Production of dissolving wood pulp (DWP) from sawdust waste

Grant Number: DST/CON 0156/2017
Grant Holder: Dr Jerome Andrew
Duration: 1 April 2020 – 31 March 2023

Other team members: Prof. Bruce Sithole (Supervisor) and Mrs Simiksha Rajcomar (PhD student)

CSIR
Touching lives through innovation

Background

- Forestry, Timber, Pulp & Paper sector important to SA economy[1]:
  - Contributes 1% to GDP, 4% to manufacturing GDP, 25% to Agric. GDP
  - Employs 170 000 people and supports the livelihoods of 700 000 people

- A Sector in decline
  - Reduced demand for traditional timber, pulp and paper products
  - Not very efficient - 90% revenue is obtained from 40% of the tree[2]
  - Large amounts of waste generated – woodchips, sawdust, bark, sludge, etc.
  - Disposed via landfilling, burning, stockpiling – not sustainable waste management
    - GHG emissions, leaching of toxic chemicals, limited landfill space
    - Additional costs incurred by sector & environmental regulations

- Support needed by the sector
  - Alternative uses for its waste
  - Create new value chains – new products + existing products
  - Competitive synergies, new markets, increased product flexibility, leading to increased income streams
  - Reduced environmental footprint of the sector

CSIR

Touching lives through innovation

[1] *Forestry Master Plan 2020-2025*

[2] *http://www.engineeringnews.co.za/article/forestry-group-moves-ahead-plan-integrate-sabie-site-extract-more-value-2013-05-10.*

Background

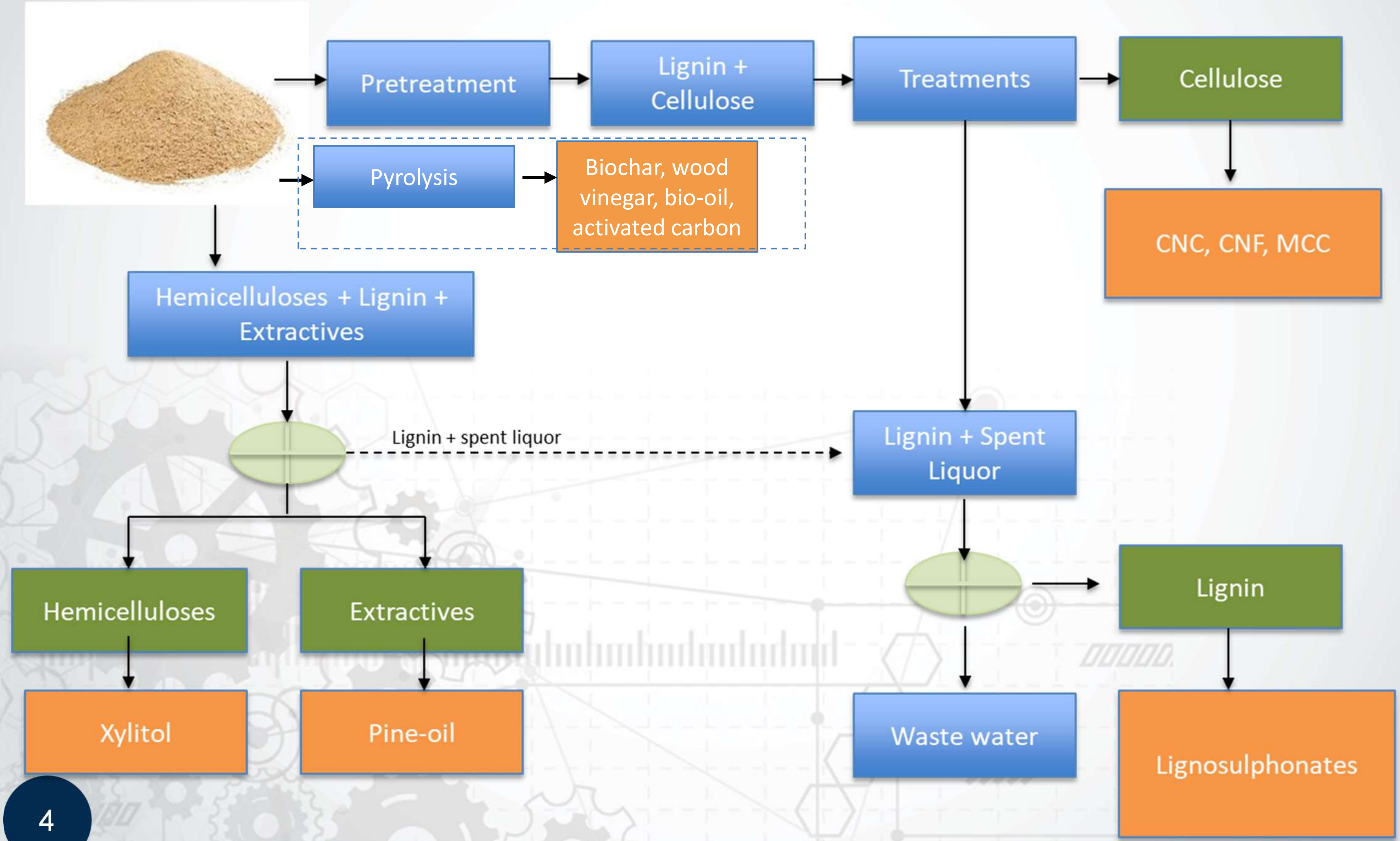
Close to 300 Sawmills in South Africa
76 registered sawmills generate 440 000 tons of sawdust per year

Stockpiling of sawdust and wood chip waste outside sawmill.

Collection of clean uncontaminated sawdust in basement of sawmill during the sawmilling operation

