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Evaluation of Biomass-to-Biofuel Conversion Technologies for Process-Scale Applications

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ABSTRACT
The depletion of fossil fuels and the concomitant harmful health and environmental impacts during exploration, production and combustion have heightened research interests in the search for more sustainable, renewable and clean sources. Biofuels, including biogas, biohydrogen, bioethanol, biodiesel, and biobutanol, are seen as a major contributor to future sustainable energy mix for domestic and industrial applications hence, significant research efforts are focused on developing biofuels using a wide range of biomass resources and conversion technologies. Biochemical, thermochemical and microbial electrosynthesis conversion approaches have been used to generate different types of biofuels from lignocellulosic biomass, grass, algae and microorganisms under lab and pilot-scale conditions. However, technological, cost and environmental challenges exist with continuous large-scale production to accommodate current and future demands. This article provides a comprehensive overview of contemporary advances in biomass conversion technologies and discusses process concerns associated with full-scale industrial systems design and development for commercial production of biofuels. It presents contemplations on key strategies to address specific challenges associated with some promising biomass conversion technologies for process scale application.

I. INTRODUCTION

The rate of production of fossil fuels to the rate of formation is significantly unbalanced making fossil fuel a non-renewable source of energy. The world consumption of fossil fuels is estimated to increase by 35% over the next 20 years to balance the power requirement of industrialisation. According to Serrano-Ruiz [1] the use of crude oil is expected to surge by 30% with about 111 million barrels per day in 2035 leading to decreasing reserves and subsequent depletion. Also, the debate for a sustainable and renewable clean source of fuel has been further strengthened due to the enormous release of greenhouse gases generated from fossil fuels [2]. The emergence of biofuel as a clean, sustainable and renewable fuel has a promising future as all attention is drawn to the bioenergy industry [3]. The United Nations Climate panel aims at reducing greenhouse gas emissions by 50-80% over the next 33 years by using biofuels [4]. Presently, the production of biofuels has received a considerable amount of research interests and governmental backing, and this is made evident in the on-going productions and marketing of bioethanol, biodiesel, biohydrogen and biobutanol fuels [3]. For instance, over the years, industrial plants have been commissioned to produce biofuels. There are commercial bioethanol plants in the United States of America and Brazil which are mainly reliant on the use of corn and sugarcane, respectively. However, the use of this edible feedstock for the production of biofuel posses a threat to the food and fibre industry [5]. In addition, the cultivation of these raw materials to feed the biofuel industry is expensive and not sustainable [6, 7]. As a result, a more sustainable and renewable source of feedstock such as lignocellulosic biomass is needed as an alternative for the production of biofuel.

Biomass are essential renewable source of energy contributing about 10-15% of the global energy pool [8]. However, as you move to the developing countries, the percentage of reliance on biomass increases from one fifth to one third [8]. The higher proportion of this biomass is used for domestic purposes such as cooking and heating, especially in the temperate zones. Although the variety of biomass from municipal, agricultural and forestry waste require extensive pre-treatment, they are a cheap and reliable source of feedstock. Also, biofuel production from biomass has less tendency to form a high amount of net greenhouse gases as compared with fossil fuels as carbon dioxide is consumed during photosynthesis in the carbon cycle for the generation of biomass [6].

Various technologies for processing of lignocellulosic biomass into biofuels are already in place. However, due to the chemical and structural complexity of the feedstock, converting to liquid fuel involves a combination of different well-integrated processes with control. For example, thermochemical conversion technology involves pyrolysis and or gasification, syngas cleaning and conditioning and subsequent transformation to liquid fuel by Fischer-Tropsch synthesis; meanwhile the biochemical processing route focuses on the production of bioethanol from the fermentation of sugars from cellulosic materials [9]. Although the underpinning principles of these conversion technologies are well documented in the extant literature their incorporation still remains a significant bioprocessing challenge; since the complexity of a feedstock introduces new difficulties [1]. For example, biomass to liquid (BTL) process involves gasifying the biomass at about 1000 ^{o}C under well-controlled oxidizing atmosphere to produce a gaseous stream which is fed to a Fischer-Tropsch, F-T unit to produce liquid fuel. The F-T unit is a very sensitive part of the process and as such sensitive to its feed which is also very difficult to control from the gasifier. Controlling the composition of the output stream of the gasifier or removal of impurities from the feed to the F-T unit require multiple steps and advanced technologies contributing directly to the challenge of integration, cost and hence, doubts in commercialization. In this article, some contemplations on key strategies to address these challenges are discussed as well as recent advances in the conversion technologies.

