

Next-generation circular waste biorefineries based on biogas utilization using renewable energy: Process composition and economic analysis

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The integration of waste-to-energy processes with CO₂ reduction and renewable energy storage was investigated. Here, we demonstrate a waste biorefinery that utilize biogas, which was previously used only for thermal energy. High value-added chemicals such as succinic acid, polyhydroxybutyrate (PHB) and synthetic natural gas can be produced by industrial microorganism from food waste and renewable energy. Water electrolysis facilities accounted for a significant portion of the investment cost. However, the rising demand for bioplastics helped shorten the payback period despite their relatively high price. The quantity and economic feasibility of final products changed according to the order of the process, and the possibility of circular waste biorefinery based on the use of biogas using renewable energy was suggested.

Keywords: Power-to-gas, Waste-to-energy, Biogas, Value-added products, Techno-economic analysis.

Introduction

Over 105 billion tonnes of organic waste—including food, agricultural, and industrial waste—are generated globally each year, releasing greenhouse gases (GHG) into the atmosphere as they decompose. Effective management and recycling of these bioresources could reduce global GHG emissions by up to 10% by 2030 [1]. As part of this global effort, South Korea has initiated strategic measures to enhance the management of organic waste, with a national target to increase biogas production by 36% through anaerobic digestion by the year 2027 [2]. Anaerobic digestion has evolved from a waste treatment method to an energy production process and further to the production of value-added products [3]. Direct methanation of biogas is one of the long-term energy storage methods that capture CO₂ from biogas and use hydrogen from water electrolysis using renewable energy [4–6]. Electro-assisted biosynthesis and biomolecule conversion are emerging technologies that integrate electrical energy with biological processes to enhance the efficiency, sustainability, and selectivity of biochemical reactions [7, 8].

Water electrolysis is a method of converting electrical energy into chemical energy. The electrons are stored

in the chemical bonds of hydrogen and can be used as a fuel or reused as electrical energy when needed. The four types of water electrolyzers are alkaline, PEM (polymer electrolyte membrane), AEM (anion exchange membrane, currently at the lab scale), and solid oxide (also at the lab scale). In order to produce hydrogen from renewable energy, it is still necessary to reduce the cost of the electrolysis facility [9].

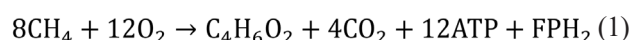
Biological methanation of biogas can use a mixed gas containing H₂S and NH₄ from biogas (high tolerance of impurities) and has the advantage of operating at a lower temperature and pressure than chemical methanation [10]. The resulting methane concentration is greater than 95% and can be inject into existing natural gas pipelines [11]. Thermophilic hydrogenotrophic methanation (THM, 60–65 °C) is primarily applied because of its faster conversion rate than mesophilic methanation [12]. Several pilot plants regarding THM were tested such as Electrochaea in the BioCat project [13], Microbenenergy in the BioPower2Gas project [14], and Copernicus P2X project. The economic analysis of THM indicated that the water electrolysis process contributes the most to the overall cost of biomethanation production [10]. However, carbon dioxide of biogas also can be converted into methane without the emission from the process and high methane content can be used for transportation fuels like natural gas [15].

Carbon dioxide from biogas can be used to produce succinic acid through the valorization of organic waste.

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Succinic acid is one of the promising platform chemicals and used as a precursor for polymer, resins, and bio-based succinic acid from renewable resources is attracting attention. *Actinobacillus succinogenes* [16], *Succinivibrio dextrinosolvens* [17], and *Basfia succiniciproducens* [18], and *Mannheimia succiniciproducens* [19] can produce succinic acid as a major end product via fermentation fixating CO₂ as a second substrate. They are capnophilic (CO₂-loving) bacteria and heterotrophs with faster growth and CO₂ fixation rate than autotrophic microorganisms. Based on the recently reported succinic acid productivity, 21.3 g/L/h [20], the CO₂ fixation rate is much faster at 4.41 Nm³ CO₂/m³/h compared to 0.001-0.032 Nm³ CO₂/m³/h by microalgae [21]. Various organic wastes were tested for succinic acid production [22], and crude glycerol, a major byproduct of biodiesel production, was one of economical feedstocks [23]. The life cycle assessment analyzed that the selling price for succinic acid production from glycerol was \$1.6-1.9 per kg, while production from corn stover was \$1.7-2.0 per kg [23].