CSIR
Touching lives through innovation

BIDF Sawdust Beneficiation Strategy



Project Aims and Objectives

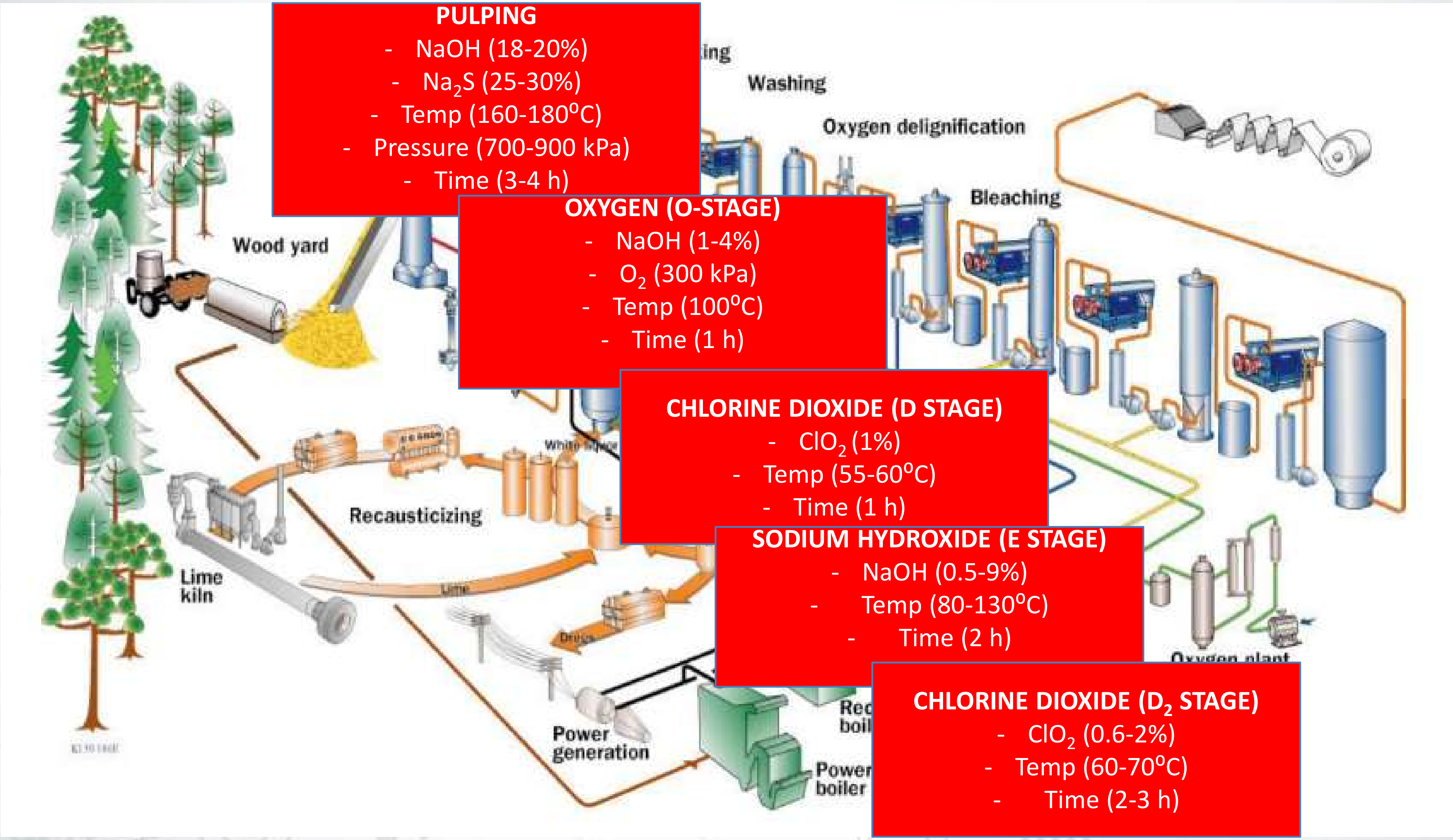
- Aim of the study: to develop novel approach to produce a dissolving wood pulp grade from sawdust waste material
  - Milestone 1:
    1.1 Optimise reaction conditions for maximum removal of lignin and hemicelluloses
    1.2 Investigate removal of hemicelluloses by hydrolysis prior to DWP production
    1.3 Characterisation of sawdust DWP and comparison to commercial pulps
  - Milestone 2:
    2.1 Determine appropriate bleaching strategies and optimize conditions based on DWP
    2.2 Characterise bleached DWP and compare with commercial DWP grades
    2.3 Undertake preliminary techno-economics to produce DWP
  - Milestone 3:
    3.1 Upscale production of DWP using 32 L bio-reactor
    3.2 Evaluate DWP for production of microcrystalline cellulose (MCC)
    3.3 Evaluate DWP for production of viscose
    3.4 Update techno-economic study to produce MCC and viscose from sawdust waste

CSIR
Touching lives through innovation

Dissolving Wood Pulp (DWP)

- DWP is a high purity cellulose product produced from wood chips
- Characterized by
  - High cellulose content
  - High viscosity and degree of polymerization (DP)
  - High purity, high brightness
  - Low lignin, pentosan and extractive content
- Two industrial processes used to produce DWP
  - Pre-hydrolysis kraft (PHK) process
  - Acid bisulphite process
- Shortage of virgin wood fibre
  - seeking alternative fibres for production of DWP

DWP Production



PULPING
- NaOH (18-20%)
- Na_2S (25-30%)
- Temp (160-180^oC)
- Pressure (700-900 kPa)
- Time (3-4 h)

OXYGEN (O-STAGE)
- NaOH (1-4%)
- O_2 (300 kPa)
- Temp (100^oC)
- Time (1 h)

CHLORINE DIOXIDE (D STAGE)
- ClO_2 (1%)
- Temp (55-60^oC)
- Time (1 h)

SODIUM HYDROXIDE (E STAGE)
- NaOH (0.5-9%)
- Temp (80-130^oC)
- Time (2 h)

CHLORINE DIOXIDE (D_2 STAGE)
- ClO_2 (0.6-2%)
- Temp (60-70^oC)
- Time (2-3 h)

- Industrial processes uses wood chips produced from trees – processes are complex, long, and energy, water and chemical intensive

DWP Markets

DISSOLVING WOOD PULP
(10 mil tons in 2023 valued at US$5.5 bil)

Acetates
US$4.02 billion
(2020)

- Films
- Cigarette tow
- Yarn & Fibre
- Plastic

MCC
US$1.1 billion
(2022)

- Pharmaceuticals
- Food
- Cosmetics
- Paintings
- Coatings

Ethers
US$6.41 billion
(2021)

- Pharmaceuticals
- Food
- Binders
- Glue

Viscose
US$65.2 million
(2020)

