



# EUROPEAN PLACE

17TH BIENNIAL TAPPI EUROPEAN PLACE CONFERENCE

20-22 May 2019 · Crowne Plaza · Porto, Portugal

## Nanocellulose and Polylactic Acid Based Multilayer Coatings for Barrier Applications

Presented by: Dr. Johanna Lahti



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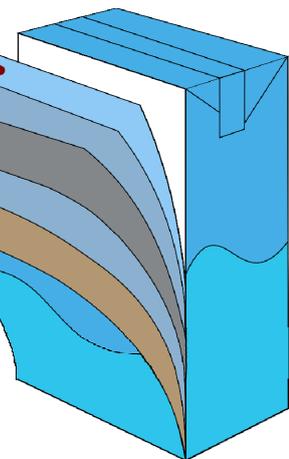
## What is Barrier Packaging made of?

Municipal solid waste is predicted to reach to about 2.2 billion tons by 2025\*



<https://bit.ly/2ryMFCG>

- Polyethylene - Internal liquid barrier
- Polyethylene - Adhesion layer
- Aluminium foil - Oxygen barrier
- Polyethylene - Adhesion layer
- Paperboard - Stability and strength
- Polyethylene - External moisture barrier



Urgent need to replace fossil-fuel based plastics with bio-based and biodegradable alternatives!

\* Kawai K. & Tasaki T., J Mater Cycles Waste Manag, 1-13, (2016)

# Objectives

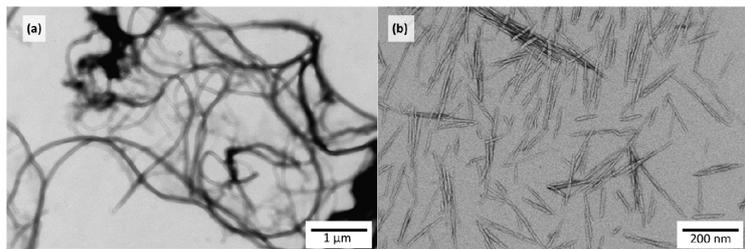
- Explore the possibility to process Nanocellulose and Polylactic Acid into a multilayer structure onto paperboard for barrier applications
- Understand barrier properties of the said multilayer paperboard
- **Demonstrate a continuous process to produce a barrier packaging that is 100% bio-based and biodegradable**

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# Nanocellulose – A promising biomaterial

(a) Microfibrillated Cellulose (MFC) from University of Maine, USA

Diameter: 20 – 60 nm;  
Length: few microns; Crystallinity:  
60 – 70%



(b) Cellulose Nanocrystals (CNCs) from Melodea Ltd.

Diameter: 5 – 20 nm;  
Length: 100 – 200 nm;  
Crystallinity: ~90%



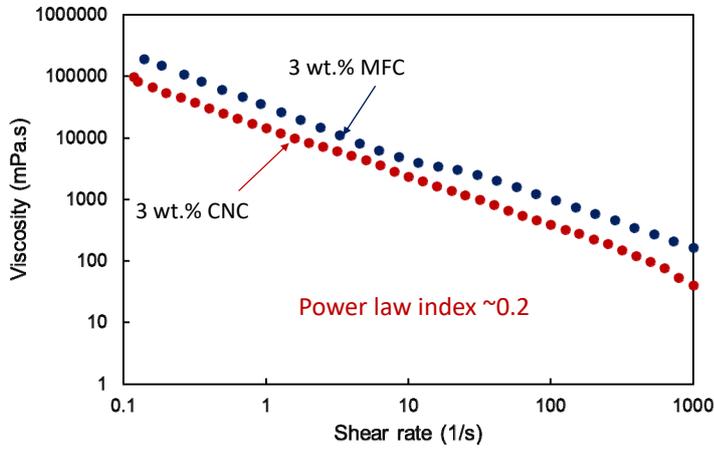
Oxygen barrier  
Mineral oil barrier  
Grease barrier



High viscosity and yield stress  
Adhesion to substrates  
Moisture barrier

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# Shear-thinning behaviour of Nanocelluloses

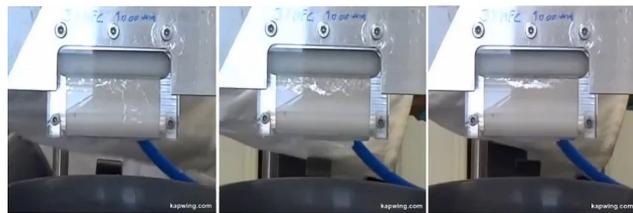


- Highly shear thinning!
- This behaviour can be leveraged by passing the suspension through a narrow slot

