

Newsletter

Direct Thermochemical Liquefaction



PyNe 51

July 2022

Finally! The first Task 34 meeting in presence after more than 2 years took place in Espoo, Finland, May 23-24th, 2022. Even if it was a hybrid meeting, it was good to see all again.

Participating countries were represented by their teamleads or their representatives offline (Daniele Castello/Denmark, Christian Lindfors/Finland, Axel Funke/Germany, Bert

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van de Beld/ The Netherlands, Michael Thorson/USA) and online (Benjamin Bronson/Canada, Francois Collard/New Zealand) and additionally Anja Oasmaa as special guest, which has strongly influenced Task 34 over the last years.

The task meeting started with a discussion of Task 34 Work Packages. All leads for work packages from the IEA proposal have been discussed and assigned among the meeting participants. The dissemination activities will be conducted by the Task lead/ KIT.

Afterwards, we considered how to further increase the visibility of Task 34 and talked about the lessons learned from the past triennium.

After a lunch break, we went on an interesting lab tour at VTT. After discussions on the past and upcoming Round Robin and its evaluation, the first day ended with an informal meeting, where the discussions and exchanges of the whole day were further deepened.

On the second day a workshop with the Finnish stakeholders was scheduled, which was very interesting and informative and a good opportunity for dissemination and networking.

Christian Lindfors has attracted many interesting Finnish stakeholders from research and industry for a joint workshop and the organization for this event was great. Find out more about the workshop in this newsletter issue

Although Task 34 has worked well and successfully in the last 3 years and has also kept in regular contact through video conferences and emails, after this physical meeting a great wind of motivation was felt, pointing to a successful coming triennium.

Then I would like to draw your attention to our homepage
<https://task34.ieabioenergy.com>.

whose renewal we were able to complete in the 1st quarter of 2022.

You will of course find our newsletter there, but also a database in which you can search for the various articles.

In addition, we have listed the Round Robins of Task 34 of the last years in another database, you can find it here:

<https://task34.ieabioenergy.com/round-robin-archive-2/>

Furthermore, you can find out what Task 34 is doing or has done, the various reports can be found under publications.

Not to forget, you can find here:

<https://task34.ieabioenergy.com/publications/pyrolysis-demoplant-database/> a database of demoplants.

Task 34 is continuing its effort to supply you with relevant DTL information and keep up the work to advance sustainable DTL applications.

Yours sincerely,
Alexandra Böhm
Task assistant

Changes in IEA Bioenergy Task 34

Some Task 34 participants changed over the course of the triennium, while others had to leave at the beginning of this year due to changes in country memberships. At this point we unfortunately have to say goodbye to some NTLs but at the same time we are also very happy to welcome new faces and with them a breath of fresh air in our team.

We are sad that we had to say goodbye to:



Linda Sandström,
Rise, Sweden



Kai Toven
Rise PFI,
Norway



Paul Bennett,
Scion, NZ



Kirk Torr,
Scion, NZ



Justin Billings,
PNNL, USA

We are really looking forward to working with you and send you a warm welcome



Michael Thorson,
PNNL, USA



Pramod Kumar,
HPCL, India



Francois Collard,
Scion, NZ

We wish all newcomers a lot of fun with us. To all those who had to leave, we wish you continued success. We hope we will stay in contact and see each other again soon, whether within Task 34 or in other places. Thank you for your work.

Ablative fast pyrolysis demonstration project in California

Brian Gannon, Biogas Energy, California, USA
Christoph Eusterbrock, Bioenergy Concept, Germany
Dietrich Meier, thermophil international, Germany

Working to address the impacts of climate change, the state of California funded a demonstration of fast pyrolysis bio-oil production that has been operating since May 2021. Biogas Energy, www.biogas-energy.com, received the grant to install and operate a 500kg/h pyrolysis facility supplied by Bioenergy Concept, www.bioenergy-concept.com, a German renewable energy technology developer.

The project has successfully made bio-oil from construction & demolition wood waste, forest residues, and orchard grindings. Disposal of these materials is a high priority for California, as the state pushes aggressively to reduce GHG emissions from biomass sources while replacing fossil fuels. Forest fires are tragically a common occurrence in California, where drought and bark beetle infestation have caused millions of tree mortalities and created a tinderbox of fire fuel. The promise of converting that waste biomass into a fuel is pushing the state to adopt pyrolysis as a

pathway to develop new waste-to-energy projects.

Samples of bio-oil were analyzed by California State University, Chico, giving baseline performance data of the equipment and confirming that the pyrolysis reactor functions to specification. Biochar samples from each feedstock were analyzed using International Biochar Initiative standards, producing exceptional results; i.e. 85% organic carbon, 4.6% ash, 360kg/cbm bulk density, 1.5% moisture content.

The 500kg/hr pyrolysis system proved easy to operate with two technicians; one monitoring the computer controls and the other performing tasks like moving materials, checking feedstock flow, etc. The system handles feedstock variability effectively, with the front-end grinder accepting everything from logs to pallets to forest grindings, and the dryer reducing moisture content using surplus heat from the reactor. The ablative fast pyrolysis reactor is robust and efficient



Fig. 1: Fast pyrolysis biooil production

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with automated controls facilitating continuous operation. This ability to process a wide variety of feedstocks with near-total automation will expedite the adoption of the technology in commercial applications.

Dr. Dietrich Meier of thermophil international, Hamburg, provided ongoing support for the project, collaborating with the CSU Chico research team and optimizing equipment performance. Dr. Meier's collaboration with Bioenergy Concept CEO Christoph Eusterbrock resulted in the delivery of a pyrolysis package that promises to address the enormous problems facing California today.

Building upon this successful project, Biogas Energy is developing several projects in California focusing initially on wood waste but progressing towards pyrolysis of biosolids and plastics as the technology is improved. Biogas Energy is also seeking to develop partnerships with technology providers and research laboratories to create marketable products for the bio-oil, such as sustainable aviation fuel, maritime fuel, and green asphalt.



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HP-HTL technology - pathway for waste valorization and plastic circular economy

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Introduction

Future of energy is always an intriguing question for the scientists as they see the depletion of natural resources around the world, forcing them to look for alternatives like non-fossilized sources and unconventional fuels. As such tremendous research is being performed toward production of waste derived fuels as a reliable replacement for drop-in fuels.

Waste streams from agriculture and municipalities may have varying compositions and properties, are however a valuable resource, that are not being fully utilized. More than 2 billion tonnes of municipal solid waste along with over 1.3 billion tonnes of food waste is being produced annually. With an annual growth rate of 10%, about 300 million tonnes of plastic waste is generated yearly. These mind-boggling figures are nearly going to double by 2050 [1] [2] [3].

Currently, US, China, India, Europe are the largest consumers and producers of plastic. On account of their intrinsic properties of easy handling, good durability and ease of shaping and sizing, plastics turned out to be an inseparable entity for mankind. Plastic is literally found everywhere, right from packaging, consumer commodities, medical equipment, electrical and electronics, to buildings, constructions, automobiles, textiles, and furniture. Thousands of tonnes of waste derived plastic is dumped every day and being a non-biodegradable material, is piling up on the earth's floor and water-bodies, thereby contributing to the existing pollution.

Literature studies earlier reported that by 2017 an estimate of about 7 billion tonnes of plastic ended up as waste out of which a mere 10% was treated, thereby leaving the rest 80% piled up in landfills. [4] As a feasible approach to treating this waste, pyrolysis of plastic to useful chemicals is explored. However,

pyrolysis requires high temperature and a thoroughly dried feedstock, that indirectly shoots up the operating/processing costs. Hydrothermal liquefaction however, is a better approach for treating wastes as it eliminates the drying stages and is able to handle moisture laden wastes and slurry wastes. Since plastic pyrolysis was the usual technique treating plastics, literature on hydrothermal liquefaction of waste plastic is very limited.

