Review Article

Furfural: A Sustainable Platform Chemical and Fuel

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Abstract
Furfural is produced from lignocellulose-biomass enriched with pentose derivatives. It is a precursor of furan-based chemicals and has the potential to become a green platform chemical. This review presents and compares the different routes and recent trends furfural production and utilization. Conventional process of furfural production from lignocellulosic biomass, especially cellulose pulp, had low conversion rate and produced lots of unused by-products. Thus, the production in industrial scale was impeded with economical feasibility. While, the recent production technologies were focused on the utilization of the feedstock and improvement of the process economics by co-production of other high value products and furfural derivatives. Regarding to the current situation, the fuel production from non-renewable fossil sources becomes more interesting to worldwide society due to the environmental concern. Furfural and its derivatives are selected chemicals to be used as alternative feedstock to substitute the use of fossil fuels. Nowadays, the improvements on production technology and process are still necessary to compete with petroleum-based products in term of economic aspects.

Keywords: Furfural, Lignocellulosic biomass, Fuel, Platform chemicals, Biorefinery

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1 Introduction

Furfural is produced from agricultural biomass enriched with pantosan (poly-pentose) by the unified process of pantosan acid hydrolysis and dehydration to furfural [1]. It is one of a major platform chemicals for many specialized chemical products, especially furfural alcohol and others derivatives. Its main applications are applied as a raw material for resin production and as a solvent in petroleum lubricant production and other purposes, for example adhesive agents and flavouring agents [2]. The furfural production in the industrial scale was established in 1921 by the Quaker Oats company using oat hulls as main raw material. Due to limited demands, the production technology had not really been improved until 1980.

The productions of value-added chemicals, fuels and energy from lignocellulosic biomass based on biorefinery concept have gained more attentions worldwide, due to the concern of environmental problems [3]. Thus, the utilization of the feedstock and improvement of process economics have been focused on integrated strategies by co-production of other high valued products together with furfural. This improved concept of production drives the economical feasibility of furfural production in industrial scale.

2 Furfural and Its Derivatives

The molecular formula of furfural is $C_5H_4O_2$ as a heterocyclic aldehyde with the closed ring structure (Figure 1). Its synonyms are: 2-furancarboxaldehyde, furaldehyde, 2-furanaldehyde, 2-furfuraldehyde, fural and furfurol [4]. It is colorless oily liquid, with the smell of almonds, and quickly darkens in color when exposed to air.

Currently, furfural is utilized as a precursor of many downstream chemical industries because it could be converted to many solvents, polymers, fuels and other intermediated chemicals. It could be hydrogenated to furfural alcohol, which is subsequently used as an intermediate chemical for ranitidine production, an anti-ulcer drug, in the pharmaceutical industry [5]. The hydrogenation of furfural alcohol yields the tetrahydrofurfuryl alcohol, which is used as a solvent for herbicide formulation in agricultural activities, for example δ-aminolevulini acid derived from 5-(chloromothyl) furfural [6]. The other important

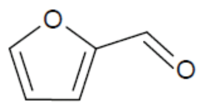

Figure 1: The molecular formula of furfural.

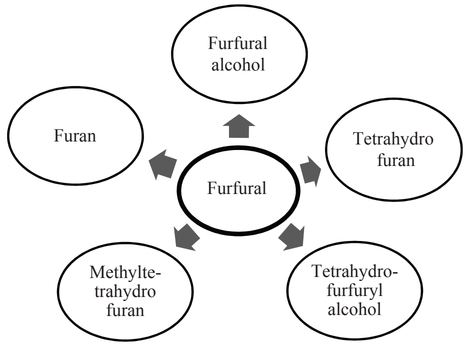

Figure 2: Furfural derivatives.

furfural derivatives are methyltetrahydrofuran and furan due to their wide applications, for example as components in alternative fuels, lithium electrodes and heat-treated commercial foods.

Furfural is mainly used in manufacturing of furfural alcohol. It is also used in synthesis of many specialty chemical products such as furan, tetrahydrofuran, tetrahydrofurfuryl alcohol, and methyltetrahydrofuran (Figure 2). Furfural is used as an extracting solvent to produce lubricant oil and as a solvent in manufacturing of anthracene and resins. As a resin, phenolic furfural is a good example, which occurs from polymerization of nitrophenol, furfural and NaOH [7]. Furfural-derived lubricant oil is produced by using Bechtel® manufacturing process to produce high-quality lubricating oil, which called as "Furfural refining" process [8]. Furfural is also utilized as a solvent for extraction and removal of undesirable compounds and impurity compounds in crude oil distillates that possibly lead to low quality of lubricating oil. Furfural solvent can remove aromatics and other compounds containing heteroatoms such as oxygen, nitrogen, sulfur and metals [9]. Using furfural as an extracting solvent in the process gives high aromatic content as by-products, which can be used for other downstream applications. Moreover, unlike phenol as extracting solvent, furfural is also a non-toxic chemical. Additionally, furfural also functions as a substrate of many derivatives, such as furoic acid that is used in sterilized and pasteurized food [10]. Furfurylamine, a furfural derivative, is used in pharmaceutical synthesis or in engine cleaning solutions. Based on multi-functions of furfural and its derivatives, furfural is considered as a major platform chemical, which is used as an intermediate and a substrate to other industrial chemicals.

