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An Overview of Recent Advances and Novel Synthetic Approaches for Lignocellulosic derived Biofuels

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ABSTRACT

One of the most important challenges to overcome at present is to reduce the dependability on the fossil fuels and replace them with green and environmental friendly substitutes. In this respect, biofuel conversion from lignocellulosic materials could prove to be an efficient process. The current review presents the most recent advances and novel synthetic approaches in utilizing lignocellulosic substrates for biofuel production, such as scheming of new/enhanced catalysts, waste valorization, using genetic modifications to improve organism traits, development and/improving the new or step integration processes, etc. Such new strategies can prove to be important tools and augment the use of renewable fuels globally. The present review summarizes the most recent strategies, advances and novel synthetic approaches offered pertaining to catalytic/non-catalytic pretreatments/transformations of lignocellulosic materials, carbohydrates based carbohydrates and technical lignin into biofuels.

Keywords: Waste valorization; biofuels; lignocellulosic biomass; catalysts

INTRODUCTION

The demand for renewable sources, especially from biomass, is increasing promptly and is expected to ascend more due to concern for the rapid depletion of fossil resources. Of particular interest are the liquid fuels due to the availability of their vast infrastructure and novel synthetic approaches gaining significant attention.

Fossil fuels used in transportation mainly comprise of hydrocarbons separated into gasoline, diesel and jet fuel. Due to their significant environmental impact, the biofuels such as bio-alcohols and biodiesel are appealingly suitable alternatives. Currently, these biofuels are produced commercially with established technology on a large scale using edible commodities like vegetable oils, starch and sugar.

To produce the biofuels, low value biomass such as crop residues and municipal waste is preferred as a primary source, whereas residual oils/fats and algae are among the secondary source.

Cellulose is a polysaccharide comprising of glucose monomer units, whereas hemicellulose being the amorphous polysaccharide contains xylose and hexoses. Generally, the approach followed for biofuel synthesis using polysaccharide is by deconstructing biomass into monomers at low temperature and then converting into platform compounds. Such compounds are either fuels themselves or require further processing to be converted into biofuels. During the biofuels production, overall energy density can be improved by extending the Carbon-Carbon bonds between intermediates and downscaling the parent feedstock oxygen content (Li et al. 2018b).

Lignin is also a major component of the lignocellulosic biomass besides cellulose. It also happens to be the most exuberant natural biopolymer. Although lignin has various types due to linkages and isolation, the technical lignin is the priority of choice on an industrial scale. Being the byproduct of paper and pulp industry or ethanol production, the technical lignin can be sustainable choice as a bulk raw material. Hence, the economy of the existing paper and pulp industries can be significantly improved by utilizing the technical lignin acquired via the industrial streams (Wendisch, Kim & Lee 2018).

In order to increase the diffusion of lignocellulosic materials, improvements in biomass pretreatment and fractionation step are mandatory. In this respect, one of the works (Ji et al. 2018) developed a multipurpose fractionation where, in the first place, hemicellulose was extracted and converted into furfural followed by lignin extraction and upgradation to phenolic monomers through hydrogenation, and finally, enzymatic hydrolization of cellulose rich solid into glucose. Similarly, another work (Yang et al. 2019) employed the method of lignin-first biorefinery where the first step involves catalytic delignification followed by generation of convertible glucan chains acquired through suspension of cellulosic microfibrils. Employing the same route, a highlighting research (Patri et al. 2019) identified synergistic mechanisms for biomass deconstruction through experimental and computational analysis, hence generating new possibilities.

