In silico Characterization of Natural Blending Agents in Conjunction with Polyhydroxybutyrate (PHB) to Solicit Biopolymer film with Improved Thermal Stability

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Abstract

Polyhydroxybutyrate (PHB) is known to exhibit limitations in terms of thermal stability. Its susceptibility to thermal degradation, particularly when exposed to high temperatures, can hinder its performance in various applications. This study addresses the thermal stability limitations of Polyhydroxybutyrate (PHB) by exploring strategies to enhance its performance through blending with biodegradable polymers. Focusing on the PHB-pectin blend, computational simulations unveil a remarkable 346.92 K increase in the glass transition temperature (Tg), surpassing pure PHB. This elevation indicates strong intermolecular interactions, potentially involving hydrogen bonding, leading to a more rigid and thermally stable polymer system. The heightened Tg has practical implications for critical applications, such as automotive components and electronic devices, where resistance to elevated temperatures is essential. Emphasizing environmental benefits, the study highlights the use of biodegradable and bio-based polymers as eco-friendly alternatives to traditional plastics, contributing to reduced plastic waste. Overall, this research underscores the potential of thermally stable polymers to both enhance material performance and address environmental concerns across diverse industries.

Keywords: Polyhydroxybutyrate, Biodegradable plastics, Thermal stability

1. Introduction

Plastic pollution has extensive and harmful repercussions on Earth's ecosystems, presenting a significant danger to biodiversity, habitats, and human well-being. As plastic waste accumulates, particularly in oceans and waterways, it wreaks havoc on marine life. Marine animals often get entangled in plastic debris or mistake it for food, leading to injuries, suffocation, and starvation (Azevedo-Santos et al., 2021). This disturbance in ecosystems results in the loss of biodiversity and the deterioration of aquatic

environments. Moreover, plastics degrade into microplastics (MPs), contaminating soil, water, and even the air we breathe. These minute particles accumulate in the food chain, potentially reaching humans through the consumption of contaminated seafood and water. While the long-term effects of microplastic exposure on human health are still under study, concerns exist regarding potential health risks. Micro/nano-plastics may pose acute toxicity, (sub)chronic toxicity, carcinogenicity, genotoxicity, and developmental toxicity (Sajjad et al., 2022) . Plastic pollution also adversely affects agricultural lands, impeding plant growth, contaminating crops, and disrupting nutrient cycles. Additionally, plastics release toxic chemicals as they degrade, polluting water bodies and posing risks to aquatic life and human well-being. The production, disposal, and incineration of plastics contribute to climate change by releasing greenhouse gases and impeding carbon sequestration processes. The economic costs associated with plastic pollution, including waste management, cleanup efforts, and damage to industries, further compound its impact on societies. Addressing plastic pollution necessitates concerted global efforts to reduce plastic consumption, enhance waste management practices, promote recycling, and develop sustainable alternatives to plastic (R. Kumar et al., 2021).

Polyhydroxybutyrate (PHB) emerges as a promising alternative to conventional plastics. PHB is a biopolymer, a type of polyester, that is naturally produced by certain bacteria through fermentation processes. What makes PHB particularly appealing as a substitute for traditional plastics is its biodegradability and sustainability (Adnan et al., 2022). Unlike traditional plastics derived from fossil fuels, PHB is a bio-based polymer that can be produced from renewable resources, such as plant sugars or agricultural by-products. When disposed of, PHB can break down naturally, leading to a reduced environmental impact compared to persistent and non-biodegradable plastics (McAdam et al., 2020). This characteristic makes PHB an attractive option for addressing the ecological challenges posed by plastic pollution and promoting more environmentally friendly practices in various industries. As research and technology continue to advance, PHB and other biodegradable alternatives have the potential to play a crucial role in mitigating the adverse effects of plastic waste on our planet. One notable drawback of Polyhydroxybutyrate (PHB) is its limited processability. PHB has a tendency to be more brittle and less flexible than traditional plastics, which can pose challenges in certain manufacturing processes (McAdam et al., 2020). The processing temperature range for PHB is also relatively narrow, making it less adaptable to certain industrial production methods. Overcoming these challenges in terms of processability is a key focus in research and development efforts aimed at enhancing the viability of PHB as a widespread alternative to traditional plastics (Moshood et al., 2022b).

Blending agents can be strategically chosen and tailored to achieve specific property improvements in polymer blends, including changes in glass transition temperature, which is critical for applications requiring specific thermal and mechanical characteristics (de Beukelaer et al., 2022). These are organic substances derived from renewable sources, such as plant extracts or biopolymers, that are strategically chosen to enhance the properties of Polyhydroxybutyrate (PHB). These agents serve as modifiers when combined with PHB, contributing to the overall improvement of its mechanical, thermal, and processing characteristics (De Sousa Junior et al., 2022). Additives such as plasticizers, nucleating agents, and elastomers improve ductility, toughness, and crystallization rate. Bio-based additives can enhance biodegradability, while processing aids and compatibilizers improve processability. Blending agents can also reduce costs and enhance thermal stability (Fredi & Dorigato, 2023).

