UDC 539.4

Modeling and Analysis of the Tensile and Flexural Properties of a Fiber-Orientated Hybrid Nanocomposite Using Taguchi Methodology

Y. Rostamiyan[a,b]

[a] Department of Mechanical Engineering, Sari Branch, Islamic Azad University, Sari, Iran

[b] Department of Mechanical Engineering, Semnan University, Semnan, Iran

yasser.rostamiyan@iausari.ac.ir

УДК 539.4

Моделирование и расчет характеристик прочности армированного волокнами гибридного нанокомпозита при растяжении и изгибе по методике Тагучи

Я. Ростамиян[а,б]

[а] Исламский университет Азад, Сари, Иран

[б] Университет г. Семнан, Иран

Оценено влияние трех независимых параметров (ориентация волокон, весовая доля наночастиц глинозема и кремнезема) на прочностные характеристики гибридного нанокомпозита из эпоксидной смолы, армированной углепластиковыми волокнами, с нанодобавками глинозема и кремнезема при растяжении и изгибе. Для планирования экспериментов использовали ортогональный набор согласно методике Тагучи. Для оценки функции отклика было изготовлено и испытано 16 образцов при запланированных комбинациях вышеуказанных параметров. Выявлен обратный эффект влияния входных параметров на соответствующие отклики, причем полученные двухмерные графики показывают, что варьирование такими двумя параметрами, как ориентация волокон – доля наночастиц глинозема и ориентация волокон – доля наночастиц кремнезема, существенно влияет на прочностные характеристики при растяжении и изгибе, в то время как варьирование параметрами доля наночастиц кремнезема – доля наночастиц глинозема не оказывает значительного влияния на вышеуказанные характеристики. Полученные диаграммы напряжение–деформация показывают, что гибридные нанокомпозиты с различной ориентацией волокон имеют более высокие прочностные характеристики при растяжении и изгибе, большие удлинения при разрушении и меньшие модули упругости, чем композиты из эпоксидной смолы как без нанодобавок, так и с нанодобавками глинозема или кремнезема.

Ключевые слова: углепластиковые волокна, ламинат, гибрид, механические свойства, методика Тагучи.

Introduction. Composite materials have been used in a wide range of industry fields like: aerospace, sports, automobile and etc. In recent decades because of their environmentally friendly nature, economical efficiency properties and superior mechanical and thermal properties compare with other kind of materials [1]. For example composites have demonstrated weight savings for aircraft structures and outstanding corrosion and fatigue-damage resistance [2]. In recent years, approximately all of the research done in the field of composite materials has focused on improving their mechanical and thermal properties by adding various additives. Different types of materials can be added to composite materials in order to achieve considered properties such as fibers, macro-, micro-, and nanomaterials. The most applicable thermoset matrices used for reinforcing composite materials are epoxy resins due to their low price, superior mechanical properties and chemical stability [3–8]. Mechanical properties of epoxy resins can be improved by adding rigid inorganic nanoparticles without affecting the glass transition temperature of the epoxy [9]. In many studies, the epoxy resin performance was improved by incorporating thermoplastic fillers, rubber agents, diluents, and nanoparticles into the epoxy, in addition to inorganic nanoparticles being used as reinforcing materials due to their low cost, ease of fabrication, and environmentally friendly nature [10]. Various nanofillers, such as silica (SiO_2) [9], clay, carbon nanotube (CNT), alumina (Al_2O_3) [11], and titania (TiO_2) [12] can be employed for this purpose. Mirmohseni and Zavareh [13] reported that adding 2.5 wt.% organically modified clay into the epoxy resin increased the tensile modulusand impact strength compared to those of the neat epoxy. Xu and Hoa [14] showed 38% improvement in flexural strength achieved by adding a low weight percentage of nanoclay to the fiber/epoxy composites. Akbari et al. [15] used liquid carboxyl-terminated butadiene acrylonitrile (CTBN) for toughening epoxy resin and 26% improvement in tensile strength was reported when 5 phr (per hundred resins) CTBN was added. Becker et al. [16] added Nanomer I.30E nanoclay into epoxy resin, which resulted in the increased fracture toughness and elastic modulus. Liu et al. [17] founded that adding nanoclay to a nanocomposite increased the fracture toughness and elastic modulus of the epoxy system without decreasing its compressive strength. Yasmin et al. [18] filled DGEBA resin with nanoclay and produced a specimen with a lower tensile strength and a higher elastic modulus compare to the neat epoxy. Ragosta et al. [19] found that adding 10 wt.% silica in to the epoxy matrix improved mechanical properties. Zheng et al. [9] added 3 wt.% of nanosilica to the epoxy matrix and described that the tensile strength increased by about 115%, and the impact strength increased by about 56%. Rosso et al. [20] filled epoxy resin with 11 wt.% of silica nanoparticles and an improvement in the tensile modulus and fracture toughness was reported.

As mentioned above, various additives can be added to composite materials. Fibers are such additives, that provide stress redistribution throughout the restoration and improve the structural properties of the material by acting as crack stoppers [21]. Glass fiber is the most commonly used fiber compared to other kinds of fibers and can improve the in-plane mechanical properties much better than the others. Panthapulakkal and Sain [22] evaluated mechanical and thermal properties of hemp/glass fiber-polypropylene composite and showed that adding glass fiber into hemp-polypropylene composite improved thermal properties. Eronat et al. [23] studied effects of glass fiber layering on the flexural strength of microfill and hybrid composites and reported that glass fiber layering of microfill and hybrid composites showed higher flexural strength, and veneering of hybrid composite with microfill composite increased the resistance of restoration. Using carbon fiber as reinforcement improves the mechanical properties of epoxy and other material matrices because of specific strength and modulus. Godara et al. [24] reinforced carbon nanotubes (CNTs) with carbon fibers and showed that the viscosity profile of the epoxy matrix reinforced with different types of CNTs indicated a strong dependency on the type of CNTs. There was also a substantial increase of over 80% in fracture toughness Mode-I for the pristine multi-walled CNTs in combination with the epoxy resin which was modified by using a compatibilizer. Bekyarova et al. [25] used carbon fiber/epoxy reinforced with carbon nanotubes and reported a great laminar strength (~ 50 MPa).

Using two or more kinds of micro- or nanoparticles as reinforcement creates hybrid nanocomposites. These kinds of composites provide higher mechanical properties and crack propagation resistance compared to those with one kind of reinforcement [26]. A large number of researches have been carried out on hybrid nanocomposites. Rostamyian et al. [27] used multi-walled carbon nanotube in present of high impact polystyrene and showed that mechanical properties such as tensile, compression and impact were improved.

