Carbon Footprint Evaluation of Polylactic Acid Production via Process Simulation

Pichan Tantichaipakorn[1], *Peerada* Yodkaew[1], *Kewalin* Preesong[1], *Aleena* Ketiam[1], and *Prakorn* Kittipoomwong[1,*]
[1]Chemical Engineering Department, King Mongkut's University of Technology North Bangkok, 10800 Bangkok, Thailand

Abstract. This study examines the design process for producing Polylactic Acid (PLA) by optimizing its production using Aspen Plus®. The objective is to enhance the quality and purity of lactide by adjusting various parameters, such as temperature. The research findings have been verified against reported data, revealing that the quantities of essential substances measured at each production stage closely match those reported in existing literatures. A sensitivity analysis reveals that lowering reactor temperatures can reduce energy consumption in both the condenser and reboiler while maintaining the quality of PLA, leading to improved cost efficiency. Additionally, the study evaluates the carbon dioxide emissions resulting from PLA synthesis, which total approximately 656 kg of CO_2 per ton of PLA. Most emissions occur during the oligomerization phase. Therefore, reducing the operational temperature during this phase to 170°C can significantly decrease the carbon footprint of PLA production. This approach helps identify an optimal operational point that maximizes energy savings.

1 Introduction

Plastic waste is one of the pressing world issues since it has become an indispensable material for human activities. In particular, the heavy use of plastic bags and plastic as packaging materials creates a tremendous volume of plastic waste. Unfortunately, only a small fraction of plastics is treated and properly managed, such as through incineration to generate electricity. The remaining plastic waste undergoes a simple landfill option with little regard to the grave consequences.

There is a pressing need for large-scale plastic waste management to eliminate as much plastic as possible from landfills, which leads to environmental pollution by contaminating water resources and exacerbating greenhouse gas emissions [1]. Although plastic wastes are mostly recyclable, the recycling efforts have fallen short of keeping pace with the growing volume of plastic waste generated. A more practical approach is necessary to resolve the problematic issues stemming from non-degradable and high carbon footprint of petroleum-based plastics. This environmental crisis has prompted the need for alternative solutions, such as employing sustainable materials like bioplastics, which are biodegradable.

Polylactic acid offers an attractive solution because of its biodegradability and sustainable material sources. The fermentation of corn or cassava starch is typically the starting step to produce lactic acid, which serves as a precursor for PLA through a polymerization reaction. The use of sustainable sources serves as a significant advantage over the option of petroleum-based raw material, which also contributes to the production of greenhouse gases, with carbon dioxide emissions around 3 kg CO_2 per kilogram of plastic [2]. On the contrary, the use of sustainable raw materials enables PLA to complete a closed carbon cycle. In other words, PLA offers a smaller overall carbon footprint by offsetting the CO_2 generated during PLA production and other phases, through the reduction of CO_2 derived from using plant-based raw materials. In addition, by carefully designing and adapting an energy-efficient production process, the greenhouse gas emissions of PLA can be further decreased compared to conventional plastics. This is even before considering the biodegradability which makes it even more attractive than conventional petroleum-based plastics from a practical perspective. Nevertheless, a challenge persists in achieving efficient degradation of PLA in industrial composting conditions.

Process simulation is an indispensable tool for understanding the impact of various process variables on achieving high yields while minimizing carbon footprints. In case of process simulation of the polylactide synthesis via lactic acid polymerization, the computational tool can emulate the complex and evolving relationships between operating parameters, including temperature, pressure, and catalyst loading, and their corresponding effects on product yield and desired specifications, including molecular weight [3].

In another case of batch production of PLA for biomedical products, a process simulation has demonstrated the ability to provide insights into the reaction kinetics and operating variables such as reactor temperature, pressure, and reactant concentration, and the product yield [4]. The authors elaborated the advantage of the computational approach in analyzing and optimizing operating conditions to achieve product specification in the article [5].

