Structure properties of wood fibre reinforced PLA composites

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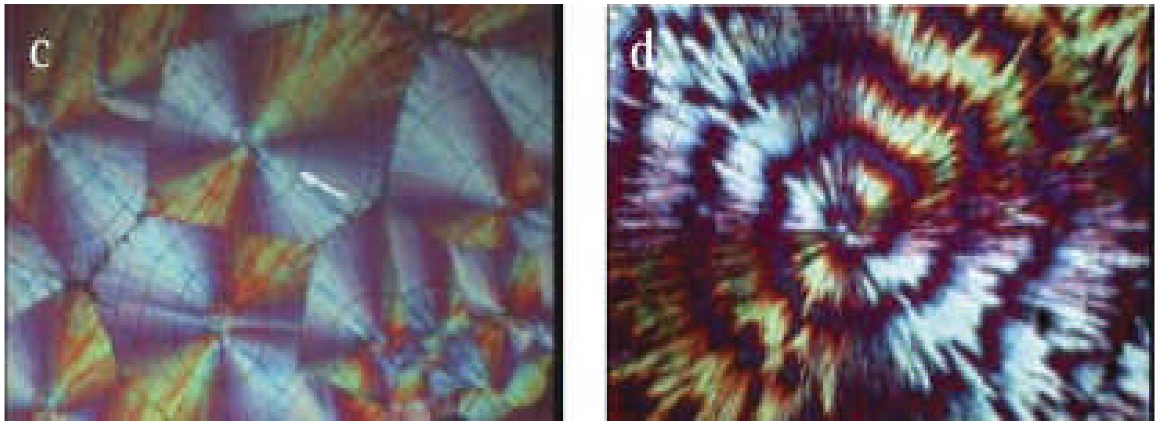
Overview

- PLA properties and processing
- PLA modification
- Fibre-filled compounds
- MFC reinforced PLA
- PLA wood fibre composites
- Use of chain extender
- Summary

Objective

Develop advanced light-weight, biocomposite based demonstrators and technological solutions with industrially desired and controlled properties combined with ecodesign features.



Spherulites in slowly cooled PLA
J. Xu et. al., Polymer 46 9176 (2005)

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PLA

PLA versatile, biodegradable polymer produced as different resin grades with a wide range of properties.

Typical datasheet values:

Module:	3.5 GPa
Tensile strength:	45-60 MPa
Density:	1.25
Tm:	150-160 °C
Tg:	55-60 °C
MFR:	5-30
Processing temp.:	190-230 °C

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PLA-related concerns

- PLA brittle
- Prone to hydrogenation
- Biodegradability: avoid abusive temperature and humidity conditions

Processing:

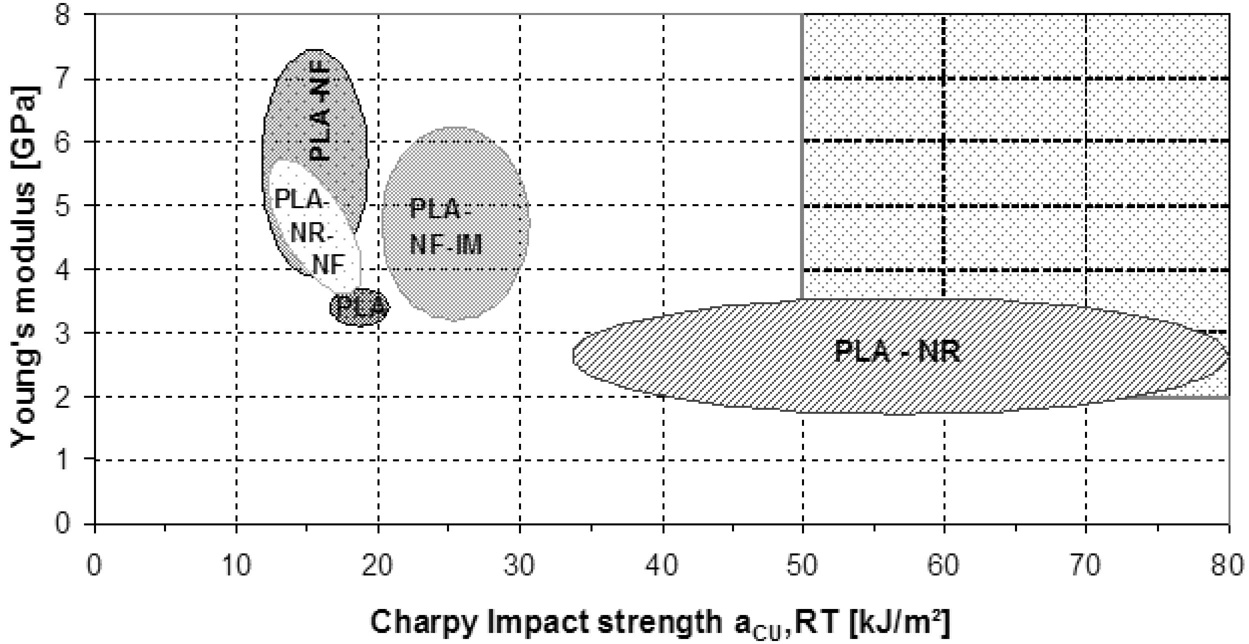
Drying : max 250 ppm humidity (2 hours at 90 °C)

