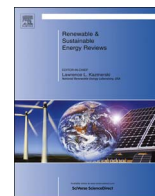




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Recent progress on catalytic pyrolysis of lignocellulosic biomass to high-grade bio-oil and bio-chemicals

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ABSTRACT

Pyrolysis converts lignocellulosic biomass to bio-oil that can be a precursor to fuel and chemicals for industries. The bio-oil contains high oxygenates fractions that deteriorate the bio-oil fuel properties. Catalysts acted to upgrade the bio-oil through selected bond cleavage reactions such as deoxygenation, cracking, decarbonylation and others reactions. Bulk and supported acid or base catalysts in biomass pyrolysis tailored the production of high-grade bio-oil. The catalytic biomass pyrolysis is an approach that is reliable for producing quality renewable fuel and chemical precursors. This paper elucidated recent studies on catalytic pyrolysis of lignocellulose biomass to renewable fuel grade bio-oil and chemicals. The review discussed the various principal activities on biomass characteristics and their potentials in pyrolysis process to produce the high-grade biofuel precursor. The possible processes used in perpetuating the pyrolysis devolatilisation of biomass are also appraised along with catalysts type, and their catalytic activities in the production of renewable bio-oil and bio-chemicals. Therefore, catalyst development for the upgrade of bio-oils from pyrolysis of biomass to renewable fuel and chemicals precursor remains a topical issue.

1. Introduction

Fossil fuels have satisfied the fast growing energy demand for transportation fuel and industrial revolution. However, increasing concern on the fossil fuel depletion and global climate change are the impetuses for the search for an alternative and carbon neutral energy resources. Attention has shifted to non-food lignocellulosic biomass, as one of the most significant renewable energy resource for liquid fuel and chemicals synthesis. The lignocellulosic biomass has been carbon neutral to fulfill the global reduction in carbon dioxide emission. Also, they are abundant and readily available to meet the global fuel demands [1]. As the precursor of fossil feedstock, the use of biomass in energy application for heating, power and transportation requires multiple steps conversion [2]. Therefore, lignocellulosic biomass from forest, agricultural and agroindustries are dedicated to liquid fuel production via thermochemical decomposition such as pyrolysis and liquefaction processes [3,4]. Depending on the desired pyrolysate (biochar or liquid oil), lignocellulosic matters are decomposed by slow or fast pyrolysis. Slow pyrolysis produces primarily biochar, the residual tar and syngas are rarely attended for any uses; the pyrolysis carbonisation is facilitated with conventional heating element or microwave irradiation [5,6].

Fast pyrolysis converts lignocellulose biomass to mainly bio-oil and other chemicals in contrast to slow pyrolysis [7]. Low cost makes focus shift to fast pyrolysis of lignocellulose biomass for bio-oil production because high-pressure liquefaction gives low yield of bio-oil at high cost [8]. Conventional fast pyrolysis produces pyrolysate liquid with fractions widely distributed. However, catalytic biomass fast pyrolysis narrows the bio-oil fractions to high-grade bio-oil. The major constituents of the pyrolysis oil are phenolic and alkylated (poly-) phenols, along with relatively small amounts of phenol, eugenol, cresols and xylenols [9]. The pyrolysis reactions involve cracking of different side-chain structures and methoxy groups of aromatic ring of the lignin component of the biomass. Heating values for the bio-oils range from 16.79 to 19 MJ/kg, close to that of oxygenated fuel such as ethanol; but lower than 40 - 45 MJ/kg for conventional fossil fuels. The bio-oils low heating values are affected by the high degree of oxygenated compounds [10]. Also, the water in the bio-oils is problematic for the direct use of the oil as a transportation fuel. The water and oxygenated compounds promoted non-volatility, high acidity, corrosiveness and aging during storage. The bio-oil experienced extreme instability and tendency to repolymerise during storage as a response to the aging, responsible for drawbacks in the bio-oil fuel properties [11]. Water and oxygenated compounds in the pyrolysis oil (bio-oil) are relatively in

