# Enhanced catalytic fast pyrolysis of biomass for maximum production of high-quality biofuels (EnCat)

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**Programmalijn:** BBE - Chemisch katalytische conversietechnologie

**Partners:** 

University of Twente (coordinator) OPRA b.v. Alucha b.v. KTH (Sweden) RISE (Sweden) BIOS (Austria) IChPC (Poland) HIG (Poland)





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### Summary

Fast pyrolysis of biomass is one of the most promising ways to directly generate liquid fuels from biomass. However, the produced pyrolysis oil may have several major drawbacks which suppress its application for power and heat generation or transportation fuels. These are: high oxygen, water and water-soluble (acids) contents, which affect negatively the acidity (corrosion effect), miscibility with petroleum-based fuels (separation of fractions), chemical stability (aging), high viscosity,low energy density and the presence of fuel bound nitrogen.

The Enhanced Catalytic Pyrolysis (EnCat) project presents and investigates a new concept for the production of high-quality bio-oil and a high yield. Because of a novel biomass pre-treatment step to be developed the concept is suitable for both woody biomass and biomass residues from agriculture, etc. The pretreated biomass will be pyrolysed in a reactor making use of deoxygenation catalysts. Simultaneously,  $CO_2$  will be captured with sorbents and via the water-gas-shift reaction in-situ hydrogen will be produced. After cleaning, the oil vapours will be mildly hydrogenated to produce a high-quality bio-oil. The high-quality oil will be used for combustion tests in both a diesel engine and a gas turbine for combined power and heat generation. Parallel to this, the bio-oil will be further upgraded by a new method of downstream hydrogenation under high pressure for production of high-grade transportation fuels. The objectives of this proposal are:

- To develop a new concept for the production of biofuels based on an enhanced catalytic flash pyrolysis process including deoxygenation and hydrogenation for the high-yield production of high-quality bio-oil from both woody and residual biomass streams;
- To test the high-quality oil in gas turbines and diesel engines for the production of heat and power;
- To further increase the applicability of the bio-oil as transportation fuel by downstream hydrogenation;
- To evaluate the new concept from biomass to biofuels with respect to sustainability and technoeconomic feasibility.

The impact of the EnCat-project is that the new biofuels concept will increase the value of the network of biomass suppliers (e.g. forest industry, farmers, SMEs from food sector, etc.) who have such biomass resources available. The industry and companies involved in biomass pre-treatment and fuel utilization for power and heat generation will extend their portfolio to new and renewable products and shall therefore become more competitive on the national and international market. The process engineering companies shall profit from widening their business by providing opportunities for petroleum chemical industries to enhance their green products. Gas turbine and diesel engine manufactures will be able to accommodate the new fuel in their equipment with minor engine changes. In the project two technology suppliers (Alucha and OPRA) are involved. To enable and speed-up the implementation of the project results, the partners will develop a plan for a demonstration project.

# Accountability (Verantwoording)

Het project is uitgevoerd met subsidie van het Ministerie van Economische Zaken, Nationale regelingen EZ-subsidies, Topsector Energie, uitgevoerd door Rijksdienst voor Ondernemend Nederland.

Voor aditionele kopieën van deze rapportage of voor aanvullende informatie kan er contact opgenomen worden met Prof.dr.ir. G. Brem via hetvolgende email adres: <u>g.brem@utwente.nl</u>

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# 1. Project information

Project acronym	EnCat				
Official project duration	46 months (1 March 2017 till 31 December 2020)				
Coordinator institution	University of Twente				
Coordinator name	Prof.dr.ir. G. Brem				
	University of Twente				
	OPRA Turbines International BV				
	Alucha Management b.v.				
Partners	KTH University				
(and also any and in 1)	RISO				
(see also appendix 1)	BIOS BIOENERGIESYSTEME GmbH				
	Institute for Chemical Processing of Coal				
	HIG Polska Sp.				

# 2. Introduction

Conventional flash pyrolysis is known and investigated since the '80-ies. Overview and reviews of this research are given in [1]-[4] and main conclusions are that bio-oil is characterized as acidic, corrosive, thermally unstable, polar and highly oxygenated. The bio-oil produced with these conventional pyrolysis technologies is not suitable for direct use as a transportation fuel or as a fuel additive; it has low heating value which is approximately 40-50% lower than that of fuel oil, it has high viscosity, substantial solids content, and it typically contains 25-30 wt-% water that cannot be readily separated, and make bio-oil immiscible with conventional fuels. Oxygen content of bio-oil ranges from 30-55 wt-% depending upon the parent biomass. The oxygenated compounds in the bio-oil, derived from cellulose, hemicellulose and lignin, are considered main contributor for most of its deleterious properties. Limited commercial successes can be seen because of the low quality of the pyrolysis oil which does not meet the requirements of the present infrastructure.

Catalytic pyrolysis can be seen as a new generation process/technology aiming at the production of a high-quality oil that is a better marketable product. So far the catalytic pyrolysis process has already been tested by some companies such as Anellotech and KiOR. GTI has also introduced a new idea called IH<sup>2®</sup> technology, which is a high-pressure thermochemical process that employs a catalysed fluidized bed hydropyrolysis step followed by an integrated hydroconversion step to directly convert biomass into high quality, fungible hydrocarbon fuels [5]. Despite these efforts no commercial breakthroughs can be noticed because the costs are too high of the complex technologies: high-pressure equipment, requirement of hydrogen production plants, and present technologies are not flexible for cheap biomass feeds such as agriculture feeds.

A further step in catalytic pyrolysis is catalytic pyrolysis in a hydrogen atmosphere. Research in this field is very young and a review is given in [6]. The conclusion from the review article is that catalytic pyrolysis in a hydrogen atmosphere provide higher yields of hydrocarbons and much slower catalyst deactivation due to coking promising compared to standard catalytic pyrolysis. The new process in the EnCat-project has gone one step further than hydropyrolysis by applying catalytic pyrolysis in a hydrogen atmosphere and simultaneously producing the hydrogen in the reactor itself using sorbents (in-situ). This new process is patented by the University of Twente and was the starting point for the project [7].

Another point that is adressed is the pre-treatment of biomass (by washing) prior to the pyrolysis process. On one hand this will make pyrolysis process more efficiently with respect to quality and quantity of the oil (less unwanted cracking). On the other hand this opens the possibility not to use only (expensive) wood, as is done in most projects so far, but also agriculture residues. This fuel flexibility makes the process very attractive for many applications in the Netherlands (wood, paper sludge, grass, straw, reed, miscanthus, etc, etc).

A final strong point is that the project also includes two applications of the pyrolysis oil in both an internal combustion engine and a gas turbine. In this way the whole chain of biomass to electricty/heat/fuel is covered. In the vast majority of previous projects the research has foucsed on whether we can use pyrolysis oil in prime movers, whereas the present project looks on how pyrolysis oil and related technologies can be optimized to make it more commercial attractive. The NL partners in the european consortium consists of knowledge insitute (UT), a pyrolysis technology supplier (Alucha) and a gas turbine supplier (OPRA). This strong consortium is a good boundary condition for a successful commercialisation of the new technology.

The Enhanced Catalytic Pyrolysis (EnCat) project has investigated a new concept for the production of high-quality bio-oil and a high yield. The idea is schematically given in Figure 1. A novel biomass pre-treatment step has been developed for this the concept which is suitable for both woody biomass and biomass residues from agriculture, etc. The pretreated biomass is pyrolysed in a reactor making use of deoxygenation catalysts. Simultaneously, CO<sub>2</sub> is captured with sorbents and via the water-gas-shift reaction in-situ hydrogen is produced. After cleaning, the oil vapours can be mildly hydrogenated to produce a high-quality bio-oil. The high-quality oil is used for combustion tests in both a diesel engine and a gas turbine for combined power and heat generation. Parallel to this, research has been carried out for further upgrading by a new method of downstream hydrogenation under high pressure for production of high-grade transportation fuels.



Figure 1, the concept of Enhanced Catalytic fast pyrolysis for the production of biofuels

The impact of the EnCat-project is that the new biofuels concept may increase the value of the network of biomass suppliers (e.g. forest industry, farmers, SMEs from food sector, etc.) who have such biomass resources available. The industry and companies involved in biomass pre-treatment and fuel utilization for power and heat generation may extend their portfolio to new and renewable products and shall therefore become more competitive on the national and international market. The process engineering companies shall profit from widening their business by providing opportunities for petroleum chemical industries to enhance their green products. Gas turbine and diesel engine manufactures will be able to accommodate the new fuel in their equipment with minor engine changes. In the project two technology suppliers (Alucha and OPRA) are involved. To enable and speed-up the implementation of the project results, the partners have developed a plan for the future roadmap.

#### The objectives of the project are:

- To develop a new concept for the production of biofuels based on an enhanced catalytic flash pyrolysis process including deoxygenation and hydrogenation for the high-yield production of high-quality bio-oil from both woody and residual biomass streams;
- To test the high-quality oil in gas turbines and diesel engines for the production of heat and power;
- To further increase the applicability of the bio-oil as transportation fuel by downstream hydrogenation;
- To evaluate the new concept from biomass to biofuels with respect to sustainability and technoeconomic feasibility.

#### General remark regarding COVID 19

COVID-19 induced restrictions especially regarding the access to laboratory and testing facilities have led to delays regarding the work at UT which had an effect on the progress of work at other partners such as e.g. IChPW. The work performed at BIOS however, could be finalised without COVID-19 related delays since no additional data from the missing test runs at UT and IChPW were needed for its completion. Also the planned seminar at the end of the project had to be replaced by a webinar. To realize all milestones the end date of the project was extended to 31 December 2020.

#### References

[1] S. Czernik, A.V. Bridgwater, Overview of Applications of Biomass Fast Pyrolysis Oil, Energy & Fuel, 18 (2004) 590-598.

[2] A.V. Bridgwater, Review of fast pyrolysis of biomass and product upgrading, Biomass and Bioenergy, 38 (2012) 68-94.

[3] A.V. Bridgwater et.al., An overview of fast pyrolysis of biomass, Organic Geochemistry, 30 (1999) 1479-1493.

