# Polymeric Biodiesel

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Biodiesel industry is expanding rapidly in accordance with the high energy demand and environmental deterioration related to the combustion of fossil fuel. However, poor physicochemical properties and the malperformance of biodiesel fuel still concern the researchers. In this flow, polymers were introduced in biodiesel industry to overcome such drawbacks. This article introduces polymeric biodiesel which is Hydroxyalkanoates methyl ester (HAME) and hydroxybutyrate methyl ester (HBME) that are sourced from carbon-enriched polymers with the help of microbial activity. Composition, production techniques, characteristics, and limitations of polymeric biodiese were explored.

polymers biodiesel polyhydroxyalkanoates hydroxybutyrate Hydroxyalkanoates

## 1. Introduction

Biodiesel fuel is conventionally sourced from four generations of feedstocks. These feedstock generations include edible oils, inedible oils, algae oil, and genetically modified crop oils. Currently, more than 75% of the global biodiesel production is produced from the first-generation feedstock which requires wide cultivation areas[1][2]. In this case, first-generation biodiesel is not green enough since the total greenhouse gas footprint for both fuel combustion and cultivation emissions may exceed that of fossil fuel diesel[3]. Moreover, the accumulating costs of farming, cultivation, production, and treatment are not feasible economically since 70–80% of the biodiesel costs are accounted for the feedstock expenses[4]. Beside the environmental impact and the economic feasibility, ethical issues concern researchers and decision-makers. Large fertile areas, greater quantities of fertilizers, and significant water resources were dedicated to grow the feedstock of first-generation biodiesel. At the same time, the food shortage problem is encountered by many countries all around the world[5]. Therefore, expanding biodiesel production from first-generation feedstock is considered one of the main reasons behind food shrink and increasing food prices. The other three biodiesel feedstock generations seem more sustainable.

Recently, polymers materials were introduced into biodiesel production to enhance the physicochemical properties and fuel performance, particularly for the second-generation feedstocks. Polymers are defined as series of molecules linked together by chemical bonds to form macromolecules. They exist naturally, e.g., proteins, wood, and rubber, and synthetically, e.g., nylon, polyethylene, and polyester[6]. They were introduced almost in every aspect of life including products of medicine, food, furniture, auto parts and so on. The biofuel industry was, and still is, one of the expanding sectors to introduce polymers materials to enhance the properties of fuel products. Polymeric biodiesel was synthesized biologically with the help of micro-organismic activity in presence of polymeric feedstock[7][8][9][10]. In addition, polymers were composited to reduce the higher content of FFA existed in second-generation feedstock oils. Moreover, they catalyze the esterification and transesterification reactions during production line of biodiesel fuel [9][10][11][12][13]. Beside catalysis, polymers were used as additives to improve the cold-flow properties and enhance the fuel performance [14][15][16][17][18]. Moreover, stable polymers were investigated as storage containers and auto parts in direct contact with biodiesel [19][20][21]. So, reviewing those studies would help to build a collective knowledge and solid background to valorize polymers in biodiesel production industry.

## 2. Polymeric Biodiesel

Biodiesel fuel can be synthesized biologically based on polymers materials produced by particular species of microorganisms. These polymers are known as polyhydroxyalkanoates (PHAs). They are produced by prokaryotic microbes under particular nutritional conditions [22]. The bio-based polymers PHAs attracted the attention in multiple applications due to their preferable economic and ecofriendly characteristics which include biocompatibility, biodegradability, diversity, and sustainability. So, they have been used in bioplastic materials, medical applications, agricultural production, and the construction sector [23]. Recently, PHAs were applied to produce the biofuel. Biological-based biodiesel was first synthesized in 2009 by Zhang et al. [24]. The research group depended on PHAs polymers to synthesize hydroxybutyrate methyl ester (HBME) and hydroxyalkanoate methyl ester (HAME) through acid-catalyzed esterification for poly-R-3-hydroxybutyrate (P3HB) and medium chain length PHA (mcl PHA) as depicted in **Figure 1**. The recovery yields were 52% and 65% for HBME and HAME, respectively. Those HBME and HAME are close to the characteristics of biodiesel fuel. The calorific values were 20 MJ/kg and 30 MJ/kg for HBME and HAME, respectively. In the following discussion, the production techniques, characteristics, and limitations of polymeric synthesized biodiesel are itemized in detail.

