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# Optimization of Mechanical and Thermal Properties of Elastomer Modified Carbon Fibers/ Phenolic Resin Composites

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## Abstract

In this study, the effect of simultaneously using of chapped carbon fibers and an elastomer as hydroxyl terminated poly butadiene (HTPB) on tensile and impact strength and thermal resistance of phenolic matrix composite was investigated and optimized experimentally. This optimization was done by designing of weight percentages of components using design expert software. To fabricate all of composite samples, mix of resole and novalac phenolic resins (80-20 wt.%) was used as matrix and the HTPB and then carbon chapped fibers were added. Also thermal behavior of composite and pure phenolic was studied through thermogravimetric analyses (TGA) and differential TGA (DTGA). The best result about tensile tests is related to composite contains 55.48 wt.% of carbon fibers and 2.2 wt.% of HTPB. Also the highest impact strength was obtained for composite contain 38.41 wt.% of carbon fibers and 12.8 wt.% of HTPB. Integration of experimental and analytical results showed the optimum weight percentages about 38.41 wt.% of carbon fibers and 2.2 wt. % of HTPB with the achievable tensile and impact strength about 6 times more than phenolic resin. The results of TGA and DTGA tests showed that the thermal resistance of phenolic matrix composite is higher than pure phenolic, so its destruction rate up to 500 ºC is less than pure matrix depend on wt.% of carbon fibers. But the effect of HTPB on thermal resistance of phenolic matrix is negligible and a little negative because of it's fully destruction at 390 ºC.

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**Keywords:** Phenolic composite, Chapped carbon fibers, Elastomer, Mechanical properties, Thermal behavior

## 1. Introduction

Phenolic resins are one of the most used industrial resins today, which despite their many advantages, have some disadvantages as low tensile strength and undesirable impact strength. Long term thermal and mechanical stability, fire and smoke characteristics and low toxicity, electrical and thermal insulating capabilities can be mentioned. Turbine engine components and parts subjected to corrosive environments, foundry, abrasives, friction and refractory are some important applications of phenolic resins [1].

However, Fragility and low tensile strength are the major weaknesses of phenolic resins. The use of additives such as clay, carbon fibers, glass fibers, elastomers etc. improves the mechanical properties as well as the thermal resistance of these resins. The use of elastomers in the structure of phenolic resins increases their toughness and resilience. The simultaneous use of carbon fibers and elastomers can be a good option for increasing the toughness and tensile strength of the phenolic based composite specimens [2].

Ma, et al. [3] have investigated the process ability, mechanical properties and flame resistance of glass fibers reinforced pultruded novalac type phenolic resin based composites. Novolac resin based composite showed good mechanical properties and higher flame resistance than the resol type. Choi, et al. [4] studied the effect of coupling agent on electrical and mechanical properties of carbon fiber/phenolic resin composites. As a result, the coupling agent improved the electrical conductivity and mechanical properties of modified composite.

The effect of high temperature and fire on phenolic resin composites properties has been investigated by Mouritz [5]. He studied the flexural strength and modulus changes of glass, carbon and Kevlar fibers reinforced phenolic, epoxy and polyester resins composites in

different thermal conditions. The results of his research showed that severe decrease in strength and modulus of the phenolic molded composites was happened due to thermal decomposition and cracking in the matrix material.

Thermoplastic polymer additives have been studied in various researches to improve the properties of phenolic resin. Polystyrene-acrylonitrile copolymer is also a suitable enhancer for the toughness of novalac resin [6]. In a study, Wu, et al. used soft segments of polyamides (nylon 6 and nylon 6,6) as a type of thermoplastic, to improve the severity of novalac resin [7]. Polyamide is a highly miscible compound with the structure of phenolic resin which also increases its impact strength due to inherent softness. Thermal degradation temperature was higher than 400°C and increased with the increasing of polyamide content.

Choi, et al. [8] have investigated about the effect of chain length of flexible diacids on morphology and brittleness of the resol type phenolic resin. In their study, the high improved toughness of modified phenolic with suberic acid, has been related to its inherently homogeneous morphology. Improvement of the mechanical properties of phenolic resin has been observed with increasing poly methyl methacrylate (PMMA) [9]. A decreasing trend of tensile strength and modulus with sequent increases in elongation at break, toughness and impact strength was concluded. The thermogravimetric analyses (TGA) results have exhibited lowering in thermal stability of the samples with increase in proportion of PMMA.

Wei, et al. [10], have used new type of elastomer nanoparticles to simultaneous improvement in impact strength, flexural strength and thermal resistance with 5 wt. %. Kaynak, et al. [11] investigated rubber toughening of resol type phenol–formaldehyde resin. It was concluded that, the toughness of acrylonitrile butadiene rubber modified and rubber-silane modified specimens were increased 63% and 50%, respectively, compared with the neat resin samples.