Oxygen produced in water electrolysis can be used to produce polyhydroxybutyrate (PHB) using methane-oxidizing bacteria. There are two pathways for PHB production using methane: the ribulose monophosphate pathway (type I) and the serine pathway (type II) (Table 1). Type I methylotrophs lack the complete TCA cycle and require the energy of acetyl-CoA, a key intermediate for PHB production while Type II need greater reducing power for serine cycle [24]. PHB accumulates during the stationary phase of growth under nutrient-limited condition, such as nitrogen, phosphate, oxygen deficiency [25]. The theoretical conversion efficiency from methane to PHB is 67% (Eq. 1) [26].



Where FPH₂ is reduced flavoprotein [26]. A large fraction of CH₄ and O₂ consumed has to be converted to CO₂ to generate reducing power such as NADH₂ and FPH₂ in type II methanotroph [26]. *Methylosinus trichosporium* [27], *Methylocystis parvus* [28], and *Methylocystis hirsuta* [29] are type II methanotrophs that use methane to produce PHB. The PHB price from methane was reported \$4.1~\$6.8/kg based on 100,000 t/a PHB production facility [29].

In this study, a combined system integrating biological methanation, succinic acid production, and PHB production for biogas valorization was investigated and simulated using SuperPro Designer software. The technology economics associated with each process were evaluated and the return on investment was compared using payback time.

Materials and Methods

The process was simulated using SuperPro Designer[®] (Version 12) software by Intelligen, Inc. The total capital investment was calculated as shown in Table S1.

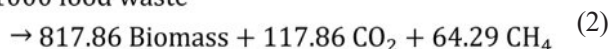
Anaerobic digestion (AD)

Anaerobic digestion is a method of recycling food waste into products such as animal feed and compost. AD installation cost was not included in the calculation because this study was based on adding another process to value the existing AD installation. In the Republic of Korea, the standard biogas production value is 100 m³ per ton of food waste. In this study, 90.6 m³ methane production per ton of raw food waste was used [30] (Table 1). The reaction mass stoichiometry of AD is Eq. (2). The daily throughput was calculated as 50 tons of raw food waste.

Table 1. The list of values used for SuperPro Designer modeling.

Process	Factors	Values
AD	Biogas production	Methane yield, 90.6 m ³ /ton of raw food waste [31]
	Methane price	\$1.35 per kilogram [49]
	CO ₂ fixation rate	400 VVD
	Methane price	\$1.35 per kilogram [49]
THM	Water electrolyzer unit cost	High-cost scenario: \$500/kW Low-cost scenario: \$380/kW
	Electricity needed (one kg H ₂ O/hr)	11 kW
	Water electrolyzer efficiency	100%
SA	Organic substrate	Crude glycerol \$0.16 per kilogram [50]
	Succinic acid production	Succinic acid yield, 0.9 g/g crude glycerol [37]
		Succinic acid productivity, 21.3 g/L/h [20]
	Theoretical CO ₂ fixation	0.21 L STP CO ₂ /g succinic acid
PHB	Succinic acid price	\$2.5 per kilogram [51]
	PHB accumulation	PHB yield, 0.55 g PHB/g methane [52]
	PHB price	\$11 per kilogram [36]