[4] M. Jahirul et.al., Biofuels Production through Biomass Pyrolysis - A Technological Review, Energies, 5 (2012) 4952.

[5] Marker T. etc., Environmental Progress & Sustainable Energy, Vol.31, No.2) DOI 10.1002/ep
[6] Resende FLP, 'Recent advances on fast hydropyrolysis of biomass', Catalysis Today, Jul 2016, pp 148-155)
[7] European patent application 14734939.3, 2015

# 3. Project approach and knowledge dissemination

Figure 2 shows the interdependencies between the WPs of the EnCat-project. In WP2 biomass is pretreated via leaching with the light fraction of the bio-oil produced in WP3. The pre-treated biomass of WP2 is used for the enhanced catalytic pyrolysis in WP3. The bio-oil from WP3 is used in WP5 for combustion test in both an engine and a gas turbine. In WP4 the bio-oil from WP3 is characterized and further upgraded to transportation fuels. The sustainability of the whole biomass to fuel chain is evaluated in WP6. Following, the project covers a whole chain approach including its evaluation by techno-economic analyses and LCAs.



Figure 2 Pert diagram of all WPs

The project results have been on a yearly basis reported to the different national fundings by the leadig partners. All stakeholders have been invited for a webinar on 26 November 2020 and the presentations of this webinar have been distributed to the participants as a replacement for the mentioned brochure in the proposal. The project results have been presented on different conferences:

- 1) Combura 2018 Symposium, Poster presentation, The Netherlands, Soesterberg, 2018
- 2) 22nd International Symposium on Analytical and Applied Pyrolysis (Pyro2018), poster presentation, Kyoto, Japan, 2018
- 3) SETAC Europe 2019 poster presentation
- 4) 7th H International Conference on Social Life Cycle Assessment: Impacts, Interests, Interactions, June 14-17, 2020
- 5) 27th European Biomass Conference and Exhibition, May 2019, Lisboa, Portugal
- 6) 28th European Biomass Conference and Exhibition, July 2020, Virtual (e-EUBCE 2020)

- 7) ASME Turbo Expo 2021
- 8) Project was presented in IEA Bioenergy Task 34 meeting in May 2017, Piteå, Sweden. Summaried catalytic pyrolysis of biomass activites of KTH was published in IEA Bioenergy Task 34 PyNe 43. Janiary 2019. Harsha Mysore Prabhakara from UT visited KTH and made test at KTH with cooperation with H2020 project BRISK2 and this was published in IEA Bioenergy Task 34, PyNe 45, December 2019.

Finally, a webinar on 26 November 2020 was organized as the planned seminar on the 28th European Biomass Conference and Exhibition in April 2020 was cancelled because of Covid-19.

In appendix 2 some photos are enclosed on information exchange.

# 4. Project results

In tis report the main results are presented per workpackage (WP).

# WP2: Biomass pre-treatment (Task leader: BIOS)

In the first project year, a basic concept for the leaching process was worked out and the general framework conditions were determined. To investigate the effects of acid leaching on biomass feedstocks BIOS developed a lab-scale test set-up and worked out a detailed test run protocol. Lab-scale leaching tests with woody biomass (Beech; Rettenmaier biomass HBS 150-500) were performed in order to identify the optimum acid leaching conditions. The influence of relevant parameters on the leaching process was investigated. The liquid to solid ratio, the acidity of the leaching liquid, the temperature and the residence time have thereby been identified as the most relevant influencing parameters. The test runs with acid leaching of woody biomass have proven the capability of this technology to remove alkali and alkaline earth metals (AAEMs) from biomass feedstocks. Also regarding other ash forming elements, a certain reduction was possible. Thus, acid leaching could reduce the ash content of the biomass significantly. In a parametric study the optimum acid leaching conditions for woody biomass were identified. The test runs showed that with acid leaching with 5 wt% acetic acid in water at 30°C, a residence time of 30 minutes and a liquid to solid ratio of 10:1 a significant reduction of AAEM in the feedstock could be achieved: Ca: 59%, Mg: 95%, Na: 96% and K: 99%.

In the second project year the acidic leaching tests have been repeated with Miscanthus. The test run results showed that the proposed optimum acid leaching conditions for woody biomass can also be applied for herbaceous biomass feedstocks except for the liquid to solid ratio, which had to be increased to 15:1. At these conditions, a significant reduction of AAEM in the Miscanthus feedstock could be reached: Ca: 73%, Mg: 92%, Na: 98% and K: 96%.

Furthermore, parameters relevant for the waste water handling have been investigated under the proposed optimum acidic leaching conditions for beech wood and Miscanthus. A detailed analysis of the used acid leaching liquid regarding relevant parameters for waste water disposal showed, that only a neutralisation of the waste water is needed for discharge into the sewage system according to Austrian legislation ("Abwasseremissionsverordnung") for both biomass feedstocks investigated.

Based on the test run results a preliminary concept of a real-scale pre-treatment process considering acid leaching has been worked out in the second year. During the design of the preliminary leaching process additional questions regarding the influence of the biomass particle size and the possibility of the re-use of the leaching liquid occurred, which have been evaluated in additional test runs in the third project year.

Figure 3 shows some results Mg, Na and K contents in the feedstock and in the treated biomass after counter-current water leaching of Miscanthus

In the final biomass pre-treatment process design, the leaching step only is considered for agricultural biomass feedstocks, because results of the enhanced catalytic pyrolysis experiments in WP3 did not show a relevant improvement for the pyrolysis of leached beech wood. However, for herbaceous biomass feedstocks significantly improved pyrolysis results are expected. In Figure 4 the main process steps of the pre-treatment process are displayed in a process flow diagram.



Figure 3: Mg, Na and K contents in the feedstock and in the treated biomass after counter-current water leaching of Miscanthus

<u>Explanations</u>: d.b.= dry basis; leaching conditions: leaching with pure water in 3 steps,  $30^{\circ}$ C and 30 min. residence time in each leaching step; leaching efficiency = (1 - element mass in the treated biomass / element mass in the feedstock) \* 100



Figure 4: Process flow diagram of the pre-treatment process

<u>Grinding</u>: In the first step, the particle size of the raw material has to be reduced. Therefore, the biomass has to be grinded in e.g. a hammer mill. In an industrial-scale process the biomass particles are expected to have a thickness of 1-2 mm and a length of 5-30 mm after grinding.

<u>Water leaching – only considered for herbaceous biomass feedstocks</u>: In the lab-scale leaching tests the counter-current leaching has been simulated in a batch process. In a real-scale process, the biomass should be leached in a continuous process. This can be achieved in an extraction tower, where the biomass particles are continuously moved through the leaching liquid in the required leaching time of 90 min. The water leaching shall be performed at a temperature of 30°C. The energy required for heating the leaching liquid to 30°C (about 25 kW per ton water) can be covered by low-temperature waste heat of the pyrolysis process (heat from condensation and cooling of the pyrolysis vapours).

<u>Mechanical dewatering – only considered for herbaceous biomass feedstocks</u>: After leaching, the biomass has a moisture content of about 75 wt% w.b. To reduce the energy requirement for thermal drying, the biomass has to be dewatered mechanically to the lowest possible value. For the mechanical dewatering e.g. a screw press can be used. The expected moisture content of the biomass after pressing is about 60 wt% w.b. The water output from the press can be recycled and used in the leaching step.

<u>Thermal drying</u>: Before entering the pyrolysis reactor the biomass has to be dried thermally to a moisture content of about 10 wt% w.b. This can be achieved in e.g. a belt dryer. The specific heat demand for a belt dryer is about 1.1 MW per ton water. The energy demand for the thermal drying process can be covered by excess heat gained from the charcoal combustion and by waste heat recovery from the exhaust gas of the gas turbine.

<u>Waste water filtratration – only considered for herbaceous biomass feedstocks</u>: Filtratable particles have to be removed from the waste water before disposal. The dispersed particles can be removed by filtration in e.g. a belt filter and can be recycled to the process.

<u>Waste water treatment – only considered for herbaceous biomass feedstocks</u>: The waste water from the leaching step has to be treated before disposal into flowing waters due to an increased Al and Fe content, a high P content and a high phenol index. For removal of phenolic components a biological treatment of the waste water in an aerobic sewage treamtment plant is applicable. For phosphor removal a precipitant (Al- or Fe-salts), e.g. Iron(III)-Chloride, has to be added to the process. The implementation of an on-site biological sewage treatment for the arising waste water from the biomass leaching process has been considered for the techno-economic evaluation (see Task 6.2).

# WP3: Enhanced Catalytic Fast Pyrolysis (Task leader: KTH)

#### Selection, preparation, and characterization of the catalysts

ZSM-5 catalyst has been widely investigated as a catalyst for oil and fuel production as well as for the production of renewable oil and chemicals from biomass. Due to the size of the micropores, the shape selectivity of pore structure, and its strong acidity, ZSM-5 catalyst induces changes in the composition of bio-oil by reducing the amount of oxygenated compound via deoxygenation reaction and promoting the formation of aromatic hydrocarbons. However, ZSM-5 catalyst is also known for its high coking formation and rapid catalyst deactivation. Some high-molecular weight oxygenates, which cannot enter the pores of microporous ZSM-5 catalyst, polymerize and form coke on the catalyst surface.

On the other hand, the amorphous aluminosilicates with ordered mesoporous structures, a large pore size, and mild acidity can convert the high-molecular weight oxygenates to a less bulky compound, give a high organic yield, as well as reduce the chances of coke deposition and pore blocking. MCM-41 is one of the mesoporous materials with high surface area and more accessible reaction sites than the traditional ZSM-5 catalyst. However, products may escape before complete pyrolysis because the pore size is too large. In addition, catalytic pyrolysis of biomass using MCM-41 catalyst results in the formation of Polycyclic Aromatic Hydrocarbons (PAHs), which is not a desirable product in the bio-oil. Hence, some studies examined the possibility for improving bio-oil quality by using both H-ZSM-5 and Al-MCM-41, or microporous and mesoporous catalysts, simultaneously.

