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Levulinic acid production from renewable waste resources: Bottlenecks, potential remedies, advancements and applications

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ABSTRACT

The rise in fuel and oil prices has driven the hunt for alternative renewable resources for production of chemical intermediates or biofuels since they provide a sustainable solution to an increasing demand. Levulinic acid (LA), one of the platform chemicals, can be produced chemically using renewable resources such as starch waste and lignocellulosic biomass which sounds to be an attractive alternative owing to its abundance and environmentally benign nature. The presence of acidic carboxyl and ketone carbonyl groups in LA impart a remarkable pattern of reactivity to levulinic acid and allow it to form several derivatives having significant applications in various fields, thereby making it a versatile green chemical. The present review deals with the current status of the technologies available for levulinic acid production, its recovery and the array of applications in several areas along with the gridlocks involved at each step of conversion process and suggest some possible remedies. Further, it not only throws light on the recent advancements in LA production approaches along with the avant-garde biotechnological approach for its biosynthesis but also depicts the current market scenario of commercial LA industry. The review also discusses the future R&D scope with an aim to enhance the yield of levulinic acid production and to make the process energy-efficient and cost-effective.

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1. Introduction

The hike in fossil fuel and oil prices and depletion of fossil resources has aggravated the energy crisis, thus, driving the search for new alternative renewable feedstocks for the production of basic chemical building blocks. A considerable part of research is focused to develop an attractive techno-economical and environmentally benign conversion process for the conversion of waste feedstock into platform chemicals on a commercial scale with a potential to replace petroleum-based fuels. Starch-rich waste and lignocellulosic biomass (henceforth referred to as LCB) have drawn much attention as feedstock for LA production owing to the renewability, abundance, low value and carbon-neutral nature. Moreover, it does not vie with the food chain due to no additional land requirement. Therefore, LCB and starch-rich waste sounds to be an ideal starting material for manufacture of fuel as 150 billion tonnes of LCB production is being reported per year while starch production is reported to be 75 million tonnes, both of which remain under-utilized [1].

Levulinic acid: Levulinic acid, also known as 4-oxopentanoic acid or gamma ketovaleric acid is a C-5 chemical and is identified as one of the key platform chemical. It is a short chain fatty acid with molecular formula $C_5H_8O_3$.

1.1. Physical properties

LA is a crystalline compound while the commercial product is yellow to brown in colour with a caramellic odor [2]. LA has boiling point of 245–246 °C, melting point of 33 °C and a density of around 1.1335 g/cm^3 [3]. LA has a refractive index of 1.4396 and is highly soluble in water, ethanol, ether, acids, chloroform, *etc*. LA has a p*K*a of 4.59 in water at 25 °C [3].

1.2. Chemical properties

LA possesses a ketone carbonyl group (C=O) and an acidic carboxyl group (COOH) which imparts it an ability to react with different functional groups to form a wide range of derivatives, thereby, making LA an ideal platform chemical [4].

LA has been produced successfully at laboratory scale but commercialization of LA has met with limited success due to certain bottlenecks *viz*. cost of raw material, equipment cost, low yield of LA due to undesirable side reactions, difficulties in efficient product recovery, catalyst recovery cost, energy-inefficiency and the process economy. LA is still being produced through petrochemical means involving use of maleic anhydride or furfuryl alcohol [5,6] which is costly and also causes environmental contamination. The review on this subject has been cited in the literature with reference to chemical process, however, the processes cited in the literature lacks in establishing a correlation with reference to environmental significance of such important building block. In the present review, various processes for LA production from starch-rich and/or lignocellulosic waste are reviewed with respect to the bottlenecks involved at different stages and pertaining solutions are also discussed. Additionally, the novel aspect of the review is that it not only brings forth the advancements in the arena of LA production and its recovery but also forecasts the newer approaches for LA production. The applications of LA and significance of its derivatives are also outlined herewith. Keeping in view the tremendous market potential of LA, its sustainable production at lower cost is of paramount significance. Ergo, the article attempts to shed light on the international status of LA commercialization and the loopholes involved in the industrial production process in conjunction with the futuristic approaches for building a viable and cost-effective LA production process.

2. Raw materials and precursors

LA can be synthesized from several different raw materials for instance, monosaccharides, precursors such as 5-hydroxymethyl furfural (HMF) and furfural, polysaccharides and renewable resources such as starch-rich waste and lignocellulosic biomass. The subsequent sub-section discusses these raw materials as starting point for LA synthesis with respect to bottlenecks, possible remedies and synthesis of these precursors in brief.

2.1. Monosaccharides

Monosaccharides, the most basic unit of carbohydrates, include the simplest sugars like glucose and fructose, which can act as starting point for LA synthesis. Fructose is readily dehydrated to 5-hydroxymethylfurfural (HMF) using acid catalyst while glucose has to be isomerized into fructose prior to its conversion to HMF. Solid catalysts like TiO_2, ZrO_2, *etc*. and enzymes like glucose isomerase may aid in isomerisation of glucose to fructose [7,8]. The HMF thus produced can be further converted to LA through different pathways.