3 Production of Furfural

Furfural is a product obtained from dehydration of xylose sugar, which often found in large quantities in hemicellulose of lignocellulosic biomass. Any feedstock containing large amounts of pentose sugars can serve as a raw material for furfural manufacturing. Thus, it is conceptually possible to utilize agricultural waste that is left unused in the fields after harvesting seasons as supply of the process. However, the contents of pentose sugars and furfurals in hemicellulose in different plants are varied, such as in almond husks, sugarcane bagasses, corn cobs have the pentosan contents of 30, 25, and 35%, respectively. Additionally, the different extraction technologies could give different yields of pentose and furfural from biomass ranging from 25 to 75% [2], [3], [11]. Many of furfural manufacturers use sugarcane bagasses and corncobs as feedstock materials. Corncob is the main supplied material used in China and Thailand.

Furfural production from lignocellulosic biomass by using three-step processing had been developed during the last century. Firstly, lignocellulosic biomass is chemically pretreated with sulfuric acid to facilitate the extraction step by modifying the recalcitrant structure of lignocellulosic biomass to facilitate the access of solvent or water to the biomass [12]–[16]. Pretreatment methods can be mainly categorized into physical, chemical, biological and physico-chemical techniques. The common method is the combination of physico-chemical pretreatment by milling and blending with chemicals such as sulfuric acid. Secondly, the hot steam is added to the reactor to hydrolyse the polymeric structures of lignocellulosic biomass to small molecules. Saturated steam efficiently hydrolyses the hemicelluloses, modifies the lignin (as an inhibitor), increases the surface area of biomass, and decreases the cellulose crystallinity and degree of polymerization [17]. Finally, furfural is refined and recovered with an azeotropic distillation column (Figure 3).

The commercial furfural production was conducted using batch or continuous processes with mineral acids as catalysts [18]. The Quakers Oats process is the initiation of furfural production in batch mode in 1921. The hydrolysis step was conducted at low temperature conditions (153°C). A screw press machine was operated to separate the spent liquor from the extracted product. Then, furfural product was refined using successively azeotropic distillation columns, composing of decantation and dehydration columns. This process was reported to have significant disadvantages, such as long residence time and high sulfuric acid concentration were required because of the low temperature [1]. Additionally, the furfural yield of 40–52% was obtained leading to unprofitable and unviable process in an economic aspect.

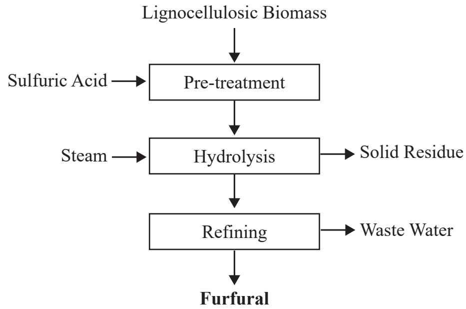

Figure 3: Conventional process of furfural production from lignocellulosic biomass.

In 1940, the production technology was developed in the batch mode to be a simple and inexpensive process. The concept of this process was similar to the Quakers Oats process, but with the better energy efficiency by reusing of the saturated furfural vapors in the boiler of the distillation column. In 1960, the continuous production process by Quaker Oats technology was designed and operated in the reactor (containing paddles to promote mixing and mass transfer) equipped with several steam injectors to obtain vaporized products and the unused residues in the discharged system. However, this process was not economically viable due to its very high maintenance cost for the drives of the auger presses.

In 1988, the SupraYield, a continuous production process, was invented using high temperature condition (220°C) and the reactor was reduced to a simple pipe. This process had many advantages including short residence time (small reactor volume needed), high concentration of furfural in the vapor products (furfural yield of 50–70%), and acid reutilization. the Suprayield process was used by South African producers. The main pitfall of this process was the high cost of investment and maintenance [2]. In 2004, the continuous process, so called Westpro-modified Huaxia technology [19], used fixed-bed reactors and continuous dynamic refining. It gave furfural yield of 55% with high purity of 99% and low amounts of by-products using a low production cost and low capital investment about only one third of other furfural technologies [4]. Nowadays, batch processing is mostly used, especially in China as the biggest furfural producers of the world, due to the advantage in labour cost. Huaxia Technology, a subsidiary of Westpro Company in California, is using a variation of the Quaker Oats technology to produce furfural in China [2].

