

*Mechanical properties and thermal conductivity of epoxy composites reinforced with recycled clamshell container waste as a micro filler (RCCF) were studied. The studies have been carried out to identify the influence of the two variables, the heating time periods (HT) within the range of 2, 4, 6 min., and wt % within the range of 1 %, 2 %, 4 % of recycled clamshell container waste that has been used as a reinforcing filler of epoxy composites. Recycling polyurethane waste aims to control and maintain a pollution-free environment, which is currently considered a difficult issue in addition to achieving low-cost aspects in preparing the composites. According to the method of no-combustion heating, the clamshell waste was converted from the natural plastic state into solids that were later made into 75 µm micro filler by grinding. Composites were ranked using grey relational analysis (GRA). The effect of each control parameter on response variables was analyzed by the Taguchi method. Using MINITAB 19 software, regression equations were obtained for each variable of mechanical properties and thermal conductivity to predict the properties of epoxy composites. The results of the addition of recycled clamshell container waste to epoxy resin show an improvement in the mechanical properties and thermal conductivity of the composites. The optimal value of the two factors was at HT2wt2, i. e. HT and wt % of 4 min and 2 %, respectively. The optimization values for the bending strength, impact strength, tensile strength, stiffness and thermal conductivity are 68.2 MPa, 10.348 kJ/m<sup>2</sup>, 21.08 MPa, 80 Shore D and 0.504 W/m·C°, respectively. The proposed Taguchi methodology based on grey relational analysis has been shown to be effective in solving multi-feature decision-making problems*

**Keywords:** recycling polyurethane/clamshell waste, mechanical properties, thermal conductivity, Taguchi method

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# OPTIMIZATION OF MECHANICAL PROPERTIES OF RECYCLED POLYURETHANE WASTE MICROFILLER EPOXY COMPOSITES USING GREY RELATIONAL ANALYSIS AND TAGUCHI METHOD

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## 1. Introduction

The current research highlights the problem of the operation costs of transportation tools (rail and airplane, etc.), which are proportional to their weight, so using light, strong composites will lower operation costs. Here, light weight and high mechanical strength are the structural properties of advanced composites. Therefore, the objectives of the current research are in line with recent research trends that urge researchers to use polymers as light materials after strengthening them with other materials or fillers of industrial or natural sources. Therefore, the effect of adding plastic waste as reinforcing materials to epoxy composites on their mechanical and thermal properties was studied. So, studies that are devoted to developing polymer composites aim to solve the weight and cost problems associated with the materials currently used.

## 2. Literature review and problem statement

Recent years have witnessed an increase in the use of polymer composites, which has led to a growing interest among researchers to produce and evaluate the mechanical and thermal properties of recycled thermoplastic composites derived from non-renewable and non-biodegradable plastics that constitute the large part of today's discarded solid waste [1]. Polymer composites through their improved properties like hardness, high specific strength, toughness, etc. have proven themselves within materials science. For example, recycled polyethylene terephthalate (RPET) is used as a matrix composition in a wide variety of polymer matrix compounds. Composite materials based on a matrix of recycled waste, in addition to being cost-effective, are also considered environmentally friendly. Recycling plastic waste for use as matrix material not only reduces waste consumption, but also addresses concerns about environmental pollution

due to post-consumer PET waste [2]. The wide use of polymer composites requires a great deal of knowledge in this field, specifically their mechanical, thermal, physical properties [3]. In this study, the researchers strengthened the epoxy with waste tire rubber (WTR) within the ratios of 1–20 % by weight to be reused as an epoxy coating. The study aims to improve the mechanical and tribological properties. The hardness property has improved by 22 % as compared to the original epoxy coating at 5 wt % WTR loading. WTR-reinforced epoxy coatings showed promising properties at optimum loading of 5 wt % [4]. Another study dealt with the development of hybrid mixed composites using epoxy resin derived from waste polyethylene terephthalate and benzoxazine bottles enhanced with bio-functional carbon. Part of the study dealt with thermal properties and thermal-mechanical behavior using different analytical methods. The results indicate an improvement in the thermal and mechanical properties and corrosion resistance. The study also concluded that this work can be considered an effective way to use waste products and sustainable biomaterials to develop high-performance hybrid composites [5]. The study dealt with the mechanical properties of epoxy composites that were reinforced with two types of natural waste as a micro filler, with different weight ratios and particle size, pollen 50  $\mu\text{m}$  and sea shells 75  $\mu\text{m}$ . The aim of the study is to recycle natural waste and study the effect of filler percentages on the mechanical behavior of epoxy composites. The study concluded that the mechanical properties of epoxy composites improved significantly [6]. Another study examined the effect of fiber content and length on the mechanical behavior of epoxy composites reinforced with bamboo fibers. The results confirmed that the flexural modulus and fracture toughness of epoxy composites increase monotonously with length and fiber content [7]. Many researchers dealt with the process of data analysis and prediction of response parameters using the Taguchi method and grey analysis, which led researchers in various fields to adopt the Taguchi method and link it to the grey relational analysis method in order to optimize the research outputs. The principle of the method is based on an idea that links the empirical and analytical concepts to determine the most influential variable in the output response to reach an improvement in system performance [8]. Another study dealt with the effect of adding varying weight percentages of tungsten and copper powder on the mechanical properties of epoxy compounds. The results showed a clear improvement in the properties of epoxy composites, and the ideal condition was recorded with a weight ratio of 5 % [9].

Our review of studies on the mechanical properties and thermal conductivity of polymer composites indicates that the available publications relate only to certain aspects of the influence of different parameters on the mechanical properties of epoxy composites. It does not exhaust the need for a comprehensive analysis of the dependence of this property of composites on a set of specific factors (filler concentration, nature of filler, temperature, etc.). All researches share the main goal, which is to improve the mechanical properties of polymers because they differ according to the different factors involved in their composition.

### 3. The aim and objectives of the study

The aim of the study is to improve the mechanical properties and thermal conductivity of epoxy composites reinforced with recycled clamshell waste micro filler.

