

Biofuels



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Abstract

Many factors contribute to make biofuel an attractive and possibly essential alternative to fossil fuel. Biofuel is fuel obtained from biomass either by direct burning or by suitable conversion processes. Sources of biomass include energy plants, agricultural processing wastes and residues and municipal wastes. A variety of processes may be applied in the production of biofuels including fermentation, gasification, pyrolysis, micro-emulsification and transesterification. Of available biofuels, bioethanol with its high oxygen content and octane no. is presently the cleanest and may be the only real renewable alternative to gasoline. Biodiesel, on the other hand, has recently attracted considerable attention as an alternative fuel to petroleum-based diesel fuel.

Keywords: Biofuel; Bioethanol; Biodiesel; Fermentation; Gasification; Pyrolysis

Introduction

During the last fifty years or so, and almost within living memory, man has witnessed an almost imperceptible change in the energy situation from surplus and cheap availability to scarcity. Renewable sources of energy have to be found in order to meet the continuously rising demand and decreasing energy reserves. Waste products and other low quality sources of energy and bottom of the barrel residues may no longer be discarded [1-3].

Bioenergy

This is energy obtained from biomass. This can be done by direct burning of wood and other energy plants, or by suitable processes by which the biomass is converted into biofuels (bioethanol, biomethanol and biodiesel) [4].

Imperatives for Bioenergy

Many factors contribute to make biofuel an attractive and possibly essential alternative to fossil fuel. The more important of these factors include:

- The increasing cost and diminishing reserves of petroleum and other fossil fuels. Biofuel on the other hand is renewable.
- The environmental consequences and rising levels of so-called greenhouse gases such as carbon dioxide and other exhaust gases in the atmosphere resulting from the combustion of fossil fuels; the combustion of biofuel on the other hand does not increase the net atmospheric levels of

carbon dioxide as it is a plant and not petroleum-derived fuel. Biodiesel in particular is a non-toxic biodegradable fuel and its combustion leads in general to reduced levels of carbon monoxide, sulphur oxides, hydrocarbons, soot and particulate matter. There may be a slight increase in NO_x emissions, but these can be controlled by delaying the injection timing in engines. Under certain conditions a biodiesel fuel with good ignitability can give lower levels of nitrogen oxides [5].

- The continuing development of processes for the production of bioethanols and biodiesel fuels that can substitute for petroleum-based fuels.
- The rapid development of sugarcane- and corn-based biorefineries producing bioethanols.
- The continuing development of biodiesel fuels produced from plant oils [6-8].

Fuels derived from grains or oilseeds will have an impact on food prices. Even at the relatively modest 2008 levels, ethanol production has caused a significant effect in the agricultural commodity markets, with corn roughly doubling in price from only a few years ago, and food and feed prices up significantly (i.e., milk prices are up 29% in 2007 alone). Spot market wheat prices, which remained under \$4/bushel for years, have risen to beyond \$18/bushel. Ramping up to a supply of 6% or more of U.S. transportation fuel using corn grain as the primary feedstock, while technically feasible, could even more dramatically affect the "food/feed versus fuel" dynamic [9,10].

On the other hand, low-cost biofuels will help keep food prices low by reducing transportation costs. For example, the cost of corn affects the price of a few items in the store, but the cost of fuels to move food products around affects the price of literally every food product. Furthermore, biofuels backers believe that cellulosic ethanol is going to end up reducing food prices because it will reduce animal feed costs and simultaneously help keep transportation costs down [11].

Direct Burning

Wood is the biggest, and most economic, source of biomass, but trees may take years to reach their full height. Interest in developing biomass as a credible and commercial energy source has focussed on selecting fast growing species that burn efficiently. There are many such species with significant potential, for example Eucalyptus and shrubs such as Euphorbia. *Leucaena leucocephala*, a plant native to Mexico, is now being grown in India and is already supplying local people with energy only three years after being planted. There is even a petroleum nut in the Philippines that burns brightly when it is ignited [12].

