

An overview of dehydration, aldol-condensation and hydrogenation processes for production of liquid alkanes from biomass-derived carbohydrates

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Abstract

We present results for the conversion of carbohydrate feedstocks to liquid alkanes by the combination of dehydration, aldol-condensation/hydrogenation, and dehydration/hydrogenation processing. With respect to the first dehydration step, we demonstrate that HMF can be produced in good selectivity from abundantly available polysaccharides (such as inulin, sucrose) containing fructose monomer units using a biphasic batch reactor system. The reaction system can be optimized to achieve good yields to 5-hydroxymethylfurfural (HMF) from fructose by varying the contents of aqueous-phase modifiers such as dimethylsulfoxide (DMSO) and 1-methyl-2-pyrrolidinone (NMP). Regarding the aldol-condensation/hydrogenation step, we present the development of stable, solid base catalysts in aqueous environments. We address the effects of various reaction parameters such as the molar ratio of reactants and temperature on overall product yield for sequential aldol-condensation and hydrogenation steps. Overall, our results show that it is technically possible to convert carbohydrate feedstocks to produce liquid alkanes by the combination of dehydration, aldol-condensation/hydrogenation, and dehydration/hydrogenation processing; however, further optimization of these processes is required to decrease the overall number of separate steps (and reactors) required in this conversion.

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1. Introduction

In the view of declining petroleum resources and rising oil prices, it is necessary to develop alternative ways to fulfill the energy needs of our industrialized society. To meet the growing demand for energy, biomass can serve as a sustainable source of renewable fuels and chemicals. In addition to domestic availability, fuels developed from biomass resources are CO₂-neutral, because CO₂ released during combustion can be utilized for biomass growth in the next cycle. A recent report from the U.S. Department of Agriculture (USDA) and U.S. Department of Energy (DOE) estimates that a sustainable production of 1.3 billion dry tonnes of biomass per year can be achieved without significant changes in agricultural practices and food supplies, leading to 20% replacement of transportation fuel demand by 2030 [1]. The primary routes for

conversion of biomass to liquid fuels include thermochemical gasification to produce synthesis gas [2], liquefaction of biomass and/or pyrolysis to bio-oils [3], conversion of sugars to ethanol and aromatic hydrocarbons [4,5], and esterification of triglycerides to form bio-diesel [2]. However, development of new technologies is critical to accelerate the development of renewable fuels from biomass. In this respect, the most effective and efficient utilization of renewable biomass resources is through the development of an integrated biorefinery, in which the energy requirements of each process are balanced with those of other processes analogous to petroleum refinery.

Recently, we have developed a process to convert sugar alcohols (e.g., sorbitol) to lighter alkanes (C₁–C₆) using a bifunctional metal-acid (Pt/SiO₂–Al₂O₃) catalyst [6]. The alkanes produced in this process contain 90% of the heating value of glucose and H₂ feed but only 30% of its mass. However, the alkanes produced in this process contain the same number of carbon atoms as the initial sugar (usually 5 or 6 carbon atoms), and hence they cannot be used for fuel

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applications due to their high volatility. As a result, we more recently developed a process that produces liquid alkanes, ranging from C_7 to C_{15} , by aqueous-phase processing of biomass-derived carbohydrates, thereby providing a renewable source for transportation fuel [7]. This process can complement the development of P-series fuel (a mixture of approximately equal volumes of ethanol, methyltetrahydrofuran and pentane-plus components) that has over 60% of its current composition resulting from renewable sources [8]. Specifically, the pentane-plus components of this fuel, which are currently derived from petroleum, can be replaced by the lighter alkanes (C_5 – C_9) produced from this technology. Moreover, the oxygenated form of saturated organic molecules or heavier liquid alkanes (C_{13} – C_{15}) from our process can serve as diesel-fuel additives. In addition, our process has an overall energy efficiency of 2.1 (ratio of heating value of alkanes to energy required to produce alkanes) as compared to bio-ethanol, which has an energy efficiency of about 1.1–1.3 [9].

As depicted in Fig. 1, the production of heavier liquid phase alkanes from carbohydrates involves a series of reaction steps starting with acid hydrolysis of polysaccharides to produce monosaccharides, followed by acid-catalyzed dehydration to form carbonyl-containing furan compounds such as 5-hydroxymethylfurfural (HMF) and furfural. Subsequently, these compounds can be condensed via aldol reaction to produce larger organic molecules ($>C_6$) by forming C–C bonds, and these aldol-products can be hydrogenated to form large water-soluble