Rostamiyan et al. [6] reported that adding nanoclay as a nanoreinforcement and HIPS as a thermoplastic phase into the epoxy resin creates synergistic effects on mechanical properties. Their results showed that new ternary nanocomposite possessing tensile, compressive, and impact strengths were improved up to 60, 64, and 402%, respectively. Geisler and Kelley [28] filled epoxy resin with alumina (Al_2O_3) and rubber particles and reported that obtained toughness values were 25% higher than those of epoxy systems having only alumina (Al_2O_3) or rubber particles. Mirmohseni and Zavareh [29] showed that by adding 2% clay and 20% polyamide, the material toughness increased by 115% and also impact strength improved as compared to neat epoxy. Kinloch et al. [30] added nanosilica and rubber microparticles to epoxy resin. They described a significant increase in toughness. Rostamyian et al. [31] used nanosilica and high impact poly styrene as reinforcement in epoxy-based hybrid nanocomposite. They reported that new ternary nanocomposite improved ultimate tensile, compression and impact strength up to 59.5, 45, and 414%, respectively, compared to those of the neat epoxy resin. Mirmohseni and Zavareh [29] showed that adding ABS, nanoclayand TiO_2 into the epoxy improved impact strength compared to neat epoxy. Rostamiyan et al. [6] also filled epoxy resin with nanoclay as a nanoreinforcement and HIPS as a thermoplastic phase and reported that impact, tensile and compression and strengths were improved by up to 402, 60, and 64%, respectively, as compared to those of the pure epoxy.

As follows from the above considerations, the mechanical properties of hybrid nanocomposites can be affected by many factors, one of which is the weight percentage of reinforcement, such as toughening agent and nanofiller [29]. The other important factor is weight percentage of a hardener. Although determination of the appropriate amount of this factor is based on the stochiometric ratio, this expectation is not far, especially in the presence of thermoplastic phase as toughening agent and also nanofiller in the epoxy resin. The probability of complete mixture of epoxy monomers and hardener would dramatically deteriorate and hence prevent a complete polymerization. The OVAT (one variation at time) is the conventional method for analyzing effective parameters of an experiment. This method can only analyze one variable at a time, while in most experimental designs, variables depend on each other and also the effect of interactions between them is important, so this method is not instrumental for deriving the truly optimum point. Leardi [32] claimed that 93% of the published papers in 2009 with general titles containing "optimization," "development," "improvement" or "effect of," employed the OVAT model. Predicting the nonlinear effect of each parameter is the important feature, which requires at least three points as parameter levels, which directly increases the number of the required experiments for model prediction and consequently increases the time and cost. There are a numerous mathematical methods of design of experiments (DOE). These methods provide the ability to evaluate the joint effect of two parameters and also the nonlinear effect of selected parameters. Also these methods can optimize the results and provide the optimum levels of each factor, in order to achieve the best result based on the desired goal. The response surface design is one of the most widespread mathematical and statistical methods for analyzing multiple factors at a time, evaluating nonlinear effect of parameters, studying the effect of interaction between factors and finally optimizing the response [33]. The Taguchi design is a section of DOE methods, which uses an orthogonal array, signal-to-noise (S/N) ratio and analyses of variance (ANOVA) for analyzing the results and determining the significant parameters and how they affect the corresponding response. This method reduces the number of generated experiments and thus reduces their conduction time and cost. For measuring the quality characterization deviating from the desired values, the Taguchi design transforms into the S/N ratio. A higher S/N ratio indicates a better experimental response of characteristics (the optimal level of the process parameter). Analyses of variance should be done to evaluate the effect of each input parameter on the response. Moreover, the ANOVA table can reveal, which particular parameter has a significant effect on the result [34]. The Taguchi method is widely used in the field of engineering. In the current study, the Taguchi method was selected for analyzing flexural and tensile strength of epoxy/carbon fiber/nanoclay/nanosilica quadratic hybrid nanocomposite. The weight percentage of nanoclay and nanosilica and also the orientation of carbon fiber were selected as independent input variables and the effect of these parameters on flexural and tensile strength properties was investigated.

1. **Experimental**.

1.1. ***Materials***. The epoxy resin utilized in this study was an undiluted clear difunctional bisphenol A, Epon 828 and provided by Shell Chemicals Co. Its epoxide equivalent weight was 185–192 g/eq. Epon 828 is basically DGEBA (Diglycidyl ether of bisphenol-A). The curing agent was a nominally cycloaliphatic polyamine, Aradur® 42 supplied by Huntsman Co. The organoclay Cloisite 30B was purchased from Southern Clay Products (Gonzales, TX, USA). The spherical silica nanoparticles with average particle size 10–15 nm and SSA (specific surface area) 180–270 m^2/g were supplied by TECNAN Ltd. The solvent used was Tetrahydrofuran (THF) with purity (GC) of more than 99% provided from Merck Co (Germany).

1.2. ***Specimen Preparation***. The laminate plates were prepared with 16 layers and different fiber orientations based on the generated experiments using the Taguchi design. For preparing each specimen carbon fiber was hand laid-up with the specific steps. In order to create homogenous mixture, the whole procedure of reinforcing the resin was carried out in a suitable solvent. Tetrahydrofuran was selected as solvent for all the mixture components, including nanosilica, epoxy resin, and nanoclay. Liquid epoxy resin was poured into an adequate amount of THF solvent, so the comparable neat epoxy specimens could be mixed using of magnetic stirrer at least 2 h with 2000 rpm. Then the solvent was completely evaporated using a vacuum Erlenmeyer. In the next stage, the mixture was homogenized by ultrasonicating (ultrasonic SONOPLUS-HD3200, 50% amplitude, 20 kHz and pulsation; on for 10 s and off for 3 s) for 8 min. At this step, 23 phr of cycloaliphatic polyamine was added as hardener according to the stoichiometric ratio. Then the mixture was degassed using the vacuum pump to remove the air bubbles. All specimens were prepared using the handy lay-up method and cured at room temperature for 24 h with the following post-curing from 20 to 130°C every 2 h with a 20°C temperature enhancement interval. Figure 1 depicts a symmetric laminate composite ply structure, while Fig. 2 displays a laminated composite specimen prepared for the current study.

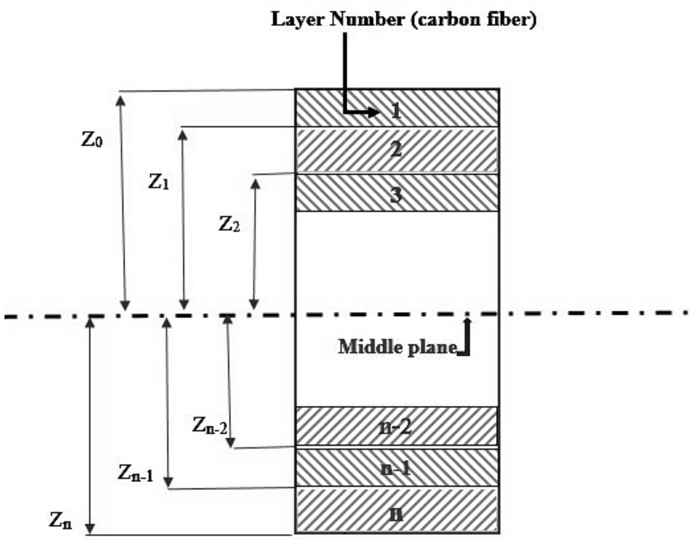


Fig. 1. Laminate stacking of plies [(±θ) 2] sym.