Biofuels

A higher market value is placed on liquid transportation fuels, mainly diesel fuel and gasoline, above other energy forms, whereby liquids are preferred over gases (natural gas) or even more over solids (coal). A modern economy cannot run without liquid fuels. Although renewably produced electricity may be increasingly a future transportation fuel option for some countries, the still cheaper internal combustion engine will be the mainstay of transportation in most countries of the world for a very long time. As Vinod Khosla, a highly successful venture capitalist and biofuels backer, points out, electric vehicles are "just toys" as far as China, India, and the developing world are concerned. Ninety seven percent of the world's transportation fuels are provided by petroleum. The world is still totally dependent on petroleum for mobility [13].

Biofuels are liquid fuels whose importance and relevance is in large measure due to the fact that they are essentially the only alternatives to petroleum-derived liquid transportation fuels. Biofuels provide potential large-scale economic and environmental advantages. They can be widely produced, some of them in large volumes, and are potentially renewable and carbon neutral. Biofuels may be produced from plants that are particularly adapted to producing hydrocarbons, but for many applications plants need not be grown specially for fuel production. Alternatively, agricultural and industrial wastes and crop residues may be used for the production of biofuels [14].

Biorefineries

A biorefinery is defined by the National Renewable Energy Laboratory [2], as a facility that integrates biomass conversion processes and equipment to produce fuels and chemicals from biomass, analogous to today's petroleum refineries. In addition

to fuels, lubricants, chemical solvents, intermediates, and polymer products a biorefinery will produce other bioproducts, such as soil amendments, food and feed components, fibres, biodegradable plastics, building materials, and various nutraceutical products [15].

The conversion of biomass to produce potential liquid fuels may be done by a variety of processes. The optimal biorefinery may not consist of only one type of conversion, such as fermentation, but may rely on a fully integrated system that employs fermentation, gasification, pyrolysis and even digestion. Gasification is high temperature conversion in the presence of limited oxygen. This produces a synthesis gas from biomass feedstocks consisting largely of CO and hydrogen. It can be used directly as a fuel for heat recovery or can be further processed by Fischer-Tropsch synthesis to methanol, dimethyl ether, mixed alcohols, and/or a mixture of hydrocarbons. The key is in the choice of catalysts that carry out the re-formation reactions, as well as the choice of feedstocks [16].

Pyrolysis refers to high-temperature heating in the absence of air or oxygen. It produces a pyrolysis oil, a complex mixture of components that is generally a low-quality fuel in itself, but one that can be upgraded by hydrocracking or catalytic cracking. Pyrolysis oils from defined biomass sources are convertible to liquid fuels as well as commodity chemicals [17].

Sources of Biomass

Energy plants

There are many plants that are particularly adapted to producing hydrocarbons, not unlike petroleum hydrocarbons. Examples of such plants include:

- a) Nonwaxy plants such as rice which have measureable quantities of cuticular hydrocarbons [4].
- b) Many desert plants that are hydrocarbon rich and which may afford clues to new types of "energy crops".
- c) Algae where the possibility of obtaining useful hydrocarbons [7] is especially intriguing, considering the ability of algae to assimilate CO₂.
- d) Grasses such as the native American prairie grass switchgrass.
- e) Woody materials such as willow and poplar.
- f) Terpenes are another potential source of tomorrow's fuels and chemical feedstocks. Turpentine, largely R-pinene, derived from the oleoresin exudate of tapped pine trees, has found many uses. *Pinus elliottii* averaged over 7kg of oleoresin per tree per year under one treatment regimen [5]. Hodges and Johnson [6] reported an alternate method of tapping *P. elliottii* using bore hole drilling. The gum rosin and turpentine have a variety of uses, and the semivolatiles fraction has potential as a liquid biofuel.

g) The guayule shrub, which is grown in marginal desert land in the American southwest, contains approximately 15% dry weight of rubber and 10% resin and produces 48% bagasse [8]. As the cost of producing the bagasse is borne by the value of the latex, the cost of the bagasse as a feedstock for biofuel is essentially zero. In addition, the bagasse is aggregated at the mill where latex extraction is carried out, so, in fact, there is a cost to not utilizing the bagasse. Feasibility studies indicate that up to 1000 gallons of ethanol per acre could be produced from this bagasse, which represents a significant additional value after profiting from the rubber latex and other potentially profitable coproducts from processing, such as termite inhibitors [9].

h) Miscanthus, which is a plant that can accumulate up to 20 tons of dry matter per acre per year.