Another approach towards this is to work with the lignin fractions individually during, after or before the pretreatment. The work following this approach (Morone, Pandey & Chakrabarti 2018) pretreated rice straw using 4 different methodologies to improve the accessibility of cellulose. When the results were compared, the alkaline wet air oxidation (AWAO) showed to have great accessibility, energy efficiency with the minimal generation of waste. In context of lignin approach, (Guo et al. 2018) converted the lignin into lignin oil with high yield using Pd/C catalyst followed by hydrodeoxygenation. Moreover, they used the biphasic reaction system to convert cellulose and hemicellulose into 5-hidroximetil furfural and furfural. Similarly, the researchers (Qiu et al. 2019) developed two catalysts (MoxC/carbon nanotube and Ru/CMK-3) to successfully retain hemicellulose and extend feedstock in lignin-first biorefinery of softwood. Finally, (Jin et al. 2018) successfully developed catalytic system to produce liquid fuel components via depolymerization of kraft lignin.

LIGNOCELLULOSIC TO BIOFUELS

PYROLYSIS

One of the most explored possibilities related to derivation of biofuels from lignocellulosic is pyrolysis. Ansari et al. (Ansari et al. 2019) was first to conduct a comprehensive pyrolytic study of cellulose, lignin and xylan. In developing bio-oils, biopolymer reaction maps prove to be an important tool in optimizing the conditions for achieving higher yields. Furthermore, (Shen et al. 2018) studied copyrolysis of nonmetallic fractions with rice husk. Such an approach helped improve the overall properties and thermal efficiency of fuels. When the heavy metal content in the char is lower, the generated syngas can be enacted for fuel synthesis. Further looking into bio-fuels, a work deserves a mention (Zhou et al. 2019) that depolymerized 3 lignin types using microwaves. The HSZ-based solid acid catalysts were used wand the results showed synergistic effect among solvent-catalyst polarity and acidity.

Charcoal on the other hand remains a popular fuel due to its viably slow pyrolytic production at low temperature. Comparatively, in one of the studies (Chandrasekaran et al. 2019), authors analyzed the efficiency of an enhanced reactor to produce charcoal in rural areas. They performed tests mainly in order to reach optimum value for the final pyrolytic temperature. Pertaining to waste valorization, the authors in the study (Zhang, Zhang & Li 2019) showed the potential of co-hydrothermal carbonization of PVC and pinewood sawdust in dechlorination of PVC and improving combustion properties of hydrochar.

LIQUID BIOFUELS

At this point of time, it has become crucial need to convert fuels from biomass. However, such a conversion needs to overcome the associated technological obstacles and loopholes for global contribution in energy matrix. For this, several studies are underway to resolve such obstacles and loopholes in biofuel conversion from lignocellulosic biomass. For instance, the study (Kaur & Kuhad 2019), pretreated the rice straw with H_2SO_4 and NaOH followed by hexose and pentose fermentation resulting in proficient and high yield of ethanol production. Furthermore, after alkali treatment, a highly pure lignin was precipitated from spent liquor, making it possible to recover all the biopolymers in a single process. Moreover, (Boboescu et al. 2019) developed a new approach of obtaining glucose from residue streams of sorghum biomass through delignification and cellulose hydrolysis. Similarly, Muthuvelu et al. (Muthuvelu et al. 2019) produced the bioethanol using 4 different lignocellulosic biomass residues. In this context, (Fan et al. 2018) explored the conversion of sugarcane molasses into ethanol observing an upturn in production, ethanol concentration and concentration rate. Compared to fermentation of pretreated SCB alone, an improvement in system liquidity, shorter fermentation time and higher yield was observed. Regarding SCB, the study (Fan et al. 2019) used mixed-feedstock fermentation to enhance the co-generation of cellulosic ethanol and methane from SCB. This approach integrated the starch-rich waste in mixed fermentation, thereby allowing to overcome the unparalleled supply of SCB and molasses. The research (Zhang et al. 2019) explored the possibility of converting residuum into bioethanol via first-refinery apple wood. The researchers used different catalysts (Ni or Ru/C) to enhance the substrate residuum.