In furfural production process from lignocellulosic biomass, pre-hydrolysed liquor is considered as unused by-products that composed of hemicelluloses, acetic acid, lignin, and furfural and can be burnt for energy production [20]. However, it has high water content (>90%) with the low heating value of hemicellulose, lignin and acetic acid. The composition of pre-hydrolysed liquor causes inefficient combustion for energy production. Therefore, the pretreatment is necessary before hydrolysis to remove lignin, to facilitate processing and to minimize the side reactions resulting in the improvement of the furfural yield. Lignin is a high molecular weight polymeric substance and is responsible of fouling of reactors. Different pretreatment methods were proposed and experimentally demonstrated to remove lignin including using alkaline treatment, flocculation, and adsorption using activated carbon or other absorbents. For acetic acid and excess water removal, several conventional techniques could be applied, for example, distillation, membrane filtration or ion exchange filtration [21]. However, each technique has different pros and cons, which requires consideration of economic viability and impacts to the environment when those unused by-products were released from the process. After pretreatment, the liquor could be further processed to the hydrolysis reaction and conversion to furfural with different production technologies as currently available [22].

Not only the lignocellulosic biomass that could be used for furfural production, but black liquor obtained from paper processing plant has also been used as raw material for furfural production using 4 step operations, including (i) lignin precipitation, (ii) hemicellulose enrichment, (iii) chemical conversion of xylan to furfural, and (iv) furfural purification [23]. In brief, lignin is precipitated from black liquor using addition of CO_2 to form $NaHCO_3$ and becomes insoluble solid precipitates. The liquor is concentrated by using ultrafiltration and the excess water and inorganic salts is removed by using nanofiltration. The xylose remaining in black liquor is converted to furfural by heterogeneous catalyst-activated reaction. Finally, furfural is purified using an azeotropic distillation and the yield of furfural could be approximately 95% [23]. Although, the concept of furfural production from pre-hydrolysed liquor and black liquor are possible. One of advantages of this concept is utilization of industrial by-products. Yet, this concept has been demonstrated only in experimental scales. The bottleneck of this process is due to the critical requirements in costly pretreatment and multi-step separation.

Due to gaining interests of industrial producers and consumers, the furfural production have been continuously improved with novel technologies. Microwave-assisted technology becomes popular for application in chemical reactions over the past years because of short reaction time and highly efficient reaction rates, which gives higher yield and better selectivity than conventional process. Serrano-Ruiz *et al.* (2012) reported that a microwave reactor improved reaction rate for furfural production by assisting on dehydration reaction, a rate limiting step of the process [24]. In comparison to the conventional heating process, 80–90% furfural conversion yield could be obtained from xylose dehydration using ionic liquid and required 4–10 hours of reaction. When using microwave heating, the reaction rate was significantly increased, and 85% furfural yield could be obtained under the operational condition at 180°C in 1 hour. However, the microwave heating was tested only in laboratory scale. To up-scale to pilot-scale or industrial-scale production, several works are needed to be investigated; for example, designing of large scale microwave heating, continuous mode of microwave generator, heat value, and electricity etc.

In addition, reactive distillation was investigated as an alternative process for furfural production by the combination of reactor and separator. The reactive distillation technology has many advantages, including immediate furfural separation from reactive zone, minimization of undesired product foaming and also heat integration benefits. Metkar *et al*., showed that the highest furfural yield at more than 75% could be obtained using reactive distillation with zeolite H-mordenite (Si/Al = 10) as solid catalyst on dehydration process of xylose [25].

4 Bio-Refinery Concept for Furfural Production

Biorefinery is a multi-disciplinary concept of process for production of the value-added chemicals, materials, energy and fuels from biomass [22]. Different strategies of co-productions were developed to fully utilize feedstock and improved process economics [26]. Biorefinery has been continuously developed since the last decade with the concept of circular economy by finding the use of unused biomass to produce desired products to reduce the impacts to the world environment. Biorefinery process has been compared to the petro-refinery process with similar purpose to produce the wide variety of products for industrial and household needs. Biomass-based platform chemicals, for example itaconic acid, levulinic acid, polyols, diacid etc. are conceptually defined as the chemicals that have potential to be divergently converted to variety of other chemicals [27]. Furfural is also a promising renewable platform chemical for wide range of end-users and producers [28].