1000 food waste

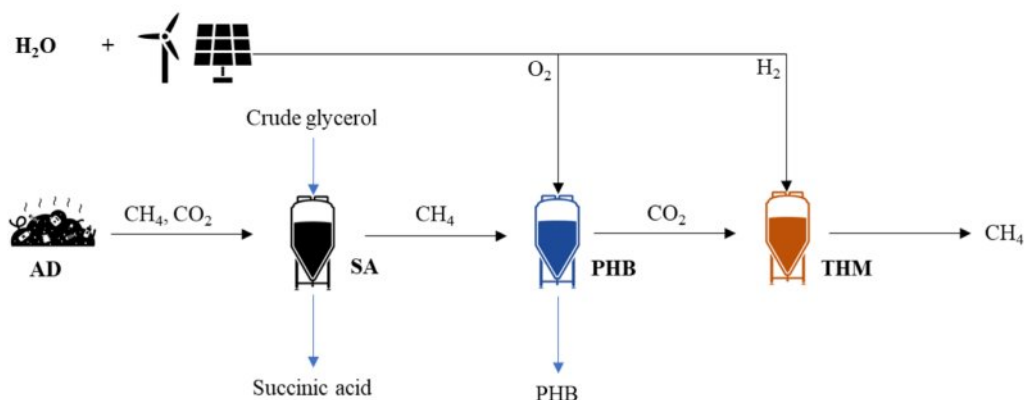


Thermophilic hydrogenotrophic methanation (THM)

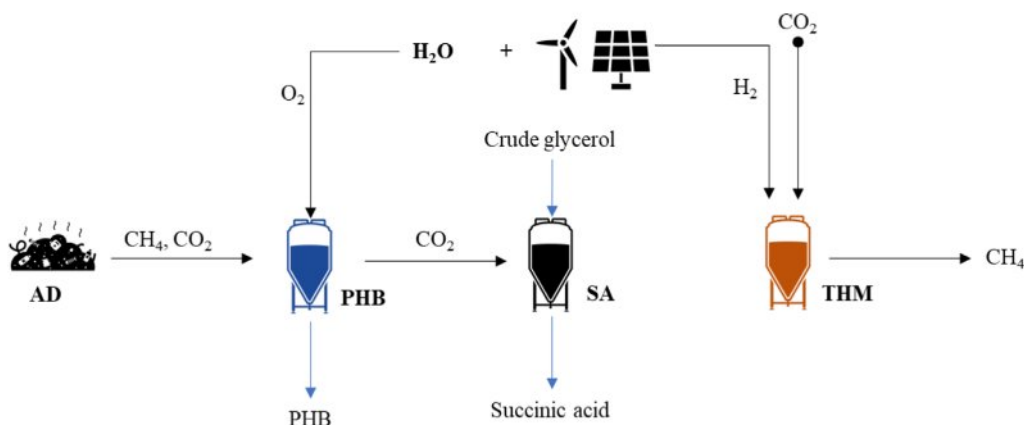
Thermophilic hydrogenotrophic methanation (THM) has been extensively studied because it has a faster methanation rate than mesophilic methanation at the

pilot scale. The main construction cost is the facility of water electrolysis [32]. The cost of water electrolysis plant was estimated at \$500/kW and the power was calculated at 5 kW per 1 Nm³ H₂/h production. A techno-economic assessment of power-to-gas estimated a payback period of approximately 15 years [32]. The cost of water electrolysis and electrical consumption was not included in the calculation because it is premised

(a) AD-SA-PHB-THM



(b) AD-PHB-SA-THM



(c) AD-THM-PHB-SA

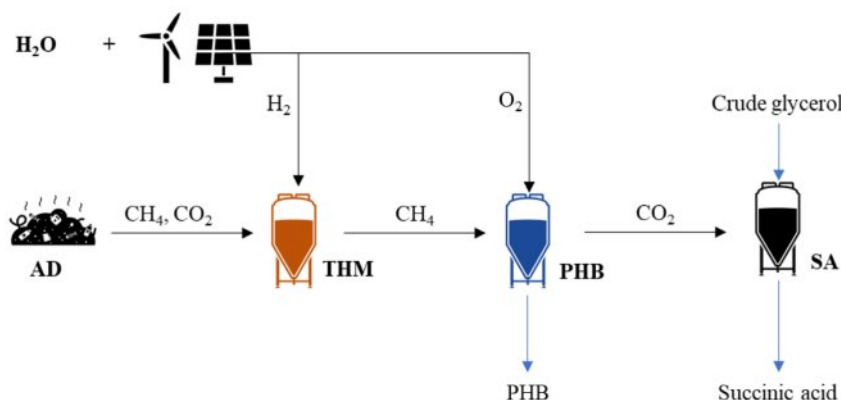


Fig. 1. Synthetic natural gas, PHB, and succinic acid production using biogas from food waste.

