PCL/PLA Polymer Composite Filament Fabrication using Full Factorial Design (DOE) for Fused Deposition Modelling

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Abstract: In this study, Polycaprolactone / Polylactice Acid (PCL/PLA) composite are used to fabricate filament wire with the specific diameter, which is in the range of 1.75 to 1.80 mm. Full factorial experimental design technique was used to study the main effects and the interaction effects between operational parameter which is (A) die temperature, (B) roller puller speed, (C) spindle speed and (D) inlet temperature. Besides that, there are two levels (-1 and +1) and the response are filament wire diameter. There are 16 numbers of runs and plus 8 centre points per blocks which makes the runs into 24 runs. From the experiment it shows that there are four factor that are significant effects on the filament wire diameter which is A, B, C and BC. The optimum parameter setting are also determined and there are 10 suggestions to achieve the target with different setting of parameter. The margin error for confirmation run is below than 15% when the parameter set at 6 Hz spindle speed, 4.99 rpm roller puller, 100.31 ºC die temperature and 79.65 ºC inlet temperature which can be noted that the confirmation run result is acceptable. The optimization parameter setting can use to continue in Fused Deposition Modelling (FDM). Filament wire from PCL/PLA are succesfully fabricated with acceptable diameter size and ready to be used for Fused Deposition Modelling process (FDM).

1.0 Introduction

Additive manufacturing has grown in the manufacturing process, where 3D printing is giving the ability to prototype and manufacture process, for example to make the difficult shape and it compromises important advantages over the traditional manufacturing process. In FDM, one of the most important parts is the filament. There are several types of filaments available in today’s market which depends on the part to produce and its functionality. There are many types of standard filaments such as Polylactic Acid (PLA), Acrylonitrile Butadiene Styrene (ABS), Polyamide (Nylon) and
Polyethylene Terephthalate (PET) which is commonly used for producing prototypes. However, there are recent studies have been carried out to produce filament from composites either from Ceramic Matrix Composite (CMCs) or Metal Matrix Composite (MMCs). The challenges developing composite filament is to produce this filament in a specific size and strength in order for the filament to be extruded through the printer successfully [1].

The most significant in the filament is that the diameter size must be uniform and constancy in order to used in 3D printing during the fabrication process and capability to optimize the right diameter to suit in the 3D printer [2]. The advantages of small diameter filament are that the filament is easy to be produce through the extruder machine besides, the printing process will be more accurate and easier.

There are a lot types of materials that are in market to produce implants such as titanium, steel, ceramic and aluminium, but nowadays, most of implants are made from polymers. There are a lot of benefits to produce implants from olymer where it reduces cost and it has a good physical properties. Polycaprolactone/Polylactic Acid (PCL/PLA), both of these polymers are most used in medical applications. These polymers are very famous in biomaterials because they are complementary on their physical properties and biodegradability. The PCL / PLA composite has been used as a tissue engineering scaffolds for regeneration, bone, ligament, orthopedic, skin, nerve and vascular tissue [3][4].

Usually, Fused Deposition Modelling (FDM) uses Acrylonitrile Butadiene Styrene (ABS) as its base materials, however, there are possibilities to produce new composites, especially from the mixture of Polycaprolactone/ Polylactic Acid (PCL/PLA) which can be an alternative material for the implant.

2.0 Methodology of research

2.1 Materials

Type of polymer used in this experiment are biodegradable polycaprolactone (PCL), medical grade of BGH600C meanwhile polylactic Acid (PLA) that used in this research is a medical grade of PTG600C. Both of this polymers are in pellet form and supplied by Shenzen Bright China Industrial Co, China.

| Designation | PCL (wt%) | PLA (wt%) |
|-------------|-----------|-----------|
| PCL30PLA70  | 70        | 30        |

2.2 Sample preparation

The PCL and PLA which is in resin form are mix manually. These uniformly dry-mixed batches are melt-blended in Brabender Plastograph machine where the temperatures were set at 160°C before it is crush into pellet form. All the compounded materials were then fed into injection section. The tensile and flexural specimens were produced on an injection molding machine with an injection temperature between 40 °C – 80 °C. Injection pressure was set to 140 MPa.