Various studies have proposed the non-modified Biobutanol as a potential replacer for gasoline. A recent report (Husin et al. 2019) shows the potential of waste sago hampas for butanol production. The researchers used separate hydrolysis and fermentation along with simultaneous saccharification and fermentation coupled with amylase and cellulose mixture for the production. In this aspect, the most recent technology is revealed by the work (Galebach et al. 2019) where in the researchers depolymerized the biomass using supercritical methanol and CuMgAl catalyst following hydro-deoxygenation over copper catalyst in a single reaction step. The researchers were able to acquire high yields of C_2-C_9 alcohols.

GASOLINE

In the study (Peng et al. 2019) the authors developed a new method to convert rice straw into C_5 and C_6, furfural and cyclic ketones with high yield using multifunctional catalysts and a biphasic reaction system. End result products could be a possible candidate for biogasoline due to the low oxygen content. The alternative approach was used by (Sousa et al. 2018) to produce kernel/palm oil based linear and branched

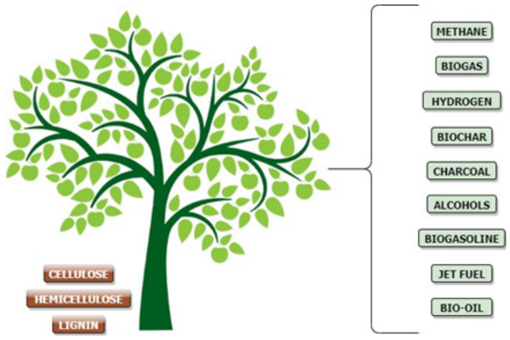

FIGURE 1. Schemetic Reprentation of Biofuels from Biomass

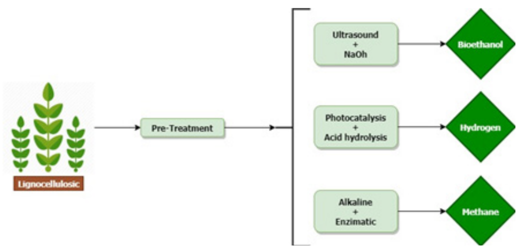

FIGURE 2. Schematic Representation of some works reported in this paper (Galindo-Hernández et al. 2018; Muthuvelu et al. 2019; Zou, Zhang & Xu 2018)

cyclic biohydrocarbons using thermally activated Hbeta zeolite catalyst with distillation ranges of aviation and transportation fuels. Finally, (Li, Otsuki & Mascal 2018) investigated the efficient conversion of levulinic ester and its products into cyclopentanes mixture. The acquired mixture showed high octane ratings and excellent flow properties.

Several works have focused on jet fuels (Pan et al. 2020; Pierobon, Eastin & Ganguly 2018) Among others, the work (Yang et al. 2018) took an interesting approach and used Grindelia squarrosa biocrude as a feedstock and a novel one-pot catalytic process to achieve a denser jet/missile fuel.

One of the promising and valuable renewable biomass-derived substrates for bioproducts is Levulinic acid (LA). Ethyl levulinate is one of the derivatives of LA and is prominently suitable liquid fuel that can easily be produced from lignocelluloses. In the work (Ghosh et al. 2018), authors investigated the combustion kinetics of ethyl levulinate and proposed its use suitably as gasoline fuel than diesel fuel, prevalently shown by literature. Cabanillas et al. (Cabanillas et al. 2019) converted methyl levulinate into γ-valerolactone (GVL) using catalysts with zeolites (BEA-75 and ZSM-5) supported zirconium. The converted product showed impressive properties for usage as fuel/fuel additive.

Many researchers in the past have studied and shown the applications of furfural and its derivatives, particularly into diesel or jet fuel. Kocharmann et al. (Köchermann, Mühlenberg & Klemm 2018) successfully produced liquid hemicellulose fractions via organosolv pulping processes. The lignin content in the fractions was apparently low. Continuous tube reactor was used to examine the conversion kinetics of hemicellulose and D-xylose into furfural. Similarly, (Bizzi et al. 2019) reported ultrasound-assisted acid hydrolysis as possibly a suitable single step alternative process for biomass conversion into furfural. On the contrary, (Malkar et al. 2019) deals with a novel catalyst based route to upgrade biomass intermediates into aldol adducts. Such adducts showed impressive enough properties for possible use as jet fuel additives. Finally, an investigation was carried out by (Feng et al. 2019) of biomass derived synthesis of 5HMF to develop a sustainable betaine-based catalysts. Besides, the catalyst, choline-O-sulfate, was developed that efficiently converted fructose into HMF.

