Biofuels through Electrochemical transformation of intermediate BIO-liquids

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Description of the deliverable content and purpose

This deliverable contains the white paper, summarising the achievements of the EBIO project, future potential of the technology, current challenges and bottlenecks as well as next steps to advance the technology. The White paper is published at the EBIO website, and in addition, uploaded to ZENODO. The links will be shared on social media.

Content

Unlocking the Future of Green Chemistry: Electrochemical Upgrading of Industrial Bioliquids - A white paper by the EBIO Consortium

In the quest for sustainable bioeconomies, industrial bioliquids like black liquor and pyrolysis liquids hold untapped potential to replace fossil-based resources. However, their complex compositions present formidable challenges for traditional processing methods. Enter electrochemical upgrading—a transformative approach that harnesses renewable electricity to selectively convert bioliquids into high-value fuels, chemicals, and materials under mild conditions.

This cutting-edge technology overcomes hurdles like high energy demands and hydrogen dependency while offering unmatched scalability, sustainability, and compatibility with existing biorefineries. By pairing innovative electrode designs with targeted transformations, such as lignin depolymerization and Kolbe electrolysis, electrochemical systems promise not just greener chemistry but a diversified and integrated bioeconomy. Dive into the possibilities of electrochemical upgrading and explore how it's shaping a low-carbon future.

Introduction

Industrial Bioliquids: Definition and Importance

Industrial bioliquids are liquid bio-based products derived from biomass feedstocks through various processes, including thermochemical, biochemical, and physicochemical methods. Typical biobased liquids produced at industrial scale are black liquor, pyrolysis liquids, lignin-derived fractions, and aqueous bio-processing streams, serving as renewable intermediates for producing fuels, chemicals, and materials, offering an alternative to fossil-based resources[1][2].

The global shift toward a circular bioeconomy has enhanced the importance of industrial bioliquids, particularly black liquor and pyrolysis liquids. Their valorization aligns with sustainability goals by reducing greenhouse gas emissions, fostering resource efficiency, and supporting waste valorization. For example, lignin-derived phenolics and phenolic acids are key precursors for manufacturing bio-based polymers, resins, and solvents[3][4].

Current Challenges in Bioliquid Processing

Despite their potential, processing industrial bioliquids into high-value products presents significant challenges due to their complex and variable composition. Pyrolysis liquids, in particular, are characterized by a two-phase system with high oxygen content, acidity, and instability, while black liquor, derived from the kraft pulping process, contains a mixture of lignin, hemicelluloses, and inorganic salts. This heterogeneity limits their direct application to combustion and gasification processes[5][6].

Many conventional upgrading methods, including hydrotreatment and catalytic reforming, face significant hurdles:

- High Energy Demand: These processes often require elevated temperatures and pressures, increasing investment and operational costs.
- Hydrogen Dependency: Hydrogen supply is a critical bottleneck for thermochemical upgrading, with implications for cost and carbon intensity.
- Limited Selectivity: Achieving precise control over reaction pathways is challenging, leading to the formation of undesirable by-products and reduced catalyst stability[7].

Moreover, the reliance on expensive catalysts and the production of aqueous waste streams further hinder the economic viability and environmental friendliness of these methods.

Role of Electrochemical Upgrading of Bioliquids

Electrochemical upgrading offers a transformative approach to address the limitations of conventional catalytic methods. By utilizing electricity to drive selective chemical reactions under mild conditions, electrochemical systems bypass the need for external hydrogen and high-pressure systems. This process holds particular promise for upgrading black liquor and pyrolysis liquids. A prior fractionation into carbohydrate-based aqueous phase and lignin-derived oil phase further facilitates selective processing.

The advantages of electrochemical upgrading include:

- Flexibility: Electrochemical systems can target specific functional groups in bioliquids, enabling tailored valorization of acids, aldehydes and ketones as well as polymeric fractions.
- Operation at mild conditions and applying electrochemical potentials instead of high temperature limits repolymerization reactions into undesired side products with low reactivity.
- Scalability: Electrochemical setups can be modular and decentralized, allowing for scalability from small farm scale to industrial levels by parallelization. This is, however, still not trivial.
- The combination of proton/electron/hydroxy ion formation by water splitting and usage for reduction and oxidation of target compounds reduces investment costs and makes efficient use of the entire electrode surface area for production of valuable compounds.
- Sustainability: When powered by renewable electricity, electrochemical processes offer a low-carbon footprint, making them integral to green chemistry initiatives[8][9].
- Application of water and commonly applied salts as electrolytes reduces the footprint of applying toxic chemicals. Especially black liquor is a suitable electrolyte and does not require any modification prior to electrochemical processing.