Polyhydroxybutyrate (PHB) blending, traditionally done in vitro through methods like melt blending and solution blending, involves controlled mixing to create desired material blends. Melt blending heats PHB to its melting point, incorporating additional PHB or modifiers during the molten state (Zhang & Thomas, 2009). Solution blending dissolves components in a common solvent, yielding a blended material after solvent removal. While valuable, these in vitro methods are time consuming and expensive which could be complemented by emerging *in silico* approaches. Computational modeling, including molecular dynamics simulations which predicts and analyzes PHB blend properties without extensive lab work. *In silico* methods accelerate screening for potential blending agents, providing insights into their impact on PHB's thermal stability. This approach enhances efficiency in exploring and understanding molecular mechanisms governing PHB blend thermal behavior. Since most of the research work fails to find thermal stability of a polyhydroxybutyrate using computational ways, this investigation is an attempt to know thermal stability of a polymer by integrating computational methods. Today, PHB remains a promising biodegradable polymer with potential applications across various industries, offering an environmentally-friendly alternative to traditional petroleum-based plastics, particularly in single-use and short-lifecycle applications (Moshood et al., 2022a). The enhancement of thermal properties in polymer mixtures is a critical focus, particularly in understanding the glass transition temperature (Tg). Tg is an intrinsic characteristic of polymers, representing the temperature at which a polymer transitions from a glassy to a rubbery state, leading to significant changes in its properties. Determining Tg experimentally is a time-consuming and costly process. Molecular Dynamics (MD) emerges as a powerful tool for investigating the atomic-scale behavior of polymers (Zheng et al., 2022). In this study, we explore into the glass transition temperature of Polyhydroxybutyrate (PHB) and various blending agents. This investigation aims to identify a blending agent or compound that not only demonstrates improved thermal stability but holds promise for enhancing the overall performance of the polymer mixture (Roudaut et al., 2004).

2. Methods

All the simulations were carried out using the Materials Studio software package of Accelrys based on the molecular dynamics method. In this study, polymer-based systems including PHB, PHB /Pectin, PHB/Alginate , PHB/Gelatin, PHB/Cellulose, PHB/Xanthan Gum, PHB/Starch, PHB/Natural Rubber and PHB/Terpenes were studied (Martău et al., 2019) (V. Kumar et al., 2021) (M, 2014). Various steps for knowing the Tg of Polymer blend are as follows:

2.1. Modelling of PHB molecular chains and different blending agents

The appropriate repeating unit is critically important for polymer in all simulations, which is related to the accuracy of the simulation and the reliability of the calculation. Generally, the more repeating units lead to the more realistic polymer. However, due to the limitation of computer resources, a longer chain may lead to a longer simulation time. Different research paper indicates molecular chain with 50 repeat units can be used for molecular dynamic simulation (Wei et al., 2019).

2.2. Modelling of PHB and blending agent system

The amorphous cell module was applied to build a PHB-blending agent model. In this work, we built different blend systems by changing polymer chain of blending agents and keeping the same polyhydroxybutyrate chain. In order to ensure that the polymer molecules had enough space for relaxation and to avoid the overlapping of chain entanglement, the initial densities of all the systems were set to 1 g/cm^3. This creates a simulation box of polymer blend (Gartner & Jayaraman, 2019).

2.3. Geometry optimization

Geometry optimization was performed via the smart minimization method. The convergence tolerance was applied using an ultra-fine with an energy convergence of 2×10^{-5} kcal/mol and displacement of 5×10^{-5} Å, and the number of iterations considered was 10000. The COMPASS force fields by group based electrostatic and atom based Vander Waals summation methods at the ultra-fine level of quality were applied to all the simulations. The purpose of this process is to eliminate the undesirable contacts and attain the lowest energy state (Dong et al., 2021).

2.4. Verification of Equilibration

Verification of equilibration in molecular dynamics simulations involves assessing whether a simulated system has reached a stable state. The temperature (T) and energy (E) are employed to judge the equilibrium of the simulated system. If the T & E are constant or fluctuate up and down only between 5% and 10%, it means that the blend system has reached equilibration (Kubitzki & de Groot, 2007).

2.5. Density Analysis at Varying Temperatures

Starting from 100 K and increasing in steps of 30 K to 500 K, 200 ps NPT simulations was performed at each temperature point. All simulation herein obeyed the following criteria i.e., Nose-Hoover-Langevin (NHL) thermostat and Berendsen barostat was utilized to control the temperature and pressure for all NPT simulations. The non-bonded Vander Waals interactions was calculated by the group-based method and the electrostatic interactions are calculated by an atom-based method with the cutoff distance of 15.5 Å. The spline width is 1 Å, and the buffer width is 0.5 Å. At the same time, three parallel runs are calculated for each case, and the results are averaged to reduce the uncertainty (Dong et al., 2021) (Guo et al., 2019).

3. Results and discussion

3.1. Building polymer chains

Further, the homopolymer chain of PHB (Figure 1) and various blending agents was created using material studio software (PubChem and Molview was used to import 3D structure of the poymers) which gave a result as:
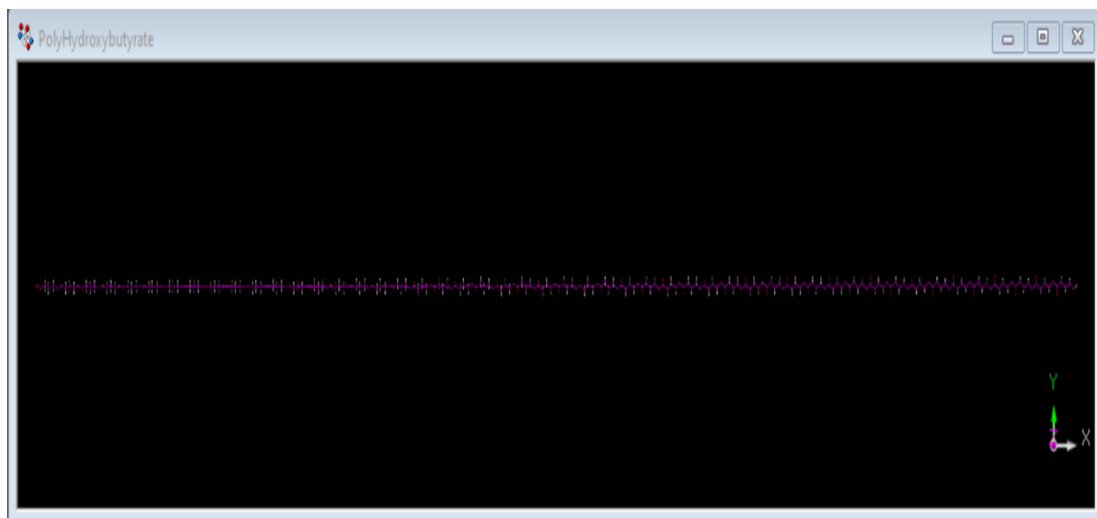

Figure 1: PHB Polymer with chain length 50; these are then used as input to the property prediction and simulation modules of Materials Studio.

