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Review

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Review

Bioethanol Production from Waste and Nonsalable Date Palm (*Phoenix dactylifera* L.) Fruits: Potentials and Challenges

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Abstract: Balancing the needs of current and future generations stimulates investment for sustainable development such as converting waste biomass into biorenewables. Sugar-based ethanol production is a well-investigated and established process, and researchers are now focusing on the transformation of cellulosic biomass to sugar and the application of non-conventional methods for ethanol production. The State of Qatar generates date palm fruit waste of approximately 4505 tons annually, excluding ornamental palms and palms outside the farms that bear nonmarketable date fruits. Date fruit molasses contains fermentable sugars, representing 75% of the total fruit mass, which can offer a good source for bioethanol production through anaerobic fermentation and distillation. On this basis, the valorization of waste date fruits can be an effective zero-waste strategy via biotransformation into bio-renewable materials, hence, contributing to the achievement of sustainable development goals. This paper reviews the potentials and challenges for the utilization of waste date fruits as a bioethanol source and assesses the abundance of waste date fruits as raw material for the conventional bioethanol conversion process. The review also identified conventional and nonconventional processes for bioethanol production and their applicability to Qatar. Finally, this confirmed ample demand for bioethanol ranging from fuel and industrial chemicals to pharmaceutical alcohol.

Keywords: date palm; *Phoenix dactylifera* L.; bioconversion; sustainable development; waste management; circular economy



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1. Introduction

Date palms are one of the oldest fruit trees in the Arabian Peninsula with the earliest evidence of cultivation dated 4000–5000 BC in southern Mesopotamia (presently Iraq) [1]. Date palm (*Phoenix dactylifera* L.) is a member of the monocotyledonous Arecaceae (former *Palmae*) family classified as a tall, evergreen, dioecious plant that is extensively cultivated because of its sweet edible fruit. Date fruits are high in carbohydrates, amino acids, and other elements that are an important source of human nutrition. Date palm trees are relatively resistant to drought and can tolerate high levels of salinity, which makes them suitable to be cultivated in arid regions such as North Africa, Middle East, and South Asia.

The world's leading date fruit producer in 2020 is Egypt with 1,690,959 tons (Figure 1, [2]). Date cultivation in Egypt has increased by more than 100% since 1993 with an estimated 15,582,000 date palm trees [3]. In the Gulf Cooperation Council (GCC) countries, Saudi Arabia topped the rank in date production. The farmers in GCC countries are growing more than 600 date varieties, and the most important and commercial varieties are Mejdoul, Fard, Khalas, Barhi, and Sukkari [4,5]. Nowadays, the national and international markets are demanding that the date producers focus on producing high-value-added date fruit varieties to meet market demands. Moreover, date producers are advised to improve productivity by cultivating higher yielding date palm trees. The characteristics of the dates, such as size, shape, color, and skin texture, are well inspected by consumers, and although

setting criteria are advantageous, this creates obstacles to date palm growers, especially during processing and marketing, e.g., grading, sorting during harvest and post-harvest activities. If the quality of the date fruits is below the standard then the fruits are rejected, which results in not only a significant loss to the farmers but also a compounded agricultural waste that needs to be addressed. It is necessarily important to convert date fruit wastes into useful products of economic value, and a waste recycling procedure must be established to attain environmental balance. The high sugar content of 75% [5,6] in date fruits offers a suitable source for bioethanol production through anaerobic fermentation. When the COVID-19 pandemic spread over the entire world, the World Health Organization (WHO) recommended the use of alcohol-based disinfectants and sanitizers to disinfect surfaces and hands when soap and water were not available. The aim of this paper is to substantiate evidence using literature on the viability of converting waste and nonsalable date palm fruits in Qatar into renewable biological resources (e.g., fuel, commodity chemicals, and bioethanol for pharmaceutical uses) and to promote the sustainable development vision of the country. Furthermore, the authors seek to address the following specific criteria regarding bio-circular economy, as well as provide a potential solution to an overarching agricultural waste problem:

- (a) sufficiency of the raw material supply and adequacy of active component content;
- (b) availability and maturity of the conversion and production technology; and
- (c) versatility of the product to society's demand.

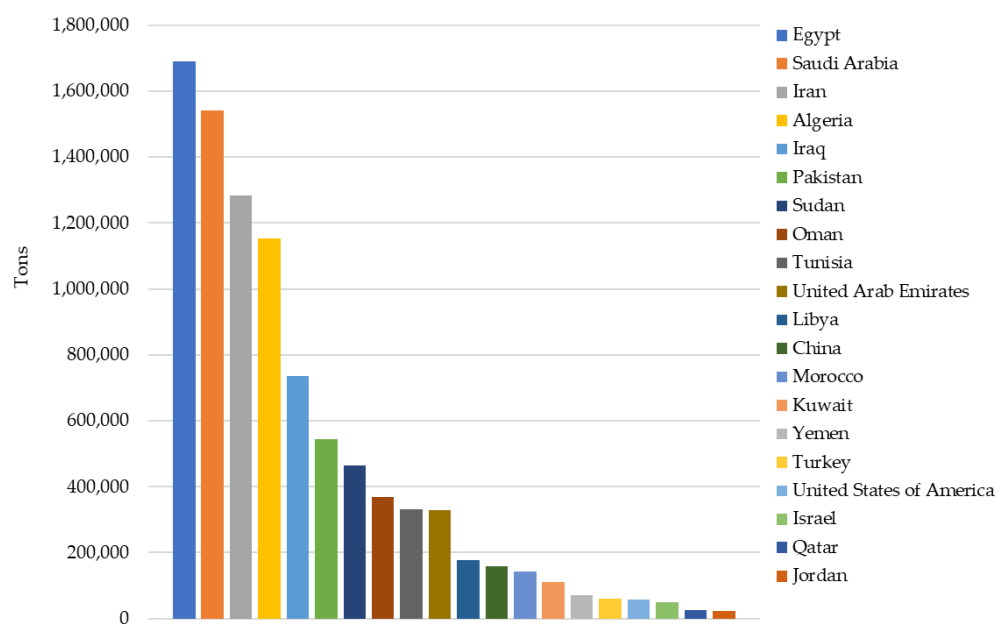


Figure 1. The world's leaders in date fruit production, adapted from [2].

Although the state of Qatar is endowed with abundant hydrocarbon resources, the state formulated a national strategy, among others, to provide framework for sustainable development. In one of the challenges stipulated in the Qatar National Vision 2030, the state is keen to create new sources of renewable wealth. In this respect, this study examines the scenario of date palm fruit production and waste generation in Qatar, the valorization of date fruit wastes and bioethanol production, and the sustainability of bulk chemical production through bioconversion and ethanol uses.

2. Scenario of Date Palm Fruit Production and Waste Generation in Qatar

2.1. Date Production in Qatar

Qatar ranks as the 19th largest date-producing country in the world with an annual production of approximately 26,000 tons of date fruits in 2020 (Figure 1) [2]. Its date palm tree reserve was estimated in 2010 as 581,336 trees in an area of 2469 hectares (ha), which

represents 71% of the total area planted under fruit trees [7]. Since then, the planted areas have increased at an average annual rate of 1.38% [4]. As of 2018, the total agricultural land of Qatar is 74,000 ha, while the total arable land is 21,000 ha (Figures 2 and 3, [8]). The production of dates is only 7.2% of the total agricultural sector, while other crops include fodder crops (41%), vegetables (34%), fruit trees (9%), and cereals (1.2%) [7]. Major date palm tree cultivations are in the north and central areas of the country where environmental conditions are favorable and the soil is characterized with low salinity.