The Aspen process simulator is a valuable tool for process simulation. It was utilized in a thesis study focusing on the production of PLA [6]. The study sought

*Corresponding author : prakorn.k@eng.kmutnb.ac.th

to improve the PLA manufacturing process. Aspen Polymer Plus was utilized to model and simulate the generation of lactic acid and the polymerization process. A dynamic model was developed to assess the stability and controllability of the process. Additionally, the heat operations within the process could be optimized using the Aspen Energy Analyzer to improve the overall heat exchanger network.

The life cycle analysis of polylactic acid offers important insights into the environmental effects associated with every phase of its life cycle. [7]. The two most critical environmental concerns are: 1) the cultivation of agricultural feedstock, such as sugarcane, and 2) the production process that converts this feedstock into PLA. Process simulation can significantly enhance production efficiency and energy usage, which in turn helps reduce CO_2 emissions and energy consumption. A comprehensive approach to process design can mitigate the carbon footprint associated with each production step while ensuring high yield and product quality. The study indicates that, in combination with efficient cultivation techniques, the overall environmental impact of polylactic acid can be substantially lowered, potentially leading to a carbon-neutral production process [8].

Process simulation serves as an effective tool for conducting a thorough design of the production process for polylactic acid. It allows for the optimization of operating conditions while also focusing on minimizing the carbon footprint associated with polylactic acid production. This research goal is to create a platform for a process simulation to analyze the operating parameters affecting polylactic acid yield, as well as the corresponding carbon dioxide emissions.

2 Methodology

There are two principal methods for producing polylactic acid: direct condensation and ring-opening polymerization. The condensation method presents challenges due to being an equilibrium reaction, which makes the removal of water during the process difficult. As a result, this limitation restricts the maximum attainable molecular weight of PLA [9]. Consequently, ring-opening polymerization has emerged as the favored method due to its capacity to provide enhanced control over the reaction and yield PLA with a greater molecular weight [10].

In the ring-opening polymerization method for producing polylactic acid, the process begins with the generation of lactide monomers. These monomers are then transformed into the final PLA product using a catalyst. The entire process is divided into three distinct stages: oligomerization, depolymerization, and polymerization. This structured approach enhances the efficiency and control over the production, resulting in high-quality PLA.

$$5(C_3H_6O_3) \rightarrow 3(H_2O) + C_6H_{10}O_5 + C_9H_{14}O_7 \quad (1)$$

In the initial oligomerization reaction, five molecules of lactic acid combine through a condensation process. During this reaction, three water molecules are eliminated, resulting in the formation of oligomers. This results in the formation of dilactic (L2) and trilactic (L3) oligomers, while water is produced as a byproduct of the reaction.

$$2(C_6H_{10}O_5) + 2(C_9H_{14}O_7) \rightarrow 5(C_6H_8O_4) + 4(H_2O) \quad (2)$$

In this stage of the process, dilactic and trilactic acid experience depolymerization, which entails the breaking of the ester bonds. When heat or a suitable catalyst is applied, these oligomers react to produce lactide and water as a byproduct. This reaction is generally reversible, allowing larger oligomers to break down into smaller lactide molecules. These smaller molecules can then be polymerized again, resulting in the formation of higher molecular weight compounds.

$$C_6H_8O_4 \rightarrow 2\,(C_3H_4O_2) \quad (3)$$

The last step in the procedure involves polymerizing lactide to produce polylactic acid. This polymerization is categorized as addition polymerization. In this process, lactide molecules break their ester linkages when they come into contact with a catalyst. Lactide units combine to form polylactic acid (PLA) through a chain-growth process that occurs under heat, often facilitated by an initiator and catalyst to commence polymerization.

The synthesis process of polylactic acid was simulated using Aspen Plus. Initially, data were collected on key chemical species such as lactic acid, oligomers, lactide, and PLA, with a focus on ensuring accuracy through the property libraries available in Aspen Plus. The Poly-NRTL model was employed to address reaction kinetics and equilibrium.

The polylactic acid production process, illustrated in Fig. 1., begins with lactic acid being dissolved in water and subsequently introduced into the primary reactor. In this reactor, a condensation reaction takes place, joining lactic acid molecules to form dilactic and trilactic oligomers, with water generated as a by-product. A portion of the oligomer products is returned to the reactor to enhance efficiency.