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Hydroxyl terminated poly butadiene (HTPB) was successfully used to modify phenolic novolac resin using resol type as compatibilizer. As the novolac- HTPB blend was chemically inert, resol was used as the compatibilizer. An 80/20 blend of novolac and resole was identified as the optimum system for these studies. According to the results of this study, the optimum amount of elastomer to improve the toughness of phenolic resin is 10% by weight [12].

Due to high toughness, elastic materials are very good additives to modify the properties of thermosets. The use of low-modulus fillers significantly reduces the phenolic resin modulus. Various sources have used different elastomers to modify the properties of thermosets. Most of these studies, have used elastomers to increase the toughness of epoxy resins, and a few have been about modified phenolic resins [13].

The use of low amounts of HTPB increases the crack growth resistance of the phenolic resin. While its use up to 10% improves the polymer bounds and HTPB dispersion and also increases the surface adhesion of the phenolic resin. Lower concentrations of HTPB improved the fracture toughness by forming two phases. Modification with 10 wt.% HTPB and above resulted in finer particle dispersion with a diffused phase morphology. Reactive compatibility is the mechanism responsible for the enhanced miscibility and strong interfacial adhesion between the two phases. [13].

In another study, Ismail, et al. [14] investigated the changs in thermal and mechanical properties of polyvinylbutyral (PVB) modified resol phenolic resin. The differential scanning calorimetry (DSC) and TGA studies showed that, the increase in PVB content results a reduced thermal stability of modified resin, at temperatures above 360 °C. The Izod impact strength of cast molded resin is maximum with PVB content of 10 phr.

Sham Aan, et al. [15] studied the effect of addition of diallylbisphenol A (DABA) on the thermal, mechanical, morphological and flame retardance behavior of Novolac

phenolic/carbon fibers composites. The 8 wt.% DABA modified novolac composites yielded more than 100% increase in impact strength compared to that of unmodified novolac composites, without compromising inherent flame retardant property.

It is concluded from the results of over mentioned studies, that the use of soft compounds in the phenolic resin structure, increases its toughness and impact ability. Also hybrid materials exhibited excellent mechanical properties in which modulus, strength, strain at break and impact strength were improved simultaneously. About 1.5 wt.% of layered silica in the phenolic matrix leads to a certain degree of exfoliation and consequently better structural and mechanical properties. Recently, multi-walled nanotubes (MWNTs) and activated carbon spheres have been used to reinforce the phenolic resin [13].

Mechanical properties modification of a thin film phenolic resin filled with nanosilica particles studied by Taheri, et al. [16]. They experimentally investigated mechanical and wears properties of three different phenolic polymer nanocomposites. For the phenolic polymer with 1 and 2 wt.% nano silica, Young's modulus was increased 7% and 12.5%, respectively in comparison with the neat phenolic resin. However, for sample with 3% nano silica this value has shown a reduction about 13.5% and 2.7% in comparison to 2% nanoparticle filled and pure resin samples, respectively. Also, hardness of nanocomposite with 2 wt.% nano silica was 27.3% more than the unfilled phenolic resin.

A new class of carbon fibers reinforced phenolic composites has been developed for thermal protection systems. This work was done by Fan and Thornton [17]. The materials incorporate thermoplastic polymer segments that were uniformly distributed throughout, and are chemically bonded to the phenolic network. They retained excellent mechanical strength at high temperatures.

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Asaro, et al. [18] studied the ablative properties of phenolic/carbon composites modified with silica particles. Mesoporous silica particles and carbon black were selected as fillers for a resol-type phenolic resin, to be used as a matrix for ablative materials. Composites with 30 wt.% of carbon black and 44 vol.% of carbon fibers achieved the lowest linear erosion rate and the highest insulation index. The composite with 20 wt.% of silica particles exhibited the lowest mass erosion rate.

Qin, et al. [19] investigated nano silica modified phenolic resin film manufacturing and properties. The effect of nano silica on the rheology and curing of phenolic resin are studied by rheometer and DSC. The viscosity of nano silica modified phenolic resin was decreased with the increase of temperature. The lowest viscosity was obtained between 70 and 90 °C. Nano silica mass loading for the improvement of mechanical properties of phenolic composites was found.

Synthesis and structure evolution of phenolic resin/silicone hybrid composites with improved thermal stability studied by J. Yun, et al. [20]. The TGA results indicate effective improvement of thermal stability of the composite compared with cured neat phenolic.