II. THERMOCHEMICAL CONVERSION TECHNOLOGIES

Thermochemical conversion technologies primarily involve high heating of feedstock to generate intermediate products including bio-oil and gases that can be enhanced into biofuels [10]. The three main types of thermochemical processing are; gasification, pyrolysis and hydrothermal liquefaction. Other thermochemical processes are available but are in essence modifications of the three mainmethods. A summary of the thermochemical processes to convert biomass to liquid fuel and gasoline has been demonstrated in Figure 1.

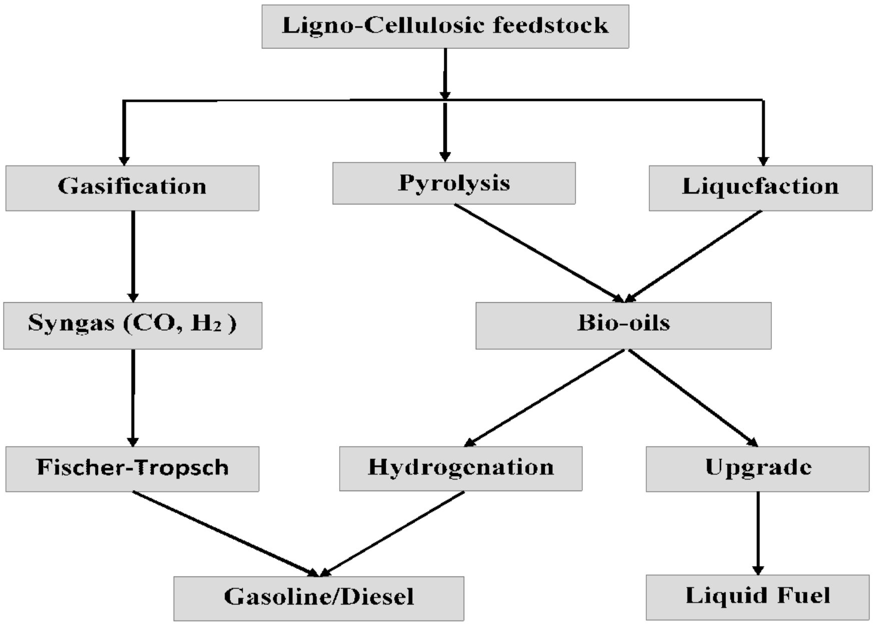

Figure 1: Summary of Thermochemical Conversion Technologies [11]

2.1 GASIFICATION

Energy inherent in biomass is made accessible by gasification. This process converts organic and fossil fuel materials in the existence of minimal air or oxygen at temperatures of 480-1650 ^{o}C. Heat is supplied directly by partial oxidation of the carbon in the feedstock. During gasification, major reactions which take place are boudouard, water gas shift, methanation, hydrogenation, carbon-water and carbon-oxygen [10]. The primary product of biomass gasification is synthetic gas or hydrogen, while there are secondary products such as liquid fuels, gaseous fuels and chemicals which are marketable. Syngas or producer gas can be converted to bioethanol and other synthetic fuels by the Fischer-Tropsch (FT) process.

Typical biomass for gasification; switch grass, elephant grass, corn husks, wood pellets, timbering wastes and bio-solids, usually have a high amount of moisture. Concomitantly, biomass feedstock with high amount of moisture content decreases the temperature in the gasifier which results in reduced efficiency of the process of gasification [6]. The biomass must, therefore, undergo some pre-treatment including drying. The size of the biomass also affects the efficiency of the gasification. Reduced and uniform sizes of the biomass fed at a constant rate to the gasifier allow for uniform distribution of heat and oxygen in the gasification unit. Air is predominantly used instead of raw oxygen for the partial oxidation of the biomass to reduce the cost of production. The utility in using gasification conversion technologies is that they are universally suitable to all plant-based feedstock. Hence, the use of lignocellulosic materials as biomass feedstock can be incorporated into the operations involving gasification [11]. Nevertheless, high amount of moisture content in the biomass [12] as well as impurities emanating from the gases [13] could reduce the efficiency in downstream Fischer-Tropsch processes requiring highly purified gas as a precursor [11].