The oligomers are subsequently moved to a second reactor, where depolymerization happens at elevated temperatures under vacuum conditions, yielding lactide, a crucial precursor for PLA. The unreacted substances are separated; some are recycled, and a small portion is eliminated to avoid contamination.

Following this, the lactide mixture is processed through two distillation columns to eliminate any remaining lactic acid and water, resulting in refined lactide. This purified lactide is then placed into a third reactor for ring-opening polymerization, employing a catalyst to generate high-quality PLA. The entire procedure includes recycling loops to optimize conversion rates and enhance overall efficiency.

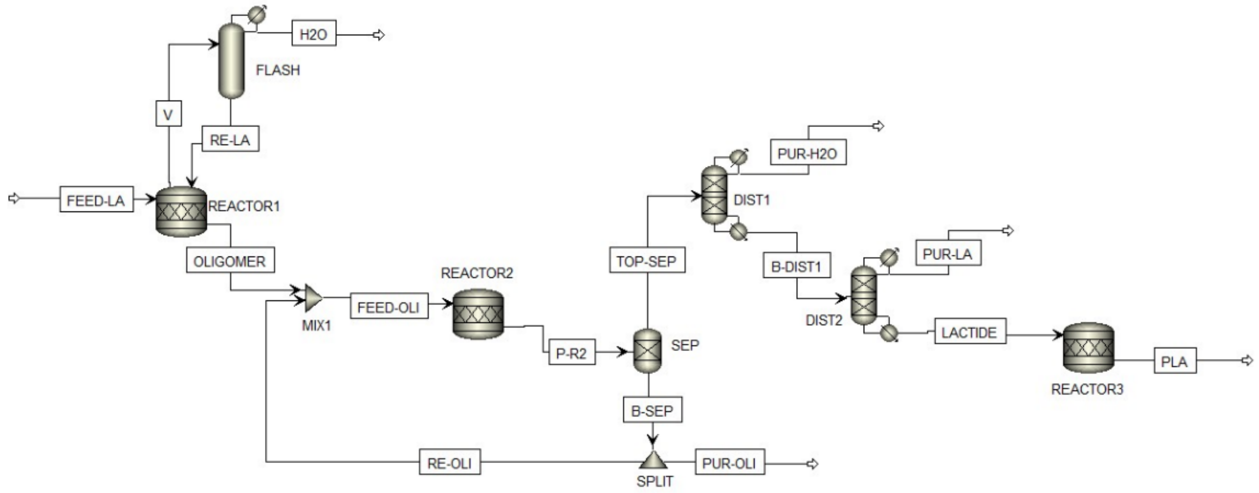

Fig. 1. Simulation flowsheet of the Polylactic acid synthesis process

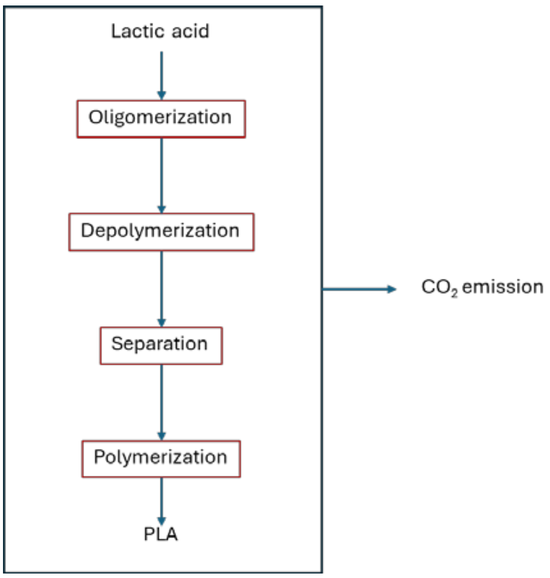

Fig. 2. System boundary

The research employs a Gate-to-Gate approach, focusing on the production process of Polylactic Acid (PLA) within the facility. It tracks the progression from the intake of Lactic Acid to the final PLA product, which is visually represented in Figure 2. To evaluate this process, a process simulator is utilized to analyze input and output balances while also measuring CO2 emissions. The study emphasizes the importance of accounting for carbon emissions related to energy consumption, particularly from grid electricity, as these contribute significantly to the facility's greenhouse gas footprint. Furthermore, it assesses emissions using natural gas as the primary energy source, highlighting the impact of heat and cooling (referred to as SCOPE 2 emissions) on the overall carbon footprint.