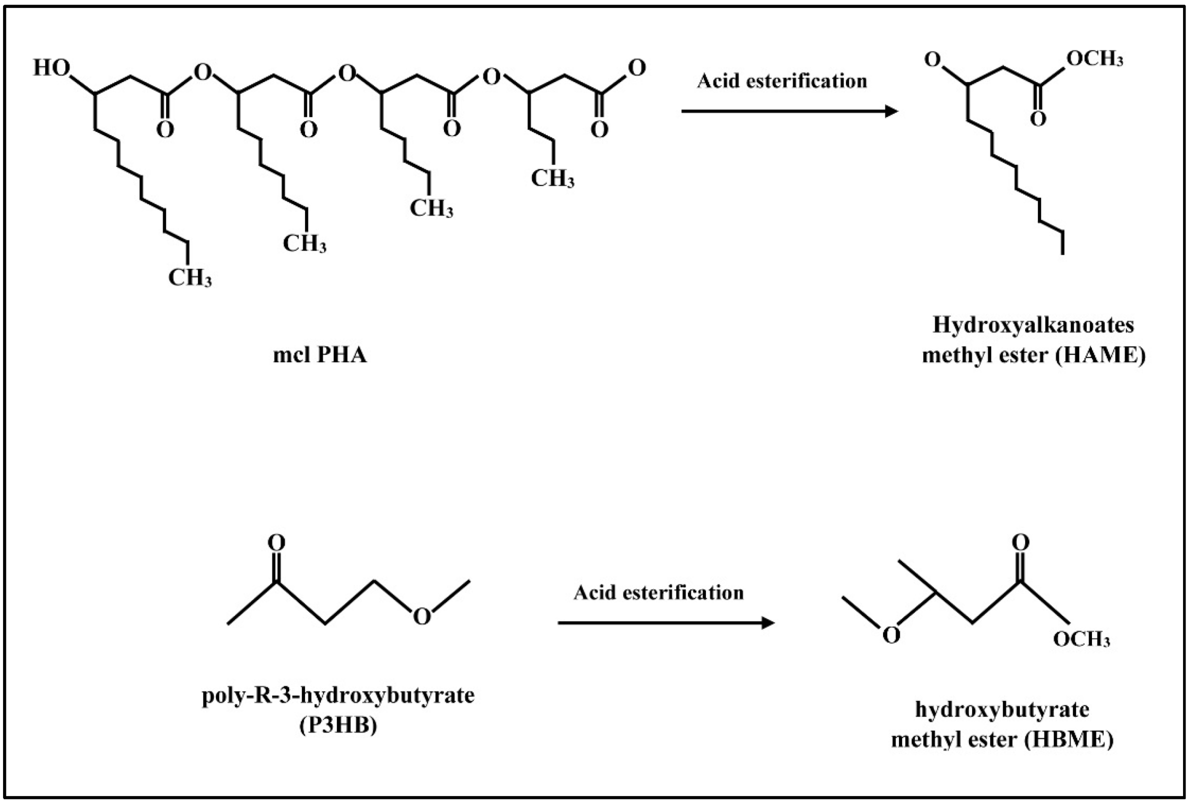

**Figure 1.** Production of polymeric biodiesel HAME and HBME from mcl PHA and P3HB.

## 2.1. Production Techniques

PHAs are polymeric ester-linked molecules consisting of considerable quantities of carbon and energy. Each molecule comprises 600–35,000 units of fatty acid monomers [25]. There are more than 150 recorded monomers, each monomer contains chain (R) group which are often saturated alkyl group or sometimes unsaturated or branched alkyl group. Based on the number of carbon atoms, PHAs are categorized into three groups, short (3–5 carbon atoms), medium (6–14 carbon atoms), and long (above 15 carbon atoms) chain length PHAs biopolymers. The difference in PHAs characteristics comes from the diversity of monomers. If the synthesis is made up of one type of monomer, it is called homopolymer while the synthesis of different types of monomers is called copolymer [26]. **Table 1** shows examples of PHAs with the sources of carbon and synthesis microbes.