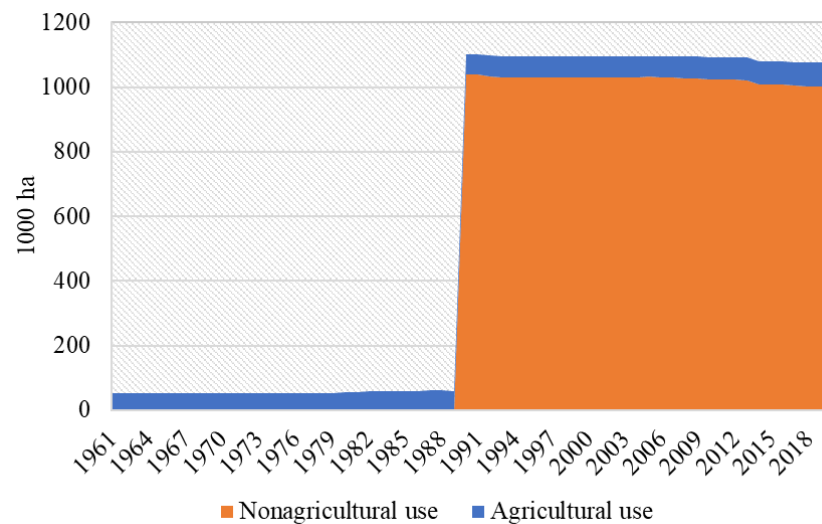


Figure 2. Land use in Qatar, adapted from [8].

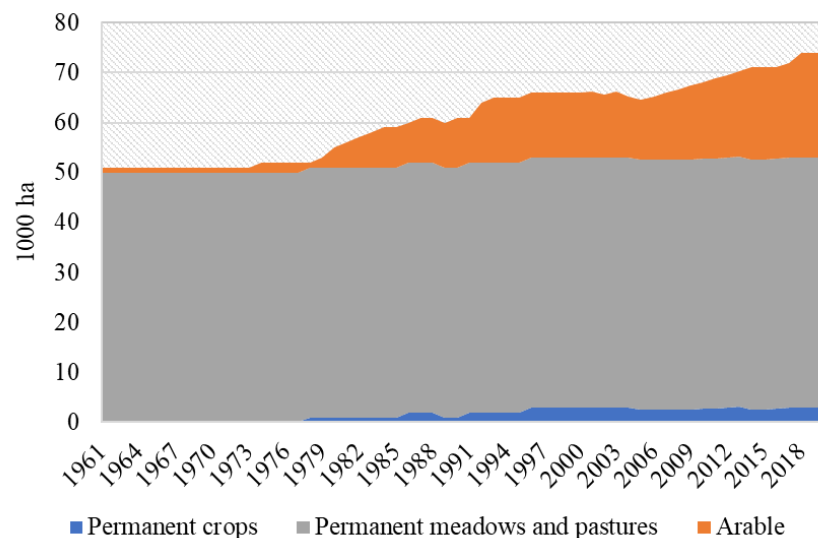


Figure 3. Agricultural area in Qatar, adapted from [8].

In 2019, Qatar imported approximately 4607 tons of dates from different countries with Oman as the top exporter [9]. Over the years, a steady increase in date production was observed (Figure 4), achieving an 82% rate of self-sufficiency, as recorded in August 2022, which is 6% higher than the past 2 years [2,10]. Qatar's agricultural self-sufficiency has increased progressively due to the government's effort on a National Food Security Program that is intended to increase domestic agricultural produce, including livestock, to reduce dependence on foreign countries. The government also holds an annual Fresh Dates Festival to showcase the farmer's products and encourage them to continue growing date fruits and improve their quality.

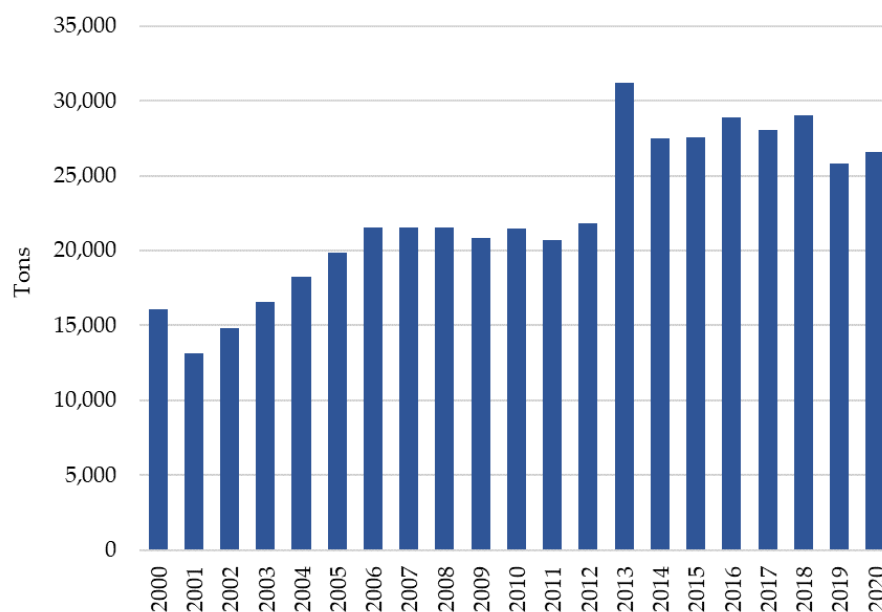


Figure 4. Two decades of date fruit production in Qatar, adapted from [2].

2.2. Agricultural Challenges in Date Production

Date palms can bear fruit within 7 to 10 years after planting, and mature trees can produce 68–176 kg of date fruits per harvest season [11]. Yield potential, however, varies depending on the characteristics of the date cultivar and agricultural management and practices. The major constraints in date production are the following [4,7]:

- a. Low-yielding date cultivars, especially among old date palm trees.
- b. Low date fruit quality and long fruit generation time.
- c. Increased number of non-productive male plants in the field.
- d. Inability to distinguish date palm varieties.
- e. Low resistance to diseases and pests.
- f. Low tolerance to soil and water salinity.
- g. Low response to fertilizers.
- h. Insufficient good water quality and irrigation system.

Other factors that contribute to low yield production are the shortage of arable land, urbanization, poor farm management and agronomic techniques, losses in date fruit harvests, improper pre- and postharvest care, lack of awareness and technology training in date palm growers, inadequate technology transfer, and collaboration/cooperation with high date producing countries. The current agricultural management and practices of date cultivars are from inherited experiences and recommendations from farmers in other date-producing countries. A promising development was the establishment of an on-going tissue culture laboratory in Qatar that uses offshoots, also known as suckers, in date propagation [1,7,12]. A more recent development in date palm cultivation is the use of somatic embryogenesis to propagate favored varieties [12]. Studies were also conducted to differentiate between date cultivars and to predict the gender of immature trees using genome sequencing [12].