Flushing procedures at start-up and close-down

PLA modifications

Toughness increased using 5-20 % impact modifiers.

- Epoxidized natural rubber
- Grafted rubber resin blends
- Aliphatic polyesters



R. Forstner et. al. ECCM13 (2008):

PLA-flax-natural rubber

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PLA composites

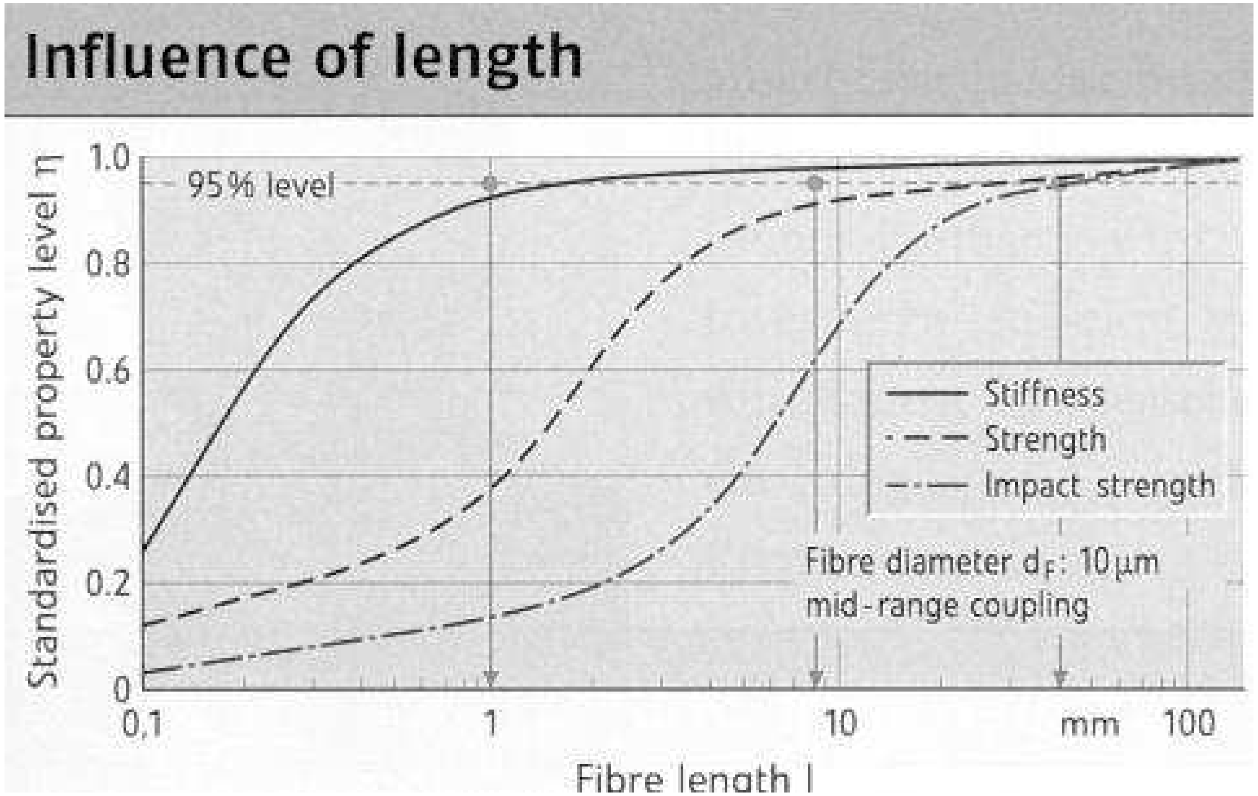
Bio-based fillers =>Improved properties and reduced costs!
PLA-flax composites 50% better tensile strength than PP-flax composited (automotive panels) [K.Oksman et al., Comp. Sci. Tech. 63, 1317 (2003)]

