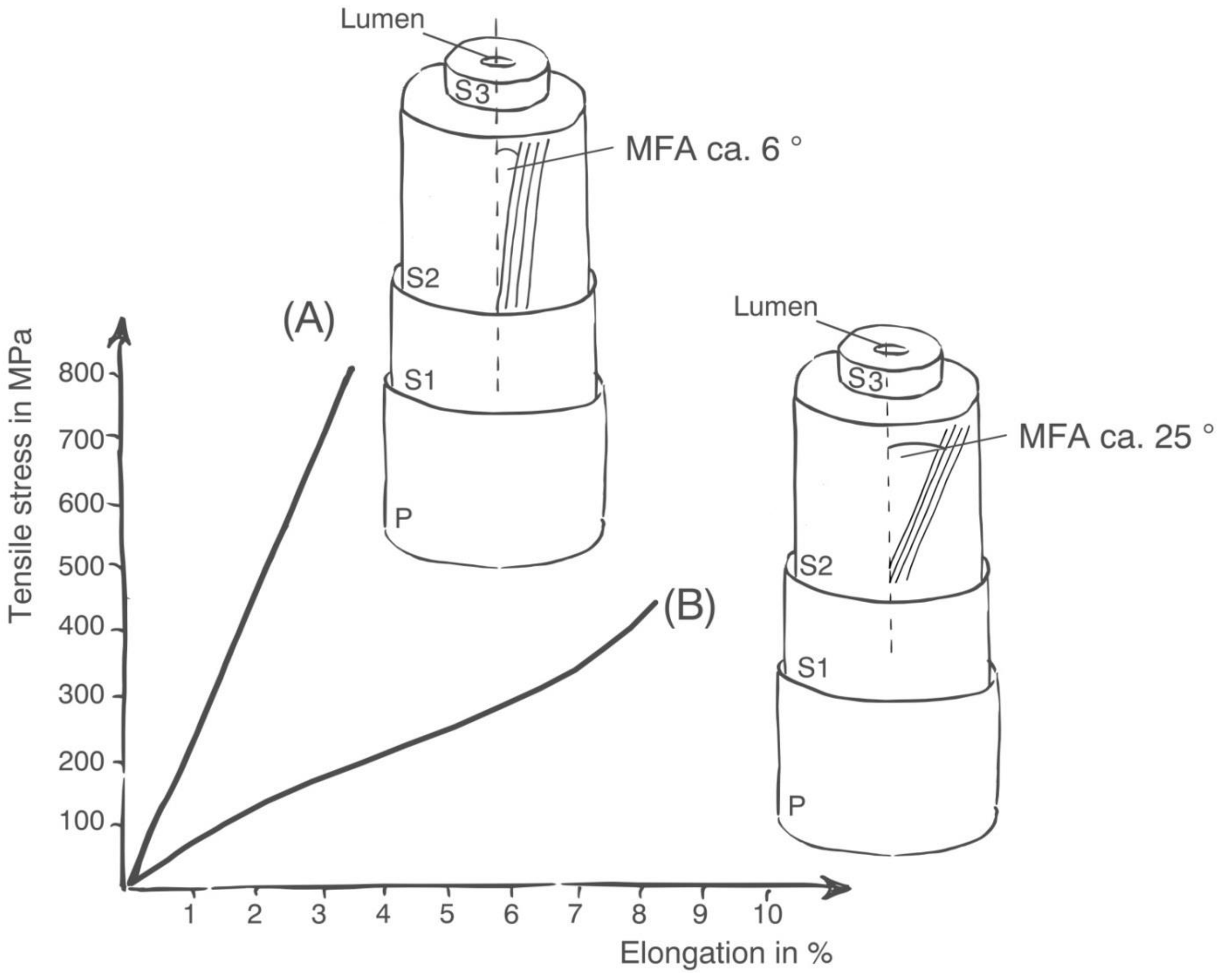


Figure 1 Flax fibers: Example of a bundle of flax fibers with kink bands in the same area.

(Baley 2004)

Stress-strain behaviour of fibres



- Stress-strain behaviour strongly influenced by the microfibril angle

Tensile stress-strain behaviour of elementary flax fibre

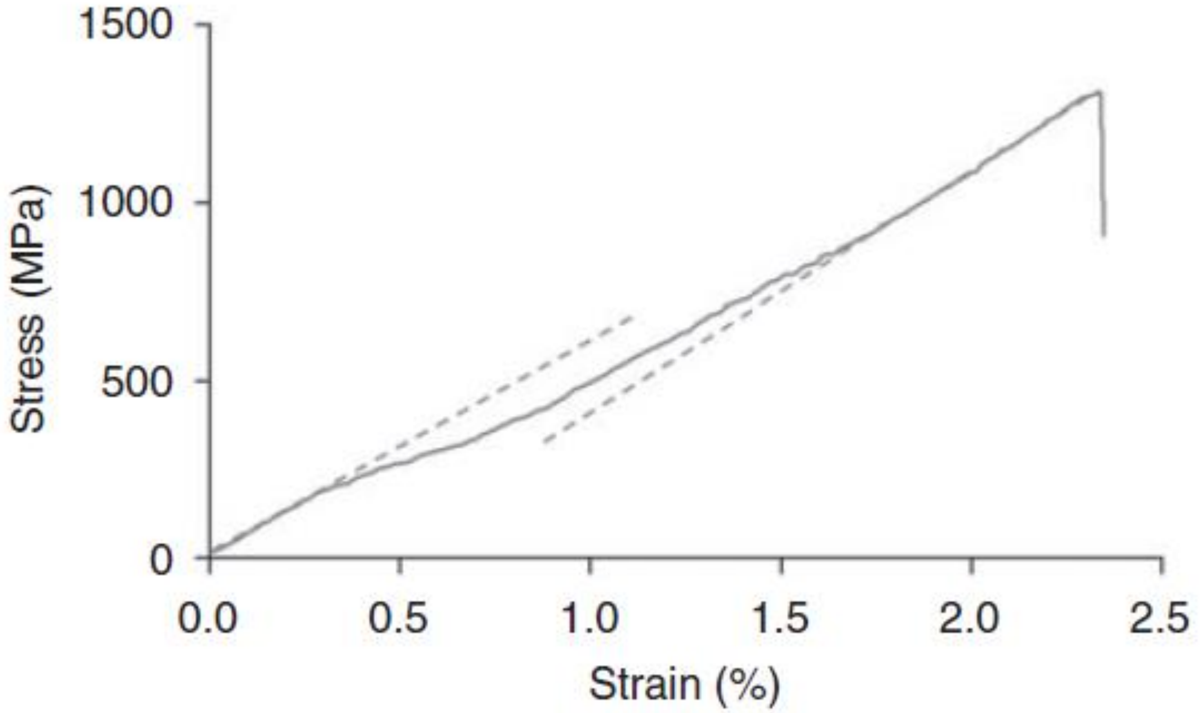
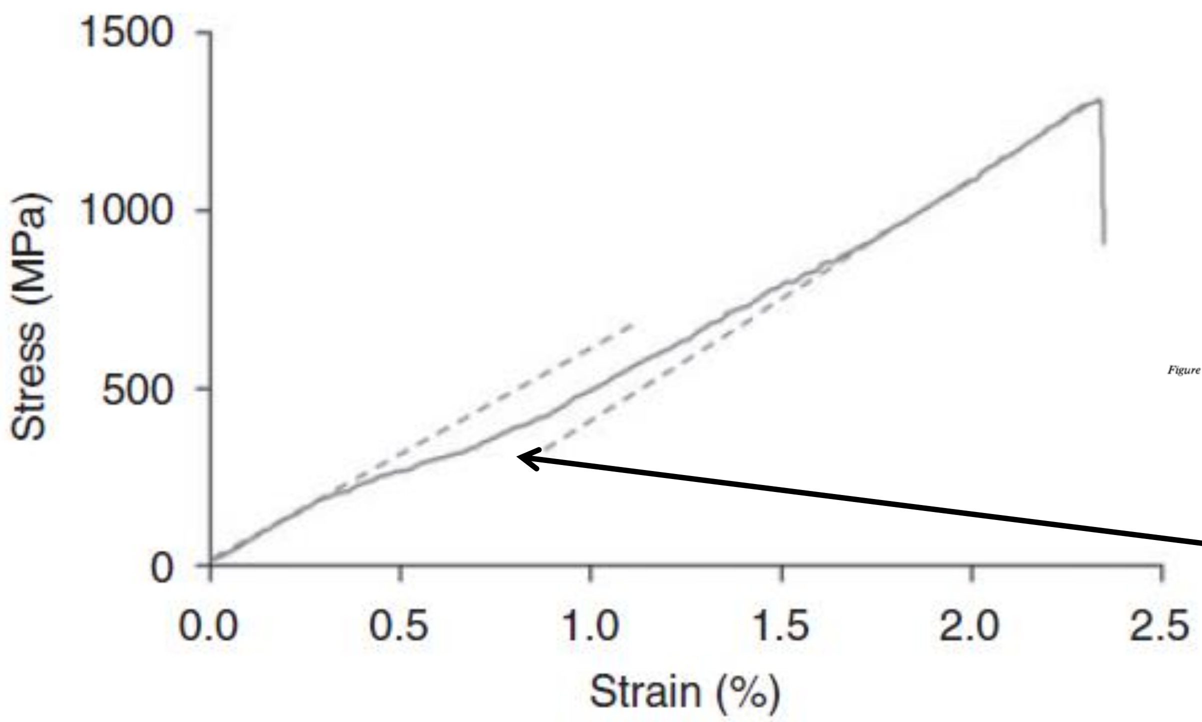


Figure 1. *Typical stress–strain curve of an elementary flax fiber.*

(Charlet et al, 2010)

Tensile stress-strain behaviour of elementary flax fibre



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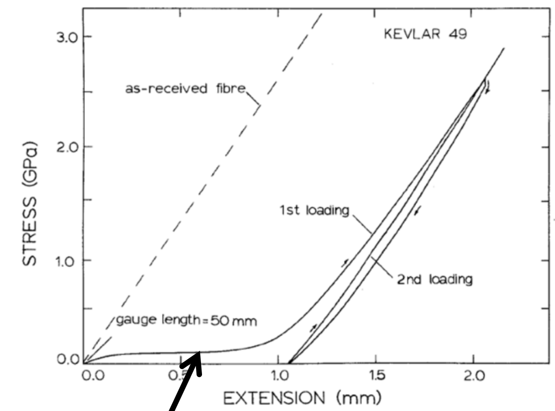


Figure 7 Tensile stress–elongation curve of Kevlar 49 fibre previously compressed ~ 3% due to matrix shrinkage.

Analogous "S" shaped form to the stress-strain behaviour

FE model proposed by Nilsson & Gustafson (2007)

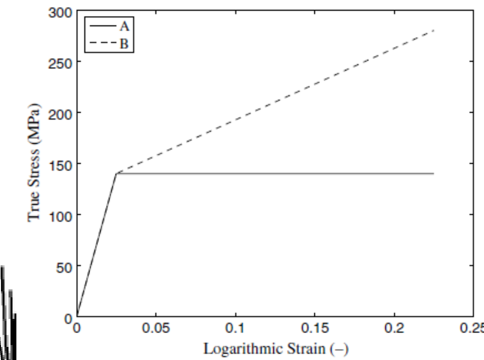


Fig. 5. Plasticity models.

