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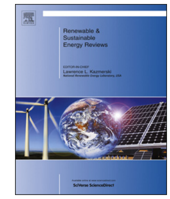

Production and catalytic transformation of levulinic acid: A platform for speciality chemicals and fuels

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ABSTRACT

Lignocellulosic biomass is a renewable and abundant source that can be used as a replacement for fossil resources in the sustainable production of speciality chemicals and transportation fuels. Over the last several decades, it has been demonstrated that one of the most effective methodology is to converse the high concentration of oxygen functionalized biomass monomers (e.g., cellulose, hemicelluloses) through de-functionalization into levulinic acid (LA) that has low oxygen content, followed by catalytic transformation of LA into fuels and valuable chemicals. This strategy currently seems to be the logical and promising alternative for sustainable development in the context of economic and environmental considerations. Besides, LA has been identified as one of the most promising platform chemicals for the sustainable production of fuels and commodity chemicals. This review is an up-to-date progress of literatures available on the subject of speciality chemicals and fuels derived from biomass through LA platform. The mechanism and current technologies for the production of LA are reviewed and compared. The potential theoretical calculation methods such as ab initio methods and density functional theories to predict the reaction pathway was also commented. The various transformation methods started from LA to speciality chemicals and fuels are critically reviewed. Among the various products, γ-valerolactone, 2-methyltetrahydrofuran and levuinate esters have been identified as promising fuels. The commercial diphenolic acid and delta-aminolevulinic acid have been widely utilized in many areas. The potential applications as well as fuel properties of these products are also discussed.

Contents

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1. Introduction

With the growth of emerging economies and increasing populations that is approximately quadrupled in the 20th century, the continued use of fossil fuels has resulted in a fast increase in global energy consumption with 16 fold [1,2]. The three most utilized fuels by industrial economies are oil, coal and natural gas [1–3], which accounted for ~87% of the global energy consumption in 2011 as shown in Fig. 1A. In the United States, fossil resources account for ~93% of the total energy consumption in the transportation sector as shown in Fig. 1B [1,2]. The global energy demand for liquid fuels was 85.7 million barrels/day in 2008 and it is estimated to be grown to ~97.6 million barrels/day by the year 2020 and ~105 million barrels/day by the year 2030 [2], while the existing energy resources of the crude oil can only support our society for another ~50 years at the current rate of consumption [3]. Besides, most speciality chemicals and fuels are directly or indirectly produced from the limited fossil resources [4–8]. To develop more sustainable routes for the production of fuels and commodity chemicals, our society has to explore and utilize alternatives such as renewable resources for this purpose. Additionally, the combustion of the primary energy sources (e.g., oil, natural gas and coal) has caused a significant increase in the concentration of carbon dioxide in the atmosphere [9–11], which has been widely considered as a greenhouse gas, trapping solar energy and changing global climate. To transform our society into a greener future, sustainable alternatives have been taken for environmental protection and rational utilization of renewable energy [9,12–16].

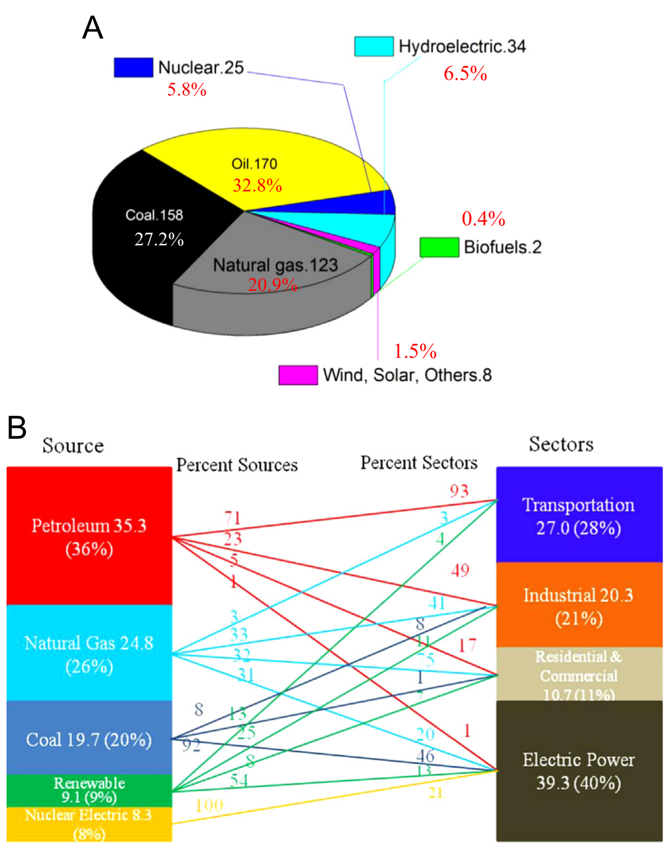

Fig. 1. Global energy consumption in 2011 (A) and Primary energy consumption in the United States by source and sector in 2011 (all numbers in quadrillion BTU, (B), modified and redraw from [2]. *Note*: the waste (4.9%) is not accounted in (A).

Biomass is the only abundant and concentrated source of non-fossil carbon that is available on Earth. The conversion of biomass to speciality chemicals and fuels has been the focus of industry and researchers within the past decade [9,17], because it not only aimed at the development of effective and environmentally benign technology, but also simultaneously solves the problem of agricultural and forestry waste use [18–23].

The DOE/NREL report, named "*Top Value Added Chemicals from Biomass*" [24] identified the twelve top chemical candidates for launching renewable chemical platforms that can be derived from biomass. Bozell et al. [25] presented an updated evaluation of potential target structures and extended the platform chemicals of biobased products from biorefinery carbohydrates. In most of the established chemical approaches being investigated, platform chemical levulinic acid (LA) [24,25], also named 4-oxopentanoic acid, was considered as one of the most promising platform chemicals from lignocellulosic biomass for fuels and chemicals (Figs. 2 and 3). It is also a speciality chemical that finds applications for several purposes, such as source of polymer resins, animal feed, food as well as components of flavoring and fragrance industry, textile dyes, additives, extenders for fuels, antimicrobial agents, herbicides and plasticizers [26,27].

LA can be obtained by the multiple steps in the hydrolysis of raw cellulose through dilute sulfuric acid treatment via

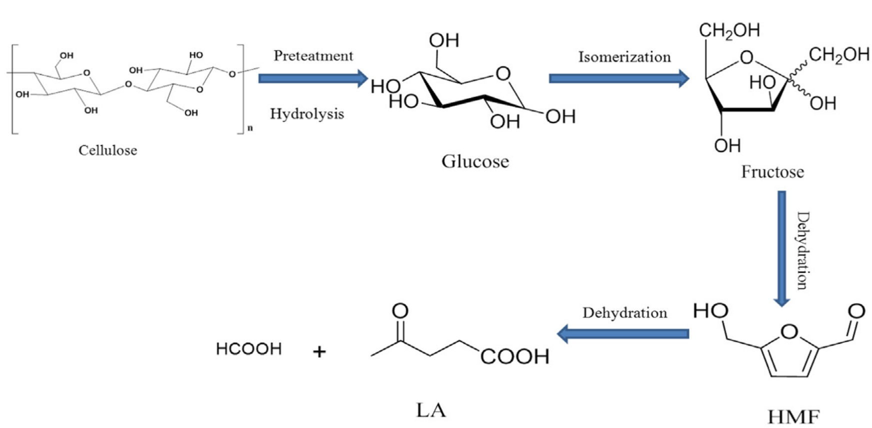

Fig. 2. Production of LA from cellulosic biomass [8,18].