Table 2. Equipment specification and cost (Free on Board).

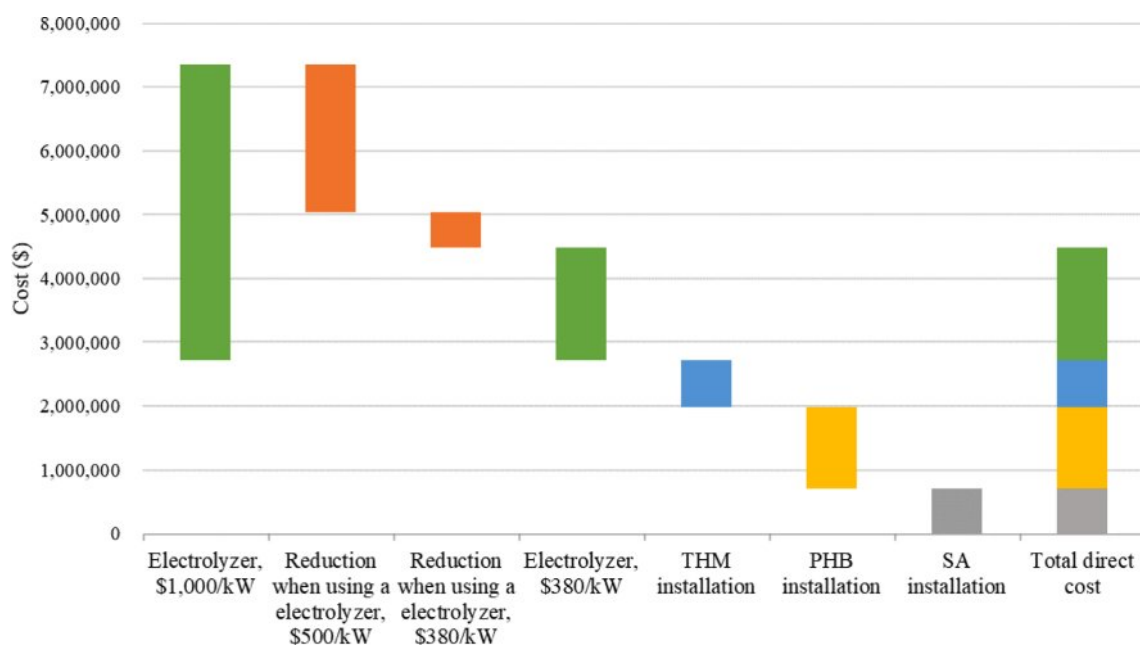
Process	Facility	AD-SA-PHB-THM	AD-PHB-SA-THM	AD-THM-PHB-SA
SA	Fermenter	\$599,000	\$599,000	\$599,000
	Clarification	\$1,000	\$1,000	\$1,000
	Succinate precipitation	\$102,000	\$102,000	\$102,000
	Sub-total	\$702,000	\$702,000	\$702,000
PHB	Fermenter	\$716,000	\$716,000	\$716,000
	Clarification	\$1,000	\$1,000	\$1,000
	PHB extraction	\$271,000	\$268,000	\$263,000
	PHB leaching	\$5,000	\$5,000	\$5,000
	PHB crystallization	\$288,000	\$288,000	\$287,000
	Sub-total	\$1,281,000	\$1,278,000	\$1,272,000
THM	Electrolyzer	\$2,318,000	\$2,318,000	\$2,318,000
	Reactor	\$733,000	\$1,500,000	\$600,000
Total		\$5,034,000	\$5,798,000	\$4,521,000
Electrolyzer percentage (%)		46.0	40.0	43.1

wastes in the combined system was the highest in the AD-THM-PHB-SA system, with a daily output of 2,250 kg CH₄. This is about 77% of the 2,920 kg CH₄ daily production in AD alone (Fig. S1). Other output carbons in the combined system were converted to value-added materials such as PHB and SA. Succinic acid production was highest in the AD-PHB-SA-THM system due to the combination of CO₂ generated from PHB production and CO₂ from AD. The SA production was 1,216 kg/day in AD-PHB-SA-THM system, compared to 596 kg/day in AD-SA-PHB-THM, 522 kg/day in AD-THM-PHB-SA (Fig. S1).