**Table 1.** Some examples of PHAs.

| Type of Polymer | Type of PHAs | Source of Carbon | Synthesis Microbe | References |
|---|---|---|---|---|
| **Homopolymer** | P3HB | Waste glycerol | *C. necator* DSM 545 | [27] |
| | | Soy cake and molasses | | [28] |
| | | Sugar of coconut, palm, rock, and toddy | *C. necator* strain A-04 | [29] |
| | | Soybean oil | *R. eutropha* H16 | [30] |
| | | Pineapple crude glycerol | *Bacillus firmus* NII 0830 | [31] |
| | | Cooking oil | *Burkholderia thailandensis* | [32] |
| | PHHp | Heptanoate | *P. putida* KTOY06 | [33] |
| | PHV | Undecanoic acid | *hydrophila* 4AK4 | [34] |
| | PHDD | Sodium dodecanoate | *P. putida* KT2440 | [35] |
| | PHO | Glycerol and sodium octanoate | *P. putida* ATCC47054 | [36] |
| **Copolymer** | P3HB-*co*-HA | Gluconate alkanoates | *Pseudomonas* sp. 61-3 | [37] |
| | P3HB-*co*-P4HB | n-alkanoic acids | *R. eutropha* H16 | [38] |
| | P3HB-*co*-HHx | Lauric acid, and oleic acid | *A. hydrophila* | [39] |
| | P3HB-*co*-P3HV | Lactose, glucose and galactose | *P. hydrogenovora* | [40] |

| Type of Polymer | Type of PHAs | Source of Carbon | Synthesis Microbe | References |
|---|---|---|---|---|
| | P3HB-*co*-P3HV-*co*-P3HHx | Dodecanoic acid and propionic acid | Recombinant *A. hydrophila* 4AK4 | [41] |
| | P(3HP-[26]4HB | Glycerol | Recombinant *E. coli* | [42] |

PHAs as stored intracellula bacterial species, capable to produce PHAs. They were classified into two groups. The first bacterial group requires excessive carbon source and limited nutritional elements of nitrogen, sulfur, magnesium, and phosphorus. *A. eutrophus*, *Protomonas extorquens*, and *P. oleovorans* species are examples of this bacterial group. In contrast, the other group such as *Alcaligenes latus*, *Azotobacter vinelandii* and *E. coli* requires excessive sources of both carbon and the other elemental nutrients [43].

P3HB and mcl PHAs are the polymeric raw materials to synthesize the polymeric biodiesel HBME and HAME. They are sourced from rich carbonic source, such as dewatered activated sludge, and isolated using particular bacterial strains. The produced polymeric materials undergo esterification through acid-catalyzed hydrolysis, in which alcohol methanol or ethanol are added in optimized ratios to complete the conversion into alkyl esters. This chemical reaction is usually catalyzed by acid catalyst, and sulfuric acid ($H_2SO_4$) is used most frequently. However, hydrochloric acid (HCl) and phosphoric acid ($H_3PO_4$) were used to catalyze this chemical reaction. In addition to the acid catalysts, base catalysis was reported to synthesize HAME and HBME from mcl PHAs and P3HB polymeric materials. After the formation of HAME and HBME, saturated solution of salted water is added and stirred intensively. Then, the mixed solution is let to settle down into two layers. The more transparent layer is biodiesel HAME and HBME that should be dewatered and filtered to remove undesirable debris [24][26]. Since the raw materials PHAs for the polymeric biodiesel is non-toxic and utilized in medical, agricultural, and industrial applications, the wastes of this acid-catalyzed hydrolysis do not form threats [44]. However, a toxicity test is highly recommended for the polymeric biodiesel and byproduct wastes in future investigations. **Figure 2** illustrates the production line of polymeric biodiesel (HAME and HBME).

