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Sustainable Production of Biofuels Using Intensified Processes

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To my family and all the people who have supported me unconditionally along the way to achieve my goals.
—Juan Gabriel Segovia-Hernández

To my beloved wife and parents
—Eduardo Sanchez-Ramirez

To my family
—Juan José Quiroz-Ramirez

To my beloved parents and brother, who have always been my unconditional support. You are my greatest inspiration to continue working to achieve my goals. Thank you for always being by my side
—Ana Gabriela Romero-Garcia

To my family and friends. Especially to my wife for supporting me to achieve my goals.
—Heriberto Alcocer-Garcia

Preface

Biofuel may refer to any form of fuel derived from biomass, and accordingly, its application can be in household energy, for electricity generation, or in the transport sector. The term biofuel in this book specifically refers to those biomass-derived fuels that can be used in the transport sector such as bioethanol, biodiesel, biobutanol, among others. Currently, bioethanol and biodiesel account for more than 90% of global biofuel use. The European Union is promoting the use of biofuels, primarily due to the savings of greenhouse gas emissions that biofuels can potentially offer. Biofuels can diversify the offer of transport fuel and are a way to raise energy self-sufficiency, diversify the production sites, and strengthen the internal agriculture of a country. Lastly, they are suitable, in many cases, for being used in current power trains and fuel infrastructures. Biomass is an attractive energy source for several reasons. First, it is renewable as long as it is properly managed, and second, it is also more evenly distributed over the earth's surface than finite energy sources and may be exploited using more environmentally friendly technologies. Biomass provides the opportunity for increased local, regional, and national energy self-sufficiency across the globe. The energy in biomass can be accessed by turning the raw materials, or feedstocks, into a usable form. Transportation fuels are made from biomass through biochemical or thermochemical processes.

Therefore, for the production of biofuels to be sustainable and economically competitive with an oil refinery, it must present low energy consumption, low operating costs, low environmental impacts, inherent safety, and good dynamic behavior in the design and operation of all equipment. For this reason, it is essential to implement strategies that aim to achieve these key objectives in the design of a biorefinery that produces liquid biofuels. Increasing awareness for energy sustainability, environmental concerns, new and unconventional feedstocks, as well as recent advances in process optimization has sparked a renewed interest in process intensification (PI). PI aims to drastically reduce the energy consumption and processing cost of the chemical processes by utilizing the synergy between multifunctional phenomena at different time and spatial scales and enhancing the mass, heat, and momentum transfer rates. There has been significant growth in the field of PI over the past decades that featured both successful industrial applications and increased research interest in academia.

This book highlights the importance of process intensification in the production of biofuels and discusses the required interdisciplinary approach to accomplish it. Authors outline intensified processes and current challenges in the production of biofuels at different levels. This book presents an overview of important ideas addressed within methodologies proposed for designing intensified processes.

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Chapter 1
Biofuels: Historical Development and Their Role in Today's Society

Abstract Have you ever imagined what life would be like without energy sources? What would the growth of civilizations have been like without fuel? How would we live today if we did not have technological developments in energy? This and other questions may arise when thinking about the development of fuels to generate energy. Currently talking about fuels is a very daily topic, about which we do not ask ourselves its origin and the evolution that these have had in history. Thought history, the human being has searched for ways of putting energy to work for them. Due to growing population, human has been looking for faster, easier, and more efficient ways to produce energy. However, for years the human being has made excessive use of existing resources, causing them to be depleted at present. Likewise, the pollution generated using these resources is also causing considerable damage to the environment at a global level. For these reasons, there is a growth in the use of alternative energy sources to reduce pollution and meet their energy needs. In this chapter will describe the origin and historical evolution of biofuels. The social, political and environmental context that originated the need for their scientific and technological development. The biofuels with the greatest demand and potential to replace fossil fuels in the medium term will be briefly described.

1.1 Historical Development of Biofuels

The history of biofuels is as old as the origin of human civilizations. Biofuels refer to plant biomass and the refined products to be combusted for energy (heat and light). Similar to fossil fuels, biofuels exist in solid, liquid, and gaseous forms [1]. Throughout history, technologies have been used and developed for the best use of biofuels, whether solid, liquid, or gaseous. Figure 1.1 shows the important stages in the use of Biofuels throughout history.

