Optimization and fabrication of pure poly lactic acid (PLA) using hot press compression moulding

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Abstract. Biodegradable poly lactic acid (PLA) samples were optimized and fabricated for flexural, tensile, hardness, and dielectric testing, according to the American Society for Testing Materials (ASTM). In order to achieve the highest value of tensile strength, flexural strength, and hardness, hot pressing compression molding was utilized to optimize PLA. Furthermore, not enough research was available regarding the preparation of PLA samples or composites using hot press compression molding. However, different processing variables such as temperature, pressure, compression time, holding and cooling time played huge roles. Each of these variables were investigated in order to optimize the PLA properties. Composites samples were also produced using the optimized processing parameters. These samples were then tested for tensile strength, which was 2.2 Mpa, flexural strength, 7.6 Mpa, Rockwell hardness C scale, 59, and dielectric constant/loss. The highest dielectric constant value achieved was 1.07810 at 1kHz frequency, and the highest amount of dielectric loss was 0.14910 at 2MHz frequency.

Keywords: Optimization, fabrication, hot press, Compression molding, poly lactic acid

1. Introduction
Polyactic acid, which is also known as polylactide (PLA), is produced using the help of microorganisms activity. It is a biodegradable polyester and comes under the category of aliphatics. PLA has gained researchers' interest in the past several years because the focus is on developing environmentally friendly composites. These polymers degrade naturally; therefore, once they are disposed pollution rate is lowered compared to non-biodegradable polymers. Polymers like polyethylene, polyvinyl chloride, polypropylene, and polystyrene can be produced cheaply, leading to exploitation in disposable packaging manufacturing. These polymers do not degrade; thus, polymer pollution has become a big issue [1, 2].

Mechanical properties of PLA usually vary around from soft elastic materials to high strength materials. Properties are usually manipulated depending on the kind of outcome that is expected. Different parameters such as crystallinity, a material formulation which includes composites, blends,
molecular weight, and orientation affect the properties differently. Even though the properties can be manipulated, PLA is a brittle material with low impact strength and elongation at break. However, other mechanical properties like tensile strength and modulus are comparable with polyethylene terephthalate (PET) [1].

PLA composite has been used with a variety of natural fibers such as flax, pulp, hemp, and kenaf [3-6]. Going through various researchers' work, it can be seen that processing temperature value varies around. In addition, processing temperature for injection molding machine varies around in the range of 170-180 °C [3]. Furthermore, in another research, it was seen that an extrusion machine was utilized, and the processing temperature value was between 180-190 °C [7]. By going through different researchers' work, it was observed that pure PLA samples or PLA composites are widely produced using injection molding and extrusion. Furthermore, different processing methods eventually lead up to different properties.

A limited amount of research was available regarding the production of pure PLA samples or composites using hot press compression molding; therefore, this research aimed to optimize PLA using a hot press compression molding machine. American Society for Testing and Materials (ASTM) (D790, D2240, D638, and D150) specimens were moulded for tensile, flexural, dielectric, and hardness tests. Various combinations and techniques were utilized to attain perfect specimens [8]. Different magnitudes of processing temperature, processing pressure, compression time, and cooling time were analyzed. Since the hot press compression molding is not a widely used technique to prepare samples for PLA; therefore, optimization is an essential step. Furthermore, in another research, it was stated that the hot press compression molding process produces samples with far better mechanical properties when compared with samples made using injection molding or extrusion [9].

2. Methodology

2.1. Mold Preparation

In order to fabricate the samples, ASTM (D790, D2240, D638 and D150) were utilized [10, 11]. The following Figure 1 and Figure 2 show the molds that were utilized and dimensions, respectively. A releasing agent was applied on the molds to prevent the sticking of samples to the mold and ease specimens' removal. GOTECH hot press compression molding machine was utilized for optimization and can be seen in Figure 3.