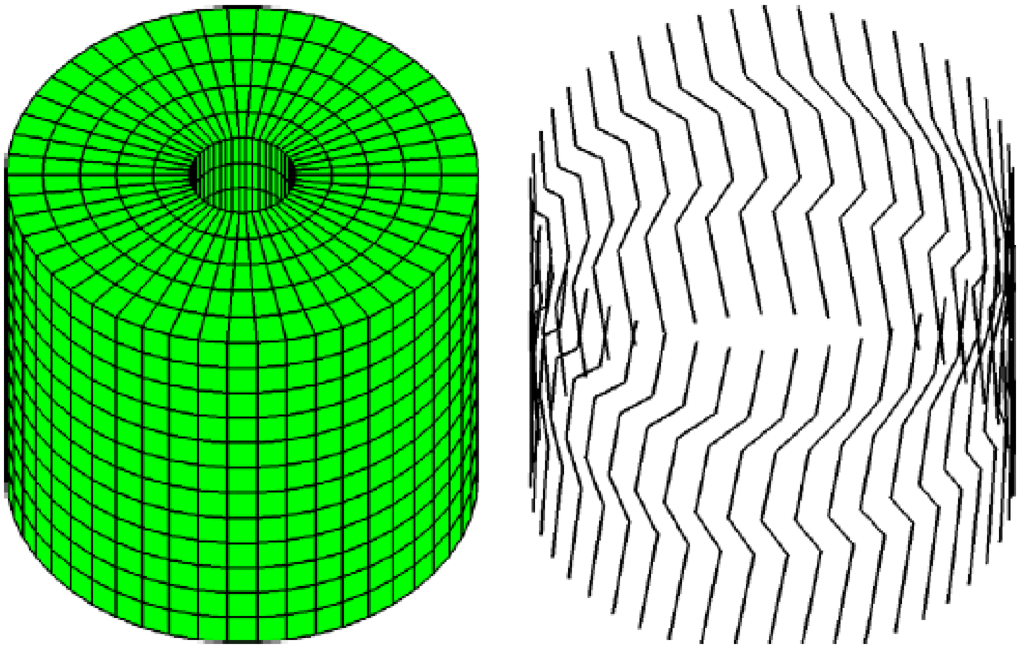


Fig. 4. Part of an FE model with dislocations, hemicellulose to the left and the embedded cellulose with dislocations to the right. Note that only the outermost truss elements are shown.

Table 1
Description of the different analyses

Model 1	Model 2	Model 3
Elastic cellulose $E_C = 130$ GPa	Elastic cellulose $E_C = 130$ GPa	Elastic cellulose $E_C = 130$ GPa
Elastic hemicellulose $E_{HC} = 5.6$ GPa	Elasto-plastic hemicellulose model A in Fig. 5	Elasto-plastic hemicellulose model B in Fig. 5
Small deformation theory	Large deformation theory	Large deformation theory

(Nilsson & Gustafson 2007)

Modelled tensile behaviour

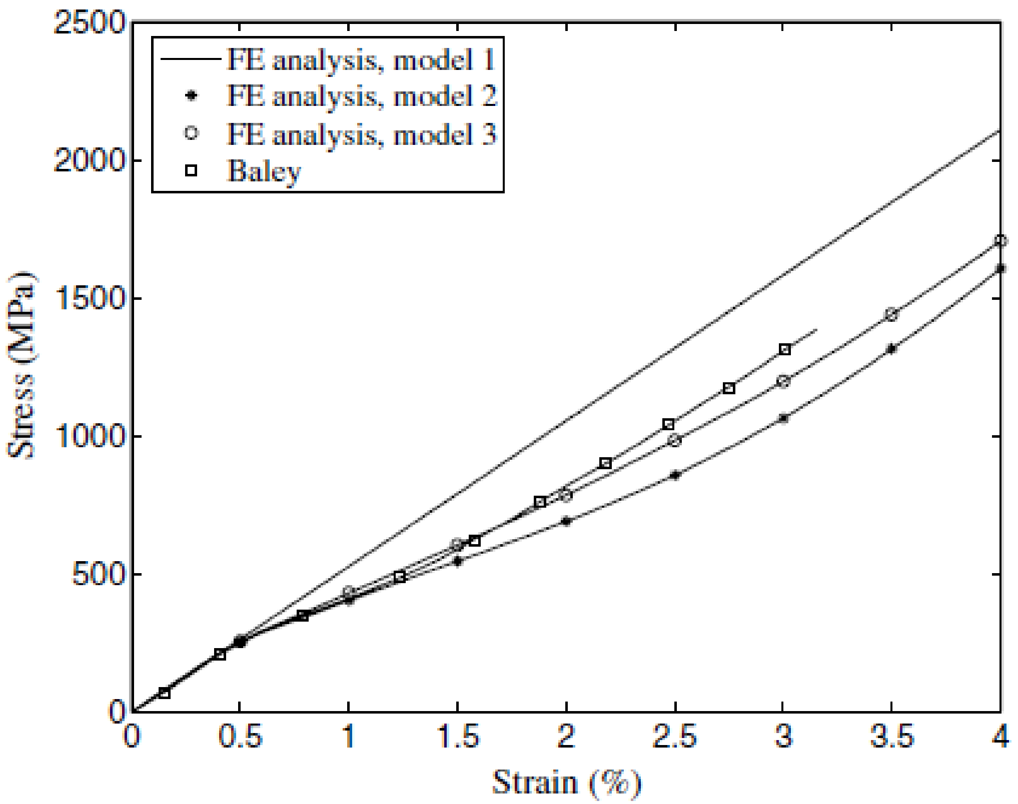


Fig. 7. Tensile behaviour of the elementary fibre.

(Nilsson & Gustafson, 2007)

Effect of the extent of damage

- Increasing levels of damage in flax fibre ultimates results in a lowering of the stiffness of the fibre
- This implies that the fibre undergoes greater strain at the defect
- This has been verified experimentally by Mott et al (1996), who showed that fibre defects acted at strain concentrators
- The same conclusion was reached by Eichhorn et al (2000)

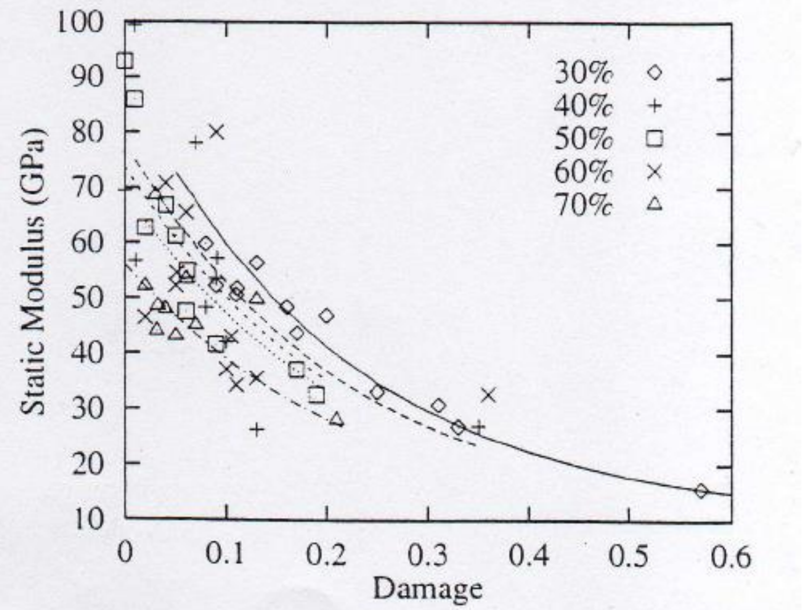
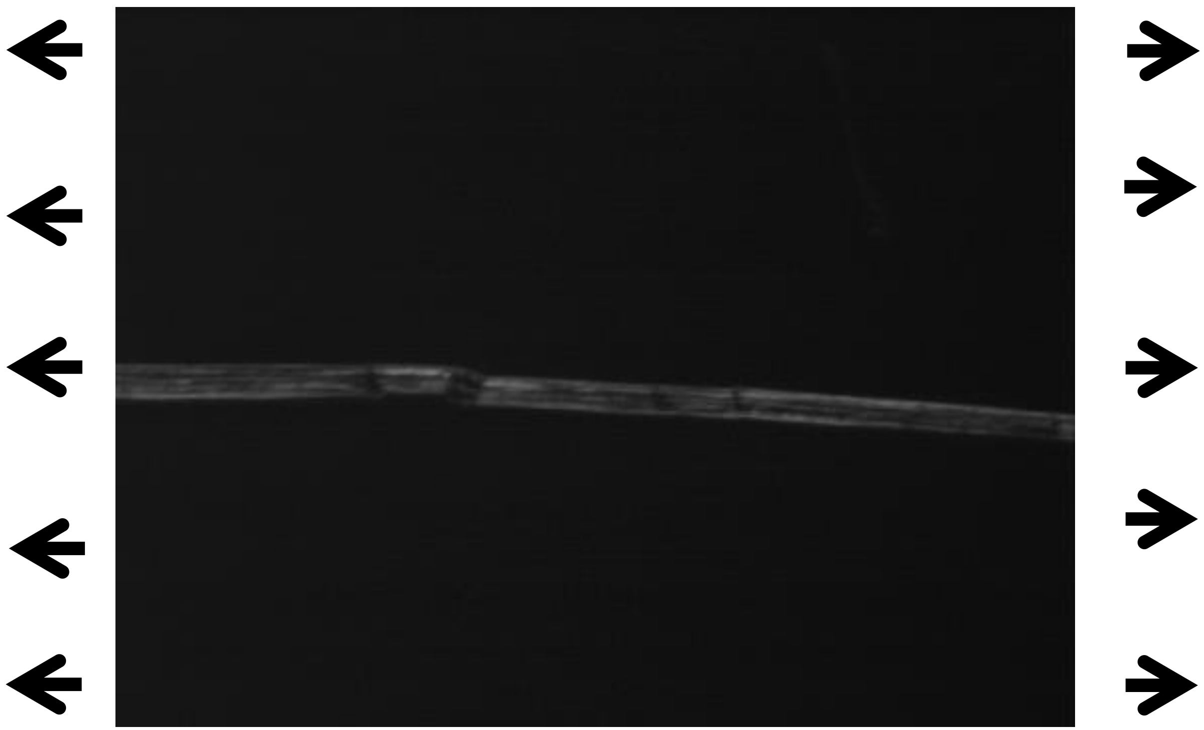
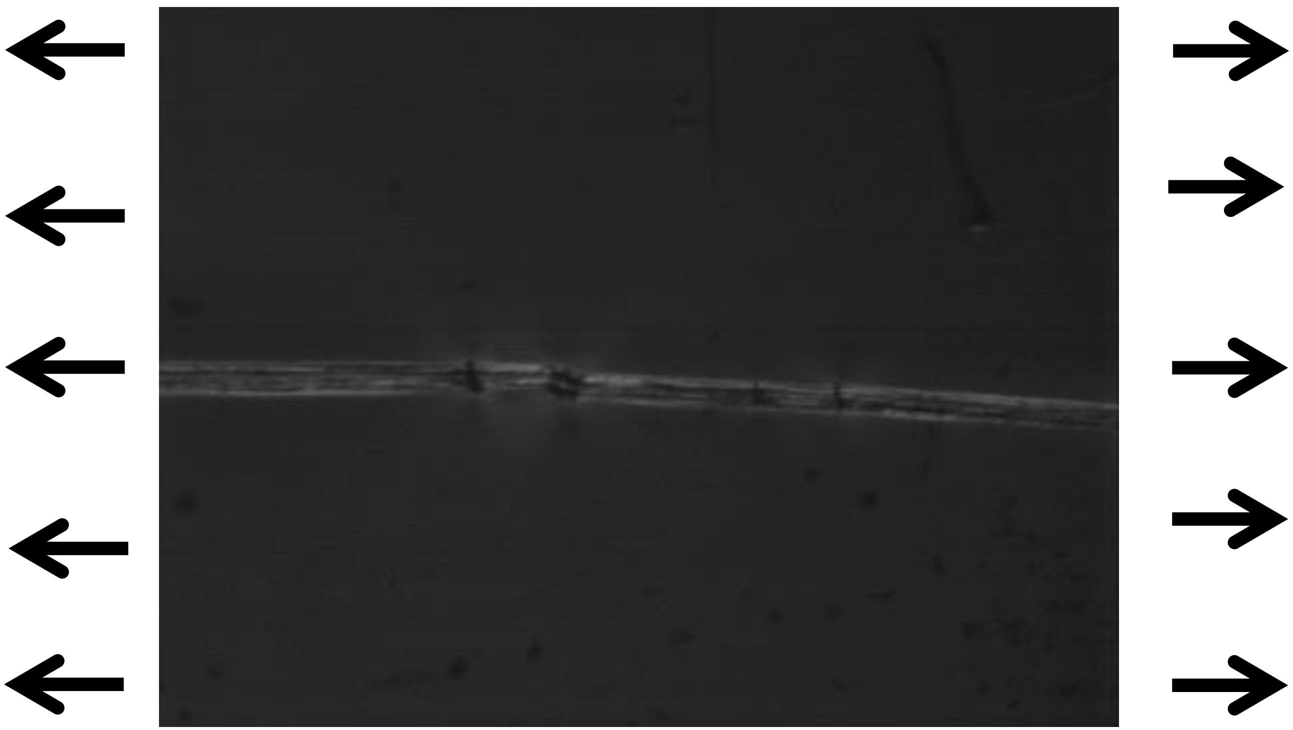
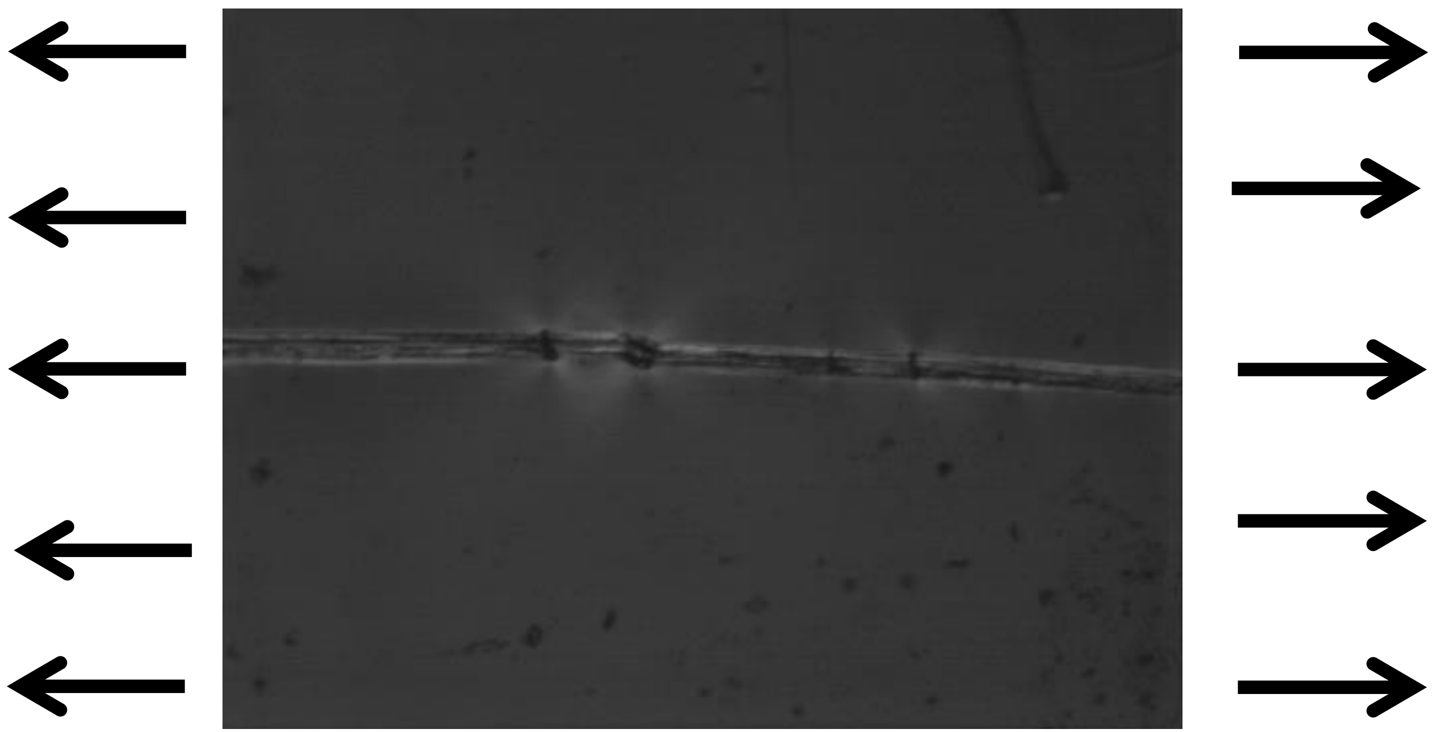