organic compounds. These molecules are then converted to liquid alkanes (ranging from C_7 to C_{15}) by aqueous-phase dehydration/hydrogenation (APD/H) over a bifunctional catalyst containing acid and metal sites in a flow reactor. Thus, dehydration of carbohydrates to produce furanic compounds and the aldol-condensation reaction play important roles in multi-step catalytic production for liquid alkanes. In this work, we address further developments in the dehydration and the aldol-condensation aspects of the above-mentioned process to produce liquid alkanes. In particular, we demonstrate that HMF can be produced in good selectivity from abundantly available polysaccharides (such as inulin, sucrose) containing fructose monomer units using a biphasic batch reactor system. The reaction system is initially optimized to achieve good HMF yields from fructose by varying contents of aqueous-phase modifiers such as dimethylsulfoxide (DMSO) and 1-methyl-2-pyrrolidinone (NMP). We further present the development of stable, solid base catalysts for aldol-condensation in the aqueous phase. We address the effects of various reaction parameters such as the molar ratio of reactants and temperature on overall yield for aldol-condensation combined with the sequential hydrogenation step. The aqueous dehydration/hydrogenation process to produce liquid alkanes is reviewed elsewhere [6,7,9]. Finally, we elucidate the effects of various reaction parameters for each of the processes involved in the conversion of sugars to liquid alkanes, and we present various opportunities to move this technology toward practical application.

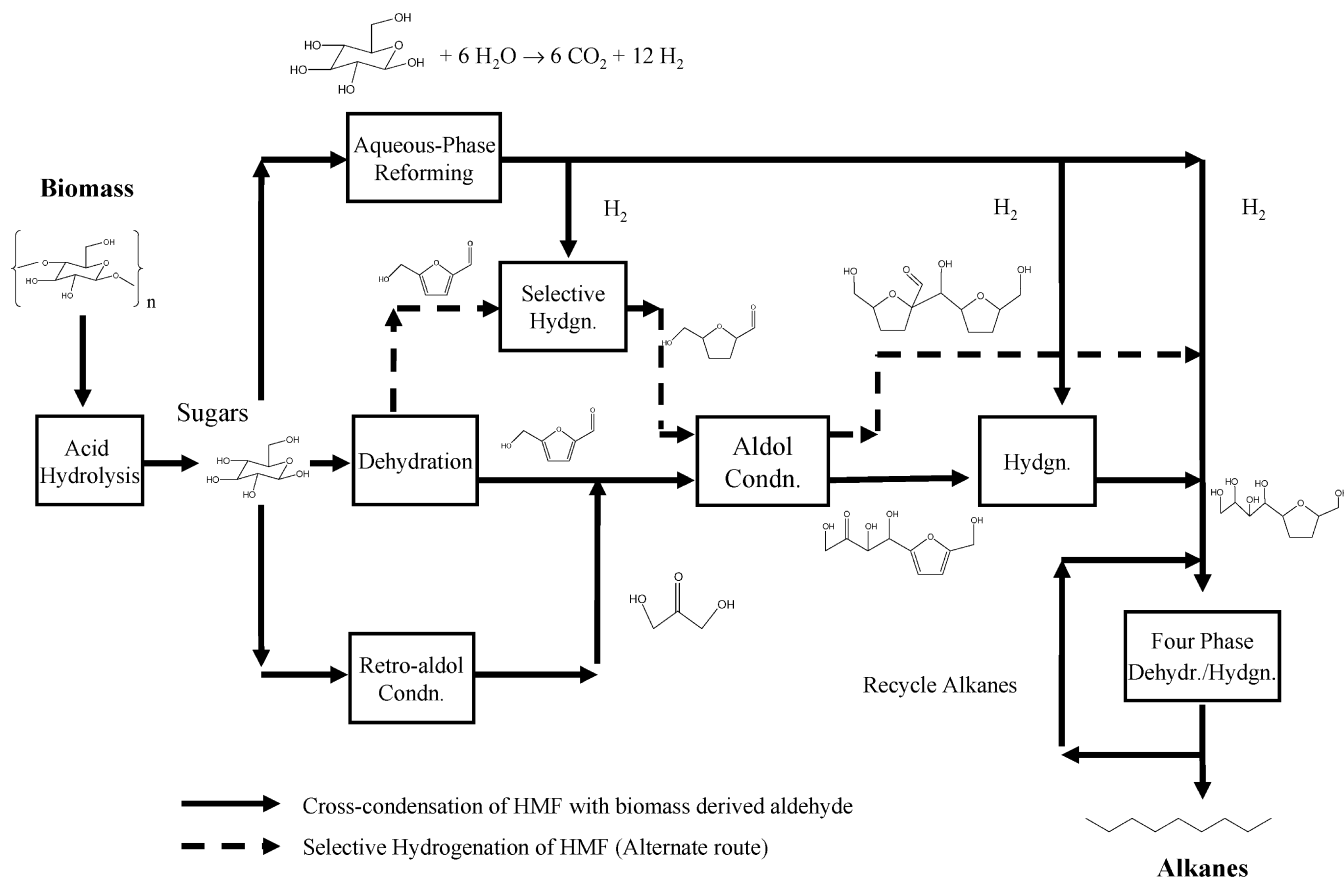


Fig. 1. Schematic diagram for production of liquid alkanes from biomass resources in a biorefinery.

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