For example, decarboxylation of organic acids via Kolbe electrolysis can remove undesirable carboxyl groups, while the electrochemical hydrogenation of sugars and furans into value-added chemicals shows potential for producing biofuels and solvents with minimal environmental impact[10][11]. Additionally, lignin depolymerization via electrochemical means has been shown to generate valuable aromatic compounds such as vanillin, vanillic acid, benzoic acid etc.[12].

The role of electrochemical upgrading extends beyond isolated reactions, seamlessly integrating with biorefinery frameworks to offer synergies with upstream and downstream processes. This integration enables the simultaneous production of biofuels, specialty chemicals, and energy, contributing to a diversified and sustainable bioeconomy.

Fundamentals of Electrochemical Upgrading

Principles of Electrochemical Processes

Electrochemical upgrading uses electrical energy to drive selective redox reactions, enabling the modification of bioliquid components under mild conditions. In direct electrochemical processes, the anode and cathode serve as catalysts for these reactions, allowing transformations such as reduction, oxidation, and hydrogenation. Indirect conversion produces activating oxidants, which are then blended with bioliquids to weaken and cleave specific chemical bonds. This versatility is essential for upgrading black liquor and pyrolysis liquids, where diverse compounds need targeted and selective conversion processes[12][13].

Figure 1: General electrolysis cell platform with 3 cm² surface area (left) and 100 cm² (right) covering different TR Levels. The production line of cells and electrodes can be scaled up to commercial scale.

Targeted Transformations for Bioliquids

For both black liquor and pyrolysis liquids, electrochemical processes can be used to selectively transform key components:

- Kolbe Electrolysis for Acid Removal: Organic acids are an essential fraction of pyrolysis liquids, acting as emulsion-stabilizer and catalysing undesired aging and polymerization reactions. They can be decarboxylated using Kolbe electrolysis into alcohols and hydrocarbons[14].
- Hydrogenation of sugars, anhydrosugars and furanes: Electrochemical hydrogenation can convert sugars and furanic compounds from the aqueous phase of pyrolysis liquids into valuable chemicals like alcohols and polyols[15][16]. The hydrogenation of the carbonyl group significantly enhances the stability of the bioliquids.
- Depolymerization of Lignins: Lignin, which makes up the oil phase of pyrolysis liquids and is also found in black liquor, can be depolymerized via electrochemical methods. This process breaks down the complex lignin structure into smaller, commercially valuable aromatic compounds[17][18].

Catalysts and Electrodes

The efficiency and selectivity of these reactions depend heavily on the choice of electrodes. For hydrogenation reactions transition metal-based electrodes such as nickel, platinum, and copper are commonly used for their conductivity and catalytic properties/19/. Recent innovations have focused on designing advanced electrodes, such as bimetallic catalysts (e.g., Cu-Pd alloys), and carbon-based materials (e.g., boron-doped diamond), which show promise for enhancing the efficiency of these transformations/20. Specifically BDD applied as anode

for Kolbe electrolysis and production of activating oxidants shows a combination of high selectivity and chemical stability against overoxidation.

Process Configurations

Electrochemical upgrading of bioliquids should be carried out in continuous Flow Reactors: These systems are suitable for large-scale operations, offering enhanced mass transfer and throughput as well as controlled conditions at the electrode surface. They are particularly well-suited for the electrochemical upgrading of black liquor and pyrolysis liquids[21][22]. Critical are the following aspects:

1. Fractionation of bioliquids into compound groups increases the process control and selectivity. Hydrogenation of water-soluble carbonylic and acidic compounds can be processed separately from lignins, which are dominant in the oil phase and are to be depolymerized.
2. Paired electrochemical conversion of reductive hydrogenation of carbonyls and oxidative decarboxylation and depolymerization results in comparable electron- and ion exchange rates. This makes pairing highly suitable for value-added production and full utilization of the electrolyser.
3. While cathodic hydrogen production cannot be avoided, anodic oxygen production is nearly entirely quenched when conducting Kolbe/non-Kolbe electrolysis, lignin depolymerization and production of activating oxidants, such as peroxydicarbonate. This facilitates the usage of simple undivided cells. The pure hydrogen produced can be valorised for hydrogenation, reduction or in fuel cells.
4. Specifically thin black liquor can be directly applied for electrochemical conversion, as it contains significant amounts of electrolytes and shows low viscosity, especially when processed at elevated temperatures of around 100 degC.
5. Pyrolysis liquids, on the other hand, require electrolytes to be added, preferably acids or other compounds which are either anyway produced or present at the biorefinery.