<u>Natural fibres</u>
Wood pulp, woof flour, flax, abaca (Manila hemp), ...
<u>Nano-fibres</u>
Micro fibrillar cellulose (MFC), microcrystalline cellulose (MCC), montmorillonite, carbon nanotubes, layered titanate, ...
w/wo surface treatment

Filler dispersion and interfacial interaction with PLA !
Good surface interaction PLA-cellulose fibres reported
(J. Ganster H-P Fink, Cellulose 13, 271 (2006))
Other authors disagree (!)

Fibre composite structure-properties

Fibre structure – length, diameter, surface, concentration, dispersion, fibre orientation.



M. Schemme,
Kunststoffe plast Europa,

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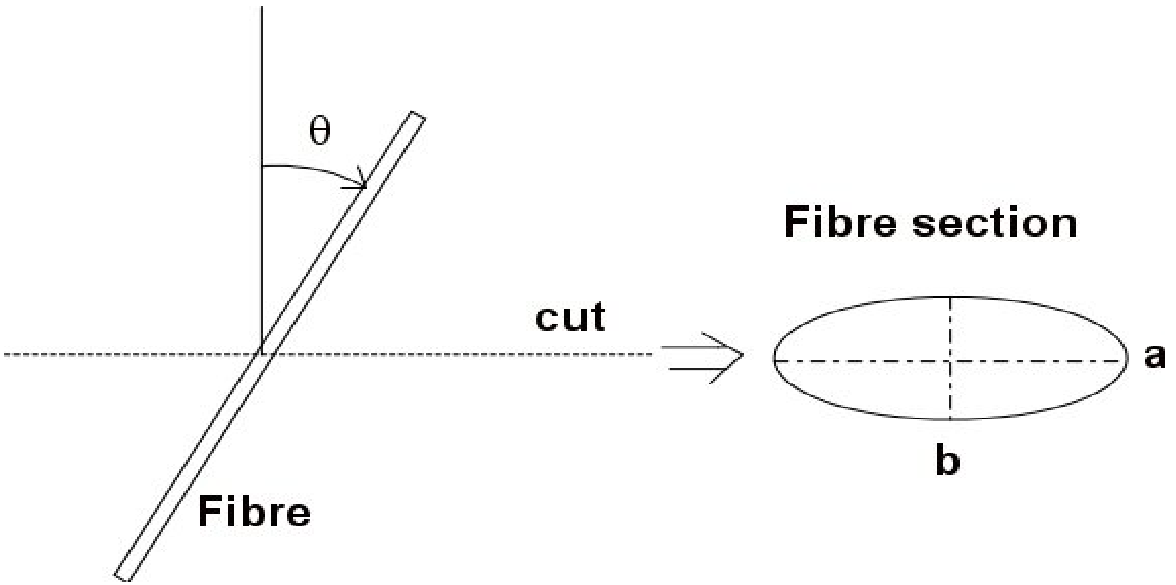
Fibre orientation

Orientation easily visualized for cylindrical glass fibres



Fibre orientation – natural fibres

Irregular natural fibres challenging!

