Biorefining of sugars via Process Intensification

Research and Innovation action (RIA) – Horizon 2020-BBI-2019-SO2-R6

Improve biorefinery operations through process intensification and new end products

D8.2 Interim Technical Report

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Lead Beneficiary	Newcastle University (UNEW), School of Engineering, Merz Court, Newcastle upon Tyne NE1 7RU, UK		
Responsible Author	Kamelia Boodhoo, UNEW, Kamelia.boodhoo@newcastle.ac.uk Fernando Russo Abegão, UNEW, Fernando.Russo-Abegao@newcastle.ac.uk		
Contributions from	Mladen Crnomarkovic, MAT, mladen@maturusoptimi.com Juan Enriquez, A-DSC, jenriquez@analisis-dsc.com Charalambos Klitis, EBOS, charalambosk@ebos.com.cy Ana Maria Lopez, IRIS, alopez@iris-eng.com Andrea Minigher, AEP, andrea.minigher@aeppolymers.com Markku Ohenoja, UOULU, markku.ohenoja@oulu.fi Karoline Wowra, Dechema, karoline.wowra@dechema.de		

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Glossary of terms and abbreviations used

Abbreviation / Terms	Description
5-HMF	5-hydromethylfurfural
CFD	Computational Fluid Dynamics
CSTR	Continuous Stirred Tank Reactor
HMC	Hemicellulose
ILCSA	Integrated Life Cycle Sustainability Analysis
KPI	Key Performance Indicator
LCA	Life Cycle Analysis
LCI	Life Cycle Inventory
PI	Process Intensification
PSE	Process Systems Engineering
SDR	Spinning Disc Reactor
TEA	Techno-Economic Assessment
TRL	Technology Readiness Level
WP	Work Package

Executive Summary

The overall aim of BioSPRINT is to develop, test and validate process intensification (PI) methods (up to TRL 4-5) for sustainable and cost-effective purification and conversion of sugars from the hemicellulose (HMC) fraction of lignocellulosic biomass to obtain biorenewable furan-based resins for novel polymeric applications.

There are four technical areas of interest in the processing of HMCs which are targeted for intensification development in BioSPRINT: (1) Upstream purification focusing on intensifying the purification and concentration of sugars; (2) Catalytic conversion of purified C5 and C6 sugars into furan-based monomers; (3) Downstream purification of the furan-based monomers; (4) Polymerisation of furan-based derivatives into novel formulations of biorenewable resins. Processing development in these four technical areas are underpinned by CFD modelling of each process step and process flow simulation of the overall biorefinery process, a Techno-Economic Analysis (TEA) and an Integrated Life Cycle and Sustainability Analysis (ILCSA). Industrially relevant HMC feedstocks derived from hard wood and straw streams generated from the production of paper pulp or biofuels are applied in this project.

Significant technical progress has been made up to M25 of this 4-year project. Key highlights include:

- In the upstream purification step, the application of nanofiltration and reverse osmosis membranes has resulted in the target recovery (80—90%) and purity (70—80%) of sugars being achieved or even exceeded with selected membranes, operating conditions and diafiltration strategies. Another PI technology based on the spinning disc concept, applied in the recovery of lignin by precipitation via solvent evaporation of an Organosolv black liquor, has been able to recover up to ca. 95% of the original lignin present as high purity solid precipitates. Thermal energy consumption in this process is 2x lower and lignin yield is at least one order of magnitude higher compared to conventional evaporative technologies. There is scope for combining membranes and spinning discs in an integrated upstream purification cascade for concentrating HMC streams, recovering purified molecules and recycling solvents.
- In the catalytic conversion work stream, novel catalyst formulations for scalable and adaptable acid catalysts have been developed with promising results, especially in the context of multiple sugars in mixed streams. Further work will focus on the development of continuous processing in bi-phasic systems using an intensified reactor technology with the novel catalyst formulations.
- In the downstream separation step, suitable solvents have been identified which can promote high reaction selectivity and efficiently extract 5HMF and furfural (more than 90% removal). Furfural and 5-HMF of high purity can be obtained from combined continuous distillation and extraction. The separation of organic and aqueous phase is also being investigated in hydrocyclones, a very promising advanced and energy efficient method, not reported to date for such an application.
- In the polymerization step, the most promising and innovative achievements so far include the development of REACH compliant furfural-based novolacs and the formulation of polyurethane foams using furfural-based polyols, with comparable or even improved properties with respect to industrial references.

Keywords: Process Intensification, lignocellulosic biomass, hemicellulose, 5-HMF, furfural, membranes, spinning disc reactor, hydrocyclone, sugar dehydration, acid catalysts, extractive reaction, biorenewable resins

1. Introduction

Lignocellulose is the type of biomass with the highest availability in the world- equivalent to *ca.* 200 billion tonnes per year globally[1]. In Europe alone, lignocellulosic biomass has an estimated annual potential of technical availability of 1.4 billion tonnes, which could be sustainably used by 2030[2], doubling the current usage. The EU biorefinery industry has grown rapidly in the last decade, with successful large scale facilities implemented across the continent for converting first generation biomass into biofuels[3]. More recently, there has been a drive to accelerate production of biorenewable energy and chemicals from more sustainable, widely available second-generation lignocellulosic feedstocks as non-food bio-based resources. However, a major increase of the usage and processing efficiency of lignocellulosic biomass for sourcing the chemical industry is necessary to reach the goal of 25 % bio-based chemicals in 2030[4].

