

Taguchi Optimization of Plasticisation Process for Glycerol and TEA Based Thermoplastic Mango Starch Biofilms

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Abstract

This investigation involves study of independent variable that influences the Young modulus of thermoplastic mango starch (TPS) as dependent response factor. The experiment was design using the Taguchi orthogonal technique with four independent variables; plasticiser type; glycerol (G), and Triethanolamine-(TEA) (T), percentage plasticiser (40 and 120 %), percentage carboxymethyl cellulose-CMC (10 and 50 %), and concentration of HCl (0.05 and 0.15 M). The result of the main effect plots for mean indicated that the gTPS-CMC1 with 268.85^a MPa is a better outcome compared to gTPS-CMC3 with 280.31^a MPa, since no significance difference was observed due to less composition requirement of CMC for gTPS-CMC1, making it more cost effective to produced with better optimum conditions. The interaction plot of the independent variables showed that for plasticiser types; when glycerol (G) was utilised a higher young modulus is observed than TEA (T) and only interacts with TEA (T) at 0.015 M HCl; 10 % CMC gives a higher response compared to 50 % CMC and showed no interaction even as the other independent variables fluctuates, and similar effect was observed for percentage plasticiser. Study concluded that the predicted mean (young modulus) is substantially consistent with the experimental observation ($R^2 = 0.6283$).

1. Introduction

The level of pollution cause by petro-plastics and high costs of production associated with bioplastic has been a disadvantage that has driven research to look for more environmental and economical alternatives. Most research is carried out without the application of statistical procedure that could help in estimating and eliminating trial and error, minimizing wastage of resources and time in production procedures^[1, 2]. Challenge facing the up-scaling commercial production of bioplastic could be due to lack of comprehensive feasibility report on the full market potential, and high cost associated with certain class of bioplastics^[3, 4]. The over dependence on unsustainable resources^[5] and time consumption could be attributed to poor experimental approach, technique, design and insufficient information on biomaterials-biopolymers. Environmental concerns has gradually wiped up sentiments, with public perception and awareness on the rise, and government policy aligning towards promoting sustainable approaches that are environmental friendly, economical, and efficient^[6]. The utilization of bioplastics is a step forward innovation to solving some of the environmental issues using cost-effective renewable substrates^[7]. This has prompted studies into the possibility of deriving raw materials from abundance agricultural by-product as feedstock for large-scale bioplastic production^[5] with low-cost^[8], and excellent performance^[9]. In addition, the production cost could be mitigated when the proper scientific approached is employed by using appropriate experiment design before commercial scale up.

With all the above factors in perspective the most viable bioplastic with potential for commercialisation are thermoplastic derived from biomass through thermomechanical process known as plasticisation. Plasticisation process can be designed with robust mathematical model that produces optimum condition for cost effective thermoplastic biomass production. Plasticisation of biomass to bioplastic

remains one of the affordable routes to obtaining sustainable plastics due to its simplicity and versatility [10, 11]. As it allowed the use of a variety of plasticiser(s) to convert rigid biomass into flexible bioplastic referred to as thermoplastic [12]. The process does not release any harmful by-product(s) into the environment and gives a material that is renewable [13]. Enhanced, economical, and efficient plasticisation process can help in producing thermoplastic starch with commercial value when a design of experiments (DOE) model is employed to derive the optimum conditions and predicts experimental outcome [14]. Scaling up the process and procedure for the production could help in creating a profitable commercial venture of thermoplastic starch if the appropriate approach of laboratory experimental design is employed which does not depend on single independent variable as practiced [15].

Therefore, the use of DOE represents a better alternative approach in scientific research compared to traditional or conventional techniques that only put experimental outputs under consideration. Most times the t-test, chi-test, analysis of variance (ANOVA) are applied to study outcomes of research investigations to ascertain certain relevant information that could be used for up-scale process and production in commercial ventures [16]. These provide information about significant differences in research data which is not enough most times, provides no information about economical advantage since it does not consider the independent input production condition(s) and product composition(s) in analysing the defined response dependent variable which is just an output.