Plants contain three primary types of carbon-based polymers, cellulose, hemicellulose, and lignin. About 70% of the total mass of plant material is sugars locked up in cellulose and hemicellulose, as five- and six-carbon sugars [12]. Cellulose, a glucose-based polymer consisting of two β -1,4 linked glucose residues (cellobiose) in their repeating units can be hydrolyzed to monomeric glucose. The microfibril scaffold of the cell wall consists of both crystalline and amorphous domains within a largely cellulosic core- a structure that limits methods for hydrolysis.

Hemicellulose, a branched polymer of predominately xylose and related pentoses, may be the "weak link" in cellulosic conversion technology. It is not readily converted to biofuels by traditional microbes in industrial-scale fermentation. The optimal use of hemicellulose-derived (C-5) sugars is important to the success of cellulosic biorefinery operations, which will depend on cofermentation of both C-5 and C-6 sugars made possible by novel microbes that metabolize both sources. Alternatively, biorefineries can be designed to isolate the slower-fermenting sugars and utilize them in other platforms, providing a broad range of products from the biorefinery (just as in petroleum refining). Any use of xylose to make either biofuels or platform chemicals. Improved yeasts and bacteria allow for utilization of hemicellulose-derived sugars, but hemicelluloses conversion is not without cost. The longer fermentation times resulting from use of xylose sugars incur significantly greater capital costs (larger fermentors). Hemicelluloses are cross-linked via ester linkages to predominately cinnamic (ferulic) acids, forming covalent linkages with lignin, making plant cell wall hydrolysis even more difficult.

Lignin, a highly branched aromatic polymer of largely guaiacyl and syringyl monomers with connecting three-carbon aliphatic sidechains, provides structural rigidity to cell wall architecture and increases its hydrophobicity-factors that, along with cellulose crystallinity, impede the breakdown of cell walls for facile conversion to ethanol. Historically seen as a waste product, lignin will likely become a valuable source of renewable energy to the biorefinery, minimizing the need to purchase

external fuel. The future may see development of other significant uses for lignin, especially as chemists make improvements in gasification, syngas reformation (i.e., Fischer-Tropsch catalysis), new plastics and adhesives, and new chemical platforms derived by utilizing lignin's phenolic-rich composition. Under suitable conditions, micro-organisms are capable of decomposing lignin as found in lignified plant materials (cornstalks, oat hulls, corn cobs and wheat straw), the rate of decomposition being as great as that of cellulose [18-20].

Agricultural processing wastes and residues

Examples of agricultural processing wastes include oat, walnut, almond, or peanut hulls. Crop residues include grain or grass straws such as wheat straw, rice straw, corn stover (the residue left behind after the corn grain has been harvested), etc. Molasses from sugar production, rice husks, cheese waste and citrus fruit skins have all been used for ethanol production. Straw and other farm residues consist predominantly (60 per cent or more) of celluloses and hemi-celluloses [21-25].

Deswarte et al. [3] isolated wax from wheat straw using supercritical CO₂ extraction. C28-31 alkanes and various sterols were predominant isolates, along with fatty acids, alcohols, and lipids. Collectively the longer chain hydrocarbons comprised 1% or more by weight in wheat straw and similar biomasses. Plant leaf waxes consist largely of C-16 and higher straight- and branched-chain alkanes. These form the cuticular waxes that play a significant role in water conservation in plants [26-28].

Municipal solid waste

Methane gas is produced by microbial activity in rubbish tips. Municipal solid waste is already converted to methane and electricity via advanced digesters and incineration systems, respectively. Cellulosics isolated from municipal solid waste are also available now to provide ethanol [29-33].