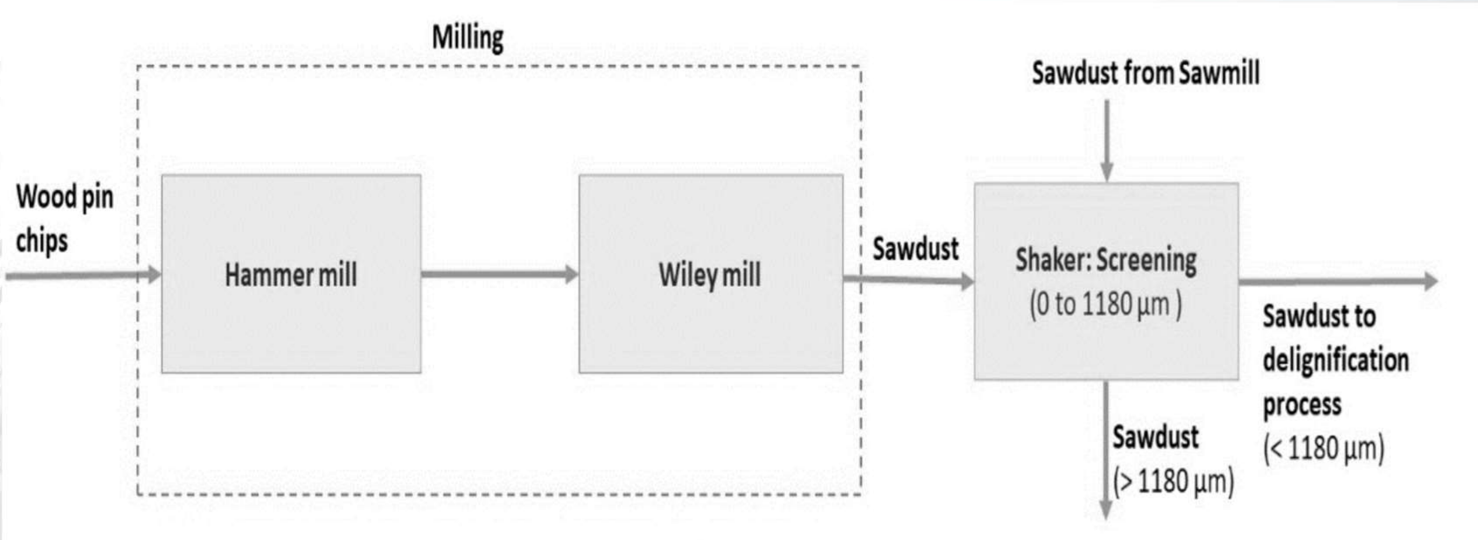
- Textile
- Sausage Skin
- Cellophane
- Sponges

Lyocell
US$1.04 billion
(2021)

- Surgical Product
- Non-wovens
- Cig Filters
- Diapers

Experimental - Sawdust to DWP

- Raw materials
  - Hardwood (*Eucalyptus grandis*) sawdust obtained from sawmill screened to < 1180µm
  - Softwood (*Pinus patula*) sawdust obtained from sawmill screened to < 1180 µm
  - Pin chips (mixed hardwood chips- 50% *E. dunnii, 40% E. grandis,* 10% *clone timber*) obtained from pulp mill – milled into sawdust < 1180 microns



Pin chips and sawdust preparation

CSIR
Touching lives through innovation

Experimental - Sawdust to DWP

- Two-stage process applied to target purity and brightness of the dissolving wood pulp
- All pulps were chemically characterised

Stage 1: Delignification
Lignin removal

→ *Delignified pulp →

Stage 2: Post delignification
Improves brightness and purity

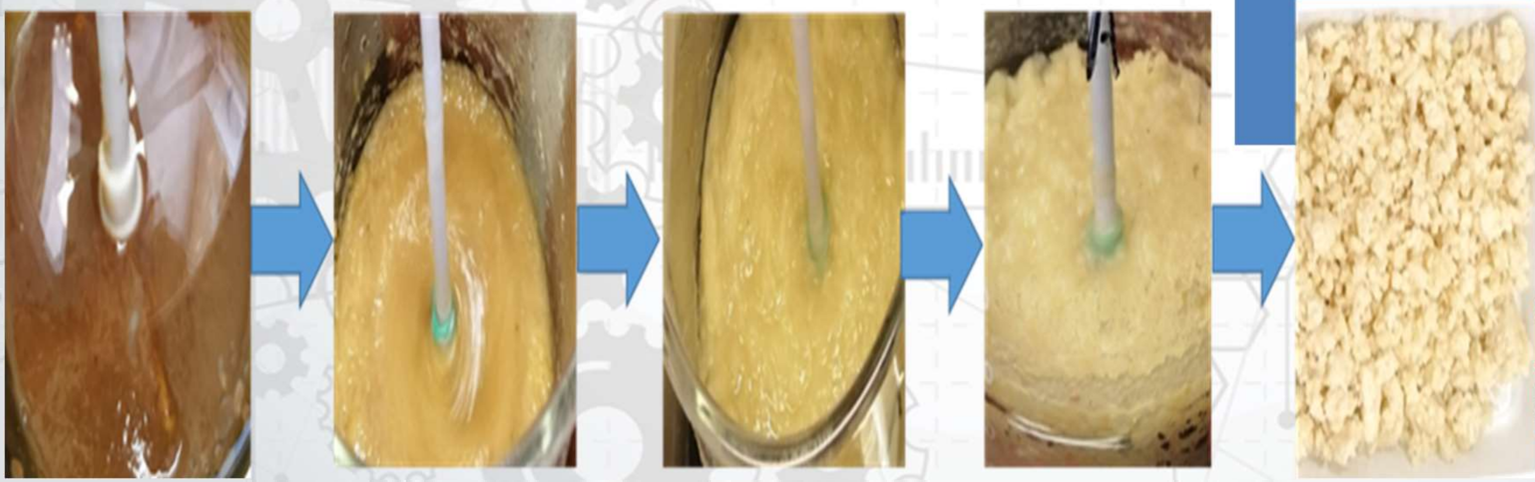
→ * Fully bleached pulp →

*** Characterization of pulps stage 1 & 2:** Viscosity, DP, brightness, S18 &S10, alpha-cellulose, hemicelluloses (HPLC), ash, lignin (AIL, ASL)

Processing conditions optimised for bench top stage 1 delignification process

Process step	Parameter	Conditions
Delignification process	Temperature	60-90°C; Δ10°C
	Time	3-6 h
	oxidant concentration	1, 2 and 3M
	Liquid to wood ratio (L: W)	1:5, 1:7.5, 1:10