Prior to the bench-scale catalytic pyrolysis experiment using H-ZSM-5/Al-MCM-41 catalysts, a kinetic study of the H-ZSM-5/Al-MCM-41 catalyst mixture and its effect on lignocellulose biomass pyrolysis is carried out. A kinetic analysis is essential to design and establish efficient, safe, and reasonable processes, including a catalytic pyrolysis process to produce biofuel from a lignocellulose biomass. A determination of the thermo-kinetic behavior of biomass allows a control of the decomposition mechanism of biomass as a function of the pressure, temperature, and combustion. The kinetic parameters of reaction are necessary to predict the reaction behaviors and to optimize the process towards the desired products during the pyrolytic degradation process. The kinetic parameters of biomass pyrolysis depend not only on the feedstock composition, but also on the pyrolytic conditions, such as the decomposition temperature, heating rate, and the presence or absence of catalysts. Thermogravimetric analysis is one of the tools to provide information on the kinetic reactions of the thermal degradation of biomass. Furthermore, information of biomass pyrolysis kinetics is important to evaluate biomass as a future feedstock for fuel or chemical production as well as for an efficient design and control of thermochemical processes.

The selection, preparation, and characterization of the catalysts have been carried out thoroughly and two scientific articles have been published. A published article entitled "Kinetic Study of an H-ZSM-5/Al–MCM-41 Catalyst Mixture and Its Application in Lignocellulose Biomass Pyrolysis," investigated the kinetic study of catalytic pyrolysis of lignocellulose biomass using catalyst mixtures consisting of H-ZSM-5 and Al-MCM-41 at ratios of 7:1, 3:1, 1:1, and 1:3. The experiments were carried out under an inert atmosphere using Thermogravimetric Analysis to gain an overall understanding of the interactions between the biomass and the catalyst. The results showed that H-ZSM-5/Al-MCM-41 ratios of 3:1 was found to work best compared to other mixtures at a temperature of 500°C.

On the other hand, inorganic contaminants and ash species contained in lignocellulose biomasses have shown a point of consideration in thermochemical conversion. This is due to these compounds will adversely affect the catalytic pyrolysis process, catalyze undesired dehydration reactions during the pyrolysis process as well as increase the yields of thermally derived char and non-condensable gases. Therefore, the chemical pre-processing step, such as a dilute-acid leaching method, is preferred before the thermochemical process. The leaching process is expected to remove physiological ash components, alkali earth, and alkali metals that inhibit the thermochemical conversion. Several attempts have been made to investigate the aqueous fraction of pyrolytic liquids, including wood-derived acids, for demineralization of biomass. Acetic acid was chosen as a model compound for the

biomass leaching process, since it is a major acid resulting from biomass pyrolysis. Hence, the leached lignocellulose biomass is seen as a potential feedstock in the conversion of biomass to bio-oil.

In addition, kinetic studies of treated lignocellulose biomass using leaching, cooperated with BIOS, were carried out in a published article entitled "The thermal degradation of lignocellulose biomass with an acid leaching pre-treatment using a H-ZSM-5/Al-MCM-41 catalyst mixture," to observe the effect of Alkali and Alkali Earth Metal (AAEM) removal on the thermochemical conversion of biomass. The acid leaching process was advantageous as a treatment prior to catalytic pyrolysis process to reach high devolatilization and reaction rate.

#### Experimental study of catalytic pyrolysis of biomass

In order to render a proper design of catalytic pyrolysis process of biomass and to provide high activity, selectivity, and longer life catalyst, researchers have altered the shape selectivity and porosity of catalysts to allow multi-step/cascade reactions to take place. Later, the study considers using a staged mesoporous and microporous catalyst to enhance the bio-oil quality. By employing mesoporous catalyst, Al-MCM-41, and microporous catalyst, HZSM-5, in staged layers, and considering a continuous flow of carrier gas, the pyrolysis vapour is constrained to pass through a layer of catalyst one after another. This system will ensure the large molecule oxygenates will be cracked into small molecule oxygenates through Al-MCM-41 before entering HZSM-5 pores.

A catalytic pyrolysis of lignocellulose biomass using a catalyst mixture of H-ZSM-5/Al-MCM-41 with ratio of 7:1 resulted in the highest organic fraction of liquid (5.66 wt-%), the highest yield of non-condensable gases (13.36 wt.%), and the lowest yield of coke (2.22 wt.%). A High Heating Value (HHV) of 34.15 MJ/kg was achieved with a carbon content of 74.90%, a hydrogen content of 8.00%, and an oxygen content of 15.00%. The degree of deoxygenation reached 65.75%, which was 13.25% higher compared to when using a single H-ZSM-5 catalyst. The favourable compounds amounted to 95.89%, which also was higher compared to a catalytic pyrolysis using a single catalyst.

The BET surface areas were exponentially enhanced when an Al-MCM-41 catalyst was introduced to the catalyst mixture. Specifically, it increased as the proportion of Al-MCM-41 in the catalyst mixture increased. After the catalytic experiments, all the BET surface areas of spent catalysts were 14.84-26.52% lower than the corresponding values of fresh catalysts, due to an accumulative coke deposition on the catalyst.

Further, the influence of catalyst regeneration on the chemical composition of the upgraded oil is one of the factors pertaining to the catalytic process. The work has been published as an article entitled 'Catalytic Pyrolysis of Lignocellulosic Biomass: The Influence of the Catalyst Regeneration Sequence on the Composition of Upgraded Pyrolysis Oils over a H-ZSM-5/Al-MCM-41 Catalyst Mixture'. For industrial application, catalyst regeneration has the benefit of controlling catalyst costs as well as limiting the generation of chemical waste. Compared to the price of fresh catalyst, the price to regenerate the catalyst is considered lower. By regenerating the catalyst, the use of new raw material has been minimized and the need for disposal has been reduced. Thus, it is important to give an indication of the yield and activity of the regenerated catalyst. Providing the performance comparison between regenerated catalyst and its fresh one allows a refinery to decide upon its regeneration or disposal.

In catalytic pyrolysis of biomass, catalyst deactivation is primarily associated with coke deposition, which is caused by the decomposition of the oxygenates derived from biomass pyrolysis and the repolymerization of phenolic oxygenates. Thus, the effect of catalyst regenerations on the product distributions and compositions of bio-oil as well as Non-Condensable Gases (NCGs) yields were investigated. After each experiment, the char and spent catalyst mixture were removed from the rector. The spent catalyst was subjected to a regeneration procedure in a fixed bed reactor by using an airflow. The furnace temperature was increased from ambient temperature up to 250°C with a ramping rate of 4.5°C min<sup>-1</sup>. Thereafter, it was kept at an isothermal condition at 250°C for 40 min. Then, the temperature was raised to 600°C with a temperature increase of 5°C min<sup>-1</sup> and kept at 600°C for 5 h. The temperature was then decreased to 105°C (instead of using an ambient temperature) to prevent moisture absorption. Finally, the catalyst mixture was stored in the oven at 105°C until the next experiment was started. Subsequently, it was fed back to the pyrolysis reactor, and the experiment was repeated. In total, ten reactions were carried out, and they were labelled as R1, R2, R3, ..., R10.

The results from the sequential catalyst regeneration of H-ZSM-5/Al-MCM-41 catalyst mixture show that the total mass balance closures calculated for the experiments vary between 95.54 wt-% and 99.43 wt.% for the non-catalytic and 94.54 wt.% and 98.67 wt.% for the catalytic experiments, respectively. The scatter in the product yields is always less than 5%, indicating reproducibility sufficient for observing trends in all similar experiments.

An increased Organic Fraction (OF) as well as a decreased water, NCG, and coke yields with the increased number of reaction cycles suggested that the acid sites per surface area of the catalyst mixture decreased, followed by its decrease in cracking abilities. The opposite trends of the OF and water yields indicated that the deoxygenation process occurred via H<sub>2</sub>O production. A curvature of trend lines was observed around the 7th reaction cycle when the catalyst mixture appeared to regain its cracking ability.

A decreased in CO and CO<sub>2</sub> yields revealed that the deoxygenation in catalytic pyrolysis with catalyst mixture occurred via decarbonylation, decarboxylation, and dehydration mechanism. The chemical formula of bio-oil changed from  $CH_{0.17}O_{0.91}$  for NC to  $CH_{0.14}O_{0.66}$  for catalytic pyrolysis experiment after 5-reaction cycle, which indicated that the oxygen in the bio-oil was decreased at the expense of hydrogen. The HHVs of bio-oils decreased as the number of reaction cycle increased, albeit the minimum value of 22.41 wt-% in R6 was still higher than the value for NC experiment. Compared to the HHVs of diesel fuel and gasoline petrol, the HHV of the produced bio-oil with catalyst mixture was still low.

As the number of reaction cycles increased, the favourable compounds yield decreased, which suggests that the catalytic activity decreased when the number of reaction cycles increased. The dominant components contributed to the yield of favourable compounds were aromatics and phenols. A decrease in aromatics and an increase in phenols with the increased number of reaction cycles was due to the decreased weak acid sites in the catalyst, which prompted the reduced coke yield and resulted in more conversion of lignin and phenol precursors into phenols.

The surface area for spent catalyst decreased by 24% of the value for the fresh catalyst. After regeneration, the catalyst regained 94% of the surface area for the fresh catalyst. The regeneration procedure could be considered to be effective.

#### Sorption enhanced catalytic pyrolysis

With respect to the concept of sorption enhanced catalytic pyrolysis tests were carried out with deoxygenation catalyst and  $CO_2$  sorbents to reveal the key parameters to optimize the process with respect to quality and yield of the bio-oil. Dolomite was selected as an CO2 Sorbent and Hydrotalcite as an Deoxygenation Catalyst.



Figure 5: Schematic of Bench scale Fludized Bed Reactor

In Figure 5, the experimental equipment used to investigate the role of dolomite and hydrotalcite in biomass fast pyrolysis. The experimental system mainly consisted of five section namely, a feeding

system, the bubbling fluidized bed reactor, solid gas separator (cyclones), cooling system and the gas analysis equipment.