Co-Processing of Upgraded Bioliquids for Biofuels Production

In EBIO two routes of biofuels production, co-Fluidised Catalytic Cracking(FCC) and co-Hydrotreatment(Co-HT) have been identified and pursued. Both routes focused on co-processing of the upgraded bioliquids with suitable refinery streams, with respect of miscibility, conversion conditions and target products:

1. Co-FCC was performed applying a microdowner unit[23]. As refinery stream vacuum gas oil was applied. It was found that the presence of bioliquids does not alter the product composition. Still, the presence of rather stable phenolic compounds require to modify the conversion conditions towards slightly higher temperatures.
2. Co-HT with light cycle gas oil has been demonstrated with excellent catalyst stability and product composition.

Scale-Up Considerations

From Laboratory to Pilot Scale

Scaling electrochemical processes for bioliquids upgrading requires various phenomena, including mass transfer, applied potential and current density window of operation. Handling a mix of organic components with a wide range of molecular weights, complicate the optimisation process significantly. Generally high flow rates are beneficial for removing bubbles and product compounds from the electrode surface. Still, high shear rates near the walls result in depletion/margination of lignin in the near wall area. In our work of lignin depolymerization flat electrode surfaces have shown sufficient current densities with values above 200 mA/cm2, even at low flow rates. Those simple electrode designs further minimize the risk for dead zones in which repolymerization or overoxidation to CO2 could occur. Still, further innovations in reactor design, such as modular and scalable electrodes, are critical for improving process efficiency[24][25]. Biofuel production, either in standalone or co-processing mode requires extended campaigns to evaluate catalyst stability and product composition and properties with respect to specs and standards.

Economic Feasibility

Electrochemical systems powered by renewable electricity, such as solar or wind, can reduce energy costs while lowering the carbon footprint. Lifecycle cost analyses of pilot-scale electrochemical upgrading demonstrate potential cost savings of up to 30% compared to traditional thermochemical methods[26]. Moreover, coupling electrochemical upgrading with waste heat recovery or integrating it into refinery processes (e.g., co-hydroprocessing and co-FCC) can enhance the economic viability of the entire biorefinery[27]. Still, the dominant cost factors for electrochemical bioliquids upgrading are the electricity costs and the electrolyser investment costs. Significant efforts need to be made to develop low-cost technologies for renewable electricity production and take further steps to reduce the electrolyser costs by standardization, mass production, replacement of costly materials etc. With respect to electrode materials, neither anode nor cathode materials have shown significant signs of deactivation. BDD as rather costly material has shown excellent stability even under extreme conditions, resulting in expected life-times of several years.

Regulatory and Environmental Considerations

Electrochemical upgrading processes produce fewer hazardous by-products compared to traditional thermochemical methods, making them attractive from an environmental compliance perspective. Lifecycle assessments (LCAs) of electrochemical processes powered by renewable energy have demonstrated a reduction in carbon intensity by up to 60% compared to fossil-based upgrading methods[28][29]. In addition, the use of an electrolyser, as compared to a combination of electrolyser for hydrogen production and high pressure hydrogenation reactor, significantly reduces safety and environmental concerns.

Integration into Biorefineries

Compatibility with Existing Biorefinery Processes

Electrochemical upgrading can be integrated into existing biorefinery frameworks to enhance biomass valorization. For example, black liquor and pyrolysis liquids can be upgraded electrochemically to valuable aromatic compounds such as vanillin, syringaldehyde and benzoic acid, reducing dependency on external chemical feedstocks[30]. In EBIO we have shown that especially thin black liquor as produced in the digester can directly be applied for electrochemical conversion. The process has been demonstrated at conditions relevant for pulp mill operation, requiring no temperature adjustments prior or after electrochemical conversion.