The scarcity of fresh water in Qatar makes it entirely dependent on desalinated seawater for potable water. Water irrigation plays an important role in the growth and development of date palms. Relying exclusively on desalinated potable water is unsustainable for Qatar's growing agriculture sector. The water requirements of date palm trees vary depending on soil texture, soil and water salinity, plant age, irrigation system, seasons, and climatic conditions (light, temperature, humidity, wind). The arid climate causes high water evaporation, thus, requiring a greater water supply for irrigating date palm trees, as well as other agricultural crops. The sandy nature of most soils implicates high drainage and leaching losses, which reduce the water-retention capacity of the soils. On average, date palms require 15,000–35,000 m³ ha⁻¹ of water per year, which is quite high and difficult

to supply in a water-scarce country [7]. The difficulty of provision of water is expected to significantly continue due to the impact of global and regional climate change, as well as the competition with domestic consumption. With the growing need for water irrigation in the agricultural sector, there is a need to address the water shortages and acquire new water sources. The use of treated sewage effluent and wastewater have the potential to supplement the growing demand of water, replacing the desalinated potable water for irrigation. In Islamic countries such as Qatar, traditional cultural beliefs and practices have hindered the use of treated wastewater except for irrigating the plants in parks and amenity areas. Even to some urban greenery landscape and vegetation, potable water is still utilized for irrigation. Sharia Law allows the use of treated sewage wastewater if deemed clean and safe and views the reuse of wastewater as part of the optimization, supplementary to the existing resources, thus, misusing it is considered a disobedience to God [13]. Adequate treatment and monitoring are, therefore, essential to ensure the safe reuse of treated water, and information campaigns in Muslim communities concerning the environmental aspects of their faith while respecting Islamic ethics are also necessary.

2.3. Date Palm Fruit Waste Sources

Date fruits from ornamental date palm trees contain heavy metals, such as Al, As, Pb, Cr, Cd, Hg, and Sr [14]. Other metallic minerals such as Cu, Ni, Mn, and Fe are essential micronutrients necessary for plant physiological processes but are toxic when their quantities are higher than the plant requirement [15]. Plants absorb heavy metals as mineral ions that are dissolved in water or soil moisture through their root system during nutrient uptake. Heavy metals are naturally found in soil, but their concentrations can be enhanced by anthropogenic activities and are location dependent. The date fruit obtained from Saudi Arabia contained high Pb and Cd in areas with heavy traffic density [16]. Ahmed et al., 2022 [14] reported the presence of Cd (<0.001 mg/kg), As (<0.009 mg/kg), and Cr (<0.005 mg/kg) in date fruits obtained from United Arab Emirates (UAE). For plants irrigated with treated sewage water, heavy metals tend to accumulate during watering of the soil. Abdulaal et al., 2017 [17] listed the heavy metals they found during their investigation on the effects of using sewage water to irrigate some known commercial date palm varieties in Saudi Arabia (Table 1). They found higher heavy metal accumulation in different varieties of date fruits irrigated with wastewater compared to using non-wastewater-based irrigation. The use of treated wastewater for irrigation purpose is a common practice in arid regions due to scarcity of water for agriculture. In Qatar, irrigation using treated wastewater did not affect the yield and fruit quality of date palm trees. No accumulation of heavy metals was observed in the leaves and date fruits when treated wastewater was used in comparison with groundwater [7].

Table 1. Detected heavy metal concentrations from different variety of fully ripened date fruits (mg kg⁻¹, adapted from [17]).

| Date Cultivar | Agwa | | Anbar | | Safawi | |
|---------------|--------|--------|--------|--------|--------|--------|
| | MW | SW | MW | SW | MW | SW |
| Cr | 0.9087 | 0.9836 | 0.3407 | 0.4799 | 0.0188 | 0.1100 |
| Cu | 1.4668 | 1.5334 | 0.2543 | 0.4466 | 0.4785 | 0.5113 |
| Fe | 17.917 | 20.565 | 16.342 | 17.958 | 14.94 | 16.077 |
| Mn | 2.4299 | 3.2355 | 0.2076 | 0.4997 | 0.7985 | 1.25 |
| Pb | 1.122 | 1.459 | 1.148 | 1.48 | 0.903 | 0.982 |
| Zn | 0.0723 | 0.0986 | 0.0967 | 0.1234 | 0.1025 | 0.1153 |

HM, heavy metals; MW, municipal water; and SW, sewage water.

The consumption of date fruits obtained from outside of the farms and the ones that received treated wastewater is still under scrutiny due to toxicological concerns associated with their growth, as well as the cultural belief perspective. These fruits are contrived

to be categorized as substandard and nonsaleable date fruits. Alternatively, problems in date palm reproductive biology can also lead to fruits with no commercial value. Inefficient pollination may cause parthenocarpic fruits or fruits that are seedless [18]. The problematic reproductive phenomenon may occur in date palms within the farm, and those palms grown not for food, such as ornamental dates, may cause date fruits to have low marketability or be nonsaleable and, therefore, become date fruit wastes.

The demand for good quality date fruits has increased in local and international markets. After sorting and grading, date fruits are categorized as low or high quality. High quality date fruits are harvested for consumption. The local market mostly absorbed low quality dates at a very low price for consumers. Low quality fruit dates can also be used as feed for the livestock sector [19]. The high-quality date fruits have high marketability; however, a high quantity of waste is also generated at various stages of the sorting of the date fruit.

3. Valorization of Waste Date Fruits and Bioethanol Production

3.1. Valorization of Waste Date Palm Fruits

Agriculture has played a significant role in the provision of food and is a major factor in the economic development of the countries around the world. In the past, the agricultural sector in the harsh arid climate of Qatar was faced with many challenges, primarily due to the lack of water for irrigation. This limited Qatar's economic activities to pearl diving, trading, and fishing [20]. Recently, Qatar discovered its oil and gas reserves, which led to the country's great wealth. Agricultural waste is generated in large quantities annually in the form of agriculture by-products and crop residues [21]. Date palm trees produce various agricultural wastes wherein each tree is estimated to produce 20 kg of dry leaves annually, date pits represent 10% of the date fruit total mass [22,23], and approximately 20% of the annual production of date fruits is lost during the post-harvest process, e.g., over-ripened date fruits, improper storage, handling, transportation, and contamination [24,25]. In Qatar, date fruit wastes could average 4504 tons annually from 2000 up to 2020 (Figure 4). Globally, date palm wastes including fruits and leaves comprise approximately 30 million tons per year [11].

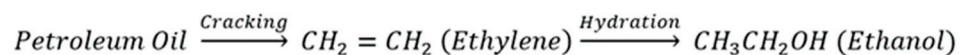
Date palm wastes consist of cellulose, hemicelluloses, lignin, and other compounds that could be bio-converted into value-added products, but they are mostly burnt in farms, causing a serious environmental threat. Low quality date fruits could be used in the processing industry to produce juice, jams, syrup, jelly, and powder by-products. They are rich in carbohydrates, nutrients, antioxidants, phenolic compounds, and fibers that are suitable for feedstock processing. However, these date residues are currently considered as waste and discarded rather than used as a resource with various possibilities for value-added products.