Other researchers studied about thermal behavior of phenolic composites such as study of characterization of phenolic resins with TGA [21], life time of HTPB in thermal conditions [22], and thermal behavior of composite based on phenolic resin and short carbon fibers [23] using DSC and TGA techniques. Also several other investigations have been carried out previously about mechanical properties of carbon fibers/phenolic composites including of tensile strength, elongation at break, flexural strength and modulus [23, 24], and impact properties [25].

In the previous studies, investigation about tensile strength of elastomer modified carbon phenolic composites is very little. Also, about simultaneous improving of tensile and impact

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strength of this kind of composites, any works not accessible. An other hand, in the most of works on properties improvements of these kinds of material, phenolic matrix used often has been based on novolac resin, which has a good mechanical property [3] and a little wt.% of resol type (max. 20%) has been used [12, 13]. Along the previous investigations, this study focused on simultaneously improvement and optimizing of impact energy absorption and tensile strength of HTPB modified chapped carbon fibers /resol based (80 wt.%) phenolic composite, having better thermal resistance than novolac type [18]. This optimization was carried out through simultaneous effects of weight percentages of carbon fibers and HTPB elastomer on mechanical properties of composite, that are the variables in design of tests and fabrication of samples. Also, the comparison of thermal resistant of the pure phenolic resin and carbon reinforced HTPB modified phenolic composite samples was studied by using TGA and differential thermogravimetric analyses (DTGA).

## 2. Experimental

### 2.1. Raw Materials

Phenolic resins (IP-502 Novalac, IL800 Resol) manufactured by Resitane Iranian company as a matrix material, carbon fibers T300 (Torray, Japan) sized by 50A/B sizing material as a reinforcement, and HTPB liquid elastomer (Iranian SKH Co.) were the main raw materials used in the fabrication of the composite specimens. Technical specifications of some raw materials were given as in Tables 1 and 2.

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Table 1- Specifications of phenolic resins used as matrix

| Resin type (wt.%) | Code | Appearance | Solvent | Additives (wt.%) |
|---|---|---|---|---|
| Novalac (20) | IP502 | Solid | Ethanol | Stearic acid (3) |
| Resol (80) | IL800 | Liquid | - | Stearic acid (3) |

Table 2- Characteristics of HTPB elastomer

| Property | HTPB |
|---|---|
| $M_n$ (average molecular mass) | 5210 |
| DPI (dispersion index) | 2.53 |
| Viscosity (cp) at 25 ºC | 2320 |
| Hydrxyl number (mg KOH/g) | 42 |
| Microstructure: vinyl; ciss; trans | 7; 64; 29 |
| $T_g$ (ºC) | -78 |

**2.2. Test Design**

In order to prevent any personal comments on the results obtained, the tests were designed using Design of Expert (DE) software “Series 8" and the central combination design using response surface methodology (RSM). Considering the widespread use in scientific research as well as the rotation capability and accurate and reliable results, this method was used to enter the variables (numerical factors) in software and registering number of the responses with their measurement units. The studied variables and their responses are listed in Table 3. Finally, after entering the variables in the software, the effect of each variable was evaluated in the form of graphs and equations of variance.

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Table 3- The wt.% of variables designed using the design expert software

| No. | Randomization | HTPB (wt.%) | Carbon fibers (wt.%) | Primary surface area ($mm^2$) |
|---|---|---|---|---|
| 1 | 7 | 7.50 | 32.50 | 45.26 |
| 2 | 1 | 2.20 | 38.41 | 45.82 |
| 3 | 5 | 2.20 | 6.59 | 46.21 |
| 4 | 2 | 7.50 | 45 | 42.87 |
| 5 | 9 | 0 | 22.50 | 46.03 |
| 6 | 6 | 12.80 | 6.50 | - |
| 7 | 3 | 7.50 | 65 | 46.21 |
| 8 | 4 | 12.80 | 38.40 | 45.22 |
| 9 | 11 | 15 | 42.50 | 47.39 |
| 10 | 10 | 7.50 | 0 | 47.22 |
| 11 | 8 | 7.50 | 22.50 | 47.30 |

It should be noted that the design of the tests was performed based on the wt.% rang of 0 to 65% of carbon fibers and 0 to 15% of HTPB to examine the effect of these two variables both simultaneously and separately. Tensile strength and impact resistance tests carried out by applying three samples from each type of composite specimens and also net resin cured samples.

## 2.3. Fabrication of Experimental Specimens

### 2.3.1. Tools and Devices

In this study, a mechanical mixer was used to make different types of phenolic matrix material by adding ethanol, acetone and stearic acid. A composite molding tool was used to make composite samples according to ASTM D651. This mold has two punches and two shoelaces made of steel 6582.