Transition of sawdust to unbleached pulp during delignification process



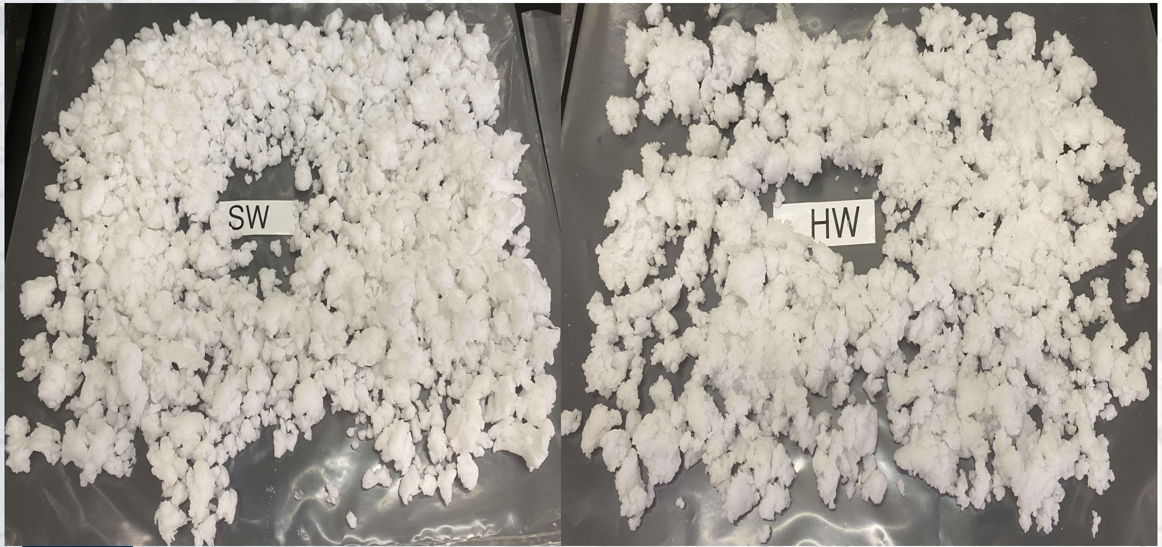
CSIR
Touching lives through innovation

Experimental - Sawdust to DWP

Bleaching - targets brightness and residual lignin and hemicellulose removal as well as establishes a high alpha cellulose content

Bleaching sequences applied: DED, DEpP, DEpD and DEDP

Pulps washed using vacuum filtration and air dried for chemical characterisation



Pulps derived after bleaching and washing process (SW-softwood sawdust pulp and HW-hardwood sawdust pulp)

Processing conditions optimised for stage 2 bleaching process

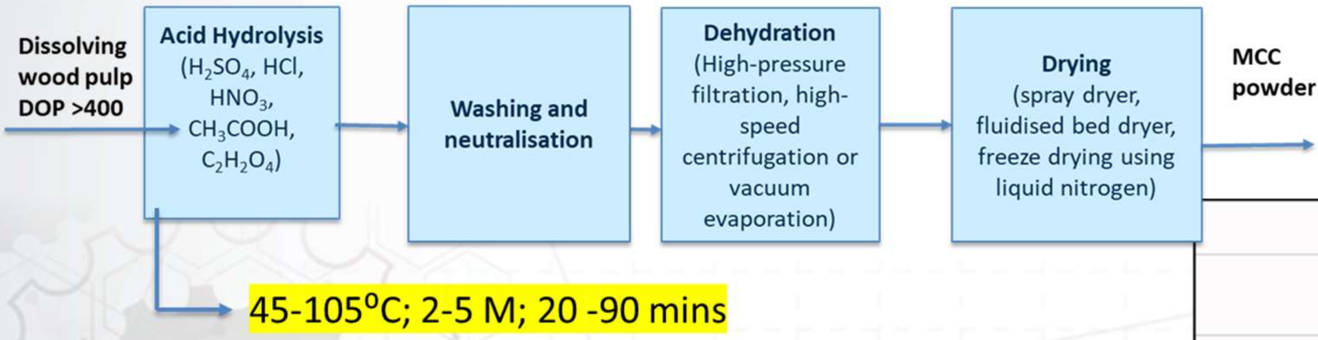
Process	Parameter	Conditions
Do-stage Chlorine dioxide	Time Temperature ClO_2 dosage	120 mins 70°C kappa factor 0.25
E-stage Alkaline stage	Time Temperature NaOH dosage	60 mins 70°C 1.5%
D1-stage Chlorine dioxide	Time Temperature ClO_2 dosage	120 mins 70°C 1.3%
E_P-stage Alkaline stage reinforced with peroxide	Time Temperature NaOH dosage H_2O_2 dosage	60 mins 70°C 1.5% 0.3%
P-stage Peroxide stage	Time Temperature NaOH dosage H_2O_2 dosage	180 mins 80°C 1.0% 0.6%

Characterisation of Bleached DWP

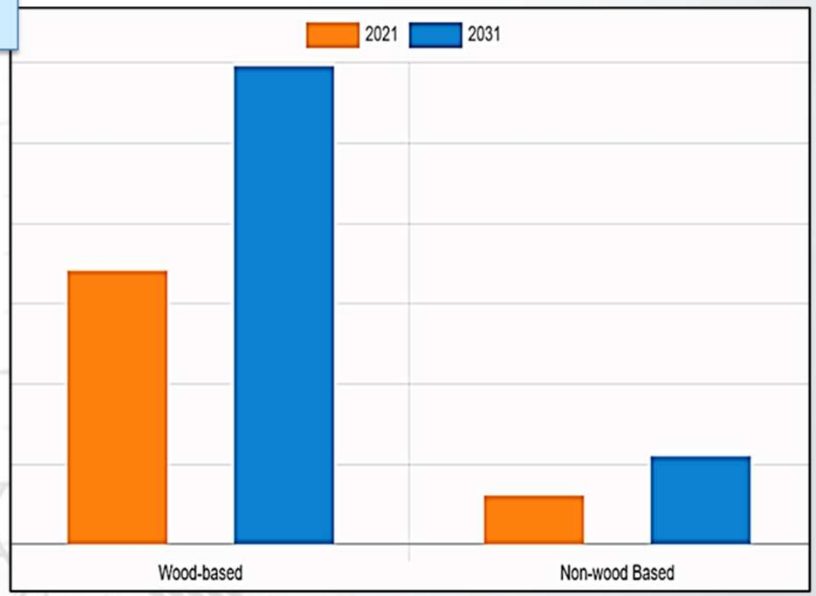
	Commercial DWP (91[α])	HW pulp					SW pulp					MHW pulp				
		UB	DED	DEpD	DEpP	DEDP	UB	DED	DEpD	DEpP	DEDP	UB	DED	DEpD	DEpP	DEDP
Micro kappa no.	N/A	3.0					5.0					4.6				
Bleaching yield (%)	N/A	-	91.6	90.4	84.3	93.7	-	91.6	84.7	84.3	89.1	-	92.5	91.2	93.1	86.8
Overall yield (%)	N/A	51.0	44.4	46.1	45.1	43.5	52.0	47.8	45.3	43.8	46.3	44.0	40.9	40.1	40.1	38.1
Viscosity (ml/g)	421	195	189	180	200	183	380	331	370	342	310	313	243	242	222	205
DP	1316	553	533	503	569	512	1213	1031	1176	1073	954	966	717	714	646	587
ISO brightness (%)	93	67.0	87.4	88.5	81.0	87.0	66.0	86.9	84.7	77.4	88.4	72.0	86.0	86.1	75.4	86.3

Microcrystalline Cellulose (MCC)

- High purity, partially depolymerised form of cellulose produced from α-cellulose
- Derived mainly from high quality pulps (obtained from wood or cotton)
- Wood is preferred raw material - *e.g.* Dissolving wood pulp (DWP)



- MCC versatility for applications: non-toxicity, bio-degradability, bio-compatibility, high mechanical strength, large surface area
- MCC high demand - challenges and limitations remain around producing low-cost MCC