Alternative method: 3D X-ray tomography

Volume: $(1.3\ mm)^3$

M. Faessel et. al, Comp Sci Tech., 65, 1931 (2005)

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MFC filled PLA

MFC as aggregates difficult to separate into nanofibrils during compounding!

➢Acetylated MFC
Synthesizing bionanocomposites: Disolving PLA in $CHCl_3$ and added MFC suspension in $CHCl_3$.
Modified viscoelastic properties and increased T_g.
(P. Tingault et al, Biomacromol. 11, 454 (2010))

➢MFC-PLA nanocomposites from sheets obtained by a papermaking-like process
A.N. Nakagaito et. al., Comp. Sci. Tech., 69, 1293 (2009)

Extruder compounding - SustainComp

Clextral 25 mm corotating twin-screw extruder

Materials

- PLA – Inego 2002D
- PLA – Inego 3251D
- PLA – Inego 8052D
- PLA – fibres of the same PLA grade as the commingled materials
- Commingled pulp – fluffy state
- Commingled pulp – sheet form (dried and not-dried)
- Commingled pulp – pelletized (from fluffy state)

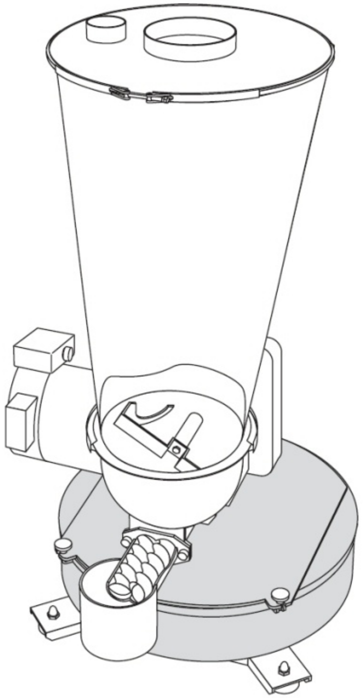
Fibres

- Fluffed sulphite fiber (WF) from Borregaard
- Pelletized WF (as received from PFI)
- MFC (micro fibrillar cellulose) – dry - Borregaard

Fibre feeding

To achieve controlled fibre handling we need to optimize:

- Hopper – cylindrical is preferred
- Agitator - fibre agitator (GF special); sword (fluffy fibres)
- Screw – spiral twin screw, coarse pitch preferred



K-Tron Twin-Screw Loss-in-Weight Feeder - KCL-T20

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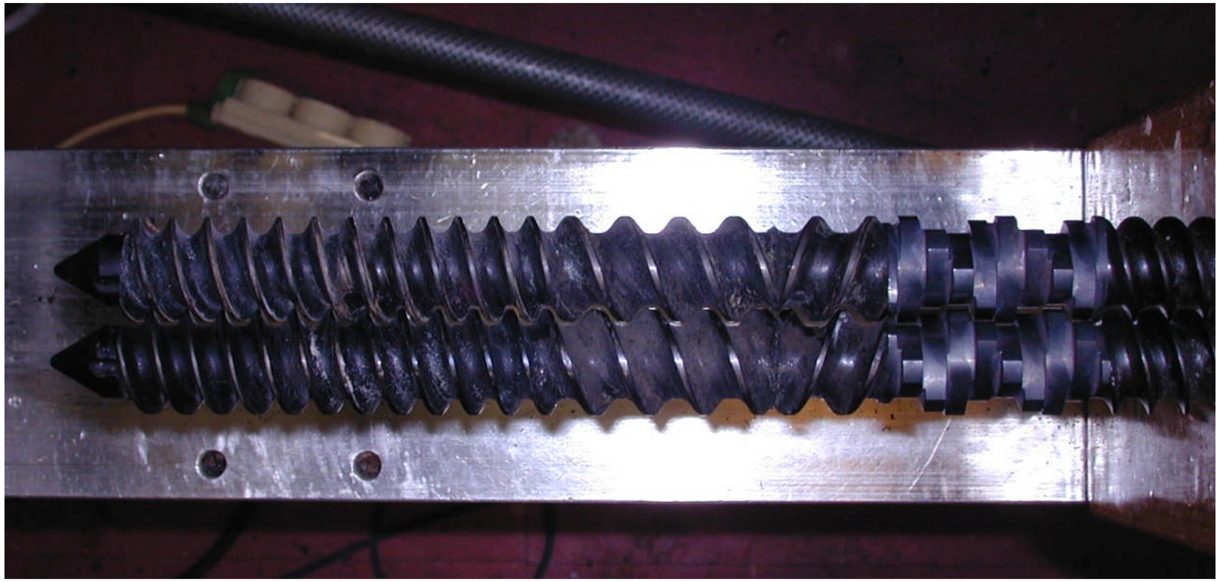
Extrusion