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large quantities; therefore it is difficult to suppress their repolymerisation that can overburden the bio-oil fuel characteristics. Water removal can increase stability and viscosity, and reduces acidity of the bio-oil, but requires expensive, sophisticated technique. To produce high-grade bio-oil and chemicals, Gong et al. [12] and Hussain et al. [13] used catalytic pyrolysis of lignocellulose biomass. Catalysts such as Zeolite (ZSM-5, HZSM-5 and FCC) [14,15], and alkaline ($\text{Na}_2\text{CO}_3/\gamma\text{-Al}_2\text{O}_3$, K_2CO_3 , $\text{Ca}(\text{OH})_2$ and MgO) [16,17] in biomass pyrolysis influences the bio-oil quality. The catalysts upgrades the quality of the pyrolysis oil through cracking, deoxygenation, oligomerisation, cyclization, aromatisation, alkylation, isomerization and polymerisation [18]. The catalytic upgrade mainly increases the heating value (energy density) of the bio-oil. Catalysts tailor thermally driven fast pyrolysis reactions to yield high quality and more stable bio-oil [19]; also, the consequences has significant influence on product selectivity by the catalysts [20]. In association with catalyst loading, pyrolysis variant conditions also influence the yield and composition of the pyrolysate. The conditions (operating parameters) are tempered to direct the pyrolysis reaction to the desired quality and quantity of bio-oil. The typical operating parameters for fixed, fluidised bed, batch and auger pyrolysers can include; Biomass loading, Biomass particle size, Nitrogen flow rate, Pyrolysis temperature and Time [21]. Also, Taarning et al. [10] reported that bio-oil compositions rest on the type of pyrolyser system, biomass feedstock, and catalyst type and the reaction conditions.

Scholars extensively discuss the main aspects of catalytic pyrolysis such as pyrolysis technology and processes, catalyst type activity and deactivation, parameters influences, biomass feedstock, and reaction mechanism. Previously, Akhtar & Saidina Amin [22] evaluated the influence of severity on zeolite catalysed biomass pyrolysis on the maximum yield of the desired bio-oil. Lappas et al. [23] appraised the application of the zeolite acid catalyst in biomass pyrolysis for the production of transportation fuel with few attentions to base catalyst in pyrolysis of biomass. Similarly, Rezaei et al. [24] reviewed the catalytic activities of several acidic zeolite catalysts for the selective production of aromatics and olefins in relation to pyrolysis conditions. Furthermore, Dickerson and Soria [25] assessed the effect of various catalyst over pyrolysis reaction with particular attention to pyrolysis reaction pathway and mechanism. However, Isahak et al. [26] reviewed catalytic pyrolysis of biomass with an exclusive focus on the type of pyrolysis and pyrolyser, pyrolysis severity in the pyrolysis of biomass to bio-oil. Galadima and Muraza [27] discussed on zeolite coke deactivation from the perspective of the catalyst topology and coke deposition on the active catalyst sites. The effect primarily reduces the yield of aromatics in the resultant bio-oil; as a consequence, the bio-oil quality as fuel precursor is seriously compromised. Furthermore, Authours in [28] appraised the influences of parameters in fashioning the distribution of the final products of biomass pyrolysis with a special focus on the pyrolysis processes and yield of biochar. Exclusively presented are the reaction pathways that establish the pyrolytic devolatilisation of structural components of biomass and the attending kinetics models that explained the devolatilisation pathways [29,30].

This paper elucidated on the prevalent dynamic innovations that expedites the production of renewable fuel and fine chemicals from the bio-oil precursor, a product of catalytic pyrolysis of lignocellulosic biomass. The report focused on research expeditions on different biomass precursors; catalysts and pyrolysis process that are susceptible to the improvement of catalytic biomass pyrolysis to high-grade bio-oil and chemicals.