Moreover, municipal solid waste comprises of a blend of plastics combined with agricultural wastes, kitchen wastes and so on. Segregating this waste into their respective groups and treating is practically not feasible. As such a co-processing technique that involves treating and further upgrading to superior products is the need of the hour.

Thermochemical techniques like hydro-pyrolysis, hydrothermal liquefaction, and liquefaction in organic solvents for the production of fuels and specialty chemicals are some of the promising routes for valorization of ligno-cellulosic biomass, algae, sewage waste and other carbon related wastes.

Herein, we report a process for the catalytic hydrothermal co-liquefaction of ligno-cellulosic biomass and plastics under conditions of about 300-350 deg C and 20-25 MPa by utilizing water as solvent and in the presence of a homogeneous proprietary transition-metal based catalyst.

By using the process detailed in this article, the decomposition temperature of plastics can be reduced, and a bio-crude oil with yield over 40% combined with an oxygenate content of less than 20% and free of sulphur, whilst giving some superior synergistic interactions between feedstock (municipal solid waste, or ligno-cellulosic biomass or food waste etc.,) and plastics can be produced.

Table 1: HTL @ Timeline

1920s	First reported inception of the concept of biomass liquefaction. Heated water along with an alkali buffering agent is used to produce liquid oil.
1970s	A biomass HTL process was developed at the Pittsburgh Energy Research Center (PERC), a pilot plant was demonstrated at the Albany Biomass Liquefaction Experimental Facility at Albany, Oregon, at a scale of 100 kg/h
1970s	HTL process was developed at Lawrence Berkeley Laboratory (LBL) that used an acid hydrolysis pre-treatment step with water as the reaction media. Scale 100 kg/h.
1985	Scientists developed correlations for the key parameters such as temperature, pressure, time, catalyst and concluded that the changes in the governing parameters has a significant impact on the product output
1994	authors introduced the extruder – feeder system to pump the high viscous liquids for the liquefaction process
1995	developed a scale up of HTL of sewage sludge operating at temperature of 300 °C and pressure of 10 MPa
1999	HTL opens up new possibilities for combined fuel production and waste water treatment in densely populated areas where large amounts of wet waste materials are available
>2000	Reputed international research laboratories started exploring the liquefaction reaction parameters that includes residence time, reaction temperature, catalyst selection and loading
>2015	Immense amount of the work pertaining to hydrothermal liquefaction of wet and dry biomass through different techniques has been carried out by many researchers
>2020	Several demonstration/commercial plants setup in Australia, Canada, Europe, India

HTL @ HPGRDC

HP Green R&D Centre (HPGRDC), Bengaluru, a research & development wing of Hindustan Petroleum Corporation Limited (HPCL), India, is actively working on several technologies for the valorization of biomass. The authors of this article are involved in developing thermochemical routes (esp. liquefaction, pyrolysis) of utilizing biomass to generate valuable chemicals and fuels. HPGRDC has setup state-of-the art biomass processing plant for the process and technology development.

Literature studies mostly report about a single biomass waste stream for the liquefaction process. HPGRDC has employed a co-processing approach for making biocrude from wastes. Different co-processing combinations were experimented. Co-processing of plastic with biomass in water in

the presence of proprietary catalyst enhanced the quality of the final biocrude and improve the solvation of biomass and plastic wastes.

The raw-materials biomass – rice straw and rice husk were sourced from nearby fields and plastic feedstock (Polypropylene, LLDPE) were sourced from local vendors in Bengaluru, India. No pretreatment was carried out for the experiments. Feed was used as it is in all runs. Liquefaction was conducted at 300-350 deg C and 20-25 MPa in all cases. Solid char phase is separated via filtration. To the liquid phase, an extraction solvent is added to separate the aqueous and oil phases. And further the oil phase is processed in rotavapor to remove the solvent and obtain final biocrude product. Figure 1, 2, 3 show the bio-crude yields at different conditions and co-processing combinations. Initial approaches were aimed at liquefying the individual wastes in the

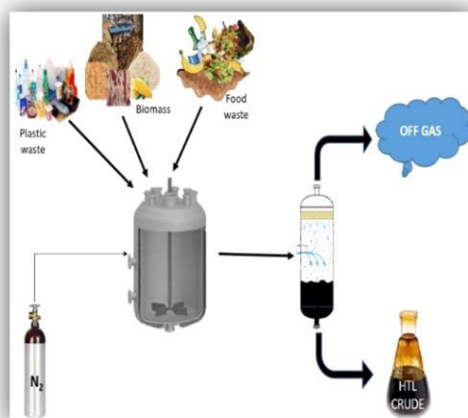


Fig. 1: Hydrothermal Liquefaction of Biomass process flow scheme

presence of in-house developed proprietary catalyst.

Figure (2) shows the oil yields for solvothermal process for producing high quality fuels and chemicals from a feedstock comprising of waste biomass (rice straw / rice husk) liquefaction in the presence and absence of catalysts.

Experiments B1 and B2 represent rice husk in the absence and presence of a proprietary transition metal based catalyst respectively. Whereas experiments B3 and B4 represent rice straw in the absence and presence of a proprietary transition metal based catalyst respectively. As shown in the figure, oil yields showed improvement with the addition of catalyst. Analysis of the bio-crude was done in GC-MS and it was found that the major

components primarily consisted of C8-C13 aromatics, phenolics and hydrocarbons. Elemental analysis of bio-crude reported oxygenates to be less than 15%.

Figure (3) shows the oil yields for solvothermal process for producing high quality fuels and chemicals from a feedstock comprising waste plastics in the presence and absence of catalysts. Experiments P1 and P2 represent polypropylene waste in the absence and presence of a proprietary transition metal based catalyst respectively. Whereas experiments P3 and P4 represent polyethylene terephthalate (PET) waste in the absence and presence of a proprietary transition metal based catalyst respectively.

As shown in the figure, oil yields showed improvement with the addition of catalyst. It

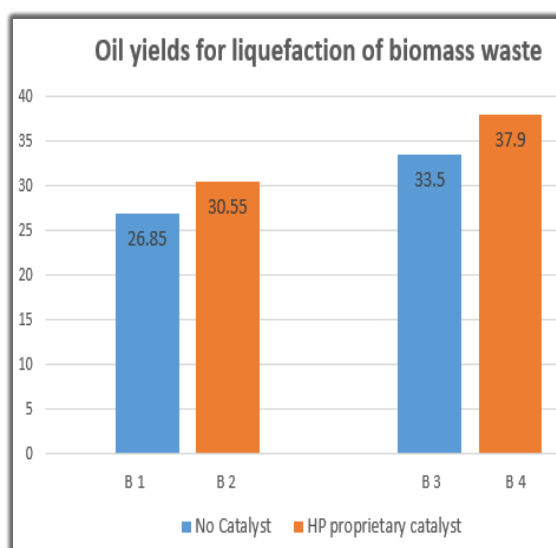


Fig 2: Oil yields of polypropylene waste and polyethylene terephthalate in presence/absence of catalyst

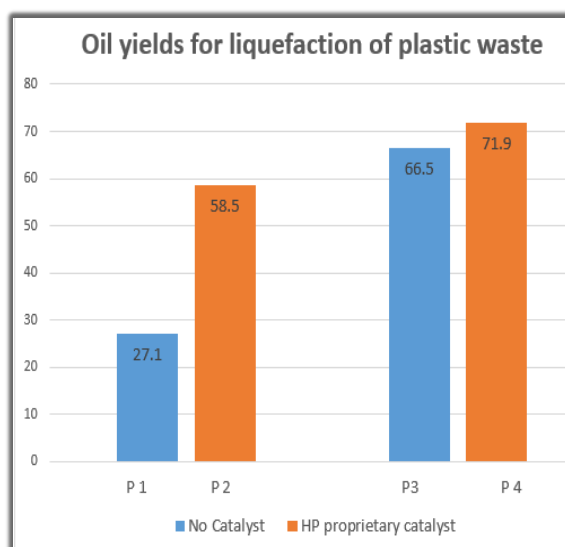


Fig 3: Oil yields of rice husk and rice straw with plastic in presence/absence of catalyst

was seen that the presence of catalyst has enhanced the solvation and gave yields over 50% in case of PP and PET as well. Analysis of the bio-crude was done in GC-MS and it was found that the major components primarily consisted olefins, benzene derivatives and alcohols.