![Figure 1](image1.png)

(a) (b) (c)

Figure 1. Shows the moulds used to fabricate (a) flexural, (b) tensile and (c) dielectric samples.
Figure 2. Shows dimensions for (a) flexural, (b) tensile and (c) dielectric samples in mm.

Figure 3. Shows GOTECH hot press compression moulding machine.
2.2. **Flexural samples**

PLA is a brittle material in nature with low percentage elongation; therefore, optimization is a critical step to achieve good mechanical properties. Furthermore, optimization is also required for hot press compression molding. Different processing variables such as temperature, pressure, time, cooling time, and even the mold's condition play huge roles in optimizing the samples. Slight variations in any of these magnitudes can affect the resultant mechanical properties.

With the help of TGA analysis, it was noticed that the melting temperature of this specific PLA used in this research was in the range of 150-155 °C, as shown in Figure 4. Therefore, the first batch of samples was kept at 155 °C, with the pressure of 140 Mpa, compression time was set at 25 minutes, and cooling time was set at 3 hours. Once the mold was removed from the machine, it was clearly visible that the samples were wrecked. This was due to the pressure value of 140 Mpa being too high. Furthermore, a temperature value of 155 °C was not enough to melt the samples completely and evenly, as seen in Figure 5. By referring to previous researchers’ work, it seemed optimum that a longer cooling time ensures that the samples will settle perfectly in the mold [12, 13].

![TGA analysis of pure PLA](image1)

**Figure 4.** It shows the TGA plot for pure PLA.

![Uneven melting of PLA at 155 °C](image2)

**Figure 5.** Shows uneven melting of PLA at 155 °C.
The second batch of the samples was placed with a processing temperature of 175 °C; however, the pressure was significantly reduced to 60 Mpa, whereas compression time and cooling time were not changed. When the mold was opened, the same overflow issue was present; however, the samples were evenly melted throughout, and bubbles were present on the surface. The overflow of the sample can be seen in Figure 6. High processing temperature was utilized in this batch because higher temperature values were used during extrusion and injection molding of PLA [14-16].

![Figure 6](image)

**Figure 6.** Shows a massive amount of overflow due to high pressure.

In the third batch of samples, the mold was replaced from a male-female setup seen in Figure 7a to flat plates, which were used on top and bottom, whereas the mold was in the middle, which can be seen in Figure 7b, underneath. Similar conditions for processing temperature, processing pressure, compression time, and cooling time were applied. With flat plates on top and bottom, overflow was reduced; however, it was not eliminated.

![Figure 7](image)

**Figure 7.** Shows (a) male-female mold used in the first runs and (b) flat plates used afterward.
In the fourth batch of samples, all the conditions were kept the same; however, the pressure was reduced further down to 30 Mpa. Nevertheless, when the mold was opened to remove the samples, overflow was reduced significantly whereas, cracks were present on the sample's surface. Due to these cracks, all the samples broke during the removal from the mold.

In the fifth batch of samples, further changes were made. Processing temperature was reduced to 160 °C, processing pressure was maintained at 30 Mpa, compression time was increased to 30 minutes, and mold was pre-heated to 160 °C, and the cooling time was still at 3 hours. With the help of these changes, even melting throughout the sample was witnessed; overflow was still present; however, cracks were removed, but bubbles and voids were present on the samples, as seen in Figure 8.

Figure 8. Shows the voids caused due to the overflow of the polymer from the mold.

In the sixth batch of samples, all the conditions were kept the same; however, the processing pressure was reduced significantly to 6 Mpa. Reducing the pressure to 6 Mpa helped in reducing the overflow even more. However, the bubbles were still not removed.