The process of recycling the date fruit wastes is an important biological process that strives to maintain the environment balance. Both date wastes and low commercial value fruits are high in natural sugar that can be transformed and converted into materials with added value, such as ethanol for pharmaceutical use or as animal feed. Fruit date syrup contains monosaccharides, such as fructose and glucose, which constitute over 80% (dry weight) of ripened date fruit [11,26]. Egyptian date fruits were found to contain 73.00% carbohydrates in the form of glucose, fructose, and sucrose, including 2.90% lipids, 3.00% protein; 13.80% moisture, 86.50% total solid, 2.13% ash, and 5.20% crude fibers. They also contain a variety of vitamins, e.g., A, B-complex, nicotinic acid; and essential amino acids, e.g., proline, glycine, histidine, valine, leucine, arginine, and aspartic acid in high concentrations, as well as threonine, serine, methionine, isoleucine, tyrosine, phenylalanine and alanine in low concentrations [27]. Another date variety, Aziza Bouzid and Assiane from Morocco, contains higher percentage of carbohydrates up to 80%, dietary fiber ranging 10.22–12.79% (dry weight basis), and essential amino acids: 184 mg lysine, 122 mg isoleucine, 98 mg threonine per 100 g dry matter [28]. These data support the important components of date fruit and, thus, are essential to explore the possible utilization of date fruit waste.

3.2. Bioethanol Production from Date Fruits

Ethanol derived from waste product biomass has an advantage over fossil fuel. Ethanol production by yeast fermentation is characterized by high selectivity and low by-product formations (Figure 5) [29]. Bioethanol is non-toxic, biodegradable, and eco-friendly and has the potential as a substitute for fossil fuel. It is an important renewable and sustainable alternative clean source of fuel. Different research has been carried out on the use of date palm fruit waste to produce bioethanol, such as Abd-Alla and El-Enany, 2012 [30], Ahmad et al., 2021 [31], Arshad et al., 2017 [32], Taghizadeh-Alisaraei et al., 2019 [11], and Zeinelabdeen et al., 2013 [26]. Date fruit syrup contains fermentable sugars that offer anaerobic fermentation, followed by distillation for bioethanol production. Various microorganisms, such as *Saccharomyces cerevisiae* [31], *Zymomonas mobilis* [33,34], *Clostridium acetobutylicum* and *Bacillus subtilis* mixture [30], *Pachysolen tannophilus* and *Pseudomonas fluorescens* [35], *Acetobacter pasteurianus* [36], *Schizosaccharomyces pombe* [37], *Leuconostoc dextranicum* [38] *Fusarium* sp. [39], including commercial baker's and alcohol yeast [40], can be used during fermentation processes to convert sugar to ethanol. The efficiency of conversion depends on various factors, such as sugar concentration, different supplements to the molasses, pH, the inoculum size of the microorganism, temperature, and time of fermentation [11,29,31,32]. Ahmad et al., 2021 [31] discovered that 38% (*w/w*) sugar in fermented molasses using a 25% inoculum size (*Saccharomyces cerevisiae*) can yield up to 11.4% (*v/v*) ethanol under the appropriate temperature of 30 °C, pH 4.5, and 72 h incubation time. Elhussieny et al., 2020 [29] genetically engineered the *Saccharomyces cerevisiae* strain (EtB20b) and found an improved ethanol yield to 19.5% with 20.8% inoculum size at 30 °C temperature, pH 6.0, and 72 h of incubation period. The mutated strain of *Saccharomyces cerevisiae* resulted in high stress tolerance and enhanced ethanol productivity and is highly favored as a sustainable solution to biofuel production. It also exhibits resistance to high incubation temperature and oxidative stresses. Another strain, *Zymomonas mobilis*, is also an excellent microorganism used in bioethanol production. It can ferment 200 g/L sugar molasses to 55.8 g/L bioethanol within 48 h [34]. Meanwhile, commercial baker's and alcohol yeasts were used due to their accessibility, inexpensive price, longer shelf life, high cells viability that could exceed 4.6×10^{10} yeast cells g^{-1} , reduced culture work-up and time, little to no microbial contamination, and high tolerance (i.e., to thermal, acid, and alcohol content) [40].

From fossil source:



Sugar fermentation route:

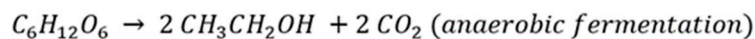


Figure 5. Ethanol formations via two different routes, adapted from [29].

3.3. Advances in Ethanol Production from Fruit Waste

Date fruits that contain high level of reducing sugars (approximately 70–80%) [41], are of great interest to the scientific community to be utilized as a source of bioethanol production through innovative procedures to bring the possibility closer to commercialization. One factor being considered is to make the process less costly, as discussed in the study conducted by Abd-Alla et al., 2012 [30]; substrate costs can make up to approximately 63% of the total cost of Acetato-Butanol-Ethanol (ABE) production, mainly due to the low efficiency of the fermenting agent (*Clostridium* in their case) to convert the substrate. In addition, maintaining the anaerobic conditions satisfactory for optimal efficiency throughout the fermentation process, such as by the addition of a reducing agent or N_2 flushing, increase the cost. Therefore, they investigated mixing in *Bacillus subtilis* DSM 4451 with the *Clostridium acetobutylicum* ATCC 824 to remove the need for maintaining an anaerobic condition for fermentation to lower costs. Their results show that *Bacillus subtilis* DSM

4451 can in fact maintain the anaerobic condition for the ABE production, as shown in Figure 6. They were able to obtain maximum ABE productivity of 0.30 g/L·h and ABE yield of 0.42 at 75 g/L. This was attributed to *Bacillus* acting as an oxygen consumer in the mixed culture to create the necessary anaerobic environment for *Clostridium*; hence, making it feasible to have a cheaper ABE production for date fruit waste.

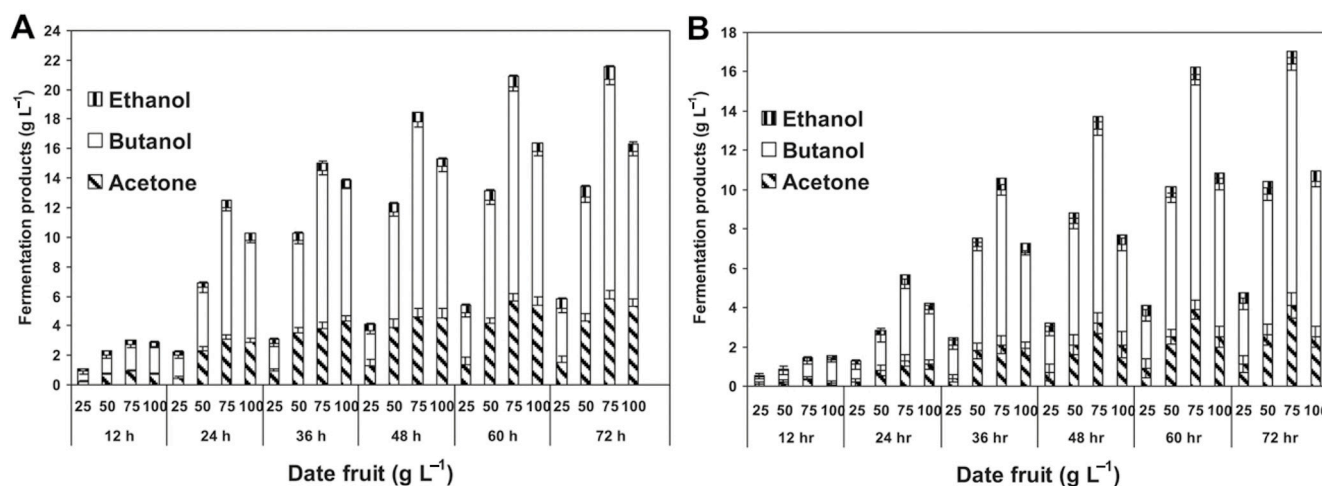


Figure 6. Production of ABE from spoiled date fruits via a mixed culture of *Clostridium* and *Bacillus* without anaerobic treatment (A) or by pure culture *Clostridium* with anaerobic treatment by the addition of L-cysteine hydrochloride and N₂ flushing (B), reprinted with permission from [30]. Copyright 2012, Elsevier B.V.