The cost of electrolyser played an important role in the

overall economic evaluation. Required electricity power was 3.9 MW (AD-THM-PHB-SA, AD-THM) and 4.6 MW (AD-SA-PHB-THM, AD-PHB-SA-THM). At the current 2020 technology level, the price of electrolyser system was calculated to be \$500/kW [8]. Electrolyser costs accounted for 40–46% of the total equipment purchase cost (Table 2).

The cost of installing water electrolysis remains high. According to IRENA, electrolyzer costs are expected to range from \$130/kW to \$307/kW with an estimated 1–5 TW of deployed capacity by 2050 [9]. The electrolyzer cost was set at \$380/kW [40], which is comparable to the installation cost of other processes and expressed

**Fig. 3.** Waterfall chart showing the overall cost reduction due to reduced electrolyzer cost in a biogas refinery system.

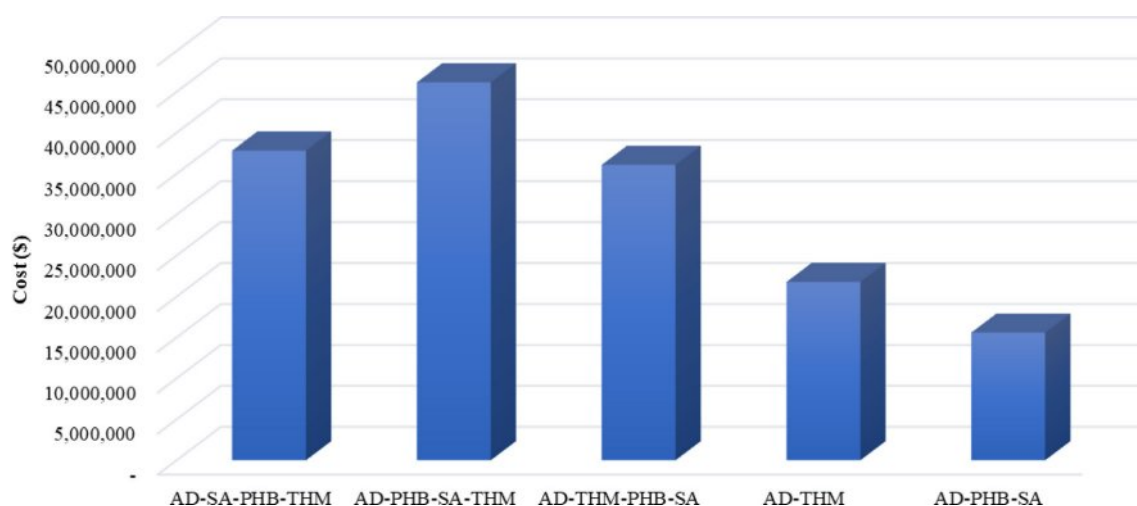


Fig. 4. Capital investment cost calculated at \$500/kW for water electrolyzer.

as a waterfall chart as shown in Fig. 3. The cost of electrolyzer of \$1,762,000 was comparable to the cost of \$1,281,000 installation for PHB production. Because of the high cost of the electrolyzer, the capital investment was higher with an electrolyzer than without it (Fig. 4). The capital investment is the sum of direct fixed capital (DFC), working capital, startup, and validation cost. The most expensive capital investment was the case of AD-PHB-SA-THM (Fig. 4). The AD-PHB-SA system had the lowest capital investment (Fig. 4).