Dynamic experiments were conducted on a bench scale fluidized bed reactor with a constant biomass feed-rate of 0.5 kg/hr., a pyrolysis temperature of 500  $^{0}$ C and at an experimental time of 60 mins. Dolomite loading (WHSV) was varied by replacing part of the quartz sand (bed material) with dolomite. WHSV is defined as the ratio of biomass flow rate to the catalyst mass in the bed. In Figure 6, the vol% of CO<sub>2</sub> measured in the effluent gas as a function of time are plotted for different WHSV. CO<sub>2</sub> was detected in the effluent gas upto a certain experimental time. After a certain time, the CO<sub>2</sub> concentration in the effluent gas increased. This rapid change is termed as "breakthrough". The breakthrough times increased from 6 to 80 min as the dolomite loading increased. Following the breakthrough, the effluent CO<sub>2</sub> volume fraction increases but does not attain a dynamic equilibrium CO<sub>2</sub> level. Hence, the bed continues to take up CO<sub>2</sub> at a lower rate than before breakthrough. We postulate that in addition to fast adsorption there is a much slower process of CO<sub>2</sub> uptake with a considerably higher capacity.



Figure 6: Effect of WHSV on CO<sub>2</sub> Concentrations during catalytic pyrolysis using Dolomite

In conclusion, Dolomite proved to be effective material for  $CO_2$  sorbent as it shifted the water gas shift (WGS) equilibrium towards H2 Production. At WHSV was 1.32, data analysis revealed that the 12,13 % of the feed Hydrogen was converted to Gaseous Hydrogen. The Average H<sub>2</sub> Volume fraction=2,83 Vol% (45,1 % on N2 free basis) in the experiment. In Figure 7, It can seen that, the introduction of dolomite into the bed resulted in CO2 sorption and Evolution of H2. CO reached a dynamic equilibrium. As discussed earlier, H2 was still present in the gas stream even after the Breakthrough was reached.

Further, the Bio-oil quality parameters were the best at WHSV of 1,32 as Dolomite mildly deoxygenated the pyrolysis vapours and yield an biooil with Alkyl Phenols and Mono-cyclopentanones. Acids were also reduced to a great extent. These results were conducted at a pyrolysis temperature of 500 °C and for an experimental time of 60 mins. The heating value of the Bio-oil was 26.52 MJ/Kg when compared to 18.1 MJ/kg for thermal pyrolysis oil measured on the bomb calorimeter. This investigation led to insights on the role of  $CO_2$  sorbent in biomass pyrolysis conditions. It opens up new possibilities for research on the effect of in-Situ hydrogen production on downstream hydrodeoxygenation processes.



Figure 7: Comparison of Gas concentrations between Thermal and Catalytic Pyrolysis

# WP4: Characterization and Upgrading of Bio-oil (WP leader: IChPW)

The aim of this WP4 was to upgrade pyrolytic oils in hydrotreating process in order to improve their functional properties to obtain transportation fuels. Expected result of this task supposed to be selecting the window of the best hydrotreatment process parameters: temperature, pressure, hydrogen and oil flow.

Pyrolysis oil valorisation tests were carried out in a batch reactor as well as in a flow installation for pressurized hydrotreatment of liquid fuels (Figure 8). The installation was equipped in a reactor with 120 cm<sup>3</sup> volume that could work up to 17 MPa and up to  $\sim$ 500°C. The temperature was regulated by two independent zone heating systems. Crude oil was fed by the pump into the reactor and hydrogen was fed from the gas bottle by using the mass flow controller. Gas and oil after reaction was removed from the reactor directly to the cooler and liquid separator.



Figure 8: Scheme of the laboratory stand for pressurized hydrotreatment of liquid fuels

Four commercial available hydrogenation and hydrodesulfurization catalysts were examined in over a dozen hydrotreatment tests: cobalt molybdenum HDMax200 catalyst by Clariant, nickel molybdenum

HDMax300 catalyst by Clariant and nickel molybdenum catalyst by Albemarle, and high nickel catalyst KUB-3 by New Synthesis Institute. Catalysts were activated before hydrotreating process. The influence of temperature in the range 200-350°C, pressure (50-170 bar), and catalyst quantity was investigated on the course of the reaction. Pyrolysis oils produced by BTG BioLiquids B.V.: BTG1, BTG2, BTG3 supplied to IChPW in sufficient quantities for tests were subjected to hydrotreatment.

Subjected to the catalytic hydrotreatment process pyrolysis oil strongly decomposes above 200°C, forming carbon deposits that settle on the surface of the catalyst, hence hindering the hydrogenation process. Under the reaction conditions tested, the preferred hydrogenation and hydrodeoxidation reactions occurred much slower than polymerization and condensation reactions leading to formation of the main product in highly viscous form often mixed with polymers substance.

The comparative BTG3 oil hydrotreating tests were carried out in the batch reactor in the presence of two pre-selected catalysts: HDMax200 and HDMax300, in the amount of 20g and 40g per 200cm<sup>3</sup> of bio-oil and without catalyst. Five tests were carried out at temperatures up to 200°C and one at 300°C, all at 160 bar. The ratio of hydrogen to bio-oil used in each experiment was 100dm<sup>3</sup>:200cm<sup>3</sup>. High viscosity substance, consistency of tar (often mixed with char) was produced in 47-60% yield as a main product. Liquid product with a 74.0-83.5% water content was obtained in a yield of 28-34%. During hydrotreating processes formation of gaseous products (11-20%) and additional amount of water (7-11%) was observed. Aqua fraction was characterized by relatively high molar ratio  $H_d/C$ (1.87-2.63) and low molar ratio O<sub>d</sub>/C: 0.05-0.13. Estimated yield of product, calculated on a dry state, was 5-9%. The molar ratio  $H_{d}/C$  in heavy tar like product was 0.76-1.18 and it was lower than in BTG3, which indicates a higher degree of product condensation compared to the starting material. Tests showed significant removal of oxygen from the oil (94-97% from water phase, 23-58% from heavy phase). The lowest deoxidation degree (23%) was obtained for the test without the catalyst (36%) and test conducted at the temperature of 300°C. Similar results were obtained for the tests carried out in 200°C in the presence of HDMax200 and HDMax300 catalyst used in different amounts. Under the tested reaction conditions, the preferred hydrogenation and hydrodeoxidation reactions strongly compete with polymerization, condensation, and hydrocracking reactions.

The best results were obtained for BTG3 oil hydrotreatment in a flow reactor in the presence of a Clariant cobalt molybdenum catalyst HDMax200 (~100cm<sup>3</sup>, H=25 cm) in temperature below 200°C at 120 bar for 4h. Oil flow was 1.2 cm<sup>3</sup>/min and hydrogen flow was 2 000 cm<sup>3</sup>/min ( $[v_{H2}/v_{oil}] = 1667$ ). Weight hourly space velocity (WHSV) was 1.8 kg oil/kg cat h. The process gave about 380 ml of product, which separated into two fraction: high water fraction (~160ml, 40%) and heavy organic one (~220ml, 55%). During the process, moderate amount of oxygen (not contained in water) was removed from both fraction: 38% from water fraction and 20% from organic one. Molar ratios Od/C was reduced in relation to the raw material used (BTG3).  $H_d/C$  in water phase was higher than in BTG3, and in the heavy phase - lower, which indicated product condensation into high-molecular organic compounds. Product with a 50% water content had a lower pH=4 and lower value of the coking number (11.70%) than highly viscous product. It was characterized by 28.45% carbon content, 59.99% oxygen content (16.11% dry matter), 8.93% hydrogen (3.41% dry matter), 0.77% nitrogen, 0,09% sulphur, and molar coefficients: Od/C=0.42 Hd /C=1.44. Highly viscous product contained 57.17% of carbon, 34.2% of oxygen (20.62% dry matter), 7.54% of hydrogen (5.83% dry matter), 0.55% of nitrogen and 0.10% of sulphur. Molar coefficients were: Od/C=0.27 and Hd/C=1.22. LHV was 22.64 MJ/kg. It was characterized by 15.3% water content, pH=5 and a high coking number (21.33%). Product left in the refrigerator for 6 weeks underwent polymerization, which resulted in a significant increase in viscosity. During the process, the deposition of carbon on the catalyst was observed to a limited extent compared to the previously performed tests.

In order to remove oxygen from the oil more effectively, an attempt was made to perform a two-stage hydrotreatment in a flow reactor in the presence of a Clariant HDMax200 catalyst. For this purpose, after the mild hydrotreating process carried out at 200°C, the first stage product was hydrogenated at a higher temperature  $\sim$ 350°C. The test resulted in a product obtained in the process contained 97.58% water. The content of the element C, H, N, S and O was respectively 0.70%; 11.08%; 0.35%; 0.09% and 86.79%. Most of the product obtained was deposited on the catalyst in the reaction space.

In hydrotreatment processes carried out in the presence of HDMax200 and HDMax300 catalyst in flow installation for pressurized hydrotreatment at temperatures below 200°C, it is possible to remove from the pyrolysis oil moderate amounts of oxygen, on average, about 25%. Under the tested reaction conditions, the preferred hydrogenation and hydrodeoxidation reactions, strongly compete with polymerization, condensation, and hydrocracking reactions. Undesirable reactions increase with the increase of the process temperature, often preventing their course, especially in the flow reactor. It seems that in order to ensure deeper removal of oxygen from the oil, it is necessary to use catalysts other than the tested ones. At the same time, it is advisable to develop additives stabilizing the obtained products, which may be reactive and show a tendency to polymerization and / or polycondensation processes with the formation of tar-like products.

The use of the distillation process as a pretreatment of pyrolysis oil before hydrogenation process, and use of the fraction between 100 and 200°C, made it possible to obtain liquid with relatively good stable physicochemical properties. Hydrogenation process was carried out in a flow reactor, in temperature below 200°C at 100 bar with oil flow - 1.2 cm<sup>3</sup>/min, hydrogen flow - 2 000 cm<sup>3</sup>/min and weight hourly space velocity - 1.8 kg oil/kg cat h. The product obtained with the yield of 75% as a result of hydrotreatment was spontaneously separated into an aqueous and organic phase (25%). Organic product was characterized by a 10.6% water content, a 63.9% carbon content, a 24.1% oxygen content (15.7% dry matter), a 8.6% hydrogen content (7,43% dry matter), LHV of 26.7 MJ/kg, and a coking number of 0.57. For this product detailed stability research and miscibility test with other fuels were performed (WP 5.2).