To achieve this aim, the following objectives are accomplished:

- studying the properties of epoxy composites reinforced with clamshell containers waste micro filler (RCCF) by determining the optimization value of each response of mechanical properties and thermal conductivity of epoxy composites under two factor effect (HT and wt%) by S/N ratio;
- studying the overall performance response through grey relational grade (GRG) outcome analysis;
- finding a regression equation that predicts the maximum values of mechanical and thermal properties based on the ideal values of HT and wt % factors associated with each case for the properties of epoxy composites specified in the current research.

### 4. Research materials and methods

Polyurethane composites include epoxy, unsaturated polyester, and phenols. Clamshell containers for food processing are one-piece containers consisting of two halves joined by a hinge area that allows the structure to come together to seal (Fig. 1). The current study used polyester clamshell. The research materials included the following:

a) Reinforcing materials: The 75  $\mu\text{m}$  micro filler was produced from the waste clamshell container, which was used as reinforcing of epoxy composites. Residues of clamshell containers were collected from the landfill of Shiftah village, Baqubah city, Diyala, Iraq;

b) Adhesive: The resin used in this work is epoxy with sika-dur32 hardener and a density of 1.1 kg/L, obtained from the chemical warehouse in Baghdad, Iraq.



Fig. 1. Clamshell container waste

The steps for preparing the fine grouting to reinforce the epoxy composites are as follows:

1. Collect clamshell waste from landfills, wash it well with soap and water, and then cut it into medium pieces.

2. The waste pieces are placed in a metal container at low heat (cooking gas) and left for a period of time (HT: the time is recorded), then it turns into a thick molten substance, noting that the melt acquires a darker color with an increase in heating time.

3. The melt is left in the container to cool at room temperature and turns into a solid substance. The resulting materials are broken down and ground well. Using a 75  $\mu\text{m}$  sieve, the required granular size is screened, the micro filler is then ready to be used as a filler for reinforcing epoxy composites.

It should be noted that every 10 pieces of clamshells turn into 50.7 grams of flour stuffing, the dimensions of one clamshell piece are  $24 \times 15 \times 6.5 \text{ cm}^3$ .



Fig. 2. The form of a melt of recycled waste after cooling and RCCF 75 μm at different heating time: *a, b* – 2 min; *c, d* – 4 min; *e, f* – 6 min

Samples formulation was done using the rule of mixture. The composition of the composite has been prepared by mixing various weight ratios (wt %) from a pre-prepared micro-filler from recycled clamshell waste (RCCF) with the base of epoxy. Heating time is 2, 4, 6 minutes and weight percentage is 1, 2, 4 % as shown in Table 1. According to the weight ratios in the research, the RCCF was added gradually to the epoxy during the process of mechanical stirring of the mixture in a plastic jar and the depended ratio in the mixing of epoxy to the hardener was 2:1. The mixture was poured into the mold, the mold was allowed to be kept at room temperature for 24 hours. A plastic mold of dimension 200×130×5 mm<sup>3</sup> was used for casting the composites. Castings were removed from the mold after 24 hours. They were cut into varying shapes and dimension according to ASTM standards for mechanical properties and thermal conductivity by CNC machine.

The mechanical behavior of tensile, impact, flexural and hardness tests was achieved on the epoxy composites. Impact test was carried out according to ASTM D256 using an IZOD impact tester. The tensile test by the universal test machine (JIANQIAO) was used according to ASTM D638. The flexural tests were performed using a three-point bend, according to ASTM 790. The hardness tests were accomplished according to ASTM Shore D. Samples thermal conductivity was as per ASTM-D790 and measured by using the Lee’s disc type (Griffin and George/England).

Table 1

Content details for the composition of composites

Sample No.	Epoxy, %	Recycled clamshell container filler (RCCF), wt %	Heating time (HT), minutes
Free (original sample)	100	0	0
Group 1	1th 752	99	1
	2th 752	98	2
	3th 752	96	4
Group 2	1th 754	99	1
	2th 754	98	2
	3th 754	96	4
Group 3	1th 756	99	1
	2th 756	98	2
	3th 756	96	4



Fig. 3. Types of specimens used in practical tests: *a* – 1 impact, 2 flexural, 3 tensile; *b* – thermal conductivity

The dimensions of the samples were adopted for the practical side according to the standard specification as shown in Fig. 3, *a* for impact, flexural and tensile tests, respectively, while the thermal conductivity samples in Fig. 3, *b* are circle shape (40×4 mm).

4. 1. Optimization procedure (S/N ratio analysis)

Using the Taguchi method, the experimental pattern was performed with the aim of analyzing the input parameters of the data to reduce the number of experiments. The procedures of the Taguchi method use the loss function to determine the deviation between the experimental values and the desired data. The loss function is converted into a signal/noise (S/N) ratio. There are three kinds of quality characteristics in the analysis of the S/N ratio, namely larger-the-better, smaller-the-better and nominalthe best. For each level of the process parameters, the S/N ratio is determined based on the S/N analysis. The purpose of this analysis is to obtain the minimum response of thermal conductivity and optimize the mechanical properties response of epoxy composites. Based on the Taguchi method, the L9 orthogonal array was selected. The smaller-the-better and the larger-the-better quality characteristics shown in (1), (2), respectively, were used to calculate the signal to noise ratios of tensile, flexural, hardness, impact strength and thermal conductivity of the epoxy composites:

$$\text{smaller – the – better : } S / N = -10 \lg_{10} \left( \frac{1}{n} \sum_{i=1}^n y_{ij}^2 \right), \quad (1)$$

$$\text{larger – the – better : } S / N = -10 \lg_{10} \left( \frac{1}{n} \sum_{i=1}^n \frac{1}{y_{ij}} \right), \quad (2)$$

where  $Y_i$  – experimental outcome of  $i^{\text{th}}$  experiments;  $n$  – number of factor level combinations,  $i$  – entire number of experiments.