GASEOUS BIOFUELS

Hydrogen fuel has gravitated much attention in recent days due to being a clean energy source. In this regard, the work of Zhou et al. (Zou, Zhang & Xu 2018) is highlighting as they used an industrially encouraging strategy to convert cellulose into hydrogen using photo and acid catalysis. On the contrary, in their work (Galindo-Hernández et al. 2018), the researchers used alkaline hydrogen to pretreat Agave tequilana bagasse followed by enzymatic saccharification with celluloses. The overall yield of methane and hydrogen was improved by 1.5 and 3.6 times due to the pretreatment with hydrolyzate secreted from saccharification process, compared to without pretreatment.

Wide and extensive studies have already taken place in the past regarding biogas generation through anaerobic digestion of lignocellulosic materials. For example, (Usman Khan & Kiaer Ahring 2020) synthesized methane by pretreating the residues via wet explosion succeeded by enzymatic hydrolysis. The results indicated that the highest yield of methane can be achieved through severe pretreatment, thus enabling the possibility of using biorefinery lignin as a substrate for biogas production. Recently, based on their modeling and experimental results, authors in their work (Yun, Ramírez-Solís & Dupont 2020) proposed a promisingly cleaner production of biomethane from lignocellulosic biomass via thermochemical conversion. The simulation results obtained from the autothermal plant showed a justified comparison of CH_4, H_2 predicted values to those modelled in literature mentioning wood gasification. In relation to biorefinery, the study (Rocha-Meneses et al. 2019) investigated the production of gaseous fuels using bioethanol production waste. The results showed the waste to possess high energy value and contribute towards overall increased output from biomass.

NEW REACTION STRATEGIES

To synthesize the jet fuel compatible biofuel, an efficient 2-step cascade process is employed. Such a process is based on catalytic system containing Pd/C and Eu(OTf)3 and incorporates NaOH-catalyzed aldol condensation followed by HDO using hydrogen. The condensation yields branched furan compounds (C11 and C12) with upto 96% of yield, following hydrogen coupled HDO to produce branched alkanes (2-methylundecane and 2-methyldecane) with upto 98% of yields (Keskiväli et al. 2017). Recently, Li et al. (Li et al. 2018a; Wang et al. 2018) have reported an alternative but more efficient way of cascade catalytic process which used solid catalysts and multiple substrates to alkylate and HDO of 2-methylfuran. They used pressure tubes to alkylate and Teflon tube containing solid catalysts for the follow up procedure of HDO. Similarly, Qui et al. (Qiu et al. 2017) simplified the cascade reactions for biofuel synthesis by performing in one-pot. They did this by producing 5-(hydroxymethyl)-2-(dimethoxymethyl) furan from fractose through 2 step dehydration process. They used a methanol solvent system employing facile catalyst (SiO2-HNO3 and DMSO). However, for more industrial applications, it is highly required to use continuous flow reactions. Kwon et al. (Kwon et al. 2019) used a fixed bed reactor with continuous flow to selectively condense 2-methylfuran and furfural to C_{15} compounds. The fixed bed was supported by phosphotungstic acid catalysts. Moreover, Weng et al. (Weng et al. 2018)higher alcohols could be used as a blending agent for diesel fuels. Herein, carbon supported phosphated ruthenium-molybdenum (RuMoP successfully converted sorbitol into renewable alkanes and alcohols using continuous trickle bed reactor system supported by Ru-Mo catalysts.