While processes are well established to valorise two of the three major components in lignocellulose, *i.e.* cellulose (for fibres) and lignin (for energy), hemicelluloses (HMCs), which typically account for 20-30 %(w/w), are relatively under-exploited[5]. The major challenge with processing of HMCs is related to the complexity of the HMC streams, which typically contain a wide range of C5 and C6 sugar monomers and their oligomers, uronic and acetic acids, residual lignin, ashes as well as pre-treatment chemicals. Extracting the sugars can therefore lead to expensive purification processes, product degradation through processing, and low yields of each of the components in the mixture. Nevertheless, HMC streams contain valuable components in the form of the C5 and C6 sugars, and it is imperative that their value is harnessed to increase biorefineries resource efficiency and profitability and to add to the range of biorenewable feedstocks available. HMC streams derived from wood industry processes (*e.g.* those employed in the production of wood fibres, pulp, or second generation biofuels) are of particular interest for immediate exploitation and valorisation. Such streams are readily available in large scale production processes and represent an under-utilised resource rich in HMCs.

Process Intensification (PI) has the potential to contribute significantly to address the challenges identified for the efficient processing of HMC by making biorefining processes faster at lower footprints, more efficient, more integrated and better for the environment. BioSPRINT aims at utilising and valorising hemicelluloses, pursuing intensified processing steps and technologies towards 'zero-waste' bio-based operations and applying an integrated biorefinery concept that embodies the cascading principle to create new HMC -derived products, so as to maximise conversion of the lignocellulosic biomass feedstock and its by-products, side streams and residual streams into higher added-value products. The BioSPRINT philosophy is based on valorising the previously-discarded or costly HMC side streams from processes employed in the production of pulp and paper or biofuels.

This report provides a summary of the technical achievements from the BioSPRINT project between M1-M25, representing the first half of this 4-year project. The technical challenges encountered and solutions implemented are also presented. Finally, the planned work in the next stage of the project is also highlighted.

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1 De Bhowmick, G. et al. (2018), Lignocellulosic biorefinery as a model for sustainable development of biofuels and value added products, *Bioresource Technology*, 247, 1144–1154.

2 Panoutsou et al. (2016) D8.1 Overview report on the current status of biomass for bioenergy, biofuels & biomaterials in Europe, S2Biom

3 Hassan, S.S. et al. (2019), Lignocellulosic Biorefineries in Europe: Current State and Prospects, *Trends in Biotechnology*, 37 (3), 231-234.

4 Objective set by the Bio-based Industries Consortium (BIC) in the 2017 Strategic Innovation and Research Agenda (SIRA).

5 Maki-Arvela, P. et al. (2011) *Chem. Rev*. 111, 5638–5666.

1.1 Mapping Projects' Outputs

Table 1: Adherence to BioSPRINT's GA Deliverable & Tasks Descriptions

<table>
<tr><th colspan="2">BioSPRINT Task</th><th>Respective Document Chapter(s)</th><th>Justification</th></tr>
<tr><td>T8.2 Technical Management</td><td>This task monitors the technical progress of the integrated BioSPRINT biorefining concept for HMCs.</td><td>Chapter 3, 4 and 5.</td><td>Focus is on tasks completed in WP1-6 in M1-M25:
- WP1 &2- Upstream Purification and Conversion of HMC sugars
- WP3 &4 - Downstream Separation and Polymerisation of 5HMF and Furfural into industrially relevant and REACH-compliant biopolymers.
- WP5 & 6- Modelling, scale-up and ILCSA system definition</td></tr>
<tr><th colspan="4">BioSPRINT Deliverable</th></tr>
<tr><td colspan="4">D8.2 Interim Technical Report
This deliverable summarises the technical achievement and key breakthrough technologies and procedures developed in WP 1-6 in the first 24 months of the project. Technical challenges encountered and solutions implemented to address these challenges are also highlighted. Planned actions in these WPs for M26-M48 leading to the Final Technical Report in M48 are also highlighted.</td></tr>
</table>

2. BioSPRINT Technical Aims and Objectives

The overall aim of BioSPRINT is to develop, test and validate process intensification (PI) methods (up to TRL 4-5) for sustainable and cost-effective purification and conversion of sugars from the hemicellulose (HMC) fraction of lignocellulosic biomass to obtain biorenewable furan-based resins for novel polymeric applications.