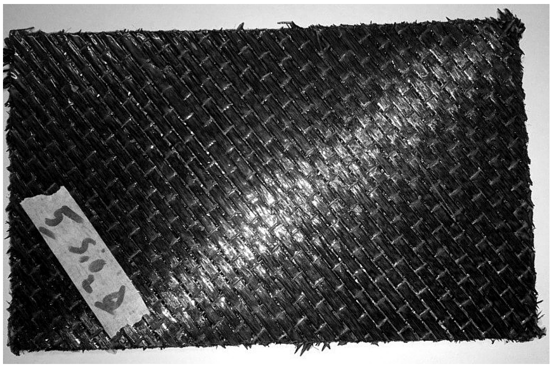


Fig. 2. Laminated nanocomposite prepared for tests.

1.3. ***Characterization***. Tensile tests in the current study were conducted according to the ASTM D: 3039. Specimens with 0 and 90° unidirectional fibers were prepared with dimensions of $15\times 250\times 1$ and $25\times 175\times 2$ mm, respectively. Other specimens were prepared with dimensions of $25\times 250\times 2.5$ mm, according to the above standard. These mechanical tests were conducted using an STM-150 universal testing machine from Santam Company (Tehran, Iran) with a load capacity of 150 kN. In addition, flexural tests were conducted based on the 3-point bending loading scheme according to the ASTM: D790. This test method covers the determination of the flexural properties of reinforced plastics, including high-modulus composites. The dimensions of the specimens were $127\times 12.7\times 3.2$ mm. Figure 3 shows the delamination process of the specimen under flexural test, while Fig. 4 shows a delaminated specimen under tensile test.

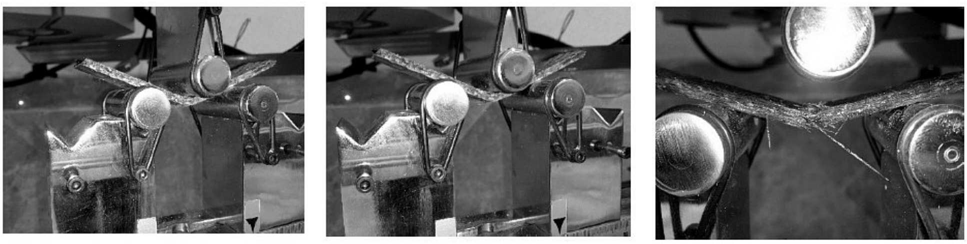


Fig. 3. Delamination process of specimen under flexural test.

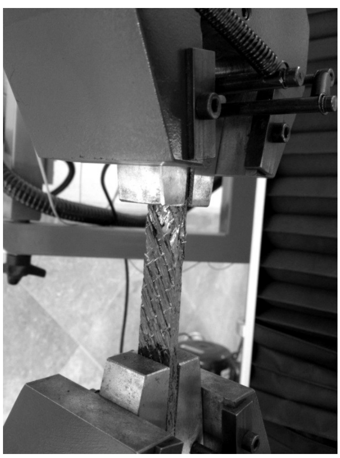


Fig. 4. Delaminated specimen under tensile test.

2. **Design of Experiment**. The Taguchi design is a lucrative method for the design of experiments and is known as "orthogonal array design." It provides a simple and effective way of analyzing and allows one to produce high-quality products with a low manufacturing cost and in less time. This method requires only a fraction of the full factorial combinations. An orthogonal array means that the design is balanced so that factor levels are weighted equally. Because of this, each factor can be evaluated independently of all the other factors, so the effect of one factor does not influence the estimation of another. This method decreases the number of experiments, as compared to other DOE methods such as full factorial design or response surface design. In this study, carbon fiber orientation, nanoclay wt.% and nanosilica wt.% were selected as input parameters and the effectiveness of each factor on tensile and flexural properties of hybrid quaternary nanocomposite was investigated. By using this method the number of experiments carried out was reduced to 16 sets instead of 64 ($4 \times 4 \times 4$ for full factorial design). The steps of the Taguchi experimental design are as follows:

(i) determining the number of levels for each parameter;

(ii) selecting the appropriate orthogonal array;

(iii) arrangement of operation parameters to the orthogonal array;

(iv) conducting experiments based on the arrangement of the orthogonal array;

(v) analysis of results usingthe signal-to-noise ratio (S/N) and analysis of variance (ANOVA).

The Taguchi method selects the optimum condition so the effect of uncontrollable factors (noise) on response attains the minimum. The Taguchi method utilizes ANOVA to determine the influence of any parameter on response and to interpret the percentage of contribution of each experimental variable. One aim of this study was to specify the most effective factors, in order to achieve the maximum enhancement of tensile and flexural properties of the mentioned quaternary nanocomposite. Three factors, such as carbon fiber orientation, nanoclay wt.%, and nanosilica wt.% with four different levels were selected for the design of experiments. Table 1 shows the independent factors and the selected levels for each of these factors. The L_{16} orthogonal array (shown in Table 2) was selected for this study according to the number of factors and their levels. Responses of the designed experiments were set to the maximum flexural and tensile strength values. The Minitab software v.16.244 was used for analyzing the results.

T a b l e 1

Factors and Levels Selected for Taguchi Design

Level	A. Fiber orientation (deg)	B. Nanoclay (wt.%)	C. Nanosilica (wt.%)
1	0	0.5	0.5
2	30	1.5	1.0
3	60	2.5	1.5
4	90	3.5	2.0

3. **Results and Discussion**. As it was mentioned above, the independent input parameters selected for the current study were carbon fiber orientation, nanoclay wt.% and nanosilica wt.% which are designated in the analysis of variance as parameters A, B, and C, respectively, whereas the selected responses were tensile and flexural properties of hybrid quaternary nanocomposite. Table 3 displays the details of 16 generated experiments using the Taguchi design and the levels of each factor in different run numbers, as well as experimental results obtained from tensile and flexural tests. According to this table, the maximum tensile and flexural strength values were obtained for the design level 2 with magnitudes of 470.3 and 26.6 MPa, respectively. The related value of input parameters for

T a b l e 2

L_{16} Orthogonal Array Used for Experimental Design

Experimental No.	Factor levels		
	A[a]	B[b]	C[c]
1	1	1	1
2	1	2	2
3	1	3	3
4	1	4	4
5	2	1	2
6	2	2	1
7	2	3	4
8	2	4	3
9	3	1	3
10	3	2	4
11	3	3	1
12	3	4	2
13	4	1	4
14	4	2	3
15	4	3	2
16	4	4	1

T a b l e 3

Experimental Design and Corresponding Responses

Experimental No.	Experimental factors			Responses	
	Fiber orientation (deg)	Clay content (wt.%)	Silica content (wt.%)	Tensile strength (MPa)	Flexural strength (MPa)
1	0	0.5	0.5	420.1	21.3
2	0	1.5	1.0	470.3	26.6
3	0	2.5	1.5	465.4	22.8
4	0	3.5	2.0	359.2	18.1
5	30	0.5	1.0	250.6	12.5
6	30	1.5	0.5	245.3	12.2
7	30	2.5	2.0	215.1	10.3
8	30	3.5	1.5	227.4	11.1
9	60	0.5	1.5	195.6	5.2
10	60	1.5	2.0	171.2	6.8
11	60	2.5	0.5	159.1	7.5
12	60	3.5	1.0	180.3	7.3
13	90	0.5	2.0	74.7	2.8
14	90	1.5	1.5	88.2	3.9
15	90	2.5	1.0	95.3	4.1
16	90	3.5	0.5	69.5	3.3

this level were 0° of fiber orientation, 1.5 wt.% of nanoclay and 1 wt.% of nanosilica. The minimum value of tensile strength occurred in design level 16 with a magnitude of 69.5 MPa and input parameters of 90° of fiber orientation, 3.5 wt.% of nanoclay and 0.5 wt.% of nanosilica. Also the minimum value of flexural strength was 2.8 MPa which occurred in design level 13 with 90° of fiber orientation, 0.5 wt.% of nanoclay and 2 wt.% of nanosilica respectively.