2.1.1 CATALYTIC GASIFICATION

Catalytic gasification is mainly concerned with the transformation of tar inevitably produced during the process to gases by catalytic cracking. For example, the gasification of biomass in a fluid bed reactor makes use of dolomite to reduce the tar content by transforming it to gases [14]. The chemical composition of the dolomites determines the tar cracking efficiency. Notably, the least tar cracking efficiency can be obtained using dolomites with the minimal composition of CaO and MgO [10].

Activated biomass can also be used as a catalyst for the removal of tar and upgrading of the product gas [15]. This is prepared by means of dry mixing or wet impregnation of the biomass with alkali salts. But considerable difficulty is encountered in the recovery of catalyst and ash disposal. Under elevated conditions of heat and pressure, a mass or coke is often deposited on catalyst. This deposited coke deactivates the catalyst. However, the use of supercritical water can be used to regenerate the active sites of the catalyst for multiple usages [16].

Table 1: Process conditions for the catalytic gasification of various biomass feedstocks and their main products [10]

FEEDSTOCK CONTAINING	GASIFICATION CONDITIONS	CATALYST	PRODUCTS	REFERENCE
Cellulose	10 mins,30 MPa, 623 K	Pt/Al_2O_3	Hydrogen gas and methane	[17]
	1 hr, 8 MPa, 573 K	SnO_2, ZnO	Carbon monoxide, carbon dioxide and hydrogen gas	[18]
	0.1 MPa, 823-923 K	$Rh/CeO_2/SiO_2$	Synthetic gas	[19]
Glucose	34 MPa, 723 K	NaOH	Hydrogen gas	[20]
Methane and carbon monoxide	80 hr, 1073 K	Ni-olivine	Rich Hydrogen gas	[21]
Saw dust	20 min, 27 MPa, 773 K	Ru/C	Hydrogen gas	[22]
	933 K	Fe/CaO	Hydrogen gas	[23]

2.2 PYROLYSIS

Pyrolysis is an energy-intensive thermal process used for the conversion of biomass without oxygen. This means combustion does not take place during pyrolysis but instead the chemical compounds decompose into other forms. During pyrolysis at elevated temperatures of 377-527 ^{o}C, reactive vapour phase products exceed their activation energy to enable a reaction process with about 350 products such as aldehydes, alcohols and aromatics. These products upon cooling form a liquid biofuel acknowledged as “bio-oil” [11].

The feedstock is pre-treated to reduce the moisture content and size before fed into the reactor. To ensure that oxygen and air are kept out, an airlock is used and the biomass in the reactor is heated indirectly [24]. After the reaction, the hot gases and char produced are separated in a gas cyclone and the char is collected. The purified gases are then quenched with cold water and the bio-oil condenses at the bottom, it is collected and stored. The uncondensed gases are then recycled into a combustor to produce fuels. The process reaction scheme is shown in Figure 2. The Bio-oil is purified and stabilized to make it suitable for downstream use. Mild Hydrotreating is used to stabilize the oil. Hydrotreating involves the use of hydrogen to extract about 90% unwanted chemicals such as sulphur, nitrogen and oxygen that may be dissolved in the bio-oils. The removal of oxygen renders the bio-oil to be less reactive with reduced acidity, and thus, safe for storage and in its use as biofuel. Additionally, the less acidity of the bio-oil makes it compatible with already existing pipelines, tanks and equipment in a plant. This also saves cost by mitigating corrosive effects. To orient the size of the bio-oil to required sizes for biodiesel among others, hydrocracking is employed.