The idea of utilizing a green energy source to reduce cost for bioethanol production from date fruit wastes was pushed forward in the study of Boulal et al., 2016 [41], where they designed a batch fermenter (Figure 7) that utilized a solar water heater for temperature control and to lower the production cost. Similar to the studies of Taouda et al., 2017 [42]; Bouaziz, 2020 [43]; and Ahmad et al., 2021 [31], they utilized *Saccharomyces cerevisiae* for the date substrate and noticed that between 24 and 48 h, the process of converting glucose to ethanol was active but suddenly decreased after 72 h. The glucose rate decreased from 13.8% to 3% after 72 h, which was determined to be caused by the accumulation of fatty acids, such as octanoic and decanoic acids, that resulted in the cessation of yeast growth. Nevertheless, the study was able to produce ethanol with a high concentration of approximately 90°, with an acceptable productivity of 250 mL/kg or 3.47 mL/kg·h at a moderate cost.

Another factor is to increase the efficiency of ethanol production by employing new technology or techniques such as Very High Gravity (VHG) technology. VHG uses a medium containing a high-sugar concentration (>250 g L⁻¹) with more than 30% solids, used in order to achieve high ethanol concentrations [11]. According to the study by Arshad et al., 2017 [32], VHG is favorable because of its high efficiency of 12–15% of maximum ethanol in contrast to the conventional fermentation of 7–8%, minimal waste generation, and low-cost operation. This can be improved by utilizing a more sugar tolerant strain and applying various strategies such as aeration, nutrient feeding, and media supplementations, for example. Therefore, they utilized a mutant strain that has a higher sugar tolerance called *Saccharomyces cerevisiae* UAF-1 for the production and applied various aeration rates with the VHG technique. They achieved up to 12% (v/v) in ethanol production using a high gravity medium with aeration, which is significantly greater than the 7–9% conventionally produced ethanol. Furthermore, the study determined that water consumption can be reduced up to 35% by using 40° Brix high gravity medium under aeration condition (Figure 8). This reduces effluent generation and cuts down distillation costs. Although this study did not utilize date fruit wastes, the technology and method shown in the study can be applied to enhance and minimize the cost of ethanol production.



Figure 7. Solar Batch Fermenter: (1) fermenter, (2) water tank, (3) solar heater, (4) control panel, (5) pump, (6) data logger, reprinted from [41].

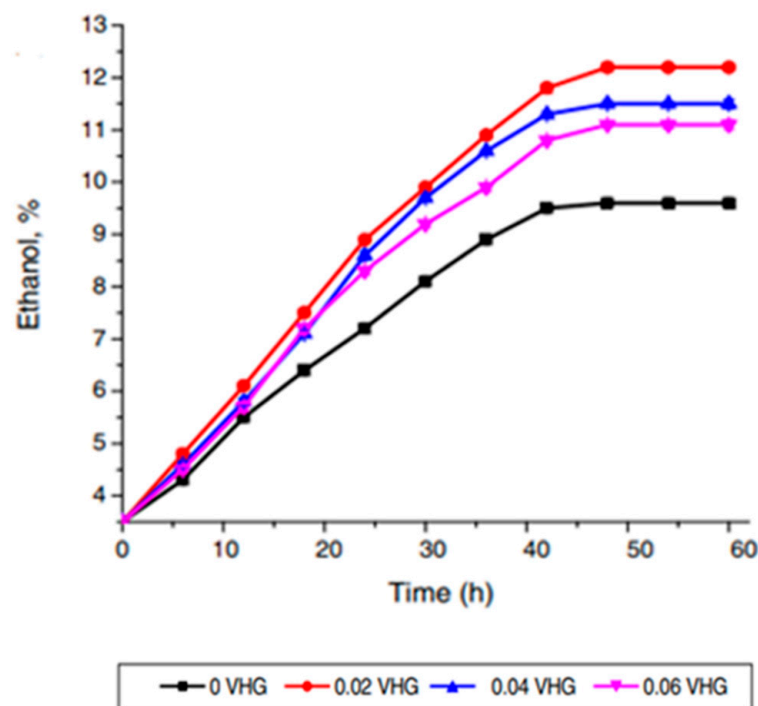


Figure 8. Ethanol production at 40° Brix at different very high gravity levels, reprinted from [32].

Modern technology and advancements in ethanol production may assist date fruit wastes to be a more efficient and sustainable source of bioethanol. Taghizadeh-Alisaraei et al., 2019 [11] discussed the technologies adapted for date waste ethanol production. Some of these developments were in the cooking, liquefaction, and saccharification processes. Saccharification is a process of breaking down complex carbohydrates into ethanol through chemical and/or enzymes means. Chemical saccharification is mostly conducted with sulfuric acid, hydrochloric acid, and nitric acid under a standardized condition of temperatures between 120–140 °C sustained for 15–30 min of reaction time. Enzyme saccharification conducted through enzymatic hydrolysis utilizes lower temperatures such as 90 °C and provides minimal degradation risks. Enzymes such as cellulases, pectinases, and amylases are used for saccharification processes on biomass [44].

Developments on these processes in ethanol production have allowed the cooking temperature to drop from operating at more than 100 °C to approximately 50 °C by synchronizing saccharification and fermentation, which is referred to as simultaneous combined saccharification and fermentation (SSF) [11]. This lowers energy consumption and cuts the costs even further by removing the need for separate equipment for saccharification. SSF provides the added benefit of decreasing the chances of bacterial contamination and reducing restrainability of high sugar concentrations to the microzyme. Yeasts that are used for ethanol production also saw some advancements; for example, sugars from the lignocellulosic biomass, which are mainly composed of glucose and xylose, can now be turned into ethanol by using an improved yeast *Saccharomyces cerevisiae* IPE003 strain [45]. Moreover, converting pentose or five-carbon sugars to ethanol can be made possible through the engineered organisms using genetically modified yeasts. The emergence of hybrid technology for the enzymatic hydrolysis process reduces feedstock conversion costs up to 19.4% and provides a synergistic effect on the pre-treatment of cellulose. In distillation, utilizing molecular sieves for ethanol dehydration rather than the azeotropic distillation helps eliminate the need for carcinogenic materials and removes one of the steps of the distillation process, resulting in energy cost reductions of up to 20%.

4. Sustainability in Bulk Chemical Production through Bioconversion and Uses of Ethanol

4.1. Sustainability in Bulk Chemicals Production through Bioconversion

The impact on the environment with global warming associated with greenhouse gas emissions and the massive exploitation of natural resources is one of the subjects of concern in sustainable development goals. Petroleum-based commodity chemicals are derived from fossil feedstock and generate a tremendous amount of global warming potential gases during production. On the other hand, countries with little to no fossil sources rely on biomass as the principal raw material for commodity chemicals such as ethanol. This implies that bio-based chemical production [46] promotes sustainability as it reduces the environmental footprint. Stoll et al., 2019 [47] laid out the differences between the two alternative ways of producing C₄ chemicals from synthesis gas utilizing petrochemical and biotechnological process routes. The petrochemical feedstocks in Figure 9 are derived from syngas, and different synthesis pathways for C₄ chemicals with microbes are outlined in Figure 10. The benefits of biological tracks were highlighted by Stoll et al., 2019 [47] and they could be prospective synthesis alternatives for complex chemicals when extensively developed.

