

Environmental Impacts of Three Polyhydroxyalkanoate (PHA) Manufacturing Processes

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This work investigates the environmental impacts of three manufacturing processes for a promising biodegradable polymeric material: polyhydroxyalkanoate (PHA). The processes studied are microbial PHA manufacturing using glucose as the carbon substrate, microbial PHA manufacturing using cheese whey as the carbon substrate, and microbial PHA manufacturing using genetically engineered plants. A lifecycle model is established for each of the processes, and the Eco-indicator 99 methodology is used in the study. Results with 10 indicators under human health, ecosystem quality, and resources are obtained and compared. Because the processes are still largely under development, many lifecycle data are rare and insufficient. A number of reference sources and reasonable assumptions are used in the calculations. The study has identified the major contributors to environmental impacts, and can quantitatively explain how each impact indicator is affected by a particular process. The study provides the information for technical decision-making for the development of environmentally-friendly PHA manufacturing processes.

Keywords Biodegradable material; Carbon dioxide emissions; Carbon substrate; Cheese whey; Ecosystem quality; Energy consumptions; Environmental impact; Glucose; Human health; Lifecycle model; Manufacturing processes; Microbial; Polyhydroxyalkanoate (PHA); Resources; Transgenic plants.

1. INTRODUCTION

Recently, researchers, engineers, and manufacturers have become to increasingly pay attention to the impacts of material manufacturing processes on our environment. Many journal articles report or discuss the search for environmentally and economically attractive materials [1], the need to reduce carbon emissions, the energy price and environmental pressures on the industry [2], the need to manufacture and process lead-free materials in the microelectronics industry [3–8], the trend to completely avoid, minimize, or carefully select cutting fluids in metal machining processes for ecology, human health, and environmentally friendly reasons [9], and the possibility to minimize energy consumption due to the current global energy crisis [10].

Polymeric materials have found more applications in many fields such as micro-systems [11, 12] and biomedical engineering. Two categories of biodegradable polymeric materials are used in the medical applications: synthetic and natural biodegradable polymers [13]. Among many natural biodegradable polymers [13, 14], polyhydroxyalkanoate (PHA) has drawn increasing attention. PHA possesses properties of biocompatibility, biodegradability, and thermo-processibility for implant and controlled drug release applications. PHA for medical implant applications has a bright future as a tissue engineering material [15].

PHA materials can be manufactured in a number of processes. To understand the environmental impacts

of various PHA manufacturing processes, a life cycle assessment (LCA) study has been performed. This article reports the LCA results on three alternative microbial PHA manufacturing processes, namely, using glucose as the carbon substrate, using cheese whey as the carbon substrate, and using genetically engineered plants.

2. PHAS

Poly-beta-hydroxybutyrate (PHB) is the most common type of PHAs and was isolated and characterized by Maurice Lemoigne from *Bacillus megaterium* in 1925 [16]. It has been found in a large number of bacteria. The bacterium *Alcaligenes eutrophus* is one of the microorganisms most frequently used for the biosynthesis of PHAs [17]. Biosynthesis of PHAs in bacteria has three distinguished metabolic phases [18]: (1) A carbon source suitable for biosynthesis of a PHA must enter the cell from the environment; (2) Anabolic or catabolic reactions, or both, convert the compound into a hydroxyacyl coenzyme A thioester (a substrate of the PHA synthase); (3) The PHA synthase uses these thioesters as substrate and catalyzes the formation of the ester bond with the concomitant release of coenzyme A. Natural organisms, such as *Azotobacter*, *Ralstonia*, and methylotropic bacteria, can be used for manufacturing of PHAs. Modified organisms such as recombinant *E. coli* [18] can be more efficient. Manufacturing of bioplastic in bacteria is limited by its high cost compared with petroleum-derived plastics [19]. This has motivated researchers to explore eukaryotic systems, especially crops, as manufacturing hosts for less expensive processes [20].