#### 4. 2. Grey relational analysis (GRA)

Taguchi design with grey relational analysis is the better solution to determine the combined response parameters. In the GRA, the measurement performance characteristic is normalized from zero to one. This process is known as a grey relational generation. Then being on the normalized data, the grey relational coefficient will be calculated. Then grey relational grade is found by averaging the grey relational coefficients. The overall performance response depends on the grey relational grade. This process converts a multi-response optimizing problem into one-response optimizing. The GRG being the objective function when smaller-the-better is a characteristic of the experimental data, the experimental data can be normalized as follows:

$$x_i^*(k) = \frac{\max x_i(k) - x_i(k)}{\max x_i(k) - \min x_i(k)}, \quad (3)$$

$$x_i^*(k) = \frac{x_i(k) - \min x_i(k)}{\max x_i(k) - \min x_i(k)}. \quad (4)$$

When larger-the-better is a characteristic of the experimental data, then the experimental data can be normalized as follows, where:

–  $i=1, \dots, m; k=1, \dots, n$ , ( $m$  is the amount of experimental data,  $n$  is the number of responses and  $X$  is the desired value);

–  $x_i^*$  – the sequence after the data preprocessing,  $x_i(k)$  is the original sequence, (the normalized value).  $\max x_i(k)$  is the largest value of  $x_i(k)$  and  $\min x_i(k)$  is the smallest value of  $x_i(k)$ .

The objective is to obtain the highest mechanical properties (larger-the better). The grey relational coefficient can be calculated from (5):

$$\xi_i(k) = \frac{\Delta_{\min} + \zeta \Delta_{\max}}{\Delta_{oi}(k) + \zeta \Delta_{\max}}, \quad (5)$$

$$\Delta_{oi} = \|x_o(k) - x_i(k)\|, \quad (6)$$

where  $\xi_i(k)$ , the identification coefficient that lies between 0 to 1 and usually taken as 0.5, is the deviation sequence of the reference sequence,  $x_o(k)$  is the reference sequence,  $x_i(k)$  is termed as comparability sequence,  $\Delta_{\max}$ ;  $\Delta_{\min}$  are the maximum and minimum values of the absolute differences ( $\Delta_{oi}$ ) of all compared sequences, respectively. From (7), the value of the GRG can be found:

$$\gamma_i = \frac{1}{n} \sum_{k=1}^n \xi_i(k), \quad (7)$$

where  $\gamma_i$  is the required GRG for  $i^{\text{th}}$  experiment and  $n$  is the number of response characteristics. The parameter combination with the highest GRG is the optimal condition for combined process parameters:

$$\gamma^{\wedge} = \gamma_m + \sum_{i=1}^o (\gamma_i^- - \gamma_m), \quad (8)$$

where  $\gamma_m$  is the total GRG,  $\gamma_i^-$  is the mean grey relational grade at the optimal level of each parameter and 0 is the number of the significant process parameters.

### 5. Results of the properties of epoxy composites reinforced with recycled polyurethane waste

#### 5. 1. Results of optimization response of epoxy composites properties using S/N ratio

Table 2 shows the values of experimental results of mechanical properties and thermal conductivity of the composites. Experiments are planned as per Taguchi  $L_9$  orthogonal array taking two factors (process parameters) such as weight percentage (wt %) of RCCF and heating time (HT) of melt at three levels. The level of wt % from 1 wt % as level 2, 1 wt % as level 2 and 4 wt % as level 3 similarly, the ranges of heating time have been taken like 2 min as level 4, 1 min as level 2, 6 min as level 3, respectively. Taguchi  $L_9(3^2)$  orthogonal array design constitutes a total of 9 experimental runs. The S/N ratio was calculated from (1), (2), the experimental results of mechanical properties and thermal conductivity are shown in Table 3. In order to compare the practical results of the samples for the current research, the results of the sample free from RCCF were added to Table 1.

Table 2

Experimental results of mechanical properties and thermal conductivity of composites

Samples	(RCCF), wt %	(HT), min	Hard.(H) (Shore D)	Impact Strength (IMP) (kJ/m <sup>2</sup> )	Flexural Strength (Fs), MPa	Tensile Strength (Ts), MPa	Thermal Conductivity (THE) (W/m.C <sup>o</sup> )	
Free (original sample)	0	0	70	7	60	25	0.58	
Group 1	1th75-2	1	2	79.26	5.59	67.79	21.51	0.566
	2th75-2	2	2	80.66	5.23	62.21	27.19	0.502
	3th75-2	4	2	79.4	9.33	74.37	15.28	0.511
Group 2	1th75-4	1	4	80.6	9.24	67.38	18.07	0.464
	2th75-4	2	4	80.46	6.13	74.21	15.22	0.477
	3th75-4	4	4	80.73	11.43	75.73	26.28	0.507
Group 3	1th75-6	1	6	81.4	5.03	62.1	26.82	0.4805
	2th75-6	2	6	78.73	5.35	66.42	14.29	0.52003
	3th75-6	4	6	78.26	11.88	70.08	24.92	0.5184

The optimum response results of the mechanical properties and thermal conductivity of RCCF reinforced epoxy compounds under the influence of two factors HT and wt % are shown in Fig. 4–9.

The optimal value of the parameters in order to achieve the highest hardness is HT2wt1. Fig. 4 shows the graph of the S/N ratio of average hardness.

The optimal value of impact strength is achieved at HT2 wt3. Fig. 5 represents the relationship for the average S/N ratio and impact strength.

The optimal parameters of the flexural strength of the composites are at HT2 wt2. Fig. 6 shows the mean S/N ratio graph for flexural strength.