2. Lignocellulose Biomass

2.1. Availability and energy prospects

The appetite for fossil fuels for energy is on the decline from the high cost, dwindling reserves and the problematic greenhouse effects. However, following the growing energy demand and drawbacks

associated with fossil fuels gives particularly lignocellulosic biomass eminent role in the international energy policy for renewable energy. Lignocellulose biomass becomes increasingly a significant source of clean alternative energy. The feedstocks are unusually diverse, cheap and abundant non-food biomass. Non-food lignocellulosic biomass constitutes an important portion of agricultural and forestry wastes, agroindustrial wastes and leftovers and municipal waste; globally, $150 - 170 \times 10^9$ t is produced annually. The global availability of agricultural residues such as wheat straw, rice straw, corn straw and sugarcane bagasse as at 2012 is 1394.39 Mt [31]. In 2010, 88.74 Mt of fresh empty fruit bunches (EFB) from palm oil mills is produced; an enormous amount of the EFB are wasted because of inefficient utilisation of the available biomass. For every palm oil produced in Malaysia, approximately 4 kg of dry biomass residue is generated. Nevertheless, power produced from palm biomass accounted for only 60 MW out of 68 MW of biomass [32]. Based on the 2008 estimate in Ghana, lignocellulosic biomass produced 72.20 TJ of energy [33]. Taken cognisance to environmental and agricultural restraints, biomass resources can provide power between 130 and 270 EJ/year in 2050. The power can satisfy 15–25% of the future global energy demand [34]. Haberl et al. [35] reported that the global bio-energy crop potential is approximately 30–120 EJ/yr. However, another study suggested a higher range from 65 to 148 EJ/yr with constraints of soil degradation and water scarcity [36]. Therefore, biomass residues and leftovers for biofuels offer an alternative option to replace fossil fuels with no additional land use. Energy and chemicals from the lignocellulose biomass residues reduce CO_2 emission, the dependence on forest wood and fossil fuels in an environmentally friendly manner. The lignocellulose biomass residues can bust green technology in mitigating global climate change and play a central role in the anticipated development of the world's sustainable source of renewable energy.

2.2. Sources of biomass residues for the production of bio-oil

Crop processing and Agro-industries produce large tonnage of biomass residues in the form of a stack, straw, bagasse, empty fruit bunches, husk and sawdust. Agricultural residues include field residues and processing residues. Harvesting of cereals and vegetables generate the substantial amount of crop residues as leftovers in the farms. The entire wastes are important sources of renewable and abundant energy resources. Abnisa et al. [37], synthesis bio-oil and other chemicals from palm oil waste, Li et al. [38] used Wood pine and Fan et al. [39] used Rape straw. Moreover, Kim et al. [40] used *Jatropha* wastes, Jae et al. [20] used wood and Naqvi et al. [41] used Paddy husk. The basis for the choice of the feedstocks for pyrolysis production of bio-oil and other chemicals depends on the structural and chemical composition and heating values of the biomass. Analytical data collected from experiments define the average value of the structural formation and chemical composition and heating value of the biomass.

2.3. Structure of lignocellulose biomass

Atmospheric carbon dioxide, water and solar energy are the precursors for the synthesis of lignocellulose biomass tissues by photosynthesis. The tissues form the structure of the plant, arranged in a complex matrix in the form of hemicellulose, cellulose and lignin.

Hemicellulose and cellulose are polysaccharide sugar; therefore, lignocellulose biomass is mainly lignin and carbohydrate. Lignin has the largest fraction of lignocellulose biomass than carbohydrate, and its composition depends on the plant species. Lignin is complex branched amorphous polymer consist of several bonds of Hydroxyl, and methoxy-substituted phenylpropane unit. Alcohols are the monomers linked to the lignin by aryl ether, and carbon-carbon bonds are the precursors for lignin biosynthesis. The structure of lignin is supported by following linkages: $\beta\text{-O-4}$, $\alpha\text{-O-4}$, 5-5, $\beta\text{-5}$, and $\beta\text{-}\beta$ linkages [15]. Pandey & Kim [42] reported the proportion of the primary linkages in lignin. The

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