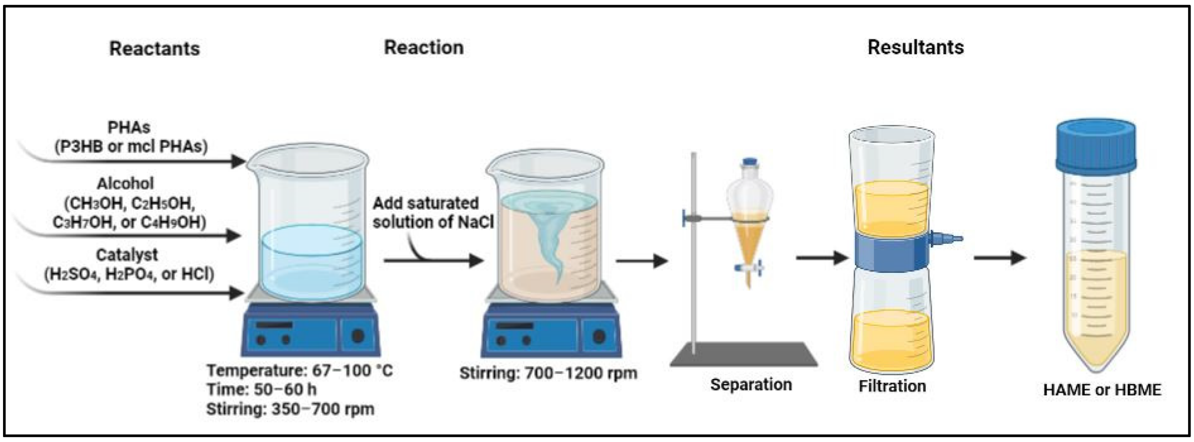

**Figure 2.** Schematic production of polymeric biodiesel HAME and HBME.

There are four parameters affecting acid catalyzed esterification to synthesize polymeric biodiesel, i.e., the type and ratio of alcohol, type and ratio of the catalyst, reaction temperature, and reaction time. Four alcohols can be used in this chemical reaction, methanol, ethanol, propanol, and butanol. However, methanol and ethanol are the most frequently used due to their preferable physicochemical properties and low costs. It was reported that alcohol methanol is more efficient for converting mcl PHAs and P3HB into alkyl esters. The reason behind this efficiency is the short chain of the alkyl group within methanol which has higher solubility and activity. For alcohol ratio, higher amount of alcohol results in higher conversion percentage [26][45]. For the catalyst type and ratio, $H_2SO_4$, $H_3PO_4$, and HCl acids were used to catalyze the esterification reaction. Literature reported usage of base catalysts for this reaction as well. Among all catalysts, $H_2SO_4$ was the most frequently used to yield high percentage of HAME and HBME. The literature reported that the maximum yield of polymeric biodiesel can by synthesized with a catalyst ratio 10–15% [26][40][46]. For the temperature parameter, it is normal to adjust the reaction temperature around the boiling point of alcohol in order to avoid alcohol evaporation. Choonut et al. [47], Keunun et al. [48], and Sangkharak et al. [49] obtained the highest yield of HBME at 67 °C. However, Zhang et al. [24] got the maximum HBME at 100 °C. For time parameter, much time is required for this esterification process. According to Choonut et al. [47], the highest HBME percentage was produced from P3HB at a reaction time 50 h. Further increasing time led to a decrease of HBME yield. **Table 2** explores the previous studies regarding HAME and HBME production with optimum values.