The common types of DOE methods have their advantages over one another and these include Response surface, Factorial, Mixture, and Taguchi methods [17]. These techniques could predict the input conditions and their corresponding output responses with response optimizer but will not give the optimum output experimental response except for Taguchi methods that do not predict input conditions but give the optimum experimental output response and predict outcome response based on the independent variables [16]. This technique measures the robustness utilised to recognize control factors and reduces unpredictability in product(s) or process(s) by minimizing the effects of uncontrollable factors (noise factors) [15, 17].

Therefore, the Taguchi technique is a simple but robust method that provides direct optimum conditions for profitable production [15]. Taguchi provides information on optimum conditions that is usually profit oriented when applied either in small, medium or large scale production due to ability to select conditions that are cost effective in thermoplastic starch production, and can help estimate both the absolute and relative error by providing predicted outcomes that can be comparable with experimental output [14, 15]. Vital information that could be derived from Taguchi technique includes; Main effect plot for Signal/Ratio and Mean, Interaction plot, predicted mean (PMEAN). Therefore, the aim of this investigation is to design experimental procedure and ascertain the optimum condition for the plasticization of Mango starch with glycerol and TEA, with young modulus as the dependent variable response applying larger-is-better Taguchi technique equation model.

2. Materials And Methods

2.1. Materials

The mango starch was provided by the Department of Pharmaceutics, Faculty of Pharmaceutical Science, Usmanu Danfodiyo University Sokoto, Sokoto, Nigeria and other reagents and tensionmeter was provided by Department of Chemical Sciences, Faculty of Sciences, Clifford University, Owerri, Abia State, Nigeria. All chemicals and reagents were used without any further purification.

2.2. Methods

2.2.1. Taguchi Design of Experiment

Taguchi Orthogonal Array Design, $L_8(2^4)$, level : 2, Factors: 4, Runs: 8, and Columns of $L_8(2^7)$ Array 1, 2, 4, 5. The Design contains 3 numerical factors (controllable), and one categorical factor plasticiser type (uncontrollable factor) as shown in Table 2.1.

Table 2.1
Taguchi Design indicating the Composition of thermoplastic mango starch biofilms

Sample Code	Plasticiser type	%CMC	%Plasticiser	HCl (M)
gTPS-CMC1	G	10	40	0.05
gTPS-CMC2	G	10	120	0.15
gTPS-CMC3	G	50	40	0.05
gTPS-CMC4	G	50	120	0.15
tTPS-CMC1	T	10	40	0.15
tTPS-CMC2	T	10	120	0.05
tTPS-CMC3	T	50	40	0.15
tTPS-CMC4	T	50	120	0.05

gTPS-CMC: glycerol plasticised thermoplastic mango starch reinforced with Carboxymethyl cellulose, tTPS-CMC: TEA plasticised thermoplastic mango starch-reinforced with Carboxymethyl cellulose, G: glycerol, T: TEA

2. 2.2. Preparation Of Thermoplastic Mango Starch Biofilm

The thermoplastic starch biofilm was prepared based on method described in previous publication ^[18] and casting technique was use to make the final biofilms by pouring the hot liquid plasticised thermoplastic starch mixture into pre-labelled Petri-dish and allowed to stand on laboratory bench for 48 hours. The composition of each thermoplastic mango starch biofilms are shown in Table 1 as derived from the Taguchi Orthogonal array design.

2. 2.3. Measurement of Young modulus

The TPS-CMC biofilms were cut into a dumb-bell shape with the help of a metallic template and the length (l_1) and breadth (b) measure with the help of a metre-ruler. The dumb-bell shape biofilms was clamped firmly unto a fabricated tensionmeter at both ends. The weight (w) needed to break the TPS-CMC biofilms at the middle and new length (l_2) before breaking was measured and recorded in triplicates ^[19].

$$\text{Stress} = \text{Force (N)} / \text{Area (m}^2\text{)} \text{-----(1)}$$

Where Force = $w \times 9.86$, and Area = $l_1 \times b$

Convert Stress from Pa to MPa. (Pa = 1×10^{-6} MPa)

$$\text{Strain} = \frac{l_2 \text{ (m)} - l_1 \text{ (m)}}{l_1 \text{ (m)}} \text{-----(2)}$$

$$\text{Young modulus} = \frac{\text{Stress (MPa)}}{\text{Strain}} \text{-----(3)}$$