Based on the research on the hydrotreatment of BTG-BTL pyrolysis oils, guidelines for a mild hydrotreating process enabling partial removal of oxygen from the oil using HDMax200 and HDMax300 catalysts in terms of the amount of catalyst in relation to the oil, reagent streams, temperature and process pressure was developed (M.4.1). Report on selected hydrotreatment process parameters and catalysts (M.4.2) and report on hydrogenated batch of bio-oil (M.4.3) are included in the IChPW REPORT NO 165/2020 on the performance of work "Enhanced catalytic fast pyrolysis of biomass for maximum production of high-quality biofuels".

## WP5 Bio-oil combustion tests (WP leader: OPRA)

#### Tests in gas turbine

This work package dealt with the application of pyrolysis oil in two common prime movers; the gas turbine and the reciprocating engine. The goal of this work package was to develop technologies to enable and improve the utilization of bio-oils in gas turbines and reciprocating engines by increasing knowledge. Both experimental tests and numerical simulations have been performed in this work package. Here detailed information on experimental tests in a down-scaled gas turbine combustor are given.

The testing has been performed in a full-scale gas turbine combustor instead of a simplified, downscaled combustor. The OPRA 3C combustor (Figure 9) used for the test has been designed specifically for burning low-calorific liquid and gaseous fuels. The volume of the combustor is significantly larger than for the conventional combustion systems. This ensures sufficient burnout time for low calorific fuels. The increase of the combustor volume can be achieved relatively easy due to the reverse-flow mounting of the can combustors. Flame stabilization is provided by the primary air, which enters the combustor through a radial swirler.

Figure 10 shows a simplified sketch of the fuel, air and flue gas flows in the combustor. The combustion air from the compressor enters at the rear side of the combustor and flows forward towards the fuel supply and the radial swirler, whereby it cools the combustion chamber walls (impingement cooling). Part of the air enters the combustor at the rear side for dilution. Inside the combustion zone a recirculation is formed to stabilize the flame.



: Fgure 9: OPRA's 3C combustor

*Fig 10: Simplified sketch of fuel, air and flue gas flows in the combustor* 

The combustor has been tested using OPRA's atmospheric combustor test rig, shown in Figure 11. The main components of the test rig are the air fan, air preheater and the combustor. The air is taken from the surroundings by the fan and it is heated up to the same temperature as the combustor inlet temperature of the OP16 gas turbine. The pre-heated air is injected into the combustor module and the exhaust gases are emitted through an exhaust stack. The combustor inlet air mass flow can be varied between 0-0.35 kg/s and heated up to a temperature of 300 °C. Natural gas fuel is supplied from the grid. The flow is accurately measured by means of an orifice flow meter and the flow is precisely controlled by an electrically actuated needle valve. Various shutoff and vent valves ensure safe operation of the system. The combustor inlet temperature, exhaust temperature and combustor air inlet pressure are measured continuously. The air mass flow is measured by dedicated equipment upstream of the air heater.



Figure 11: Atmospheric combustor test rig

The liquid system (shown in Figure 12 consists of three fuel tanks, the ethanol and pyrolysis oil tanks are pressurized by shop air to supply the fuel to the nozzle. The diesel tank is at atmospheric pressure and connected to a fuel pump. The pyrolysis oil supply can be heated up to a maximum of 85 °C by an electric heater. The flow is precisely controlled by an electrically actuated needle valve. Various shutoff valves ensure safe operation of the system. Turbine flow meters measure the flow accurately. The liquid fuel is not filtered in the system, so the fuel is tested as supplied.



Figure 12: Liquid fuel system in atmospheric combustor test rig

The full-scale gas turbine combustor has been tested using pinewood fast pyrolysis oil, produced by BTG, hereafter referred to as wood pyrolysis oil. Important parameters for further optimization of the gas turbine combustor are obtained, including combustor efficiency, pollutant emissions, temperatures and operating envelope.

A significant impact of the nozzle and atomization has been found for the liquid fuels. Initial tests have been performed with conventional nozzle types. The combustor operated stable for ethanol and diesel with the conventional nozzle. Operation with wood pyrolysis oil only with the conventional nozzle resulted in unstable combustion. Bad atomization with this type of nozzle resulted in incomplete combustion, whereby the flame stretched far outside the gas turbine combustion chamber (Figure 13). The atomization of the high viscosity pyrolysis oil resulted in bigger droplets, which polymerized and could not be burned within the residence time given in the combustor. As a result, significant amounts of unburned fuel and dust have been found in the exhaust channel after operation with bad atomization (Figure 14). Furthermore, clogging of the conventional nozzle was found while operating on pyrolysis oil.





3:Fig 13: View to combustor outlet with bad : Fig 14:Dust and unburned fuel traces in the atomization of wood pyrolysis oil exhaust channel after operation on pyrolysis oil with bad atomization

The main objective of the test runs was to gain data regarding the temperatures at different positions in the combustor and regarding the flue gas composition at combustor outlet. At first glance, these measurements appear as standard tasks, however, when considering the framework conditions in a gas turbine combustor, they turn out to be extremely challenging. It has to be considered that in the combustor temperatures of up to 1,600°C prevail and that due to the radiation of the flame and of the air-cooled combustor walls suction pyrometers have to be applied to gain reliable temperature measurement results. Moreover, very high local gas velocities exceeding even 60 m/s occur. Furthermore, due to the small dimensions of the combustor commercial suction pyrometers are not applicable.

Due to these constraints, such measurements have not been performed yet by other institutions and therefore, a new methodology had to be worked out by BIOS and special suction pyrometers and high temperature gas sampling probes had to be developed and constructed. Regarding the latter a quenching probe has been constructed. In a quenching probe, the temperature of the flue gas is quickly reduced to below 800°C to inhibit further reactions of CO with  $O_2$  in the extraction line to the analysers. If this is not done, as in a simple not cooled extraction probe, most likely a considerable share of the CO (more than 60%) is oxidised before reaching the analyser.

In Figure 15 the different measurement positions are presented. Temperature measurements should be performed at several positions close to the combustor wall (positions 1, 3, 5 in

Figure ) and on its axis (positions 2, 4, 6 in Figure ) by applying purpose built suction pyrometers. The flue gas composition  $(O_2, CO_2, NO_X, CO, OGC)$  as well as the temperature at combustor outlet should be measured in the middle of the outlet (position 8) and 2 cm from the wall (position 7).



Figure 15: Measurement positions in the gas turbine combustor.

Explanations: PA ... primary air; SA ... secondary air; FG ... flue gas

Moreover, relevant operation data were recorded by the process control system. Regarding the temperature data recorded by the process control system it should be noted that the data obtained from the unshielded thermocouples (see also Figure ) should only be assessed qualitatively.

Bio-oil was delivered from the supplier BTG/BTL (NL) in 30 litre canisters. A picture of a sample taken from the bio-oil as well as results of its analyses are presented in Figure 16.

7	Parameter	<b>Bio-oil utilised</b>	unit
	Density	1,153	kg/m <sup>3</sup>
E 500	С	60.56	wt. % d.b.
100	Н	4.81	wt. % d.b.
SISA SISA	Ν	0.15	wt. % d.b.
1 COLORE	S	0.0082	wt. % d.b.
Martin Bornor	0	34.46	wt. % d.b.
Carl Contraction of the second	Ash content	0.01	wt. % d.b.
	Water content	28.00	wt. % w.b.
	GCV	24.3	MJ/kg d.b.
	NCV	16.1	MJ/kg w.b.

Figure 16: Picture, chemical and physical data of the bio-oil utilised during the turbine combustor tests

The gas turbine combustor of OPRA can operate with natural gas, ethanol and pyrolysis oil fed from different supply lines. During combustion of natural gas (reference case), very stable operating conditions at nominal load conditions of the combustor could be achieved. Due to the stable operation, it has been decided to perform one temperature measurement over a period of about 10 minutes per position.

The average temperatures over a measurement period showed no pronounced temperature profiles along the radius of the combustion chamber. At combustor outlet, the wall-near temperatures were significantly lower than the temperatures in the middle axis. This was due to wall cooling effects at the outside positions as well as the fact that secondary combustion air injected downstream the cone at the end of the combustion chamber is in this region not fully mixed with the flue gas and forms an oxygen rich and cooler strain in the wall-near section.

During the test runs with bio-oil, after ignition with natural gas and as soon as stable operating temperatures at the outlet of the combustor were reached, operation with pyrolysis oil (bio-oil) was initiated. The entire procedure of switching the fuels proceeded manually and took less than one minute. With one bio-oil tank filling the combustor could be operated for 10 to 15 minutes, then operation was switched to natural gas again, the tank is refilled and operation was switched again to bio-oil combustion.

The temperatures measured with the suction pyrometers (mean values) were significantly below those for natural gas combustion due to the lower heating value of bio-oil. Moreover, in contrast with the rather uniform temperature distribution over the combustion chamber during natural gas combustion, pronounced radial and axial temperature profiles occurred.

Compared with natural gas combustion the  $NO_x$  emissions were slightly lower and the CO emissions were slightly higher. As also recognised for natural gas combustion, the  $O_2$  content at the wall-near measurement position 7 was significantly higher than the one at position 8. Moreover, in contrast with natural gas combustion,  $NO_x$  is mainly formed from nitrogen contained in the bio-oil (fuel-N) and due to the comparably low temperatures only to a minor extent via thermal  $NO_x$  formation. In this respect, the main objective of the CFD simulations should be to improve the air staging settings of the combustor to achieve lower  $NO_x$  emissions.

Based on the measurement data regarding bio-oil supply, bio-oil composition and air supply, mass, element and energy balances over the bio-oil combustion process have been calculated. Comparisons of the calculated and the measured flue gas compositions showed a reasonable closure and thus the reliability of the data collected could be proven.

#### Combustion modelling of char burnout for pyrolysis oil

Here a CFD model of catalytic pyrolysis oil combustion was developed based on the available literature and taking also into account char combustion. The model consisted of water, phenol and char

with ration matching the estimated properties of the new catalytic pyrolysis oil. An Eulerian-Lagrangian formulations was used, where the gas flow was modelled as continuous phase and liquid pyrolysis oil particles contained a char were model using discrete phase, see also Figure 17. The model was executed in Open FOAM computational open source software using geometry of OPRA's combustor. The model was further validated with the experiments performed at OPRA Turbines showing good qualitative agreement between numerical and experimental data.