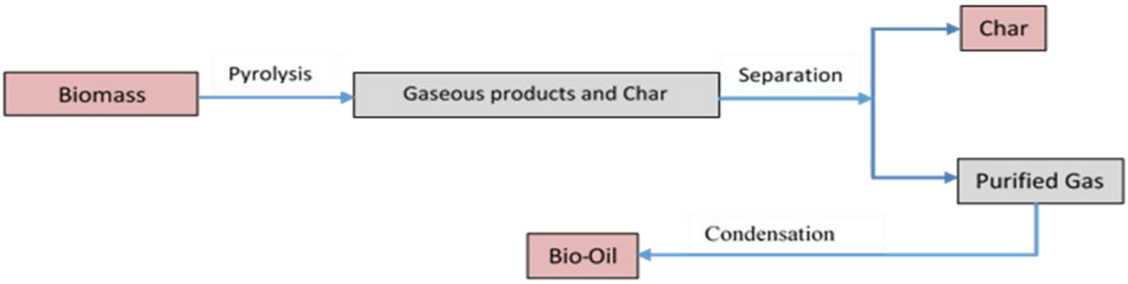

Figure 2: Pyrolysis of Biomass to Bio-oil

According to the heating rate and temperatures, pyrolysis can be classified into;

- Slow pyrolysis; 0.1-1 ^{o}C/s and 300-400^{o}C
- Fast pyrolysis; 10-200 ^{o}C/s
- Flash pyrolysis; >1000 ^{o}C/s

Amongst these, fast pyrolysis is the most commercial [6].

2.2.1 FAST PYROLYSIS

Slow pyrolysis has previously been used in the conversion of biomass at minimal heating rate and protracted duration. The disadvantage with this process is that increased resident time can result in further cracking of the primary products, lower yield and undesirable chemical properties in the produced biofuel [25]. Also, minimal heating rates and protracted duration necessitates the use of higher energy requirements [26]. Thermal depolymerization and decomposition of biomass devoid of air/oxygen at temperatures of 350-700^{o}C is known as fast pyrolysis [10]. The products of fast pyrolysis depend on the heating rate, biomass composition and temperature. Fast pyrolysis is the most commercial because at temperatures above 400 ^{o}C the yield of bio-oil is increased, and it requires particle sizes of less than three (3) millimeters for efficient pyrolysis [10]. On the other hand, flash pyrolysis (10,000 ^{o}C/s) is often not used in the conversion of biomass to bio-oil because at higher temperatures of about 1000 ^{o}C, the bio-oil begins to decompose. Fast pyrolysis enhances the yield of bio-oil obtained including contents of the target compounds. This can be done by monitoring the operating temperature set point at about 500^{o}C and minimal residence time. Maintaining a temperature of about 500 ^{o}C in the pyrolysis reactor with minimal residence time of typically less than two (2) seconds and the rapid removal and cooling of the products enhance the pyrolysis process [27]. The advantage of fast pyrolysis is that it can directly produce a liquid fuel [28].

2.2.2 CATALYTIC PYROLYSIS

Taking the separation between catalyst, liquid products, and corrosion into consideration, the use of zeolites and other solid catalyst in pyrolysis have recently received much attention [10]. ZSM-5, Al-MCM-41 catalyst is widely used in the pyrolysis of lignocellulosic biomass. In particular, hydrocarbons can be produced in considerable quantities by fast pyrolysis of biomass over these catalysts [29]

Table 2: Summary of feedstocks, process conditions and main products of catalytic pyrolysis.

Feedstock	Catalyst	Reactor used	Reaction conditions	Main products	Reference
Corncob	$ZnCl_2$	Quartz reactor	340 ^{o}C	Furfural	[30]
Pine wood	$Fe_2(SO_4)_3$	Conical spouted-bed reactor	500 ^{o}C	Furfural	[31]
Sawdust	NaOH	Conical spouted-bed reactor	500 ^{o}C	Acetol	[31]
Pine sawdust	H-ZSM-5	Conical spouted-bed reactor	400-500 ^{o}C, under N_2 flow	C_4 hydrocarbons	[32]

2.3 HYDROTHERMAL LIQUEFACTION

Biomass reacting at high temperature under high vapour pressure is termed hydrothermal liquefaction. Thermal depolymerization is a phenomenon that occurs during this process. Thermal de-polymerization involves the decomposition or break down of the biomass at high temperatures. Hydrothermal Liquefaction of biomass is ideal for processing high moisture biomass. Pressures are elevated to keep water in either supercritical or liquid state. The resulting liquid typically consists of glycol-aldehyde dimers, 1-3-

dihydroxyacetone dimers, anhydro-glucose, soluble polyols, furfural and phenolic. Feedstock for this technology are; cellulose, hemicellulose, lignin, aquatic biomass, microalgae, waste animal manure and human sewage.