3 Result and Analysis

3.1 Process modelling of PLA production

First, the oligomerization undergoes in REACTOR1 where lactic acid is used as a feedstock to produce L2 and L3 oligomers at a rate of approximately 11,500 kg/h. Subsequently, the L2 and L3 are fed into REACTOR 2 for a depolymerization reaction, yielding lactide at a rate around 9,100 kg/h. Following this, two distillation units are employed to purify lactide, resulting in approximately 8,900 kg/h of refined product. Finally, refined lactide undergoes a ring-opening polymerization in REACTOR 3, generating polylactic acid at an output rate of about 8,900 kg/h. Overall, this synthesis process achieves a total yield of 77% in the production of PLA.

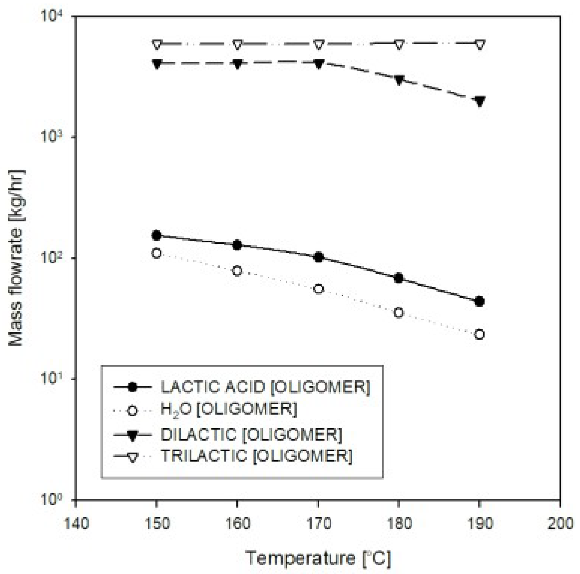

Fig. 3. Mass flow rate of various components in the OLIGOMER stream of REACTOR1 as a function of temperature.

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The production rate of components from REACTOR1 exhibits a clear temperature-dependent behavior, as illustrated in Figure 3. As the temperature increases from 150°C to 190°C, there is a notable reduction in the levels of unreacted lactic acid and water, signaling improved conversion efficiency and effective water removal. The flow rate of dilactic acid initially rises, peaking at 4100 kg/h at 170°C, but subsequently declines sharply to around half that amount by 190°C. In contrast, the flow rate of trilactic acid continues to increase with temperature, rising from 5892 kg/h to 5942 kg/h. This suggests that while higher temperatures favor the production of longer-chain oligomers, excessively high temperatures can adversely affect the yield of shorter-chain products such as dilactic acid.

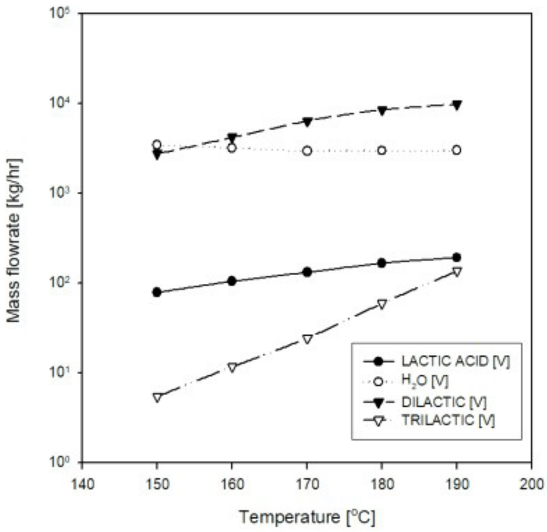

Fig. 4. Mass flow rate of various components in the V stream of REACTOR1 as a function of temperature.