NEW CATALYST SYSTEMS

Great potential has been shown by many novel homogenous and heterogeneous catalyst systems in synthesizing renewable biofuels and making them a commercially possible (Bender, Dabrowski & Gagné 2018; Murphy & Xu 2018; Singh 2018; Sudarsanam et al. 2018; Trombettoni et al. 2018; Zhang, Song & Han 2017). As such, Liu et al. (Liu et al. 2018a) produced a catalytic system based on HCL and $ZrO(OH)_2$ and used it to effectively catalyze the fructose to 5-ethoxymethylfurfural (gasoline additive) and EMF to 2,5-bis(ethoxymethyl)furan. Furthermore, Xu et al. (Xu et al. 2017) found that using Mn_2O_3, as high as 96% of yield could be achieved of C_{10} oxygenates in aldol condensation of furfural with mild conditions. Similar high yields of C_9 and C_{10} alkanes were obtained over Pd/C and Pd-FeO_x/SiO_2 catalysts. Likewise, Liu et al. (Liu et al. 2018b) could achieve >90% yield of condensation intermediates via aldol condensation of different furfural species. The researchers used cyclic ketones over NaOH catalyst and mild conditions. Subsequently, at mild conditions, HDO condensation intermediates were catalyzed into cycloalkanes (gasoline) using amorphous zirconium phosphate fused with Pd/C and resulted in 76% of yield. Furthermore, HDO has also been effective in conversion of sorbitol into gasoline products having high octane number with 70% yield using catalysts supported on zirconium phosphate (Kwon et al. 2018). On the other hand, carbon supported Ru-Mo catalysts were shown to possess low affinity towards C-C bond of sorbitol. Low affinity made these catalysts highly selective for C_6 products in liquid and gas phases (Weng et al. 2018) higher alcohols could be used as a blending agent for diesel fuels. Herein, carbon supported phosphated ruthenium-molybdenum (RuMoP.

BIOMASS-DERIVED FUEL MOLECULES

Fuel molecules derived from biomass packing high energy have emerged to be new and novel materials (Zhang et al. 2018). Tang et al. (Tang et al. 2017) were the first to demonstrate and synthesized jet fuel range cycloalkane (trimethyl-cyclohexane). They coupled aqueous phase of glycerol and HDO of isophorone to achieve a good yield of 67%. In the same way, utilizing the easily accessible feedstock, Liu et al. (Liu et al. 2019) employed a 2 steps conversion method to produce a mixture of C_{12} and C_{18} branched polycycloalkanes from cellulose. The mixture possessed high density and yield of 74.6%. In the first step, they selectively converted the cellulose to 2,5-hexanedione, which was then transformed into polycycloalkanes over a dual bed catalyst system via aldol condensation-hydrogenation-HDO. Wang et al. (Wang et al. 2017b) used the similar approach to synthesize high density cyclopentycyclopentane with a high yield of 70%. This biofuel is a novel high density jet or rocket fuel and was synthesized using hemicellulose derived cyclopenanone as a substrate.

BIOFUELS FROM TECHNICAL LIGNIN

Two of the approaches are commonly taken in order to synthesize lignin-derived fuels; one-pot depolymerization and bio-oil upgradation. The bio-oil upgrade is accomplished by degrading biomass through chemical or biochemical means (Becker & Wittmann 2019; Ren et al. 2019). Hydrocarbons can be converted into transparent liquid form under reductive conditions, with carbon numbers from C_6-C_9 and C_{12}-C_{17} (Shao et al. n.d.; Wang et al. 2017a). To contain the radicals from repolymerizing and condensing, genreally a solvent based lignin hydro treatment is performed. However, solvent free approaches are preferred due to the associated issues of cost and recycling.