In addition to the exploitation of industrial microbiology, biomass is gaining momentum in sustainable development. Biomass, as a dense-packed carbohydrate in the form of cellulose, can be converted to fuel or feedstock. Sheldon, 2018 [48] discussed the conversion of polysaccharides into commodity chemicals through bio- and chemocatalytic transformations. The championed conversion mechanism is simplified into a graphical model in Figure 11, published in his article. Furthermore, Haveren et al., 2008 [49] listed bulk chemicals, each with potential bio-based feedstock, that are worth exploring. The bulk chemicals with potential sources that may be applied in Qatar are listed in Table 2.

Table 2. Potential bulk chemical production from bio-based feedstocks, adapted from [49].

| Bulk Chemicals | Bio-Based Feedstocks |
|-----------------|----------------------|
| Ethylene | sucrose/sugar |
| Ethanol | sucrose/sugar |
| ethylene glycol | glycerine |
| 1,4-butanediol | glycerine |
| Benzene | lignin |

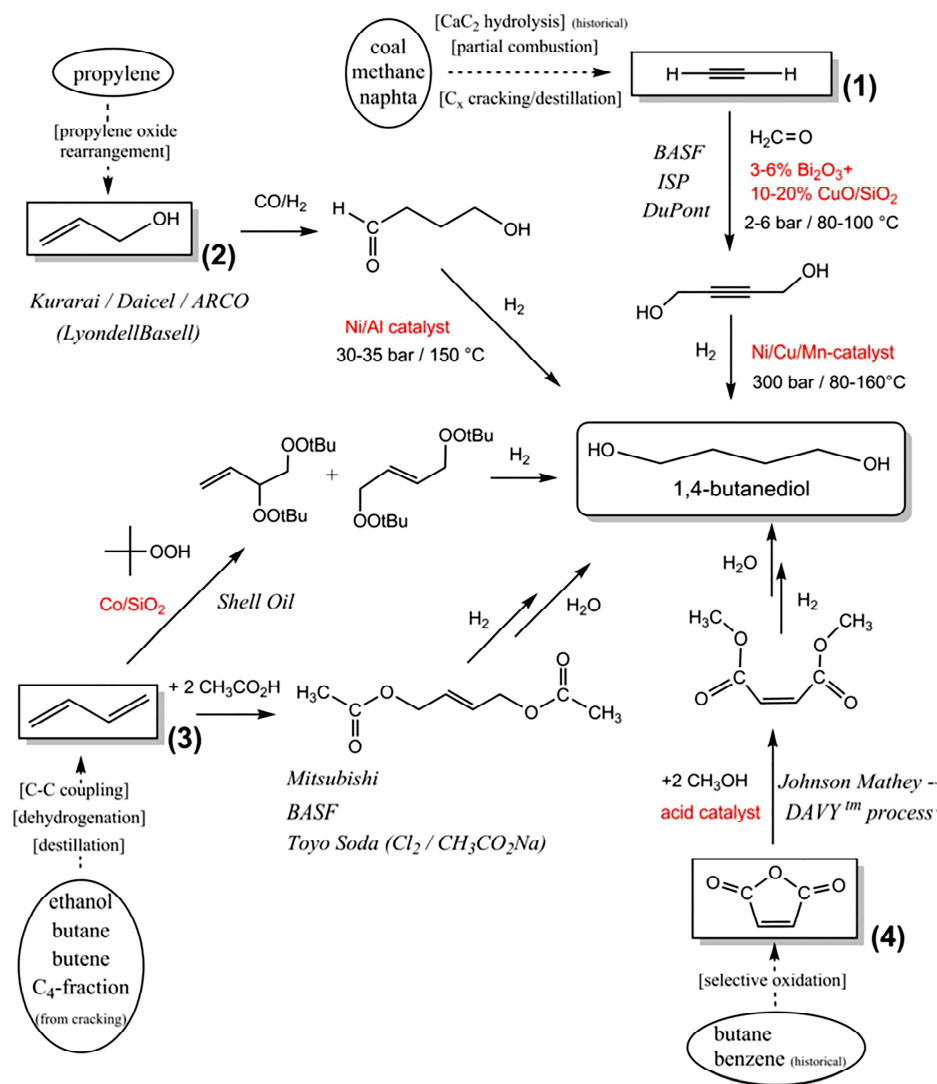


Figure 9. Production routes to 1,4-Butanediol via petrochemical feedstocks: (1) acetylene, (2) allyl alcohol, (3) butadiene, and (4) maleic anhydride, reprinted with permission from [47]. Copyright 2019, American Chemical Society.

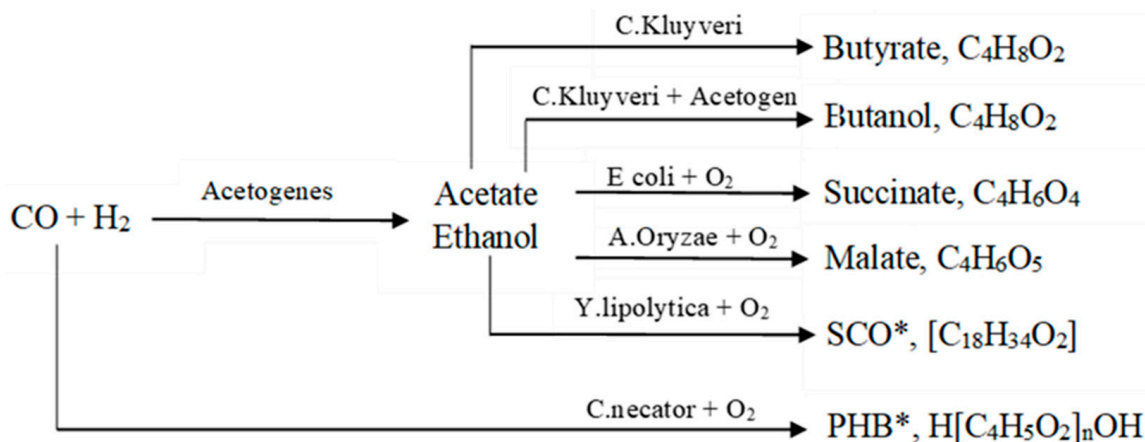


Figure 10. Microbial pathways of C₄ chemical from syngas; * SCO = single cell oil, PHB = polyhydroxybutyric acid, reprinted with permission from [47]. Copyright 2019, American Chemical Society.