The specific technical objectives are illustrated in

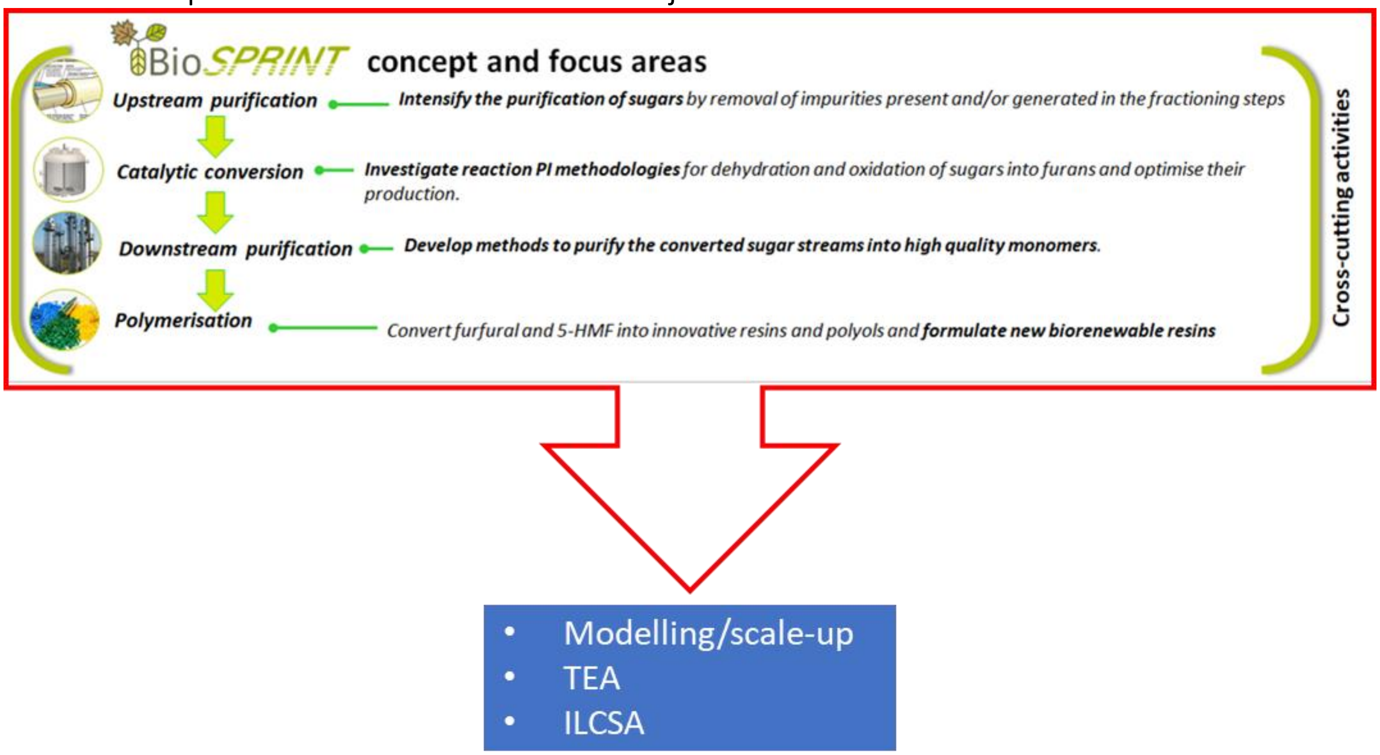

Figure 1. These are related to intensified upstream purification (WP1), intensified catalytic dehydration of C5 and C6 sugars (WP2), efficient downstream separations of 5-HMF and furfural obtained from sugars (WP3) and polymerisation of sugar-derived biobased molecules into biorenewable resins (WP4). Techno-economic viability and replicability of the new biorefinery concept based on developments in WPs 1-4 will be assessed using process simulation tools and validated by integrating PI technologies at TRL 5 (WP5). Lastly, in WP6, an integrated sustainability assessment will be conducted to demonstrate the sustainability and safety of the final processes and biorenewable products.

Targeted feedstocks for processing in BioSPRINT involve HMC streams derived from hard wood and straw streams emanating from industrial processes typically employed in the production of paper pulp or biofuels. These existing streams have explicitly been selected due to their industrial relevance, and the opportunity they provide to work with different lignocellulosic feedstocks and processes, each with their unique characteristics and challenges. Such streams are usually not valorised at all, or if they are, this is limited to biofuel and power generation uses. Therefore, valorisation of these streams as proposed in BioSPRINT will offer the opportunity to greatly reduce the feedstock-to-waste and waste-to-energy flows in the biorefinery context.