Comparing with the technical and economic aspects of PHAs, their life cycle impacts on the environment are relatively less known. It is reported that using corn stover

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and corn grain for manufacturing PHA would reduce the environmental impacts of the PHA manufacturing [21]. Using fermentation-derived PHA would not appear to be a sustainable approach [22]. It is the use of biomass power (not renewable feedstock) that makes the product preferable to PE from a greenhouse gas point of view. The use of renewable resources to produce the PHA plastic has the added benefit of sustainability [23]. A life cycle analysis of PHA derived from soybean and glucose revealed that the CO₂ emission and life cycle inventory (LCI) values of energy consumption were smaller for the PHA copolymer from soybean oil than for P(3HB) from glucose [24].

3. THE METHODOLOGY USED

The methodology used in this study is the life cycle assessment (LCA) technique based on the ISO 14040 series of standards. An LCA study consists of four phases: (1) goal and scope definition, (2) inventory analysis, (3) impact assessment, and (4) interpretation [25].

The goal and scope of an LCA study should be clearly defined and consistent with the application [26]. The goal of this LCA study is to identify the environmental impact of three PHA manufacturing processes from cradle to factory gate.

The LCI (life cycle inventory) analysis phase is related to the calculations and collection of data [26]. Because the processes studied here are still largely under development, many LCI data are rare and insufficient. Therefore, a number of reference sources and reasonable assumptions are used in the calculations, as detailed in Section 4. A model is established in a GaBi software system [27] for each of these three processes.

The life cycle impact assessment (LCIA) generally has several mandatory elements (choosing of characterization methodology and impact categories indicators, calculating of the indicator results, etc.) that convert LCI results to the collection of indicator results for different impact categories. There may be optional elements for weighting or normalization of the indicator results, etc. [28]. In this study, the Eco-indicator 99 methodology [29] and 10 indicators under human health, ecosystem quality and resources are chosen to be used.

An LCA study completes with the interpretation phase by evaluating the results of the LCIA of the product system and presenting them to meet the requirements described in the goal and scope of the LCA study [30]. In this study, results in the form of 10 indicators under human health, ecosystem quality, and resources are obtained, and they are presented and compared in Section 5 of this article.

4. LCA OF THREE PROCESSES

The following three scenarios are modeled and analyzed in this study:

Scenario 1: Microbial PHA manufacturing using glucose as carbon substrate;

Scenario 2: Microbial PHA manufacturing using cheese whey as carbon substrate;

Scenario 3: PHA manufacturing from transgenic corn.

4.1. Scenarios 1 and 2

The microbial PHA manufacturing has three stages: fermentation, recovery, and granules. At the fermentation stage, recombinant *E. coli* harboring the *R. eutropha* PHA synthesis genes is a good candidate [31]. This process accumulates PHA content greater than 8% of dry cell weight. *E. coli* can utilize various carbon sources including sucrose, lactose, glucose and xylose, which allows manufacturing of PHA from cheap agricultural byproducts such as whey, molasses, and hemi-cellulose hydrolystate. The fermentation is conducted under agitation and aeration in a jar fermenter. Steam is needed in the pretreatment for heat sterilization. The carbon source is fed continuously. Heat generated by aerobic oxidation is cooled by cooling water to keep the culture medium at a constant temperature. In this study, the energy consumed is electricity at agitation, fermentation temperature, aeration, and water cooling processes.

There are two methods for PHA recovery: by surfactant-hypochlorite digestion or by treatment with the dispersion of chloroform and hypochlorite [32]. The former method is considered in this study. In this method, surfactant solution is added to the fermentation broth, followed by hypochlorite digestion in a flow-through manner. PHA is separated by centrifugation from aqueous solution having the dissolved non-PHA cell material. During this process, energy consumption is electricity for centrifugation, washing, and high-pressure homogenization. Steam is consumed in the evaporation and spray drying process. The LCI of the fermentation process is given in the first two columns of Table 1 with data from Refs. [22, 32, 33]. After fermentation and recovery, the PHA granules are dried by a spray dryer to obtain the end product. The LCI for this process is summarized in the last two columns of Table 1 with data from Refs. [22, 32].