Individual liquefaction of wastes in the presence of catalyst gave satisfying yields. Further exploration was done for co-liquefaction of biomass and plastic wastes.

Figure (4) shows the oil yields for solvothermal process for producing high quality fuels and chemicals from a feedstock comprising of 1:1 ratio of biomass and waste plastics for liquefaction in the presence and absence of catalysts. Experiments M1 and M2 represent rice straw with polypropylene in the absence

and presence of a proprietary transition metal based catalyst respectively. Whereas experiments M3 and M4 represent rice husk with polypropylene in the absence and presence of a proprietary transition metal based catalyst respectively. As shown in the figure, oil yields showed improvement with the addition of catalyst. Analysis of the bio-crude was done in GC-MS and it was found that the major components primarily consisted of olefins and naphthenes.

Conclusion

HPGRDC has setup state-of-the-art equipment in par with the latest technology for conducting research in the areas of waste valorization, especially in the biomass upgradation field via thermochemical methods like hydrothermal liquefaction and

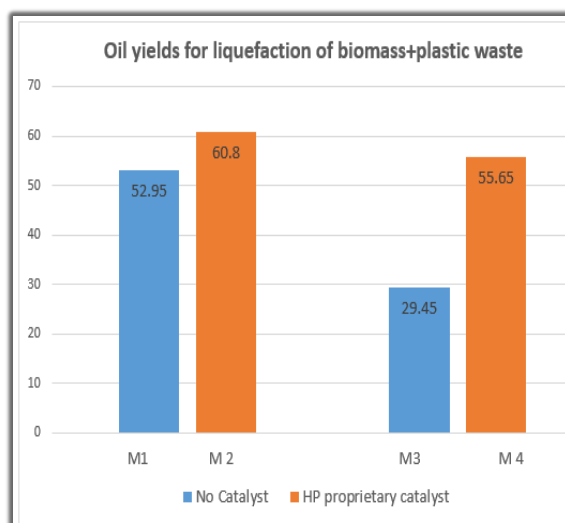


Fig 4: Oil yields of rice husk and rice straw with plastic in presence/absence of catalyst

pyrolysis. The in-house prepared proprietary catalyst can effectively enhance the solvability of hard-to-degrade plastics along with biomass in water at relatively lower temperatures to produce biocrude containing less oxygenates and valuable chemicals that can be used as fuel blends. The process presented in the article overcomes the usual difficulties and environmental issues caused by the currently employed techniques for disposal or treating of municipal wastes, plastic wastes and agricultural wastes. At the same time, this process is an economic path for the recycling of hard-to-degrade plastic wastes.

As such it avoids the need to separate different kinds of wastes for individual treatment, thereby providing the advantage of co-processing municipal, agricultural-waste with plastics and hence reduces the carbon footprint.

As a way forward to the existing research, HPGRDC is planning for a larger-scale continuous catalytic hydrothermal liquefaction of biomass technology, and more experimentation towards converting biocrude to useful transportation fuels and also looking forward to collaborate with member countries of Task 34 for developing waste valorization routes to manufacture drop-in fuels.

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Important commercial steps towards bio-based gasoline from pyrolysis oil

Linda Sandström, Rise Energy Technology, Sweden

The Swedish refinery company Preem will soon initiate the production of biobased gasoline, produced from sawdust, at the Lysekil refinery. In the coming two years, 50 000 ton pyrolysis oil will be processed at the refinery.

Preem has a vision to produce 5 million cubic meters of renewable fuels in 2030. Just recently the company announced that it sharpens its climate goal by ten years, meaning that it aims to be climate neutral along the whole value chain already by 2035.

- We sharpen our climate goals because we want to, we can, and we must. Our generation has a responsibility towards coming generations to ensure a sustainable transition says Magnus Heimborg, CEO at Preem, in a press release.

The production of forest-based renewable gasoline is one important step to reach this goal. Large scale production of bio-diesel is already a reality through the production of

HVO (hydrotreated vegetable oils), while the production of bio-gasoline is today much less developed.

At the Pyrocell facility in Gävle, Sweden, saw dust is converted to pyrolysis oil. Pyrocell is a joint venture between the refinery company Preem and the wood industry company Setra. The pyrolysis plant, situated at the Setra Kastet saw mill, was constructed with support from the Swedish Environmental Protection Agency and has just been commissioned; production started in September 2021. The facility is designed to produce about 25 000 ton of biobased pyrolysis oil per year and the target is that all this pyrolysis oil will be upgraded to renewable fuel at Preem's refinery in Lysekil, Sweden.

– Pyrocell is a unique effort that enables a sustainable value chain from forest to tank. We replace the fossil oil and contribute to an increased share of renewables in transportation fuels, says Pontus Friberg, working chairman of the board at Pyrocell.



Fig. 1: Picture of the Pyrocell pyrolysis plant

The first short-term test of feeding pyrolysis oil to the fluidised catalytic cracker (FCC), where the upgrading to bio-gasoline is to be performed, has recently been performed at the Lysekil refinery in a demonstration project supported by the Swedish Energy Agency. In this test a few hundred tons of pyrolysis oil was treated as a mixture of 2 percent pyrolysis oil and 98 percent fossil feedstock.

Deeper evaluation of the performed test will also reveal the resulting renewable content of the produced gasoline. This since the cracker also produces gas and diesel. The fate of the green carbon atoms will be tracked using carbon-14 analysis of the product streams from the cracker.

– The test lasted a couple of days and showed that the quality of the produced gasoline and diesel was not affected by the pyrolysis oil, says Katarina Persson, development engineer at Preem.

Bio-based pyrolysis oil differs from fossil oil in many ways, for instance through a high viscosity, a high density and a low pH. It is also not miscible with fossil feedstock, due to the high oxygen content. The inorganic content also differs from fossil oils; bio-based pyrolysis oil contains metals such as sodium, potassium, magnesium and calcium, which can cause catalyst deactivation.

– Further research and development is needed to enable large quantities of pyrolysis oil to be processed at the refinery while maintaining a low rate of catalyst deactivation, says Olov Öhrman, R&D manager at Preem.

The pyrolysis oil also contains chlorides, which can cause corrosion of the equipment at the refinery. Preem has therefore installed monitoring equipment to track how the equipment is affected by the pyrolysis oil.

The next step in this venture is to perform a long-term test where 50 000 ton of pyrolysis oil will be processed during two years. This corresponds to two years of pyrolysis oil production from the Pyrocell pyrolysis plant

and will result in approximately 25 000 ton of renewable products, mainly gasoline. If the results are successful, Preem sees a bright future ahead.

- The market exists and is just waiting. There is a huge demand for bio-gasoline, says Katarina Persson.



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Demonstration of the fractionation of fast pyrolysis bio-oil by liquid-liquid extraction on pilot scale

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Biomass is a valuable, sustainable feedstock for the production of chemicals and materials. For the optimal utilization of bio-resources, fractionation on the basis of functionalities is often preferred. The approach presented here is applying a two-step, thermochemical fractionation process (TCF). In the first step the biomass feedstock is converted by fast pyrolysis into a liquid product. Fast pyrolysis is characterized by the fast and short heating of the biomass resulting in a partial depolymerisation of the feedstock, but retaining the key chemical functionalities in the liquid. In the second step the liquid is separated at low temperature by a multistep, liquid-liquid extraction process. The fractions - pyrolytic sugars, pyrolytic lignin and extractives- consist of components derived from the de-polymerization of extractives, cellulose, hemicellulose and lignin, see Fig. 1. Subsequently, each of the fractions can be used as a raw-material in bio-based materials or as a feedstock for further (electro-)chemical or biological conversion.