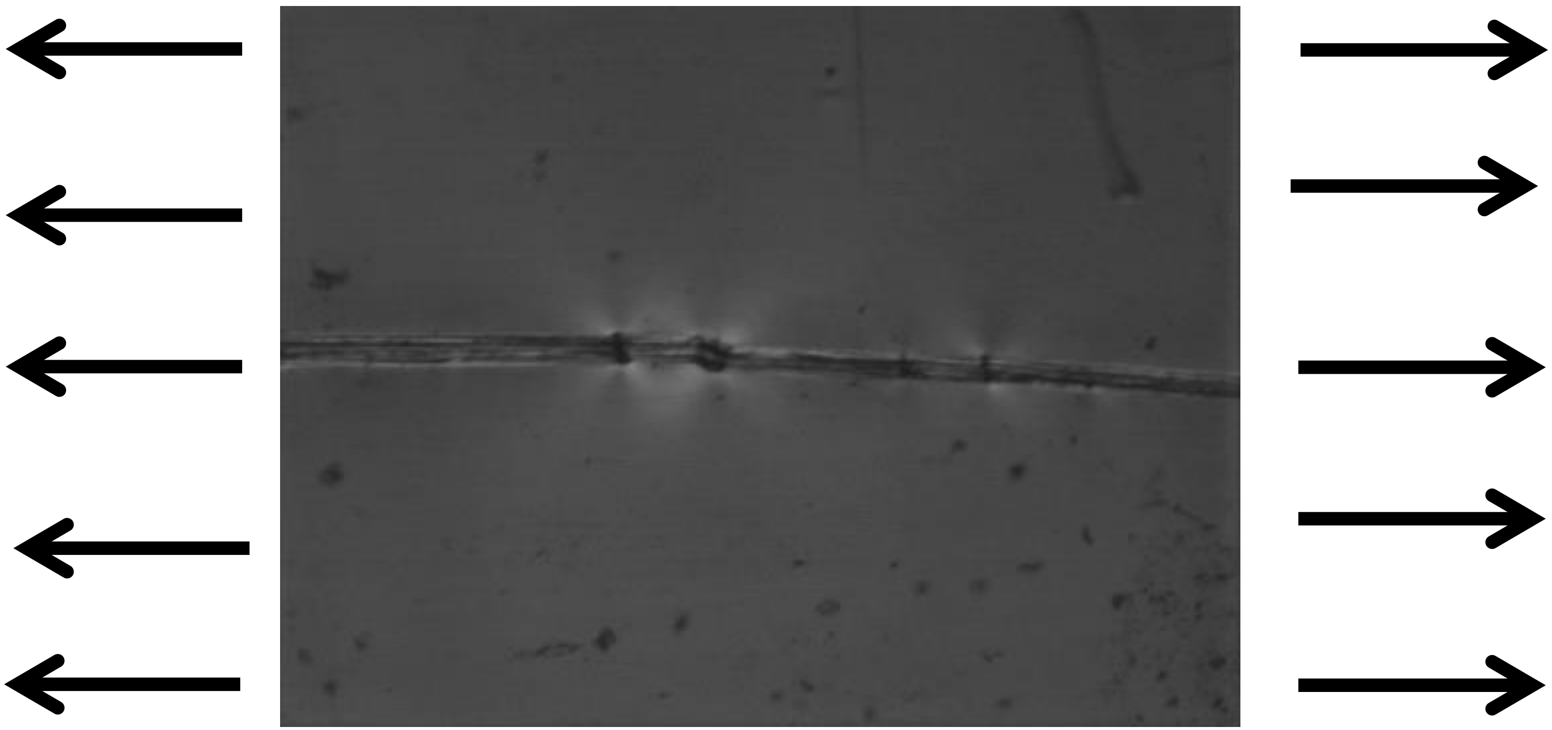
FIGURE 4. Static modulus and damage for flax ultimates.

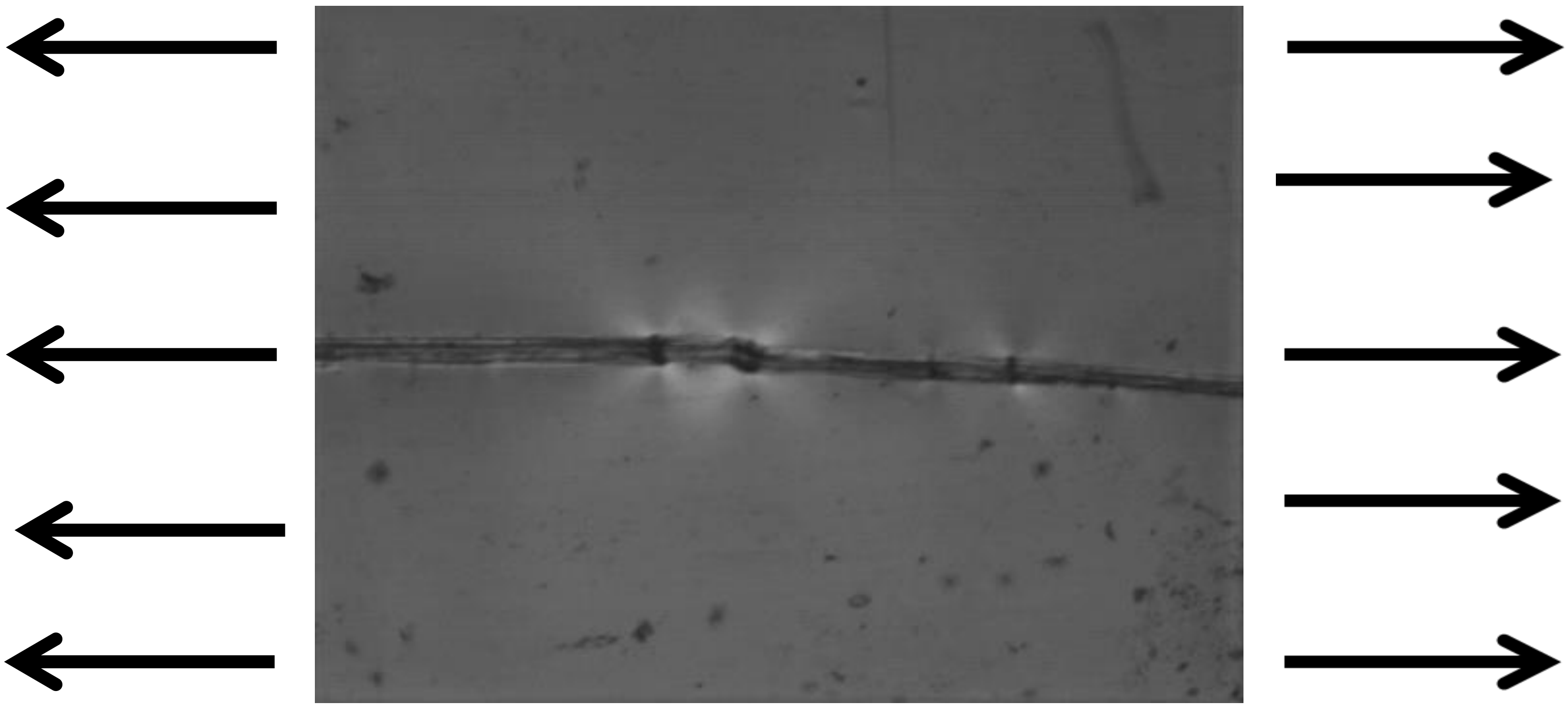
(Davies & Bruce, 1998)