Based on biorefinery concept, in 1990, the Biofine process, which is the patented process to convert biomass to furfural and levulinic acid [29]. The Biofine process comprises of two reactors, including a downstream pre-mixer for steam hydrolysis reaction and a well-mixed reactor for levulinic acid production. This process could provide a theoretical yield approximately of 70% from C_5 sugars, which is about 50% conversion of the biomass [30]. Based on this co-production concept, the paper mill sludge was used as feed for the Biofine process at 3,000 ton per year for the commercial plant in Caserta, Italy. In this process, furfural and levulinic acid were produced from the stripping or distillation, which required large amounts of boiling water and energy [26].

Vedernikovs process was developed in 2001 by the Latvian State Institute of Wood Chemistry to enhance furfural yields and minimize cellulose loss. This process required small amount of concentrated acid and salts used in hydrolysis and dehydration reactions. The advantages of this process are the increase of furfural production yield from 55% to 75% based on the theoretical value and the 5-fold reduction of cellulose degradation degree [26]. Then, the commercial plant was built in Iran in 2006 using Vedernikovs process [30]. Recently, this technology claims that the theoretical yield can be increased up to 90% when ethanol is produced from the residual cellulose.

The mixtures of C_5 and C_6 sugars in lignocellulosic biomass are hydrolysed by addition of mineral acid and subsequently dehydrated to furanic aldehydes, furfural, hydroxymethyl furfural and 5-methylfurfural [31]. The hydrogenation of furfural or hydroxymethyl furfural gives furfural derivatives with similar boiling points and octane numbers to ethanol which suitable for using as an additive in the alternative fuels. However, it is not appropriate to be used directly as a drop-in fuel because of the short carbon chain, so C-C re-forming reaction is necessary [32]. The synthesis of longer-chain hydrocarbons from furfural is formed by aldol condensation and dimerization reactions followed by hydrodeoxygenation, which can produce C_8 to C_{13}+ alkanes [26].

Ershov *et al*. revealed that all the furfural derivatives have a wide range of oxygenated compounds that are suitable as fuel components [33]. They have less oxygen content than octane boosters used today (ethanol, methyl tert-butyl ether, and N-methylaniline) and cause high volumetric heat of combustion (from 25.2 to 30.7 MJ/L). Moreover, the properties of furfural derivatives are low heat of vaporization, low boiling point (ranges from 31.1°C to 189.0°C) and low crystallization points (below 50°C) that appropriate for using as octane boosters and antiknock additives. Based on the engine test, 2-methylfuran and furfurylamine showed physicochemical and functional properties to be promising octane boosters.

5 Furfural Market

In 2001, furfural world production was more than 280,000 ton per annual. At this time, the main producer of furfural was China at rate of 200,000 ton per year (equivalent to 71.42%), followed by Dominican Republic and South Africa at 32,000 and 20,000 tons per year, respectively. In Thailand, about 8,500 tons per year were produced [4]. The global market growth rates for furfural and its derivative are in the range of 2.47–3.1%

per year with the main furfural market size and price of 370,000 tons per year and 1,500 USD per ton, respectively [34]. The major consumptions were dedicated to furfural and furfural alcohol at 50,000–100,000 and 130,000 tons per year, respectively. The biggest consumers of furfural and its derivatives are USA, Japan, and Europe at 48,000, 21,000, and 19,000 tons per year, respectively. For pricing, each furfural derivative has quite big variation related to downstream requirements in the markets. The price was ranged in 900–1,500 USD/ton for furfural, 1,400–1,800 USD/ton for furfural alcohol, 1,000–2,000 USD/ton for furan resin, and 5,600–6,000 USD/ton for tetrahydrofuran [4].

According to a new report published by Transparency Market Research (TMR) in 2017, the global market of furfural derivatives was valued at around 2.0 USD/kg and was anticipated to expand at a compound annual growth rate (CAGR) of over 3.1% from 2018 to 2026 [35]. The market value in Asia Pacific is anticipated to expand at a CAGR of over 3.5% during the forecast period [35]. Increasing demand for furfural derivatives in bio-based products drives the majority of global market in pharmaceutical and oil refining industries.

6 Conclusions

Furfural is used as a platform chemical due to its versatile capability to convert to varieties of high value-added products. It could be produced from agricultural biomass rich in pantosan. However, the complexity of these compound mixtures requires costly and complex pretreatment steps which makes this route of production unprofitable. is a precursor of furan-based chemicals. The improvement of co-production process of furfural and other platform chemicals could be an interesting alternative methodology to obtain economical feasible process. The improvements on technology and process strategies based on biorefinery concept are still essential to make furfural and its derivatives competitive with the petroleum-based products.

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