Figure 17: Oxygen content in the gas phase and particles temperature in the discrete phase during combustion of pyrolysis oil

To investigate a behaviour and performance of the gas turbine during the combustion process several parameters including char content in the fuel droplet, electrical load, oxygen content, air swirl velocity and openings/closing of the secondary air holes was done. Generally, the results show that completeness of the combustion and char conversion increases with the reduction of the char content within the droplets (i.e. by decreasing the char content from 1 wt.% to 0,1 wt.% the char conversion factor was doubled). Similar effect was observed for increasing the air swirl velocity and air flow inside the combustor. Here, the improvement comes mostly from a longer residence time of the particles staying in the combustion chamber, as they swirl more than in the conventional case, and effect of an additional air which has been delivered to the regions where there is sufficient amount of fuel to combust but there is not oxygen left. Both effects have to be, however, considered carefully since it was observed that after bypassing certain conditions the improvement outcome diminishes. It was not observed that additional secondary air will improve the combustion process. Yet, it should be noted that the exact mass flow of air through secondary air holes was estimated thus this effect in industrial application can differ from the obtained numerical data. Furthermore, an increase in the air inlet temperature resulted in linear increase of the char conversion process.

Based on the findings, a new design of the combustor was proposed with enhanced air swirl inlet velocity and temperature. By applying the new conditions the char conversion rate has been improved from 37% to 83%.

# WP6: Full-scale design, techno-economics and sustainability (leader: RISE)

#### **Techno-economic analysis**

In order to optimise the process design of the EnCat process with respect to economic apsects, a detailed techno-economic analysis of the whole process has been performed in two stages.

In the second project year a preliminary techno-economic evaluation has been performed based on a basic plant scheme worked out within WP6 in cooperation with the other project partners, also including the biomass pre-treatment based on acid leaching. BIOS worked out questionnaires for the specicifc system units (biomass pre-treatment, pyrolysis process and gas turbine) and forwarded them to the project partners UT and OPRA. In the filled-in questionnaires the partners provided information regarding the expected investment costs, the operating and consumption costs, maintenance and labour costs as well as the expected lifetime, awaiting energy demand and space requirement for the different units. Based on the input given by the partners, the preliminary economic evaluation of the EnCat

process has been performed. Upgrading of the bio-oil produced has not been considered for the techno-economic analysis as no reliable data had been available.

The results of the preliminary economic evaluation of the EnCat process showed the need for optimisation measures to improve the economic viability of the process. Due to the comparably high consumption based costs for the biomass pre-treatment based on acidic leaching and high costs for catalytic material for the pyrolysis process no economically feasible result could be achieved.

For the final techno-economic evaluation the design of the pre-treatment process has been adapted according to WP2 and the whole process and system design have been optimised based on the experimental results from WP2 and WP3 as well as literature data. In the following the final EnCat process is briefly described.

Leaching is only considered for herbaceous biomass feedstocks. To reduce the consumption based costs continuous counter-current leaching with water is considered instead of an acidic leaching process (see Task 2.2).

Biomass pyrolysis is conducted in a circulating fluidised bed reactor. The bed material applied consists of a mixture of catalyst (1.980 kg) and sorbent (4.020 kg) materials, which shall facilitate insitu hydrogen production, deoxygenation and CO2-capture. To reduce the consumption based costs of the pyrolysis process, for the final economic evaluation the use of cheaper catalyst (hydrotalcite) and sorbent (dolomite) materials has been considered. The bed material and the char produced in the pyrolysis process are separated from the pyrolysis gas in a cyclone placed downstream the pyrolysis reactor and forwarded to a char combustor (bubbling fluidised bed). There, the char is oxidised and the bed material is heated up to the desired temperature needed to run the pyrolysis process. Moreover, the sorbent is regenerated before re-entering the pyrolysis reactor. Due to losses and the deactivation of the catalyst, 2% of the total bed material have to be replaced per day. Based on literature data and experimental results of partner UT, this replacement rate has been reduced compared to the preliminary evaluation. By this measures and the utilisation of cheaper sorbents and catalysts (see above), the costs for the bed material could be reduced significantly compared to the preliminary evaluation.

The pyrolysis vapours leaving the pyrolysis reactor resp. the cyclone are separated by condensation and cooling to 30°C into bio-oil (heavy fraction), an aqueous fraction (light fraction) and non-condensable gases. The aqueous fraction can not be used for the leaching process (as planned in the initial plant concept) due to its low content of acids. A disposal without treatment is not possible due to the high contents of organic compounds and the low pH-value. Therefore, the injection into the char combustor is considered for thermal treatment. The non-condensable gases are used in a gas turbine for electricity production.

For the final economic evaluation two different feedstocks, beech wood and Miscanthus, have been considered. For beech wood a fuel input of 9.0 t/h d.b. has been defined by the consortium. Due to the lower gross calorific value (GCV) of Miscanthus the fuel input for this application had to be increased to 9.2 t/h d.b. to gain the same energy input (and also output) at the pyrolysis reactor.

In the preliminary concept of the EnCat process, as a first approach, the utilisation of the produced bio-oil in a gas turbine for in-situ heat and power generation was considered. For the final evaluation the concept has been adapted towards the production of bio-oil for external use. Only the non-condensable pyrolysis gases are used in a gas turbine for in-situ electricity production. Before entering the gas turbine the non-condensable gases are compressed. The compressor is part of the gas turbine unit. The net electric capacity of the gas turbine amounts to about 1.5 MW.

To further increase the energy efficiency of the whole EnCat process, an ORC-process utilising waste heat from the gas turbine and the charcoal combustion has been implemented into the plant concept. In the ORC the flue gases of the char combustor and the gas turbine (in total about 26.3 MW) are used to produce additional electricity, hot air for the biomass drying and also hot water for e.g. district heating. The electricity production of the ORC amounts to about 3.3 MW. By ORC implementation, the electric efficiency of the EnCat process is increased from about 3% to about 10% (related to the NCV of the feedstock material entering the process).

Due to the higher energy demand for drying for the application of Miscanthus (moisture content after the leaching process is about 75 wt% and after dewatering about 60 wt%) less heat is available for district heating than for the application of beech wood (no leaching is considered, moisture content of beech wood is about 45 wt% before drying). In case of Miscanthus a heating capacity of about 6.5 MW and in case of beech wood of about 13.1 MW are available for district heating.

In the final design of the EnCat process, the biomass leaching (based on pure water) is only considered for Miscanthus. After leaching a filtration step of the waste water is considered to remove filterable matter and to recycle the filtered biomass particles to the process. Experimental results from WP2 indicated the need of an additional waste water treatment after the leaching step due to increased Al and Fe contents, high P content and high phenol index of the waste water (see Task 2.2). To evaluate the impact of the additional costs for the waste water treatment in an on-site biological sewage treatment plant, for the application of Miscanthus two different application cases have been considered:

- Under the assumption that by filtration the waste water is cleaned sufficiently, the used leaching water is discharged into the sewage system (limiting values may be less strict in other countries than in Austria)
- For cleaning the water for discharge into flowing waters in a nearby river a biological waste water treatment plant and additionally required chemicals for P elimination are considered (in this case no costs for water are considered as river water is used for leaching)

The process flow diagram of the final EnCat process, which formed the basis for the economic analysis, is presented in Figure 18. It has to be emphasised, that with the present EnCat process layout biomass is converted into heat and electricity and, as the main product, bio-crude oil, which shall be utilised as a substitute for fossil crude oil in refineries.



Figure 18: Process flow diagram of the EnCat process

In a full cost calculation according to VDI 2067 the annual capital bound costs, maintenance costs, consumption based costs, operation based costs, fuel costs and auxiliary energy have been evaluated for the EnCat process for the application cases defined. The expected total investment costs for the EnCat process, the resulting total costs per year including revenues from electricity and heat production and the resulting bio-oil production costs are presented in Table 6.

Table 1: Expected total investment costs, annual costs und bio-oil production costs for the EnCat process for Miscanthus and beech wood

Application		Miscanthus (water leaching)	Miscanthus (water leaching with waste water treatment)	beech wood (no leaching)		
Total investment costs	[€]	45,450,000	55,390,000	42,350,000		
Capital bound costs	[€/a]	2,387,728	2,913,089	2,225,844		
Maintenance costs	[€/a]	1,029,879	1,114,420	976,833		
Operation based costs	[€/a]	724,936	767,206	711,911		
Consumption based costs	[€/a]	2,072,981	1,004,834	923,560		
Fuel costs	[€/a]	5,449,106	5,449,106	8,647,850		
Electricity costs [€		2,275,750	2,379,189	2,023,618		
Total costs per year	[€/a]	13,940,380	13,627,844	15,509,617		
Revenues electricity production	[€/a]	4,192,204	4,192,204	4,192,204		
Revenues heat production	[€/a]	2,028,972	2,028,972	4,086,748		
Total costs incl. revenues	[€/a ]	7,719,204	7,406,668	7,230,665		
Annual bio-oil production	[t/a]	14,672	14,672	14,672		
Bio-oil NCV	[MJ/kg]	28.97	28.97	28.97		
<b>Bio-oil production costs</b>	[€/MWh]	65	63	61		

The maintenance costs are approximated to 2% of the total investment costs. The operation based costs include labour costs (450.000 €/a) and costs for insurance. In the consumption based costs include costs for water (incl. disposal), for the biomass leaching in the pre-treatment process, costs for chemicals needed for the waste water treatment and costs for the replacement of the bed and sorption material of the pyrolysis unit. The electricity demand for the process is based on data from manufacturer specifications, literature data and data provided by UT and OPRA.

In Figure 19, a breakdown of the annual costs for the EnCat process to the different cost groups is displayed. The figure shows, that the major part of the annual costs is related to the fuel costs.

Based on the framework conditions and assumptions for the final economic evaluation of the EnCat process, bio-oil production costs of  $63 - 65 \notin$ /MWh for the application of Miscanthus and  $61 \notin$ /MWh for the application of beech wood result. These bio-oil production costs are in the range of the costs for crude oil including a CO<sub>2</sub>-tax of 100  $\notin$ /t CO<sub>2</sub> (69  $\notin$ /MWh).