2.3 Filament preparation

The Polymer nano-composites that crushed into pallet size later is fed into single screw extruder machine which have been set along with a water bath and also a roller puller machine as shown in Figure 1.
2.4 Full Factorial Design (DOE)

The Design Expert Software version 6 is used in this experiment to discover the main effects for the parameters that involve in the filament fabrication process. Analysis of Variance (ANOVA) remained proficient in finding the conditional on variables that affect the machining appearances and to create the optimum situation. There is four parameter use in this experiment, Table 2 shows the range of parameter factor and level were carried out in two level high and low. The high was design in (+1) and for low was designed in (-1). The response is very important because from that the result of this experiment will be obtained. The incline of the factor that continued in demand to recognized the response which is the filament wire size. Two level full factorial design have four factors (A, B, C and D) and each factor to be studied at 2-level. There are altogether 24 runs with added 8 center points per blocks. The full factorial experiment run is shown in Table 3.

**Table 2:** The optimized factor using DOE

| Factor code | Name               | Units | Low (-1) | High (+1) |
|-------------|--------------------|-------|----------|-----------|
| A           | Die temperature    | ºC    | 100      | 160       |
| B           | Roller puller speed| rpm   | 2        | 6         |
| C           | Spindle speed      | Hz    | 2        | 6         |
| D           | Inlet temperature  | ºC    | 60       | 100       |
Table 3: Full factorial experiment runs

| Std | Run | Block | Factor (A) Die temperature (ºC) | Factor (B) Roller puller speed (RPM) | Factor (C) Spindle speed (Hz) | Factor (D) Inlet temperature (ºC) | Response Filament size (mm) |
|-----|-----|-------|-------------------------------|-------------------------------------|--------------------------------|-----------------------------------|-----------------------------|
| 9   | 1   | PCL70PLA30 | 100                           | 2                                   | 2                             | 80                               |                             |
| 15  | 2   | PCL70PLA30 | 100                           | 6                                   | 6                             | 80                               |                             |
| 13  | 3   | PCL70PLA30 | 100                           | 2                                   | 6                             | 80                               |                             |
| 21  | 4   | PCL70PLA30 | 130                           | 4                                   | 4                             | 65                               |                             |
| 18  | 5   | PCL70PLA30 | 130                           | 4                                   | 4                             | 65                               |                             |
| 8   | 6   | PCL70PLA30 | 160                           | 6                                   | 6                             | 50                               |                             |
| 10  | 7   | PCL70PLA30 | 160                           | 2                                   | 2                             | 80                               |                             |
| 7   | 8   | PCL70PLA30 | 100                           | 6                                   | 6                             | 50                               |                             |
| 3   | 9   | PCL70PLA30 | 100                           | 6                                   | 2                             | 50                               |                             |
| 5   | 10  | PCL70PLA30 | 100                           | 2                                   | 6                             | 50                               |                             |
| 22  | 11  | PCL70PLA30 | 130                           | 4                                   | 4                             | 65                               |                             |
| 4   | 12  | PCL70PLA30 | 160                           | 6                                   | 2                             | 50                               |                             |
| 16  | 13  | PCL70PLA30 | 160                           | 6                                   | 6                             | 80                               |                             |
| 24  | 14  | PCL70PLA30 | 130                           | 4                                   | 4                             | 65                               |                             |
| 2   | 15  | PCL70PLA30 | 160                           | 2                                   | 2                             | 50                               |                             |
| 19  | 16  | PCL70PLA30 | 130                           | 4                                   | 4                             | 65                               |                             |
| 17  | 17  | PCL70PLA30 | 130                           | 4                                   | 4                             | 65                               |                             |
| 20  | 18  | PCL70PLA30 | 130                           | 4                                   | 4                             | 65                               |                             |
| 1   | 19  | PCL70PLA30 | 100                           | 2                                   | 2                             | 50                               |                             |
| 14  | 20  | PCL70PLA30 | 160                           | 2                                   | 6                             | 80                               |                             |
| 6   | 21  | PCL70PLA30 | 160                           | 2                                   | 6                             | 50                               |                             |
3.0 Result & Discussion.

3.1 Two Level Full Factorial Design.
The complete result of all 24 runs of experiment are shown in Table 4. From the experiment, it can be seen that there are two results that produce the filament with diameter size in range of 1.75mm-1.80mm which are run number 6 and 13. The parameter setting is when the die temperature at 100°C, the roller puller speed at 6 RPM, the spindle speed at 6 Hz. The differences is only at the inlet temperature where at run 6 the temperature is at 50°C meanwhile at run 13 the temperature is at 80°C.