Bioethanol

Bioethanol with its high oxygen content and octane no. is presently the cleanest and may be the only real renewable alternative to gasoline. In 2005, the United States passed Brazil and became the world's number one ethanol producer. There is evidence to indicate that the use of ethanol in cars will lead to enhanced engine performance and reduced emission of CO. Only minor modifications to engines are needed for cars to run on fuel blends of gasoline with up to 30% ethanol. New cars can also be made with engines designed to run on pure ethanol [34]. Large-scale use of bioethanol will require conversion of sugary crops or cellulose-based feedstocks (Cellulosic ethanol).

Bioethanol may be produced from sugary and starchy crops such as sugar beet, potatoes, corn, cassava and Jerusalem artichokes. Present technology relies largely on the fermentation of sugars or starch to ethanol using fermentation technology that has been known for many years. The cost of enzymes for hydrolysis has been reduced considerably, and other processing costs continue to improve as market economics and competition

between producers evolves. This represents a maturing of pre-existing technologies rather than fundamental new breakthroughs in the underlying science.

For cellulosic biofuels the processing steps can be broadly outlined as pretreatment (to open up the cell wall materials for enzymatic attack), enzymatic hydrolysis (to convert the cell walls to sugars), and fermentation (to convert the sugars to ethanol or other biofuels). In the ammonia fibre expansion (AFEX) pretreatment process [13], biomass plant material is cooked with hot concentrated ammonia under pressure. When the pressure is released, the ammonia evaporates and is recovered, and the treated biomass is then more available for conversion to sugars. Ammonia treatment tends to minimize sugar degradation that occurs in acid pretreatments. About 98-99% of the ammonia is recovered; the remainder can serve as a nitrogen source downstream for fermentation organisms. In the consolidated bioprocessing process enzyme production and fermentation are combined in a single organism.

Wood alcohol (methanol)

Alternative platform chemicals and biofuels from cellulose, such as butanol and methanol, are advantageous in several ways.

Table 1: Comparison of properties of biodiesel and diesel fuel.

Plant oil	Density g/cm ³	Viscosity (37.8 °C) mm ² /s	Density g/cm ³	Flash Point (°C)	Nett Calorific value 106×kJ/m ³	Nett Calorific value 106×kJ/m ³
Peanut	0.883	4.9	54	176	33.6	19
Soybean	0.880-0.885	4.5	45-56	178	32.7-33.5	19,20
Babassu	0.879	3.6	63	127	31.8	19
Palm	0.872-0.880	5.7	62-70	164	32.4-33.5	19,20
Sunflower	0.86	4.6	49	183	33.5	19
Rapeseed	0.882	4.2 (40 °C)	51-60	-	32.8	20
Diesel fuel	0.830-0.840	3.5 (40 °C)	51	-	35.5	20

Botanical research suggests that there might be over 300 different plant species that can give diesel substitutes. The cultivation of many of these plants may not be easy. For instance, the castor oil plant is no more than a minor crop because it gives such poor yields and the seeds of the plant contain not just castor oil, with its precious ricinoleic acid, but ricin, one of the world's most potent toxins.

Plant oils are largely esters of glycerol and fatty acids. Plant oils or oil blends may be used as alternative diesel fuels as has been suggested by many workers. However, the direct use of plant oils is generally considered to be unsatisfactory and impractical for both direct-injection and indirect type diesel engines. This is related not only to the polyunsaturated character of the triglycerides and the acid composition and free fatty acid content of such oils and their high viscosity and low volatility, but also to various other problems associated with their use such as carbon deposits, lubricating oil thickening and gum formation due to the oxidation and polymerization of the oils during storage and combustion [14]. Consequently derivatives

These and other products of biomass conversion capture, contain, and recycle the excess carbon dioxide and methane effluents that contribute to global warming. Methanol is an alternative liquid transportation fuel and potentially a hydrogen source for fuel cells. Although methanol from wood is technically feasible, methanol from coal seems to be the favoured current source, certainly in coal-rich countries such as China where expanded coal-to-methanol production facilities are under development. Efficient conversion of CO₂ to methanol by hydrogenation or electrochemical conversion represents a research challenge.