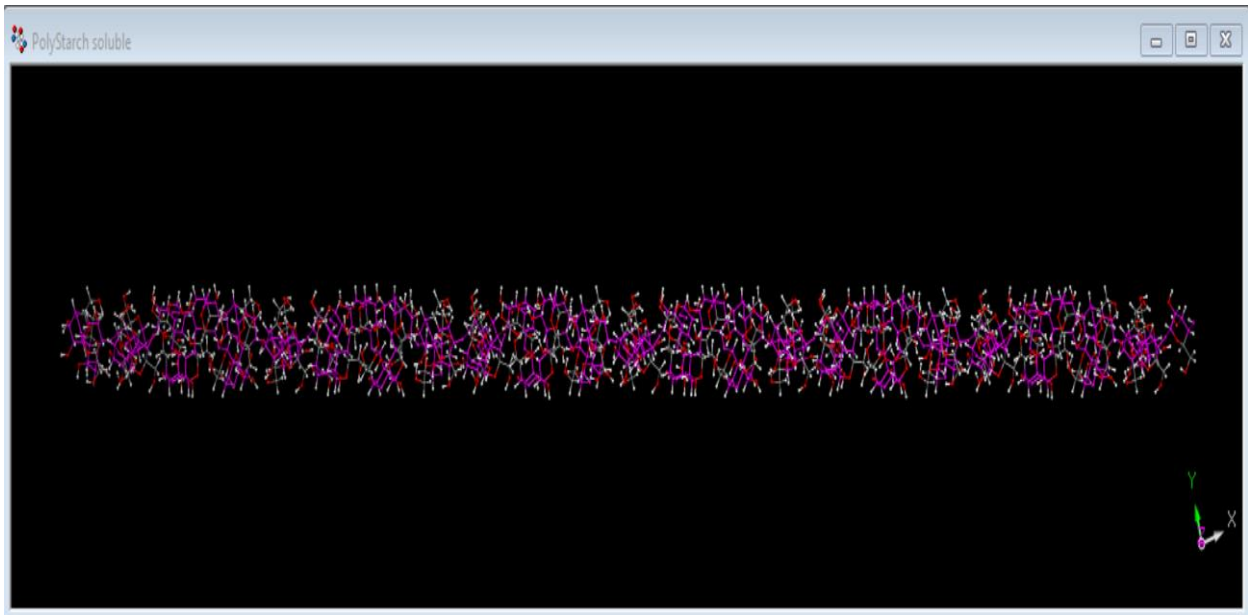The homopolymer chain of 3D structure of one the blending agent i.e. starch was created (Figure 2).

Figure 2: 3D Structure of Starch with chain length 50; Chains can be constructed from repeat units built using the Sketcher, taken from the extensive repeat unit library or by importing desired structures Similarly the polymer chains of all other blending agents like pectin, alginate, cellulose, starch, gelatin, terpene, fibrin, cellulose etc. were prepared.

3.2. Creation of Simulation Boxes

The amorphous cell module was used to build a PHB-blending agent model. In this work, we built different blend systems by changing polymer chain of blending agents and keeping the same PHB chain. In order to ensure that the polymer molecules had enough space for relaxation and to avoid the overlapping of chain entanglement, the initial densities of all the systems were set to 1 g/cm^3. This creates a simulation box of polymer blend. Then equilibrated simulation boxes were used for further studies (Figure 3).
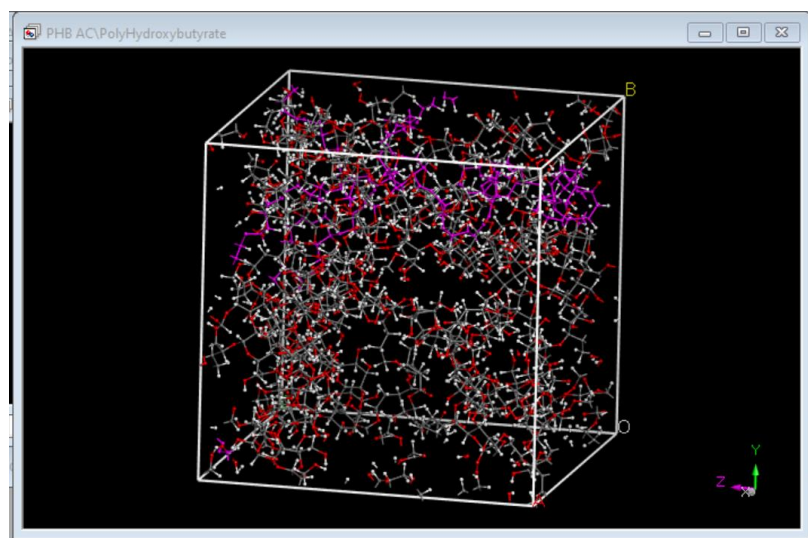

Figure 3: Equilibrated simulation boxes including PHB and starch; In this study, we created blend systems by altering the polymer chains of blending agents while maintaining a consistent polyhydroxybutyrate chain.