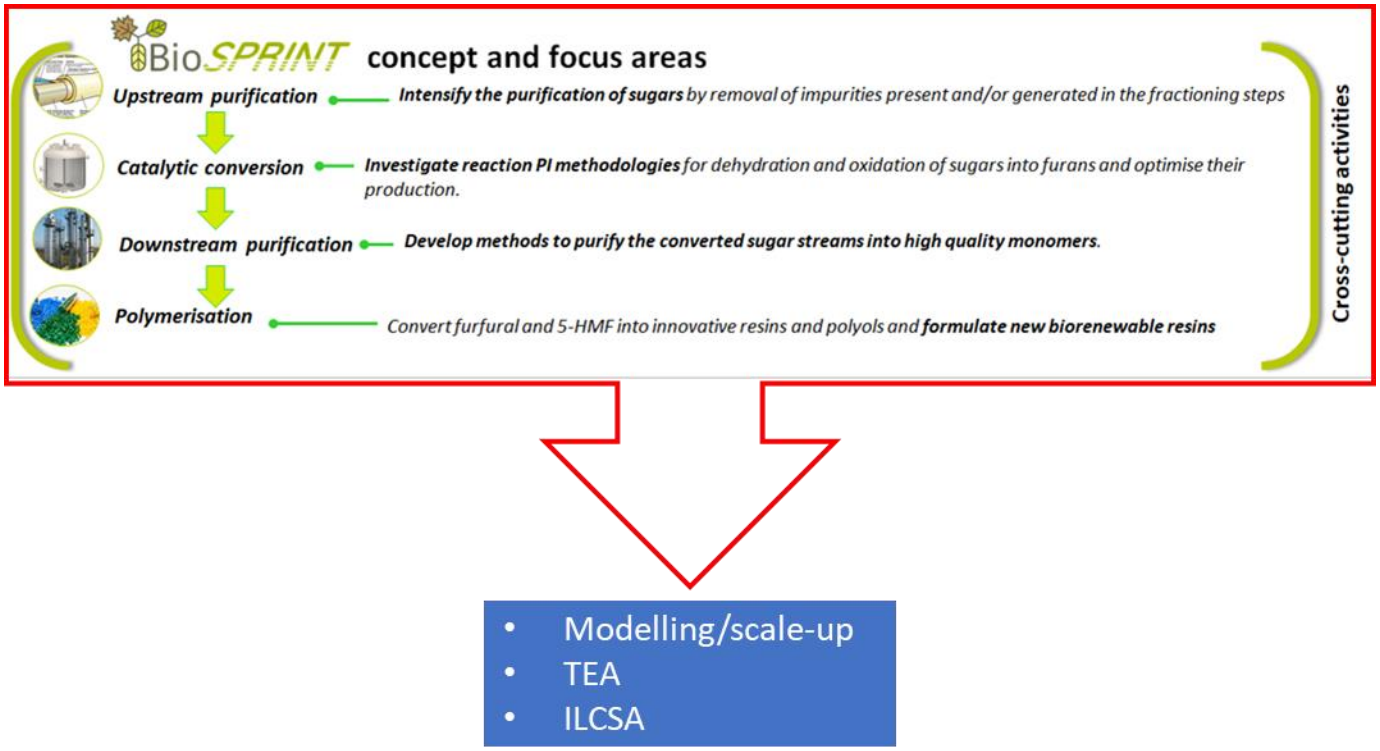

Figure 1. Technical focus areas of BioSPRINT.

3. Scientific and/or technological achievements in M1-M25

3.1 Work Package 1 – Intensification of Upstream Purification

Two technologies have been investigated for the upstream purification of the HMC feedstocks: membrane separation and concentration, and spinning disc technologies.

The applicability of different nanofiltration and reverse osmosis membranes has been studied for two real/industrial HMC streams. The experimental work indicates that the target recovery (80—90%) and purity (70—80%) of sugars can be achieved or even exceeded with right decision on membranes, operating conditions and diafiltration strategies. The efficient removal of particular short-chain acids, furan derivatives and solvents from the HMC streams was also observed. Membrane systems show limited concentration abilities, with volumetric concentration factor around 2.5 before excessive fouling of the membranes occurs. It seems, however, that the fouling is reversible. Membranes have also been proposed together with SDR to build different kind of novel upstream purification cascades.

The spinning disc technology has been applied in the recovery of lignin by precipitation via solvent evaporation of black liquor emanating from an Organsolv HMC stream. Up to *ca*. 95% of the original lignin present is recoverable as solids precipitate under carefully selected operating conditions of temperature, flowrate, and disc speed to maximise disc residence time. The precipitates are mainly in the form of individual particles with minor agglomeration for easier downstream processing. They have also been determined to be of high purity. Thermal energy consumption is 2× lower and lignin yield is at least one order of magnitude higher on the spinning disc compared to conventional evaporative technologies such as CSTRs combined with falling film evaporators. This performance enhancement is attributed to significantly higher rates of heat and mass transfer achieved in the flow of micrometers-thick films of black liquor on the rotating disc. Short and controllable residence times on the spinning disc allowed the integrity of individual particles to be maintained, even at high operating temperatures.

Sugar precipitation from HMC feedstock has also been studied by antisolvent precipitation technique in the spinning disc reactor. The preferred solvent, identified from a solvent screening study, results in ~35% recovery of C5 and C6 monomeric and oligomeric sugars in solid precipitates of high purity (>95%) obtained in a single stage mixing process on the spinning disc. The antisolvent to feed stream ratio contacting each other on the spinning disc had the greatest impact on sugar recovery of all parameters studied in the mixing process.