Stream "V" primarily comprises L2 oligomers and water, with an increasing concentration of dilactic acid at elevated temperatures, indicating enhanced efficiency of the condensation reaction as illustrated in Figure 4. Meanwhile, the "OLIGOMER" stream is predominantly made up of dilactic and trilactic acids, with trilactic becoming more prevalent at temperatures exceeding 170°C. However, at these higher temperatures, the quantity of dilactic acid significantly decreases due to its evaporation into stream "V," resulting in a reduced mass in the "OLIGOMER" stream. This loss poses a challenge as it diminishes the availability of reactants for further reactions. Therefore, to minimize product loss and optimize process efficiency, it is recommended to maintain the reactor temperature at 170°C.

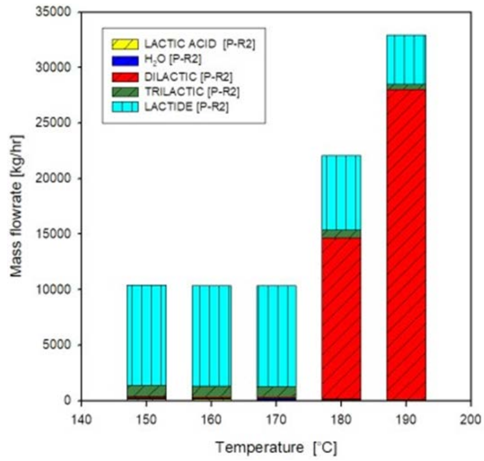

Fig. 5. Mass flow rate and component distribution in the P-R2 stream of REACTOR2 as a function of temperature.

The operating conditions of oligomerization in REACTOR1 significantly impact the compositions present in both REACTOR2 and REACTOR3, with temperature playing a crucial role in this process. It is essential to maintain an operational temperature between 150 and 170°C to stabilize oligomer concentrations. However, an increase in temperature to the range of 180 to 190°C leads to substantial changes, primarily due to an uptick in the recirculation of dilactic acid. Furthermore, at these elevated temperatures, the flow rate of the "RE-OLI" recycled stream, which returns unreacted L2 and L3 to REACTOR2, decreases, indicating an effective conversion process.

As illustrated in Figure 5, the increase in temperature correlates with a rise in the stream P-R2 from REACTOR2. This aligns with anticipated feed patterns and supports the overall mass balance continuity. Nonetheless, the higher temperature conditions result in an increased presence of trilactic acid, suggesting that some incomplete conversion may occur. This aspect necessitates careful monitoring during the downstream separation processes.

Recent validation against prior studies shows that substance quantities remain consistent throughout different production stages. Specifically, the mass flow rate of OLIGOMER from REACTOR1 corresponds well with earlier observations [5], confirming OLIGOMER as the primary component, alongside smaller amounts of lactic acid and water. Our model predicts a water mass flow rate of 55 kg/h, notably lower than the 72 kg/h reported in other studies, indicating improved efficiency in water removal. Furthermore, as depicted in Figure 6, our model achieves an oligomer production rate of approximately 10,000 kg/h, which exceeds the reported production figures of 8,469 kg/h and 8,531 kg/h from previous research [3, 6]. These findings underscore the advantages of optimized reaction conditions, resulting in increased oligomer yields and decreased levels of unreacted lactic acid in the system.

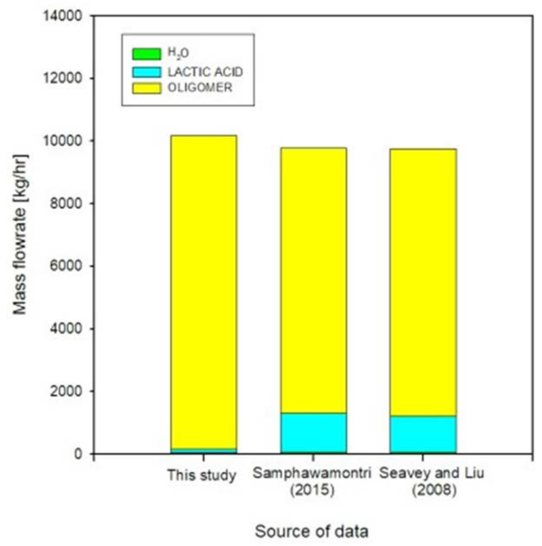

Fig. 6. Comparison of mass flow rates and component distributions in oligomerization across various studies.