Table 3

S/N ratio of experimental results as per Taguchi L<sub>9</sub> orthogonal array

Group of composites No.	Samples	(RCCF), wt %	(HT), min	S/N ratio (dB) of (H) (Shore D)	S/N ratio (dB) of (IMP), kJ/m <sup>2</sup>	S/N ratio (dB) of (Fs), MPa	S/N ratio (dB) of (Ts), MPa	S/N ratio (dB) of (THE) (W/m·C°)
Group 1	1 <sup>th</sup> 75-2	1	2	37.981	14.948	36.623	26.653	4.944
	2 <sup>th</sup> 75-2	2	2	38.133	14.370	37.428	28.688	5.986
	3 <sup>th</sup> 75-2	4	2	37.996	19.398	35.877	23.683	5.832
Group 2	1 <sup>th</sup> 75-4	1	4	38.127	19.313	36.571	25.139	6.670
	2 <sup>th</sup> 75-4	2	4	38.112	15.749	37.409	23.648	6.430
	3 <sup>th</sup> 75-4	4	4	38.141	21.161	37.585	28.393	5.900
Group 3	1 <sup>th</sup> 75-6	1	6	38.213	14.031	35.862	28.569	6.366
	2 <sup>th</sup> 75-6	2	6	37.923	14.567	36.446	23.101	5.679
	3 <sup>th</sup> 75-6	4	6	37.871	21.496	36.912	27.931	5.707