The first biofuel used was the wood and other plant material, it is not clear when human being started the controlled use of fired for heat and light. Nevertheless, archaeologists identified charred animal bones and stone tools in wood ash in Wonderwerk Cave of Kuruman Hills in South Africa, providing evidence of controlled fire

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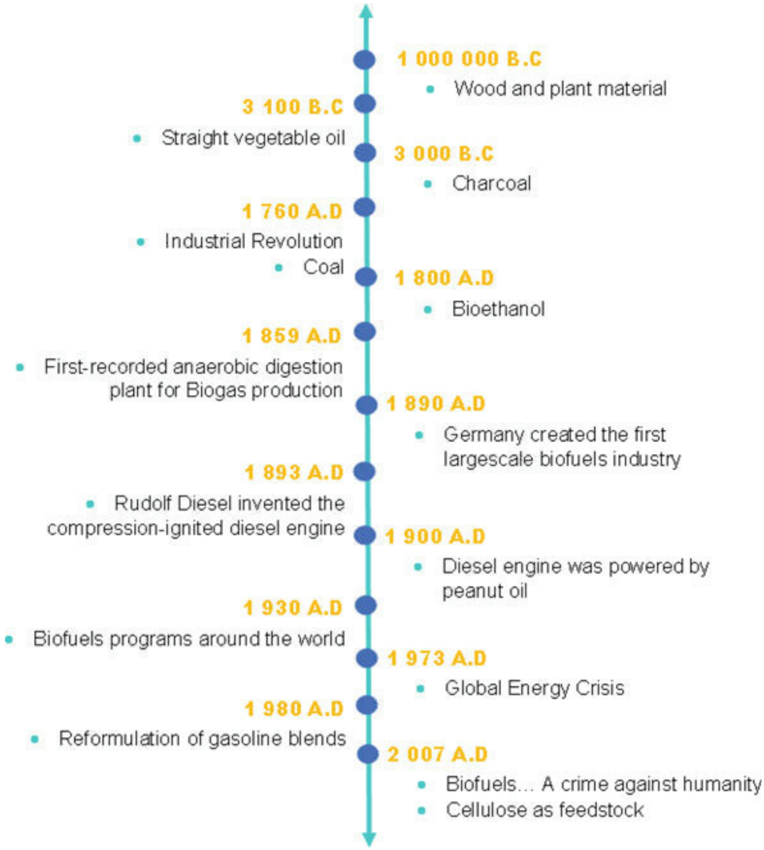

Fig. 1.1 Important stages in the use of biofuels throughout history

use by prehistoric "mankind" creatures one million years ago [2]. At first, wood was burned for warming, light, and cooking. Then the heat from fired was used to transform materials or change its form to make them more useful, this is the case of metals. Before nineteenth century, the wood continues being used as the primary fuel for cooking and heating.

The first liquid biofuel used of which there is a record was the use of vegetable oil. This was used as fuel for ceramic lamps in ancient Egypt. Emphasizing the use of olive, castor, and rapeseed oils. Vegetable oil and animal fats were widely used in oil lamps for lighting before the invention of gas lights and electric lights in late eighteenth century, but for making biodiesel it did not occur until 1930s [3].

Back to 3000 B.C., human beings started to use charcoal (which is formed by the slow pyrolysis of wood) in metallurgy to smelt ores for copper and iron. Charcoal was the designated governmental fuel for cooking and heating in China's Tang Dynasty in 700 A.D. During 1931–1960 in China and the World War II in Europe when gasoline was scarce, many automobiles were powered by wood gas, a mixture of carbon monoxide and hydrogen generated by partially burning charcoal in a gasifier [1]. Nowadays, charcoal is still a valuable product used for cooking as a barbeque briquette. As well it is used for heating, air and water purification, art drawing, and steel-making.

As human population increased over the time, the dependence on fuels increased as well. During this transition, fossil fuels played a significant role in society and for the development of new technologies. Coal had been used since the second millennium B.C., but its potential used had not been discovered by that time. Around 1760 A.D. it was found that coal can be used as a powerful source of energy, and with this began a new stage in the history, the Industrial Revolution. The industrial revolution marked the beginning of a new era in the use and exploitation of fuels. During the initial stages of industrial revolution, biofuels powered the first lamps and internal combustion engines. The shift from biofuels to petroleum products like kerosene and gasoline as the primary fuel source took place in the 1860s for oil lamps and in the twentieth century for automotive fuels [4]. Coal continued to be used in great quantities until the twentieth century.