3.3. Geometry Optimization

Geometry optimization was the next step. Before starting the simulation, the energy of the system's computational cell needs to be minimized. The "**Smart Minimizer**" method is a computational technique used to find the lowest-energy configuration of the system (Jing et al., 2022) . Here is the image of pop-up which appears when geometry optimization completes.
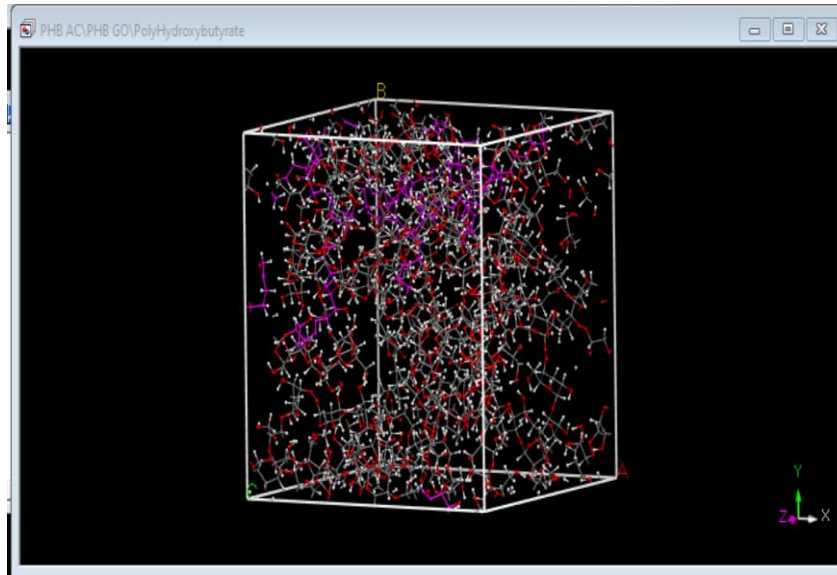

Figure 4: Geometry optimization was done to get a structure with stable configuration; this process is crucial for removing unfavorable contacts and achieving the lowest energy state, which represents a stable configuration of the models

3.4. The Glass Transition (Tg)

The glass transition temperature (Tg) serves as a pivotal parameter influencing the properties and potential applications of polymers. Currently, the free volume theory of the glass transition, originally proposed by Fox and Flory, remains the most widely adopted explanation for this phenomenon. At room temperature, polymer chains are in a frozen state due to the lower energy associated with chain segment motion. Notably, the variation in free volume is minimal with a temperature increase below Tg (Zheng et al., 2022). as the system temperature surpasses Tg, the polymer's free volume experiences a swift expansion, triggering a transition from a stationary to a motion state. Tg marks the temperature at which the polymer system undergoes a sudden and significant change in free volume. Typically, the specific volume versus temperature curve closely mirrors that of free volume versus temperature. Consequently, the specific volume versus temperature serves as a useful indicator for determining the inflection point when the polymer undergoes the transition from a rubbery to a glassy state upon cooling. The specific volume versus temperature curve for the pure polyhydroxybutyrate phase is depicted in Figure 5. With the visual inspection of the computed data points, the inflection point of Tg is obtained by a rough estimation (Zheng et al., 2022) (Kalogeras & Brostow, 2009).

Subsequently, the computed data points are segregated into two categories, aligning with the regions below (glassy) and above (rubbery) this approximation. Each segment of the computed dataset undergoes linear regression to establish a straight line. The point of inflection is determined by the intersection of these two regression lines. The plot depicting specific volume versus temperature for pure Polyhydroxybutyrate exhibits a noticeable break indicative of vitrification. Figure 5 illustrates that the