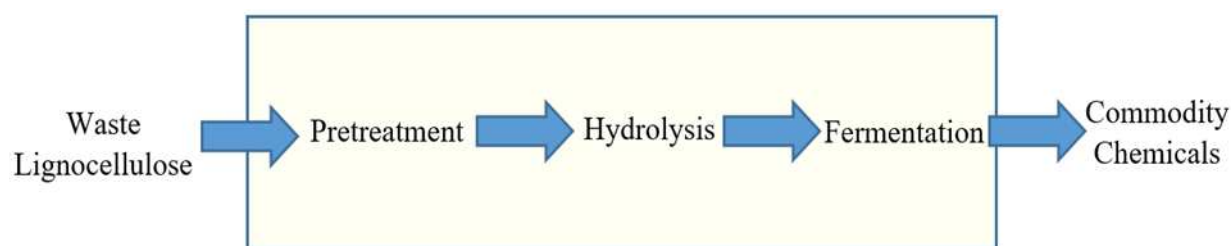


Figure 11. Carbohydrate to commodity chemicals model, adapted with permission from [48]. Copyright 2019, American Chemical Society.

4.2. Uses of Ethanol

The uncertainty of increase in prices for petroleum-based fuels from diminishing reserves and shifting political geography have prompted the scientific community to seek a more sustainable solution, that is, biofuels. One of the most commonly used compounds for biofuel is ethanol, which when blended with gasoline creates “gasohol”, which is a high octane and water-free alcohol. The most common blend of gasohol is E10, that is, 10% ethanol and 90% gasoline. The presence of oxygen in the ethanol molecule helps combust the fuel perfectly, which in turn, reduces air pollution [50]. However, higher ethanol in blends requires modification of the conventional spark ignition engine for optimum performance [51]. Other known blends are E15 (15% ethanol), E20 (20% ethanol), and E85 (85% ethanol), or flex fuel that can only be used by flexible fuel vehicles [52]. Ethanol can be produced in two different ways, that is, either from fermentation of materials such as corn and straw or from petrochemical feedstocks, primarily by the acid-catalyzed hydration of ethylene. The latter method that relies on fossil fuels is far from being a clean and sustainable method of production, thus, ethanol produced from the former is usually referred to as “bioethanol” [53]. Bioethanol is commonly looked upon, especially when ethanol is derived from agricultural wastes that will not compete for food demand, because it provides the key advantages of being harmless, recyclable, renewable, and eco-friendly [31]. In 2016, the global bioethanol production reached a staggering 100.2 billion liters and was predicted to be around 134.5 billion liters by 2024 with United States being the largest producer (Figure 12) [54]. This infers that more countries are recognizing and emphasizing the potential of bioethanol not only for biofuel but also for the wide variety of applications that greatly appeal to countries such as Qatar that envisions itself having better food security, more sustainable environment, and a diverse economy in the near future.

In industry, ethanol can be found to have a broad application. Ethanol is used in the cosmetics industry where it is present in many hair styling products, perfumes, deodorants, and foundations, but it is strictly observed that the alcohol used is denatured ethanol to prevent misuse. Ethanol is used in cosmetics because it is volatile, solubilizing, and facilitates penetration, that is, the active ingredients present in the product are able to penetrate into the superficial layer of the skin and promote deposits on the skin or hair [55].

Furthermore, ethanol is also known to serve many purposes in medicine. As a solvent, it dissolves the active ingredient in medicines that are insoluble, such as in the case when chlorofluorocarbons (CFCs) in inhalers were replaced with hydrofluorocarbon (HFA), utilizing ethanol as a co-solvent to solubilize the active ingredients and excipients with the new formulation. Ethanol is present in a wide variety of concentrations in medicines, from less than 0.1 g in 5 mL (2%) as a flavoring agent in oral medicine to an infusion of paclitaxel for an adult containing 20 g of ethanol (equivalent to 1 glass of wine) [56]. In addition, ethanol is also used as an extraction solvent in herbal medicine to obtain specific compounds, such as in the study conducted by Hikmawanti et al., 2021 [57], where various concentrations of ethanol (50%, 70%, and 96%) were utilized to extract antioxidant compounds from Katuk leaves. As an antidote, ethanol can serve as a cheaper and readily available alternative to fomepizole to remedy ethylene glycol poisoning and methanol poisoning by acting as a competitive inhibitor for alcohol dehydrogenase [58].

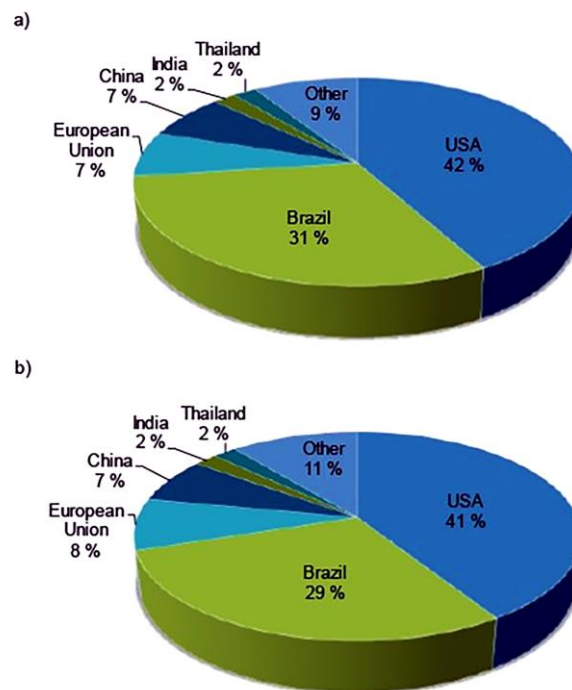


Figure 12. Predicted world's bioethanol production (a) and consumption (b) by 2024, reprinted from [54].

As an antiseptic, ethanol is commonly used in hand sanitizers for its bactericidal and anti-fungal effects. It works by dissolving the lipid bilayer membrane and denaturing the proteins, which is effective against to most bacteria and viruses [59]. It became one of the routine standards of care to deter the presence or growth of bacteria on the skin, thus, becoming a key element of infection control procedures in healthcare. During the outbreak of COVID-19, hand sanitizers were in great demand, such that not only the pharmaceutical companies but also chemical, brewery, and perfumery industries started producing hand sanitizers [60].

In Qatar, there has been a huge demand for ethanol-based disinfectant since March 2020, which was the height of the COVID-19 outbreak. However, supply was limited, and as an example, there was an advertisement looking for ethanol manufacturers to supply up to 1000 tons [61]. The increasing awareness of communities towards personal hygiene to combat viruses, even long after vaccines were issued to the population, caused a tremendous rise in the hand sanitizer market. As of 2020, the global hand sanitizer market is valued at 4 billion US dollars and is estimated to grow at a 3.6% rate between 2021 and 2027. The effectiveness of alcohol-based sanitizer against surface pathogens, as well as the recommendation of the government health agencies in using it to combat the virus, drives the expected compound annual growth rate to 3.8% from 2021 to 2027 [62].

5. Synthesis and Discussion

The study incontestably affirmed that sufficient evidence in both contemporary and seminal peer-reviewed literature corroborates the benefit of bio-converting waste date palm fruits into bioethanol to foster sustainable development in Qatar. As an illustration, sustainable development is the core focus of the three fundamental concepts derived in this study, depicted as the subset of the three circles in the diagram shown in Figure 13. The three circles correspond to the three concepts discussed, which are waste sugar-rich biomass (waste date palm fruits), valorization, and bioethanol. All three main elements in the diagram complementarily affect each other and clearly contribute to the enhancement of the sustainability goal. Sustainability pertains to conserving natural resources for future generations and mitigating environmentally destructive activities, while at the same time fulfilling the present demands of society. It encompasses the three pillars of society: envi-

ronment, economic, and social. In this study, the valorization of waste materials endorses a zero-waste strategy to help conserve the environment, and it employs bioconversion as an environmentally sound technique to produce profitable products that meet society's needs; and bioethanol is a renewable liquid substance that has many domestic and industrial uses that originated from waste sugar-rich biomass.