In this study, the depolymerization step focused on optimizing lactide production while reducing the formation of undesirable byproducts. The outcomes achieved were found to be comparable to those reported in two other studies as shown in Figure 7. Notably, the absence of water and lactic acid during the process indicates a more efficient separation method. However, the detection of some oligomers suggests potential inefficiencies within the depolymerization process or the associated distillation units. Although the yield of lactide obtained in this study is lower than that found in other literature, the purity of the product is notably higher, underscoring an improvement in product quality.

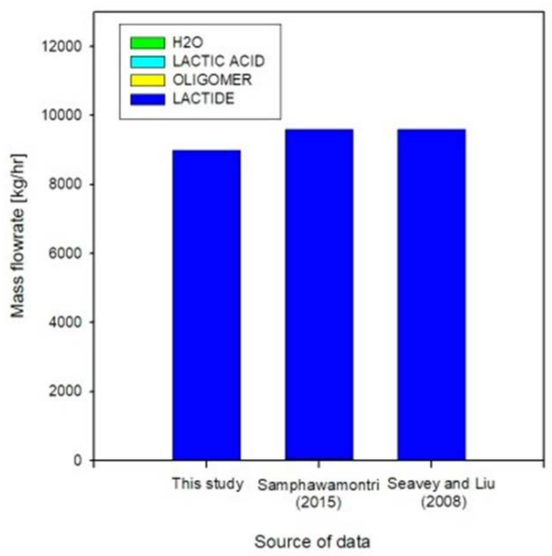

Fig. 7. Comparison of mass flow rates and component distributions in depolymerization across various studies.

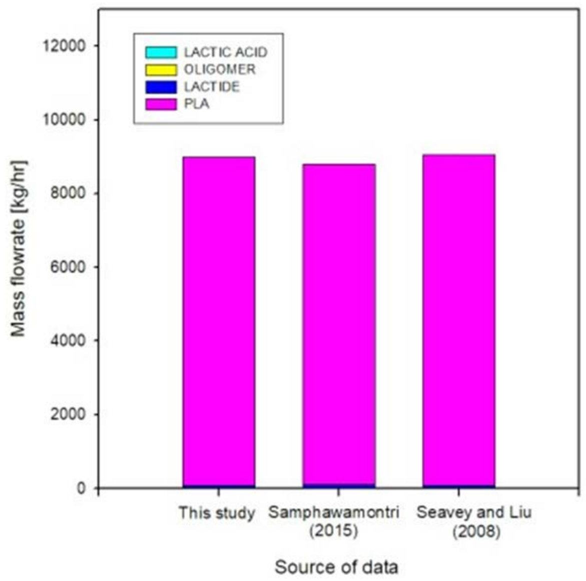

Fig. 8. Comparison of mass flow rates and component distributions in ring-opening polymerization across various studies.

The ring-opening polymerization phase effectively produces polylactic acid, yielding 90 kg/h of lactide and 8,896 kg/h of PLA. These outcomes are consistent with previous research, such as that conducted by Samphawamontri [6], which reported lactide residuals of 93 kg/h and a PLA yield of 8,693 kg/h. Additionally, Seaviy and Liu [3] documented a lactide production rate of 95 kg/h alongside a PLA yield of 8,963 kg/h. As shown in Figure 8, our process notably distinguishes itself by generating a clean product stream that eliminates lactic acid and oligomers, emphasizing the significance of optimized reaction conditions and separation processes. While our lactide content is slightly lower, our model demonstrates a promising approach for synthesizing high-purity PLA.