This TCF approach is an alternative to pyrolysis with staged or fractional condensation. In that case the production of the various fractions is an integral part of the pyrolysis process by operating a number of condensers at different temperatures. A separation of components on the basis of boiling point/vapor pressure will be obtained. This is principally different from the TCF process in which a separation on the basis of solubility in specific solvents is achieved, and a better separation on basis of functionalities can be expected in case of TCF.

Pilot plant design & Operation

Prior to designing and building the pilot unit extensive testing was performed on lab- and bench scale. The bench-scale unit has an input capacity of 12.5 kg of FPBO per hour and is operated in continuous flow. All the FPBO fractions produced in this unit were used for initial product development. Furthermore, operation of the unit provided the required data and experience to enable the design of the pilot plant.

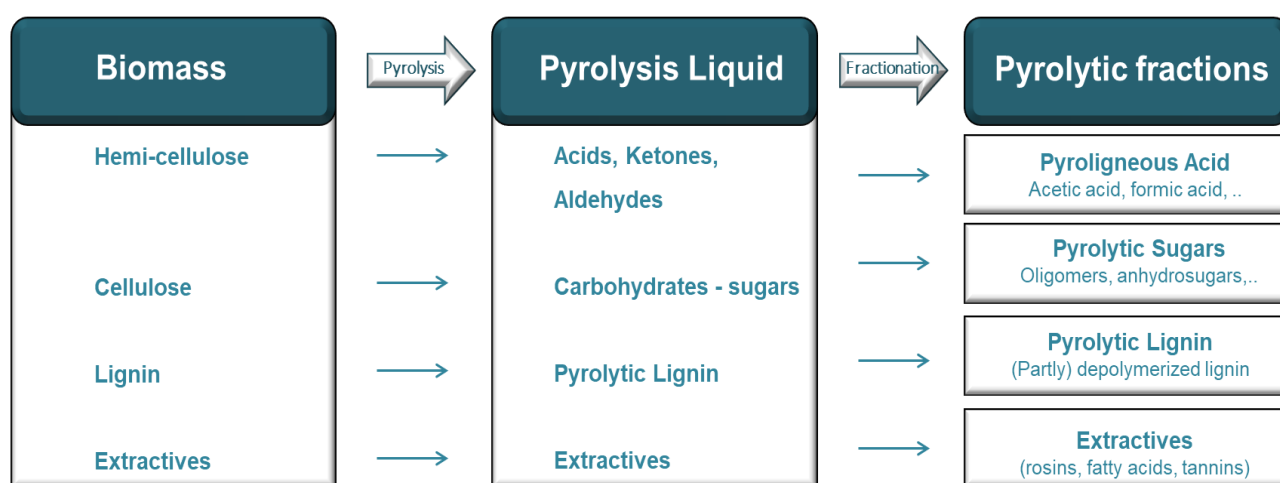


Fig. 1: Illustration of the 2-step Thermo Chemical Fractionation (TCF) concept.

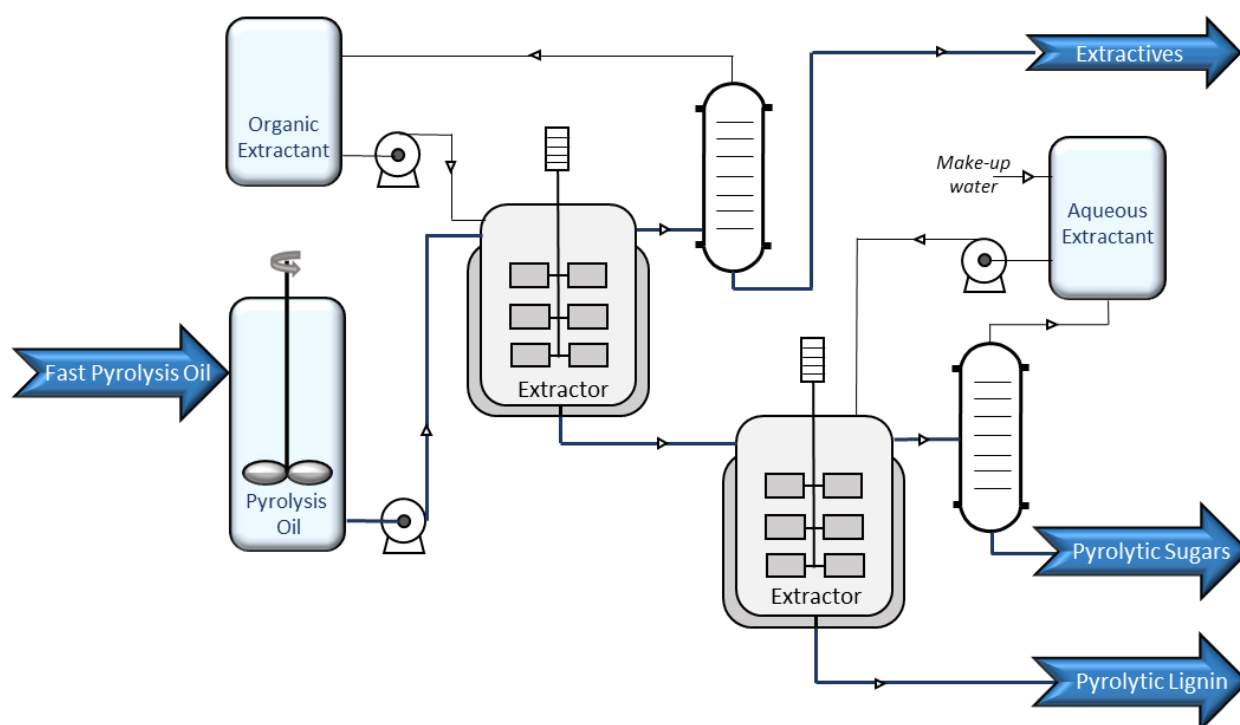


Fig. 2: Simplified process flow diagram of the TCF process

The design capacity of the pilot unit equals 3 ton of FPBO input per day, which is roughly a ten-fold scale-up of the bench-scale unit. It includes two extraction units; the first one to remove extractives from the oil, the second one to separate the pyrolysis oil in a lignin and sugar fraction. Extractants (both organic and water) can be recovered and eventually recycled to the process. The sugar fraction can be further concentrated, and the liquid lignin can be divided in a solid lignin and a phenolic fraction. A simplified process flow diagram is shown in Fig. 2; a photo of the unit in Fig. 3.

The pilot plant was commissioned by the end of 2018, and several campaigns were conducted in 2019-2021. Normally, only one or two unit operations are operated simultaneously, and recovery of extractives is only performed if needed for the specific FPBO (i.e. rich in extractives). Currently, actual capacity achieved for the lignin/sugar separation is 120 kg/h which is very close to the design capacity. The products obtained in the pilot plant show very similar properties compared to those from the bench scale unit.

Product properties

Table 1 shows typical properties of the products obtained by the fractionation of the FPBO. Primary products are aqueous sugars, liquid lignin and extractives; concentrated sugars, solid lignin and phenolics are only obtained by aftertreatment of the primary products.

Applications

The fractions produced in TCF can be used in a range of applications and dedicated development work was performed within the *Bio4Products* project. Inhere, the fractions were a.o. used to partly replace fossil based raw-materials in various phenol-based resins, and as an active component in formulations for wood modification and in foundry resins. Recently, the *NewWave* project has started; in this project the fractions are used as sustainable raw materials in four existing manufacturing lines, largely replacing fossil based raw materials and substituting toxic chemicals like formaldehyde and creosote. The manufacturing lines will produce engineered wood panels, furan base-

Table 1: Indicative properties of the products obtained by the fractionation of FPBO

			CAS: 2414605-13-1		CAS: 2411004-28-7 / 2411004-20-9			
			Aqueous	Concentrated	Liquid	Solid	Phenolics	
C	wt%	44	18	49	54	71	63	76
H	wt%	7	10	7	7	6	8	10
Carbon residue	wt%	17	5	22	30	36	5	2
Water content	wt%	23	63	5	13	<1	2	<1
MW	g/mol	-	-	-	~850	~1300	-	-
LHV	MJ/kg	16				28	26	35

chemicals, polyols and polyurethanes, and modified/engineered wood with the goal to enhance the sustainability of building materials in the construction industry. The four manufacturing are interlinked, and output from one line will further improve the sustainability of the other. Besides the production of sustainable products, water re-use, end-of-life recycling options and efficient use of byproducts are an integral part of the

project and will result in an almost zero-waste approach.