In the seventh batch of samples, the baking paper was utilized between the mold and plates. Utilizing baking paper, making sure that samples were not sticking with the plates. Furthermore, the baking paper helped to cool samples evenly. With the help of previous runs, all the magnitudes were finalized. The processing temperature value was set at 160 °C, processing pressure was set to 6 Mpa, compression time was set to 30 minutes, mold was pre-heated to 160 °C, and cooling time was 3 hours. When the mold was removed and opened, it was witnessed that all five samples were perfect, and can be seen in Figure 10. To be certain, another batch of samples was placed in a hot press compression molding machine with the same conditions, and the second run was also successful. Step by step procedure to prepare the mold with baking paper can be seen in Figure 9. Therefore, optimization of flexural samples was completed. The samples were removed from the mold carefully and were placed in an airtight container for two weeks before testing [17]. These samples were then tested using GOTECH universal testing machine. All five samples were tested, and the average value was recorded [15]. Moreover, the flexural strength was calculated using the equation shown:

\[ \sigma = \frac{3FL}{2wd^2} \]  

\( \sigma \) = Flexural strength  
\( F \) = Force
\[ A = \text{Surface area} \]
\[ L = \text{Length of the sample} \]
\[ w = \text{width of the sample} \]
\[ d = \text{depth of the sample} \]

**Figure 9.** Shows the step-by-step guide to achieving perfect samples using baking paper.

**Step 1:** Flat plate was placed with baking sheet on top of it. Then the mould was placed with PLA powder.

**Step 2:** Another baking sheet was placed on top of the PLA powder.

**Step 3:** Flat metal plate was placed on top of the baking sheet and the sample was placed inside the hot press compression moulding machine.

**Figure 10.** Shows perfect flexural sample achieved using optimization.
2.3. Tensile samples

Optimization of flexural samples made things much clearer. It was noticed that the processing temperature range for even melting throughout the sample was achieved in the range of 160-170 °C while maintaining the processing pressure around 6 Mpa. It was noticed that compression time could be adjusted between 20-30 minutes, and cooling time can range from 2 hours to 24 hours. Furthermore, with baking paper, bubbles were removed, and the overflow was controlled significantly.

With this knowledge, the first batch of tensile samples was placed inside the hot press compression molding machine. Processing temperature was set at 160 °C, processing pressure was at 6 Mpa, hot press machine was pre-heated to 160 °C, compression time was 30 minutes, cooling time was 3 hours, and just like flexural samples, the baking paper was utilized. It turned out that the samples were not melted evenly, which led to the cracking of samples, as seen in Figure 11a. Uneven melting occurred due to the higher amount of PLA volume present in the tensile mold than flexural mold.

With the second batch, all of the conditions were kept the same; however, compression time was increased from 30 minutes to 40 minutes. This change was considered in order to improve the melting of PLA and remove the cracks. By increasing the compression time, cracks were removed; however, samples were still not melted properly; therefore, they broke while being removed from the mold, as seen in Figure 11b. Furthermore, voids were present on the surface of the samples as well.

The third batch of samples was placed, and two changes were made. Processing temperature was increased from 160 to 170 °C, and compression time was reduced back to 30 minutes. Samples in the mold showed even melting; however, bubbles were present, as seen in Figure 11c. Moreover, the samples cracked, and some broke as soon as pressure was removed. This showed that a temperature of 170 °C makes the samples more brittle.

The fourth batch of samples was placed, and the processing temperature was reduced to 165 °C. Even melting was witnessed, and there were no cracks. However, again when pressure was removed, samples cracked, as seen in Figure 11d.

Constant cracking and breaking of samples led to the inspection of mold. With the help of inspection, it was concluded that the mold is slightly bent; therefore, when pressure was removed, the samples would crack or break. Mould was replaced, and the fifth batch of samples was placed. Processing temperature was set at 165 °C, and mold was pre-heated to 165 °C, processing pressure of 6 Mpa was applied, compression time was 30 minutes, and cooling was done for 3 hours at room temperature. When the mold was removed, it was witnessed that all five samples were perfect. Perfect optimized tensile sample can be seen in Figure 12. Samples were removed carefully and placed in an airtight container for two weeks before testing [17]. Mechanical testing was carried out using GOTECH universal testing machine and hardness was found using SHORE hardness meter. Hardness values were then converted to the Rockwell hardness C scale. All five samples were tested for tensile strength and hardness, and the average value was recorded [15]. Moreover, the tensile strength was calculated using the equation shown:

\[ \sigma_{\text{tensile}} = \frac{F}{A} \]  

\[ \sigma_{\text{tensile}} = \text{Tensile strength} \]
\[ F = \text{Force} \]
\[ A = \text{Surface area} \]
Figure 11. Shows the optimization process of tensile samples, (a) uneven melting and cracks, (b) cracks and voids, (c) cracks and voids, and (d) minor cracks and bubbles.

Figure 12. Shows perfect tensile samples achieved using optimization.

2.4. Dielectric

Fabrication of flexural and tensile samples provided a clear idea regarding the magnitudes. Optimization of dielectric samples was the easiest among all of the optimizations. The processing temperature was set at 165 °C, and the hot press compression machine was pre-heated to 165 °C. Processing pressure was set at 6 Mpa, compression time was 25 minutes, and the cooling time was approximately 3 hours. Moreover, cooling was done at room temperature. The baking paper was utilized the same way it was utilized for tensile and flexural samples. When the mold was opened, it was witnessed that all the samples were perfect. There were no cracks, bubbles, or uneven melting of the samples. A perfect optimized sample can be seen in Figure 13. Samples were placed in an airtight container for two weeks before they were tested [17]. These samples were then tested using the
Agilent E4980-A LCR meter shown in Figure 14. Four samples were tested for dielectric constant and dielectric loss, and the average value was taken and plotted [15]. Equation used is shown underneath:

\[
C = \frac{\varepsilon_0 \varepsilon_r A}{t}
\]  

(3)

A = area of electrical conductor
\( \varepsilon_0 = \) dielectric constant of free space \( (8.854 \times 10^{-12} \text{ F/m}) \)
\( \varepsilon_r = \) dielectric constant of the insulator layer
\( t = \) thickness of the insulator layer

**Figure 13.** Shows a perfect dielectric sample achieved using optimization.

**Figure 14.** Shows Agilent E4980-A LCR meter used for testing dielectric samples.
3. Results and Discussion

3.1. Optimization
As explained in previous sections, the optimization of pure PLA was achieved for flexural, tensile, hardness, and dielectric samples. ASTM (D790, D2240, D638, and D150) standard molds were utilized to optimize the samples. Several magnitudes, such as the processing temperature at which the hot press compression molding machine was set, melted the PLA, and manufactured the composites. Compression time is the time at which the PLA was heated at the processing temperature. The cooling time is the time taken for PLA to reach room temperature from processing temperature. Furthermore, compression pressure was applied to the mold to make sure it was closed and sealed properly. Optimized magnitudes can be seen in Table 1. Issues in samples such as uneven melting, bubbles, cracking, and voids due to overflow were faced and each of these issues were solved, and therefore, optimized samples were achieved.

Table 1. Shows optimized magnitudes for flexural, tensile, and dielectric samples.

| Magnitudes         | Flexural | Tensile | Dielectric |
|--------------------|----------|---------|------------|
| Processing temperature (°C) | 160      | 165     | 165        |
| Processing pressure (Mpa)   | 6        | 6       | 6          |
| Compression time (min)      | 30       | 30      | 25         |
| Cooling time (h)            | 3        | 3       | 3          |
| Pre heat                   | Yes      | Yes     | Yes        |

All of the issues, such as uneven melting and cracks in the sample, were eliminated by adjusting magnitudes like temperature and pressure. However, issues like bubbles and voids in the samples were still present. With the help of baking paper, these issues were solved. The utilization of baking paper between the plates slowed down the cooling time. Without baking paper, the top and bottom plates had to be cooled down because the PLA was sticking to the plates.