Twin-screw for better dispersion.

BUT: A trade-off with fibre breaking which limits properties!



190 °C, 150 rpm, 30-50 g/min

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Material degradation during processing

Fibre length

Sample	Fibre length (mm)	Stdev (%)
Comming. pellets	0.73	
IM-Comming. Dried Sheet	0.16	± 0.6
IM-Comming pelletized	0.17	± 1.5
Wood fibres	1.62	
Wood fibres pelletized	0.95	
IM-2002D + 10% WF	0.26	± 5.0
IM-2002D + 30% WF	0.33	± 0.0
IM-2002D + 40% WF	0.20	± 0.5

No significant PLA degradation seen !

Fourier transform infra-red (FTIR) spectroscopy

- Commingled material
- Commingled- compounded and injection moulded

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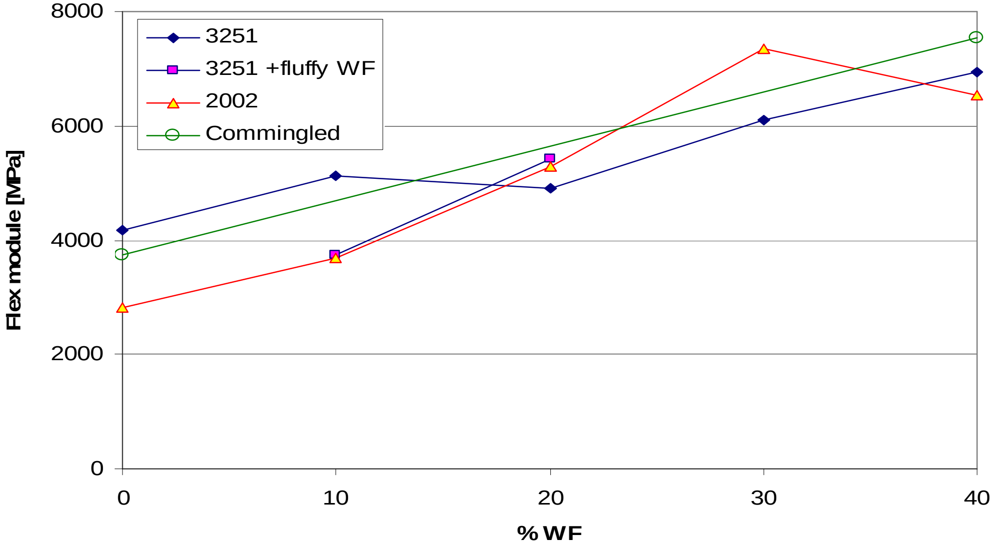
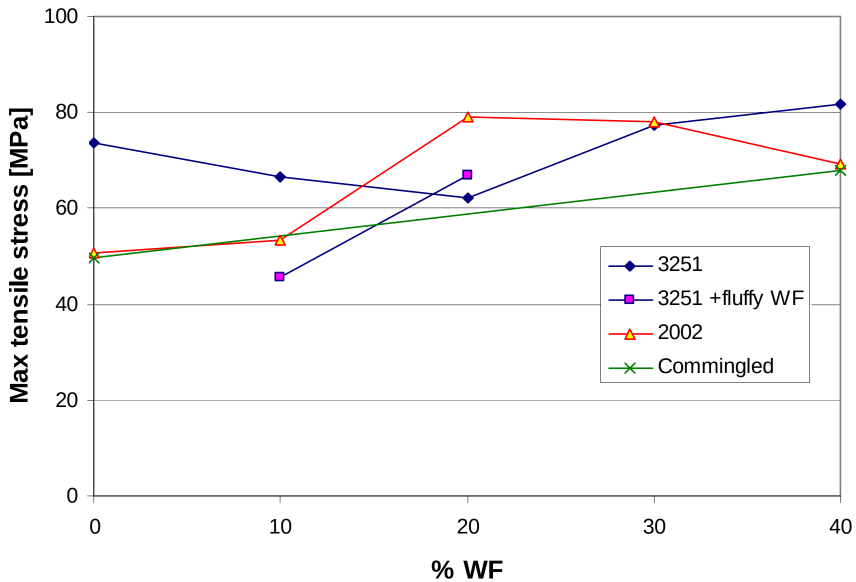
PLA wood-fibre properties