The spinning disc technology development is supported by the application of process systems engineering (PSE) tools including computational fluid dynamics (CFD) which is being applied to analyze in detail the film distribution on the disc as it is spinning. A detailed knowledge and understanding of the wavy motion of the fluids on the disc is being developed. Two types of discs are being tested in WP1, a smooth or flat surface and a grooved surface to enhance the mixing and heat and mass transfer between the fluids. CFD analyzes the application of SDR technology for both the sugar and lignin precipitation case studies which involves two different mechanisms for precipitation as highlighted above. For the membrane systems, a multi-level modeling approach has been established. Detailed flow models of a lab-scale membrane unit have been developed with CFD. A dynamic diafiltration plant model has also been developed to evaluate the parameter uncertainties and control strategies of a multi-stage membrane plant.

Additionally, the control strategies of the lab scale and the scale up of the system have been developed based on UOULU's and UNEW's experience in their membrane and spinning disc technologies and processes. In the SDR system variables like disc speed, flowrate and temperature were identified as key process parameters and are recommended to be monitored and controlled. In the membrane system the controlled variables are the pressure, the feed flow rate and the temperature.

3.2 Work package 2 –Intensification of Catalytic Conversion

Machine learning methods for catalyst evaluation have been implemented and in particular, procedures suitable for variable and modelling method selection have been developed allowing a very good accuracy in the predictions of catalyst performance with model structures as simple as possible. The importance of the used catalyst descriptors and reaction conditions were analysed in comparison to the predicted conversions, selectivities, and yields, allowing findings of non-obvious descriptors with high predictive power. Machine learning methods were first studied with a literature dataset[6] and later adapted to the BioSPRINT case. In particular, machine learning and high-throughput screening methods had not been seen in use for development of catalysts for sugar dehydration reactions, which means there is a large chance of discovering optimised operating conditions.

Catalyst formulations for scalable and adaptable acid catalysts have been developed and information is being prepared for full publication soon. Comprehensive kinetic models for C5 and C6 sugar dehydration using commercial zeolite catalysts have been generated and showed promising results in terms of modelling of conversion of complex feedstock streams. Until now, the kinetic models in literature were mostly for single sugars, and mainly for xylose and glucose. In BioSPRINT, this work has taken a step forward by developing and experimentally validating models with multiple sugars in mixed streams.

Development of continuous processing has been initiated through mixing studies using intensified reactor technology, enabling a preliminary understanding of the mechanics of mixing in the system and acquisition of data required for energy balances and scale-up. Mixing processes in the specific type of reactor selected for this project have not been published in literature yet, and the aim is to obtain data to complete mechanical energy balances for process scale up. Therefore, the work undertaken in BioSPRINT with this technology will enable better understanding and characterization of its performance in liquid/liquid and liquid/solid processes.

PSE tools are applied in WP2 to analyze the behavior of the catalytic reactor. CFD is used to analyze the flow patterns. The CFD studies analyse not only flow and operating conditions, but also agitator design. Additionally, the CFD analyses have shown that the flow in the cells can considered laminar with disruption due to agitation. The reactor is beign modelled utilizing the kinetic data provided by the project to evaluate dynamic responses and develop process control methodologies. This dynamic model is being used to identify the scale-up challenges and control strategies that can successfully maximise yield and selectivity.

3.3 Work package 3 – Intensification of Downstream Purification

In WP3, where the downstream separation after the catalytic conversion of C5 and C6 sugars in bi-phasic system will take place, the separation of both organic and aqueous phase is being investigated in hydrocyclones, a very promising advanced method, not reported to date for such an application.

[6] Uusitalo, P.; Sorsa, A.; Russo Abegão, F.; Ohenoja, M.; Ruusunen, M. Systematic Data-Driven Modeling of Bimetallic Catalyst Performance for the Hydrogenation of 5-Ethoxymethylfurfural with Variable Selection and Regularization. Ind. Eng. Chem. Res. 2022, 61 (14), 4752–4762. https://doi.org/10.1021/acs.iecr.1c03995

Design methodologies for hydrocyclones have been tested and validated experimentally for a comprehensive development and optimisation of the process in the ongoing project tasks. CFD tools have been applied to analyze the flows within the hydrocyclones and good progress has been made to match the performance achieved experimentally.

Whist individual separation technologies already exist for C5 and C6 furan-based derivatives, separation of combined furfural and 5-HMF are at the moment non-existent. Also, the choice of the solvent for the furfural extraction is extensively discussed in the literature, but less information is available for 5-HMF extraction, and for the separation and purification of both furan products by the same solvent. Suitable solvents were identified based on reaction and/or separation performance, paying particular attention to partion coefficients, separation and reaction selectivities, and to sustainability and toxicity issues. Screening by experiments has narrowed down the selection of solvent candidates that can be used for effective removal (more than 90%) of furfural and 5-HMF from both streams leaving the bi-phasic reactor as well as recycling of the solvent back to the reactor. The separation processes were modelled and simulated to further narrow down the selection before refining the selection through reaction and separation experimental work. By using combined continuous distillation and extraction, furfural and 5-HMF of high purity or a mixture of both can be achieved. Further work is still necessary to complete the modelling part of both systems and evaluate potential benefits in terms of purity, energy consumption and costs. The results will be disseminated through international journal publications in due time.