Table 4: The respond result for filament size diameter

| Run | Factor (A) Die temperature (°C) | Factor (B) Roller puller speed (RPM) | Factor (C) Spindle speed (Hz) | Factor (D) Inlet temperature (°C) | Response Filament size (mm) |
|-----|---------------------------------|--------------------------------------|-------------------------------|----------------------------------|-----------------------------|
| 1   | 100                             | 2                                    | 2                             | 80                               | 1.18                        |
| 2   | 100                             | 6                                    | 6                             | 80                               | 1.63                        |
| 3   | 100                             | 2                                    | 6                             | 80                               | 1.98                        |
| 4   | 130                             | 4                                    | 4                             | 65                               | 1.61                        |
| 5   | 130                             | 2                                    | 6                             | 65                               | 1.61                        |
| 6   | 160                             | 6                                    | 6                             | 50                               | 1.77                        |
| 7   | 160                             | 2                                    | 2                             | 80                               | 1.36                        |
| 8   | 100                             | 6                                    | 6                             | 50                               | 1.53                        |
| 9   | 100                             | 2                                    | 6                             | 50                               | 0.98                        |
| 10  | 100                             | 6                                    | 2                             | 50                               | 2.49                        |
| 11  | 130                             | 4                                    | 4                             | 65                               | 1.63                        |
| 12  | 160                             | 6                                    | 2                             | 50                               | 1.12                        |
| 13  | 160                             | 6                                    | 6                             | 80                               | 1.76                        |
| 14  | 130                             | 4                                    | 4                             | 65                               | 1.57                        |
| 15  | 160                             | 2                                    | 2                             | 50                               | 1.70                        |
| 16  | 130                             | 4                                    | 4                             | 65                               | 1.59                        |
| 17  | 130                             | 4                                    | 4                             | 65                               | 1.59                        |
| 18  | 130                             | 4                                    | 4                             | 65                               | 1.59                        |
| 19  | 100                             | 2                                    | 2                             | 50                               | 1.43                        |
| 20  | 160                             | 2                                    | 6                             | 80                               | 2.73                        |
| 21  | 160                             | 2                                    | 6                             | 50                               | 2.81                        |
| 22  | 160                             | 6                                    | 2                             | 80                               | 1.11                        |
| 23  | 100                             | 6                                    | 2                             | 80                               | 0.93                        |
| 24  | 130                             | 4                                    | 4                             | 65                               | 1.53                        |
3.1.1 Analyzed in of Variance (ANOVA) for Filament Wire.

All the data were analyzed in variance (ANOVA). Figure 2 shows the half normal plot graph. From the figure, all the specimens are located far from line that have a significant effect which are point A, B, C, and BC. Table 5 below shows the ANOVA summarize for the next interpretation.

![Half Normal plot graph](image)

**Figure 2**: The half normal plot graph for filament wire diameter PCL70PLA30

| Source       | Sum of squares | DF | Mean Square | F value | Prob > F |
|--------------|----------------|----|-------------|---------|----------|
| Model        | 4.94           | 4  | 1.24        | 36.75   | < 0.0001*|
| A            | 0.28           | 1  | 0.28        | 8.28    | 0.0100   |
| B            | 1.39           | 1  | 1.39        | 41.25   | < 0.0001 |
| C            | 3.05           | 1  | 3.05        | 90.85   | < 0.0001 |
| BC           | 0.22           | 1  | 0.22        | 6.64    | 0.0190   |
| Curvature    | 0.018          | 1  | 0.018       | 0.54    | 0.4735** |
| Residuals    | 0.61           | 18 | 0.034       |         |          |
| Lack of Fit  | 0.31           | 11 | 0.028       | 0.67    | 0.7318** |
| Pure error   | 0.29           | 7  | 0.042       |         |          |
| Cor Total    | 5.56           | 23 |             |         |          |

The model of F-value is 36.75 indicates the model is significant. There are only a 0.01% chance that a model F-value this larger could occur due to noise. The value of Prob > F less than 0.0500 indicates that the model is significant. In this case, only point A, B, C, BC are significant model terms because the value is below than 0.0500. The Curvature F-value is 0.54 implies the curvature. The curvature measure of difference between the average of the center point and average of the factorial point. This design is significant relative to noise. The Curvature Prob F> value has chance 47.35%, this is the large value and might occur due noise. The Lack of Fit F-value is 0.67, this is not
significant relative to the purse error. Prob > F there is a 73.18% chance, this is large could occur due to noise. Non- significant is the best result because when reject the factory are not involved the value is close to 1.