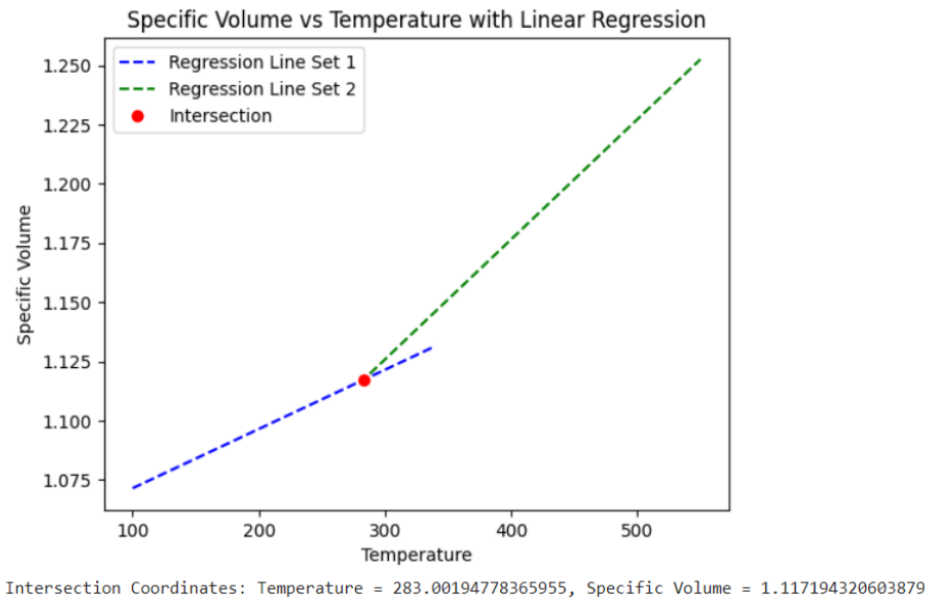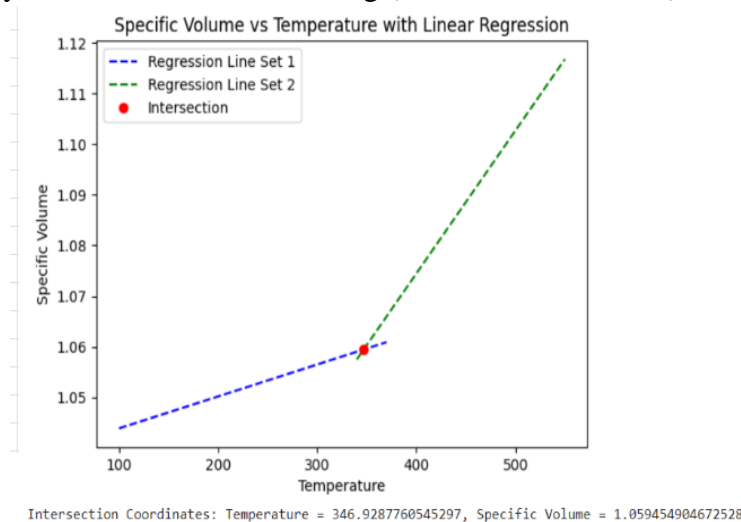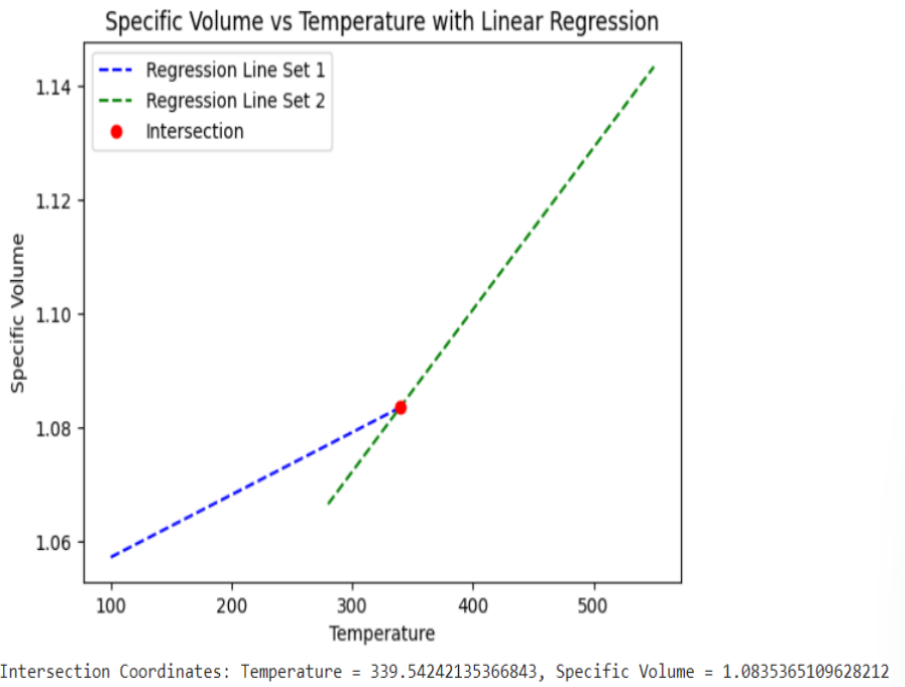glass transition temperature (Tg) of pure PHB, as determined through MD simulation, stands at 283.00 K.

Figure 5: Specific volume vs temperature graph showing glass transition temperature of PHB

The glass transition temperature of the PHB is obtained by the intersecting point which is 283.00 K (Figure 5). It is slightly higher than the experimental value of the PHB that is 275-279K. Comparing with the MD simulations, the possible reason is that an excess free volume in the vicinity of the PHB surface induced by the preferential segregation of chain end group of PHB brings about a lower experimental Tg value.
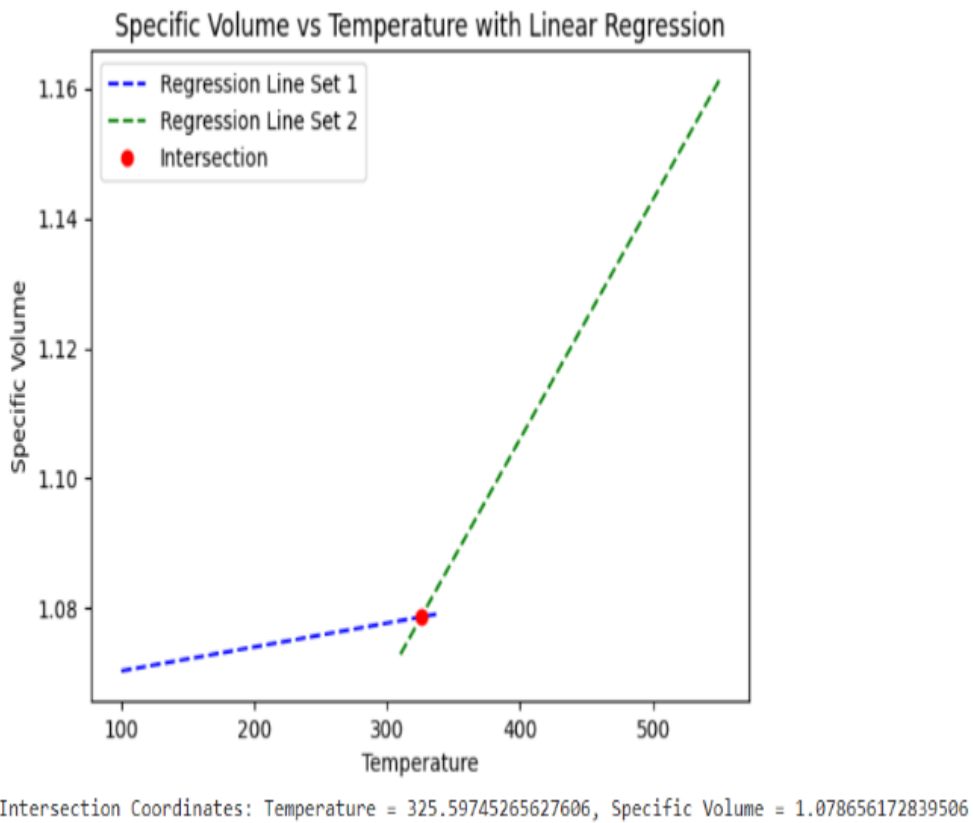
As the method of calculation of glass transition temperature of polymer mixture is the same as that of simple polymer, the same procedure was performed on PHB and its blends. The next graph shows the glass transition temperature of the PHB-Pectin blend is 346.92 K which is significantly higher than the glass transition temperature of the PHB i.e. 275-279K. This predicts that this blend can have the increased thermal stability as a result of increase in Tg (McAdam et al., 2020).
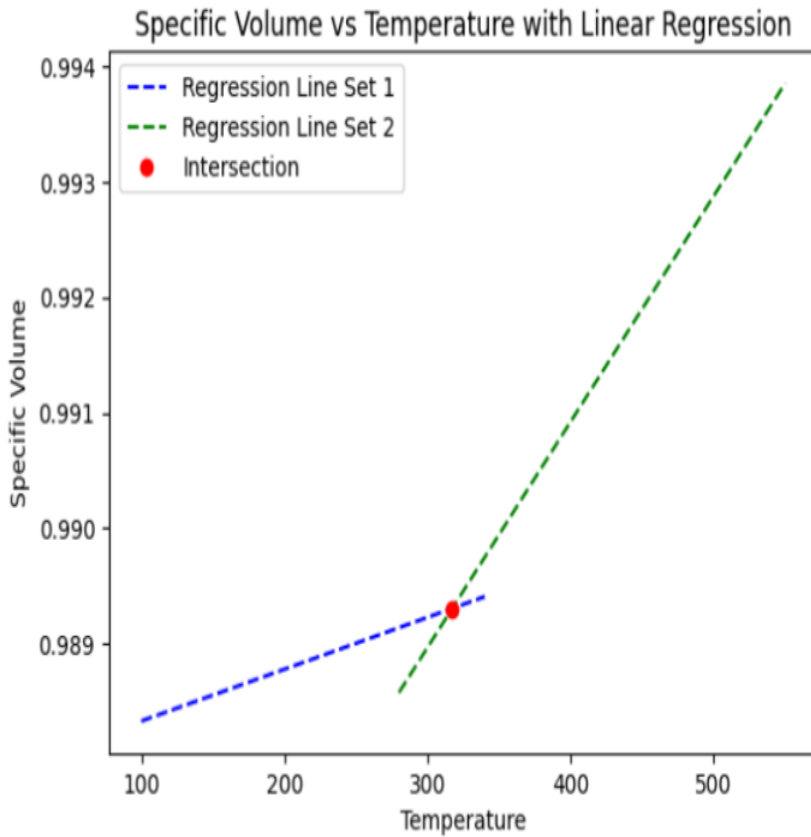