3.4 Work package 4 – Development of Biorenewable Resins and Polymerisation Intensification

In WP4, where furfural and 5-HMF are being converted into polymers for industrial use (in particular, resoles, novolacs and Mannich polyols), the most promising and innovative achievements so far are:

i) REACH compliant furfural-based novolacs, which have been synthesised through a chain growth polymerisation protocol, which allows to finely control molecular weight and kinetic of polymerisation and avoid undesired homo-polymerisation of furfural;

ii) the synthesis of Mannich polyols from furfural;

iii) the formulation of polyurethane foams using furfural-based polyols, with comparable or even improved properties with respect to industrial references.

WP4: REACH compliant furfural-based novolacs. There are no furfural-based novolacs in commerce and the scientific literature reports only very limited examples[7,8,9]. In WP4, a chain growth polymerisation protocol has been developed to control the reactivity of furfural (thus avoiding undesired homo-polymerisation). After obtaining a cardanol-based novolac using appropriate amounts of paraformaldehyde, the controlled addition of furfural allows to synthesise novolacs which are "polymers" according to REACH, therefore compliant to the regulation and exempt from registration.

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[7] R. Srivastava, D. Srivastava, "Utilization of Renewable Resources in the Synthesis of Novolac Polymers: Studies on its Structural and Curing Characteristics", Int. J. Res. Rev. Eng. Sci. Technol., 2(2): 22-25, 2013.

[8] D.K. Mishra, B.K. Mishra, S. Lenka, P.L. Nayak "Polymers from renewable resources. VII: Thermal properties of the semi-interpenetrating polymer networks composed of castor oil polyurethanes and cardanol-furfural resin", Polym. Engin. Sci., 36(8): 1047-1051, 1996.

[9] S. Gopalakrishnan, R. Sujatha "Synthesis and thermal properties of polyurethanes from Cardanol-furfural resin", J. Chem. Pharm. Res., 2(3):193-205, 2010.

Mannich polyols from furfural. Although furfural, according to literature, can be used in the synthesis of benzoxazines and hot melt adhesives, there are no examples in literature of furfural-derived Mannich polyols. They have been obtained in WP4 using a two-step method *i.e.* firstly converting furfural and an alkanolamine into an oxazolidine, and then reacting the latter with a phenolic compound to obtain a Mannich polyol. This route circumvents the issues related to the reactivity of furfural (which leads to formation of solid products insoluble in any solvent) and allows the recovery of Mannich polyols, which can be used in polyurethane foams.

Polyurethane foams obtained from furfural-based polyols. Although examples of furan and tannic-furanic foams are shown in literature, there is no evidence of polyurethane foams containing furfural derivatives. In WP4, furfural-based liquid novolacs and Mannich polyols have been combined to formulate rigid polyurethane foams which, apart from the characteristic color, have good overall performance and have enhanced properties (e.g. compression strength) with respect to industrial references.

3.5 Work package 5 – Validation, Process Simulation & Integration

In the initial stages of BioSPRINT, we have determined the boundaries of the existing/reference and improved biorefineries that are in line with definitions, settings and system descriptions outlined in task T6.1 of WP6. A new concept of a conventional (*i.e.* non-intensified) biorefinery process is introduced, representing the reference against which the improved/process-intensified biorefinery will be benchmarked. The non-intensified biorefinery assumes the use of proven technologies (TRL5 and above) to convert hemicellulosic sugars into value products such as polyols, resins and polymers. Within this concept, we identified processing steps that are within the scope of BioSPRINT's research and development activities: i) upstream purification of hemicellulose stream, ii) catalytic conversion of sugars to furans, iii) downstream processing of furans, iv) conversion of furans to value products (resins and polymers). In subtask ST5.1.1.2, a comprehensive AspenPlus simulation model has been developed for the conventional biorefinery, including all the above-listed process steps. A detailed description of the conventional biorefinery model, including technology assumptions, is given in the deliverable D5.1.

Modelling and simulation activities have been extended to the existing biorefineries cases to include the pretreatment of lignocellulosic material and the production of hemicellulose streams. We considered three biorefinery cases that use different pretreatment technologies and lignocellulosic feedstocks: i) UPM's biorefining HMC stream, ii) FhG's Organosolv fractionation of hardwood, and iii) an industrial steam explosion process of wheat straw. The simulation models of the existing biorefinery cases are used to characterise the three different hemicellulose streams in terms of composition, volume, economic value, and allocated emissions. Such information will be a key input for the integrated lifecycle sustainability assessment to be undertaken in WP6.