Table 6 below shows the ANOVA and a regression summary of filament wire diameter fabrication. The Predicted R- Squared, Adjusted R-square and R- Square value is 0.8127, 0.8867, and 0.8909 respectively. This values are in reasonable because the value were close to 1. For the Adequate Precision the value must greater than 4 is desirable. In this adequate result, the value is 18.831 indicating an adequate signal. The Adequate Precision is measure the signal to the noise ratio and this model can be used to navigate the design space.

Table 6: ANOVA and regression summary filament wire diameter for PCL70PLA30

| Response model      | Quadratic model |
|---------------------|-----------------|
| R-squared           | 0.8909          |
| Adjusted R-Squared  | 0.8667          |
| Predicted R-squared | 0.8127          |
| Adequate Precision  | 18.831          |

The quadratic regression equation might be expressed as:

Final Equation in terms of Coded Factor:

\[
\text{Filament wire diameter} = 1.65 + 0.13*A - 0.29*B + 0.44*C - 0.12*B*C
\]

Final Equation in terms of Actual Factor

\[
\text{Filament wire diameter} = 0.3191 + 4.39583E*A - 0.029062*B + 0.33656*C - 0.029531*B*C
\]

3.1.2 Graphic Plots Analysis Filament wire Diameter

The analysis continues with the graph plots. The factor among spindle speed, roller puller, inlet temperature and die temperature were observed better in the normal plot of residuals graph, residuals vs. predicted graph and Outlier T graph. The graphs plot are shown in Figure 3, 4 and 5 respectively. Figure 3 shows that the residuals point lies on a straight line indicating that the errors are distributed normally. Meanwhile, the plots in Figures 4 and 5 shows that no obvious pattern and unusual structure and all the results fall in the acceptable range. Therefore, it can be concluded that the model proposed was adequate and could proceed for further analysis. At this point, only significant effects were analyzed.
Figure 3: The normal plot of residuals graph of filament wire diameter PCL70PLA30

Figure 4: The residuals vs. predicted graph of filament wire diameter PCL70PLA30

Figure 5: The outline T graph of filament wire diameter PCL70PLA30
Figure 6 shows the contour and interaction graph among (B) roller puller speed and (C) spindle speed. It have a strength interaction between two factors to fabricate the filament wire diameter. When the spindle speed was increased and the roller puller speed was decreased, the diameter size of filament wire produced is in a large size. When the spindle speed was decrease, the reverse result was found and the diameter of filament wire that produce in a small size.

This design of effect were contributed through the die swell outcome which is significant miracle defining to size or shape and quality performance of screw speed of extruder polymer. Meanwhile, the roller puller speed is important in fabricating of filament wire diameter in required range. If the roller puller setting was in a higher speed, it is possible that the wire that pull out from the die with a high speed will create the size of the wire that came out is in a small diameter. In order to achieve the required size of wire diameter, both of this parameter need to adjust in a perfect manner as both are correlated [5].

**Figure 6:** Contour and interaction graph among (B) roller puller speed and (C) spindle speed on filament wire diameter PCL70PLA30
3.1.3 Optimization Condition
The optimum condition for the filament wire diameter were obtained from the 2 level factorial design. The combination from few parameters were combined which is to achieve the filament wire diameter in the range 1.75 mm. From the result, there are 10 suggestions to achieve the target with different setting of parameter. Meanwhile, the combination of all suggestions were in specific desirability value because all value is closed to 1. Table 7 shows the suggested setting parameter for 1.75mm filament wire diameter.

Table 7: Suggested setting for filament wire diameter

| Number | Die temperature (°C) | Roller puller speed (rpm) | Spindle speed (rpm) | Inlet temperature (°C) | Filament wire diameter (mm) | Desirability |
|--------|----------------------|--------------------------|---------------------|------------------------|-----------------------------|--------------|
| 1      | 100                  | 4.97                     | 6.00                | 79.96                  | 1.75316                     | 0.987        |
| 2      | 100.31               | 4.99                     | 6.00                | 79.65                  | 1.74992                     | 0.984        |
| 3      | 160.00               | 2.00                     | 2.83                | 50.00                  | 1.75492                     | 0.984        |
| 4      | 124.99               | 5.52                     | 6.00                | 80.00                  | 1.74992                     | 0.972        |
| 5      | 113.96               | 5.24                     | 5.95                | 80.00                  | 1.74992                     | 0.972        |
| 6      | 100.00               | 4.98                     | 6.00                | 74.71                  | 1.75492                     | 0.966        |
| 7      | 160.00               | 2.00                     | 2.82                | 52.62                  | 1.74662                     | 0.956        |
| 8      | 148.56               | 6.00                     | 6.00                | 60.66                  | 1.75417                     | 0.939        |
| 9      | 160.00               | 4.19                     | 3.99                | 50.00                  | 1.75137                     | 0.901        |
| 10     | 100.00               | 4.98                     | 6.00                | 60.12                  | 1.75137                     | 0.901        |