To enable or simplify product research and development at reasonable scale, REACH (1-10 t/y) and PPORD registrations have been filed for the lignin and sugar fractions. Product research on extractives have been limited so far and only tests at lab scale have been carried out.



Fig 3: Photo of the FPBO fractionation pilot plant at BTG.

Acknowledgement

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Upgrading of biomass pyrolysis volatiles to syngas a using 3D-printed lattice-structured catalyst in an electrified reformer

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Introduction

Pyrolysis is a process in which organic matter thermochemically decomposes under anaerobic conditions.

Pyrolysis of biomass forms a variation of condensable gases (commonly referred to as bio-oil or tar), non-condensable gases and biochar [1]. Biochar is a valuable bioproduct of pyrolysis, it can be used to replace carbon positive fossil fuels and as fertilizer [2].

The combination of bio-oil and condensable gases is referred to as pyrolysis volatiles, they contain hundreds of organic compounds and water. The presence of water and oxygenated compounds create undesired properties like low heating value, incomplete volatility and acidity.

Further treatment (i.e. upgrading) is thus required for future applications, like fuel and H₂ production [3]. In terms of H₂ production, there is plenty of H₂ in the pyrolysis volatiles (bio-oil and non-condensable gases), that can be cracked (reformed) for high H₂ and CO production, which is the key element to obtain a highly selective H₂ production. The renewable H₂ is used as an energy carrier and as a feedstock in many industrial processes (such as in the chemical, energy and transportation sector) and can play an essential role into the CO₂ neutral industrial processes.

Catalytic steam reforming

CSR refers to a series of catalytic reactions between the pyrolysis volatiles and steam that produce syngas, a combination of H₂, CO and CO₂. Steam reforming is of endothermic nature, therefore hydrogen production is favored by high temperatures. [3].

In the conventional technology, the catalyst (usually, a Nickel-based material) is loaded into a number of tubes placed inside a furnace. A large-scale industrial reformer consists of several dozens of tubular tubes with a length exceeding 10 meters, the combustion (energy source) must occur at a higher temperature than the reaction temperature to generate the necessary inward heat flux into the reformer. This is due to the poor heat transfer phenomena lead uneven heat distribution inside the reactor [4].

The mass transport phenomena show also limitations in catalytic reactors, low effectiveness factor and diffusion-limited reactions due to laminar flow and bad mixing have been reported in the literature [5, 6]. In addition, coke formation coming from decomposition of pyrolysis volatiles deposits on the surface of the catalyst, blocking the active sites and thus leading to catalyst deactivation.

3D-printed catalysts

Additive manufacturing (3D printing technology) can aid some of these bottlenecks. Lattices are defined by an elementary structure (e.g. octet- and cubic-truss) that is repeated in the 3D space. Lattice structures can be optimized for high surface area (lattice design), and good heat transfer properties (material choice), which are desired in heterogeneous catalysis [7, 8]. The lattice structure promotes better mixing, remedying the diffusion-governed mass transport in the conventional technology [9, 6].

A material with good thermal properties can be chosen in order to address the heat

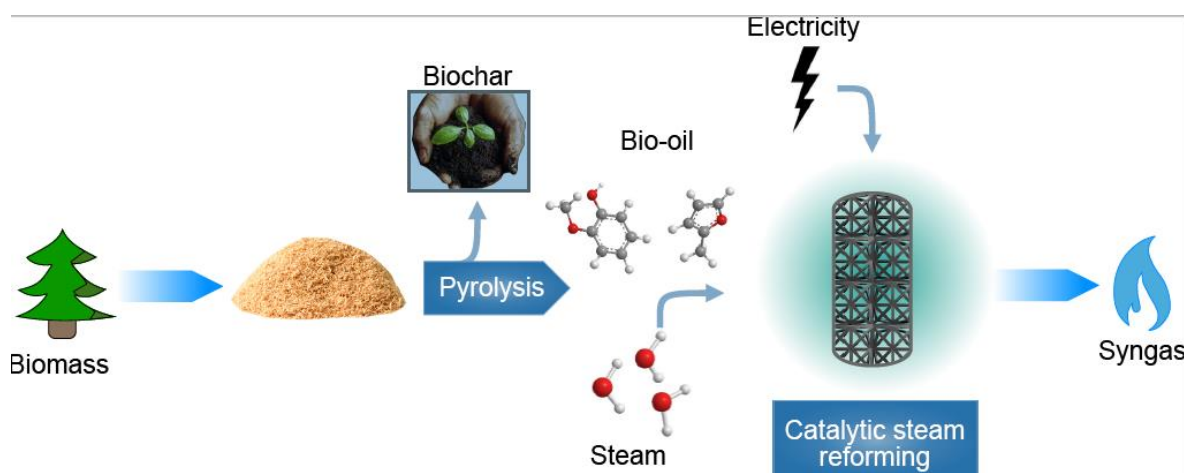


Fig. 1: From biomass to syngas using electrified catalysis

transfer limitations and the high porosity results in low pressure drop [10]. In a further attempt to increase the energy efficiency, direct electrification has shown potential to result in better heat flux and therefore higher energy efficiency [4, 11, 12]

Process design

The biggest challenges for the implementation of syngas production from renewables are catalyst deactivation (which affects the syngas production) and low energy efficiency (which affects energy costs). The use of biomass pyrolysis volatiles together with the electrification of the process has the potential to become a carbon neutral process for rich H_2 syngas production with high energy efficiency.

The presented process has the potential to take syngas production from biomass into industrial scale.

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Liquid organic hydrogen carrier for upgrading of pyrolysis oil – Insights from a lab-scale batch reactor study with dibenzyltoluol

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High potential can be seen in liquified biomass for substitution of fossil resources for material and energetic utilisation. Some challenges exist during production and utilisation of liquified biomass. High oxygen content and the complexity of the biomass structure leads to oxygen-rich oils with a heterogeneous composition (Behrendt et al. 2006). For valorisation of biomass-derived oils in conventional industrial pathways it is necessary to adjust these oils in their properties to the fossil resources. This includes to reduce the amounts of heteroatoms like oxygen and the complexity with different substance classes.

During further upgrading processing of the oils, polymerization can occur due to acids and oxygen-rich functional groups, resulting in the formation of coke. This can be prevented by the utilisation of hydrogen as an upgrading agent. Liquid hydrogen-donating systems, like tetrahydronaphthalin and formic acid, are able to decrease the repolymerization and coke formation during pyrolysis oil upgrading (Churin 1991, Kleinert et al. 2008).

A further alternative to provide hydrogen for an upgrading reaction is the utilisation of liquid organic hydrogen carrier system (LOHC). Dibenzyltoluol (DBT), a widely used heat transfer oil, is recommended as a typical LOHC-system with a high thermal stability (up to 395°C). Towards tetrahydronaphthalin, DBT is able to deliver more hydrogen with previous complete hydrogenation to perhydro-DBT (Preuster et al. 2017). The advantages of LOHC utilisation, comparing pressurized hydrogen, are the easier handling and safe storage in combination with temperature stability, lesser toxicity and the higher availability of hydrogen (Arlt et al. 2017). The transfer hydrogenation and dehydrogenation to provide hydrogen in various degrees were shown in literature with ruthenium (150°C, 50 bar) and platinum (270 – 310°C, atmospheric conditions) catalysts as well (Brückner et al. 2014, Müller et al. 2016). Besides the complete hydrogenated structure (18H-DBT) several intermediated states with different degree of hydrogenation exists (6H and 12H). The degree can be estimated and precalculated by the density of the DBT/perhydro-DBT system.