However, in terms of revenue, all cases were not big different (Fig. 5). Income is divided into three categories: revenues, carbon credit, and food waste overhead income. The prices of methane, PHB and succinic acid were calculated at \$1.35/kg, \$11/kg and \$2.5/kg, respectively. The carbon credit was set \$15.89/ton CO₂ in a Korea-Emissions Trading Schemes (ETS) and methane ETS was calculated 25×CO₂ ETS. Food waste overhead income includes waste tipping fee (\$100/ton FW) under waste management system. In Korea, it costs an average \$100 to process 1 ton of food waste through a resource recycles program, including correction, transportation, and treatment [41]. AD-PHB-SA-THM and AD-PHB-SA show high revenue income. Income from the carbon credit and tipping fee accounts for 41.2–50.4% of the total income amount (Fig. 5). The daily production of methane is the highest in all case (Fig. 5a), but PHB was the most profitable because of its high price (Fig. 5b).

The total capital investment, operation cost of each combination process, and the economic analysis of the scenarios were shown in Table 3. The payback period was determined based on total revenue compared to capital investment and operating costs (Table 3). The payback time is 7.4 year for AD-SA-PHB-THM and 2.9 year for AD-PHB-SA (Table 3).

The investment cost of water electrolysis was the most expensive part in whole processes suggested in this study. Renewable energy sources such as solar and wind power

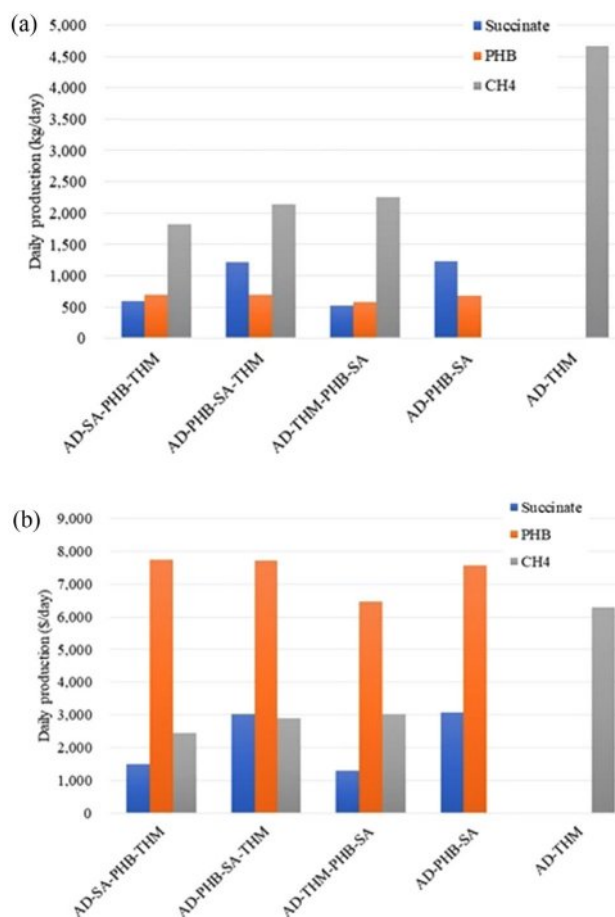


Fig. 5. Daily production amount (kg/day, a) and revenues (\$/day, b) from each system.

can be used as a power source for water electrolysis. Green hydrogen costs range from \$3/kg to \$6.55/kg, while fossil-based hydrogen is \$1.80/kg [42]. The generated hydrogen can be used as a reducing agent for chemicals production by microorganisms and oxygen can be used as

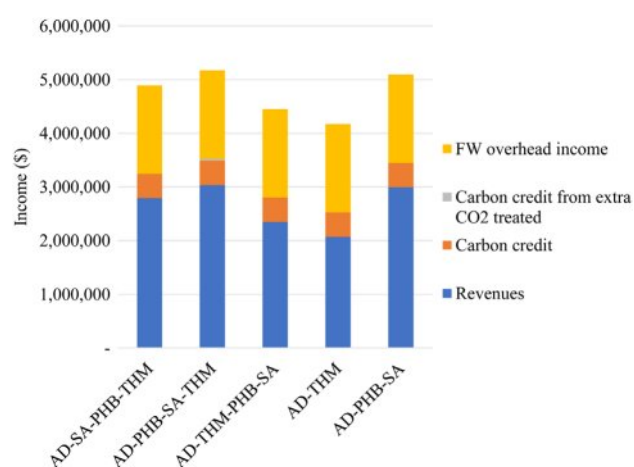
Table 3. Economic parameters of each process from 50 tons of food waste.