Demand of raw material sources for producing MCC (SUMANT and SHARMA, 2023)

MCC Applications

MICROCRYSTALLINE CELLULOSE (MCC)
(190 000 tons)

- In 2023, the global MCC market valued at US$ 1.2 billion. Anticipated to double to US$ 2.1 billion by 2030

Pharmaceuticals (39%)
- Capsules
- Pellets
- Tablets
- Sachets
- Suspensions
- Emulsions

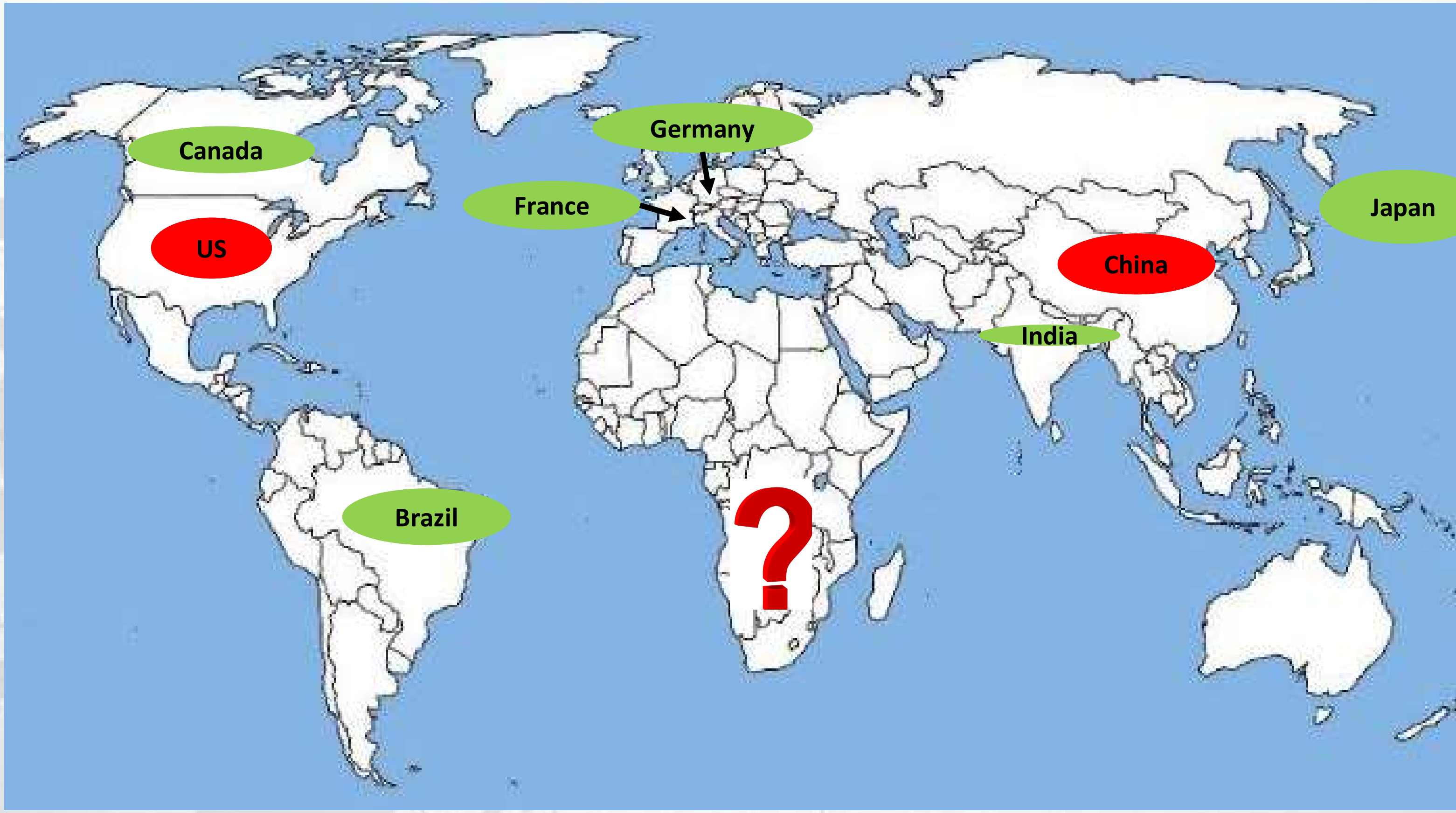
Food Manufacturing (30%)
- Baked Products
- Fat-free
- Diary
- Canned Meat
- Frozen Foods
- Condiments

Cosmetic & Personal Care (15%)
- Skin care
- Hair care
- Oral Products
- Makeup

Paints & Coatings (7%)
- Rheology modifiers
- Non-stick coatings

MCC Global Production

- China and the USA have been leading in global producers of MCC
- Other significant players include India, Brazil, Canada, Germany, Japan and France.
- No known producers in Africa - although South Africa home to one of the largest DWP producers in the world