(a)

Intersection Coordinates: Temperature = 339.54242135366843, Specific Volume = 1.0835365109628212

(b)

Intersection Coordinates: Temperature = 325.59745265627606, Specific Volume = 1.078656172839506
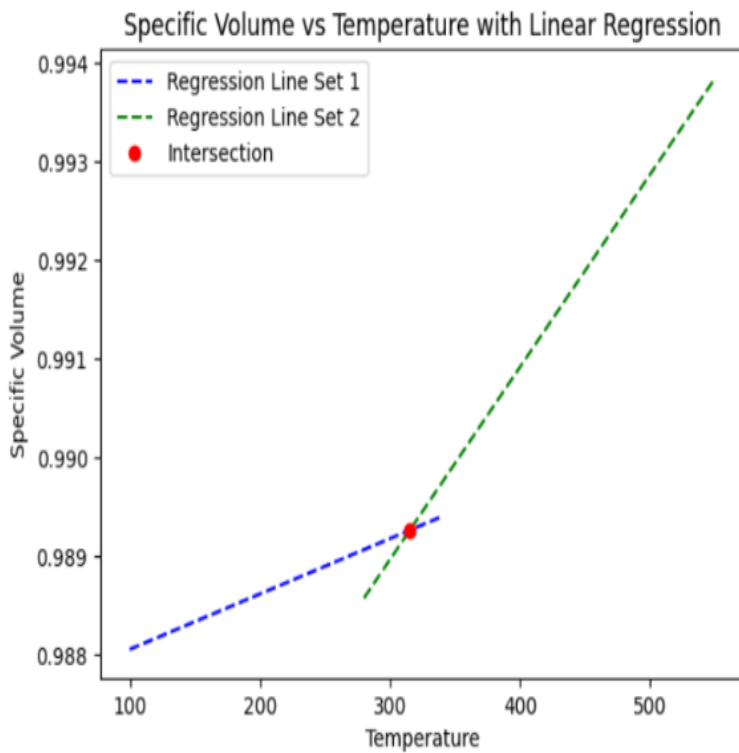
(c)