Parameters	AD-SA-PHB-THM	AD-PHB-SA-THM	AD-THM-PHB-SA	THM	AD-PHB-SA
Capital Investment (\$)	35,673,000	41,619,000	32,268,000	17,746,000	15,592,000
Operating Cost (\$)	312,000	366,000	320,000	155,000	192,000
Total Income (\$)	6,759,298	8,255,433	6,377,298	4,174,298	7,277,298
Revenues (\$)	4,657,000	6,116,000	4,275,000	2,072,000	5,175,000
Carbon credit (\$)	452,298	489,433	452,298	452,298	452,298
Overhead income (\$)	1,650,000	1,650,000	1,650,000	1,650,000	1,650,000
Gross Margin	0.954	0.956	0.95	0.963	0.972
Return On Investment	0.136	0.142	0.141	0.17	0.34
Payback Time	7.4 year	7.0 year	7.1 year	5.9 year	2.9 year

an electron acceptor for aerobic microorganisms. Using biogas, biological conversion to value-added chemicals such as succinic acid and PHB can be profitable addition to renewable biomethane production as shown in this study. Revenue was determined by the process sequence in the overall process flow, market price, operating costs, and additional substrate requirements for biological production. This study suggested that biogas can be used to produce a variety of compounds biologically, and that different processes can be combined to generate revenue according to substrate-product, so that the net profit can be different depending on the process sequence.

The production of electricity from biogas in the world 91,819 GWh in 2019 and it charge 16.5% in bioenergy including solid biofuels (69.8%) and renewable municipal waste (12.3%). To increase the profits of biogas, bioplastics can be a candidate material that can provide economic benefits along with electricity production. Biological recycling pathways, such as anerobic digestion and aerobic composting, constitute a circular plastic economy. For plastic production, renewable energy supply such as photovoltaics and wind power can cut greenhouse gas emissions [43]. Bioplastics production is attractive because its current high price. PHB revenue from methane was calculated to be 2.2 times that of electricity revenue from methane [44]. Our results also showed that bioplastics production is more profitable than biogas refining due to market price difference. High product costs of bioplastics can be lowered by using waste. By adding a modular production facility to the biogas plant, the range of biogas utilization can be expanded. The result of life cycle assessments in AD with bioplastics reduced greenhouse gas (GHG) by an average of 55% [45].

CO₂ and renewable resources are the solution for developing more sustainable polymers [46]. They can be obtained from AD of a typical waste management system and overcome cost-inefficient problems currently encountered in sustainable polymers production. The biogas utilization for plastics production means carbon capture and it can turn GHG emissions to carbon utilization. Incineration is used to treat non-biodegradable

**Fig. 6.** Income composition for each process.

plastics such as polyethylene, polypropylene, polystyrene, poly (vinyl chloride) and poly (ethylene terephthalate) [43], and the CO₂ generated can be used to produce succinic acid (Fig. 6). A recent report has shown that a combination of biomass and CO₂ utilization can achieve net zero-emission plastics with operating costs similar to linear carbon pathways [47]. Carbon capture company, LanzaTech has developed scalable carbon-negative approach for the production of acetone and isopropanol using synthetic biology [48]. Transitioning from a linear to a circular economy requires reusing products, and biogas must be integrated into a carbon circular system. Biogas can contribute not only to energy production but also to the circular plastics economy, by processing the possibility of producing biodegradable plastics circular economy.

Conclusions

Bioplastics production from the biogas is suggested for circular plastic economy in addition to biomethane production using hydrogen from water electrolysis by renewable energy. The cost of water electrolyser was the largest investment, and a major factor in prolonging the

payback period. Through the production of high value-added bioplastics, economic benefits can be increased and a contribution to the circular carbon economy can be achieved.

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