Carbon substrate is the main material in PHA manufacturing. Two manufacturing processes for the carbon substrate are assessed: glucose made from corn grains and cheese whey as the byproduct in cheese making.

4.1.1. Glucose Made from Corn Grains. During corn farming, carbon dioxide intake comes from the air. There is

TABLE 1.—LCI of fermentation and recovery processes [22, 32, 33].

Parameter	Data (per kg-PHA)	Parameter	Data (per kg-PHA)
Raw materials		Raw materials	
Glucose	3.768 kg	Triton-X100	0.153 kg
Water (excluding recycled cooling water)	261	NaOCl	2.961 kg
NH ₃	0.109 kg		
Whey	65.67 kg		
Energy and steam		Energy and steam	
Aeration	1.27 kWh	Evaporation	0.33 kg
Agitation	0.32 kWh	High-pressure homogenization	1.97 kWh
Media sterilization	0.45 kg	Spray drying	2.00 kg
Cooling water	0.76 kWh	Centrifugation and washing	0.50 kWh
Emission			
CO ₂	1.91 kg		

also carbon dioxide emission to the air through agronomic affairs due to the use of agrochemicals, such as herbicides, insecticides, fertilizer, and fuels. Corn wet mill is assumed to be at the same location of corn farming. By wet milling, corns are processed into glucose together with corn oil. This process consumes electricity and leads to carbon dioxide emissions. The LCI for glucose making is summarized in the first two columns of Table 2 with data from Refs. [21, 24, 34].

4.1.2. Cheese Whey Made from Cheese Making. Cheese whey is a waste made from cheese making and thus has a low price, making it a promising substitute to glucose as a carbon substrate in PHA manufacturing. Milk making by farming contains farm operation and animal food preparation [33, 35]. The LCI for whey making is summarized in the last two columns of Table 2 with the data from Ref. [34].

4.2. Scenario 3

Compared to relatively mature microbial PHA manufacturing, producing PHA from transgenic plants is at the lab-manufacturing stage. However, its potential superiority in environment and cost has attracted increasing research interest. The corn farming process is similar to that in the glucose manufacturing process, but the output is corn stover contained PHA. The corn stovers are transported to the PHA factory to produce PHA granules by extraction and compounding processes. C_4 – C_{11} alcohol is the chemical used for PHA extraction. The model built for C_4 – C_{11} alcohol is based on butanol manufacturing with the consumed amount of C_4 – C_{11} alcohol, and includes the energy requirements for compressed air [23]. The compounding step produces PHA granules for specific application requirements.

The LCI for corn stover is given in the first two columns of Table 3, and the LCI for extraction and compounding is summarized in the last two columns of Table 3 [23]. The dried stovers after PHA extraction can be burned for steam and electricity generation. The steam and electricity

TABLE 2.—LCI of glucose making and whey making [21, 24, 34].

Parameter	Data (per kg-glucose)	Parameter	Data (per liter milk)
Input			
CO ₂ intake from air	−1.47 kg	Milk from farm	1 L
Energy	2.5 MJ	Electricity	61.4 Wh
Nitrogen fertilizer	28.78 g	Water	$1.55 \times 10^{-3} \text{ m}^3$
Phosphorous fertilizer	12.72 g	Heat	0.49 Wh
Potassium fertilizer	14.37 g	Output	
Herbicides	0.57 g	Whey	0.876 kg
Insecticides	0.03 g	Yellow cheese	0.103 kg
Lime	5.28 g	Cream	0.070 kg
Diesel	0.157 MJ	COD	3.914 g
Gasoline	0.037 MJ	N	0.216 g
Liquid petroleum gas	0.21 MJ	P	0.082 g
Electricity	0.027 MJ		
Natural gas	0.047 MJ		
Output			
Corn	1.46 kg		
CO ₂ emission	0.15 kg		

TABLE 3.—LCI of corn stover, extraction, and compounding [23].