Sensitivity analyses revealed, that the most critical values for an economically feasible result of the EnCat process are a cheap catalyst and a high catalyst lifetime to achieve a competitive bio-oil price.

*Figure* shows, that the major part of the annual costs is related to the fuel costs and that the share of the fuel costs on the total costs can be reduced from 56% to about 40% when utilising Miscanthus instead of beech wood. The total bio-oil production costs are for Miscanthus 3 to 6% higher than for beech wood (due to the costs for the leaching process), however, this could in future be compensated or even overcompensated when utilising cheaper agricultural residues as feedstock material. It can therefore be concluded, that the leaching process significantly increases the feedstock flexibility of the EnCat plant and therefore provides the option to utilise cheaper but in terms of AAEM contents problematic fuels as well. Feedstock selection and purchase can therefore be performed depending on seasonal availabilities and seasonal prices of a rather broad spectrum of different biomass feedstocks

With the optimisations measures considered for the final techno-economic analysis an economically feasible result is possible for the EnCat process regarding the production of a crude oil substitute under consideration of a CO2-tax of  $100 \notin t CO2$ .



Figure 19: Breakdown of the annual costs for the EnCat process to the different cost groups

#### Sustainability

A screening life cycle assessment was done in the beginning of the project to identify the environmental hotspots and several recommendations on process improvements were given. All results documented in a screening LCA report. In addition, an LCA/SLCA workshop was conducted in project meeting, Mölndal. In the workshop, RISE presented the screening LCA



Figure 20: System boundary of the study. The functional unit is 1 MJ of pyrolysis oil produced. The pyrolysis process in this chart included the combustion and gasification of char and catalyst regeneration.

results to all partners and process improvements activities are discussed. For the SLCA, all partners discussed the potential social impacts of the EnCat process and are engaged in selecting important impact categories for the SLCA study. An SLCA was carried out based on the input from all partners and the results were presented in project meeting, Hengelo. An abstract was written and submitted to SLCA conference. In next stage, a detailed life cycle assessment study was conducted to assess the environmental impacts of the final process design of EnCat project.

For detailed LCA, the functional unit of the study has been set to production of 1 MJ bio-oil produced from the system for transportation fuel (figure 20). The studied system is a cradle-to-gate system, including operations needed for production of biomass, pre-treatment of biomass, pyrolysis process and recovering of flue gas. Inventory information have been collected mainly from project partners BIOS, UT Twente, KTH and OPRA. Generic data, such as electricity and transport have been taken from the Ecoinvent database and literature. The focus of the results has been on global warming potential, but also acidification, eutrophication and photochemical oxidation have been assessed.

LCA results (Figure 21-22) show that so far, the global warming potential of the bio-oil generated from EnCat process (cradle-to gate) is 73.5-79.8  $CO_2$  eq- g/MJ. It is about six or nine times higher than diesel, petrol, and heavy/light fuel if only considering the production of the oil. However, if including the combustion of the oil, the global warming potential of the bio-oil are comparable with other fuel oil.

The environmental hotspots in terms of the global warming for EnCat process is the electricity use in the pyrolysis process. This followed by the wastewater treatment for the aqueous fraction emitted from the process. For other impact categories (photochemical oxidation, acidification, and eutrophication), the emissions from aqueous fraction contributed most to environmental impact.



Figure 21: Global warming potential for the case of Twente pyrolysis when wood chips are used as a raw material. Only processes with more than 1% total impact are shown.



Figure 22: Global warming potential for the case of Twente pyrolysis when miscanthus is used as a raw material.

The bio-oil generated from EnCat has lower in all impact categories for wood biomass compare to miscanthus biomass. This is because wood chips have no leaching process involved in the pretreatment. On the other hand, the choice of catalyst and sorbents does not affect the final results as the impacts of catalyst and sorbents are insignificant (i.e. contributing 0.1% to the GWP). Recovery of flue gas in the system for biomass pre-treatment, which was identified as hot spot in the first LCA round, reduced all impacts categories: global warming (20-44%), photochemical oxidation (25-71%), acidification (31-66%) and eutrophication (1-2%).

For future process improvements, it is recommended

a) Use internal electricity in the pyrolysis process

b) Study on how to handle the wastewater (aqueous fraction) emitted from the process

c) Study on the possibility of generating other products from the system (e.g. fractionation of the oil products to produce chemical components with high  $CO_2$  footprint)

d) Consider biomass from waste stream (i.e. zero starting impact) to reduce the impact of feedstock preparation

# 5. Roadmap for further development

The roadmap towards a commercial plant is divided in 4 stages (figure 23). EnCat is the current project: tests performed at 0.1-1 kg/hr scale. TRL 3-4. EnCat 2.0 is pilot scale testing stage: 10-100 kg/hr feedrate. EnCat 3.0 for demonstration purpose (1t/hr) and finally EnCat 4.0 the first commercial unit (10t/hr).



#### EnCat 2.0: pilot scale testing & product application

Table 2 shows that, in order to reach overall TRL of 4 on all process steps, the focus should be on developing the following process steps: (1) the catalyst regeneration, (2) Catalytic Pyrolysis Oil (CPO) application tests (lab and pilot). However, to enable CPO product tests, Catalytic pyrolysis (3) oil production at pilot scale is required (TRL5). Furthermore, to improve plant economics test with cheaper feedstock (4) are recommended, for locations in countries where a tax of 100Euro/t  $CO_2$  is not implemented yet. (see ETA in 6.2)

topic	Process	Task	Improves Encat
1	catalyst regeneration	Study and validate catalyst stability, regeneration factor, replacement factor.	Technically: TRL 4. Catalyst lifetime improvement enhances economics and reduces environmental impact of EnCat products
2	CPO application	CPO application test in - Gas turbine - IC engine - CPO post-upgrading	Technically: TRL 4 Confirms economic values of EnCat products as bio-fuel and green chemical building blocks
3	Catalytic pyrolysis	Produce large amounts of PO, pilot scale to enable PO application test	Catalytic pyrolysis: TRL5. Note that integration of several key process steps are required. (Catalytic pyrolysis, catalyst regeneration, quench). Also biomass pre- treatment on pilot scale will be required
4	Feedstock	Test with low cost organic waste streams	Economically.

Table 2: overview of main technical topics to be addressed in EnCat 2.0

While focusing on the technical topics listed, the goals of EnCat 2.0 should be:

- Improve CPO quality: Production of CPO with O-content <20%. At this level (stable), the CPO is a commercial product used by chemical industry.
- Product application fuel 1: demonstration on gas turbine: stable run (co-)combustion CPO
- Product application fuel 2: demonstration on IC engine
- Product application: CPO liquids as Green chemical building blocks
   Utilization of light PO fraction: reform to bio-based acetic acid solution

#### EnCat 2.0 consortium

Within the current EnCat consortium the knowledge and the test facilities are present to reach the goals of EnCat 2.0. By using the existing automated Alucha Pilot plant for CPO production in EnCat 2.0, a faster and cost efficient development towards EnCat 3.0 will be possible due to gains in control and level of process integration. External parties should be welcomed: potential CPO clients should be enabled to perform product application tests.



#### EnCat 3.0: integrated (heat) larger scale DEMO plant

After reaching the EnCat 2.0 goals, an integrated plant unit on demonstration level will be developed, designed, build, commissioned and tested extendedly. Plant automatization and process control will be based on the pilot plant used in EnCat 2.0. Plant flue gas emission measurement will be included. Again, the ETA will be updated. With a capacity of 1.0 ton per hour (on dry base) this unit, at TRL 7, will enable more users of CPO to perform application tests for longer periods. It will also demonstrate the CPO product is stable, on costumer required specifications. The 1t/hr "*preliminary design*" of EnCat can serve as a basis for the DEMO Plant (see figure 24).

*EnCat 4.0: first commercial plant.* With the lessons learned form EnCat 3.0, the first commercial plant shall be developed, designed, build, commissioned, tested extendedly and start running for production. Where required, additional tests will be performed at the 1t/hr plant. At the end of EnCat 4.0 The system is complete and qualified (TRL 8) and is ready to be proven in operational environment (TRL 9). Finally an estimated timeframe for the roadmap in 4 stages is presented in table 3.

Title	Scale	biomass feed rate [kg/hr]	20	20	2021		2022		2023		
EnCat	lab	0.1-1									
EnCat 2.0	pilot	10-100									
EnCat 3.0	demo	1,000									
EnCat 4.0	commercial	10,000									

Table 3: overview and time schedule of stages for further development of the EnCat concept

# 6.Sustainability and relevance for society (TKI-BBE programme, in Dutch)

Dit project past binnen de programmalijn "Chemische katalytische conversietechnologie" van het TKI BBE programma. Bij het te ontwikkelen katalytische pyrolyseproces wordt de biomassa omgezet in een hoogwaardige brandstof die geschikt is voor gas turbines en dieselmotoren. Tijdens de voorbehandeling van de biomassa wordt de biomassa gescheiden in een organisch gedeelte dat gebruikt wordt voor de pyrolyse en een niet-organisch gedeelte dat gerecycled kan worden als mineralen. Hierdoor wordt de biomassa volledig omgezet in vermarktbare produkten (energie en materialen). Het nieuwe proces betreft katalytische pyrolyse waarbij een katalysator toegepast wordt om de pyrolysedampen in de reactor te deoxygeneren, waardoor een olie geproduceerd wordt die weinig zuurstof bevat, minder corrosief is, stabieler is, en een hoge verbrandingswaarde heeft (vergelijkbare eigenschappen met fossiele olie). Bij het deoxygeneringsproces wordt de in biomassa aanwezige zuurstof aan koolstof gebonden en vormt  $CO/CO_2$ . De gevormde  $CO_2$  wordt in dit nieuwe proces afgevangen met een sorbent en hierdoor wordt evenwicht van de water-gas-shift reactie helemaal naar de waterstof kant verschoven. M.a.w. er wordt extra waterstof geproduceerd. Deze waterstof wordt in een nabehandeling voor hydrogeneren gebruikt om de kwaliteit van de olie nog verder te verhogen tot transportbrandstof. Op den duur kant ditzelfde proces ook gebruikt worden voor de productie van duurzame chemicaliën of een combinatie van brandstoffen en chemicaliën.