**Table 2.** Optimum values for polymeric biodiesel production HAME and MBME.

| Polymeric Biodiesel | Polymers Source | Reaction Parameters | | | | | Yield (%) | References |
|---|---|---|---|---|---|---|---|---|
| | | Alcohol | Catalyst | Ratio % (C in A) * | Temperature (°C) | Time (h) | | |
| HAME | mcl PHA | Methanol | $H_2SO_4$ | 15 | 100 | 60 | 65 | [24] |
| HAME | mcl PHA | Methanol | $H_2SO_4$ | 10 | 67 | 60 | 68 | [49] |
| HBME | P3HB | Methanol | $H_2SO_4$ | 15 | 100 | 60 | 52 | [24] |
| HBME | P3HB | Methanol | $H_2SO_4$ | 10 | 67 | 60 | 40 | [50] |
| HBME | P3HB | Methanol | $H_2SO_4$ | 10 | 67 | 50 | 70.7 | [47] |
| HBME | P3HB | Methanol | $H_2SO_4$ | 10 | 67 | 60 | 65 | [48] |

* C in A: catalyst in alcohol.

## 2.2. Characteristics and Limitations

Polymeric biodiesel (HAME and HBME) has physicochemical properties almost close to biodiesel fuel. In spite of being sourced from low-cost feedstocks like sewage sludge or wastes, HAME and HBME offer valuable, ecofriendly, and sustainable biodiesel fuel. Unlike vegetative and animal-based biodiesel, the polymeric biodiesel does not require extensive purification process. The calorific value of this pure biologically derived fuel reached 20 and 30 MJ/kg for HBME and HAME, respectively. The heating value can be improved to 30 and 35 MJ/kg for HBME and HAME respectively by adding 10% ethanol [24]. The polymeric biodiesel has preferable performance characteristics. The start ignition is faster than vegetative and animal-based biodiesel. In addition, just short combustion time is needed to fully burn. Plus, this fuel produces very little trace amounts of exhaust smoke. The reason behind these preferable performance characteristics refers to the high oxygen content of HAME and HBME fuels which helps to combust the full fuel within short time and produce neglectable smoke. Besides, the polymeric biodiesel contains neither nitrogen nor sulfur. That is why its emissions are more ecofriendly since it is free of nitrogen oxides and sulfur oxides. Thus, sustainable biodiesel fuel with favorable characteristics is offered by biopolymers materials derived from low-cost feedstocks which are not competitive with food resources [26]. **Table 3** explores the characteristics of polymeric biodiesel HBME.

**Table 3.** The physicochemical properties of polymeric biodiesel (HBME) (adapted from [48][49][50]).

| Physicochemical Properties | Unit | Value |
|---|---|---|
| Density at 20 °C | $Kg/m^3$ | 900 |
| Viscosity 20 °C | $mm^2/s$ | 4 |
| Pour point | °C | 1 |
| Flash point | °C | 68.5 |
| Heating value | MJ/kg | 25.1 |
| Cetane number | - | <1 |
| Octane number (RON) | - | 62.2 |
| Oxygen content | %wt | 41 |
| Oxidative stability at 100 °C | h | 8.13 |

Despite the preferable characteristics of biologically derived polymeric biodiesel, there are some drawbacks that may limit using HAME and HBME as an alternative fuel. The conversion percentage of mcl PHAs and P3HB into alkyl esters is lower compared with vegetative and animal-based biodiesel. Zhang et al. [24] recorded conversion percentages for the first time as 52% and 65% for HBME and HAME, respectively. However, 70% recovery yield was recorded later by Choonut et al. [47], Keunun et al. [48], and Sangkharak et al. [49]. Beside the low conversion percentage, much time is required to reach the maximum yield. The optimized results of Choonut et al. [47] revealed that only 12.8% yield of HBME can be obtained from P3HB after 10 h of esterification reaction. The yield increased to 70.7% once the reaction time was doubled five times into 50 h. However, the HBME yield was decreased beyond 50 h of reaction time. The polymeric synthesized esters of HBME and HAME have a lower cetane number compared with conventional diesel and biodiesel since they are highly oxygenated compounds. Moreover, their vaporization latent heat is high. Thus, much enhancement is needed before fueling engines with polymeric biodiesel [24].

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