Parameter	Data	Parameter	Data (per kg-PHA)
Regular corn			
Harvest stover (dry basis)	5546 kg/ha	Raw PHA extraction	
Stover yield (dry basis)	9224 kg/ha	Stover (dry basis)	6.8 kg
		Stover residue after extraction	5.8 kg
Stover harvest rate	0.6 kg/kg	Electricity	8.8 MJ
Grain dry content	0.845 kg/kg	Stream	12.5 kg
Grain yield	8950 kg/ha	Alcohol (loss)	0.08 kg
Ratio of dry grain to dry stover	55/45 kg/kg		
Modified corn			
Corn grain	8503 kg/ha	PHA compounding	
Harvested stover (dry basis)	5269 kg/ha	Titanium oxide	0.003 kg
Grain yield loss	0.05 kg/kg	Electricity	2.4 MJ

can be reused in the PHA manufacturing to decrease the total energy consumption. Because emissions and energy conversion data for burning dried corn stover are not found, they are not considered in this work.

4.3. Basic Data

The electrical power grid for the above processes is estimated using power grid mix BUWAL. The steam consumption is estimated using the data of the polymerization plants (APME, one of steam producers). These data are available in the database of GaBi 4.0. Only transportation of the main materials (the glucose and cheese whey used in the fermentation and the corn stover used in the extraction process) is taken into account. Transportation of relatively minor materials (the chemicals such as NH₃ used in fermentation) is ignored.

5. RESULTS AND DISCUSSION

This work uses the Eco-indicator 99 methodology, which standardizes a set of indicators for calculating and characterizing the environmental damages and resource depletion. The environment termed as a set of physical, chemical, and biological parameters influenced by man is the conditions to the functioning of man and nature. These conditions are classified into three damage categories: ecosystem quality (acidification/nitrification, eco-toxicity), human health (carcinogenic effects, climate change, ozone layer depletion, radiation, respiratory [inorganic], respiratory [organic]), and sufficient supply of resources (fuels, minerals) [29]. Each category is represented by the relevant eco-indicators. GaBi 4 is used in this work to obtain results in the three categories using the processes and LCI data reported in Section 4.

Figures 1–3 show the relative contributions of impact factors in each damage category and compare the total impacts of the three PHA manufacturing scenarios reported in Section 4. In the human health damage category, the respiratory (inorganic) impact indicator dominates, and climate change ranks second. The other four indicators are relatively insignificant. In the ecosystem damage category, the acidification/nitrification impact indicator contributes most to the total impact. In the resource damage category,

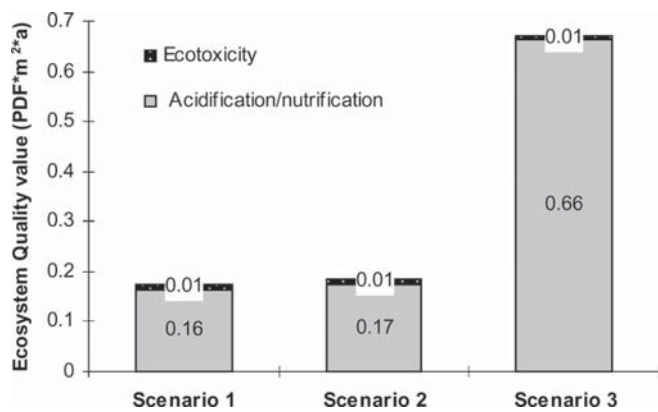


FIGURE 1.—Ecosystem damage values.