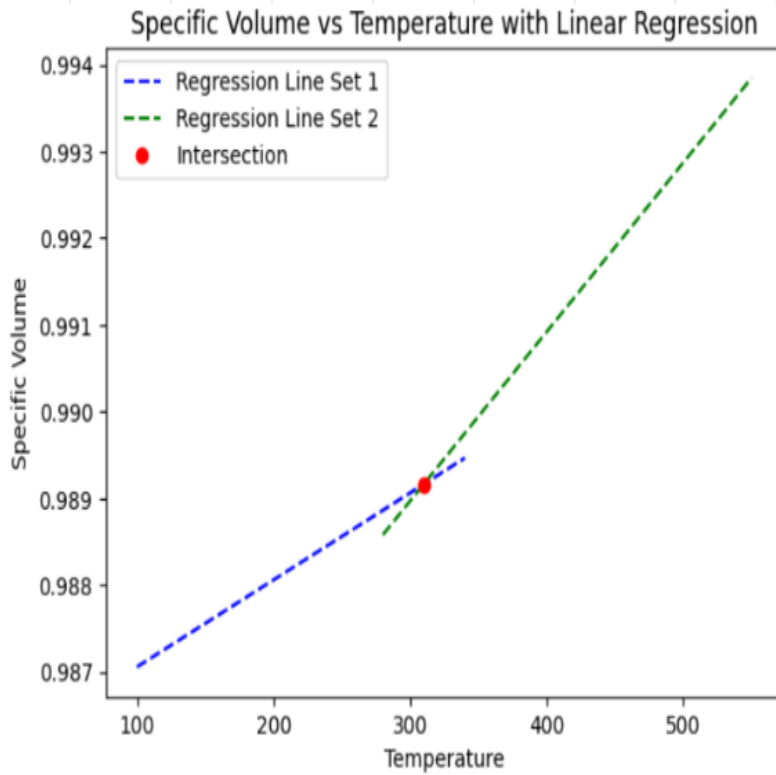Intersection Coordinates: Temperature = 317.298909360333, Specific Volume = 0.9893082488338372
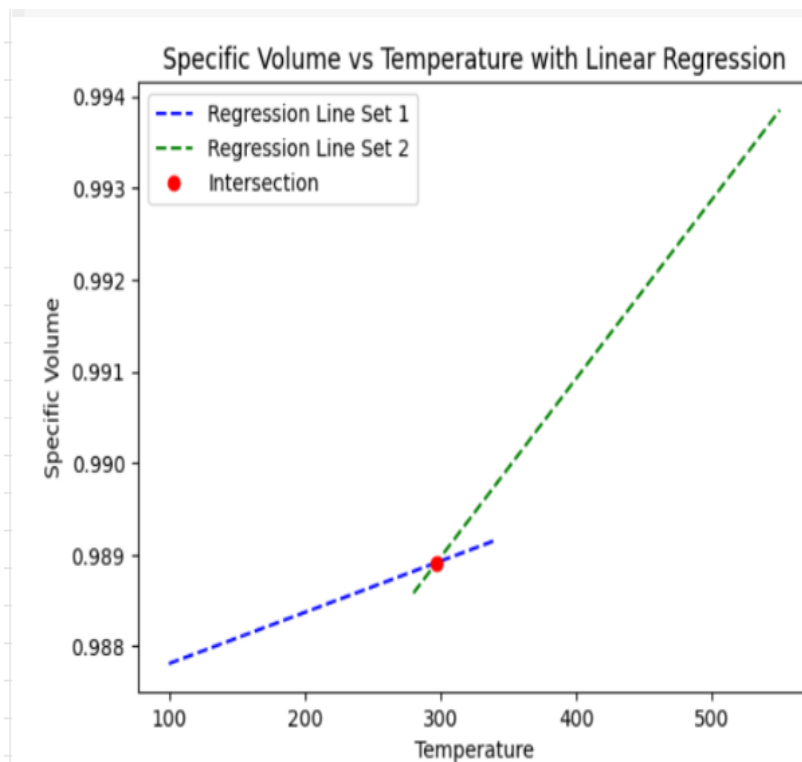
(d)

Intersection Coordinates: Temperature = 315.26444772860486, Specific Volume = 0.9892685250485994
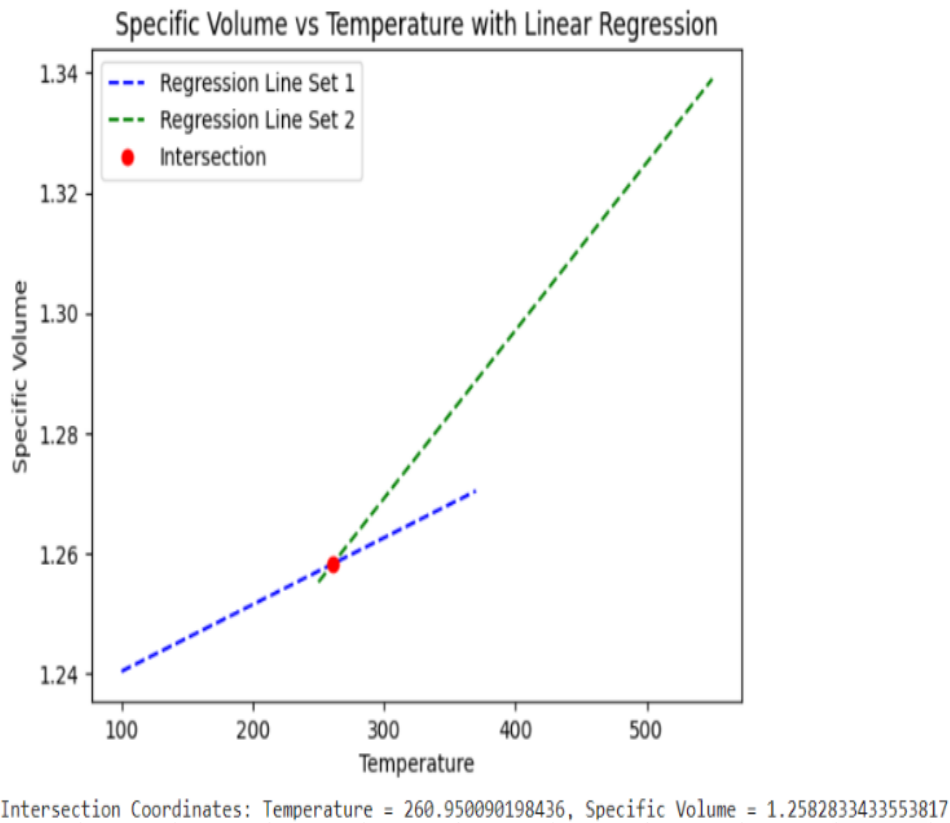
(e)