In the 1800s, biofuels were available primarily for lighting purposes. For those times, another biofuel that gained strength at the beginning of the nineteenth century was bioethanol. Although the production of alcohol by fermentation dates to the beginning of civilizations before Christ, alcohol was used only as a drink and not for fuel. By 1860, thousands of distilleries produced tens of millions of gallons of alcohol per year for lighting in the USA and Europe [5]. Years later the United States government imposed a tax per gallon of alcohol. This meant an increase in the cost of alcohol as fuel, so that the use of biofuels was overshadowed due to high costs, causing significant growth in the petroleum industry.

Backing on time, biogas was used since the beginning on the tenth century B.C. to prepare warm bath water. In the beginning of 1800, it was discovered that flammable gas from cattle manure ponds was methane. Years later, around 1859, it was constructed the first recorded anaerobic digestion plant for biogas production in India [6]. Nevertheless, different studies were carried out to study anaerobic digestion as a science and select best anaerobic bacteria and digestion conditions for promoting methane production.

Meanwhile in Europe, there was not competition between the use of bioethanol and petroleum as a fuel due to there were no taxes for the ethanol industry. Some countries like Germany and France, wanted to find alternatives to fossil fuels. Before World War I, Germany created the first largescale biofuels industry in the world, by using potatoes to produce bioethanol. Studies in biofuels continued demonstrating that bioethanol could be produced from any vegetable matter that could be fermented.

During the nineteenth century there developed some forms of internal combustion engine. In 1893 the German engineer Rudolf Diesel invented the compression-ignited diesel engine. He conducted several tests with vegetable oil fuels, with the aim that farmers could produce their own fuel. Based on this idea, in 1900 the French company Otto demonstrated a diesel engine powered by peanut oil [1]. However, attention to the development of vegetable oil-based fuels has been overshadowed by the high availability and low price of gasoline and diesel.

Around 1890 till 1914, some countries governments such as Germany, France and England were worried about the longevity of oil reserves and the unpredictable nature of oil supplies [7]. Due to this uncertainty, in Europe and the United States the farmers were encouraged to produce alcohol once again in Europe and US congress lifted alcohol tax in 1906. Moreover, at the beginning of the twentieth century, engineers were evaluating the advantages of ethanol against gasoline. As a result, it was found that ethanol could be used in both high and low compression engines, while gasoline could be used just in low compression engine. Between 1907 and 1909, the US Department of Agriculture conducted more than 2000 engine and fuel tests concluding that "a gallon of alcohol will develop substantially the same power in an internal combustion engine as a gallon of gasoline, due to superior operating efficiency" [4]. This efficiency was later denominated as the "octane" rating.

Meanwhile, between the years of 1900 and 1930 more than 30 nations began with a "biofuels program" by promoting the use of biofuels and push the growth of agriculture. Tropical countries like Brazil and Philippines developed new markets for sugar cane. Among with the successful growing of biofuel application, the first generation biorefineries occurred in Brazil [8]. Therefore, the number of distilleries in Brazil producing fuel-grade ethanol increased. Meanwhile in Cuba, it was produced ethanol as an additive for gasoline.

The World War II brings innovation in biofuels, German fuel production was derived from non-petroleum sources, while the allies had plenty of oil for the war effort [9]. By that time, the use of alcohol as fuel was a question of necessity, leaving behind the question of costs and efficiency. There was reported the use of different kinds of vegetables oils such as: nuts, tea leaves, cotton and cabbage seed. In China and India, where food was scare, it began to be used alcohol blends. As the war was ended, cheap imported oil was once more readily available and alcohol blends were marketed sporadically. Around those years, different nations wanted to push forward the biofuels production. By 1952 this idea was abandoned due to the increasing availability of cheap oil from the Middle East [4]. Therefore, most of the alternative biofuel programs around the world had been abandoned as far too costly in comparison.

After World War II, oil became in the main fuel globally. For more than twenty-five five years the world was totally dependent on oil from the Middle East. This situation changed in 1973, due to the war between countries in the Middle East. Generating an energy crisis at international level, giving the opportunity to look again at biofuels as an alternative to the use of oil as a fuel. Energy crisis spurred a widespread search for alternative energy sources. Around the same time, Brazilian bioethanol production from sugarcane began to increase. While in the USA, bioethanol production from corn for blending with gasoline gain interest for research.