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## Slot flow

3wt.% MFC



Increasing pressure drop (and shear rate) →

3wt.% CNC



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## Substrate

- Pigment coated paperboard
  - Smooth and closed surface – better coating holdout at low coat weights
  - Sufficient porosity – for back side drying
  - Wet-strength – has to handle excessive amounts of water
  - Temperature tolerance
- **Poor adhesion to Nanocellulose coatings!**
- Thin layer of **cationic starch** primer coating (< 1 g/m<sup>2</sup>) to improve adhesion

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## Polyactic Acid (PLA)

- Sourced from corn sugar, potato starch and sugarcane
- Barrier against **water vapour**, mineral oils and grease
- Poor barrier against oxygen
- Thermoplastic – hot-melt extrusion

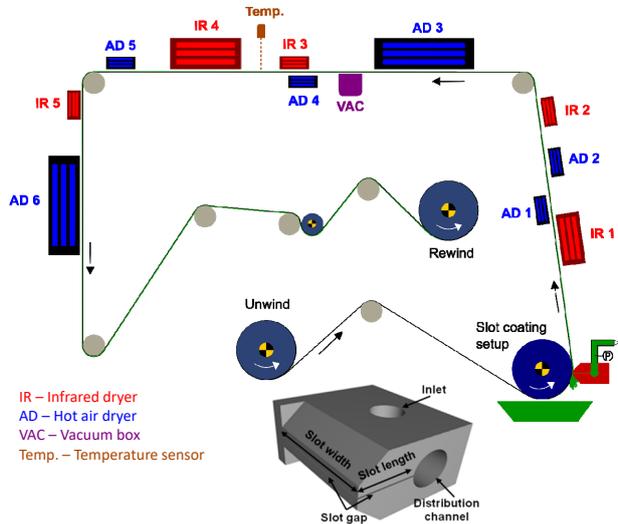


<https://bit.ly/2Hz0ASC>

**Is there a benefit in combining Nanocellulose and PLA?**

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## Slot-die coating of Nanocellulose



- Mini-pilot scale custom built setup based on RK Koater
- IR & hot air drying
- 150 mm web, 1-50m/min speed
- Custom fitting of slot die (length: 34 mm, width: 74 mm, slot gap: 50 – 1000  $\mu\text{m}$ )
- Adjustable gap between substrate and slot-lip for coat weight control

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## Slot-die coating of Nanocellulose



- 2.5 wt.% MFC + 5% CMC\* (of MFC dry weight) as plasticizer
- 3 wt.% CNC + 20% Sorbitol<sup>†</sup> (of CNC dry weight) as plasticizer
- Two coat weights for each suspension ~ 6 (L) and 12 (H)  $\text{g}/\text{m}^2$

\*CMC – Finnfix 4000G (CP Kelco)

<sup>†</sup>Sorbitol – D-sorbitol – 99% (Sigma-Aldrich)

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# Extrusion coating of PLA

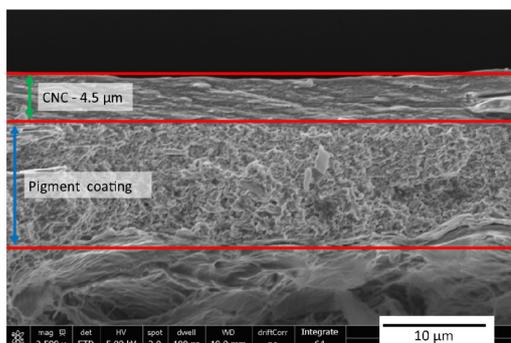


Pilot scale R2R extrusion coater at Tampere University

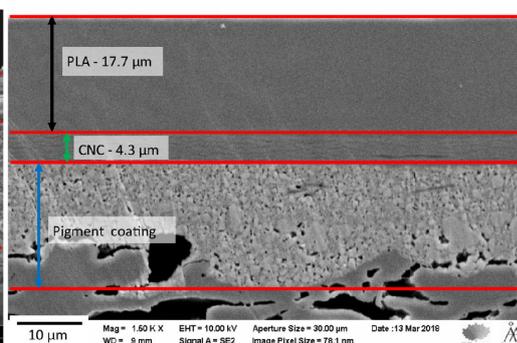
- 23 g/m<sup>2</sup> of PLA coated on top of Nanocellulose coating
- 15 g/m<sup>2</sup> of LDPE coated on top of Nanocellulose as reference
- Corona treatment to improve adhesion at Nanocellulose/PLA (or LDPE) interface