3.1. ***Signal to Noise (S/N) Ratio***. The Taguchi method recommends the use of S/N ratio which is measured by the deviation of characteristics from their target value. In this case the aim is to improve and increase the obtained value of tensile and flexural strength and, so it seems that a larger response is better and the S/N ratio will be defined as follows:

$$\mathrm{S/N} = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i^2}\right),$$

where n is the trial repetition and y_i is the result of the ith experiment for each trial.

Results of the mean S/N ratios for the three input factors at their designed levels are shown in Table 4. Figure 5 shows the S/N graph for different levels of the three selected factors.

T a b l e 4

Magnitudes of S/N Ratio

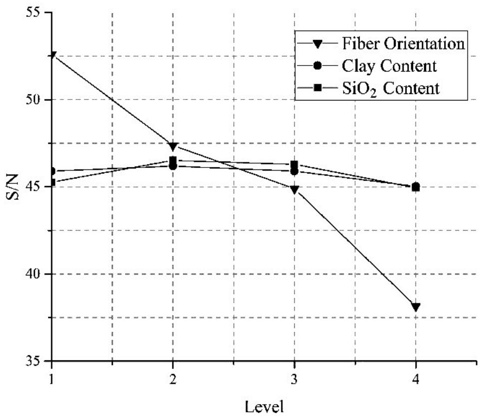
Experimental No.	Experimental factors			S/N ratio	
	Fiber orientation (deg)	Clay content (wt.%)	Silica content (wt.%)	Tensile strength	Flexural strength
1	0	0.5	0.5	52.46	26.56
2	0	1.5	1.0	53.44	28.49
3	0	2.5	1.5	53.34	26.92
4	0	3.5	2.0	51.19	25.10
5	30	0.5	1.0	47.95	21.93
6	30	1.5	0.5	47.78	21.58
7	30	2.5	2.0	46.64	20.00
8	30	3.5	1.5	47.12	20.82
9	60	0.5	1.5	45.80	13.97
10	60	1.5	2.0	44.65	16.65
11	60	2.5	0.5	44.02	17.50
12	60	3.5	1.0	45.10	17.26
13	90	0.5	2.0	37.38	8.29
14	90	1.5	1.5	38.88	11.82
15	90	2.5	1.0	39.55	12.25
16	90	3.5	0.5	36.77	10.37

3.2. ***Analysis of Variance for S/N Ratio***. The Taguchi method uses analysis of variance for estimating the effect of input parameters on corresponding response. For this purpose Taguchi method evaluates the significance of each parameter according to its probability value (*P*-value). In the current study, the ANOVA analysis was carried out based on confidence level $\alpha = 0.05$. So the terms with probability $P \geq 95\%$ ($\alpha \leq 0.05$) are significant and those with *P*-value less than 95% ($\alpha \geq 0.05$) are not considered effective on the selected response. Table 5 displays the ANOVA results for tensile and flexural strength properties. As seen the fiber orientation was the most effective parameter on both responses with a probability value of $P > 99\%$. Nanoclay wt.% was significant on tensile and flexural

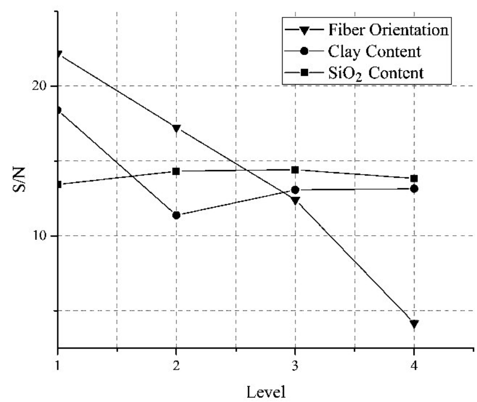
Table 5

Analysis of Variance for S/N Ratio for Tensile and Flexural Strengths

Source	DF	Seq SS	Adj SS	Adj MS	*F*	*P*
Tensile strength						
A	3	431.566	431.566	143.855	712.62	0
B	3	3.060	3.060	1.020	5.05	0.044
C	3	7.023	7.023	2.341	11.60	0.007
Residual error	6	1.211	1.211	0.202	–	–
Total	15	442.860	–	–	–	–
Flexural strength						
A	3	562.532	562.532	187.511	310.32	0
B	3	8.807	8.807	2.936	4.86	0.048
C	3	13.031	13.031	4.344	7.19	0.021
Residual error	6	3.626	3.626	0.604	–	–
Total	15	587.966	–	–	–	–



a



b

Fig. 5. Plot of S/N ratio as a function input parameters for tensile (a) and flexural (b).

strength with *P*-value 96%. Moreover nanosilica wt.% affected both tensile and flexural strength with *P*-values of 96 and 98% respectively. So it can generally be concluded that variation in weight percentage of nanosilica had a greater effect on the flexural strength than tensile.

3.3. ***Main Effect Plot for S/N Ratio***. Figures 6 and 7 display the main effect plot of S/N ratio on tensile and flexural strength properties respectively. These types of plots determine how a parameter affects the corresponding response. According to Fig. 6, the tensile strength decreased while continuously increasing the degree of fiber orientation and had a small increase with increasing the weight percentage of nanoclay and nanosilica and then decreased slightly. From Fig. 7 it is obvious that nanosilica wt.% and nanoclay wt.% had a similar effect on flexural strength and the nanosilica wt.% decreased the magnitude of flexural strength more than tensile, also increasing in the degree of fiber orientation decreased the flexural strength. Comparing the effect of nanoclay wt.% in both graphs, it shows that the magnitude of tensile strength at level 4 was higher than level 1, while the flexural strength at level 4 was lower than that at level 1.

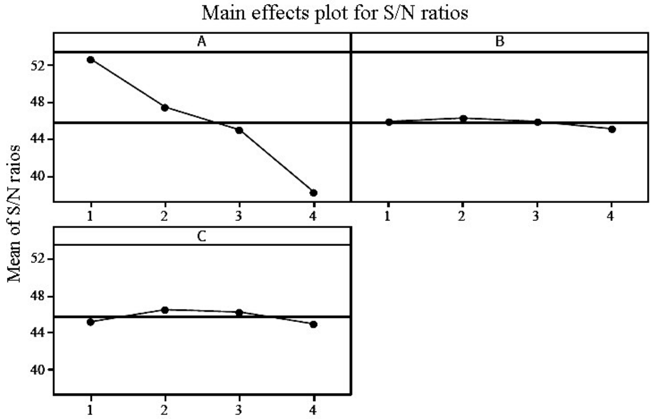


Fig. 6. Main effect plot of S/N ratio for tensile strength.