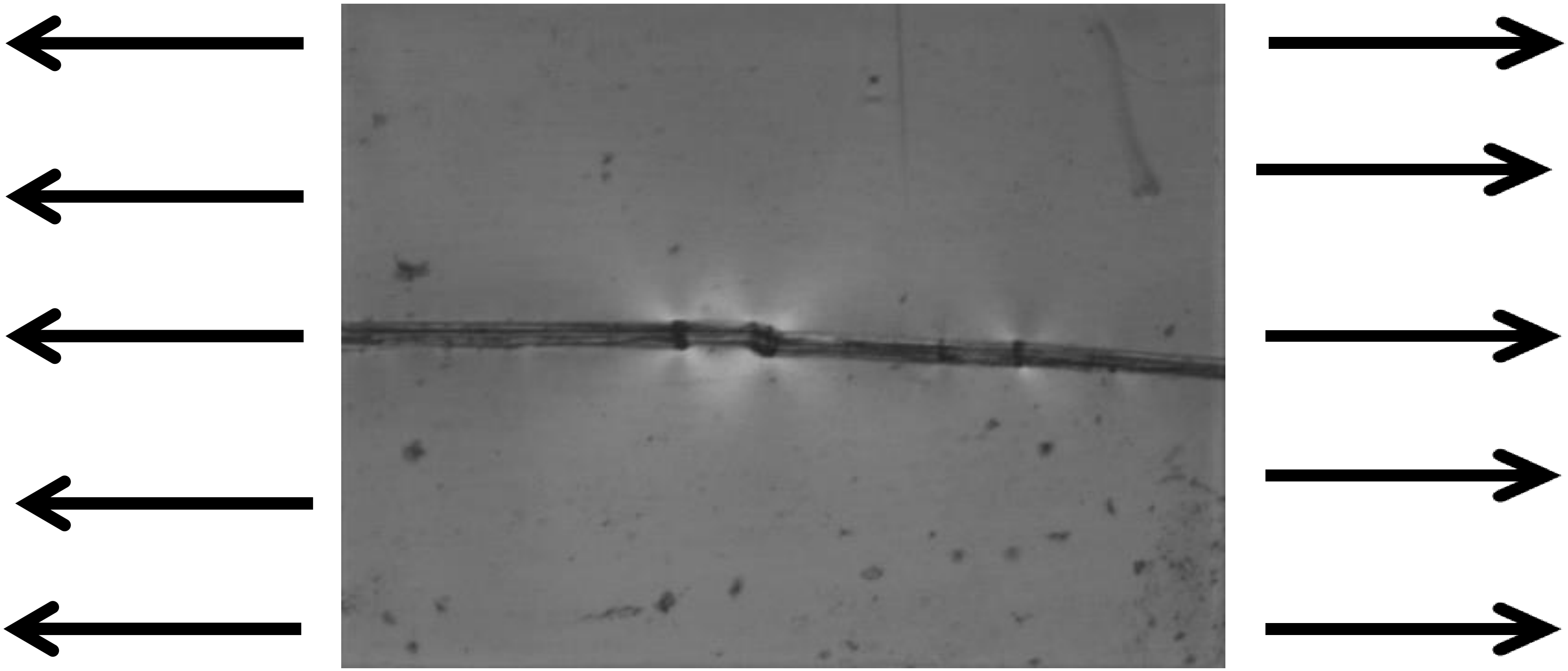


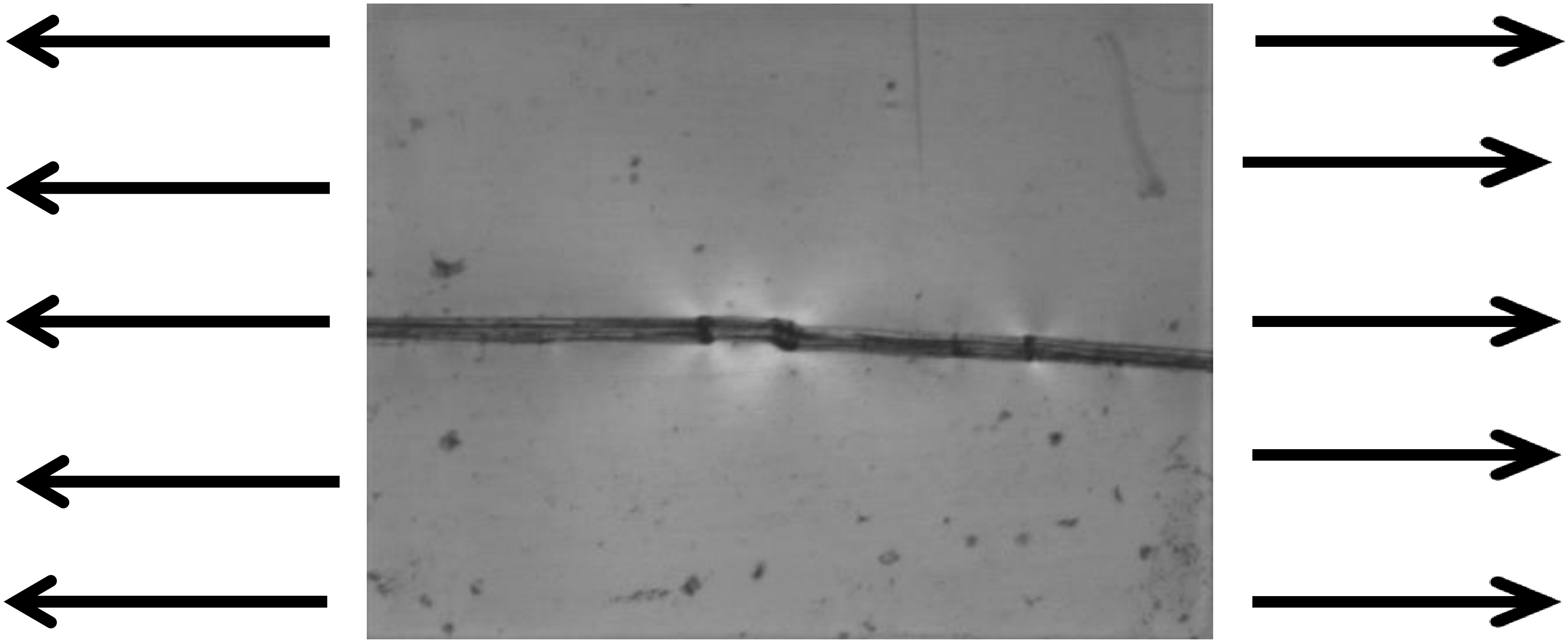






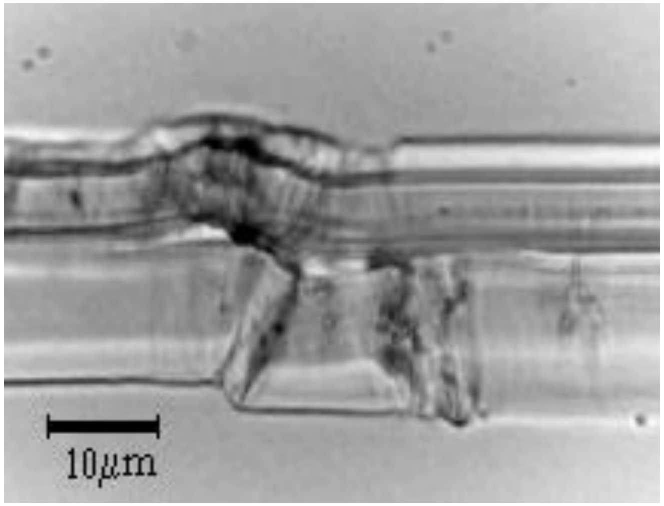


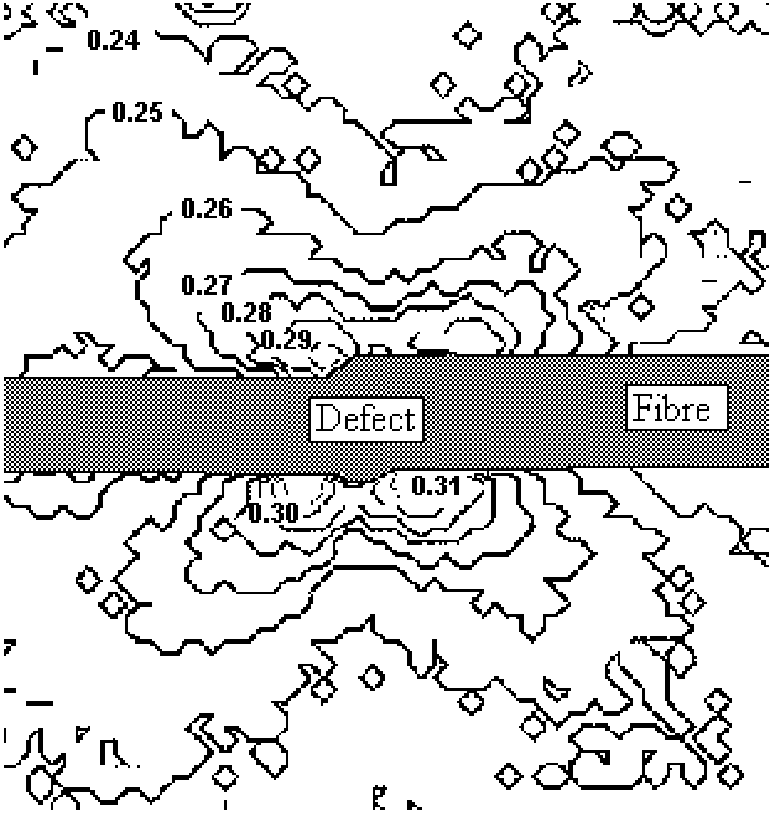




Effect of fibre damage (dislocations) in hemp fibre-epoxy composites

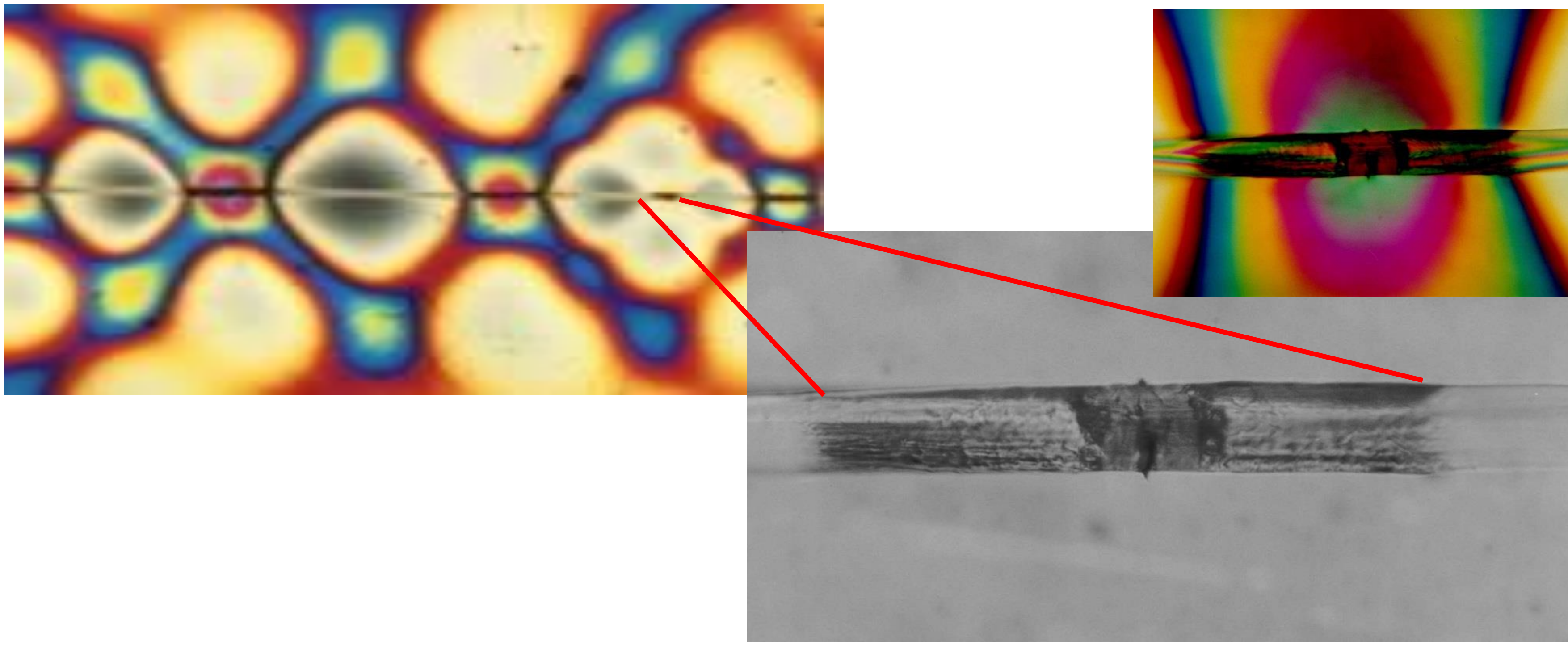
Shear stress distribution in an epoxy matrix adjacent to a defect in a strained specimen at small deformation