2. 2.4. Measurement of Absolute and Relative Error

Mathematical the absolute and relative error are determined using equation shown below ^[20, 21].

$$\text{A.E} = \text{PMEAN} - \text{EMEAN} \text{-----(4)}$$

$$\% \text{ R.E.} = \frac{\text{A.E}}{\text{PMEAN}} \times 100 \text{-----(5)}$$

Where A.E: absolute error, R.E: Relative error, PMEAN: Predicted Mean (Young modulus), and EMEAN: Experimental Mean (young modulus).

2. 2.5. Statistical Analysis

The observed experimental outcome (YM) was analysed using Analysis of variance (ANOVA) to determine the significant difference at 95% confidence interval. The Taguchi designed experiment was optimized using two step optimization processes. Step 1 used the Larger-is-better signal-to-noise ratio (S/N) to identify those factors that reduces variability to maximise the response calculated as shown in equation (6). Steps 2 recognized control variables which shift the Mean to objective and have a small or no effect on the signal-to-noise ratio ^[17]. The main effect plot of mean and Signal-noise ratio, interaction plot, and the predicted mean i.e. predicted young modulus where derived and analysed using the higher-is better signal-to-noise ratio model of Taguchi method in Minitab 17 software suite.

$$\text{S/N: } -10 \times \log \left[\sum (1/Y^2)/n \right] \text{-----(6)}$$

Where Y = response for a given factor level, n = sum of responses in the factor level combination.

3. Result

Table 3.1 indicates the result of Young modulus derived from the experimental determination while Table 3.2 and 3.3 shows the Taguchi Analysis of Young modulus response with respect to the independent variable Plasticiser type, CMC (%), Plasticiser (%), and HCl (M).

Table 3.1
Young Modulus (Response) with Grouping Information using the Tukey Method at 95% Confidence Interval

Sample Code	Plasticiser type	CMC (%)	Plasticiser (%)	HCl (M)	Response (Young Modulus) (MPa)
gTPS-CMC1	G	10	40	0.05	268.849 ± 01.569 ^a
gTPS-CMC2	G	10	120	0.15	155.596 ± 27.369 ^{bc}
gTPS-CMC3	G	50	40	0.05	280.309 ± 08.280 ^a
gTPS-CMC4	G	50	120	0.15	091.114 ± 01.791 ^{de}
tTPS-CMC1	T	10	40	0.15	176.404 ± 13.785 ^b
tTPS-CMC2	T	10	120	0.05	124.993 ± 39.413 ^{cd}
tTPS-CMC3	T	50	40	0.15	106.636 ± 02.734 ^{cd}
tTPS-CMC4	T	50	120	0.05	041.347 ± 05.161 ^e
Mean ± Standard deviation, Means that do not share a letter are significantly different					

Table 3.2
Response Table for Larger-is Better Signal-to-Noise Ratios

Level	Plasticiser type	%C MC	% Plasticiser	HCl (M)
1	45.14	44.82	45.76	42.95
2	39.94	40.26	39.32	42.13
Delta	5.20	4.57	6.43	0.82
Rank	2	3	1	4

Table 3.3
Response Table for Means

Level	Plasticiser Type	% CMC	% Plasticiser	HCl (M)
1	199.0	181.5	208.1	178.9
2	112.3	129.9	103.3	132.4
Delta	86.6	51.6	104.8	46.4
Rank	2	3	1	4

4. Discussion

From Table 3.1, the result should that gTPS-CMC3 has the highest young modulus of 280.31 MPa (response) compared to gTPS-CMC1 of 268.85 MPa but are not significant difference ($p > 0.05$), but both are significantly higher compared to the biofilm derivatives ($p \leq 0.05$). 155.596 MPa was observed for gTPS-CMC2 that is less than 176.40 MPa for tTPS-CMC1, and higher than 124.99 MPa for tTPS-CMC2, and 106.64 MPa for tTPS-CMC3 but indicates no significant difference ($p > 0.05$), while gTPS-CMC4, tTPS-CMC2 and tTPS-CMC3 showed no significant difference ($p > 0.05$) but are significantly lower than tTPS-CMC1 ($p \leq 0.05$). 0.41.35 MPa was observed for tTPS-CMC4 is significantly the lowest response except for gTPS-CMC4 with 0.91 MPa where is showed not significant difference ($p > 0.05$).