The presence of cellulose and or hemicellulose yields more bio-oil. The formation trend for bio-oil productionusing hydrothermal liquefaction is lipids>proteins>carbohydrates [10]. At higher temperatures, lignin shows a higher hydrothermal liquefaction temperature than hemicellulose and cellulose.

2.4 TORREFACTION

Torrefaction also known as mild pyrolysis is a technology which is employed to improve biomass properties, upgrade the quality and make it a more attractive biofuel. The underlying principle of torrefaction is the removal of oxygen under atmospheric conditions with a final solid product. The oxygen to carbon ratio is lowered in the torrefied biomass compared to the original biomass.

The Fischer- Tropsch process and entrained flow gasification process was employed to produce liquid biofuels [33], torrefaction along with other pre-treatment methods was used during this process. It was concluded that torrefaction has an edge over fast pyrolysis and conventional pelletization and enhances the economic viability of synthetic transportation fuels [34]. Torrefied biomass has a lower weight and density compared with the original biomass due to the reduction in moisture content. This technology therefore has a promising potential for minimizing costs and energy used in long distance transportation of products [35].

2.5 RECENT DEVELOPMENTS, CHALLENGES AND THE WAY FORWARD IN THERMOCHEMICAL CONVERSION TECHNOLOGIES

The pyrolysis process does not require panoptic pre-treatment stages which is common in sugar hydrolysis. However, the bio-oil produced requires extensive pre-treatment before it can be efficiently used in internal combustion engines [11]. This calls for the development of an alternative method for treating the high acidity and oxygenate content of the bio-oil at a cheaper cost. An emerging technology is to couple a pyrolysis reactor with other catalytic reactors such as steam reformer and hydrogenation [10]. A micro-scale pyrolysis reactor can be built from this combination [36]. Gasification on a large scale is in use in South Africa (SASOL). This company employs the Fischer Tropsch synthesis to convert syngas to bioethanol. Recently, it has been demonstrated that Hydrogen gas and carbon monoxide can be produced through aqueous phase reforming of glycerol at temperaturesbelow 347 ^{o}C [37] and subsequently upgrading of syngas with FT synthesis [38]. Organic compounds consisting of glycoaldehyde dimers, 1,3-dihydroxyacetone dimers, anhydroglucose, 5-HMF, Furfural, organic acids and phenolic compounds are produced during hydrothermal liquefaction of biomass. This complex mixture formed cannot be easily separated [10], hence a Novel separation technology needs to be developed for the efficient extraction of these products [39].

III. BIOCHEMICAL CONVERSION

Biochemical conversion involves the production of sugars from the hydrolysis of cellulose and hemicellulose which are further processed into biofuels by the action of catalysts [40]. Biological processes are essentially microbial digestion and fermentation. The biochemical process converts biomass to mainly ethanol, biobutanol and methane [41]. The various stages of this process have been illustrated in Figure 3.

3.1 ALCOHOLIC FERMENTATION

Alcoholic fermentation is the breakdown of carbohydrates in biomass to release sugars and subsequent conversion of the sugars to ethanol.[25, 42]. The stages of production as depicted in Figure 3 involve pre-treatment, hydrolysis of cellulose and hemicellulose to release sugars, fermentation of sugars by microorganisms to produce ethanol and butanol. The ethanol diluted solution is purified through distillation to remove impurities and water and hence concentrated ethanol of about 95% volume is obtained [41, 42]. Again, concentrated ethanol of 99% and better are attainable at the cost of energy.

3.1.1 PRE-TREATMENT

Lignocellulosic biomass is made up of cellulose and lignin existing in the form of a matrix chained by hemicellulose [43]. The hemicellulose serves as a barrier to the cellulose matrix. Pre-treatment which combines both physical and chemical process is used to break this recalcitrant barrier to give access to enzyme catalyzed cellulose conversion [44].