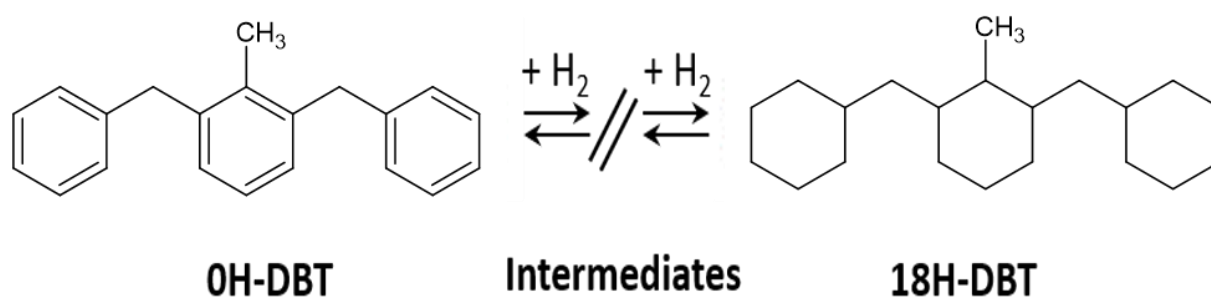


Fig. 1: Transfer hydrogenation of Dibenzyltoluene isomer via intermediate stages

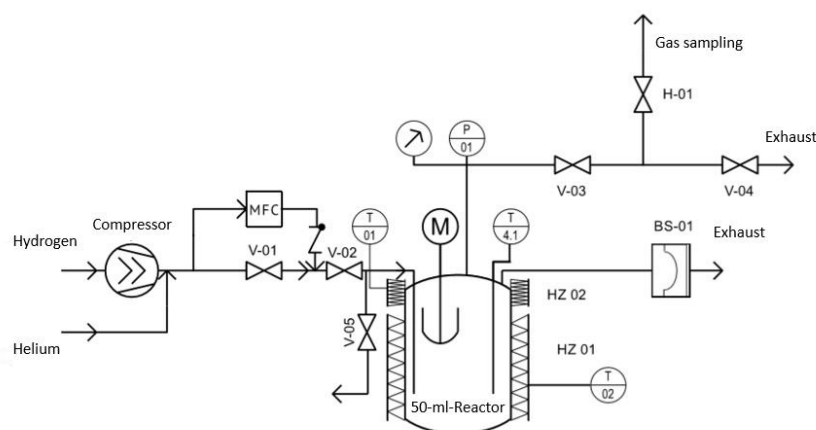


Fig. 2: Process flow chart of the lab-scale batch equipment (left) and 50-ml-reactor (Paar Instruments, series 4560) with heating wristbands, steering system and gas inlet and exhaust at the reactor head (right)

We firstly studied the hydrodeoxygenation (HDO) of model substances like guaiacol and a slow pyrolysis oil from beech wood (proFagus GmbH, Bodenfelde). Larger amount of perhydro-DBT were produced in a 600-ml-batch autoclave system (data not shown) with platinum (3.2 mm pellets, 0.5 wt. % platinum on Al_2O_3). The HDO experiments were carried out in a 50-ml-batch system, as shown in figure 2). The system was filled with perhydro-DBT, either with guaiacol or slow pyrolysis oil (produced from beech, proFagus GmbH, Bodenfelde) in a ratio of near 4 (DBT/guaiacol, pyrolysis oil, w/w). The platinum catalyst was added in an approximate ratio of 0.025 mol % regarding to the weighted DBT. A typical run was conducted at several temperatures (230°C; 290°C; 350°C) with a reaction time of 4 h. The degree of dehydrogenation and reaction mixture composition were estimated by GC-MS/FID measurements.

Figure 3 (left) shows the degree of dehydrogenation of the perhydro-DBT under consideration of reaction temperature and the used substances (guaiacol, pyrolysis oil resp.). At temperatures under 350°C the degree of hydrogenation is similar to the blank trials without model substances. At 350°C the degree of hydrogenation decreases to two-thirds of the degree of blank trials.

Considering the molar ratio of guaiacol to perhydro-DBT of 1:3 (mol/mol) then a large part of the guaiacol reacts by decreasing

hydrogenation degree. In contrast, the degree of hydrogenation of the pyrolysis oil behave not so consistent. Although exactly the same ratios between pyrolysis oil, perhydro-DBT and catalyst were weighed the degree of hydrogenation can not be calculated as simply as at the guaiacol case. Nevertheless, the degree decreases in the same extent.

Therefore, it can be assumed that larger portions of the oil have also been hydrogenated. This can also be concluded from the composition of low molecular weight reaction products (Figure 3 right). Typical hydrogenation products of guaiacol are cyclohexane, benzene, 1,2-dimethoxybenzene and the isomers of cresol. According to the pyrolysis oil the single substances were summarized to the classes hydrocarbons, phenols, guaiacols. Further classes result from the composition of the pyrolysis oil. It can be observed, that oxygen-containing substances disappears during the hydrogenation treatment of the pyrolysis oil. Therefore, the amount of hydrocarbons and benzenes increase at the composition of the processed pyrolysis oil. Supports also the side reaction of DBT degradation and coke formation. As an undesired side effect, the production of coke increases with the treatment temperature and the utilisation of solid platinum catalyst. Simultaneously an increase in the amount of DBT-degradation products were observed. There is the assumption that the commercially available platinum catalyst.