Figure 7 shows the ramps plots for suggestion number 1, where the parameter setting value must set on optimization conditions to achieve goals. Where the die temperature, roller puller speed, spindle speed, and inlet temperature value must set at 100.00 °C, 4.97 rpm, 6.00 Hz, 79.96 °C respectively.

3.2 Confirmation run for Filament Wire Diameter
The confirmation run are needed to compare the result between the theoretical and experimental value for the size of wire in the range 1.75 mm-1.80mm. From the optimization 2 level factorial design,
there are 10 runs suggested to achieve the target. From the parameter range, run 2 were selected for the confirmation run to evaluate the value of margin error. Table 8 shows the confirmation run selected from the 2 level factorial design.

**Table 8: Confirmation run selected (run 2)**

| Number | Die temperature (°C) | Roller puller speed (rpm) | Spindle speed (rpm) | Inlet temperature (°C) | Filament wire diameter (mm) | Desirability |
|--------|-----------------------|---------------------------|---------------------|------------------------|-----------------------------|--------------|
| 2      | 100.31                | 4.99                      | 6.00                | 79.65                  | 1.75                        | 0.986        |

By referring to the filament size in Table 8, the filament wire diameter for run 2 is 1.75mm. The confirmation run was repeated for 5 times, in order to identify the best setting on single screw extruder machine and roller puller machine. The actual test result in confirmation run is 1.75mm diameter size, the comparison of theoretical prediction result and the actual test result can be a final consideration for the optimal parameter predicted allowed in range. The margin error value of actual and predicted result was set less than 15%.

Table 9 shows the result of confirmation run. The parameter were set at 6 Hz spindle speed, 4.99 RPM roller puller, 100.31 °C die temperature and 79.65 °C inlet temperature. This optimizes setting for a confirmation run in a 95% prediction interval (PI) are reasonably accurate and the error value for filament wire diameter in the range 0 - 15%. The percentages are below than 15% and the confirmation run result is verified as valid to the existence of an optimal point. From this experiment, PCL/PLA filament wire succesfully fabricated with exact size, strength and properties to be used in Fused Deposition Modelling (FDM) machine.

**Table 9: Confirmation run result for filament wire diameter**

| Run | Wire diameter (PCL70PLA30) | Actual (mm) | Predicted (mm) | Error (%) |
|-----|---------------------------|-------------|----------------|-----------|
| 1   | 1.76                      |             |                | 0.57      |
| 2   | 1.77                      |             |                | 1.14      |
| 3   | 1.75                      | 1.75        |                | 0         |
| 4   | 1.76                      |             |                | 0.57      |
| 5   | 1.78                      |             |                | 1.69      |
|     | Average error (%)         |             |                | 0.79      |

**4. Conclusion**

Filament wire from Polycaprolactone/ Polylactic Acid (PCL/PLA) composites are succesfully fabricated with the aid of two level full factorial design using Design of Expert version 6 software. From the study, it can concluded that:

1. From the experiment there are 2 out of 24 runs that gives a promising result as the filament produce falls into an acceptable range. The parameter setting is when the die temperature at 100°C, the roller puller speed at 6 RPM, the spindle speed at 6 Hz. The differences is only at the inlet temperature where at run 6 the temperature is at 50°C meanwhile at run 13 the temperature is at 80°C.
2. The Analyzed of Variance (ANOVA) for filament wire diameter indicate that the significant effect were the die temperature (A), roller puller speed (B), spindle speed (C) and roller puller speed with spindle speed (BC). For the curvature and lack of fit value, there were not significant for the response. In curvature, if the value is significant, an additional run was needed. The lack of fit value must be closes to 1.

3. In order to achieve the target required diameter size of the filament which is 1.75 ± 0.05 mm, there are 10 suggestion of parameter setting for the all the 4 factors.

4. The confirmation run are conducted using the value from optimal condition that is suggested. The setting for confirmation was 6Hz for spindle speed, 4.99 RPM for roller puller, 100.31 °C for die temperature and 79.65 °C for inlet temperature. From the confirmation run result, the percentage average error are below 15% and its is acceptable.

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