Omdat zowel de katalyse als de  $CO_2$ -absorptie in de reactor zelf plaatsvindt, betreft het een compacte technologie. En omdat tevens de processen bij atmosferische druk kunnen plaatsvinden resulteert dit in relatief lage investeringskosten, wat deze technologie tot kosten-efficiënt maakt. In WP6 van het project zal een ontwerp, kostenbegroting en roadmap voor commercialisatie ontwikkeld worden.

Doordat in de keten een voorbehandeling van de biomassa (wassen) is opgenomen, is het proces geschikt voor zowel houtachtige biomassa als voor reststromen afkomstig vanuit de landbouw.

Door gebruik te maken van duurzame biomassa (kort-cyclisch) en geen gebruik te maken van fossiel (hulp) brandstoffen is dit nieuwe proces volledig duurzaam. Op deze manier worden duurzame brandstoffen, mineralen en eventueel chemicaliën geproduceerd. De  $CO_2$  die uiteindelijk vrijkomt bij de toepassing van deze duurzame brandstoffen draagt niet extra bij aan het broeikaseffect. Deze geproduceerde  $CO_2$  wordt namelijk weer gebruikt voor het groeien van de biomassa (fotosynthese). Dit geldt zowel voor kleine als voor grote schaal toepassingen.

De geproduceerde energie vervangt een equivalent van fossiele brandstoffen en draagt zo bij aan de verlaging van de uitstoot van fossiele  $CO_2$ . Een schatting van de besparing van fossiele brandstoffen en dus vermeden  $CO_2$  wordt hieronder kort weergegeven.

Voor houtachtige biomassa gaan we uit van 99% organisch materiaal. Hiervan wordt circa 50% omgezet in olie en 30% in gas. Beide producten zijn vermarktbaar als brandstof. De overige 20% is char en is nodig voor het proces zelf. Het overall rendement van het proces bedraagt dus 80%. Per ton biomassa wordt zo 14 GJ aan fossiele brandstoffen bespaard en wordt de uitstoot van  $CO_2$  verlaagd met bijna 2 ton  $CO_2$  (er vanuit gaande dat bij de verbranding van steenkool circa 0,13 ton  $CO_2$  per GJ uitgestoten wordt). Als we er verder vanuit gaan dat in NL op termijn 25 pyrolyse installatie's met een input van 20 ton/h biomassa gerealiseerd worden, dan is daar circa 4 Mton biomassa voor nodig. En dat vervangt dus 56 PJ aan fossiele brandstoffen en vermijdt 8 Mton uitstoot van fossiele  $CO_2$  per jaar. Uiteraard kan dit proces verder uitgerold worden binnen NL en daar buiten. E.e.a. zal afhangen van de behaalde efficiencies, kwaliteit van de brandstoffen en de te behalen kostprijsreducties.

Voor reststromen afkomstig van de landbouw kan een vergelijkbare schatting gemaakt worden. Er vanuit gaande dat de reststromen voor de helft uit organisch materiaal bestaan en voor het overige uit mineralen, inert, etc, dan kunnen de getallen die hierboven staan ongeveer gehalveerd worden wat de vervanging van fossiele brandstoffen en de reductie van  $CO_2$  betreft. Daarentegen komen bij dit proces ook mineralen vrij die weer vermarktbaar zijn. Door de inzet van deze duurzame mineralen worden ook weer fossiele brandstoffen vermeden en wordt de  $CO_2$  emissie verder gereduceerd. Dit hangt echter sterk af van de soort mineralen die in zich in de reststromen bevinden. Hiervoor zal in het project een afschatting gemaakt worden.

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# Appendix 1

#### **Description of partners**

A consortium of 8 partners from 4 ERA-NET Bioenergy countries has been formed. Within this consortium, the whole process chain is covered with acknowledged experts and unique partners. Starting at the biomass fuel provided by Holmen, fast pyrolysis process research and development by UT, KTH, BIOS, IChPW and HIG, testing the concept by Alucha, and ending at the utilisation of biooil in a gas turbine by OPRA and in a gas engine by IChPW, with also full LCA analysis by RISO. The combination of different fields of experience, approaches and know-how from the different partners shall lead to significant added values from the international cooperation Consequently, the international cooperation within the ERA-NET project can provide much better resources, expertise and knowledge transfer than any solely nationally conducted project.

In general,

- A new full chain network within bio-oil production and application from biomass via catalytic fast pyrolysis including academic partners, researcher institutes and industries, will be established. This will form the competences for later demonstration and commercialisation of the concept.
- Specific expertise from the partners will accelerate the development of the whole system
- Expertise needed cannot be covered by a single country only
- Direct industrial cooperation will result in relevant additional know-how gain for all partners

For individual countries and partners:

#### The Netherlands

The Dutch government wants to reduce the greenhouse gas emissions with 80-95% in 2050. Biomass can play in major role in reaching this goal as biomass is the only carbon-containing renewable fuel that can directly be used for the production of hydrocarbons (transport fuel). The UT has a long experience with research in field of biomass and especially with biomass pyrolysis. Also the involved Dutch industrial partners have a strong experience in the bio-energy field. OPRA has developed and successfully rig-tested a gas turbine combustor and fuel injector for conventional pyrolysis oil and is the only gas turbine manufacturer who is able to utilize bio-oil in its turbines. By demonstrating the utilization of the pyrolysis oil in this project OPRA foresee new market opportunities globally, which will lead to increased sales and subsequently an increase in the work force in the Netherlands.In addition, OPRA will be able to demonstrate and promote it's technology to biomass rich countries within Europe, which is expected to accelerate the introduction of this technology. Alucha has successfully developed a commercial scale plastic waste pyrolysis plant for Stora Enso Group and a 10kg/hr pilot scale pyrolysis reactor of paper sludge. Currently, a 100 kg/hour Demo scale pyrolysis reactor is being constructed for SCA. Added value for The Netherlands is the cooperation with the Swedish, Austrian and Polish partners who bring in their experience on catalytic pyrolysis, hydrogenation, biomass pretreatment, diesel engines and social LCA. Exchange of PhD- and MScstudents and coworkers between the different countries is foreseen.

#### Sweden

In Sweden, one of the mainly challenges to reduce  $CO_2$  emissions is the transportation section, which consumes 37% of total energy. Biomass is the only renewable energy which is able to convert to liquid fuel directly. Successful performance of this project can contribute to the aiming. Sweden also have a long history on researching and development of biomass thermal conversion and especially on gasification. All involved partners from Swedish side own strong experiences on biomass thermal conversion from different aspects. Expertise and competence from catalytic pyrolysis from KTH, LCA on bioenergy from RISO will be shared with project partners.

Added value for Sweden is that the experiences and competences on the catalyst development, upgrading bio-oil and generating power from bio-oil via gas turbine and gas engine, which are weak and not so activate yet till now, can be strengthened and promoted. Another added value is the young researcher training, a new PhD student will be enrolled, and this person will move to the Netherlands at least one month within the project.

#### Austria

Relevant current focus areas of Austrian bioenergy related R&D objectives are methods and processes to improve the storage and combustion properties of biofuels (focus on 2<sup>nd</sup> and 3<sup>rd</sup> generation biofuels) as well as the production of secondary energy carriers from biogenic raw materials such as agricultural and industrial residues by e.g. pyrolysis. Consequently, BIOS shall contribute with its vast experience in biomass fuel characterisation and feedstock pre-treatment to the project. A second main task shall be contributions to the improved utilisation of bio-oil in gas turbines whereby the long lasting experiences of BIOS regarding CFD simulations, especially regarding NOx emission formation simulations, shall be utilised within the project. Moreover, BIOS (based on its engineering expertise) shall contribute with techno-economic analyses to the development of an economically sound final process.

Austria shall gain added values from the project especially from the cooperation with the partners from Sweden and the Netherlands by increasing the knowledge regarding catalytic pyrolysis and hydrogenation as well as the optimisation of gas turbines for the operation with bio-oil. This shall form the basis for first decentralised pyrolysis based CHP systems in the future, shall strengthen the biomass pyrolysis research activities in Austria and shall enhance industrial networking in the field.

#### Poland

In Poland the challenge is the  $CO_2$  emission reduction from the power generation sector and to use more biofuels in the transportation sector. The added value for Poland is the cooperation with the Swedish, Austrian and Dutch partners who bring in their experience on catalytic pyrolysis, biomass pre-treatment, and gas turbines. The cooperation in these targets strengthens the exchange of knowledge and experience. Every project partner is responsible for different stages of the production chain. From the Poland's point of view the participation of the small company HIG Polska is crucial for developing the technology. It is a dynamically developing small enterprise from the petrochemical and waste utilization industry. The company produces high-quality n-alkanes from polyolefin wastes. In order to produce high-quality products (e.g. cosmetic kerosene and paraffin), the hydrorafination process of the raw product from pyrolysis is implemented. Nowadays, the new plant for hydrorafination with a processing capacity of hydrocarbons up to 200 tones/month is under construction.

The Institute for Chemical Processing of Coal (IChPW), is a research and development company, which is involved in the research on the efficient use of fossil, renewable and alternative fuels. IChPW has vast experience in the hydrogenation of different oils under pressurized conditions. The laboratory stand for high pressure hydrogenation will be applied for this purposes equipped with a reactor with a volume 0.2dm<sup>3</sup> with industrial catalysts.

In the last 4 years IChPW has conducted research on combustion of process gas from biomass gasification both in dual fuel and gas engines. The research has been conducted in two scales 35kWel and 350kWel, both applications were for combined heat and power production. The research focused on development of process gas treatment methods, tailor-suited for application of gaseous fuels in piston engines. One of the major topics that has been realized was also the modernization of engine intake as well as fuel injection systems, for increasing the efficiency of the combustion process.

# Appendix 2: Photos







Pyne 45

Harsha Mysore Prabhakara visited KTH and made test at KTH, published in IEA Bioenrgy Task 34, PyNe 45, December 2019