Some physical pre-treatment methods include extrusion, comminution, irradiation and chemical pre-treatments include acid hydrolysis, alkaline hydrolysis, Carbon dioxide explosion, oxidative process and the use of ionic liquids. Chemical pre-treatment methods are normally preceded by physical and mechanical treatments such as chipping and grinding to make material handling easier [44].

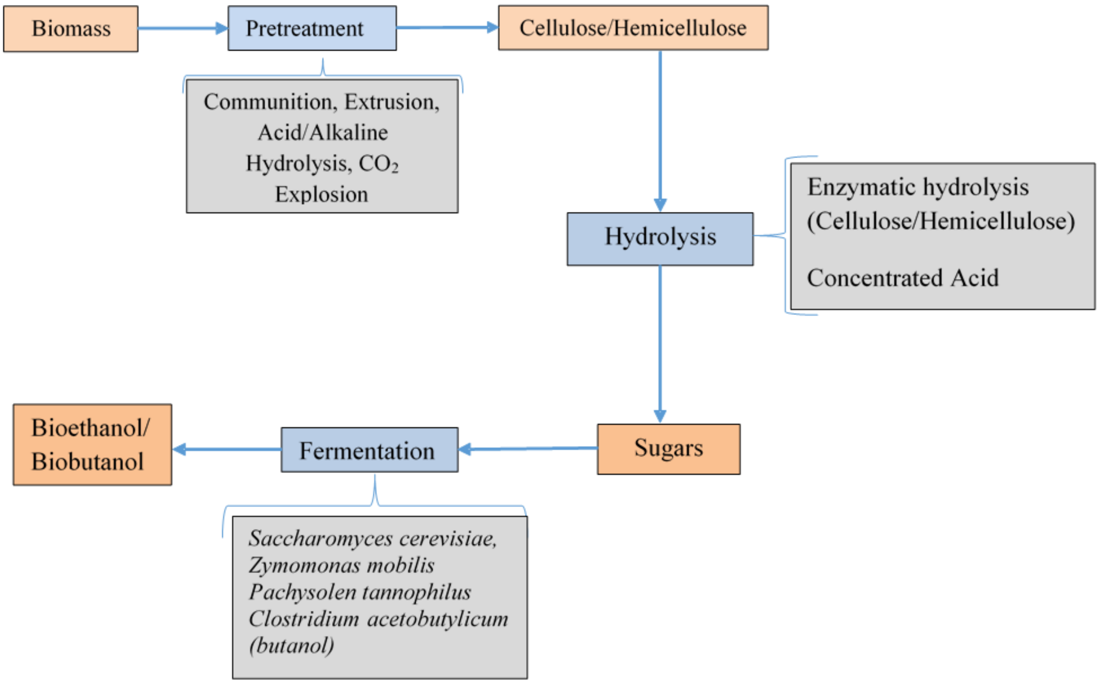

Figure 3: Schematic diagram of fermentation of lignocellulosic biomass to bioethanol or biobutanol

3.1.2 HYDROLYSIS OF CELLULOSE

The hydrolysis of cellulose and hemicellulose to pentose and hexose sugars can either be done with concentrated acid or enzyme catalyst. However, due to the numerous disadvantages of the use concentrated acid such as corrosion, further degradation of sugars into furanladehydes and organic acids and also the requirement of high amount of alkali in subsequent neutralization have made enzymatic hydrolysis a better choice. Cellulase and hemicellulase enzymes are used for the hydrolysis [43].