Table 3.2 and 3.3 are consistent with each other from the Delta ranking of the controllable factors on the mean and signal-to-noise ratio, the both table indicates that plasticiser (%) has the highest effect on the Mean and Signal to Noise ratio. Followed by Plasticiser type, and thirdly CMC (%) and lastly with HCl. This observation is expected to be consistent with main plot mean and signal-to-noise ratio.

From Fig. 3.1 and 3.2, the main effect plot of mean and signal-to-noise ration showed that glycerol plasticised thermoplastic mango starch hydrolysed with 0.05 M HCl, and plasticised with 40 % glycerol and reinforced with 10 % CMC indicates the optimum condition for the production of CMC reinforced thermoplastic mango starch which is equivalent to gTPS-CMC1 with young modulus i.e. mean of 268.85 MPa and signal-to-noise ratio 51.055. The results indicates that selection of optimum conditions is factor that depends on the smallest input of independent variable that gives the highest output response. Result indicates that gTPS-CMC1 with 268.85 MPa is lower than gTPS-CMC3 with 280.31 MPa signal-to-noise ratio but the analysis of variance reveal that both outcomes are the same but the choice of gTPS-CMC1 by Taguchi approach over gTPS-CMC3 does not depend on the highest output response but on the lowest possible combination of inputs i.e. independent variables that gives a output that are significantly viable or economical to produced. gTPS-CMC3 will required 50% CMC, 40 % glycerol and 0.05M HCl to give 280.31 MPa; and gTPS-CMC1: 10% CMC, 40% glycerol and 0.05M HCl is required to give 268.85 MPa. the choice of gTPS-CMC1 over gTPS-CMC3 lies in the amount of CMC required as input from the main effect plot, because the amount of required to give 280.31 MPa gTPS-CMC3 is 50%, while 10% is

required to produced 268.85 MPa for gTPS-CMC1 with no significant difference in outcomes ($p > 0.05$). this represent a reduction in production cost since a lesser amount of CMC is required to produced gTPS-CMC1. The observation is consistent with previous study on carboxymethylation process using Taguchi technique by Agwamba et al [22].

Figure 3.3 showed that type G and T has no interaction between then when CMC increases from 10 to 50 %. Increase in plasticiser 40 to 120 %, but when the HCl was increased from, 0.05 to 0.15 M, the G and T interacted at 0.15 M HCl. The CMC at 10 and 50% showed no interaction with G and T, 40 to 120 % plasticiser, and 0.05M to 0.15 M HCl. The % Plasticiser at 40 and 120 % showed no interaction from for plasticised type from G and T, % CMC from 10 to 50 %, and HCl from 0.05 to 0.15 M. HCl at 0.05 and 0.15 M showed no interaction for CMC from 10 to 50% but showed interacted at T for plasticised type, and 120 % plasticiser.

According to Henseler et al., [23], the R^2 value of 0.6283 observed in this study indicates that the predicted mean is substantially consistent with the measured experimental observation. Three (3) of the observed mean (predicted and experimental) i.e. the predicted mean of gTPS-CMC1, gTPS-CMC3, tTPS-CMC2, and tTPS-CMC4 are significantly different from the experimental mean ($p \leq 0.05$) which represents about 37.5 % of the result indicating that the remaining 62.5 % are not significant different ($p > 0.05$). This showed that the model fit for deriving predicted mean.