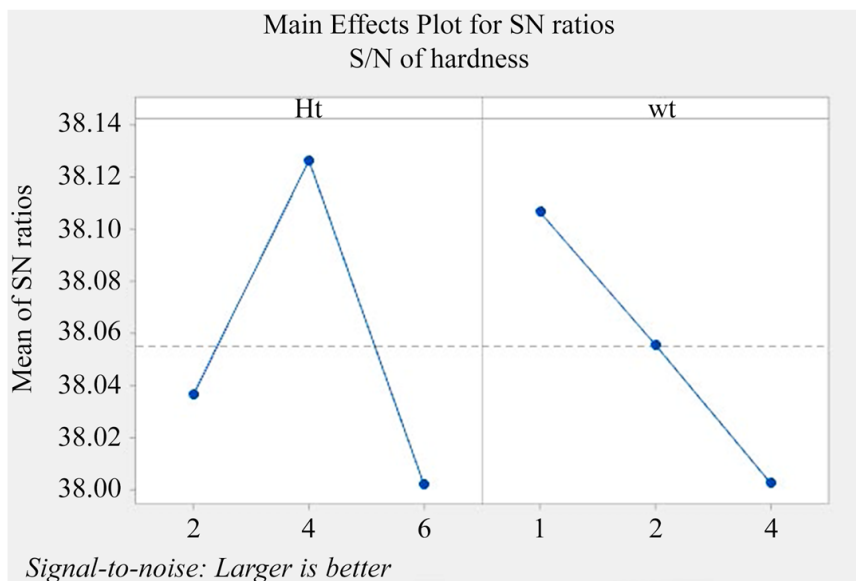


Fig. 4. Mean S/N ratio graph for hardness

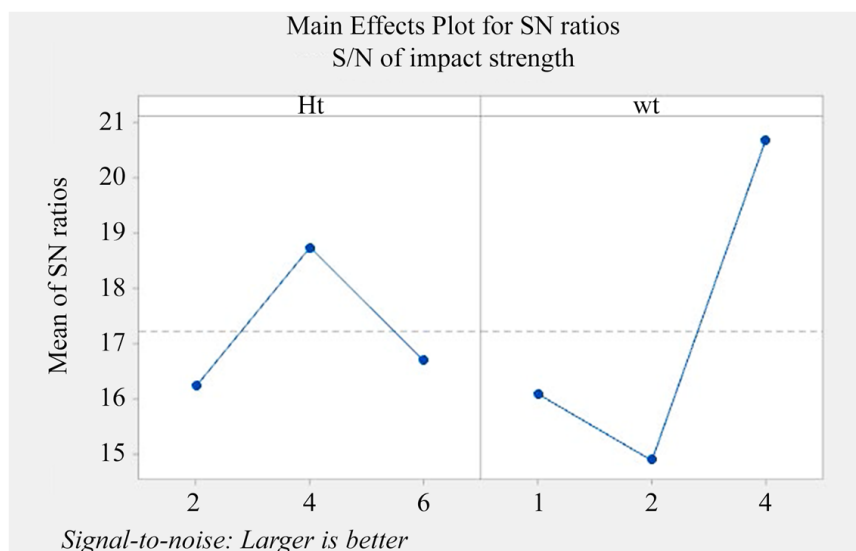


Fig. 5. Mean S/N ratio graph for impact strength

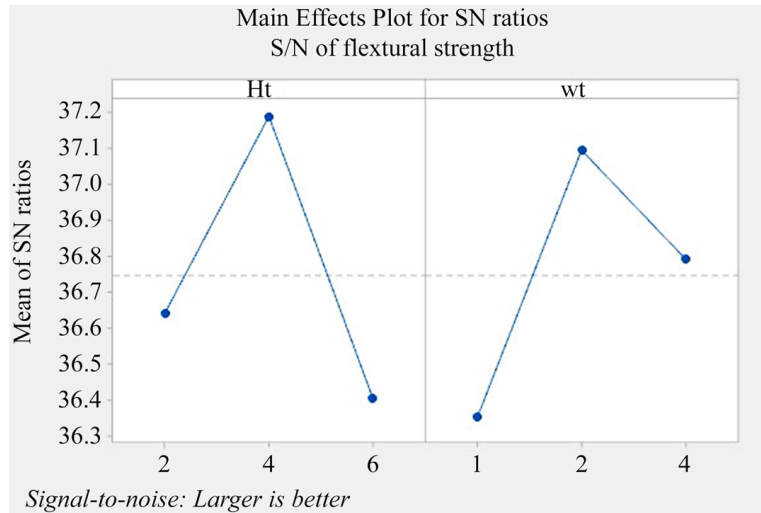


Fig. 6. Mean S/N ratio graph for flexural strength

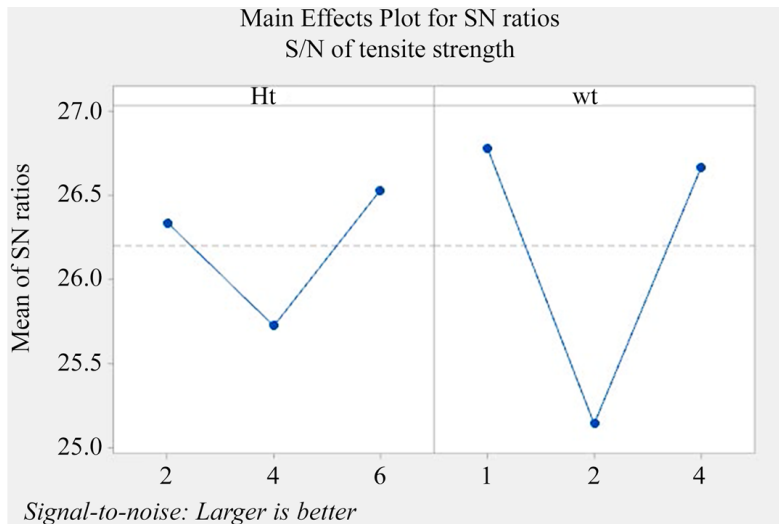


Fig. 7. Mean S/N ratio for tensile strength

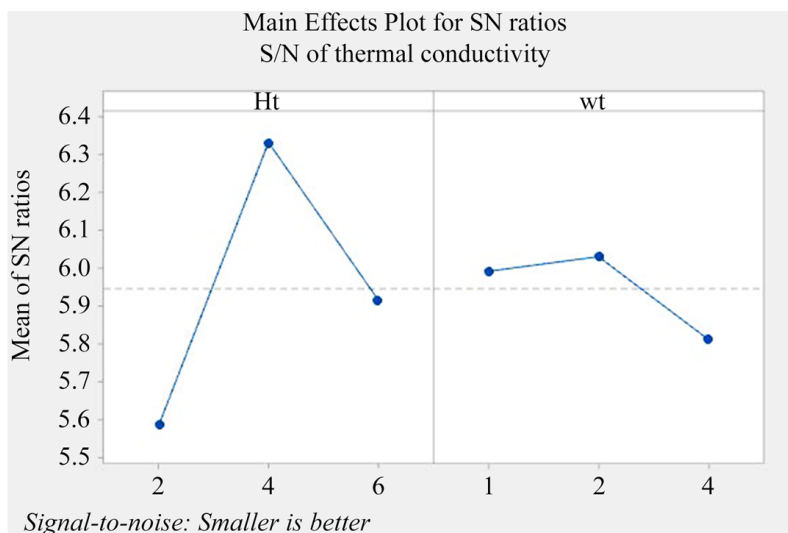


Fig. 8. Mean S/N ratio for thermal conductivity

Fig. 7 shows the mean S/N ratio for tensile strength and the optimal parameters of the flexural strength of the composites are achieved at HT3 wt1.

Fig. 8 shows the mean S/N ratio for thermal conductivity. It can be noted that the optimum factors for thermal conductivity are HT2wt2.

It is clear that all the optimal responses to the mechanical and thermal properties of epoxy compounds in the above figures are achieved at H and F varying effect. This effect is related to the nature of the property.

**5. 2. Results of the overall performance response of composites properties by determining the response of grey relational grade**

By adopting the procedure described in [10], a grey relational analysis was performed. The methodology was adopted to find the multi-response of parametric optimization.

Step 1: the data obtained from the S/N ratio analysis shown in Table 3 were used to calculate the grey relational generations (Normalized) shown in Table 4 with smaller- and larger-the better, given in (3), (4).

**Grey relational generations (Normalized)**

Composites Groups No.	Comp. No.	(H) Larger-the-better	(IMP) Larger-the-better	(Fs) Larger-the-better	(Ts) Larger-the-better	(THE) Smaller-the-better
Group 1	1th 75-2	0.323	0.123	0.442	0.636	1.000
	2th 75-2	0.768	0.045	0.909	1.000	0.396
	3th 75-2	0.368	0.719	0.009	0.104	0.486
Group 2	1th 75-4	0.749	0.708	0.411	0.365	0.000
	2th 75-4	0.705	0.230	0.898	0.098	0.139
	3th 75-4	0.790	0.955	1.000	0.947	0.446
Group 3	1th 75-6	1.000	0.000	0.000	0.979	0.176
	2th 75-6	0.152	0.072	0.339	0.000	0.574
	3th 75-6	0.000	1.000	0.609	0.864	0.558

Step 2: the grey relational coefficient was calculated using (5) from the normalized data as shown in Table 6. The value of the distinguishing coefficient is taken as 0.5 as the equal weight has been given to all quality characteristics. The deviation sequence results are calculated using (6) as illustrated in Table 5.

**Table 5**

**The deviation data ( $\Delta oi$ )**

Composites Groups No.	Composites No.	Evaluation of ( $\Delta oi$ )				
		H	IMP	Fs	Ts	THE
Group 1	1th 75-2	0.677	0.877	0.558	0.364	0.000
	2th 75-2	0.232	0.955	0.091	0.000	0.604
	3th 75-2	0.632	0.281	0.991	0.896	0.514
Group 2	1th 75-4	0.251	0.292	0.589	0.635	1.000
	2th 75-4	0.295	0.770	0.102	0.902	0.861
	3th 75-4	0.210	0.045	0.000	0.053	0.554
Group 3	1th 75-6	0.000	1.000	1.000	0.021	0.824
	2th 75-6	0.848	0.928	0.661	1.000	0.426
	3th 75-6	1.000	0.000	0.391	0.136	0.442

Step 3: the result of grey relational grade (GRG) was calculated using (7) from the results data of grey relational coefficients. The results of grey relational grade (GRG) are illustrated in Table 6. The 3<sup>th</sup> 75-4 composite is represented as Rank No. 1. Then, this result is used for optimizing the multi-response parameters as it is converted to a single grade.

**Table 6**

**Grey relational coefficient and grey relational grade (GRG) data**

Composites Groups No.	Composites No.	Grey relational coefficient					GRG	Rank
		H	IMP	Fs	Ts	THE		
Group 1	1 <sup>th</sup> 75-2	0.425	0.363	0.473	0.579	1.000	0.568	5
	2 <sup>th</sup> 75-2	0.683	0.344	0.846	1.000	0.453	0.665	2
	3 <sup>th</sup> 75-2	0.442	0.640	0.335	0.358	0.493	0.454	8
Group 2	1 <sup>th</sup> 75-4	0.666	0.631	0.459	0.440	0.333	0.506	7
	2 <sup>th</sup> 75-4	0.629	0.394	0.830	0.357	0.367	0.515	6
	3 <sup>th</sup> 75-4	0.704	0.918	1.000	0.904	0.474	0.800	1
Group 3	1 <sup>th</sup> 75-6	1.000	0.333	0.333	0.959	0.378	0.601	4
	2 <sup>th</sup> 75-6	0.371	0.350	0.431	0.333	0.540	0.405	9
	3 <sup>th</sup> 75-6	0.333	1.000	0.561	0.787	0.531	0.642	3

**Table 4**

Step 4: from the grey relational grade (GRG) value, the influence of all parameters at various levels is shown in Fig. 9. From the value of GRG, the influence of each parameter at various levels is plotted in Fig. 9. The mean GRG is illustrated in Table 7.

