



Review

Catalytic reactions of gamma-valerolactone: A platform to fuels and value-added chemicals



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ABSTRACT

Gamma-valerolactone (GVL), which is accessible from renewable lignocellulosic biomass, has been identified as one of the most promising platforms for the sustainable production of fuels and value-added chemicals. This review aims at recent advances in the catalytic production of GVL from biomass and further upgrading of GVL to fuels as well as value-added chemicals. The first part briefly reviews recent advances for the production of GVL from biomass. The second and third sections critically review and identify current technologies for the efficient production of GVL. The mechanism, different types of advanced homogeneous and heterogeneous catalysts employed have been compared and broadly categorized. Challenges and areas that need improvement are also highlighted in the corresponding area. The fourth section concentrates on potential applications and the upgrading of GVL to fuel additives, diesel fuels and value-added chemicals. The final section offers a summary and future perspective in the field.

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Nomenclature

LA	levulinic acid
BL	butyl levulinate
GVL	γ -valerolactone
EL	ethyl levulinate
HMF	5-hydroxymethylfurfural
ML	methyl levulinate
BF	butyl formate
FAME	fatty acid methyl esters
DMF	2,5-dimethylfuran
ILs	ionic liquids
MTBE	methyl tert-butyl ether
WHSV	weight hourly space velocity
MTHF	2-methyltetrahydrofuran
Zr-Betal-100	Betal zeolite with the Si/Zr ratio of 100
THF	tetrahydrofuran
DOE	department of energy
Y_{GVL}	yield of γ -valerolactone
NREL	National Renewable Energy Laboratory
1,4-PDO	1,4-pentanediol
ZrO ₂ -VS	acid-tolerant ZrO ₂

1. Introduction

It has been predicted that 20% of transportation fuel and 25% of chemicals by 2030 will be produced from lignocellulosic biomass, which is the only renewable carbon source capable of replacing fossil fuels [1–3]. Lignocellulosic biomass, consisting of three main structural units (i.e., cellulose, hemicellulose and lignin), is a complex fibrous material that can be found in the cell walls of plants as shown in Fig. 1 [4–6]. As an alternative carbon source, lignocellulosic biomass offers some more attractive benefits over petroleum [7,8]. Lignocellulosic biomass is cheap, abundant and has widespread worldwide availability. It is reported that 1.3 billion tons of dry biomass can be produced per year in the United States alone [8]. More importantly, the lignocellulosic biomass is sustainable and its derived monomers can be transformed into fuels and numerous valuable chemicals [9–14]. The main pathway of the conversion of biomass feedstocks into renewable fuels was shown in Fig. 2 [13,15]. Over the last several decades, catalytic conversion of biomass has got significant growth and will likely increase in the coming years [16–21].

Due to the structural and chemical complexity of lignocellulosic biomass, variously advanced processes have been employed to convert it into fuels and chemicals [12,13,20]. Current tech-

nologies mainly involve two major routes: thermal processing and aqueous phase processing (Fig. 2). Thermal processing including gasification and pyrolysis, is well-suited for the production of fuels, where inaqueous phase processing provides attractive paths for the production of intermediates that can be subsequently upgraded to value-added chemicals and fuels for the transportation sector [13,20]. Biomass gasification often provides clean combustion, compact burning equipment, and high thermal efficiency [22–25]. However, the gasification process is quite complex and sensitive, where the fuel is bulky and frequent refueling is often required for continuous running of the system. Besides, the way of dealing with the residues such as ash and tarry condensates is a time consuming and tough job [20,22,24]. Unlike gasification, pyrolysis process consists of a thermal degradation of the initial solid biomass into gases and liquids without an oxidizing agent [21,26]. The products of biomass pyrolysis primarily consist of biochar, bio-oil and gases including CH₄, H₂, CO and CO₂ [27,28]. Pyrolysis offers a flexible and attractive way of converting solid biomass into an easily stored and transported liquid fuels [29]. However, the cost of pyrolysis is quite high compared to the production of fossil fuel. Despite the rapid development over the last few decades has been reached, many techno-economic barriers (e.g., the cost of biomass pyrolysis) need be reduced [30,31].

In comparison, aqueous phase processing of biomass seems more promising and attractive. The DOE/NREL report, named “Top Value Added Chemicals from Biomass” [32] has identified a list of top value-added chemical candidates for launching renewable chemical platforms derived from biomass. Along the line of these candidates, GVL has attracted considerable attention over the last several decades and will likely flush in the coming years due to its attractive physical-chemical properties and unique fuel characteristics as depicted in Table 1 in comparison with ethanol fuel. Over the last a few decades, a number of researchers have developed different advanced methods for improving the synthesis of GVL from lignocellulosic biomass [10,16,21,32]. GVL is stable in water and in the presence of air, and thus can also be produced from C₆ and C₅ sugars through LA intermediate. Besides, GVL, a widely used food additive, exhibits the most typical characteristics of an ideal sustainable green solvent for the production of either energy or carbon-based consumer products [33]. In addition, GVL does not hydrolyze under neutral conditions and does not form a measurable amount of peroxides in a glass flask under air in weeks, making it a safe material for the industrial application [33,34]. A comparative study of GVL and ethanol as fuel additives with a mixture of 10 v/v% GVL or ethanol and 90 v/v% 95-octane gasoline [33,34], has shown very similar fuel properties and it has been suggested as an attractive liquid fuel.

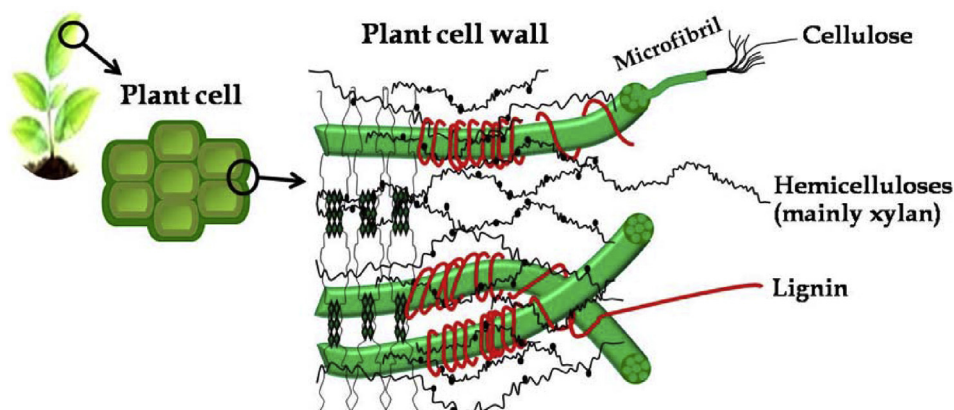


Fig. 1. Lignocellulosic biomass composition in plants. Reprinted with permission from Ref. [6].

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