



Recent advances in catalytic transformation of biomass-derived 5-hydroxymethylfurfural into the innovative fuels and chemicals



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ABSTRACT

In contrast to the nonrenewable fossil resources, biomass, the only renewable resource of organic carbon in the nature, is considered as a special kind of inexhaustible feedstocks, which can be used for the synthesis of numerous valuable products in a sustainable manner. Among many biomass-derived products, 5-hydroxymethylfurfural (HMF) is identified to be a crucially important versatile compound due to its marvelous structure that is composed of an aldehyde group, a hydroxyl group and a furan ring. Hence, HMF possesses a very strong chemical reactivity, and it can be further transformed into a wide variety of value-added derivatives. In recent years, the synthetic methods, physicochemical properties and commercial prospects of HMF-based conventional derivatives such as 2,5-dimethylfuran (DMF), 5-ethoxymethylfurfural (EMF), ethyl levulinate (EL), long chain alkane (LLA), levulinic acid (LA), 2,5-diformylfuran (DFF) and 2,5-furandicarboxylic acid (FDCA) have been intensively reviewed by many researchers. However, up to now, the preparation of HMF-based innovative derivatives such as 2,5-dihydroxymethylfuran (DHMF), 2,5-dihydroxymethyltetrahydrofuran (DHMTHF), 1,2,6-hexanetriol (HTO), 1,6-hexanediol (HDO), 1-hydroxyhexane-2,5-dione (HHD), 3-hydroxymethylcyclopentanone (HMCPN), furan-2,5-dimethylcarboxylate (FDMC), maleic anhydride (MA), 5-hydroxy-5-(hydroxymethyl)furan-2(5H)-one (HHMFO), 5-alkoxymethylfurfural (AMF), 5,5-oxy-(bismethylene)-2-furaldehyde (OBMF), 5-arylaminomethyl-2-furanmethanol (AAMFM), 2,5-furandiamidine dihydrochloride (FDADHC), 1-alkyl-5-hydroxy-2-(hydroxymethyl)pyridinium (AHHMP), 5,5-bis(hydroxymethyl)furoin (BHMF), 5-(dialkoxymethyl)-2-furanmethanol (DAMFM), 5-chloromethylfurfural (CMF), 5-alkanoyloxymethylfurfural (AOMF) and furfuryl alcohol (FFA) has not yet been comprehensively summarized. In order to fill this gap, the latest studies and advancements on the preparation of HMF-based innovative derivatives via various catalytic approaches such as hydrogenation, oxidation, etherification, amination, condensation, halogenation, esterification and decarbonylation are systematically outlined and discussed in this review. Furthermore, a few potential research trends in the future studies are also proposed to provide some useful ideas for the further preparation of HMF-based innovative derivatives in a much more green, simple, efficient and economical way.

1. Introduction

Nowadays, approximately 86% of fuels and 96% of chemicals in the world are produced from the nonrenewable fossil resources such as coal, petroleum and natural gas that make tremendous contributions for the speedy development and prosperity of human society [1–5]. However, with the progressive increase of global product demands and the continuous decrease of worldwide fossil resources alongside with the growing concerns about environmental pollutions and carbon

emissions, it is very urgent and necessary to search for the renewable resources to reduce the dependence on the nonrenewable fossil resources [6–10]. In contrast to water energy, wind energy, solar energy, ocean energy and geothermal energy, biomass, possessing a range of excellent merits such as abundant, cheap, diverse and widespread, is the sole carbon-based renewable resource in the nature and then is regarded as the best choice for the production of numerous valuable products in a much more sustainable manner [11–18]. Therefore, in recent years, the relevant researches on the conversion

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of renewable biomass into valuable products have attracted extensive attention in the scientific, industrial and governmental communities.

Among various biomass-derived products, 5-hydroxymethylfurfural (HMF), which can be synthesized by the dehydration of fructose [19–26], glucose [27–35], sucrose [36–40], inulin [41–46], starch [47–50] and cellulose [51–56], is considered to be one of the most appealing and promising platform compounds [57–62], this is because that it contains a marvelous and reactive structure involving an aldehyde group, a hydroxyl group and a furan ring and then can be further transformed into high-quality fuels such as 2,5-dimethylfuran (DMF) [63–72], 5-ethoxymethylfurfural (EMF) [73–82], ethyl levulinate (EL) [83–90] and long chain alkane (LLA) [91–95] and high-value chemicals such as levulinic acid (LA) [96–105], 2,5-diformylfuran (DFF) [106–115] and 2,5-furandicarboxylic acid (FDCA) [116–125]. More excitingly, in addition to the above-mentioned conventional derivatives, many innovative derivatives such as 2,5-dihydroxymethylfuran (DHMF), 2,5-dihydroxymethyltetrahydrofuran (DHMTFH), 1,2,6-hexanetriol (HTO), 1,6-hexanediol (HDO), 1-hydroxyhexane-2,5-dione (HHD), 3-hydroxymethylcyclopentanone (HMCPN), maleic anhydride (MA), 5-hydroxy-5-(hydroxymethyl)furan-2(5H)-one (HHMFO), 2,5-furandimethylcarboxylate (FDMC), 5-alkoxymethylfurfural (AMF), 5,5-oxy-(bismethylene)-2-furaldehyde (OBMF), 5-arylaminoethyl-2-furanmethanol (AAMFM), 1-alkyl-5-hydroxy-2-(hydroxymethyl)pyridinium (AHHMP), 2,5-furandiamidine dihydrochloride (FDADHC), 5,5-bis(hydroxymethyl)furoin (BHMF), 5-(dialkoxymethyl)-2-furanmethanol (DAMFM), 5-chloromethylfurfural (CMF), 5-alkanoyloxymethylfurfural (AOMMF) and furfuryl alcohol (FFA) can also be prepared from HMF (Fig. 1). Currently, much more interest and attention are paid to the production of HMF-based conventional derivatives, and their synthetic methods, physicochemical properties and commercial prospects have been extensively reviewed by many researchers [126–140]. However, to the best of our knowledge, there is no special review on the production of HMF-based innovative derivatives. Furthermore, it is particularly necessary to point out that massive new technologies are increasingly applied and many important achievements are continuously obtained in this research area. Hence, a comprehensive and real-time review is also needed.