Total mean grey relational grade=0.561

Step 5: to determine the improvement in the GRG, confirmation experiments are performed from the initial parameter setting to the optimal parameters obtained in the epoxy composites using RCCF addition.

The predicted grey relational grade can be determined using (8). The GRG at optimal value becomes 0.609, which is close to the predicted value of 0.670. Table 8 shows the confirmation run, it appears that the GRG of both responses such as mechanical properties and thermal conductivity is significantly improved (0.041) through the setting of optimal combination. The above analysis shows that the parameters improved the minimum values of thermal conductivity and the maximum values of mechanical properties by analyzing the grey relationships based on the Taguchi method. The optimal grey relational grade is predicted at the selected optimal setting controllable variable. The most significant parameter with the optimal level was already selected as HT2 followed by wt3. The HT2wt3 is an optimal parameter combination of the mechanical properties and thermal conductivity of RCF/epoxy composites obtained by means of GRG and was considered as a confirmation test.

Finally, it is shown clearly that the mechanical properties and thermal conductivity of RCCF/epoxy composites were significantly improved by 0.041, a good agreement between the actual and predicted GRG was obtained.

**Table 7**

**Main effects on grey relational grade**

Factors	Mean grey relational grade			Max-Min	Rank
	Level 1	Level 2	Level 3		
HT	0.5741	0.5935	0.5155	0.0780	2
wt %	0.5668	0.4905	0.6258	0.1353	1

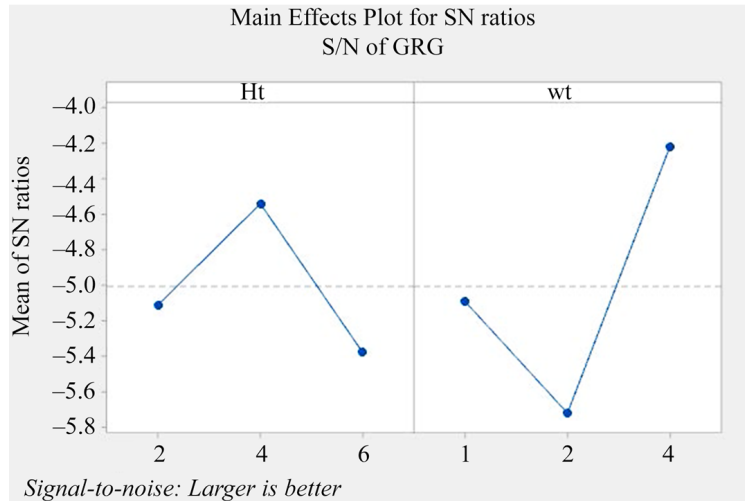


Fig. 9. Main effect of grey relational grade (GRG)

Table 8

Results of the confirmation test

State of parameter	Setting level	H	IMP	Fs	Ts	THE	GRG
Initial controllable parameters	HT1wt1	79.26	5.59	67.79	21.51	0.566	0.568
Optimum controllable parameters	HT2wt3	79.42	10.37	70.28	21.489	0.5049	0.609
Grey relational grade predicted	HT2wt3	81.072	12.123	75.185	23.078	0.477	0.670

Note: Improvement in GRG=0.041

5. 3. Results of regression models of epoxy composites properties

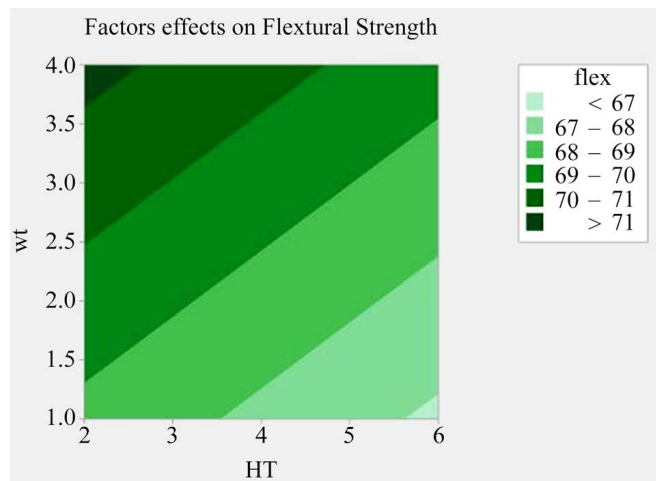
Regression analysis is used for modeling and analyzing several variables due to the presence of a relationship between a dependent variable with one or more independent variables. In the present study, the dependent variables are the mechanical properties and thermal conductivity of epoxy composites, whereas the independent variables are the weight percentage of recycled clamshell and heating time. By using MINITAB 19 software, the regression models for each output dependent variable of mechanical properties and thermal conductivity were obtained. The optimal values HT2 wt2 obtained from grey relational analysis as shown in Fig. 9 were used to determine the optimum performance of the epoxy composites as follows.

5. 3. 1. Regression equation for flexural strength

$$Fs = 68.4 - 0.48HT + 0.86wt. \tag{9}$$

The contours for the flexural strength of the epoxy composites reinforced with RCCF are shown in Fig. 10.

The optimal value of flexural strength is 68.2 MPa, which was obtained from (9).



5. 3. 2. Regression equation for impact strength

$$IMP = 3.26 + 0.176HT + 1.596wt. \tag{10}$$

The regression equation (10) of impact strength is represented in Fig. 11.