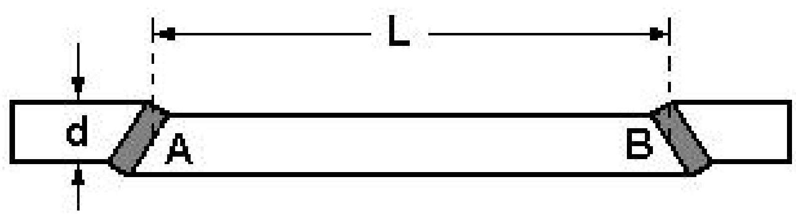
(Hughes et al. 2000)

Matrix shear stress post fracture



Polarised light micrograph of a failed single filament composites showing fibre-matrix de-bonding in regions of high shear-stress concentration adjacent to fibre defects (and fracture)

Model for a fibre containing dislocations



(Hughes et al. 2007)

- Continuous fibre acts as a series of shorter fibres or segments
- Dislocations act as the loci of microstructural failure, resulting in
  - fibre fracture
  - fibre-matrix de-bonding
  - matrix cracking

Matrix shear stress distribution

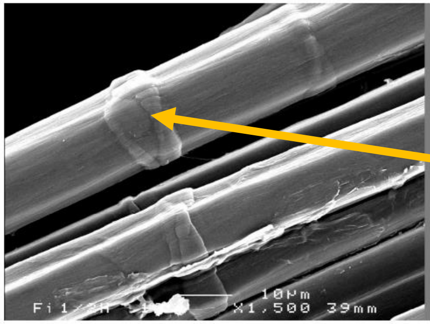
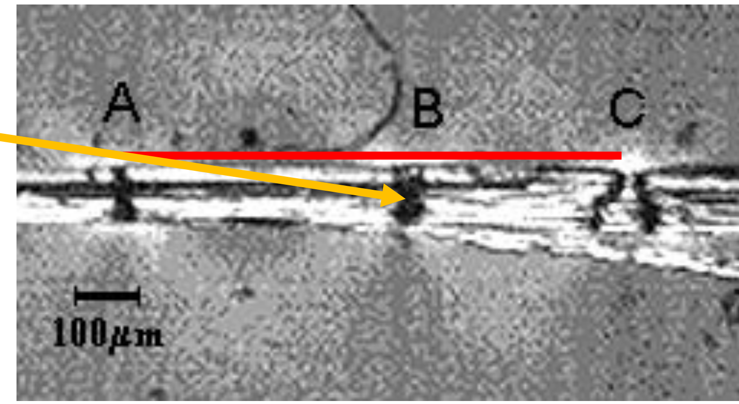
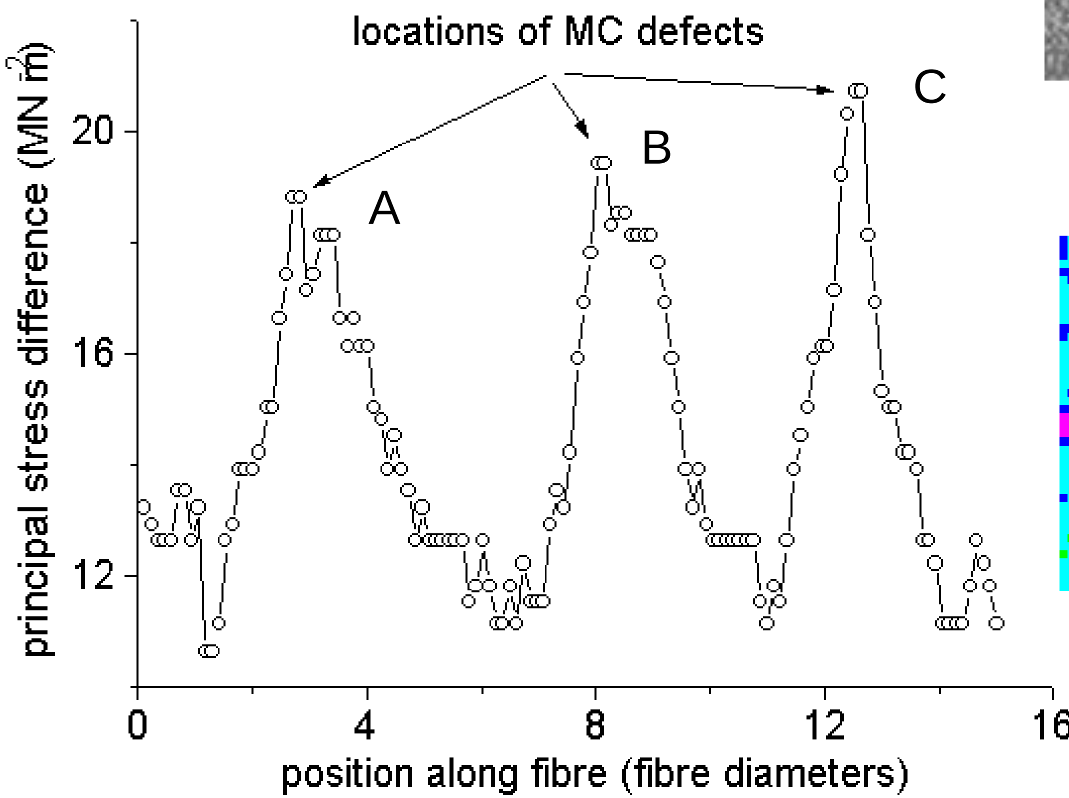


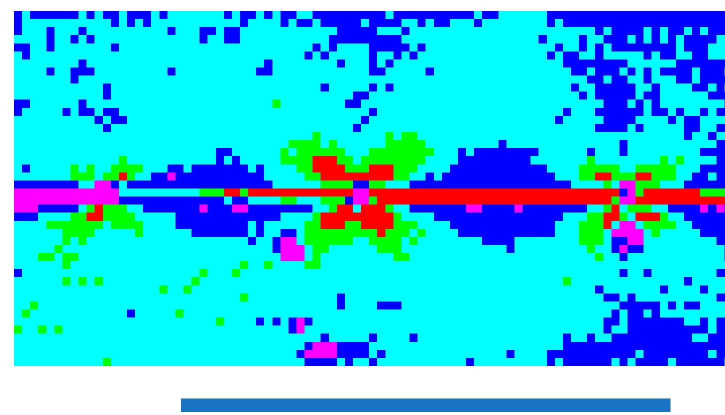
Figure 1 Flax fibers: Example of a bundle of flax fibers with kink bands in the same area.

Fibre defects



Single flax fibre composite





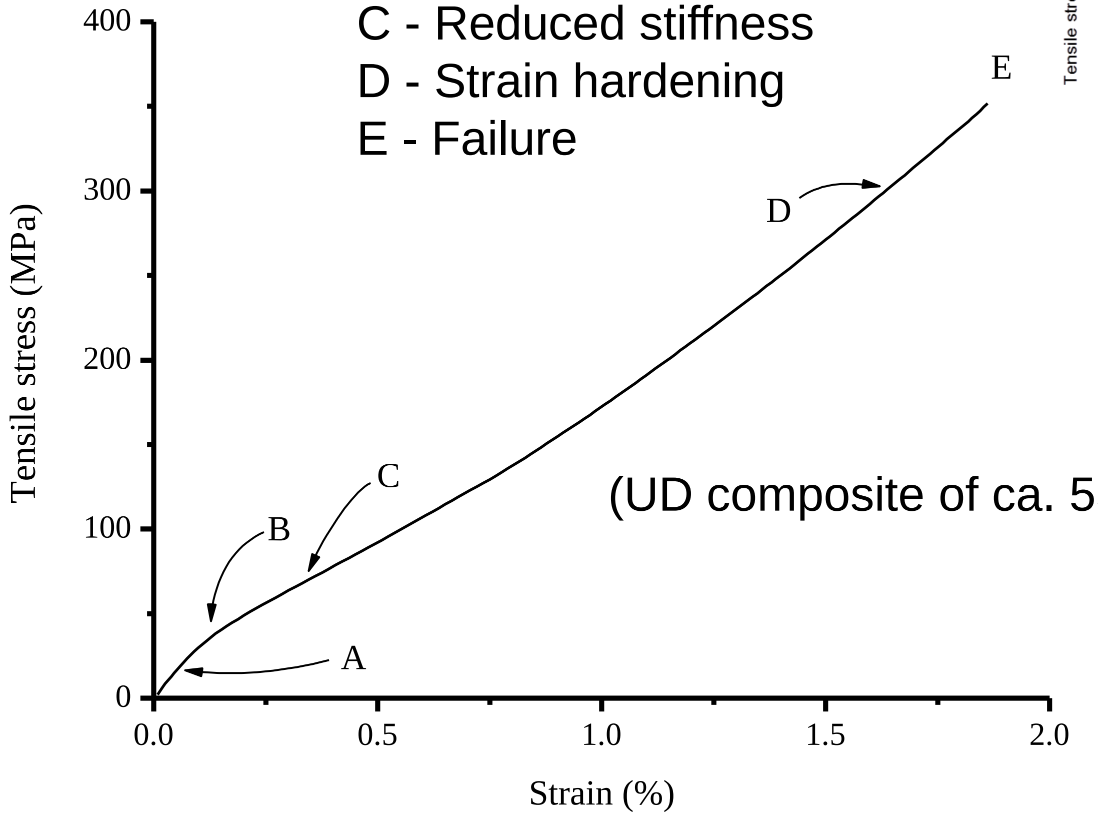
(Eichhorn et al 2001)

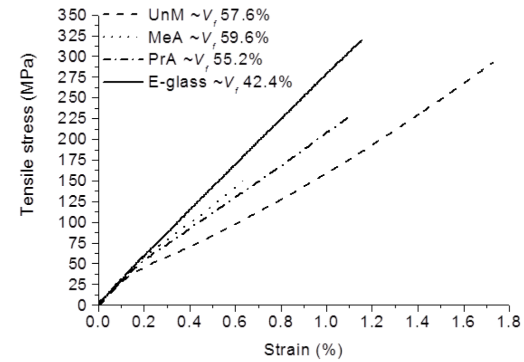
How are composite properties affected?