Intersection Coordinates: Temperature = 309.8594807822443, Specific Volume = 0.9891629906198119

(f)

Intersection Coordinates: Temperature = 297.14141561875977, Specific Volume = 0.9889146646345414

(g)

Intersection Coordinates: Temperature = 260.950090198436, Specific Volume = 1.2582833433553817

(h)

Figure: Specific volume vs temperature graph showing glass transition temperature of a) PHB and Pectin b) PHB and Alginate c) PHB and Gelatin d) PHB and Cellulose e) PHB and Xanthan Gum f) PHB and Starch g) PHB and Natural Rubber h) PHB and Terpene

The glass transition temperatures of various polymer blends were investigated through molecular dynamic simulations, revealing insightful trends in thermal behavior. In the presented graph, the polymer blend with pectin exhibited the highest Tg which was 346.92 K. PHB along with alginate shows the glass transition temperatures of 339.54 K and gelatin shows glass transition temperatures of 325.59 K, demonstrating notable thermal stability. Moving towards other Tg values, xanthan gum (315.26 K) and starch (309.85 K) contributed to the polymer blends, showcasing their elevated impact on the glass transition temperature. PHB and Natural Rubber showed slight increase in Tg i.e., 297.14 K. Remarkably, the lowest glass transition temperatures was observed in the PHB blend incorporating terpene, which was at 260.95 K. This result suggests that the inclusion of terpene as a blending agent significantly influences the glass transition temperature, showcasing its potential to lower the thermal transition point of the polymer blend.

In summary, the molecular dynamic simulations provide a comprehensive understanding of the glass transition temperatures in these polymer blends, offering valuable insights for tailoring thermal properties based on the choice of blending agents.

The result obtained is summarized as follows. In which we can see the highest and the lowest glass transition temperatures of the Polymer Blends.

Table 1: Glass Transition Temperatures of Polymer Blends

S.No.	Proposed Polymer	Blending Agents	Glass transition temperatures of Polymer Blends	
1.	PHB	Pectin	**346.92 K**	**Highest**
2.	PHB	Alginate	339.54 K	
3.	PHB	Gelatin	325.59 K	
4.	PHB	Cellulose	317.29 K	
5.	PHB	Xanthan Gum	315.26 K	
6.	PHB	Starch	309.85 K	
7.	PHB	Natural Rubber	297.14 K	
8.	PHB	Terpene	**260.95**	**Lowest**

In conclusion, the study conducted computational simulations to investigate the glass transition temperature of polyhydroxybutyrate (PHB) blended with various biodegradable polymers such as pectin, alginate, gelatin, cellulose, xanthan gum, starch, natural rubber, and terpene. The glass transition temperature of PHB and pectin shows the maximum increase in Tg which indicates a better thermal stability. Strong intermolecular interactions between the polymer components can lead to a higher Tg. In the case of PHB (polyhydroxybutyrate) and pectin, the presence of hydrogen bonding or other attractive forces between the polymer chains can increase the overall energy required for the polymer chains to transition from a glassy to a rubbery state. This study demonstrate the potential for accurate and reliable benchmark simulation Tg values.

The significant increase in the glass transition temperature (Tg) observed when blending PHB (polyhydroxybutyrate) with pectin, compared to the pure PHB polymer, underscores the substantial impact of the polymer blend on thermal properties. Specifically, the Tg of the PHB-pectin blend experienced a remarkable increase of 346.92 K, surpassing the Tg of pure PHB, which showed Tg of 283K. This notable enhancement in Tg suggests strong intermolecular interactions and compatibility between PHB and pectin, possibly involving hydrogen bonding or other cohesive forces, leading to a more rigid and glassy state in the blended polymer system making a more thermally stable. Thermally stable polymers can withstand higher temperatures without undergoing significant degradation (Zheng et al., 2022). This property is essential in applications where PHB may soften, deform, or degrade at elevated temperatures, such as in automotive components, electronic devices, and aerospace applications. Polymer materials with high thermal stability tend to have improved durability and longevity. This is advantageous in applications where materials are subjected to prolonged exposure to high temperatures, UV radiation, or harsh chemical environments. Such polymers can contribute to longer product lifespans and reduced material replacement frequency (Barra et al., 2023).

4. Conclusion

Polyhydroxybutyrate (PHB) is known to exhibit limitations in terms of thermal stability. Its susceptibility to thermal degradation, particularly when exposed to high temperatures, can hinder its

performance in various applications. The aim of this study is to explore strategies to enhance the thermal stability of PHB, specifically through the incorporation of various blending agents. By investigating the glass transition temperature (Tg) of PHB blended with substances such as Pectin, Starch, Cellulose, Alginate, Terpene, Gelatin, and Xanthan Gum using Molecular Dynamics (MD) simulations, we seek to identify potential solutions to address PHB's thermal stability issues. The findings not only shed light on the effects of these blending agents on the mechanical properties of the polymer but also offer valuable insights into improving its overall thermal stability. At the same time, the scale of simulation is just within nanosecond and nanometer, which is far deviating from the experimental scale. Despite the disadvantages of the MD simulations, our findings in the present work not only unveil the promising effects of PHB-Pectin on improving the mechanical properties of polymer blend, but also provide significant guidance to improve the thermal stability of polymer. The calculated results of specific volume along with temperature show that the Tg of PHB when mixed with lending agents progressively increase. Results from the molecular dynamic simulation of PHB and various blending suggest that Pectin-PHB blends have better thermal stability when compared with other biopolymers. At the same time, the scale of simulation is just within nanosecond and nanometer, which is far deviating from the experimental scale. Despite the disadvantages of the MD simulations, our findings in the present work not only unveil the promising effects of PHB-Pectin on improving the mechanical properties of polymer blend, but also provide significant guidance to improve the thermal stabiliaty of polymer.

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