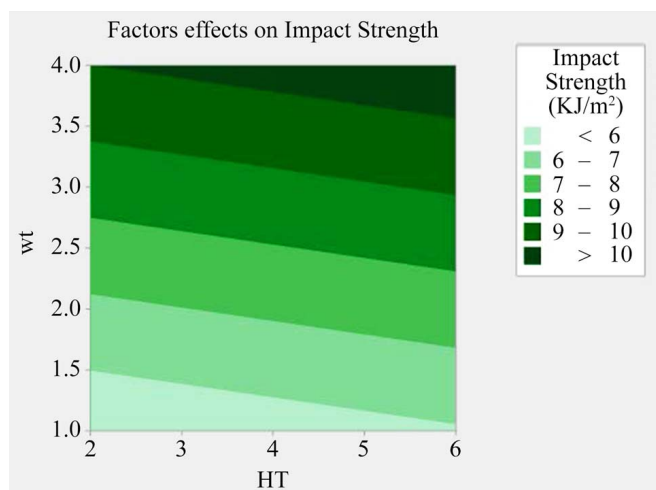


Fig. 11. Contour plots for impact strength



The contours for the impact strength of the epoxy composites reinforced with RCCF are shown in Fig. 11 and the optimal value is 10.348 kJ/m<sup>2</sup>.

**5. 3. 3. Regression equation for tensile strength**

$$T_s = 19.82 + 0.17HT + 0.24wt. \tag{11}$$

The regression equation (11) of tensile strength is represented in Fig. 12.

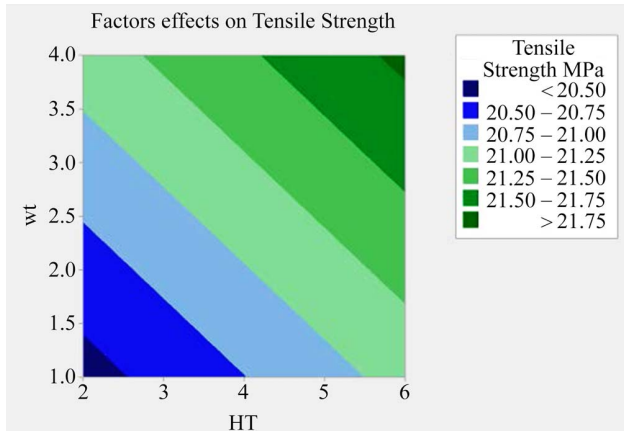


Fig. 12. Contour plots for tensile strength

The optimal value of the tensile strength of the epoxy composites/RCCF is 21.08 MPa as shown in the contour in Fig. 12, which was obtained from (11).

**5. 3. 4. Regression equation for hardness (Shore D)**

$$Hard = 80.97 - 0.077HT - 0.308wt. \tag{12}$$

The regression equation (12) of hardness is represented in Fig. 13.

The optimal value of the hardness of the epoxy composites/RCCF is 80 (Shore D) as shown in the contour in Fig. 13.

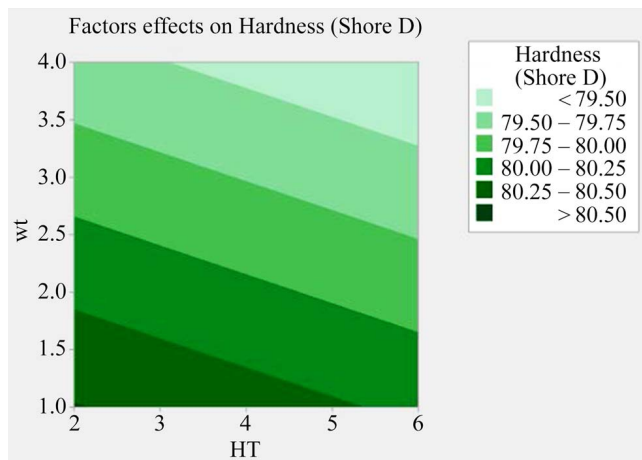


Fig. 13. Contour plots for hardness (Shore D)

**5. 3. 5. Regression equation for thermal conductivity**

$$Therm = 0.5173 - 0.00501HT + 0.0033wt. \tag{13}$$

The regression equation (13) of thermal conductivity is represented in Fig. 14.

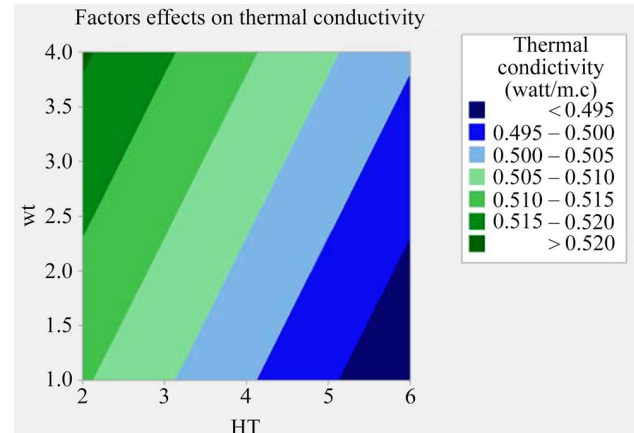


Fig. 14. Contour plots for thermal conductivity

The optimal value of thermal conductivity of the epoxy composites/RCCF is 0.504 W/m·C° as shown in the contour in Fig. 14.