Biodiesel

Biodiesel is fatty acid methyl esters produced from plant oils. This has recently attracted considerable attention as an alternative fuel. Compared to petroleum-based diesel fuel, it has similar viscosity, higher cetane number and flash point but slightly lower calorific value (Table 1). Rudolf Diesel, the inventor of the Diesel engine, used peanut oil to fuel one of his engines at the Paris Exhibition of 1910. In 1912 he wrote: "The use of vegetable oils for engine fuels may seem insignificant today. But such oils may become in the course of time as important as petroleum and the coal tar products of the present time".

that approximate the properties of hydrocarbon-based diesel fuels need to be developed from plant oils before they can be considered as viable diesel fuel alternatives.

Several processes have been developed for the utilization of plant oils and oil wastes for the production of biodiesel fuel including pyrolysis, micro-emulsification and transesterification. Pyrolysis refers to high-temperature heating in the absence of air or oxygen. Although pyrolysis of plant oils gives in general products that are chemically similar to petroleum derived diesel fuel, some of the properties of these compounds may not be acceptable such as pour point and carbon residue and ash contents. In addition, the removal of oxygen during thermal processing eliminates the environmental benefits of using an oxygenated fuel [16].

The pyrolysis of triglycerides with the aim of obtaining products suitable for diesel engines was investigated by a number of workers. Thermal decomposition of triglycerides produces compounds of several classes including alkanes,

alkenes, alkadienes, aromatics and carboxylic acids. The nature and relative amounts of such compounds depend largely on the type of vegetable oil used, as different vegetable oils give on pyrolysis widely different compounds. Pyrolysis of soybean oil for instance gives a lower viscosity and higher cetane number oil containing 79% carbon and 12% hydrogen [15].

Microemulsions are isotropic, clear or translucent thermodynamically stable dispersions of oils, water, a surfactant and often a small amphiphilic molecule, called a cosurfactant. Ziejewski et al. [18] prepared a non-ionic emulsion of 53.3% alkali-refined and winterized sunflower oil, 13.3% 190-proof ethanol and 33.4% 1-butanol. This emulsion had a viscosity of $6.31 \times 10^{-6} \text{ m}^2/\text{s}$ at 40 °C, a cetane number of 25, a sulphur content of 0.01%, free fatty acids of 0.01% and ash content of less than 0.01% [18]. Lower viscosities and better spray patterns were obtained by increasing the amount of 1-butanol. Schwab et. al. found that 2-octanol is an effective amphiphile in the micellar solubilisation of methanol in triolein and soybean oil [17].

Biodiesel may be derived from plant oils by the transesterification of triglycerides to their corresponding methyl esters. The resulting biofuel has a lower viscosity and better physical properties and its engine performance is improved. Transesterification, also called alcoholysis, is the displacement of alcohol from an ester by another alcohol in a process similar to hydrolysis, except that an alcohol is employed instead of water. Suitable alcohols include methanol, ethanol, propanol, butanol and amyl alcohol. Methanol and ethanol are used frequently, but transesterification with methanol, i.e. methanolysis, is most often preferred because of lower cost and other, physical and chemical, advantages.

The transesterification reaction may be represented by the equation. It consists of three consecutive, reversible reactions, where three moles of alcohol per mole of triglyceride are required to produce 3 moles of fatty methyl esters and one mole of glycerol. The production of glycerol as a by-product (roughly 1kg of impure glycerol is produced for every 10kg of biodiesel) is a vexing issue with transesterification. The potential oversupply of glycerol from expanded biodiesel production represents a short-term hurdle but it may represent on the long run an opportunity for chemists and chemical engineers in developing new glycerol-based products. Purified glycerol has value in cosmetics and paints. It can also be converted to methanol, ethylene, and propylene glycol, solvents, and many other products using existing, but not necessarily commercially viable, processes. Glycerol derivatives may be applied as plasticizers in many polymer formulations or more directly as fuel additives. Glycerol can also be a substrate for enzymatic conversion or fermentation routes to fuels such as methane [10] and many of the fermentation products mentioned above. Specifically, if glycerol were to become as inexpensive as glucose as a fermentation feedstock, one could envision metabolic pathways for fermentation conversions that are more efficient than sugar conversion, with fewer moles of CO₂ effluent.