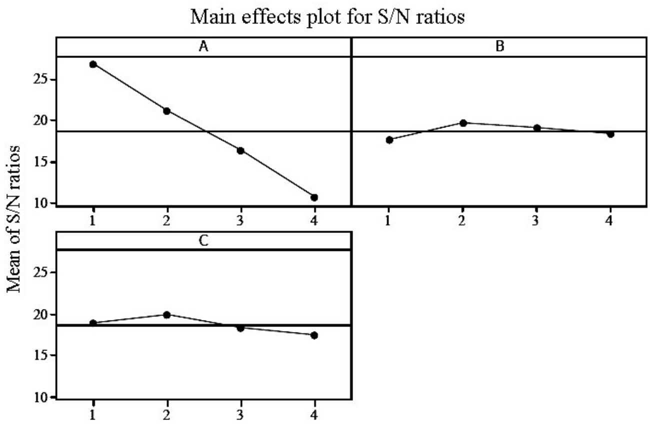
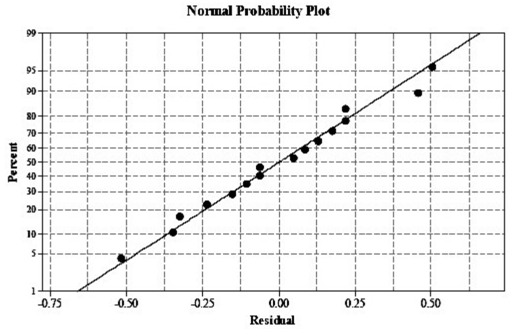


Fig. 7. Main effect plot of S/N ratio for flexural strength.

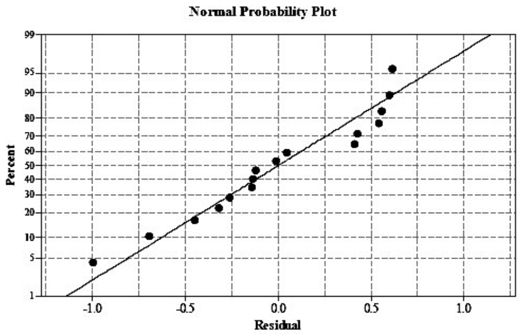
3.4. ***Normal Probability Plot for S/N Ratio***. Figure 8 depictss the normal probability plot of S/N ratio obtained from the analysis of variance for tensile and flexural properties respectively. These types of plots help us to determine whether a particular distribution fits collected data and allows the comparison of different specimen distributions. Falling of the plotted points close to the fitted distribution line and close together means that the selected distribution has a good and acceptable fitness. From two parts of this figure it can be seen that the plotted points for both responses have fallen close together and also close to the fitted distribution line, so the selected distribution fitness is good for both tensile and flexural strength but it is more fitted for tensile as opposed to flexural as seen in Fig. 8.

3.5. ***Plot of Residuals versus Fitted Values for S/N Ratio***. Plot of residuals versus fitted values for tensile and flexural strength properties is shown in Fig. 9. From this figure it can be generally obtained that the residuals for both responses had scattered on the display randomly, therefore the model proposed was adequate and there were no reasons to suspect any violation of the independence or constant variance assumption. As seen the dispersion of residuals for the flexural strength was better than tensile.

3.6. ***2D Contour Plots for Tensile and Flexural Strength Properties***. At this stage, 2D contour plots have been used in order to evaluate the effect of interaction between parameters on flexural and tensile strength properties of desired hybrid quaternary nanocomposite. These plots are also useful for comparing the effect of main factors with the results obtained from analysis of variance and main effect plots. The graph shows how

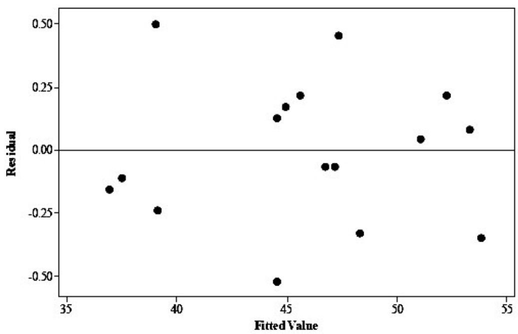


a

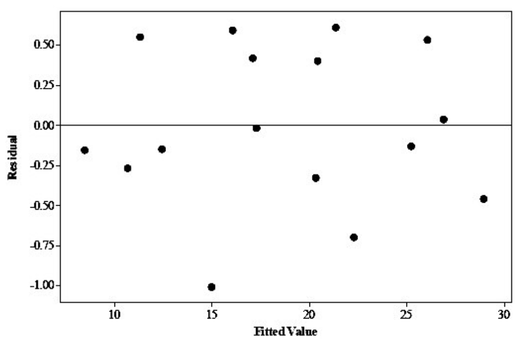


b

Fig. 8. Normal probability plot of S/N ratio for tensile (a) and flexural (b) strengths.



a

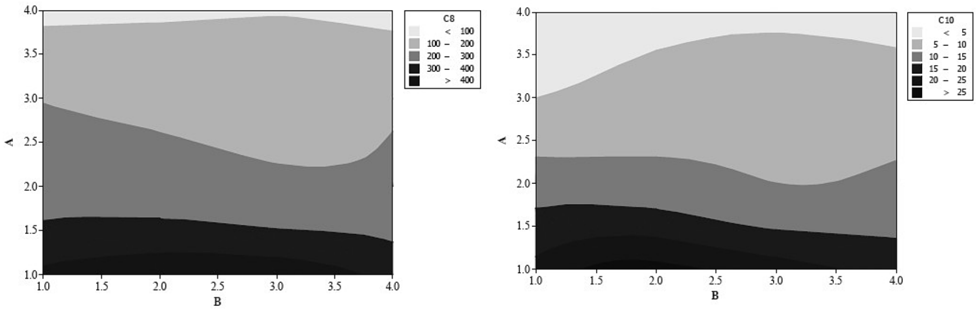


b

Fig. 9. Plot of residuals versus fitted values for S/N ratio for tensile (a) and flexural (b) strengths.

the selected parameter changes the response: the color varies with variation of the response magnitudes, while the range of this variation can be seen in the graph legend.

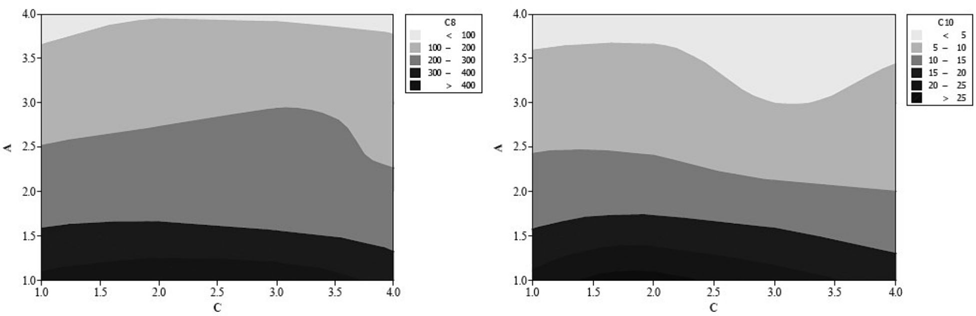
3.6.1. *Effect of Fiber Orientation and Nanoclay*. Figure 10 depicts the 2D contour plots for evaluating the effect of fiber orientation and nanosilica on corresponding responses which are indicated as A and B, respectively. As seen, an increase in the degree of fiber orientation results in the reduction of both tensile and flexural strength values, while an increase in wt.% of nanoclay provides a slight increase in the flexural strength followed by its reduction, whereas the tensile strength exhibits a continuous reduction. More, a significant interaction between fiber orientation and nanoclay is observed, while a simultaneous increase of both variables reversely affected two interested responses, according to Fig. 10.



a b

Fig. 10. 2D contour plots for fiber orientation and nanoclay for tensile (a) and flexural (b) strengths.