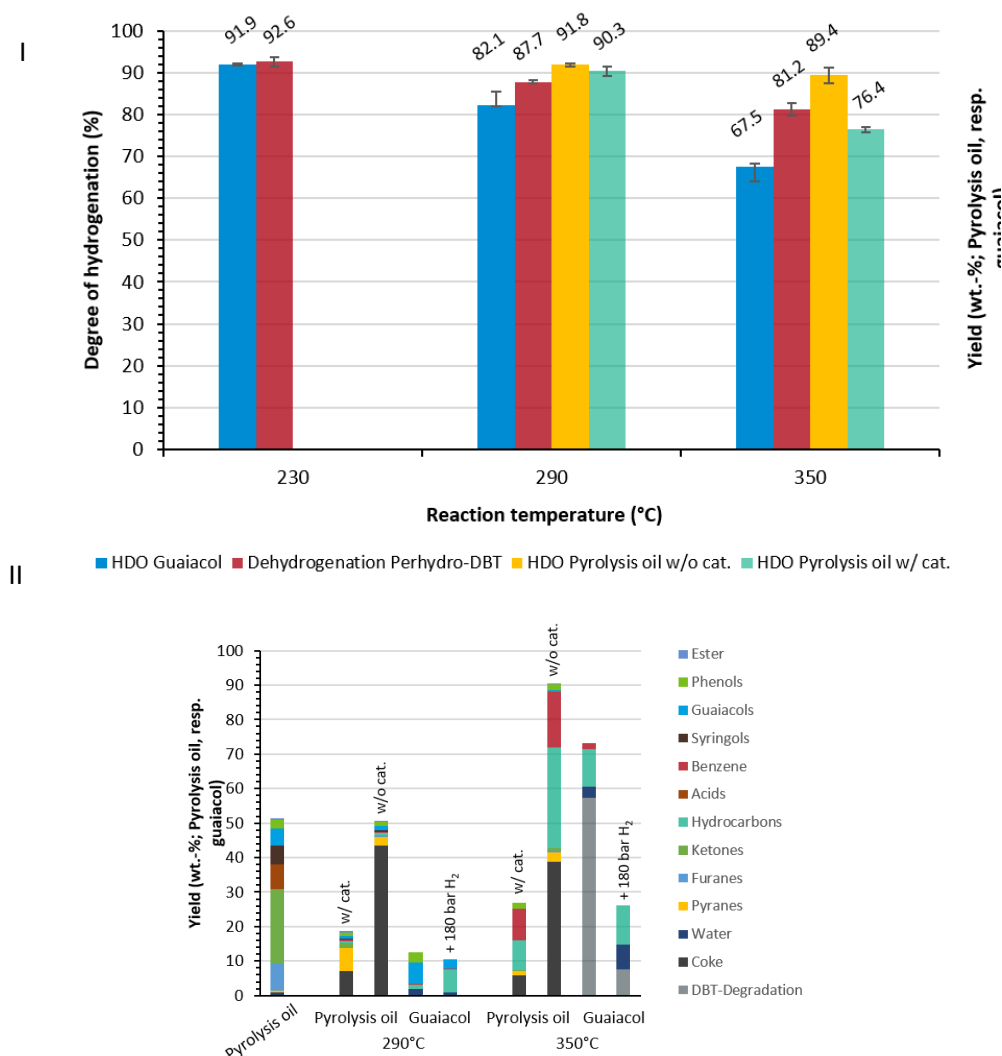


Fig. 3: Degree of hydrogenation (I) and composition of reaction products (II, based on weight portion) of guaiacol and pyrolysis oil hydrogenation trials with DBT (molar ratio 1:3, mol/mol) and platinum catalyst (0.0025 mol Pt per mol guaiacol and 0.17 g per g pyrolysis oil respectively)

Other studies show higher selectivity of tailored catalysts without DBT degradation in remarkable extent. However, the hydrogenation functionality of DBT in combination with a platinum catalyst can be observed at the formation of hydrocarbons and benzenes during the treatment with model compounds. Obviously, the application of an additional hydrogen pressure reduces the formation of degradation products.

Certain degree of hydrogenation of the trials with additional hydrogen show similar decreases during the treatment (data not shown). However, the following challenges still arise from the following results. During

the trials with the model substances only small amounts of the reaction products could be recovered and analysed. Furthermore, DBT degradation products are difficult to distinguish from the hydrogenation products of model substances and pyrolysis oil as well.

Therefore, the next work would have to consist of an adjustment of the reactor design to capture the volatile reaction products for balancing and analysis in higher extent. Subsequent trials with tailored catalysts and marked DBT are also conceivable.

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Pyro2022 in Ghent/ Belgium

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The International Conference on Analytical and Applied Pyrolysis finally is back to real life with its 23rd edition taking place in Ghent/ Belgium from May 15th to 19th – the Pyro2022. After an involuntary switch to digital format in 2020 due to the ongoing CoVid pandemic (itself an excellent event), Wolter Prins, Frederik Ronsse and their organizing committee managed to put together a wonderful event that was indeed a worthy start back to face to face presentations and discussions! Two parallel sessions over the course of four full days were realized in the honourable ‘Het Pand’ of Ghent University, with the possibility to join in online for those that could not make it in person.

Not much to surprise, the contributions reflected the strong dynamics in the field of plastic pyrolysis which can be observed since several years, both in terms of fundamental research in the field of reactions and kinetics as well as developments towards application. Nonetheless, biomass pyrolysis was well represented with many excellent talks and there was also a dedicated session on hydrothermal processes. There is still a very strong (and much needed!) movement to

elucidate reaction networks along with confining associated analytical methods. Naturally, this also helps developing in-depth characterization of liquid products from thermochemical liquefaction.

Another observation relevant to the field of DTL are the ongoing activities to develop reactions and catalysts for the production of value added compounds *via* (fast) pyrolysis – as nicely pointed out by Erik Heeres and Qiang Lu – cumulating in the concept of a pyrolysis biorefinery. Many promising candidates and pathways for their production have been identified and further validation at higher TRL is ongoing.

Interesting to note is also the increased use of quantum chemistry for the investigation of pyrolysis reaction networks. William ‘Bill’ Green gave an inspiring talk on how to move forward in this field; not by giving an answer but rather with an excellent overview of the possibilities and limits the different approaches have, pointing out to potential synergies. This view was also reflected by other contributions and we can be sure to see more from this field in the coming years.





Finally, Robbie Venderbosch provided a fascinating overview of the history of fast pyrolysis technology developments. While giving credits to many (if not all?!) of the important pioneers in this field, he came to a somewhat surprising conclusion looking at the mere numbers. Out of the five endeavours that made it to commercial fast pyrolysis demonstration units, only two failed. This positive evaluation makes fast pyrolysis a very successful technology!

fruitful talks and discussions that were enabled by finally meeting in person again.



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Ghent is a wonderful place to stay and the organizing team did an excellent job, which added to the great atmosphere of the conference. Not to forget about all those



Task 34 Workshop May 24th 2022, Espoo, Finland

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VTT, Finland

Task 34 workshop was held on 25th May 2022 in Espoo, Finland, connected to the task meeting. Axel Funke from KIT Germany gave an introduction to IEA Bioenergy and especially to Task 34.

Joakim Autio from Valmet presented “Developments and applications of fast pyrolysis technologies”. Valmet is leading developer and supplier of process technologies, automation and services for the pulp, paper, and energy industries. Valmet is active in several fields, like biomass and waste gasification, thermal and catalytic pyrolysis, lignin extraction, pellets, and biomass pretreatment. The target in biomass pyrolysis is to prepare product for co-processing in conventional refinery or biorefinery to replace fossil feedstocks. In LignoCat project industrial piloting is ongoing for Valmet pyrolyzer with catalytic upgrading. In pilot tests at VTT efficiency and yields have been in expected range, total energy efficiency over 80% including side stream utilization and energy yield over 40% to biocrude. Valmet has also cooperation with Circa on biochemical production with pyrolysis. [1]. Valmet will offer the main machinery and equipment and actively contribute to the development of Circa’s production processes of bio-based

renewable solvents. The bio-based solvents will replace harmful petroleum-based solvents used in a wide variety of industries.

to Valmet's leading company project through the EU’s Recovery and Resilience Facility The Finnish national funding agency Business Finland has granted a total of EUR 20 million (RRF). In addition to that Valmet plans to invest EUR 40 million into the project during the upcoming four-year period.

The leading company projects are Business Finland's largest single funding decisions. If successful, the projects will lead to outgrowths on a business level that will be worth several billion euros, industrial investments, significant impact on employment, growth in export earnings and tax revenue, and positive effects on the environment. In addition, Business Finland plans to fund the partners of each leading company with EUR 50 million. A total of 12 leading projects including Valmet, and Neste are currently underway. [2]

Ahmad Kalantar from Neste presented Neste's activities in the renewable field “Transforming refineries to minimise carbon footprint and maximise handprint”. The target is to substitute fossil oil i.e. by electric cars,



Fig 1.: Participants of the task 34 Workshop

increase biofuel use, and Power to X. Renewable H₂ and Power-to-X include refinery transformation. Vision for 2035 include renewable H₂ units, Power to X, CCU, CCS, waste heat to district heating, and reduced net CO₂ emissions. Energy sources as renewable and recycled hydrocarbons for road transportation, aviation, polymers, and chemicals are under change. Presently they are fatty acids (HVO), within 3-5 years recycled waste plastics, within 5-10 years lignocellulosics and municipal solid waste, and after ten years algae and Power-to-X. Waste and residue feedstocks availability is expected to grow to over 35 Mton/a by 2030. Neste's focus is on scalable solutions with lignocellulosic fuels to chemicals, and materials. Algae and MSW will be scalable feedstocks for aviation fuels, renewable H₂ and PtX. Impurities and bio-oil stability in co-refining are the biggest research challenges.