3.2 Carbon footprint evaluation of polylactic acid production

An analysis of CO_2 emissions associated with the various stages of polylactic acid production reveals that oligomerization is the largest contributor, emitting 274 kg of CO_2 per ton of PLA, as shown in Figure 9. This significant emission is primarily due to the high energy requirements for heating and maintaining the necessary reaction conditions. Following oligomerization, the depolymerization phase generates 169 kg of CO_2 per ton, driven by the energy needed to convert oligomers into lactide. The distillation process contributes the least, accounting for 72 kg of CO_2 per ton, while the polymerization phase emits 141 kg of CO_2 per ton, stemming from the energy-intensive thermal and catalytic processes involved. These findings suggest that improving energy efficiency—such as optimizing the operating temperature in Reactor 1—could significantly reduce the overall carbon footprint of PLA production.

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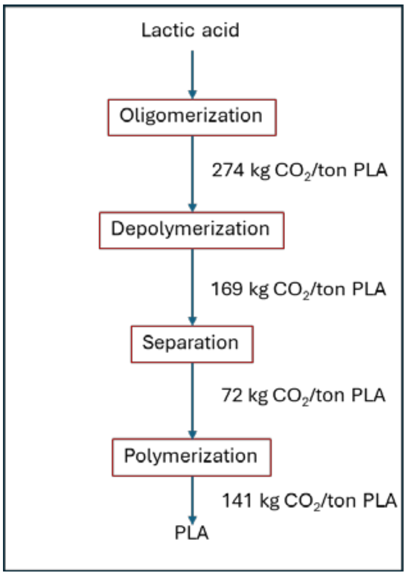

Fig. 9. CO_2 emissions across various stages of PLA production.

The simulation results indicate that oligomerization temperature significantly influences both CO_2 emissions and lactide production as illustrated in Figure 10. An optimal temperature of around 170°C or lower was identified, where both CO_2 emissions and lactide output consistently increase. However, temperatures above 170°C led to a notable increase in CO_2 emissions and a sharp decrease in lactide production. This decline is attributed to the reduced formation of shorter-chain products, like dilactic acid, during the oligomerization reaction at higher temperatures, negatively affecting the overall production of lactic acid and polylactic acid.

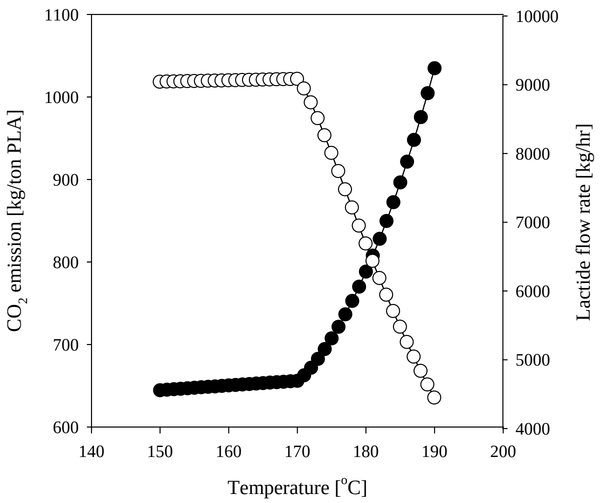

Fig. 10. The overall CO_2 emissions and mass flow rate of lactide in relation to oligomerization temperature.

At an oligomerization temperature of 170°C, the production of polylactic acid results in approximately 656 kg of CO_2 emissions per ton. This figure aligns closely with a previously reported estimate of 691 kg CO_2 per ton [7]. The slight difference in these estimates can be attributed to the current study's use of a natural gas-based energy source, which produces less CO_2 compared to the fossil fuel utilized in the earlier research. It is important to note that these findings originate from idealized conditions and may not fully capture the complexities encountered in real-world scenarios. Factors such as energy losses, equipment wear, and variability in operations could potentially increase emissions. Despite this, the consistent results from both studies enhance the credibility of the simulation methods and the analysis of CO_2 emissions conducted.