# SEM Cross-sections

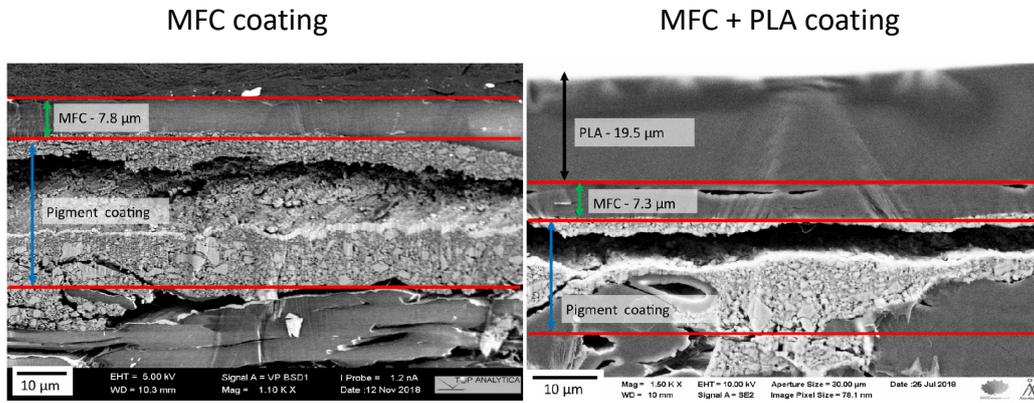
CNC coating



CNC + PLA coating

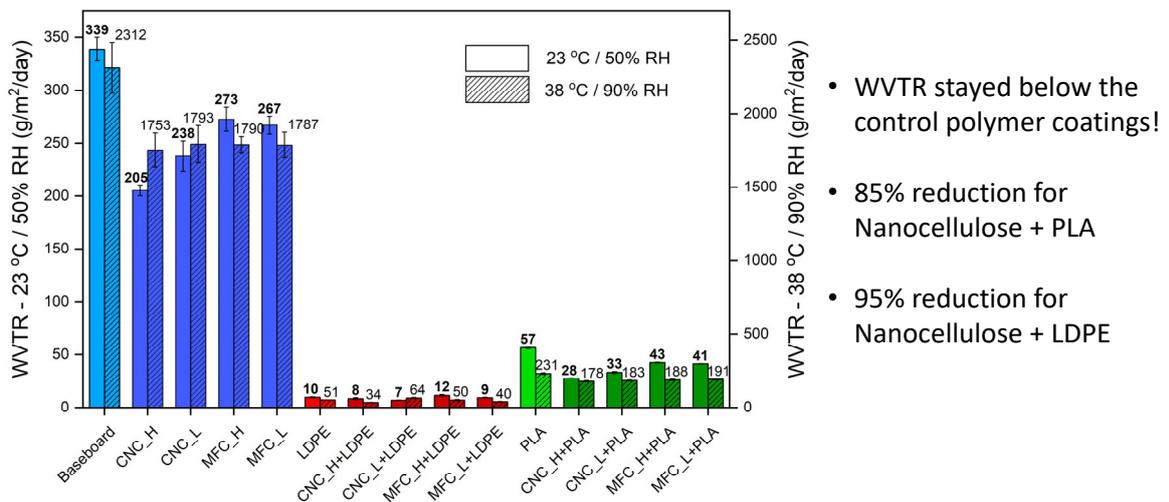


# SEM Cross-sections



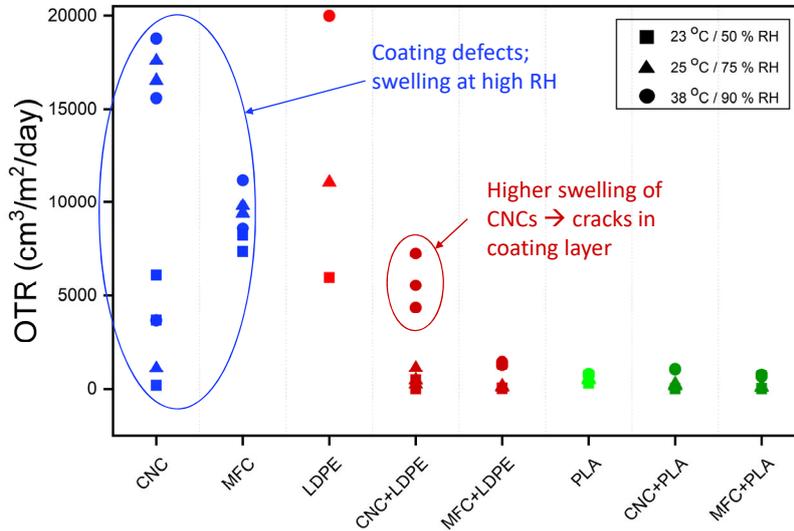
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# Water vapour transmission rate (WVTR)