**6. Discussion of results optimization of composites properties reinforced with recycled micro filler**

According to the data of the current research, the conditions of the factors that led to the improvement of the results of the mechanical properties and thermal conductivity of RCCF micro-filler reinforced epoxy composites are clearly shown in Fig. 4–14. From Fig. 4, the ideal state of hardening of the epoxy composites is achieved when the parameters are HT2wt1, which means HT of 4 min and wt % of 1%. This may happen because longer heating causes the recycled materials to lose elasticity and turn into more brittle and hard materials. Also, Fig. 5 describes the optimal factors to maintain the maximum impact strength achieved at the HT2 wt3 state in which the maximum weight % of RCCF is about 4 % and HT is about 4 min. The decrease in impact strength with increasing heating time may be due to the increase in RCCF brittleness, which leads to a decrease in the interatomic bonding. An increase in the fine filler concentration leads to the filling of the interstitial voids in the composite structures, thereby increasing the impact strength. It can be seen that the impact strength is high at the higher factor of HT. Fig. 6 shows the optimal parameters of the flexural strength of the composites at HT2 wt2, i. e. HT of 4 min and wt % of 2 %. An increase in the weight percentage of the RCCF epoxy reinforcement increases the bending resistance, so it is high at the HT. In Fig. 7, the optimum tensile strength values are achieved when the factors are HT3 wt1. This condition is achieved when the heating time is 6 min and wt % is 1 %, while the lowest tensile values are at 2 min of HT3 and 2 % of wt %. In this state, the weight ratio factor appears to have the most influence on the tensile property compared to the highest value of the heating time factor. This behavior may be caused by the retention of epoxy's elastic properties at low percentages of fillers added. In Fig. 8, it can be noted that the optimum factor for thermal conductivity is HT2wt2. It means that HT at 4 min and wt % at 2 % are the optimum factors for lower

thermal conductivity. The lowest thermal conductivity of the epoxy composites is achieved at the highest value of heating time (4 minutes) compared to the highest value of wt % (2 %), which may be due to the good diffusion of the added micro-filler between the epoxy atoms. Fig. 9 shows the main effect of grey relational grade (GRG).

The optimal parameter formulation is selected based on the higher GRG value, which means a stronger correlation to the reference sequence and better performance. Thus, the optimal tuning for multi-responses is HT2wt3, i.e. 4 min of HT and 4 % of wt %. In Fig. 9, the higher value of mean grey relational grade (GRG) represents the maximum value of mechanical properties of epoxy composites and the minimum value of thermal conductivity. The variance of the maximum and minimum results of mean grey relational grade (GRG) for turning parameters was 0.078 for the heating time (HT) factor and 0.1353 for the weight percentage factor (wt %) shown in Table 7. This result indicates that the wt % factor has a major effect on multi-response compared to the HT factor.

The contours for the mechanical properties and thermal conductivity of the recycled clamshell filler reinforced epoxy composites are shown in Fig. 10–14. They indicate how a change in HT and wt % affects the properties of the recycled clamshell waste micro-filler/epoxy composites. The contour levels shown in Fig. 10, 11 revealed that the flexural strength ( $F_s$ ) of 69–70 MPa can be achieved using HT of 4 minutes and wt of 2 %. While the impact strength (imp) of 9–10 kJ m<sup>2</sup> can be achieved using HT of 4 minutes, wt of 4 %. The contour plots shown in Fig. 12, 13 revealed that the tensile strength ( $T_s$ ) of 21–21.25 MPa and hardness (hard) of 80.25–80.5 can be achieved using HT of 6 minutes, wt of 1 % and HT of 2 minutes, wt of 4 %, respectively. Also, the contour plots in Fig. 14 revealed that the thermal conductivity (HT) of 0.5–0.505 W/m·C° can be achieved using HT of 4 minutes, wt of 2 %. The use of plastic waste as a reinforcing material for epoxy composites improved their mechanical properties, as the results of the current research agree with what the researchers reached in [4], where the mechanical properties of epoxy composites improved after adding plastic waste prepared from tires at various wt%.

The presented study considers only one type of polymer composites (epoxy). Furthermore, it is limited to the investigation of the mechanical properties and thermal conductivity of epoxy composites. Further research could explore the thermal physical properties of a wide range of practically important polymer composite materials. In addition, a range of physical and mechanical properties should cover composites, as well as study influencing factors at larger ranges to include values greater than suggested in the current research regarding the HT and wt % factors.

## 7. Conclusions

1. The present research studied the effect of HT and wt % on the behavior of 75 micro filler (RCCF) prepared from recycling the clamshell waste as a filler to enhance

epoxy resin on the mechanical properties and thermal conductivity of epoxy composites. The optimal parameter formulation is selected based on the higher GRG value, which means a stronger correlation to the reference sequence and better performance. Thus, the optimal tuning for multi-responses is HT2wt3, i.e. 4 min of HT and 4 % of wt %. The higher value of the mean grey relational grade (GRG) represents the maximum value of the mechanical properties of epoxy composites and the minimum value of thermal conductivity. The variance of the maximum and minimum results of mean grey relational grade (GRG) for parameters was 0.078 for the heating time factor (HT) and 0.1353 for the weight percentage factor (wt %). This result indicates that the wt factor has a major effect on multi-response compared to the HT factor.

2. Through the overall performance response to the results of the relational degree (GRG), it becomes at the optimal value of 0.609, which is close to the expected value of 0.670, from which we conclude that the GRG for each of the responses such as mechanical properties and thermal conductivity is greatly improved by 0.041 by adjusting the optimum combination. Then the optimal value of factors is HT2wt2, which ensures optimal mechanical properties and low thermal conductivity. Hence the grey relational analysis based on the Taguchi method for the optimization of the multi-response problem is a very useful tool for predicting the mechanical properties and thermal conductivity of recycled clamshell container waste (RCCF) reinforced epoxy composites. Moreover, the adoption of the use of recycled clamshell waste products as a filler in epoxy composites achieved positive results by improving the properties.

3. Regression equations were obtained for the mechanical properties and thermal conductivity of epoxy composites under the influence of HT2wt2 factors. From the contour, which is the optimal performance, the maximum values for the properties of epoxy composites were determined, where the optimization values for the bending strength, impact strength, tensile strength, stiffness and thermal conductivity temperatures are 68.2 MPa, 10.348 kJ/m<sup>2</sup>, 21.08 MPa, 80 Shore D and 0.504 W/m·C°, respectively. The results of the current research proved the improvement of the mechanical properties and thermal conductivity of epoxy compounds reinforced with RCCF filler clearly compared with the original properties values of epoxy without addition. The present research demonstrates how to use Taguchi parameters design for optimizing mechanical properties and thermal conductivity with minimum cost and time to industrial readers. The current research suggests an analogous study to the current one, with the addition of recycled filler from waste plastic prepared by traditional methods to compare the results and their effect on improving the thermal and mechanical properties of epoxy composites.

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