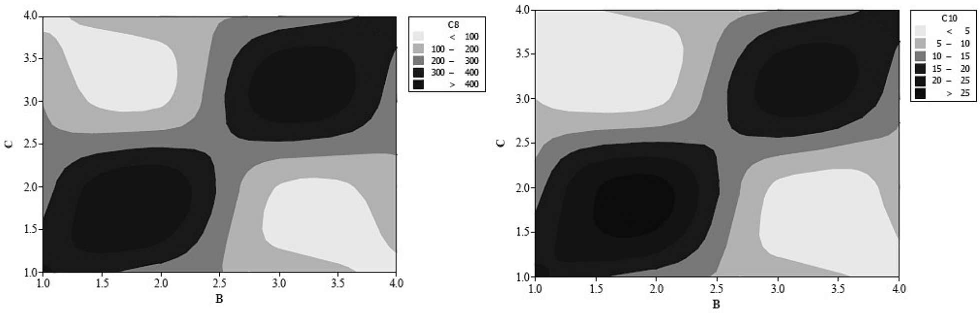
3.6.2. *Effect of Fiber Orientation and Nanosilica*. Figure 11 depicts the 2D contour plots drawn for the effect of fiber orientation and nanosilica. From these graphs it can be obtained that the fiber orientation had a continuous reverse effect on both responses, while an increase in the nanosilica wt.% resulted in an initial slight increase of the flexural and tensile values and their further reduction. Moreover, a simultaneous increase in the magnitudes of both parameters decreased the tensile and flexural strength properties, which is similar to the effect of fiber orientation and nanoclay. The interaction between fiber orientation and nanosilica was significant.



a b

Fig. 11. 2D contour plots for fiber orientation and nanosilica for tensile (a) and flexural (b) strengths.

3.6.3. *Effect of Nanoclay and Nanosilica*. Figure 12 displays the 2D contour plots for nanosilica and nanoclay for tensile and flexural strength, respectively. As is seen, the increased value of each factor decreased both the required responses, but simultaneous



a b

Fig. 12. 2D contour plots for nanosilica and CNT for tensile (a) and flexural (b) strengths.

increase in the magnitudes of both factors had no obvious effect on the tensile and flexural properties.

From the above discussion it can be concluded that increasing the levels of all input parameters has reverse effects on both of the studied responses and reduces the response of obtained values. Therefore, in order to achieve higher magnitudes of tensile and flexural strengths, lower degrees of fiber orientation and lower contents of nanosilica and nanoclay should be incorporated for preparing the required hybrid nanocomposite.

3.7. ***Stress–Strain Plots***. Stress–strain plots were generated and the mechanical properties of desired hybrid nanocomposite were measured with simple nanocomposites and pure epoxy. Hybrid nanocomposite specimens were prepared with different fiber orientations, weight percentage of nanoclay and weight percentage of nanosilica and compared with nanocomposites with single nanoparticle and neat epoxy.

Figure 13a displays the stress–strain dependence for design level 2 with 0° of fiber orientation. 1.5 wt.% of nanoclay and 1 wt.% of nanosilica. Two single nanoparticle nanocomposites were considered with 1.5 wt.% of nanoclay and 1 wt.% of nanosilica. It can be seen that, pure epoxy had the lowest value of tensile strength and the hybrid nanocomposite showed a higher value of tensile strength and elongation at break compare to the two other nanocomposites mentioned. Also it failed at a higher value of tensile strength compared to others. Moreover hybrid nanocomposite specimen had a lower elastic modulus compared to other specimens according to the gradient of stress–strain graphs.

Figure 13b depicts the stress–strain dependence for design level 6 with 30° of fiber orientation. 1.5 wt.% of nanoclay and 0.5 wt.% of nanosilica. Two single nanoparticle nanocomposites were prepared 1.5 wt.% of nanoclay and 0.5 wt.% of nanosilica. From this section it can be seen that, the hybrid nanocomposite had a higher value of tensile strength and elongation at rupture, as compared to nanoclay-epoxy, nanosilica-epoxy, and pure epoxy. Moreover, the hybrid nanocomposite specimen exhibited a lower elastic modulus, as compared to other specimens, according to the gradient of stress–strain graphs, and failed at a higher value of tensile strength.

Stress–strain graphs for design level 12 with 60° of fiber orientation. 3.5 wt.% of nanoclay and 1 wt.% of nanosilica are shown in Fig. 13c. As seen, the values of tensile strength for hybrid nanocomposite were higher than those of nanosilica-epoxy; nanoclay-epoxy and pure epoxy, while they had lower elastic moduli, as compared to three other composites, according to the gradient of graphs. They also failed at higher values of the tensile strength, similar to the earlier described cases. In general, the results discussed of this section demonstrate the reverse effect of studied factors on the tensile properties and are in agreement with observation made during the analysis of variance and results of 2D contour plots.

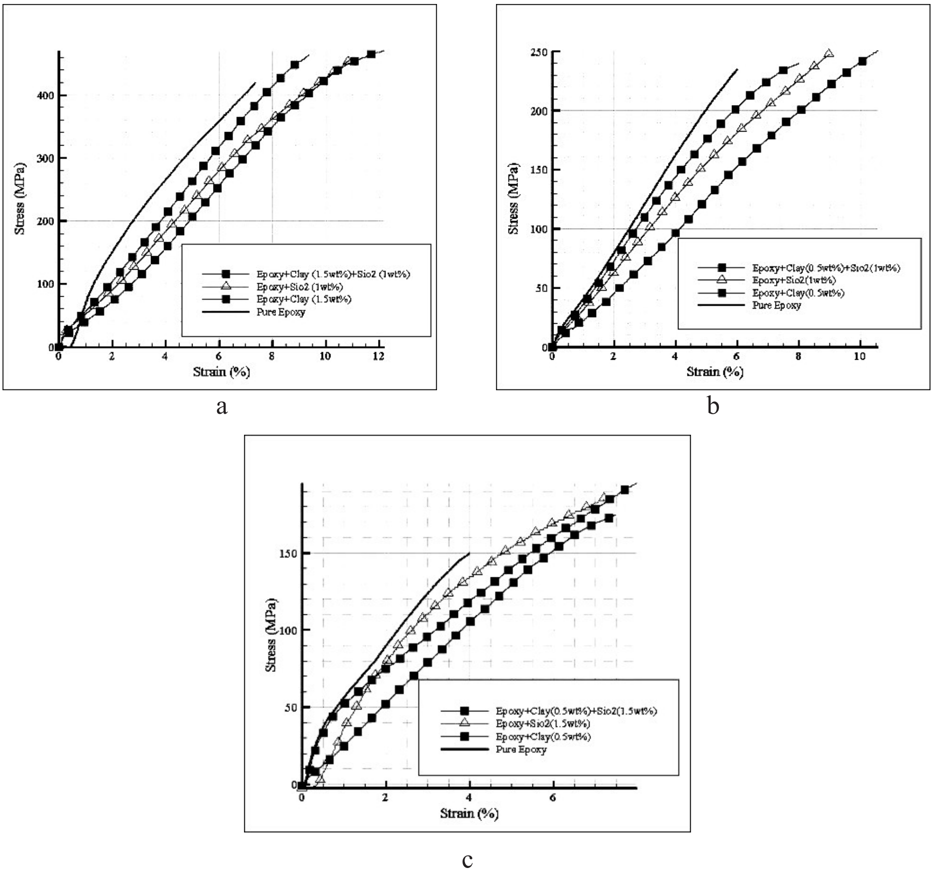


Fig. 13. Stress–strain plots for different fiber orientation: (a) 0°; (b) 30°; (c) 60°.