- Unidirectional composites manufactured from flax fibre in an epoxy matrix
- Fibres modified to improve fibre-matrix adhesion
- Various fibre volume fractions
- Tensile properties investigated

Deformation behaviour

A - Initial linear region
B - Yield point
C - Reduced stiffness
D - Strain hardening
E - Failure





(UD composite of ca. 55% volume fraction)

(Hughes et al. 2007)

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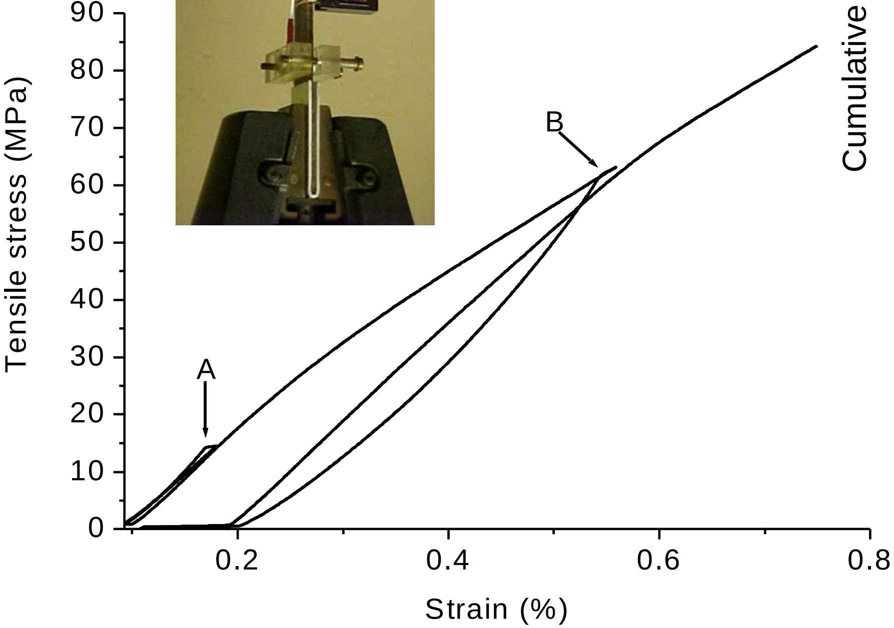
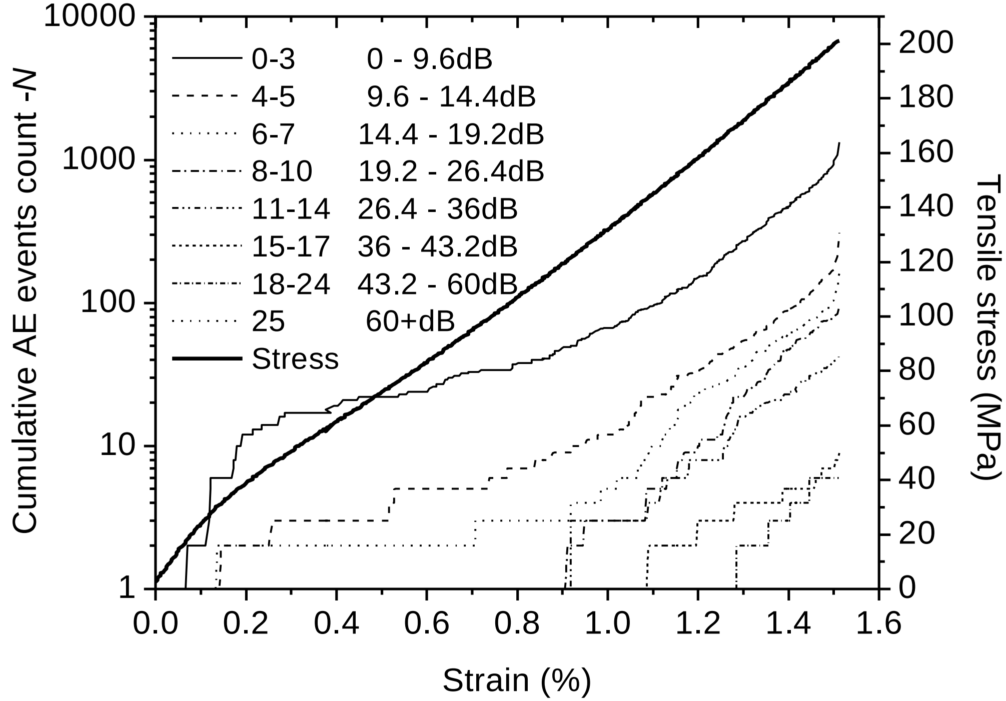
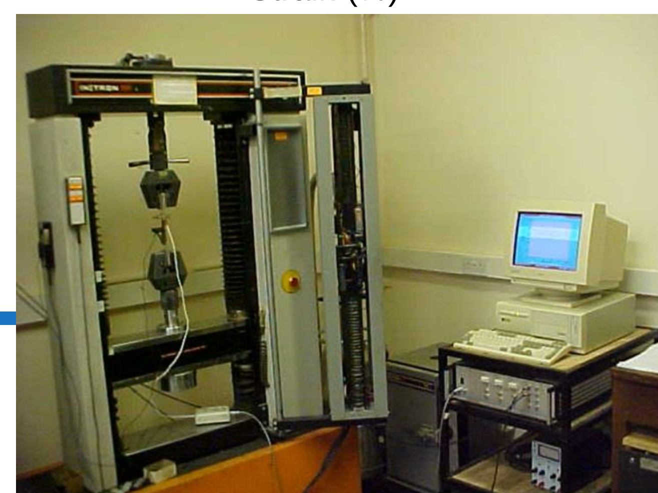
Irreversible (plastic) deformation

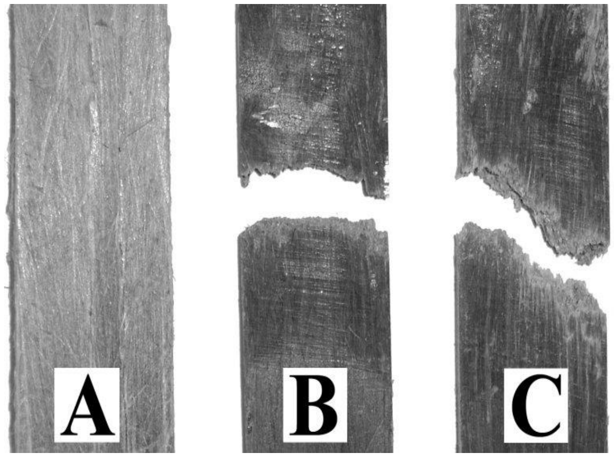


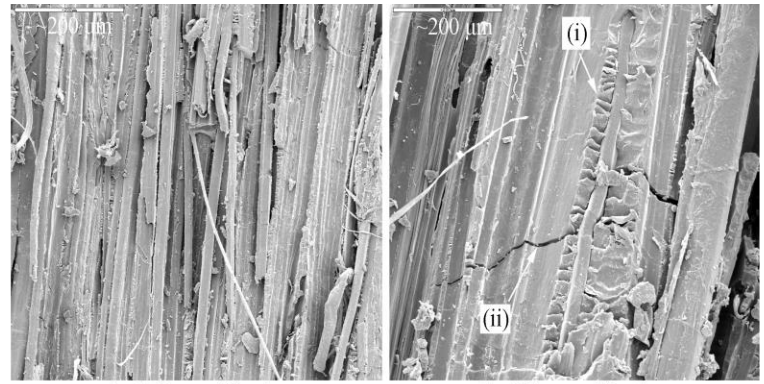


(Hughes et al. 2007)

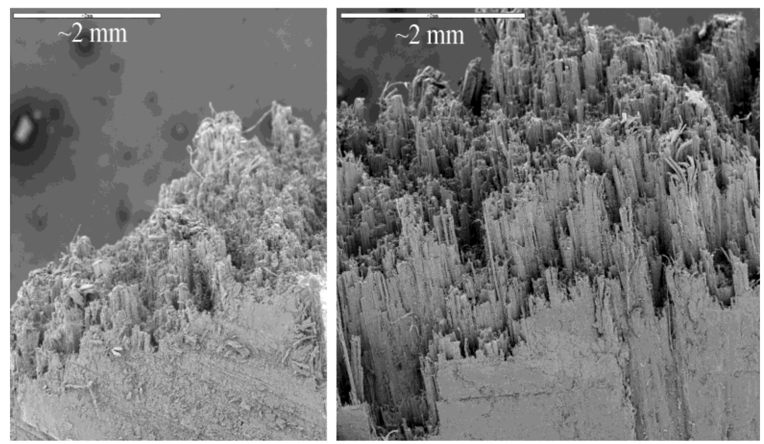


Fracture behaviour





A x170 magnification   B x500 magnification



A x20 magnification   B x25 magnification

Increased adhesion reduces interfacial de-bonding and results in a change in the fracture behaviour: ductile to brittle

(Hughes et al. 2007)

Effect of changing interfacial properties

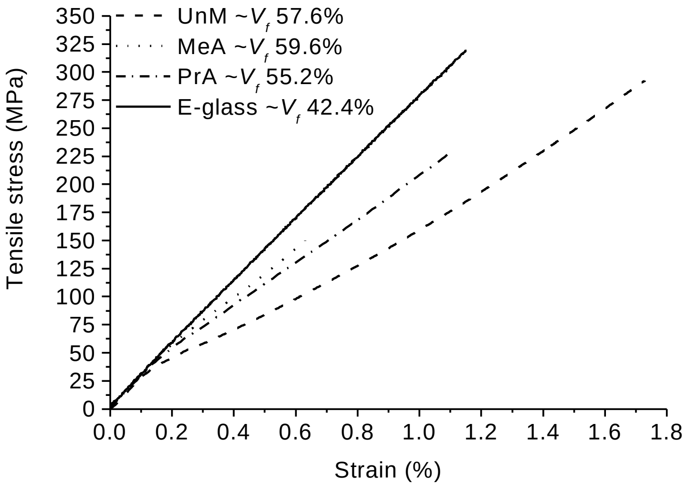


Table VV: Analysis of the influence of fibre-matrix adhesion upon yielding behaviour

Reinforcement type	Modulus			Yield point	
	Young's Modulus (GN m^{-2})	Tangent modulus (GN m^{-2})	Difference (%)	Yield strain (%)	Yield Stress (MN m^{-2})
UnM	28.96 (1.72)	13.82 (0.98)	52	0.12 (0.01)	35.89 (3.55)
PrA	27.41 (2.26)	18.56 (3.23)	32	0.18 (0.02)	51.32 (6.61)
MeA	26.69 (2.52)	18.10 (3.00)	32	0.18 (0.06)	50.47 (13.73)

(Hughes et al. 2007)

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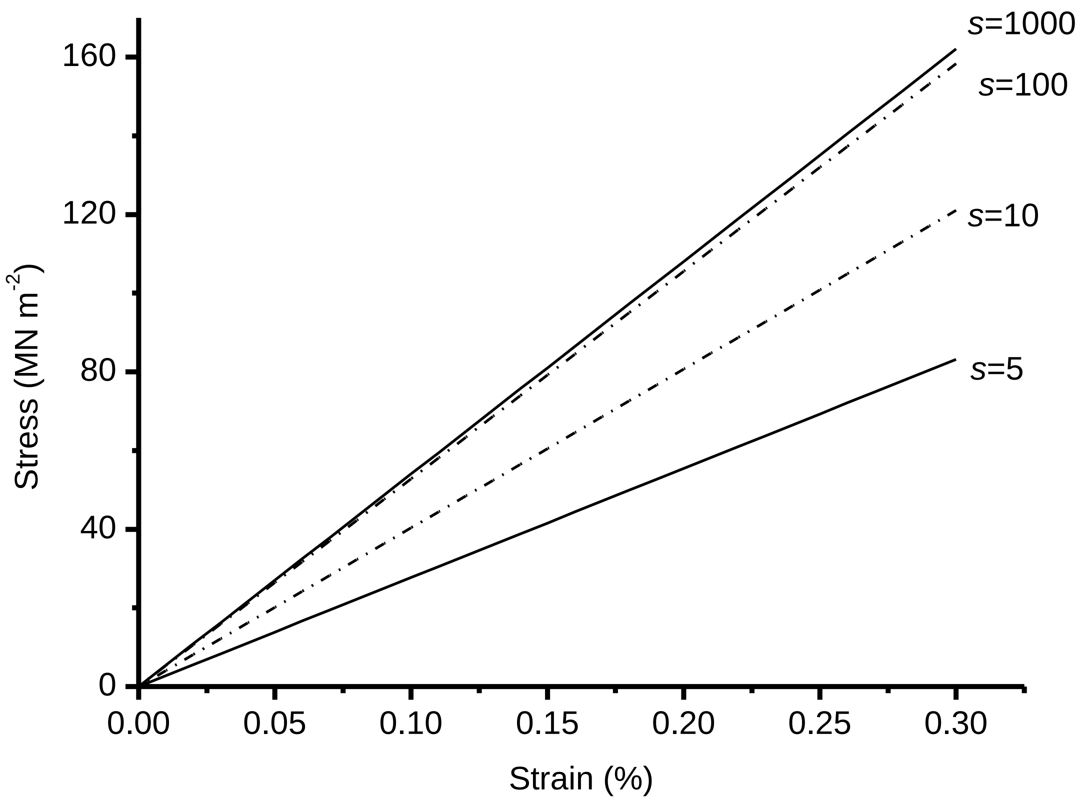
Effective fibre aspect ratio