Furthermore, we have defined the key performance indicators (KPIs) to compare and assess the conventional and improved biorefinery's cost-effectiveness, productivity, energy, and water usage. KPIs for the conventional biorefinery have been calculated and reported in D5.1.

We have also initiated modelling of the intensified biorefinery. Process information and experimental data were collected from partners in WP1 to WP4 and compiled into Aspen model inputs. In the coming period, MAT will build the simulation model of the intensified biorefinery and calculate KPI.

3.6 Work package 6 – Integrated Life Cycle Sustainability Assessment (ILCSA)

The main aim of WP 6 is a thorough Integrated Life Cycle Sustainability Assessment (ILCSA) considering effects of the Process Intensification (PI) methods selected in BioSPRINT along the HMC valorisation value chain. Work performed within this workpackage included definitions, settings and the description of the qualitative system as shown in deliverable D6.1. A reference case has been defined, considering a 'standard biorefinery' (which delivers the HMC stream) and a 'conventional biorefinery', where the HMC stream is currently not valorised and is considered as an underutilized stream. In contrast to the standard biorefinery, the HMC stream is valorised by the BioSPRINT process. Three different standard biorefineries/case studies will be considered since each of them uses a different pre-treatment approach and/or biomass feedstock. The description of the systems under study is continuously updated and revised based on further BioSPRINT technology development. Discussions on definitions and boundaries are done in coordination with WP5 and the other technical work packages. A final report on descriptions of definitions and settings is to be finalised in M32. Currently, data on mass and energy balances of the existing and conventional biorefineries are being extracted and collected from the simulation models. Furthermore, the development of a concept for adaptations to IFEU's Life Cycle Inventory (LCI) databases was initiated in order to devise the LCA methodology and develop data exchange templates for mass and energy balances.

4. Technical Challenges and Solutions

Foreseen and unforeseen risks are monitored and mitigated throughout the project through a risk management strategy outlined in task 8.3, which is regularly updated and was already shared as part of deliverable reports D8.4 and D8.5. In addition to this, during the course of the first 24 months of the project, technical challenges were encountered which required, or are undergoing, resolution. Table 2 below highlights the key technical challenges and solution strategies encountered for each work package.

Table 2: Technical Challenges and Solutions in period M1-M25

WP	Technical Challenge/Issue	Solution Strategy	Status†
1	Permeate flux decrease caused by fouling.	Membrane fouling studies were performed. The results show that the main cause for fouling was concentration polarisation which can be reduced by increasing the cross-flow velocity of the feed. Additionally, the initial permeate flux was recovered after water flush and chemical cleaning.	S
1	Sugar recovery by antisolvent precipitation method on the spinning disc is not as high as desirable.	The scope for implementing a hybrid process to recover and recirculate the feedstream in a multistage mixing process is being considered.	O
2	Too high conversion in aqueous phase led to low selectivity towards furans.	Overactive catalysts formulations were considered and catalyst loadings adjusted in order to tune activity to a suitable range of conversations so differentiation of catalyst performance in aqueous reactions could be performed.	S
2	Reaction is very sensitive to temperature control making operation inside local exhaust ventilation cabinets difficult to control due to variable air flows and external surfaces cooling.	Control tests were conducted with dehydration of synthetic sugar mixtures in the different platforms with parallel reactors to identify the variability of results. Where issues were identified, the systems were equipped with additional insultation and air shielding devices. Tests are carried out in duplicate to identify spurious experiments and outliers. Machine Learning and design of experiments methods for high-throughput experiments are data-rich and therefore able to cope with the occasional spurious data points without compromising the quality of the models obtained.	S
2	Analytical methods published in literature had not been tested for all	Literature was extensively researched to identify different chromatography methods suitable to analyse reactants, products and by-products in	S

	compounds and solvents of choice.	different solvent systems. Methods were screened and redeveloped to suit the specific solvent chemistries in use. When required, gas chromatography methods were also used in addition to liquid chromatography methods.	
2	Mixing and heat transfer data for the reactor technology selected were not available in literature.	Mixing and heat transer studies have been/are being carried out to fully characterise the flows, mixing and heat transfer time-scales, and to obtain quantitative data for energy balances and heat transfer coefficients. CFD simulations are being validated against experimental work to obtain robust scale up models.	O
2/3	Large number of solvent candidates for reaction and/or separation potentially leading to too time consuming experimentation.	Solvent screening and selecitone exercises were first carried out by researching partion data in literature, determined partion coefficients experimentally and simulating separation potential. A limited selection of solvents has been idenfied and is not being experimentally tested and implemented in processed development.	S
3	In the study of flows in the prototype hydrocyclones, one of the main challenges is the difficulty in quantifying wall roughness.	A CFD parameterisation study on wall roughness has been performed to understand/minimise its influence in the hydrocyclone.	S
4	Lower reactivity of 5HMF and furfural has been observed in the formation of phenolic resins and polyols.	Changes in formulations, reagent concentrations and operating conditions in conventional and intensified reactor system have been/are being implemented to enable reasonable polymerisation rates.	O
4	Self-polymerisation of 5HMF and furfural hinders high yields of phenolic resins and polyols.	Reaction conditions have to be carefully chosen to limit self-polymerisation.	O

†S – Successfully completed, U-Unsuccessful/Aborted/Incomplete, O-Ongoing, F-Forecasted.