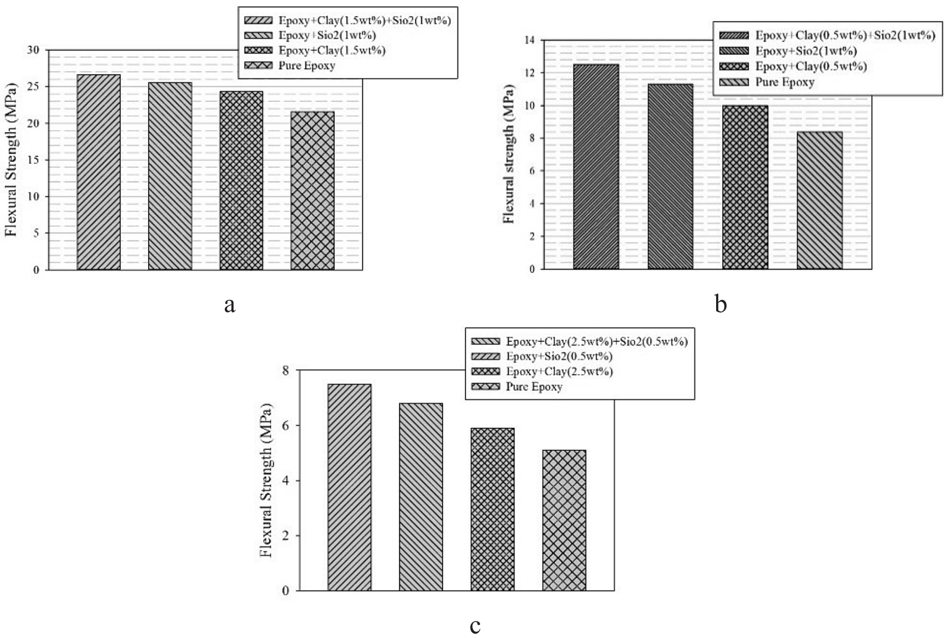


Fig. 14. Plot of flexural strength for different fiber orientation: (a) 0°; (b) 30°; (c) 60°.

Figure 14 display the plots of flexural strength for 0, 60, and 90° of fiber orientation and feature the design levels used for preparing hybrid and simple nanocomposites. The analysis of three plots in Fig. 14 implies that the hybrid nanocomposite exhibits the highest flexural strength, whereas the epoxy-nanosilica and epoxy-nanoclay nanocomposites are the next in the respective decreasing order. Thus, the epoxy-nanosilica nanocomposite was more effective than epoxy-nanoclay one, which fact is in agreement with earlier observations made during the analysis of variance in previous sections. Also, a pure epoxy had the lowest value of flexural strength. Moreover, reduction of magnitude of all mentioned nanocomposites can be observed with increase in the fiber orientation value.

Conclusions. In this study, the effect of three independent parameters on tensile and flexural properties of hybrid quaternary nanocomposite was evaluated. Input selected variables were carbon fiber orientation, weight percentage of nanoclay and weight percentage of nanosilica. The Taguchi orthogonal array design was selected for designing the experiments and 16 specimens were prepared and tested based on designed levels for each response. Main effect plot and also analysis of variance for S/N ratio indicated that the most effective parameter was carbon fiber orientation which decreased both responses continuously. Nanosilica and nanoclay contents decreased the tensile strength and flexural strength properties. Also the analysis of variance showed that nanoclay and nanosilica affected the tensile strength more than the flexural one due to related probability values. Moreover, from 2D contour plots it was obtained that two component interactions between fiber orientation and nanosilica and also between fiber orientation and nanoclay were effective for both responses, whereas interaction between nanoclay and nanosilica was not an effective term. In addition, stress–strain plots indicate that hybrid nanocomposites with different fiber orientations manifest higher values of tensile strength, as compared to those of epoxy-nanosilica, epoxy-nanoclay composites and pure epoxy, and generally have higher values of tensile strength and elongation at rupture, but lower elastic moduli. Finally, the flexural strength of a hybrid nanocomposite is higher than that obtained for epoxy-nanoclay and epoxy-nanosilica nanocomposites, whereas the pure epoxy has the lowest value of flexural strength.

Резюме

Оцінено вплив трьох незалежних параметрів (орієнтація волокон, вагова частка наночастинок глинозему і кремнезему) на міцнісні характеристики гібридного нанокомпозиту з епоксидної смоли, армованої вуглепластиковими волокнами, з нанодобавками глинозему і кремнезему при розтязі та згині. Для планування експериментів використовували ортогональний набір згідно з методикою Тагучі. Для оцінки функції відклику було виготовлено і випробувано 16 зразків при запланованих комбінаціях вищевказаних параметрів. Виявлено зворотний ефект впливу вхідних параметрів на відповідні відклики, причому отримані двовимірні графіки показують, що варіювання такими двома параметрами, як орієнтація волокон – частка наночастинок глинозему й орієнтація волокон – частка наночастинок кремнезему, істотно впливає на характеристики міцності при розтязі та згині, в той час як варіювання параметрами частка наночастинок кремнезему – частка наночастинок глинозему не має значного впливу на вищезгадані характеристики. Отримані діаграми напруження–деформація показують, що гібридні нанокомпозити з різною орієнтацією волокон мають більш високі характеристики при розтязі та згині, великі подовження при руйнуванні і менші модулі пружності, ніж композити з епоксидної смоли як без нанодобавок, так і з нанодобавками глинозему або кремнезему.