The Fig. 3.4, also indicated that the predicted mean and experimental mean of gTPS-CMC1, gTPS-CMC3, tTPS-CMC2, and tTPS-CMC4 are significantly different ($p \leq 0.05$) because the relative error are approximately higher than 10 % and gTPS-CMC2, gTPS-CMC4, tTPS-CMC1, and tTPS-CMC3 indicated a relative error that are approximately less than 10% and are not significantly different ($p > 0.05$). Therefore, the Taguchi technique model could be modified to help estimate of level accuracy in an experimental results. The relative error derived from the Taguchi Prediction model relates to the significance difference between the predicted and experimental mean derived from the analysis of variance (ANOVA) of experimental mean response.

5. Conclusion

The use of Taguchi technique for design and analysis gives the optimum independent production parameters, and predict almost accurately the response. This represents an efficient and economical avenue to mitigate the expensive nature associated with bioplastic processing and production in either large or small scale and if emulated can help reduce cost production in commercial venture. Furthermore, the larger-is-better signal-to-noise model can be modified to include relative error, which could further help give credence to the outcome of scientific investigation. Therefore, it can be said that Taguchi technique is a versatile tool that could be use for optimization and estimate errors in outcome of plasticisation process of glycerol and TEA plasticised thermoplastic mango starch with optimum conditions that could be used for up-scale commercial production.

Declarations

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Figures

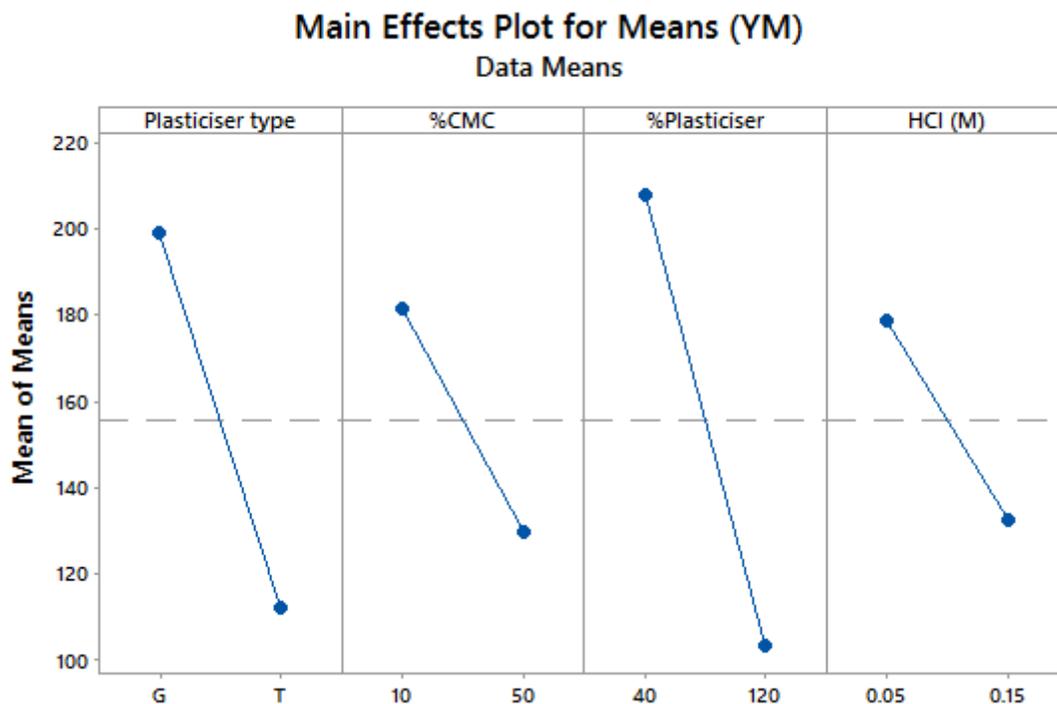
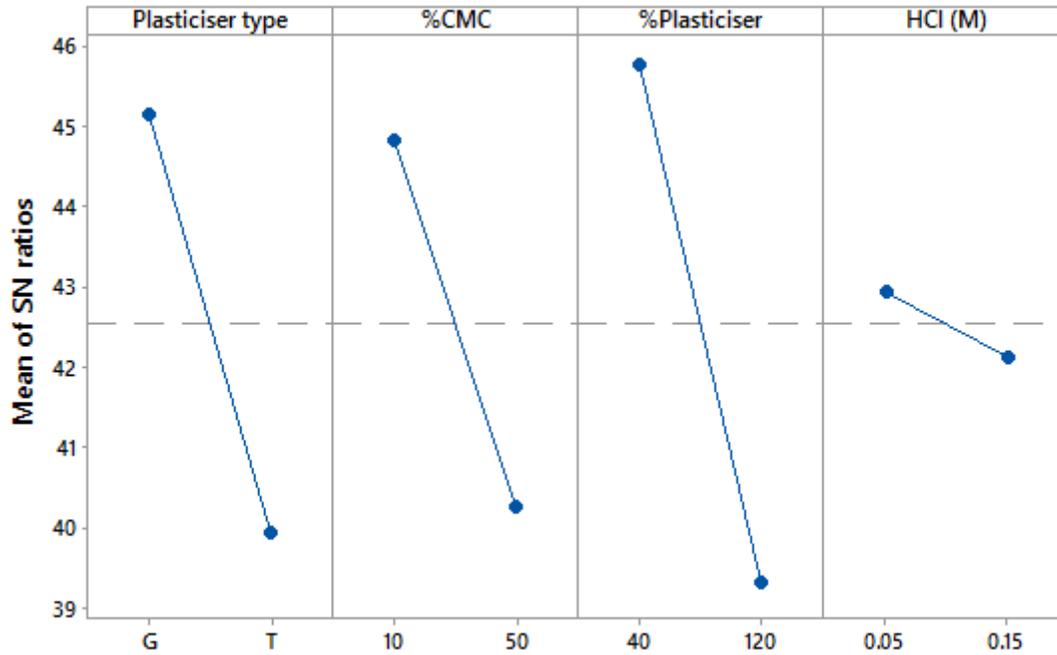