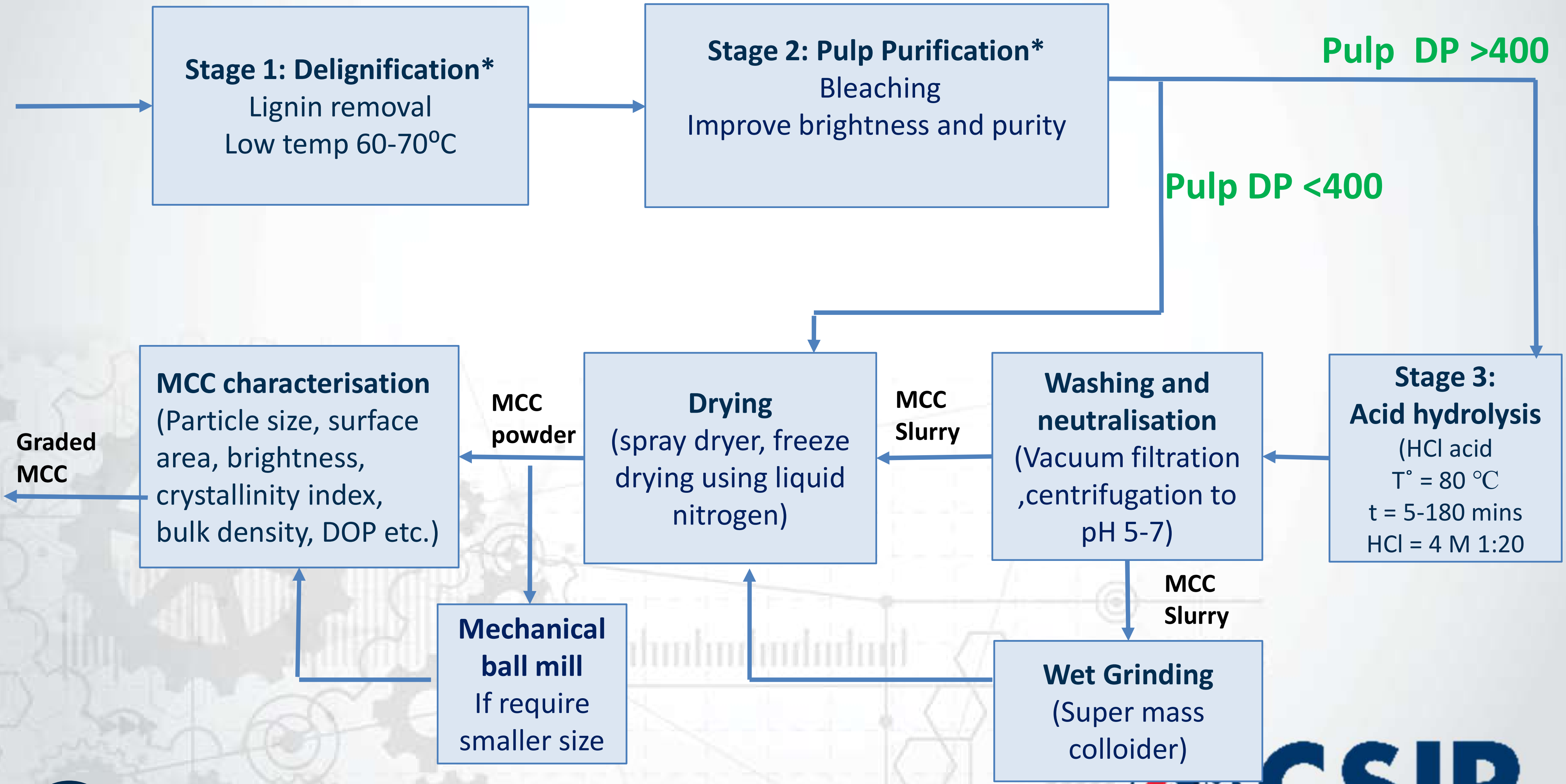
MCC Local Market in South Africa

- India, Germany, Taiwan and China are leading exporters of MCC to SA
- MCC retails for R1000 – 2000 per kg

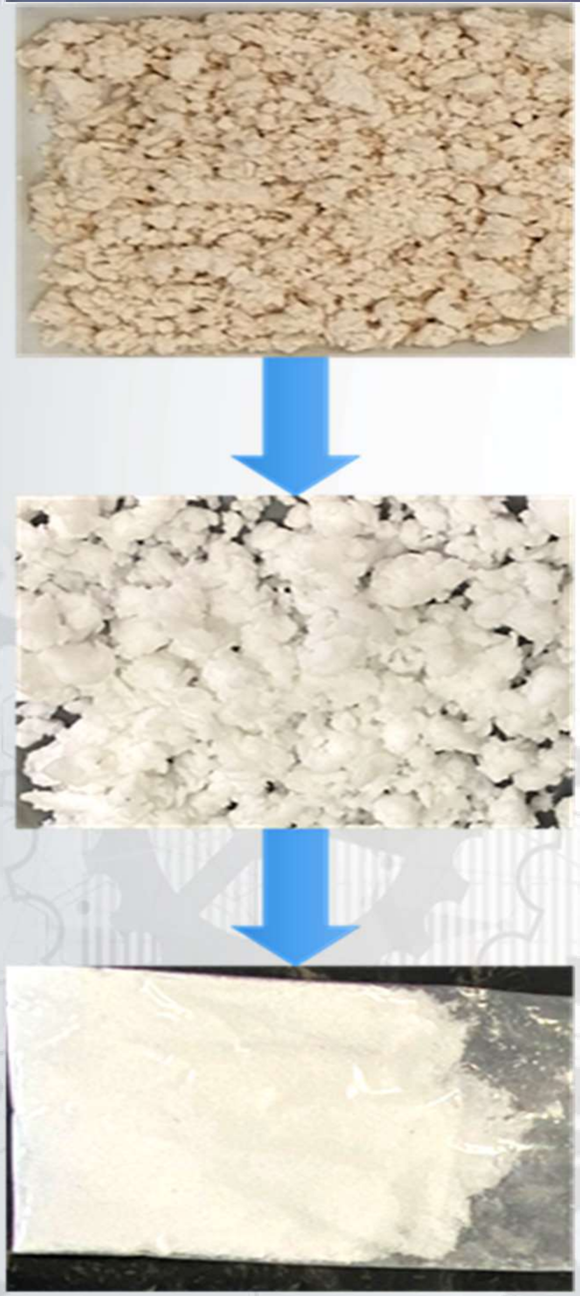
Year	Tonnage
2020	222.5
2021	169.5
2022	201.8
2023 (Jan-Mar)	80.0

CSIR
Touching lives through innovation

Experimental Overview



Properties of MCC produced



	Commercial MCC	91α MCC*	HW MCC	SW MCC	MHW MCC
Acid hydrolysis		80 °C, 4 M HCl,120 min	80 °C, 4 M HCl, 5 min	80 °C, 4 M HCl, 120 min	80 °C, 4 M HCl, 5 min
Overall yield (%)	nd	nd	45	44	34
Acid hydrolysis yield (%)	-	95	98	96	90
Viscosity (ml/g)	127	129	109	146	124
DP	334	343	279	393	325
Monosaccharides					
Arabinose (%)	nd	nd	nd	nd	nd
Galactose (%)	nd	nd	nd	nd	nd
Glucose (%)	91.4	97.4	98.9	93.7	94.4
Xylose (%)	1.34	0.00	0.61	0.00	2.74
Mannose (%)	0.12	0.00	nd	2.98	nd

*nd- not detected (less than detection limits)

91α MCC produced from 91α DWP

- CrI measured 88-90% for prepared MCC , 93% for commercial MCC

Pilot scale MCC production

	HW	SW	MHW
Conditions	(3 M,60°C,4 h)	(3 M,60°C,6 h)	(3 M,70°C,3 h)
Yield (%)	46.0	51.2	35
Rejects (%)	1.35	0.22	1.5
Micro kappa no.	5.4	3.6	4.3

