



Integration of heterogeneous and biochemical catalysis for production of fuels and chemicals from biomass

Ian Wheeldon, Phillip Christopher and Harvey Blanch



The past decade has seen significant government and private investment in fundamental research and process development for the production of biofuels and chemicals from lignocellulosic biomass-derived sugars. This investment has helped create new metabolic engineering and synthetic biology approaches, novel homogeneous and heterogeneous catalysts, and chemical and biological routes that convert sugars, lignin, and waste products such as glycerol into hydrocarbon fuels and valuable chemicals. With the exception of ethanol, economical biofuels processes have yet to be realized. A potentially viable way forward is the integration of biological and chemical catalysis into processes that exploit the inherent advantages of each technology while circumventing their disadvantages. Microbial fermentation excels at converting sugars from low-cost raw materials streams into simple alcohols, acids, and other reactive intermediates that can be condensed into highly reduced, long and branched chain hydrocarbons and other industrially useful compounds. Chemical catalysis most often requires clean feed streams to avoid catalyst deactivation, but the chemical and petroleum industries have developed large scale processes for C–C coupling, hydrogenation, and deoxygenation that are driven by low grade heat and low-cost feeds such as hydrogen derived from natural gas. In this context, we suggest that there is a reasonably clear route to the high yield synthesis of biofuels from biomass- or otherwise derived-fermentable sugars: the microbial production of reactive intermediates that can be extracted or separated into clean feed stream for upgrading by chemical catalysis. When coupled with new metabolic engineering strategies that maximize carbon and energy yields during fermentation, biomass-to-fuels processes may yet be realized.

Address

Chemical and Environmental Engineering, University of California Riverside, Riverside, CA 92521, United States

Corresponding authors: Wheeldon, Ian (iwheeldon@engr.ucr.edu), Christopher, Phillip (christopher@engr.ucr.edu)

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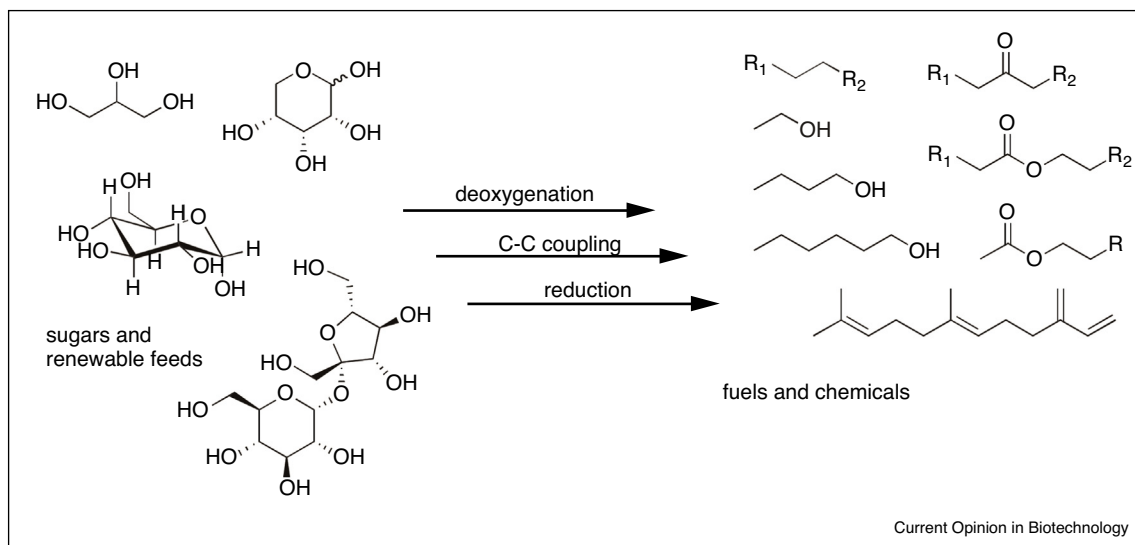
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Introduction

The impact of recombinant DNA technologies on the production of fuels and chemicals from renewable resources has been remarkable. It has been driven in part by the reduced environmental impact that bioprocessing provides over petroleum- and syngas-based chemical synthesis and refining. Today, biotech-produced industrial chemicals represent over \$125 billion in revenue in the US with an annual growth of over 10% [1]. Biofuels produced from renewable resources, primarily 10% ethanol blends used in gasoline, represent a significant component of this growth and US biodiesel production from plant oils increased to over 1.8 billion gallons in 2013. Because of compatibility issues with the existing fuels infrastructure, higher ethanol blend ratios may be problematic and there has been growing interest in the production of second-generation biofuels that resemble gasoline components, such as C₄–C₁₀ branched chain hydrocarbons and alcohols. Second-generation biofuels ('advanced' biofuels) are anticipated to have an additional advantage over ethanol beyond infrastructure compatibility, in that less energy may be required for their phase separation from the fermentation broth compared to the recovery of ethanol by distillation [2].