NEW REACTION STRATEGIES

Hita et al. (Hita, Heeres & Deuss 2018) used a solvent free approach to hydrotreat the technical lignin via inexpensive Fe-based limonite catalyst. They achieved 41 wt% of yield with mostly alkylphenolics and aromatics, which can aid to biofuel production. Similarly, Wang et al. (Wang et al. 2017a) produced cyclohexane derived alkanes (jet fuel/diesel range) using one-pot HDO of softwoof lignin. The process was carried out using abundant earth metals such as Zn, , Ni, Fe, Cu and along with Ru on HY zeolite catalysts. In the synthesis of hydrocarbon fuels using technical lignin, the one-step approach has proved to show promising results but the associated problems often cause the low productivity, which can be overcome by elongating the residence time and increasing catalyst-lignin ratio.

Bio-oil is a complex compound mixture formed under severe conditions during biomass degradation. HDO with hydrotreating catalysts can be used to upgrade this complex mixture to the quality level of traditional fossil fuel pools. Usually, conventional hydrotreating catalyst based HDO process requires high pressurized hydrogen but recent novel methods have been proposed that use noble/transition metals supported on molecular sieves to overcome this need (Li et al. 2017). Furthermore, it was possible to successfully upgrade the fast pyrolysis lignin vapors using conventional hydrotreating catalysts such as HZSM-5. An organic liquid with 70 wt% oxygen deprived aromatics was synthesized. The key to minimize the repolymerization reactions during pyrolysis depends upon intimate contact between catalyst and lignin, which requires further research to investigate (Zhou et al. 2016).

Anderson et al. (Anderson et al. 2017) has reported a tandem lignin-first biorefinery method, further extended by Rinaldi and Cao et al. (Cao et al. n.d.; Rinaldi 2017). This method, termed as reductive catalytic fractionation, makes the use of active stabilization and hence selectively extracts lignin from the whole biomass. The extracted lignin is an efficient phenolic resource. Such alternative approaches as this may effectively be used in flow reactors to improve an overall mass and energy balances. The hydrocarbons derived from lignin could therefore be an efficient and green substitutes for gasoline, diesel and aviation fuels.

NEW REACTION STRATEGIES (BIOCHEMICAL)

Recently, biochemical conversion of technical lignin into lipids is seen as a significant route with potential for biofuel synthesis. Such technical lignin based bio-converted lipids can possibly replace chemicals originating from fossil fuels i.e. a better sustainable approach for biodiesel production. Among the different species of bacteria, *Rhodococcus opacus* was shown to produce oxygen/laccasses treated lipids from technical lignin such as Kraft lignin. The concentrations of lipids were as 0.07 and 0.15 g/L (Wei et al. 2015; Zhao et al. 2016). However, the former study investigated only one oxygen treatment condition. A recent study expanded on the oxidative depolymerization and its resulting effect on bioconversion of lignin (Abdelaziz et al. 2019). Combined oxygen treatment and fermentation is shown to enhance the lignin bioconversion but needs further investigation. The highest lipid production from lignin to date (1.83 g/L) was achieved using fed-batch fermentation integrated with combinatorial pretreatment (Liu et al. 2018c) Nevertheless, a similarly high lipid concentration was produced using lignin from corn stover, which is chemically different from kraft lignin.

CONCLUSIONS AND FUTURE PERSPECTIVES

At present, the field of biomass conversion is witnessing many interesting developments. New catalysts and reaction systems are helping enhance the biomass conversion and utilization. There are mainly 2 different approaches towards valorization of biomass; dedicated approach and or using side products. The former approach promises an augmented production while the later one carries the most near term promise. One of the major highlights of the present review is the paper and pulp industry and possible strategies to synthesize the renewable fuels from the cellulose and hemicellulose parts of the wood. Keeping in consideration the proposed novel approaches, many technology related questions can be posed here such as if valorization should be brought in use of the technical lignin streams available at paper and pulp industry or should more novel alternatives be considered as priority? Finally, if the paper and pulp industries are willing to implement such novel strategies? Another important aspect to keep in mind is the sustainable need of silviculture for sizable amount production of renewable fuels. It is obvious that the use of biomass derived biofuels is just the part of the solution when volumes of transportation fuels required worldwide is considered in mind.

DECLARATION OF COMPETING INTEREST

None.

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