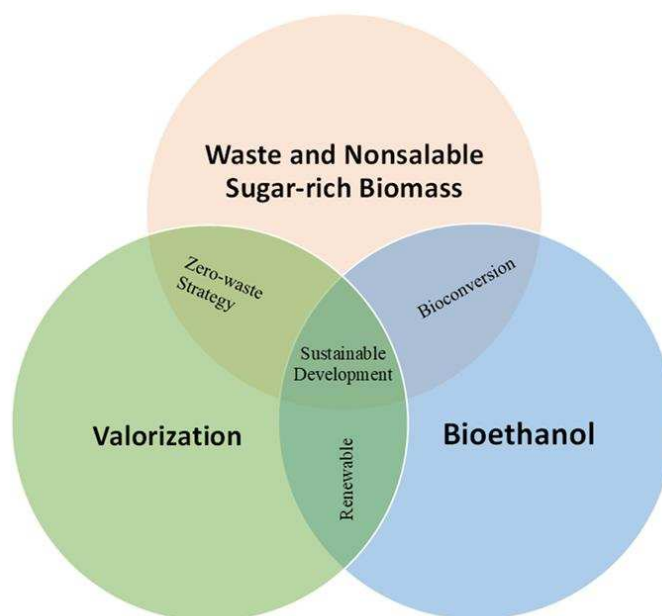


Figure 13. Illustration of fundamental concepts on sustainable development.

The study shows that there is a significant quantity of available waste date fruits coming from the different farm areas of Qatar. Farm date palms are cultivated to produce fruits that are rich in carbohydrates, comprising over 75% (dry weight basis) per ripe fruit. The sugar content is on average approximately 80–90% of the carbohydrate, and the balance makes up the complex carbohydrate or cellulosic fiber. Because of harsh weather conditions, acute water scarcity, and effects attributable to cultural and agronomic management, Qatar on average generates date palm fruit waste of 4505 tons annually. The ethanol that could be derived, considering the amount of sugar is identical with good quality date fruits, might satisfy the pharmaceutical or antiseptic alcohol demand of 1000 tons that was highlighted by Globartis in the year 2020 [61]. However, the fruits coming from ornamental date palms and palms outside the farms that bear nonmarketable date fruits are not included in the statistics and, hence, are yet to be studied. Ornamental date palms, including the ones outside the farms, are normally irrigated with treated wastewater. Although there is no evidence yet of heavy metal presence, it is still subject to further investigation, as over the course of time, heavy metals may accumulate in palms and probably deposit in the fruits. Moreover, cultural and religious beliefs prohibit the human consumption of date fruits coming from date palms irrigated with treated wastewater. Hence, the date fruits produced by these nonfarmed palm trees that are nonsaleable and unfit for human consumption are deemed as wastes.

Another finding that this study upholds is the number of studies conducted in producing bioethanol from date palm fruits. Back in year 2000, Zohri and Etnan [63] developed a fermentation process to manufacture bioethanol from substandard date palm fruits and surplus dates to be used in white vinegar production. The process is applied in one of the leading food companies in Al-Ahsa, Saudi Arabia. The company continues to be a leading vinegar manufacturer in the country in the present. Bioethanol synthesis from sugar sucrose is widely studied and understood and normally employs *Saccharomyces cerevisiae* as the yeast species for fermentation. Several studies worked with developing *Saccharomyces cerevisiae* strains; one of them applying genetic modification for the purpose of enhancing

ethanol production, wherein inhibition initiates at 6% ethanol concentration [31], making yeast material resistant to high alcohol concentration. To some degree, high ethanol concentration during yeast fermentation is toxic to yeast, resulting in the termination of yeast activity. Because of this problem, there are studies that utilize different microorganisms, even bacteria. Examples of these include *Zymomonas mobilis*, *Clostridium acetobutylicum* and *Bacillus subtilis* mixture, *Tannophilus* and *Pseudomonas fluorescens*, *Acetobacter pasteurianus*, *Schizosaccharomyces pombe*, and *Leuconostoc dextranicum* (refer to Section 3.2). Notwithstanding, the conversion efficiency still depends on many process factors such as sugar concentration, nutrient concentration, pH, the inoculum size of the microorganism, temperature, agitation rate, and fermentation time. On the other hand, Taghizadeh-Alisarei et al., 2019 [11] considered the sugar and starch fermentation method as a first-generation process that is a well-established bioethanol production process with industrial applications. The first-generation method is not sufficient to address the agricultural wastes that are complex, such as the cellulosic components of date fruits, date pits, and even plant parts of the date palms. Research studies are now geared towards the second-generation method, which incorporates non-conventional techniques such as very high gravity (VHG) fermentation, simultaneous combined saccharification and fermentation (SSF), semi-simultaneous saccharification and fermentation (SSSF), and separate hydrolysis and fermentation (SHF) (refer to Section 3.3). The second-generation production process deals with breaking down complex carbohydrates to simple sugars for conversion to bioethanol. Thus, the second-generation method is used in combination with the first-generation process for sugar fermentation.

Finally, this paper also reveals that bioethanol has many applications. Bioethanol is simply ethanol derived from the fermentation of sugar-rich biomass. It is the same ethanol as the one produced by the hydration of ethylene. The latter is a fossil derived alcohol, whereas the former is from renewable sources, which is the reason bioethanol is under the spotlight recently due to combustion emission benefits. It is a common belief within the scientific community that the inherent presence of oxygen within its molecular structure facilitates complete combustion, thus, carbon monoxide and unburnt hydrocarbon in the exhaust are reduced. However, as far as NO_x emission is concerned, particularly at high ethanol concentrations in gasohol blends, the results are still inconclusive [52]. On the other hand, ethanol as a commodity chemical is used as a gelation agent for carrageenan industries [64], intermediates for the production of other chemicals such as the dehydration of ethanol to ethylene [65], and as an industrial solvent. Ethanol is also used in cosmetics as a base owing to its good dispersion and penetration effects in skin. In addition, ethanol is well-known today as an antiseptic or disinfecting substance since the outbreak of the COVID-19 pandemic in the year 2020. Its efficacy as a virucide covers a wide spectrum of clinically relevant viruses at high concentrations of ethanol [66]. That said, the demand for ethanol-based hand sanitizers will continue to exist not only in Qatar but worldwide.

6. Conclusions and Future Prospective

Qatar has a long-term goal of conserving its natural resources for the future generations while meeting the needs of current generation. As part of this goal, the country could outweigh the exhaustion of non-renewable resources with the exploration and development of new sources of renewable wealth. One potential that this paper demonstrates is the utilization of waste date fruits as a bioethanol source. Waste sugar-rich biomass, valorization, and bioethanol have complementary influences in enhancing sustainable development. Ethanol is an important commodity chemical for fuel, industry, and pharmaceutical uses. In conclusion, this study has established the abundance of waste date fruits as feedstock material that encompasses both the farm waste fruits and nonsaleable fruits. The study also has identified both the conventional processes, which are mature and applicable in the Qatar context, and the nonconventional processes of bioethanol production. Finally, there is ample demand for ethanol from the society at large, ranging from fuel and industrial chemicals to pharmaceutical alcohol.

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