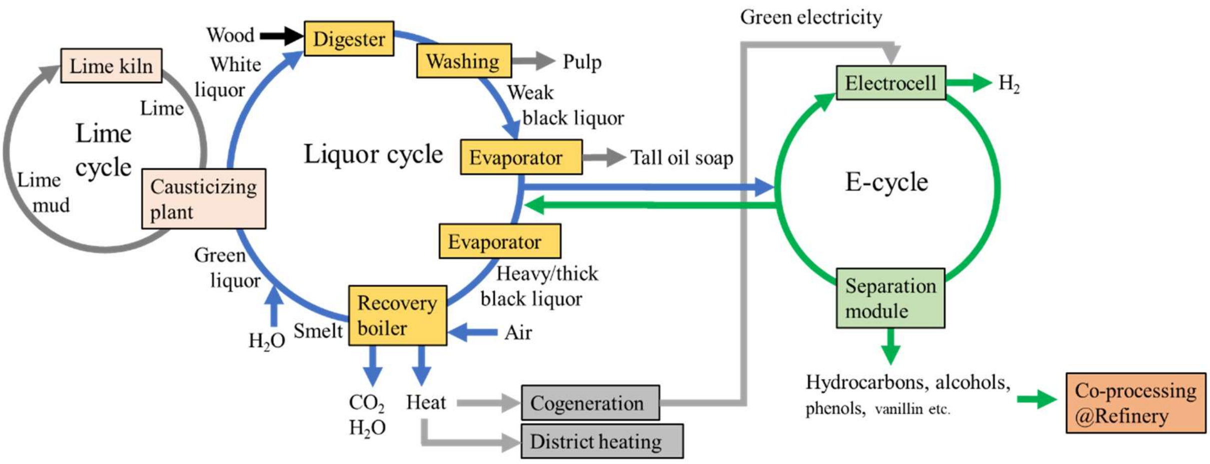

Figure 2~~1~~: Simplified integration of an electrochemical cycle into a kraft pulp mill, based on the kraft pulping scheme of Tran and Vakkilainen.[31]

Integration with fast pyrolysis requires crude bioliquid fractionation as well as addition of electrolytes. Products from electrochemical upgrading are blended prior to catalytic upgrading and co-processing.

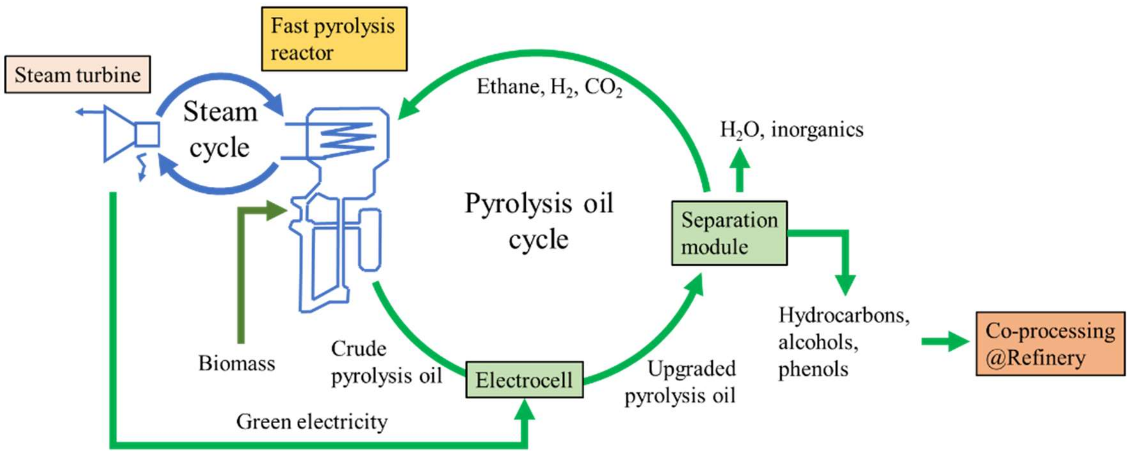

Figure 3~~2~~: Excess electricity produced in a pyrolysis unit for intermediates for advanced biofuels. In blue the existing Empyro-type pyrolysis plant is presented.

Modular and Decentralized Systems

Electrochemical systems are well-suited for decentralized deployment in biorefineries, enabling localized upgrading of bioliquids and reducing transportation costs[32]. These systems can be powered by renewable energy sources, such as wind or solar or available hydropower, further improving the environmental and economic sustainability of biorefineries[33].