1. M. Xu, J. Hu, X. Zou, et al., "Mechanical and thermal enhancements of benzoxazine-based GF composite laminated by in situ reaction with carboxyl functionalized CNTs," *J. Appl. Polym. Sci.*, **129**, 2629–2637 (2013).
2. M. M. Schwartz, *Composite Materials Handbook*, McGraw-Hill, New York (1992).
3. P. C. LeBaron, Z. Wang, T. J. Pinnavaia, "Polymer-layered silicate nanocomposites: an overview," *Appl. Clay Sci.*, **15**, 11–29 (1999).
4. A. Fereidoon, A. H. Mashhadzadeh, and Y. Rostamiyan, "Experimental, modeling and optimization study on the mechanical properties of epoxy/high-impact polystyrene/multi-walled carbon nanotube ternary nanocomposite using artificial neural network and genetic algorithm," *Sci. Eng. Compos. Mater.*, **20**, No. 3, 265–276 (2013).
5. Y. Rostamiyan, A. Fereidoon, A. Ghasemi Ghalebahman, et al., "Experimental study and optimization of damping properties of epoxy-based nanocomposite: Effect of using nanosilica and high-impact polystyrene by mixture design approach," *Mater. Design*, **65**, 1236–1244 (2015).
6. Y. Rostamiyan, A. B. Fereidoon, A. H. Mashhadzadeh, and M. A. Khalili, "Augmenting epoxy toughness by combination of both thermoplastic and nanolayered materials and using artificial intelligence techniques for modeling and optimization," *J. Polymer Res.*, **20**, 1–11 (2013).
7. Y. Rostamiyan, A. Fereidoon, A. H. Mashhadzadeh, et al., "Using response surface methodology for modeling and optimizing tensile and impact strength properties of fiber orientated quaternary hybrid nano composite," *Composites Part B: Engineering*, **69**, 304–316 (2015).
8. Y. Rostamiyan, A. B. Fereidoon, "Preparation, modeling, and optimization of mechanical properties of epoxy/HIPS/silica hybrid nanocomposite using combination of central composite design and genetic algorithm. Part 1. Study of damping and tensile strengths," *Strength Mater.*, **45**, No. 5, 619–634 (2013).
9. Ya. Zheng, Yi. Zheng, and R. Ning, "Effects of nanoparticles SiO_2 on the performance of nanocomposites," *Mater. Lett.*, **57**, 2940–2944 (2003).
10. J. Ma, M.-S. Mo, X.-S. Du, et al., "Effect of inorganic nanoparticles on mechanical property, fracture toughness and toughening mechanism of two epoxy systems," *Polymer*, **49**, 3510–3523 (2008).
11. S. H. Lim, K. Y. Zeng, and C. B. He, "Morphology, tensile and fracture characteristics of epoxy-alumina nanocomposites," *Mater. Sci. Eng. A*, **527**, 5670–5676 (2010).
12. S. Sinha Ray and M. Okamoto, "Polymer/layered silicate nanocomposites: a review from preparation to processing," *Prog. Polym. Sci.*, **28**, 1539–1641 (2003).
13. A. Mirmohseni and S. Zavareh, "Epoxy/acrylonitrile-butadiene-styrene copolymer/clay ternary nanocomposite as impact toughened epoxy," *J. Polym. Res.*, **17**, 191–201 (2010).
14. Y. Xu and S. V. Hoa, "Mechanical properties of carbon fiber reinforced epoxy/clay nanocomposites," *Compos. Sci. Technol.*, **68**, 854–861 (2008).
15. R. Akbari, M. Beheshty, and M. Shervin, "Toughening of dicyandiamide-cured DGEBA-based epoxy resins by CTBN liquid rubber," *Iran Polym. J.*, **22**, 313–324 (2013).
16. O. Becker, R. J. Varley, and G. P. Simon, "Thermal stability and water uptake of high performance epoxy layered silicate nanocomposites," *Eur. Polym. J.*, **40**, 187–195 (2004).
17. W. Liu, S. V. Hoa, and M. Pugh, "Organoclay-modified high performance epoxy nanocomposites," *Compos. Sci. Technol.*, **65**, 307–316 (2005).

18. A. Yasmin, J.-J. Luo, and I. M. Daniel, "Processing of expanded graphite reinforced polymer nanocomposites," *Compos. Sci. Technol*., **66**, 1182–1189 (2006).

19. G. Ragosta, M. Abbate, P. Musto, et al., "Epoxy-silica particulate nanocomposites: chemical interactions, reinforcement and fracture toughness," *Polymer*, **46**, 10506–10516 (2005).

20. P. Rosso, L. Ye, K. Friedrich, and S. Sprenger, "A toughened epoxy resin by silica nanoparticle reinforcement," *J. Appl. Polym. Sci.*, **100**, 1849–1855 (2006).

21. P. K. Vallittu, "Flexural properties of acrylic resin polymers reinforced with unidirectional and woven glass fibers," *J. Prosthetic Dentistry*, **81**, 318–326 (1999).

22. S. Panthapulakkal and M. Sain, "Injection-molded short hemp fiber/glass fiber-reinforced polypropylene hybrid composites – mechanical, water absorption and thermal properties," *J. Appl. Polym. Sci.*, **103**, 2432–2441 (2007).

23. N. Eronat, U. Candan, and M. Türkün, "Effects of glass fiber layering on the flexural strength of microfill and hybrid composites," *J. Esthet. Restor. Dent.*, **21**, 171–178, discussion 179–181 (2009).

24. A. Godara, L. Mezzo, F. Luizi, et al., "Influence of carbon nanotube reinforcement on the processing and the mechanical behaviour of carbon fiber/epoxy composites," *Carbon*, **47**, 2914–2923 (2009).

25. E. Bekyarova, E. T. Thostenson, A. Yu, et al., "Multiscale carbon nanotube–carbon fiber reinforcement for advanced epoxy composites," *Langmuir*, **23**, 3970–3974 (2007).

26. M. F. Uddin and C. T. Sun, "Improved dispersion and mechanical properties of hybrid nanocomposites," *Compos. Sci. Technol.*, **70**, 223–230 (2010).

27. Y. Rostamiyan, A. Fereidoon, M. Rezaeiashtiyani, et al., "Experimental and optimizing flexural strength of epoxy-based nanocomposite: effect of using nano silica and nanoclay by using response surface design methodology," *Mater. Design*, **69**, 96–104 (2015).

28. B. Geisler and F. N. Kelly, "Rubbery and rigid particle toughening of epoxy resins," *J. Appl. Polym. Sci.*, **54**, 177–189 (1994).

29. A. Mirmohseni and S. Zavareh, "Modeling and optimization of a new impact-toughened epoxy nanocomposite using response surface methodology," *J. Polym. Res.*, **18**, 509–517 (2011).

30. A. J. Kinloch, R. D. Mohammed, and A. C. Taylor, "The effect of silica nano particles and rubber particles on the toughness of multiphase thermosetting epoxy polymers," *J. Mater. Sci.*, **41**, 1293–1293 (2006).

31. Y. Rostamiyan, A. H. Mashhadzadeh, and A. SalmanKhani, "Optimization of mechanical properties of epoxy-based hybrid nanocomposite: effect of using nano silica and high-impact polystyrene by mixture design approach," *Mater. Design*, **56**, 1068–1077 (2014).

32. R. Leardi, "Experimental design in chemistry: A tutorial," *Analytica Chimica Acta*, **652**, 161–172 (2009).

33. C. Zhang and W. K. Wong, "Optimal designs for mixture models with amount constraints," *Statist. Probab. Lett.*, **83**, 196–202 (2013).

34. Y. Rostamiyan, A. B. Fereidoon, A. Omrani, and D. D. Ganji, "Preparation, modeling, and optimization of mechanical properties of epoxy/HIPS/silica hybrid nanocomposite using combination of central composite design and genetic algorithm. Part 2. Studies on flexural, compression, and impact strength," *Strength Mater.*, **45**, No. 6, 703–715 (2013).

Received 06. 04. 2015