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## Oxygen transmission rate (OTR)



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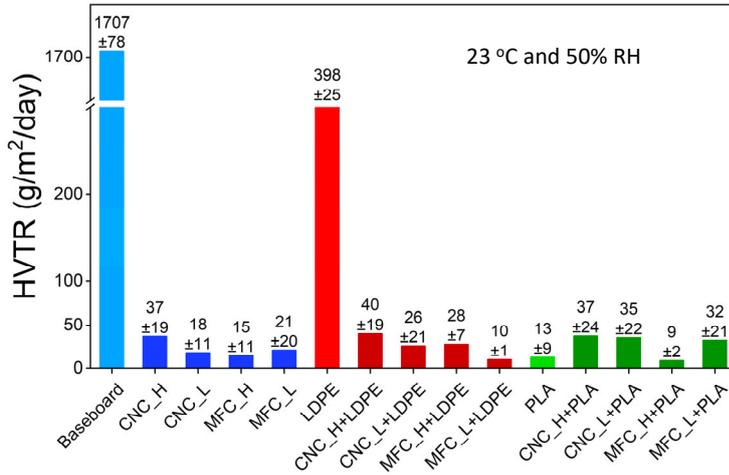
## Oxygen transmission rate (OTR)

	23 °C / 50% RH	25 °C / 75% RH	38 °C / 90% RH
LDPE	-	-	-
LDPE+CNC	15 ± 4	398 ± 98	6117 ± 1228
LDPE+MFC	15 ± 2	123 ± 18	1357 ± 73
PLA	334 ± 32	509 ± 41	771 ± 67
PLA+CNC	7 ± 1	204 ± 37	1065 ± 30
PLA+MFC	11 ± 2	100 ± 16	751 ± 62

- 98% reduction for Nanocellulose + PLA compared to just PLA at 23 °C / 50% RH
- CNCs swell more than MFC at high RH → higher OTR
- Nanocellulose is protected from moisture only from one side!

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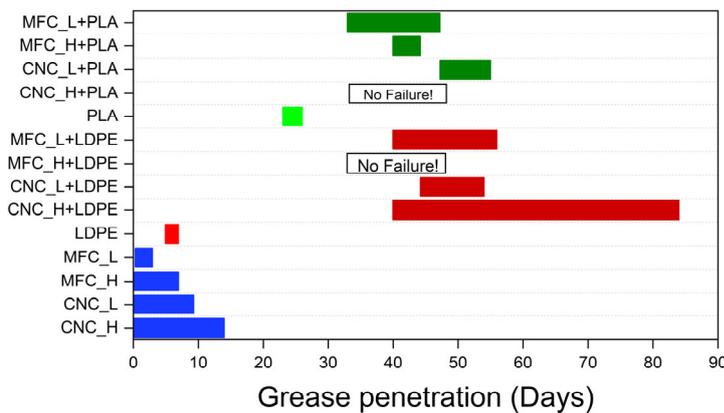
# Heptane vapour transmission rate (HVTR)



- 98% reduction for Nanocellulose + PLA (or LDPE) compared to baseboard
- Considerable improvement over just LDPE coating
- Sensitive to coating defects

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# Grease barrier

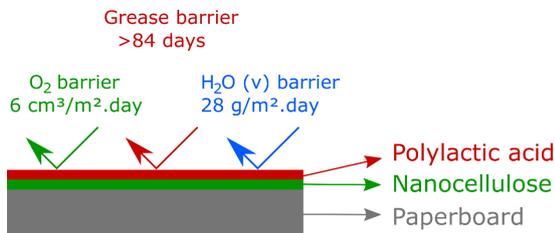


Start point of bar → 1<sup>st</sup> sample fails  
End point of bar → 6<sup>th</sup> sample fails

- Olive oil at 40 °C; six parallel samples
- 5-fold increase compared to Nanocellulose alone
- 10-fold increase compared to LDPE alone
- 2-fold increase compared to PLA alone
- Sensitive to coating defects!

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# Conclusions



- Two continuous coatings methods used in tandem:  
Slot-die coating of Nanocelluloses (MFC/CNCs)  
→ Extrusion coating of PLA (or LDPE)
- Extrusion coating helps fill in the coating defects in Nanocellulose layer
- The multilayer paperboard is 100% bio-based and biodegradable

## Future work

- Backside extrusion coating to further protect Nanocellulose at high RH
- Influence of convertability (creasing and sealing) on barrier properties
- Coating with polymers other than PLA

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Thank You!

### References:

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