3.1.3 FERMENTATION

The hydrolysate undergoes fermentation to produce ethanol by microorganisms. There are different microorganisms but the most widely used commercially is yeast [25, 42, 44]. Other microorganisms used are *Zymomonas mobilis, Pachysolen tannophilus* and *Escherichia coli* [43-46]. Biobutanol is produced from anaerobic fermentation using *Clostridium acetobutylicum[47, 48].*

3.2 ANAEROBIC DIGESTION

The process of anaerobic digestion involves the conversion of biomass waste into biogas. Generally, the waste contains high moisture about 80-90% [49]. The conversion of this liquid waste into biofuel occurs in three successive stages in the absence of oxygen [41]. The first stage, known as the hydrolysis stage involves the breaking down of complex carbohydrates mainly cellulose into sugars. The second consecutive stage which is the fermentation stage involves the action of bacteria which convert the sugars into alcohols, hydrogen gas, carbon dioxide and other acidic compounds [42]. Subsequently, the gas which is made up of hydrogen and carbondioxide is converted into mainly methane in the last stage called methanogenesis as shown in the figure 4 below [42, 49].

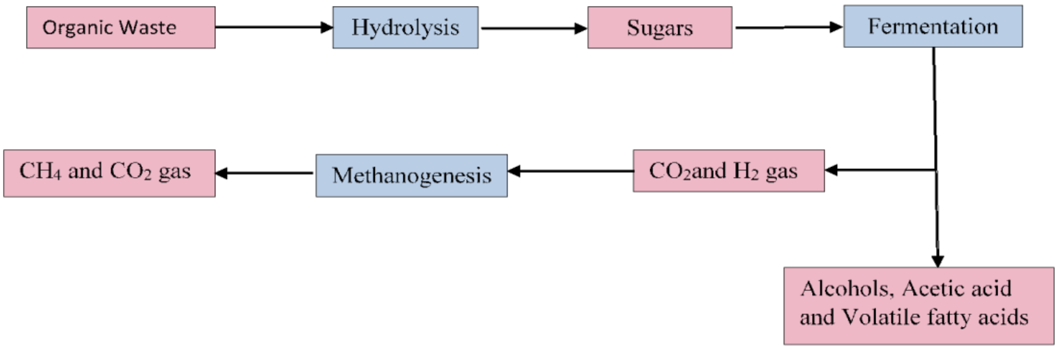

Figure 4: Schematic diagram for anaerobic digestion of organic waste

3.3 RECENT DEVELOPMENTS, CHALLENGES AND THE WAY FORWARD IN BIOCHEMICAL CONVERSION TECHNOLOGIES

Though biochemical conversion technologies have been in existence for quite a while, the general recalcitrant nature of lignocellulosic biomass poses a major challenge to the hydrolysis stage[40].Hence, reducing the efficiency of that stage. Another key challenge is the effectiveness of the process of conversion of sugars into biofuels and its purification. In addition, dealing with a wide variety of feedstock calls for improved effectiveness of cellulose enzymes and fermentation organisms [50].

As part of overcoming these challenges and advancing the quality and quantitative production of biofuels, the Department of Energy, United States of America, has set-up two process demonstration units; one at Lawrence Berkeley National Laboratory which accepts diverse feedstock to demonstrate the efficiency of novel biofuel production processes and the other at the National Renewable Energy Laboratory to evaluate the efficiency and fuel yields for different feedstock blends, process designs and conditions [40].

Additionally, the Bioenergy Technologies Office in the United States of America has begun research in the following areas:

- ➢ enzymes for the hydrolysis to release sugars from cellulose and hemicellulose
- ➢ microorganisms for the conversion of sugars (both pentose and hexose)
- ➢ advanced catalytic conversion of sugars into biofuel blendstocks.

IV. CONCLUSION

Fuels obtained from petroleum fractions have a relatively low cost of production and hence extensive research must be carried out in other to penetrate the existing petroleum-based market. Biofuels from lignocellulosic biomass have the potential of producing sustainable fuels and chemicals. Bioethanol and Biodiesel have been produced on commercial scales from first generation biomass such as corn, sugar cane, canola and soybeans. These first-generation feedstocks are obtained from the edible fraction of food crops and hence not a sustainable source of feedstock. This limitation has brought about the development of second and third generation biofuels. Integration of the various thermochemical and biochemical process technologies creates a lucrative new process for biofuels production. This Integration can create a complex process flow and thus rigorous studies will have to be carried out to determine the optimal operating conditions for the processes. Small particle size and high oxygen to carbon ratio of biomass is a key challenge in its conversion to biofuels. Torrefaction therefore improves these properties and increases the energy density of the biomass.

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