During the 80's most unleaded gasoline in the USA, was made using a petroleum refining process called "severe reforming", which boosted the levels of benzene, toluene and xylene compounds from 25% to as high as 40% of the fuel [10]. High levels of these carcinogenic compounds in fuels worried environmental policy markets. When these aromatic chemicals were burned in car engines, they not only emitted carcinogens and toxics into the air but highly volatile, photo-reactive chemicals that contributed to urban smog as well. By 1990, the USA Government created the Clean Air Act and empower the Environmental Protection Agency in order to find alternatives to set the increasing environmental problem. The cleaner alternative was that fuels would contain a class of octane boosting compounds such as methanol, ethanol and natural gas [4]. Which contain lower concentrations of aromatic compounds when burned, and reformulated gasoline blends. These "oxygenates" compounds raised gasoline's oxygen content, promoted more thorough combustion of fuel, reduced pollution and at the same time raised octane.

After the growing development of the biofuel program in the USA, where they used corn to produce biofuels, the question began to be stronger: Is it correct to use food crops for the production of fuels? It was till 2007, that Jean Zigler a special rapporteur for food rights, accuses biofuels production from food crops as a "crime against humanity" [11, 12]. The fuel or food issue, becomes a complex situation worldwide arising the idea that the technologies advances should be capable to use agricultural waste to produce fuels. In this way, the goal was to put food and human needs first, local development including fuels second and fuel exports third. For mor than a century, researchers has been concerned about how to evite scarcity of food and develop biofuels from non-food materials. Around the early twentieth century, information about the use of cellulose as a feedstock for biofuels production began to increase. As the years went by, the biofuels potential of cellulose, the most abundant organic material on earth, was a recurrent theme in scientific literature [4]. Nowadays, due to the impact of biofuels on climate change, food rights and sustainability. There is the necessity to develop optimal and sustainable systems that can be capable to produce remarkable results.

1.2 Biofuels: A Key Component for Society Development

Through history, it is possible to understand the importance that biofuels have had from the development of the first civilizations to the present. Research into biofuels has progressed considerably in the last century, focusing in the fields of agriculture, transport and chemical engineering. Nowadays, due to the increasing concerns about global climate change, energy security, high oil prices, and declining oil reserves it has been prompted a shift toward biofuels as a feasible and renewable alternative to fossil fuels. The history of development and research in the area of biofuels has always been affected by political and economic issues. Such factors continue to influence biofuel decisions worldwide.

Currently, despite considering the use of biofuels as an alternative to solve the environmental problems mentioned above. It is important to consider the environmental, economic, political, and social implications of using these. Consequently, the government of a country faces a difficult *trilemma*: how to stimulate the growth of the biofuel industry while protecting food security and preserving environmental sustainability simultaneously [13]. To understand the situation, it can be said that the direct competition effect, will lead the scarcity of certain food crops that are the feedstock for biofuel production. This situation will push the prices up for these inputs and therefore the increase of both food and energy markets. The increment in cost production will applied not only for those that are direct inputs but also for those that are close substitutes. The indirect competition effect further exacerbates increased food prices due to the reallocation of land from food to biofuels: as land becomes scarce to produce agricultural commodities, it leads to a lowered supply of certain foods, which then creates excess demand for food, ultimately creating upward pressure on food prices [14].

The increment in food prices can be translated as a direct issue of food security. By rising the food prices not only the economies of less developed countries will be treated. Consequently, people in low-income regions and food-deficit countries are more vulnerable to the issue of food security than the ones in developing countries. About the environmental implications, the rapid development of biofuels can lead to serious degradation of agricultural land [15]. By clearing native ecosystems to release land for rapid biofuel production it has been proved that there is an increment in carbon dioxide emissions by burning or decomposing biomass and oxidizing humus. If so, then it will undoubtedly suppress any greenhouse gas benefits of biofuels for decades or even centuries to come [16]. Overall, the global development and utilization of biofuels will continue to increase, in order to maximize the potential of biofuels it is necessary enhance the existing technologies for biofuels production. Nowadays, the current global biofuel production needs to be sustained at a 10% output growth rate per year until 2030 to meet the goals of sustainable development scenarios (SDS). Furthermore, transport biofuel consumption needs to almost triple by 2030 to be on track with the SDS. Unfortunately, the world transport biofuel production increased by only 6% until 2019, and at this stage, only a 3% annual production growth is anticipated over the next 5 years [15].

1.3 Conclusions and Perspectives

In this chapter, a summary of how biofuels have evolved over the years was shown. The story about the use of biofuels is as old as the origin of man. Since the beginning of humanity, biofuels have been of vital importance for the development of civilizations. Throughout history, it is possible to observe that research and development in biofuels has been affected according to political, social and economic issues. Nowadays, these factors continue to set the pace of development in the area of biofuels. In order to increase biofuels production, several international biofuels trades have been created, enhancing economic welfare internationally. As well, it is needed to enhance policies to promote biofuel trades and to enhance the production of the next-generation biofuel. As well there is an imperative necessity to seek for improve the existence technologies or develop new once that allow human being to enhance biofuels production and position them as an alternative or substitutes for the predominant fuels.