Transesterification reactions are usually catalyzed by acid or alkali catalysts. Transesterification using alkali catalysts gives high levels of conversion of triglycerides to their corresponding methyl esters in short reaction times. Alkali-catalyzed transesterification proceeds approximately 4000 times faster than that catalyzed by the same amount of an acidic catalyst, and is thus most often used commercially [28]. Alkalis used for transesterification include NaOH, KOH, carbonates and alkoxides such as sodium methoxide, sodium ethoxide, sodium propoxide and sodium butoxide.

For alkali-catalyzed transesterification, the glycerides and alcohol must be substantially anhydrous because water causes a partial reaction change to saponification, which produces soap [29,31,32]. The soap consumes the catalyst and reduces the catalytic efficiency, as well as causing an increase in viscosity, the formation of gels and difficulty in achieving separation of glycerol. According to Freedman et al., sodium hydroxide or sodium methoxide may react with moisture and carbon dioxide in air thereby diminishing their effectiveness and reducing the ester yields [32]. The free fatty acid content of the refined oil should be also as low as possible [30,31]. Ma et al. [30] suggested that the free fatty acid content should be below 0.5% [30]. Higher molar ratios of alcohol to vegetable oil result in greater ester conversion in a shorter time. Freedman et al. [32] found that in the transesterification of different vegetable oils such as soybean, sunflower, peanut and cotton seed oils, increasing the molar ratio from 1:1 to 6:1 increased significantly ester conversion, with the highest conversion being achieved at a 6:1 molar ratio. Similar results were also reported by Feuge and Grose in the transesterification of peanut oil with ethanol, where the molar ratio was increased from 3:1 to 6:1 [31] and by Krisnangkura and Simamaharnnop in the transesterification of palm oil with methanol where sodium methoxide was used as a catalyst [33,34]. Transesterification by acid catalysts is in general much slower than that by alkali catalysts. On the other hand, acid-catalyzed transesterification can be more suitable for glycerides that have relatively high free fatty acid contents and more water. In some cases where a lower grade oil such as sulphur olive oil is used, acid-catalyzed transesterification may be necessary as reported by Aksoy et. al [23].

Acids used for transesterification include sulphuric, phosphoric, hydrochloric and organic sulphonic acids. Acid-catalyzed transesterification may be carried out in a conventional reaction or in situ. In the conventional reaction the acidified alcohol reacts with the purified oil extracted from the oil-bearing material, whereas the oil-bearing material contacts the alcohol directly in the in situ transesterification and extraction and transesterification proceed within the same process and alcohol acts both as an extracting solvent and transesterification reagent. In situ transesterification with good results was reported by a number of workers using different oils such as soybean oil [26], rice bran oil [27] and sunflower oil. In situ transesterification of sunflower oil with acidified methanol produced fatty acid methyl esters

in yields significantly greater than yields obtained with the conventional reaction [24,25].

Enzymatic transesterification using lipase is also an attractive transesterification process for biodiesel fuel production, where the glycerol produced as a by-product can easily be recovered and the fatty methyl esters easily purified. In the transesterification of triglycerides with methanol using *Rhizopus oryzae* lipase as catalyst, the triglycerides and partial glycerides are first hydrolyzed by lipase to partial glycerides and free fatty acids, respectively, after which methyl esters are synthesized from free fatty acids and methanol [22]. This reaction mechanism suggests that in contrast to alkali-catalyzed methanolysis, free fatty acids contained in used oils can be easily converted to methyl esters.

The main hurdle to the commercialization of enzymatic transesterification is the cost of lipase production. As a means of reducing the cost, the use of whole cell biocatalysts immobilized within biomass support particles has been suggested as immobilization can be achieved spontaneously during batch cultivation, and in addition, no purification is necessary. The lipase production cost can be further lowered using genetic engineering technology, such as by developing lipases with high levels of expression and/or stability towards methanol.

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