(Hughes et al. 2007)

Other considerations about reinforcement

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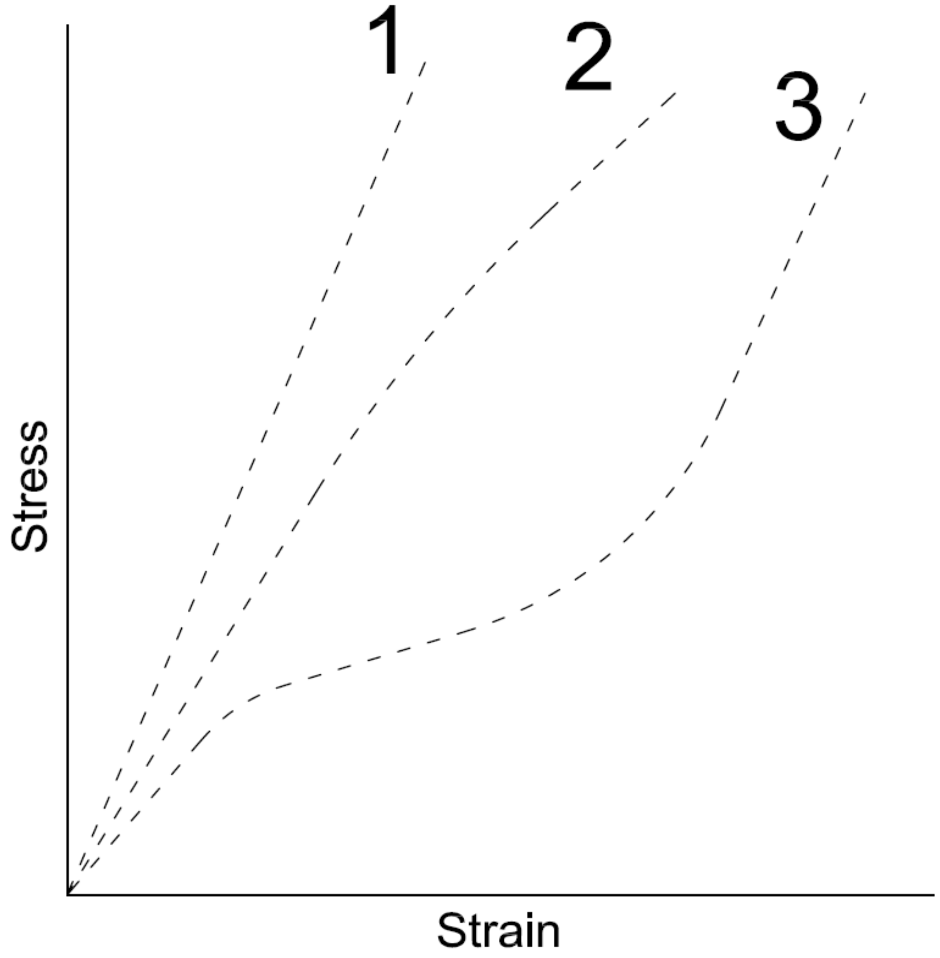
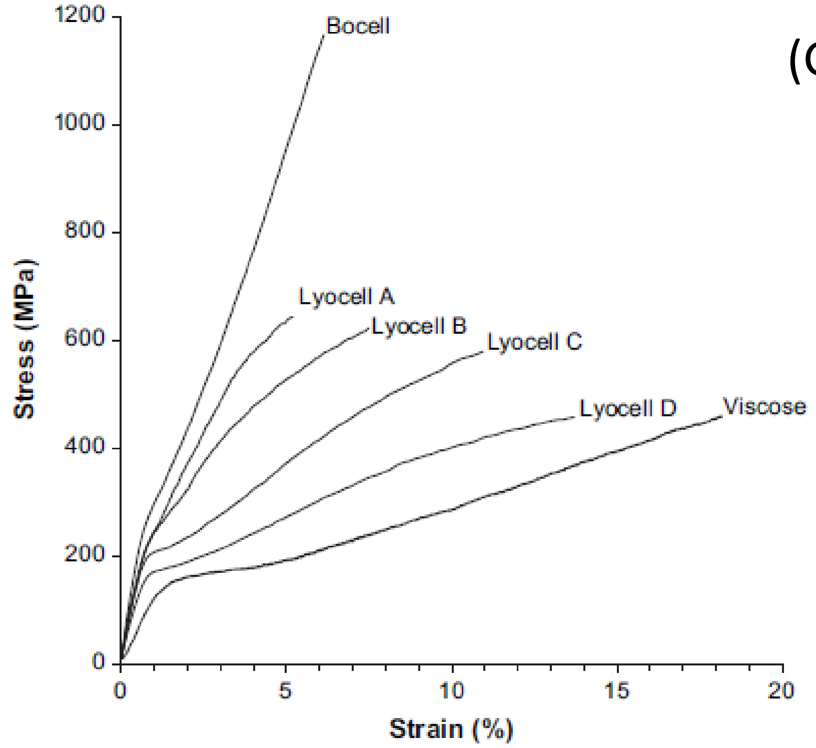
Regenerated cellulose fibre-based composites

- Continuous fibres
- Regular cross-section
- Significantly less variability than natural fibres
- Defect free?
- Potential for modification
- Various manufacturing options: conventional matrices (epoxy, unsaturated polyesters etc.) or single polymer composites ("all-cellulose composites" - ACC)

Tensile behaviour of hemp fibre



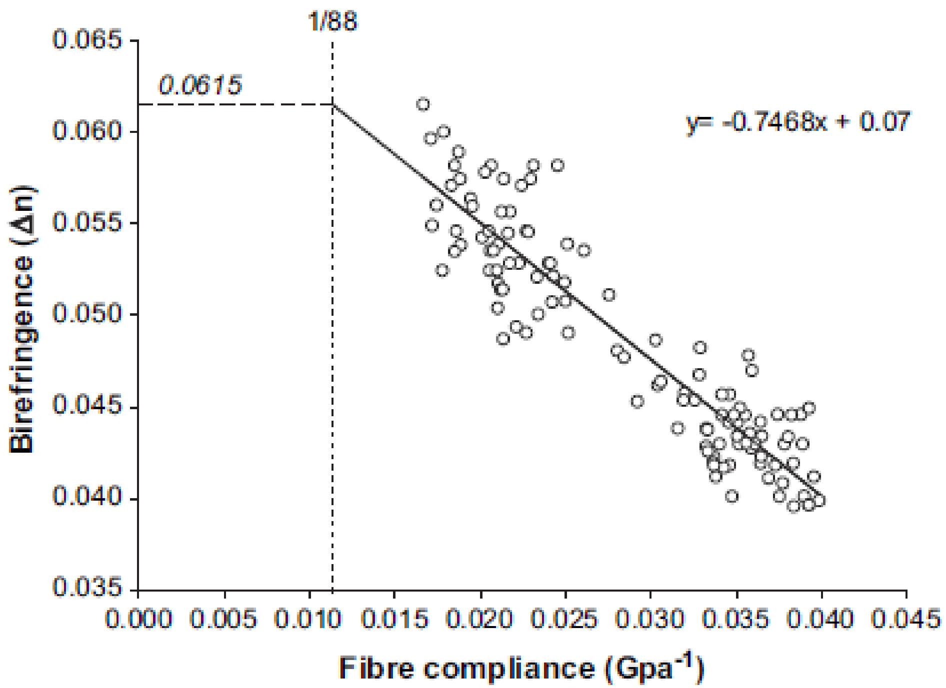
Stress-strain behaviour of regenerated cellulose fibre



(Gindl et al 2007)

- High strain to failure can provide good ductility
- Pronounced yield point

Compliance vs orientation



Greater orientation leads to stiffer fibre – good for composite reinforcement

(Gindl et al 2007)

Natural vs Lyocell fibre

- Epoxy matrix composites

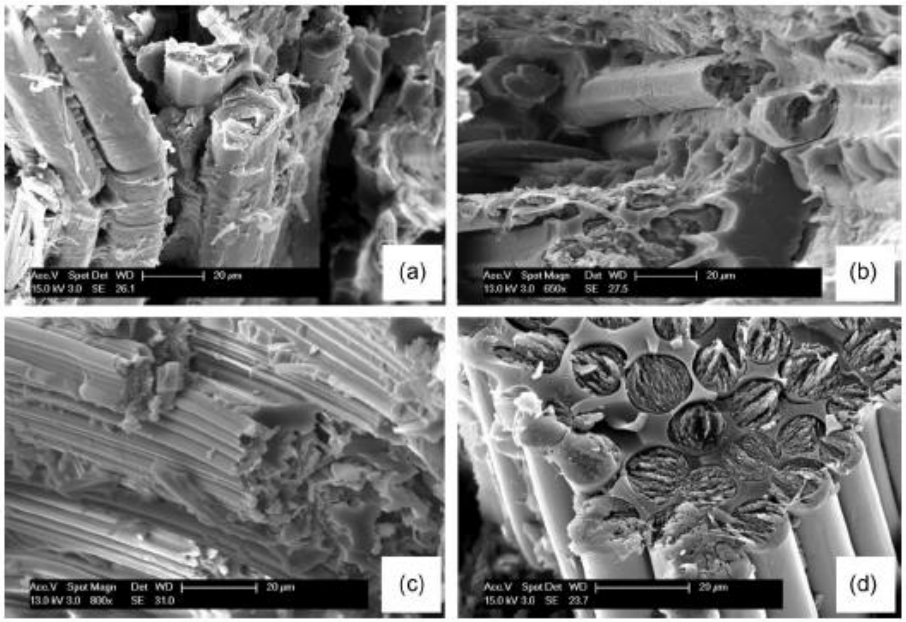
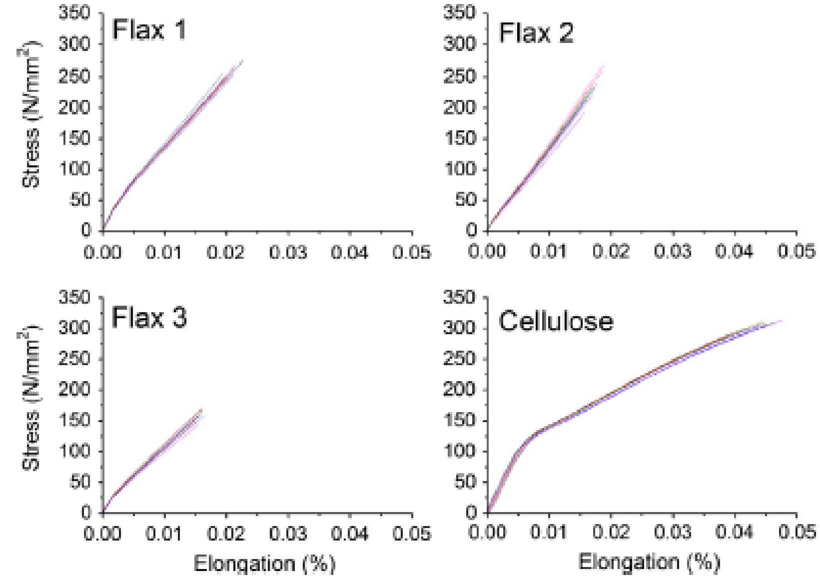


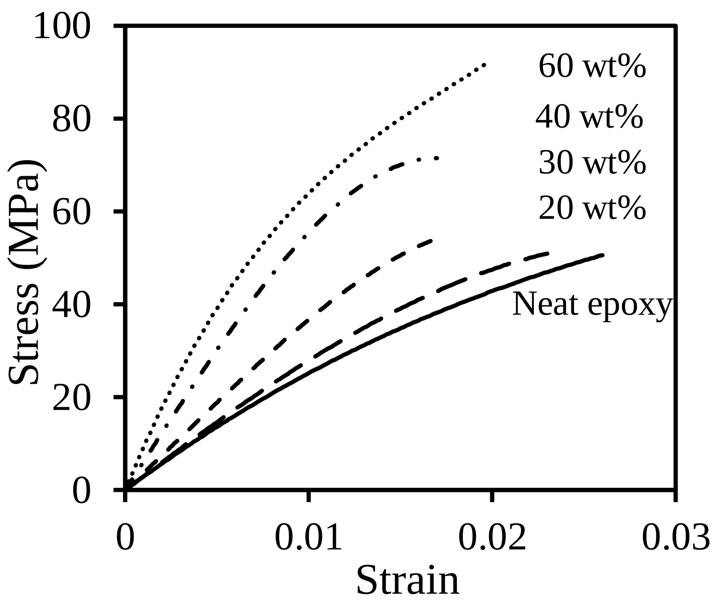
Fig. 4. Longitudinal tension fracture surfaces of (a) Flax1, (b) Flax2, (c) Flax3 and (d) Cellulose reinforced specimens.