- Scale up using 32 L reactor processing 1 kg sawdust

CSIR
Touching lives through innovation

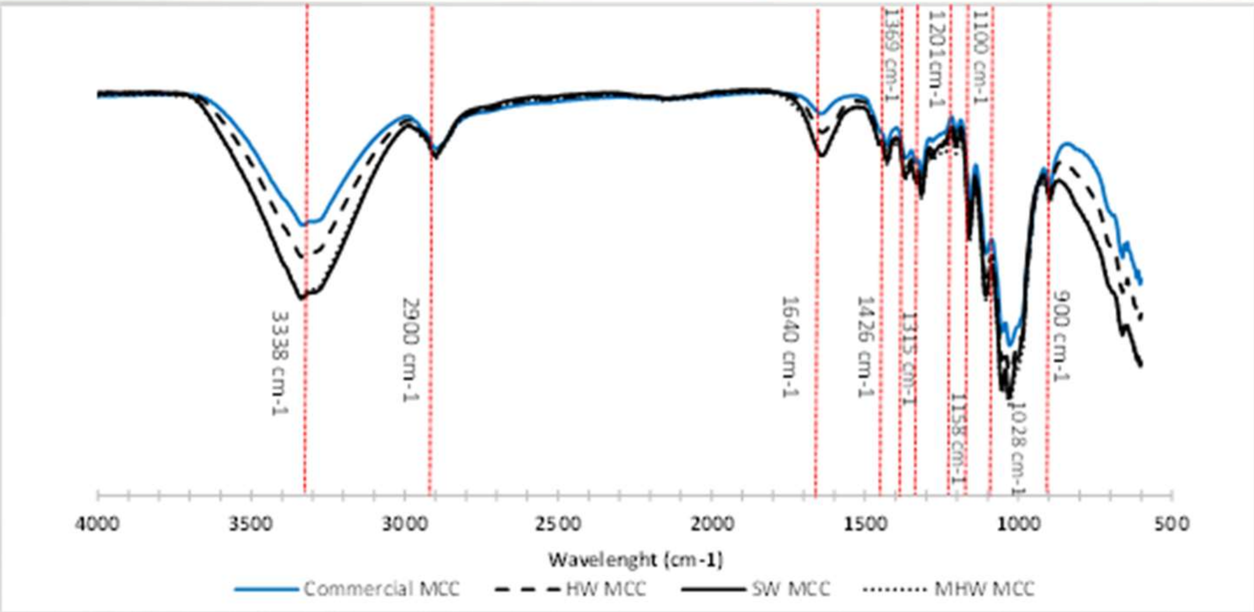
Pilot scale MCC production

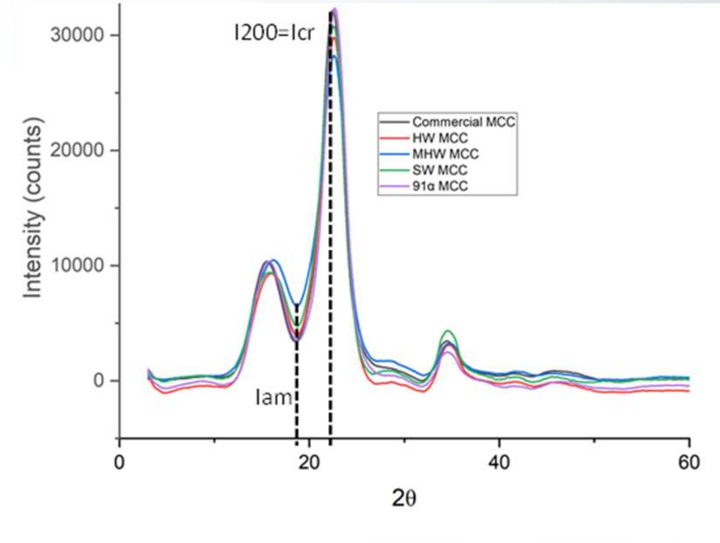
	Commercial MCC	91α MCC	HW	SW	MHW
Acid hydrolysis conditions	-	(4 M,80°C, 120 mins)	(4 M,80°C, 60 mins)	(6 M,80°C,120 mins)	(4 M,80°C, 5 mins)
Overall yield (%)	-	-	42	41	30
pH	7.35	7.23	6.72	6.75	5.08
CrI (%)	89	87	84	84	76
LOD (%)	4.5	7.90	10	23	15
Intrinsic viscosity (ml/g)	142	129	131	147	102
DP	383	343	347	398	257
ASL (%)	0.30	0.67	0.29	0.51	0.32
AIL (%)	Cannot be measured for bleached products				
Monosaccharides					
Arabinose (%)	0.0	0.0	nd	nd	nd
Galactose (%)	0.0	0.0	nd	nd	nd
Xylose (%)	2.10	0.0	nd	nd	nd
Mannose (%)	0.0	0.0	0.0	1.83	0.0
Glucose (%)	95.7	97.5	87.5	83.6	78.1

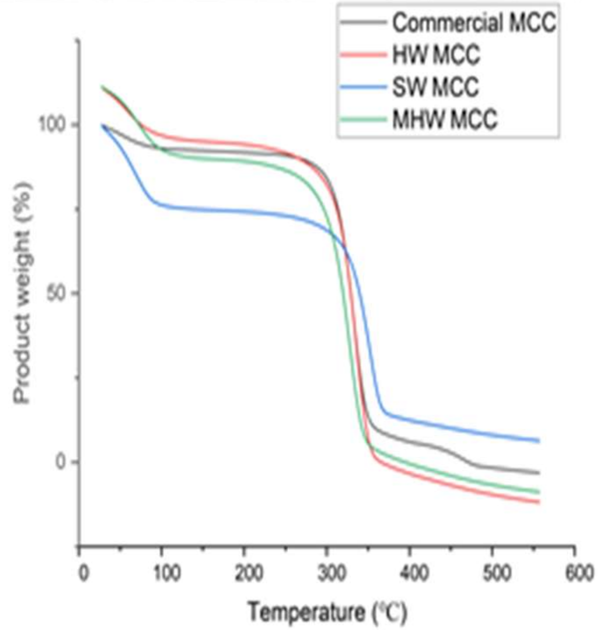
*nd- not detected (less than detection limits)
91α MCC produced in laboratory using 91α DWP

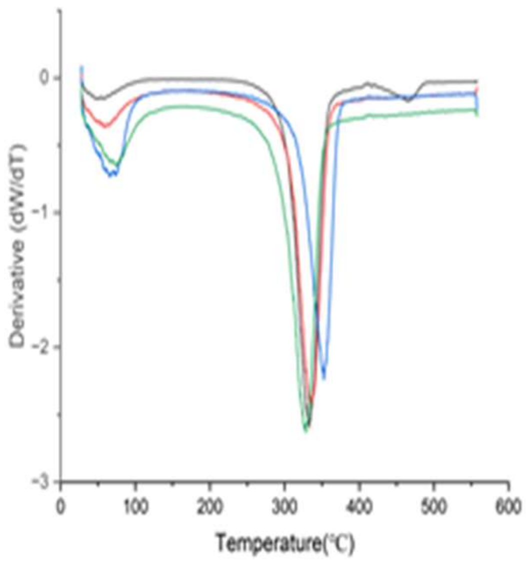
CSIR
Touching lives through innovation

Pilot scale MCC production









- MCC match with Avicel PH101 commercial MCC
- Onset temperatures of MCC (250-289℃) similar to commercial MCC at 280℃
- Max degradation temp = 325℃
- Crystallinity Index close match to commercial MCC