4 Summary

The study investigates the synthesis of polylactic acid (PLA) using Aspen Plus process simulation software, highlighting its potential for industrial applications. It outlines a three-step reaction process comprising oligomerization, depolymerization, and ring-opening polymerization, ultimately yielding a high-purity PLA output of 77%, which is consistent with current benchmarks. A significant aspect of the findings is the optimization of reactor temperatures, particularly during the oligomerization phase, which proves to be most effective at 170°C. This temperature not only enhances product yield but also minimizes energy losses and prevents the degradation of raw materials. From an environmental perspective, the oligomerization stage plays a pivotal role in reducing the carbon footprint. The analysis indicates CO_2 emissions of approximately 656 kg per ton of PLA, in line with existing literature. Overall, the study underscores the importance of integrating process simulation with CO_2 emission analysis, reinforcing the value of such models in the evaluation of industrial processes.

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References

1. R. Geyer, J. R. Jambeck, K. L. Law, Production, use, and fate of all plastics ever made. Sci Adv. **3**, e1700782 (2017). https://doi.org/10.1126/sciadv.1700782
2. E. T. H. Vink, K. R. Rabago, D. A. Glassner, P. R. Gruber, Application of life cycle assessment to Nature Works™ polylactide (PLA) production, Polymer Degradation and Stability. **80**, 403 (2003). https://doi.org/10.1016/S0141-3910(02)00372-5.
3. K. C. Seavey, Y. A. Liu, Polylactide and Nylon-6,6 Polymerization Process Modeling in Polymers Plus. In Step-Growth Polymerization: Process Modeling and Product Design, (John Wiley and Sons, New Jersey, 2008)

4. G. Martinez, A. Lasprilla, B. Lunelli, A. Jardini, R. Filho, A computer tool for the development of poly (lactic acid) synthesis process from renewable feedstock for biomanufacturing. Comp. Aid. Chem. Eng. **29**, 346 (2011).
   http://dx.doi.org/10.1016/B978-0-444-53711-9.50070-5
5. G. Martinez, A. Lasprilla, M. Barbosa, A. Jardini, R. Filho, Modeling and simulation of Poly-lactic acid synthesis in batch process for biomedical applications. Computer Aided Chemical Engineering. **30**, 997 (2012).
   http://dx.doi.org/10.1016/B978-0-444-59520-1.50054-3
6. Y. Samphawamontri, Process Modeling and Simulation of Polylactic Acid, Master thesis, Kasetsart University, Department of Chemical Engineering (2015)
7. A. Morão, F. Bie, Life Cycle Impact Assessment of Polylactic Acid (PLA) Produced from Sugarcane in Thailand, Journal of Polymers and the Environment, **27**, 2523 (2019).
   https://link.springer.com/article/10.1007/s10924-019-01525-9
8. U. Suwanmanee, T. Leejarkpai, Y. Rudeekit, T. Mungcharoen, Life cycle energy consumption and greenhouse gas emissions of polylactic acid (PLA) and polystyrene (PS) trays, Kasetsart Journal - Natural Science. **44**, 703 (2010)
9. L. Mou, J. Li, Y. Lu, G. Li, Juan Li, Polylactic acid: A future universal biobased polymer with multifunctional performance from monomer synthesis, and processing to applications: A review. J. of Hazardous Materials Advances. **18**, 100757 (2025).
   https://doi.org/10.1016/j.hazadv.2025.100757
10. M. C. D'Alterio, I. D'Auria, L. Gaeta, C. Tedesco, S. Brenna, and C. Pellecchia, Are Well Performing Catalysts for the Ring Opening Polymerization of l-Lactide under Mild Laboratory Conditions Suitable for the Industrial Process? The Case of New Highly Active Zn (II) Catalysts. Macromolecules. **55,** 5515 (2022).
    https://doi.org/10.1021/acs.macromol.2c00719
11. Rahmayetty, Y. Whulanza, Sukirno, S. F. Rahman, E. A. Suyono, M. Yohda, and M. Gozan, Use of Candida rugosa lipase as a biocatalyst for L-lactide ring-opening polymerization and polylactic acid production, Biocatalysis and Agricultural Biotechnology. **16**, 683 (2018).
    https://doi.org/10.1016/j.bcab.2018.09.015