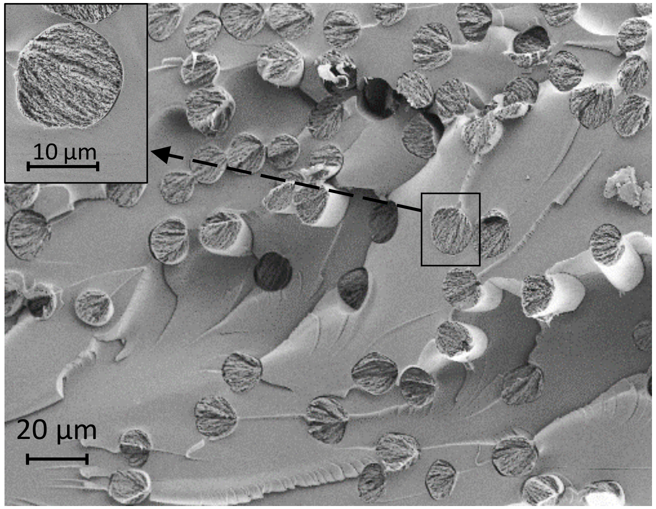
(Santamala et al (2016). Composites Part A-Applied Science and Manufacturing. 84: 377-385)



	Epoxy [31]	Flax1	Flax2	Flax3	Cellulose	E-Glass [42]
Longitudinal						
Fibre volume (%)		34.23	31.18	32.66	54.08	45
Modulus (GPa)	3.5	19.5 ±3.67%	16.3 ±6.89%	14.9 ±6.22%	20.5 ±7.92%	38.6
Strength (MPa)	73	254 ±5.76%	230 ±8.74%	157 ±4.93%	302 ±3.23%	1062
Elongation (%)	3.5	2.05 ±5.89%	1.71 ±6.21%	1.57 ±3.07%	4.32 ±6.66%	2.75
Transverse						
Fibre volume (%)		36.17	32.91	35.63	55.01	45
Modulus (GPa)	3.5	4.93 ±4.30%	2.81 ±8.28%	3.86 ±8.73%	4.51 ±3.34%	8.27
Strength (MPa)	73	32 ±6.30%	11 ±13.56%	17 ±10.40%	17 ±3.23%	31
Elongation (%)	3.5	0.75 ±7.40%	0.45 ±16.04%	0.53 ±13.59%	0.39 ±7.33%	0.38

Ioncell-epoxy composites



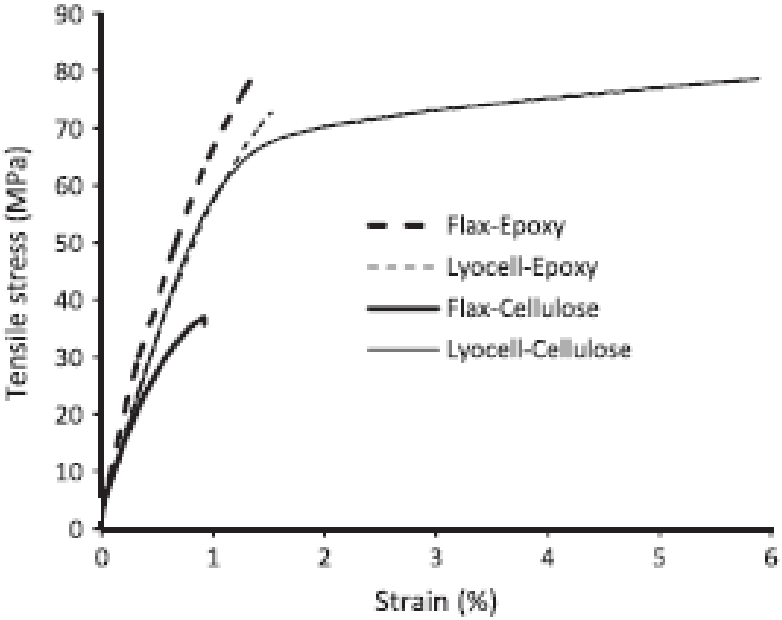


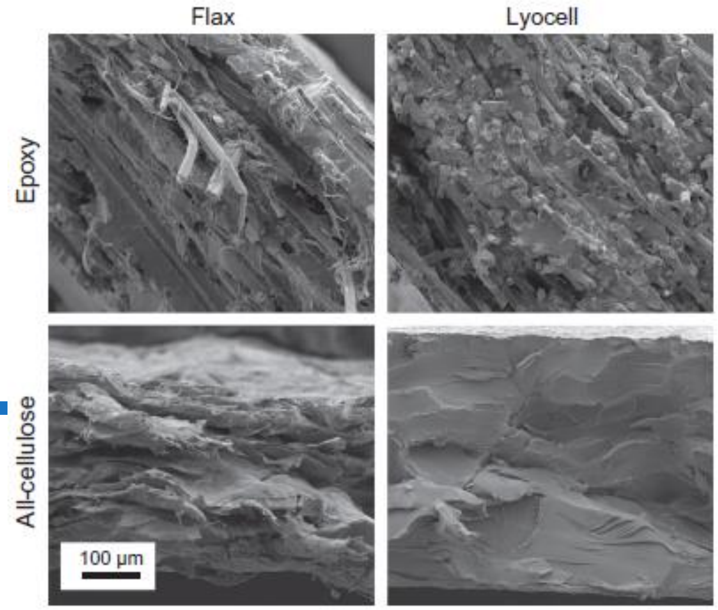
Fibre loading (wt%)	Young's modulus (GPa)	Tensile strength (MPa)	Yielding strength (MPa)	Yielding strain (%)	Strain at break (%)
0	2.2 ± 0.4	50.2 ± 6.5	13.3 ± 4.9	0.66 ± 0.4	4.1 ± 0.5
20	3.0 ± 0.5	58.3 ± 12	22.4 ± 3.4	0.86 ± 0.25	2.8 ± 1.2
30	4.5 ± 0.5	72.5 ± 7.7	36.7 ±16.1	0.88 ± 0.36	2.1 ± 0.2
40	4.9 ± 1.2	62.7 ± 16.5	39.5 ± 1.6	0.7 ± 0.17	1.6 ± 0.4
60	7.2 ± 0.8	102.8 ± 28	30.3 ± 11	0.49 ± 0.14	4.0 ± 1.8

(Unpublished data)

Traditional matrix vs ACC

- ACCs can be prepared by ‘selective dissolution’ of fibre or by mixing of reinforcement in dissolved cellulose, followed by regeneration
- Various cellulose solvents have been investigated
- Unidirectional fibre reinforced composites prepared from Lyocell or flax fibre with either epoxy or ‘cellulose’ matrix
- Clearly very different microstructures created when selective dissolution employed
- Failure of epoxy-matrix composites dominate by matrix properties





(Gindl-Altmutter et al (2012). Compos. Sci. Technol. 72(11): 1304-1309)

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ACCs via solvent infusion processing (SIP)

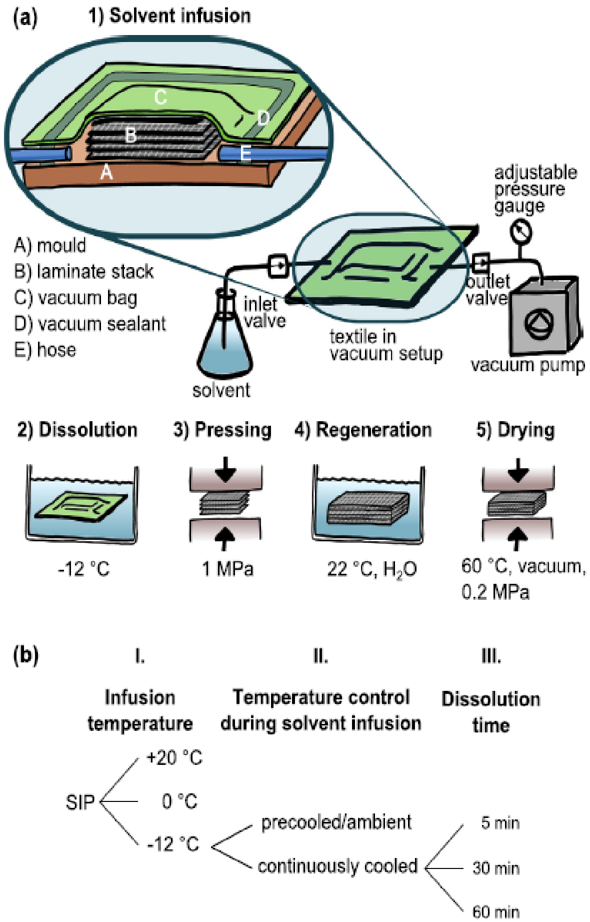
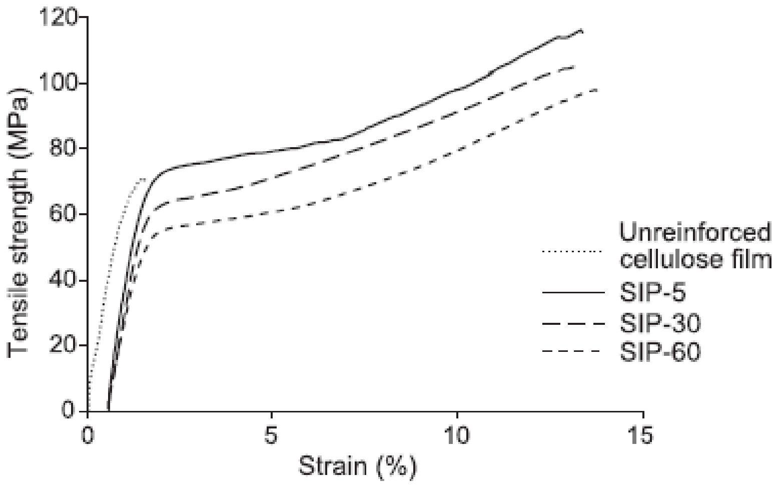


Fig. 1. (a) Schematic of solvent infusion processing (SIP) using aqueous NaOH/urea solution as solvent. (b) Schematic of the three stages of optimisation of the processing parameters for SIP using NaOH/urea.

- Woven Cordenka® fibre textile
- Solvent: NaOH/urea
- Dissolution time/temperature varied



(Dormanns et al (2016) Composites Part A-Applied Science and Manufacturing 82: 130-140)

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