Gerhard Muggen from BTG Bioliquids told about BTG Bioliquids Pyrolysis Technology. BTG Bioliquids is technology provider and have delivered the Empyro plant in Netherlands, GreenFuelNordic (GFN) plant in Finland and Pyrocell in Sweden. The Empyro plant started the bio-oil production already in March 2015, while GFN started the bio-oil production December 2020 and Pyrocell in September 2021. GFN Oy is a biorefining company based in Finland. They use sawdust as feedstock for pyrolysis. Future plan is to use shavings and other potential bark-free feedstocks.

Present application for the oil is heating and later transportation fuels and feedstocks for chemical industry. GFN has the plan to increase the capacity by 7 new plants from BTG Bioliquids and Technip in future. Pyrocell, a joint venture of Setra and Preem, also uses sawdust as feedstock. FPBO production capacity is 24,000 t/a. Preem Lysekil refinery will co-process the FPBO produced at Pyrocell. Until now, they have co-fed more than 2,000 tons, which corresponds to 1-3 % biofeed. There were questions on co-processing details. The nozzles for co-processing can be delivered by UoP (USA), or Technip (NL). The temperature at nozzles is not an issue.

Vaibhav Agrawal from Tampere University of Applied Science presented their EU H2020 project BL2F [3] (2020-23) on hydrothermal Liquefaction. The project targets to develop a high-quality, drop-in biofuel from kraft black liquor to be used in the aviation and shipping sectors. Research areas include 1) Salt separation, 2) Solids/salt handling, 3) Water handling, 4) Gas handling, and 5) Hydrogen production. Bio-oil has been produced in a continuous unit (feeding rate about 13 l/h) with reactor size up to about 3 liters. Salt removal and product analyses are on-going. Industrial partners are The Navigator Company (NVG), Ranido, Valmet, Neste, and Avinor. Research partners are: PSI, SINTEF, SINTEF-ER, Tampere University, KIT, Brunel University London, and VTT.

Riinu Walls from Meriaura told on sustainable Shipping. Meriaura Group is a Finnish family owned group of companies that focuses on shipping and shipping related services. Their cargo volume is 3 million tons annually, that equals 3 % of total volumes of Finnish foreign trade sea transportation. The mission is to provide solution-oriented transport service to partners in special and project cargoes, and products of bio-, circular economy. As the first sea carrier, Meriaura has introduced almost carbon neutral transport contracts. Meriaura EcoVoy-concept is based on low-consumption, biofuel-powered EcoCoaster vessels. When using waste-based biofuel, transports' lifecycle emissions are 92 – 96% lower than with fossil fuels. Meriaura exploits side streams of foodstuff industry, such as crude fish oil, along with already once utilized vegetable oils. The production is audited by DNV GL and has a sustainability system certificate. Green maritime future target is to reach 100% carbon neutrality. The concept is based on hybrid propulsion that combines bio-oil and battery technology. Meriaura is involved in a common reed project in Finland coordinated by Turku University of Applied Science. The interests in the project are common reed logistics, substrates for greenhouses, biochar and better conditions of the archipelago and climate. In Finland there is around 100 000 hectares of common reed. The amount of reed is approximately 5 tonns

of dry substance per hectare. Dry reed is most suitable for pyrolysis and biochar use. Meriaura also participates Finnish national project BIOFLEX on “Production of sustainable storable liquid fuels for flexible power generation and marine transport (2020-2024)” [4] lead by VTT. The feedstocks are biomass and waste plastics and technology used is fast pyrolysis. The overall goal of the BioFlex project is to develop a least-cost solution for the production of sustainable and storable liquid

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What happened 20 years ago?

It is interesting to see how the field of direct thermochemical liquefaction developed over the years. We are thus presenting one example highlight from the PyNe newsletter twenty years ago in this regular feature....:



Entrained Flow Gasification of Bio-oil

By Robbie H. Venderbosch, Bart van de Beld and Wouter Prins, BTG biomass technology group b.v., The Netherlands.



Syngas from biomass
To produce synthesis gas from biomass, several options are possible, such as direct oxygen-blown pressurised biomass gasification. Entrained flow gasification of bio-oil may be an interesting new option, because bio-oil offers the advantages that:

- It is a uniform clean liquid and easy to handle, especially pumping, atomisation and pressurisation.
- bio-oil production can be adapted to local conditions of biomass price and availability. Its utilization for large scale gasification can be centralized at an appropriate location, and
- It reduces transportation costs and
- most existing entrained flow gasifiers can probably be used.

Some of these advantages are summarised in Table 1. Research on both routes including the bio-oil gasification is justified which is why BTG is gasifying bio-oil.

Bio-oil gasification
The BTG entrained-flow gasifier used is shown in Figure 1. It consists of a feeding section for liquids and air, a gasifier, and a cooling section. Air, and mixtures of oxygen and air,

are introduced through a dedicated inlet. The bio-oil flow rate can be varied from 0.1 to 2 kg/hr. The gases produced are cooled, and condensed water is weighed. Gas samples are analysed by GC.

Results of analysis of the 'tar' (by SPA) will be published in a separate paper, together with results derived for enriched air (50% oxygen) and pure oxygen. Only the air gasification results are discussed here. Figure 2 shows results for two temperatures, 1000°C (filled symbols) and 1100°C (open symbols) as a function of the Equivalence Ratio (ER). The solid lines represent the equilibrium composition of H_2 , CO, CH_4 , and CO_2 .

Both CO and H_2 concentrations are significantly lower than the equilibrium values. However, H_2 : CO ratios of 0.8 to 0.9 are obtained (Figure 3), and these are very close to the theoretical values. CH_4 concentrations decrease from 10 vol.% down to zero, while CO_2 concentrations increase from zero to 5 vol.%. A relatively high CH_4 concentration in the product gas is beneficial in energy production, but is not desired in syngas. Literature data suggests that thermal decomposition of CH_4 occurs at temperatures only above 1250°C: higher gasification temperatures are required to eliminate the CH_4 .

Advantages of direct biomass gasification	Advantages of bio-oil gasification
<ul style="list-style-type: none"> • Cheap feedstocks are sometimes available • Biomass is a known feedstock • Energy efficiency of syngas production up to 80% • Close coupling of biomass conversion and green fuel synthesis allows intensive system integration • Gasification has been 'demonstrated' up to a scale of 30 MW_{th} (Värnamo, Battelle) 	<ul style="list-style-type: none"> • Bio-oil production is a cheap pre-treatment step • Liquid products are easy to handle • Energy efficiency of bio-oil gasification is over 85 % • De-coupling biomass conversion from bio-oil processing offers cost reduction opportunities • Oil gasification with pure oxygen is proven technology • Bio-oil contains no ash • Pressurisation of bio-oil is easy • Bio-oil syngas cleaning is cheap (less tar and methane) • Problems related to feedstock variations are avoided

Table 1. Comparison of biomass and bio-oil gasification for FT synthesis.

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Upcoming Events



19. + 20. October 2022

4th European Conference Future of Biofuels, Copenhagen, Denmark
<https://fortesmedia.com/future-of-biofuels-2022,4,en,2,1,17.html#details>



24. – 26. October

International Conference on Biofuels, Bioenergy and Bioeconomy, Vancouver, Canada
<https://frontiersmeetings.com/conferences/biofuels/>



9. + 10. November 2022

ACI's 11th European Biomass to Power, London, UK
<https://www.wplgroup.com/aci/event/european-biomass-to-power/>



10. + 11. November

3rd International Conference on Biofuels and Bioenergy, Paris, France
 Theme : Exploring New Trends in Biofuels and Bioenergy for Sustainable Development
<https://crgconferences.com/biofuels/>



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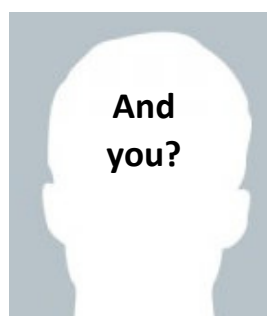
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Contact your national representative in the IEA Bioenergy ExCo to assess the benefits of joining Task 34.

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Task 34: Direct Thermochemical Liquefaction



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