Improved stiffness (as expected)

Increasing trend for strength afo WF content

=> PLA-fibre adhesion

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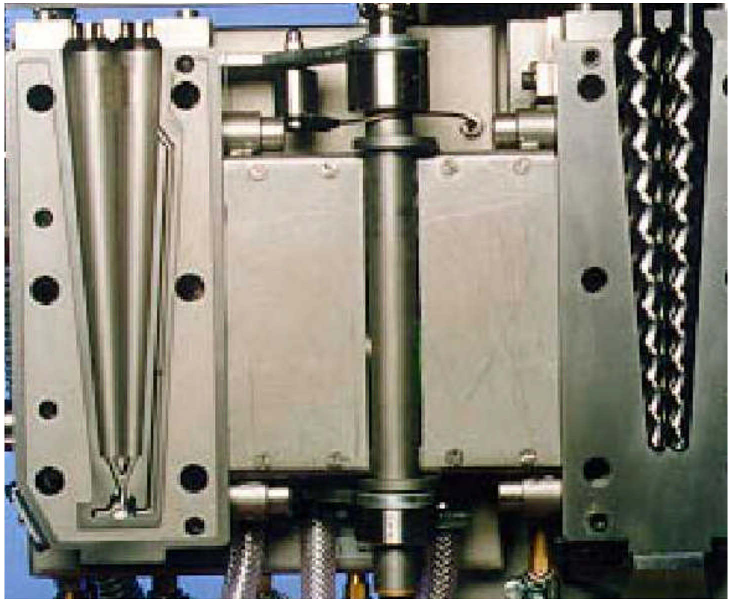
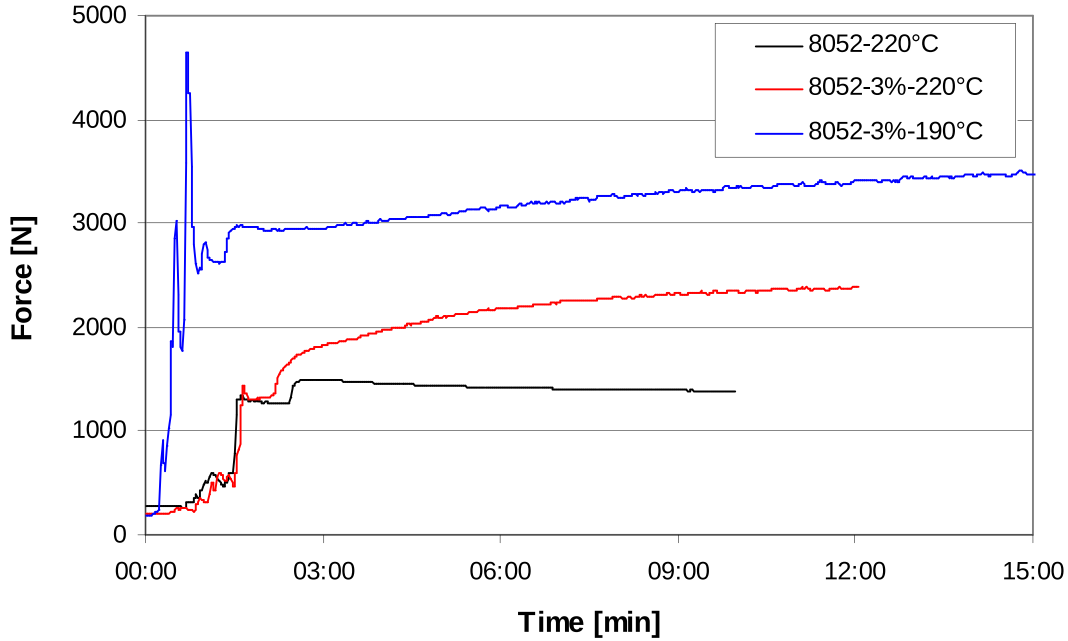
Compounding and chain extender