According to the present actual situations and recent research advances, the state-of-the-art studies on the production of HMF-based innovative derivatives via various catalytic approaches are system-

atically summarized and discussed in this review, which is organized in the following order: catalytic transformation of HMF via hydrogenation; catalytic transformation of HMF via oxidation; catalytic transformation of HMF via etherification; catalytic transformation of HMF via amination; catalytic transformation of HMF via condensation; catalytic transformation of HMF via other reactions. Moreover, some potential research trends in the future studies are also prospected to supply a few useful ideas for the further production of HMF-based innovative derivatives. All in all, we hope that this review can not only raise much more concerns about HMF-based innovative derivatives but also offer theoretical references and technical supports for the industrialization of HMF-based innovative derivatives.

2. Catalytic transformation of HMF via hydrogenation

2.1. DHMF

DHMF, which can be produced by the selective hydrogenation of aldehyde group in HMF, is widely used as an intermediate for the synthesis of resins [141], fibers [142], foams [143], drugs [144], polymers [145–147] and crown ethers [148–150]. Traditionally, DHMF is readily obtained by the stoichiometric hydrogenation of HMF with the help of sodium borohydride (NaBH_4) [151–153]. However, this strategy has some problems such as the treatment of reductive agents and the production of equivalent salts. To overcome these drawbacks, considerable efforts have been devoted to seek the appropriate catalytic systems that are able to effectively and selectively hydrogenate HMF into DHMF (Table 1).

In 2012, Balakrishnan et al. [149] studied the selective hydrogenation of HMF over carbon-supported platinum (Pt/C) in ethanol. Under 14 bar molecular hydrogen (H_2), the moderate DHMF yield of 82.0% was obtained at 23 °C for 18 h. Following this study, the selective hydrogenation of HMF in water was explored by Ohyama et al. [154], Chen et al. [155] and Tamura et al. [156] in 2013. By using alumina-supported gold ($\text{Au}/\text{Al}_2\text{O}_3$), zirconia-silica-supported ruthenium ($\text{Ru}/\text{ZrO}_2\text{-SiO}_2$) and silica-supported rhenium oxide and iridium ($\text{Ir-ReO}_x/\text{SiO}_2$) as catalysts, DHMF yields were noticeably increased to 96.0%, 90.4% and 98.0% at 120, 25 and 30 °C for 2, 4 and 6 h under 65, 5 and 8 bar H_2 , respectively, which was due to the synergistic actions between metals with small sizes and supports with basic properties in the

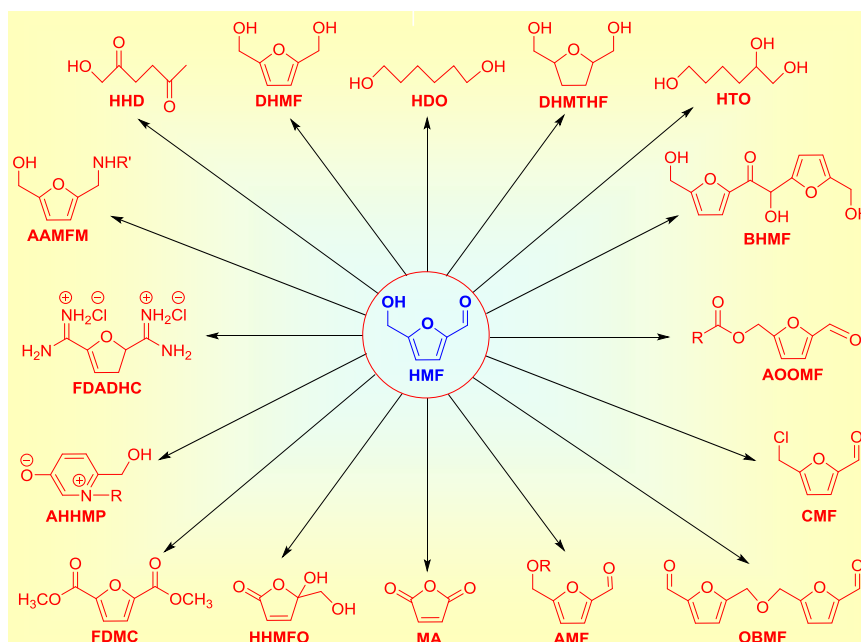


Fig. 1. Catalytic transformation of biomass-derived HMF into various innovative derivatives.

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