CSIR
Touching lives through innovation

Conclusions

- A grade of pulp similar to commercial DWP was produced from sawdust using a low temperature process (60℃)
- The degree of polymerization (DOP) of pulp was not suitable for viscose due to low viscosity of the pulp
- However, MCC was successfully produced
- Properties of synthesized MCC compared well with commercial MCC

Financial Report

ACTIVITY BASED (FINANCIAL REPORT)	
GRANT NUMBER:	DST/CON 0156/2017
TOTAL AWARDED GRANT AMOUNT (excl. VAT):	R2 454 888
FINAL EXPENDITURE ON PROJECT (excl. VAT):	R2 454 888
UNSPENT FUNDING	R0

Touching lives through innovation

Project Team - supported

Researcher name	Gender	Race	Nationality	Institutional affiliation
Jerome Andrew	Male	Indian	South African	CSIR
Jonas Johakimu	Male	Black	Burundian	CSIR
Londani Musimanyana	Female	Black	South African	CSIR
Pule Seemela	Male	Black	South African	CSIR
Christabel Thangwane	Female	Black	South African	CSIR
Prabashni Lekha	Female	Indian	South African	CSIR – since resigned

Student name	Gender	Race	Nationality	Degree completed	Student number
Simiksha Rajcomar (née Balkissoon)	Female	Indian	South African	PhD	209502586

Project Team – not supported

Researcher name	Gender	Race	Nationality	Institutional affiliation
Prof. Bruce Sithole	M	Black	Canadian	UKZN
Kersch Naidoo	M	Indian	South African	CSIR – AAF
Nelisiwe Ngubane	F	Black	South African	CSIR intern
Minenhle Mfiki	M	Black	South African	CSIR Intern
Philasande Machi	M	Black	South African	DUT – WIL intern
Pumela Jafta	F	Black	South African	YES Intern
Lethabo Gaebee	M	Black	South African	YES Intern
Nelisiwe Ngubane	F	Black	South African	CSIR intern
Minenhle Mfiki	M	Black	South African	CSIR Intern

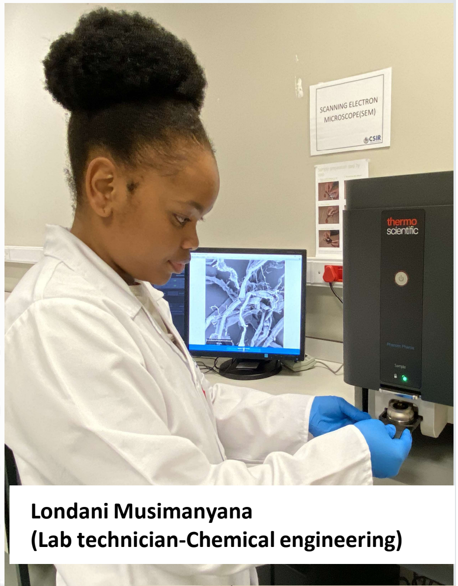
Summary of Deliverables

Deliverables	Contracted	Final Published	In-Draft	Comment
Dissertations	1	1	0	Two year embargo not to publish the thesis requested from UKZN
Journal Papers	2	1	1	Submitted to Journal
Technical Reports	1	1	0	
Conference Papers	2	2	0	
Technology Packages	1	1	0	Tech Demo TRL 6
Other	0	1	0	Poster presented at Sustainable Bioenergy & Processes, Cape Town, South Africa, 13-15 December 2021
TOTAL	7	6	1	

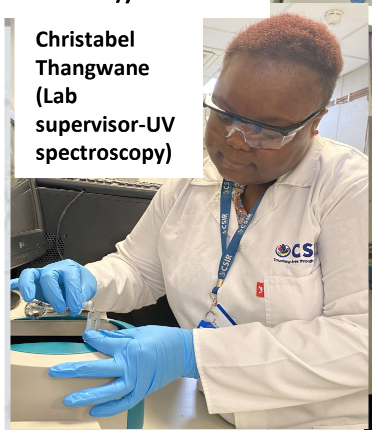
Acknowledgments

- Waste Research, Development and Innovation Roadmap for funding of the project
- CSIR BIDF staff/interns/post graduate students: assistance with conducting experimental and analytical training/testing.
- CSIR BIDC: conducting pilot experimental trials and analytical services
- Mondi Durban, South Africa: Analytical services
- Sappi Technology Centre: Analytical services
- CSIR Agro processing department: assisting with spray drying

Pule Seemela (Lab technician-Chemistry)

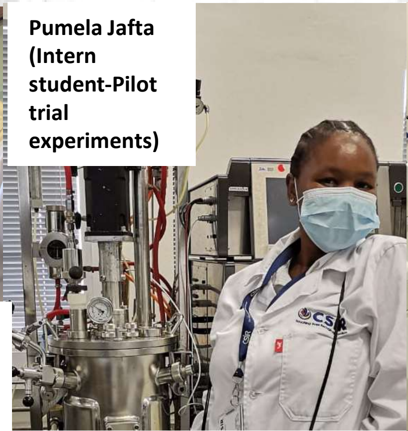


Londani Musimanyana (Lab technician-Chemical engineering)

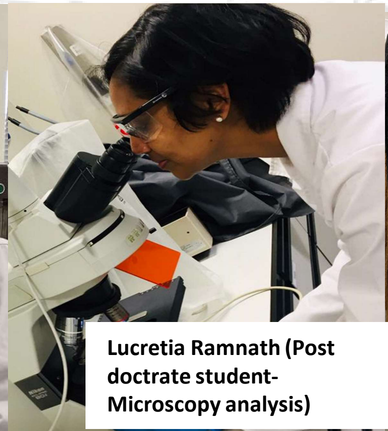


Christabel Thangwane (Lab supervisor-UV spectroscopy)

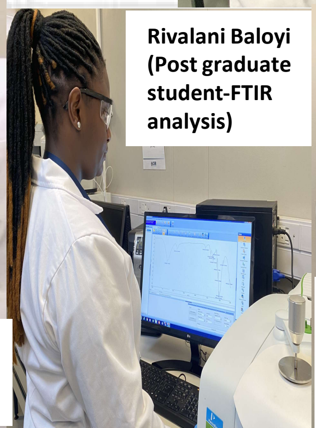
Minenhle (Intern student-Ash analysis)



Pumela Jafta (Intern student-Pilot trial experiments)



Lucretia Ramnath (Post doctrate student-Microscopy analysis)



Rivalani Baloyi (Post graduate student-FTIR analysis)

Philsande Machi- (Intern student-experimental tests)

CSIR

Touching lives through innovation