- PLA composites generally brittle !
- A chain extender improves melt strength and possibly final properties. CESA-extend (Clariant -Joncryl (BASF) epoxy functional acrylic polymer)
- Average residence time in extruder is 3-4 min => CE reaction not complete (But, subsequent injection moulding adds to the residence time)
- CE reaction kinetics will depend on actual thermo-mechanical history including dispersion.
- On-line study under realistic conditions can be performed on a mini batch extruder - DSM Midi2000
- Test with Inego 8052D w/ 0 and 3% CE

Reactive extrusion – on-line kinetics

Batch extruder – Midi2000





➢ **Force reflects CE reaction**

Polymer viscosity reflected by
force excerted at cell below the cavity

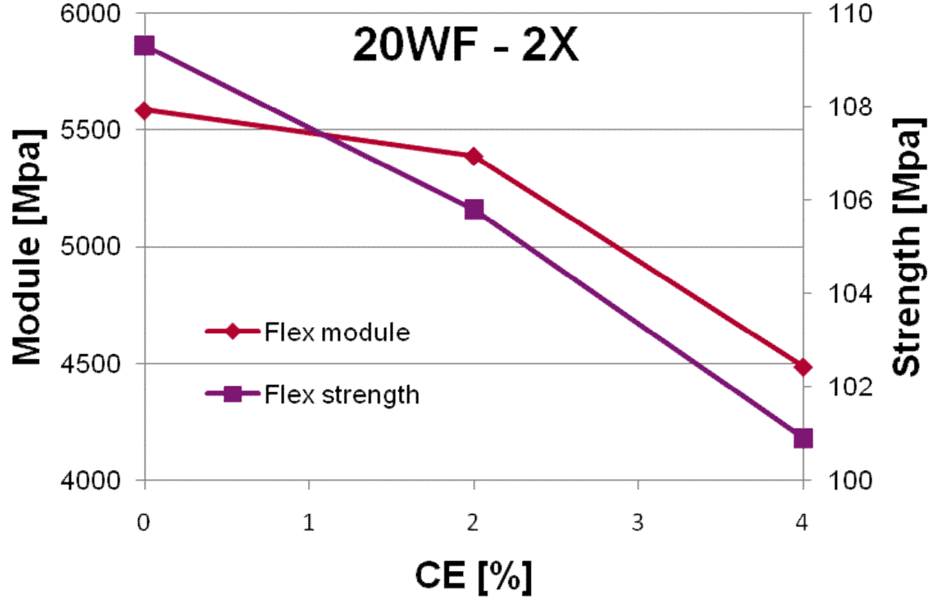
PLA wood-fibres and chain extender

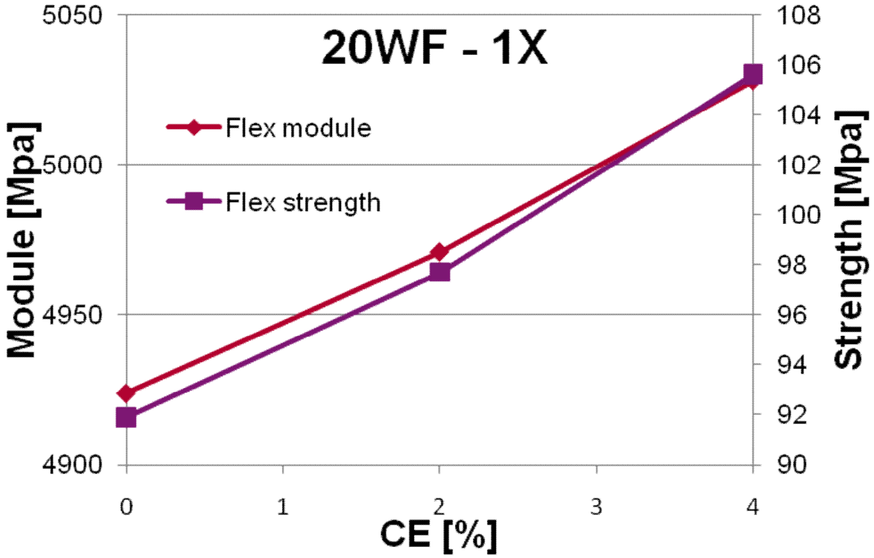
Inego 3251

CESA-Extend

Clextral: 1X and 2X

40 rpm, 190 °C





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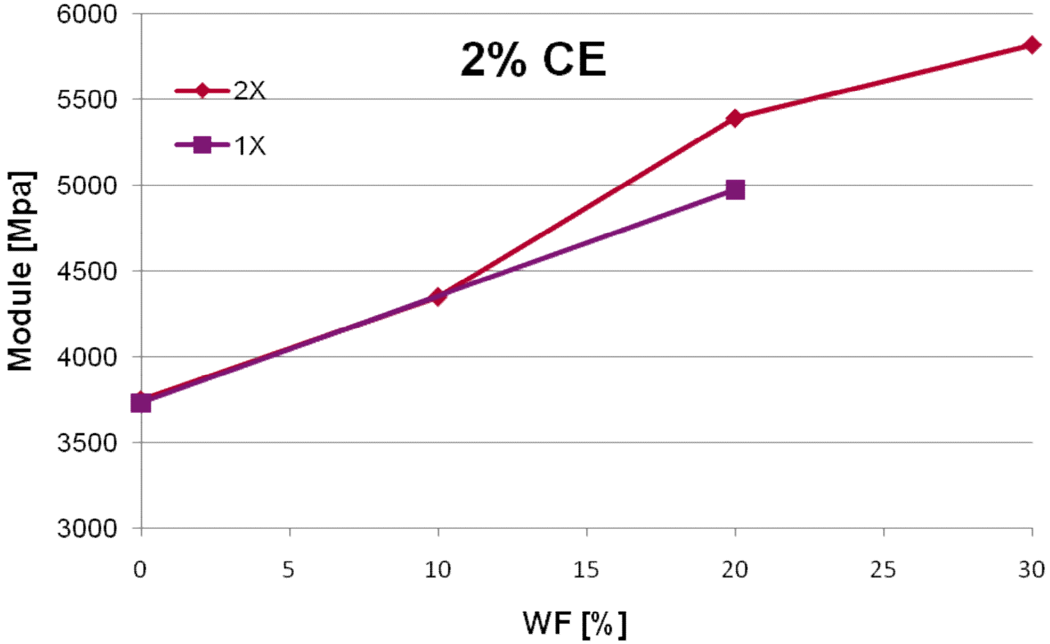
PLA wood-fibre and chain extender



More effect of 2X extrusion than of chain extender

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Summary

- Large possibilities for modifying PLA properties
- MFC promising. Wet-mixing a potential technique
- Compounding commingled material destroys effect on fibre length
- Effects of PLA-WF adhesion observed
- Next: Further material modification