5. Conclusions and Further Actions

The technological achievements in BioSPRINT in the first 25 months represent a step forward in developing innovative technologies and processes for application in the European bio-based industry. These developments have demonstrated potential for reducing the environmental impact compared to existing technologies and processes. For example, the development of more efficient separation and purification steps for isolating sugars has a direct impact on reducing the environmental footprint of such processes by reducing processing time, energy, water and solvents consumption. Furthermore, the small processing volumes in the intensified technologies such as SDR in WP1 and an alternative reactor technology in WP2 will contribute to enhanced process safety when handling hazardous and flammable materials such as ethanol and other organic solvents.

The mentioned contributions highlight the need for interdisciplinary approaches for the innovative development of processing technologies. In relation to resource depletion and environmental impacts, a number of technologies that are currently being investigated in BioSPRINT have the potential to reduce energy consumption and physical footprints in comparison to conventional technologies, such as stirred reaction vessels, decanters for liquid-liquid separation, shell and tube heat exchangers, and batch reactors, amongst others. By intensifying heat and mass transfer processes, these innovative technologies enable processes to rely on lower driving forces, and therefore, smaller temperature and concentration differences can be employed, whilst still achieving high transfer and reaction rates.

The extractive-reaction process of converting sugars into furan-based derivatives under bi-phasic extraction conditions being studied in WP2 has the potential to reduce formation of by-products, leading to higher yields, and thus reducing the needs for further purification downstream. Due to intensification of the liquid-liquid separation using hydrocyclone (centrifugal force) instead of conventional liquid-liquid decanter, the separation time will be reduced as well as a size of the separation unit, allowing enhanced recovery of reactive intermediates.

Continuous processing is envisaged, starting from the upstream purification steps, passing through the bi-phasic reactor, intensified liquid-liquid separation, and along to the downstream purification, namely distillation of the organic phase with extraction of the furans (furfural, 5-HMF). Process intensified separation in a continuous mode with smaller processing volumes and milder process conditions, compared to batch processes, will have positive effect on process safety. Therefore, the inventories of hazardous substances in the decanter units and in the distillation columns will be substantially reduced.

Significant potential exists for reducing or eliminating process steps which will have process, economic and environmental benefits. These and further social, economic and environmental potentials will be investigated in detail in work package 6 depending on the achievements of the technical work packages 1-4.

Future work planned until M48 includes the following:

WP1: In the upcoming stages of the project, feedstock variability analysis and immediate upstream implications will be reported in D1.2. Integrated SDR and membrane processing for upstream purification for both FhG and UPM streams will be assessed for transfer to scale up studies in WP5. Water and solvent recovery strategies for the intensified upstream purification and concentration will also be evaluated in order to reduce waste and improve the sustainability of the upstream processes.

Upstream PSE and scale-up recommendations will be made based on the separation unit's virtual twin, identified disturbance & control variables and instrumentation setup.

WP2: Future work in WP2 will focus on completing the development of hybrid extractive-reaction processes and intensification using continuous reactor technologies. Solvent selection will be narrowed down and integrated with the separation processes in WP3. CFD and dynamic modelling of the intensified process will be completed to create methods for accurate prediction of scale-up challenges and optimise process control.

WP3: Further experimental work will test the processing sequences simulated and predicted to validate downstream separation models and methodologies. Energy intensification and integration will be investigated, and identification of downstream scale-up challenges for the intensified technologies will be carried out.

WP4: Further experimental development of new polymers based on 5HMF and furfural converted and purified in WP2 and WP3 will be implemented. Strategies for intensified polymerisations of novolac polyols and resins will also be assessed and implemented experimentally.

WP5: In the next project period, the focus will be on finalising the economic assessment of the conventional biorefinery for input in WP6 and updating the economic KPIs. The improved/intensified biorefinery simulation model will also be developed and the KPIs will be calculated. The new biorefinery concept will be optimised based on techno-economic viability. A desktop study will be carried out to assess the replicability of the PI concept with other industrially relevant biorefinery streams. Finally, all PI technologies developed in BioSPRINT will be integrated and validated at TRL 5 using an actual hemicellulose stream obtained from UPM's process.

WP6: The qualitative system description of the system under study is continuously updated and will be finalised in M33 described in D6.2. Adaptions of LCI database and collection of mass and energy balances will be finalised and activities on techno- and socio-economic assessment completed. LCA models will be build and based on these information assessments on environmental, socio- and techno-economic assessment will be done. Furthermore, product and process safety assessments will be completed. Finally, the Integrated Life Cycle Sustainability Assessment (ILCSA) will be presented.