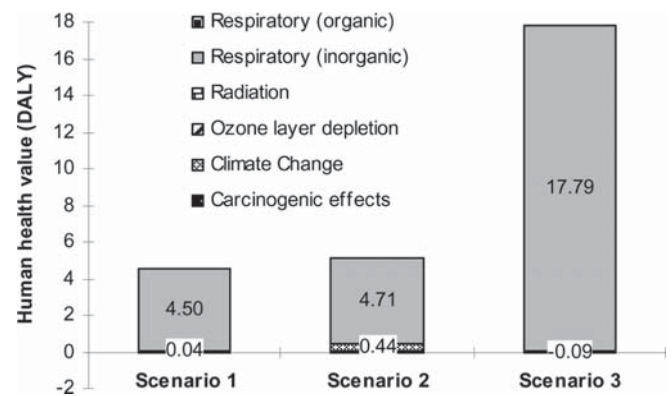


FIGURE 3.—Human health damage values.

the fossil fuel impact indicator dominates, compared with minerals. Scenarios 1 and 2 have similar results, while Scenario 3 has much higher impacts.

To identify the main contributors to the total impacts in the damage categories, the contribution percentages of the eco-indicators are calculated and are compared. The respiratory (inorganic), acidification/nitrification, and fossil fuels impact indicators are the major contributors causing the significant differences in the total impacts of the three scenarios in the three damage categories. As shown in Table 4, among the major contributors, the impact generated during the steam raising process is the most significant to the total impact of respiratory (inorganic) and acidification/nitrification, especially in Scenario 3. The electricity generation process has major impacts on the fossil

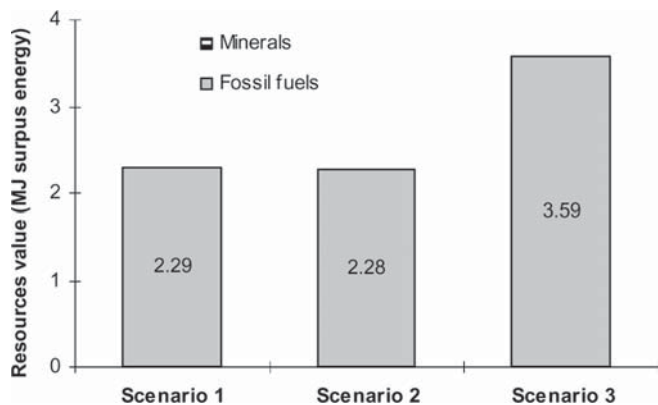


FIGURE 2.—Resource damage values.

fuels in Scenarios 1 and 2. For climate change, the carbon dioxide intake effects from the corn farming processes of Scenarios 1 and 3 can offset the carbon dioxide emission from other processes. The compensation effect is most obvious in Scenario 3.

6. CONCLUSIONS

- 1) The dominant contributor to the environmental burden in the PHA manufacturing processes investigated is energy, especially steam. High steam consumption leads to high impacts on fossil fuels, respiratory (inorganic), and acidification/nitrification.
- 2) With the current method, PHA manufacturing from transgenic plants is less beneficial over the full cradle-to-gate life cycle than microbial PHA manufacturing with glucose and whey.
- 3) However, it has superior performance in CO₂ emissions and energy consumptions. Its CO₂ intake at the corn forming stage can more than compensate its CO₂ emission, and its polymer extraction and compounding processes consume power much less than the fermentation and recovery processes in the microbial PHA manufacturing.
- 4) The main cause of its high life cycle impact is its high steam consumption, requiring 4.5 times more than that of microbial PHA manufacturing. Using renewable energy source would make PHA manufacturing from transgenic plants significantly better. One method is using the residual corn stovers after extraction for electricity and steam [23].

TABLE 4.—Contribution percentages (%) of major impact indicators.

	Fossil fuels		Respiratory (inorganic)		Acidification/Nitrification		Climate change		
	Steam	Power	Steam	Power	Steam	Power	Steam	Power	Farming
Scenario 1	27.6	62.9	86.3	13.3	89.2	10.3	33.5	128.6	-157.1
Scenario 2	27.7	63.2	82.5	12.7	84.1	9.7	3.3	12.7	72.5
Scenario 3	78.1	20.4	98.2	1.7	98.6	1.3	72.2	31.3	-204.7

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