Figure 1

Main Effects Plot for Mean (Young modulus)



Signal-to-noise: Larger is better

Figure 2

Data Means Main Effects Plot for Signal-Noise Ratio

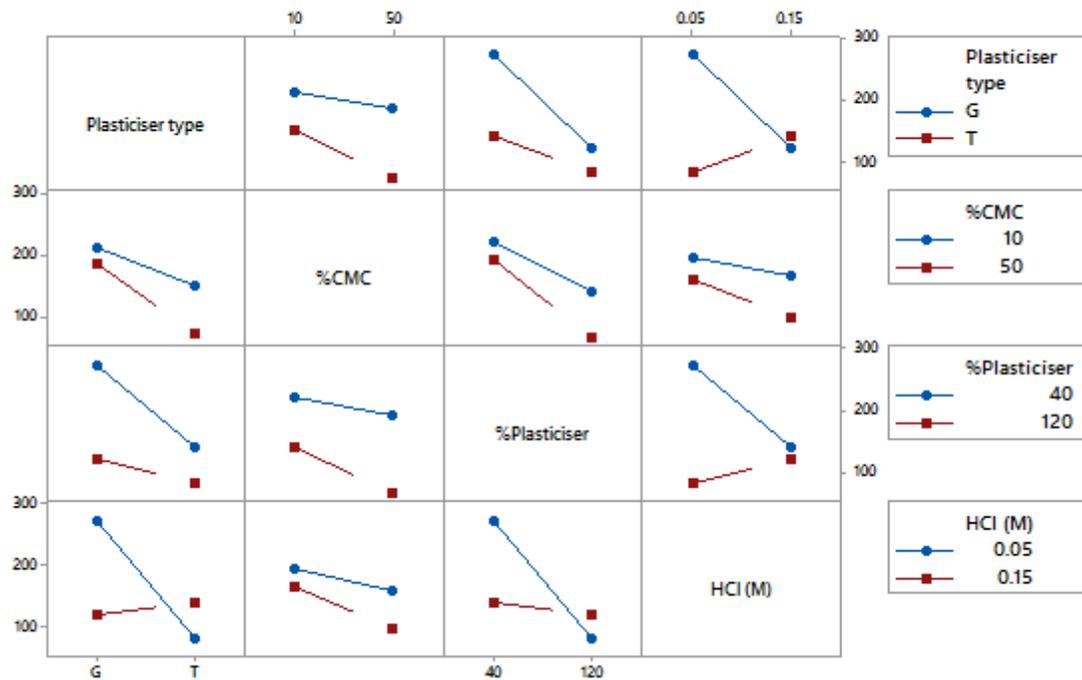


Figure 3

Data Means Interaction Plot for Young Modulus