The renewable resources available for the production of industrial chemicals and fuels include corn-derived sugar, glycerol, sucrose (sugar cane) and potentially lignocellulosic sugars (glucose, xylose). These sources provide carbohydrates ranging from C₃ to C₆ as raw materials. Oils, such as palm oil and vegetable oils, can also be used to produce biodiesel. The conversion of sugars to biofuels or industrial chemicals with carbon lengths greater than C₆ requires C–C bond formation and, in the case of fuels and some chemicals, oxygen removal to produce compounds more reduced than the feedstock (Figure 1). Depending on reaction scheme, reduction through hydrogenation may also be necessary. These reactions are possible in both a biological context and through chemical catalysis. Large scale heterogeneous hydrogenation, decarboxylation, and deoxygenation reactors are commonplace in the chemical and petroleum-based fuel industries, and C–C coupling is well established through Fischer–Tropsch, methanol-to-gasoline (MTG), and other technologies. Where chemical catalysis is lacking, bioprocessing excels. Microbial processes can uptake and metabolize sugars and lipids from impure and mixed raw materials streams and can selectively hydrogenate, decarboxylate, and deoxygenate multi-functional chemical species.

Figure 1



General reaction scheme for the conversion of sugars and renewable carbon sources into fuels and chemicals.

In this review, we argue that a combined approach that integrates chemical and biological catalysis is a potentially viable way forward for biofuels and the renewable production of industrial chemicals (Table 1). By exploiting the advantages of each technology, it may be possible to effectively convert sugars, biomass-derived or otherwise produced, into highly reduced long chain and branched hydrocarbons at high yields and low costs. For example, the high energy costs of biological C–C coupling can be avoided by building fuel-length carbon chains with low-grade heat to drive heterogeneous condensation reactions. Recent reviews on this topic have focused on biomass upgrading chemistries and catalysts [3*,4,5], and design rules for coupling biological and chemical catalysis have been proposed [6*]. We begin our review by discussing the advantages of the respective catalytic systems and current work focused on addressing their disadvantages. We finish our discussion by using recent examples of integrated chemical and biological processes to identify the pressing technological and process design challenges.

Microbial biosynthesis of fuels and chemicals: the effective use of raw materials, but reduction, deoxygenation, and native metabolism reduce yield

A common view of biology is that it excels at synthesizing complex, multifunctional compounds and materials. In many respects this is true. Native metabolic pathways convert simple sugars into complex, chiral secondary metabolites (*e.g.*, penicillin) and long, monodispersed polymer chains (*e.g.*, proteins, polypeptides, nucleic acids, *etc.*). But these synthetic pathways are often one of many,

with the production of any single compound often occurring at low yield and requiring significant separations to achieve useful purities. Bioprocesses do not always follow this common view, but instead successful processes can be developed from the catabolism of sugars into simple alcohols and acids (*e.g.*, ethanol, butanol, lactate, and succinate). These processes are viable because theoretical and practical energy and carbon yields are high and microbes can use complex raw materials streams as a low-cost source of sugars [7]. With respect to biofuels, ethanol from corn- and sugar cane-derived sugars (in the US and Brazil, respectively) is dominant. Production of lignocellulosic biomass-derived sugars still face technological challenges to overcome biomass recalcitrance and the high cost of enzymes for saccharification [8,9].

The microbial production of long chain and branched industrial chemicals and fuels requires anabolic reactions to extend chain length and, in the case of fuels, reduction through selective oxygen abstraction. Metabolically, these reactions are costly. C–C bond formation can occur through fatty acid biosynthesis and other pathways at the expense of cellular energy in the form of adenosine triphosphate (ATP). Microbial oxygen abstraction can be accomplished by CO_2 formation or by H_2O production. In the latter case, reducing equivalents (hydride ions, typically NAD(P)H) are formed from additional sugars, effectively via an aqueous reforming reaction. Oxygen is subsequently removed from the target product as water. Oxygen abstraction thus reduces the yield of product from sugar, either by loss of carbon from the sugar (CO_2 production) or by the requirement for additional sugar to abstract oxygen as water.

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