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Chapter 2
Process Intensification and Circular Economy

Abstract Process intensification is a valuable strategy to enhance the performance of production processes. It may allow reductions in costs and environmental impact, and enhancements in terms of operability and safety. Although the PI philosophy and methodology have a relatively long history in the scientific field, the ideas of this philosophy fit well with the current trends of sustainability and circular economy; since both ideas, in short, seek the reduction of resource use, the reduction of waste, and the continuous and circular use of raw materials. To ensure the sustainability of biofuel purification, it is important to develop processes with low environmental impact, which can also be allowed through the development of intensified technologies. This chapter presents how the intensification of processes is directly related to sustainability, circular economy, and the principles of green chemistry. Finally, a summary of the intensified technologies for obtaining liquid biofuels is shown.

2.1 Introduction

Relationship between industry and environment is crucial for industrial business performance. Environmental impacts have incrementally increased pressure on industrial businesses. Looking back to the beginning of the industrial revolution, mass production of goods was enabled by new manufacturing methods resulting in products with high availability and low costs. Consequently, due to new consumer societies and staggering growth in industrial activity, emissions to environment, solid waste generation and landfill have become increasingly severe [1]. In addition, due to a growing world population and especially strong middle-class growth the demand for resources is expected to rise rapidly indicating a rising consumption of natural resources. Since planet earth's resources are limited the requirements of exponential economic and population growth cannot be met [2]. In this scenario, it is not only the challenge of environmental pollution that is becoming acute but the challenge of global resource scarcity as well. These circumstances confront manufacturing industry to simultaneously cope with the pressure of environmental regulations,

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challenges of resource price volatility and risks in resource supply, in addition to their daily business.

The concept of circularity, especially in terms of closed material loops, is not a concept of novelty originating from recent developments, but has been emerging now and then throughout the history: Before the industrial revolution, i.e. in times of craftsmanship and hand production methods, waste as unwanted or unusable material was virtually unknown [3]. The circular economy (CE) has received increasing attention from policymakers globally as a concept that can support the goals of reducing overconsumption of natural resources while delivering economic benefits [1]. A recent meta-definition which is based on an analysis of 114 definitions of the term reads: "A [CE] describes an economic system that is based on business models which replace the 'end-of-life' concept with reducing, alternatively reusing, [and] recycling [...] materials in production/distribution and consumption processes, [...], with the aim to accomplish sustainable development, which implies creating environmental quality, economic prosperity and social equity, to the benefit of current and future generations" [4]. In this context, the need for the chemical industry to develop processes which are more sustainable or eco-efficient has never been so vital [5]. The successful delivery of green, sustainable chemical technologies at industrial scale will inevitably require the development of innovative processing and engineering technologies that can transform industrial processes in a fundamental and radical fashion [5]. To achieve a rapid design and implementation of process design in a CE context, process intensification (PI) can be a promising approach to develop more sustainable processes. The definition of PI has thus evolved from the simplistic statement of 'the physical miniaturization of process equipment while retaining throughput and performance' [6] to the all-encompassing definition 'the development of innovative apparatus and techniques that offer drastic improvements in chemical manufacturing and processing, substantially decreasing equipment volume, energy consumption, or waste formation, and ultimately leading to cheaper, safer, sustainable technologies' [7]. Several other definitions with slight variations on the generic theme of innovative technologies for greater efficiency have since emerged [8]. The reduction in scale implied by intensification has many desirable consequences for chemical engineering operations. First, the lower mass- and heat-transfer resistances enabled by the reduced path lengths of the diffusion/conduction interfaces, coupled with more intense fluid dynamics in active enhancement equipment, allow reactions to proceed at their inherent rates. By the same token, the more rapid mixing environment afforded by the low reaction volumes should enable conversion and selectivity to be maximized. Residence times of the order of minutes and seconds may be substituted for the hour-scale processing times associated with large conventional batch operations, with beneficial consequences for energy consumption and process safety. PI covers a wide range of processing equipment types and methodologies, as aptly illustrated in Fig. 2.1 [9].

Many of the equipment types classed as 'intensified technologies' have long been implemented in the chemical industry, such as compact heat exchangers, structured packed columns, and static mixers. More recent developments include the spinning disc reactor (SDR), oscillatory baffled reactor, loop reactor, spinning tube-in-tube