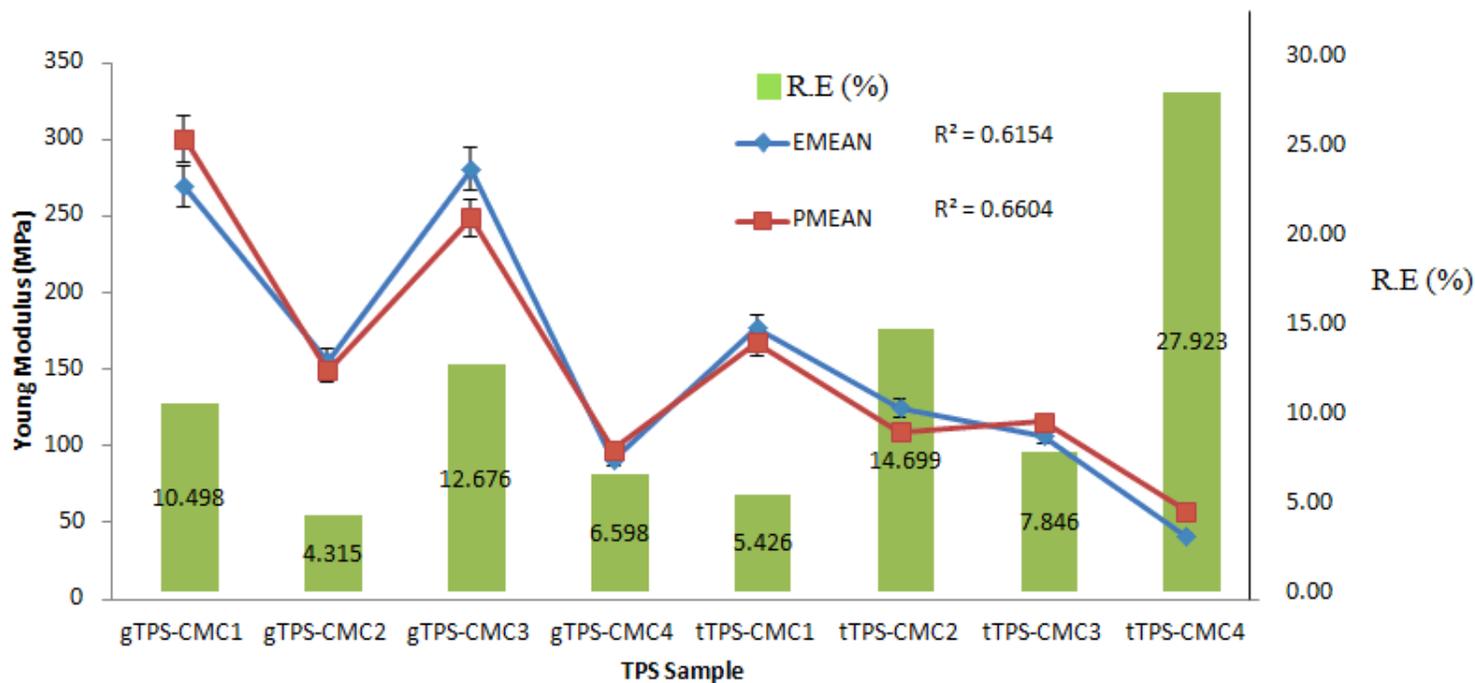


Figure 4

Experimental and Predicted mean (Young Modulus) and Relative error. Key: R.E is absolute value where R.E: Relative error, PMEAN: Predicted mean and EMEAN: Experimental derived mean.