As can be seen, a high amount of processing temperature was not utilized in this optimization paper. The optimized temperature of 160-165 °C is 5-10 °C higher than PLA's melting temperature. A higher temperature than this range would cause the PLA to vaporize, leaving the presence of bubbles and voids. Furthermore, temperature values higher than 165 °C caused samples to crack while extracting them from the mold. Therefore, higher temperature values were not used. If 155 °C, which is the melting temperature of this specific PLA, was used, uneven melting was witnessed.

Hotpress compression molding is not a commonly used technique to produce samples for PLA. More commonly utilized methods are injection molding and extrusion. Therefore, optimization on PLA holds huge importance when using hot press compression molding [9].

3.2. Mechanical properties
Flexural, tensile, and dielectric samples were placed in an airtight container for two weeks before any testing. Samples were tested for tensile and flexural strength using GOTECH universal testing machine (UTM). The hardness was tested using the SHORE hardness meter, and the value was converted to the Rockwell hardness C scale. These values can be found in Table 2.
Table 2. Shows mechanical properties of pure PLA.

| Mechanical properties       | Pure PLA |
|----------------------------|----------|
| Tensile strength           | 2.2 Mpa  |
| Flexural strength          | 7.6 Mpa  |
| Rockwell hardness C scale  | 59       |

PLA is a brittle material with low impact strength and elongation at break. However, these properties can be improved with fibers synthetic or natural rubber and even with fillers, as it has been shown by several researchers [1-5]. Furthermore, these specific properties are relevant for this specific blend of PLA made using this specific optimization.

3.3. Dielectric properties

Dielectric samples were tested using the Agilent E4980-A LCR meter. Four samples were tested, and all four values were recorded. The average of the four values was used to calculate the dielectric constant and dielectric loss. The equation shown underneath was used for calculation. Furthermore, the dielectric loss values and constant were plotted against frequency, and the plots can be seen in Figure 15 and Figure 16.

![Dielectric Constant for neat PLA](image-url)

Figure 15. Shows dielectric constant plot for pure PLA.
According to the plots shown regarding dielectric constant and dielectric loss, several trends were noticed. The highest dielectric constant value was 1.07810 at 1 kHz frequency, whereas the highest amount of dielectric loss value was 0.14910 at 2 MHz frequency. The dielectric constant showed a gradual decrease in the range of 20 Hz – 200 Hz. However, this trend can be explained using electronic polarization or atomic polarization. However, the dielectric constant showed a gradual increase in the range of 200 Hz – 1 kHz with 16.3%. This trend can be predicted due to the shaking or better known as orientational polymerization. Furthermore, the charge storing capacity was also increased once the frequency value reached 200 Hz. The dielectric constant value showed a gradual decrease in the range of 1 kHz – 2 MHz due to interfacial polymerization with a percentage of 6.6%. Whereas, in the range of 1 kHz – 2 MHz dielectric loss was increased at a percentage of 95.7%. An increase in dielectric loss can be explained due to the increased vibration occurring in the molecules. Furthermore, this trend can also be predicted if microscopic defects were present on the surface or inside the sample [18, 19].

4. Conclusion

Optimization and fabrication of pure PLA was a big task. As mentioned earlier, hot press compression molding is not a widely utilized technique to fabricate PLA samples. However, this paper focused on optimizing and fabricating PLA samples using hot press compression molding. Magnitudes such as processing temperature, processing time, processing pressure, cooling time, and molds were constantly changed depending on each batch’s outcome. Optimized samples were prepared at a processing temperature of 160-165 °C, the processing time of 25-30 minutes, processing pressure of 6 Mpa, the cooling time of 3 hours, and ASTM (D790, D2240, D638, and D150) molds were used. However, mechanical properties were not that high, knowing that PLA is a brittle material with low impact strength and elongation at break. Furthermore, looking at the dielectric constant and loss plots, it can be predicted that a high amount of dielectric loss can be expected when a higher frequency is used; therefore, the dielectric constant decreases. At the same time, low frequency leads to high dielectric constant and low dielectric loss.
5. References

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