Energy Integration

Electrochemical systems can also benefit from energy integration within biorefineries. Surplus electricity or heat generated by other refinery processes can be used to power electrochemical upgrading, enhancing overall efficiency and reducing operational costs[34].

Lifecycle Analysis

LCAs of biorefineries incorporating electrochemical upgrading have shown a 50% reduction in carbon intensity compared to standalone thermochemical methods[35]. These findings highlight the importance of integrating electrochemical processes to optimize the environmental performance of biorefineries.

Case Studies and Applications

Electrochemical Decarboxylation of Carboxylic Acids

A continuous flow electrochemical small pilot reactor was successfully tested for the Kolbe/non-Kolbe electrolysis of aqueous electrolytes containing single acids and mixtures. The effect of acid type on the process selectivity has been demonstrated[36].

Anodic Lignin Depolymerization into Mono- and Oligomeric Aromatic Compounds

Electrochemical oxidation of lignin into phenolics has been successfully demonstrated at pilot scale, producing a range of aromatic compounds such as vanillin, syringaldehyde, vanillic and benzoic acid[37].

Electrochemical Hydrogenation of Aldehydes and Ketones

Electrochemical hydrogenation of carbonyl functionalities has shown to significantly enhance the stability of bioliquids against aging and coking[38]. The suitability of transition metal and carbon-based cathode materials has been demonstrated.

Integration of Renewable Energy

The focus of EBIO was to demonstrate electrochemical processes under conditions of constant voltage or current. The exploration of fluctuating availability and costs of renewable electricity was not targeted within the project. In this respect, the process is suitable for cases such as electricity production from excess heat at biorefineries and hydropower [39]. In principle the option for alternating operation based on the availability of cheap renewable electricity is an option, but requires significant cost reductions of electrolysers, as they are not utilized 24/7, and the application of a certain small potential at all times to minimize oxidation of cathode materials under low loads.

Challenges and Research Opportunities

Technical Challenges

Challenges such as electrode durability, reaction efficiency at scale, and the need for low-cost renewable electricity remain critical areas for development. Further research into advanced electrocatalysts, improved reactor designs, and the optimization of process parameters will be key to enhancing the efficiency of electrochemical upgrading[39][40].

In the specific conversion process of Kolbe electrolysis of acetic acids, methyl-radicals are produced, which can cause degradation of BDD electrodes. In addition, when applying transition metal electrodes for oxidative lignin depolymerization, the combination of high voltages and high pH electrolytes is required to achieve high current densities, but can result in leaching of metals into the electrolyte.

Research Opportunities

Future research opportunities include the exploration of novel electrochemical techniques for the selective upgrading of complex bioliquids, as well as the integration of electrochemical upgrading with other sustainable technologies, such as biogas production or waste-to-energy processes[41][42].

Specifically the identification and optimisation of stable electrode materials for electrochemical direct oxidation and oxidative decarboxylation reactions is of high importance. Critical raw materials are to be avoided if possible.

For a paired electrochemical concept the ultimate goal is to operate in undivided electrolysers. This requires smart electrolyser concepts with products that can be processed together in downstream operations. Examples are reduction of cellulose- and hemicellulose-based carbonyl functions paired with phenols produced from lignin. Alternatively, the anodic oxidation processes are paired with cathodic hydrogen production.

Integration of electrochemical conversion processes into biorefinery concepts requires significant efforts, with respect to crude bioliquid and product fractionation and purification. Especially the development of efficient technologies for the separation of higher value compounds from a complex bioliquid matrix is of high importance.

Conclusion

Electrochemical upgrading of bioliquids holds significant promise for the efficient and sustainable processing of industrial bioliquids such as black liquor and pyrolysis liquids. By leveraging electrochemical techniques such as Kolbe electrolysis, carbonyls hydrogenation, and lignin depolymerization, these bioliquids can be converted into high-value chemicals and fuels. When integrated with co-hydroprocessing and co-FCC technologies applying established refinery processes for fuels and chemicals production, electrochemical upgrading can contribute to a more sustainable bioeconomy, reducing reliance on fossil fuels and promoting circular resource use. Further research and innovation will be essential to fully realize the potential of electrochemical upgrading and its role in future biorefinery ecosystems[43][44].

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