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In order to determination of curing thermal cycle (heating temperature and time) of phenolic composite, kinetic of curing process studied by using of differential scanning calorimetry (DSC) device (Nezsch - F3 Maya200 model). Also for determination of B-stage optimum time of phenolic composites, flowing tests of some samples were performed. Fig. 1 shows the tools of flow test of phenolic composite.

Fig. 1- Tools of flow test of composite samples for determination of B-stage optimum time.

A Bottenfield hot press (HP) machine with 100 tons load and complete equipment such as thermocouples and thermostats were used to make the composite samples. Chisel and ring elements of HP system, are connected to the mold by the refractory cables to heating it, during pressing and molding of the composite specimen. The current intensity in these elements is between 500 and 800 A. A thermocouple and thermal thermostat were also used to indicate and control of temperature of the metallic mold, up to 160 °C.

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TGA and DTGA analysis of molded phenolic resin (S-0) and composite sample (S-1) were carried out by using a TGA analyzer (UK polymer laboratories, PL1500) having the range of ambient temperature up to 800 ºC with the rate of 20 ºC/min.

Tensile strength tests performed by using tensile test machine (Instron, model 6025) with 20 KN loading unit. Traction braces are used to hold the specimens on the traction device in accordance with ASTM D 651 as shown in Fig. 2. Also Izod impact test device (Zwick, model 5113) was used to perform impact tests of samples.

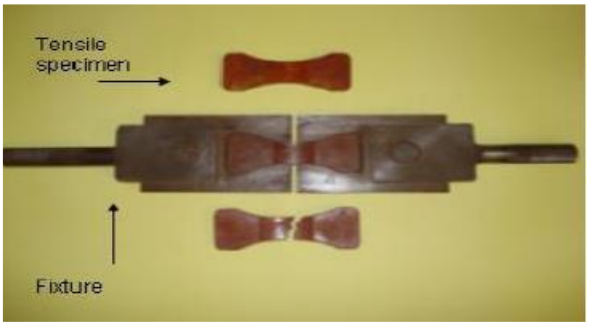


Fig. 2 - Closure of tensile strength in accordance with ASTM D 651.

### 2.3.2. Fabrication of Composite Samples

Initially in order to determination of B-stage condition and curing thermal cycle used for making of samples, the DSC analysis and also flowing tests of S-0 and S-1 samples were performed. But to avoid prolonging the content, their description will be omitted. Results of DSC analyses and flow tests consist choosing cycle of 3 h at 80 ºC for B-stage and 160 ºC during 2 h for purposed curing process.

For fabrication all of samples, mix of 80 wt.% resole and 20 wt.% novalac phenolic resins was placed in an electric shaker for 3 h. In each sample to the extent specified in the design of the tests, the HTPB was added to the mixture of resol and novalac matrix and placed in an ultrasonic mixer for two ten-minute intervals. The carbon chapped fibers were then pressed into the obtained blend, spread on a plastic wrap, placed in an oven (Frishluft, Germany) at 80

°C for 3 h reaching B-stage phase. Finally, in order to forming and purposed curing, precomposite material was transferred to the mold, and placed in the press machine for 2 h under a temperature of about 160 ºC. At the end, the machine was switched of and the mold was cooled and finally, the piece was removed from the mold. This procedure was performed 3 times for all samples specified by the test design method and then each sample was prepared according to the related standard for tensile and impact tests.

### 2.4. Test of Specimens

#### 2.4.1. Thermogravimetric (TG) Test

The TG analysis of two molded cured samples including of the pure phenolic matrix (S-0) and a composite sample (S-1) containing 32.5 wt.% of carbon fibers and 7.5 wt.% of HTPB and 60 wt.% of phenolic matrix, were studied comparisional at 50% humidity according to ASTM E1131 method.

#### 2.4.2. Mechanical Tests

Tensile strength tests performed at ambient temperature and humidity of 41% and according to ASTM D638-10 standard. Traction braces are used to hold the specimens on the traction device in accordance with ASTM D 651. As the same way, Izod impact test was performed at the ambient temperature and 50% humidity under 2.7 J pendulum force, according to ASTM-D 256.

## 3. Results and Discussion

### 3.1. Thermogravimetric Test Result

The Figs. 3 and 4 show the comparisional results of TGA and DTGA of S-1 and S-0 samples. As shown in Fig. 3, the TGA diagram of the S-1 sample shows three slope change

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zones, which appear as peaks in the DTGA diagram shown in Fig. 4 included of 3 zones. First zone is included of the range of ambient temperature to 300 °C. About 20% of weight loss occurred, which could be due to the volatility of volatiles in the sample, such as alcohol, or volatiles due to initial reactions of H and OH and log-out, in accordance with reference [21].

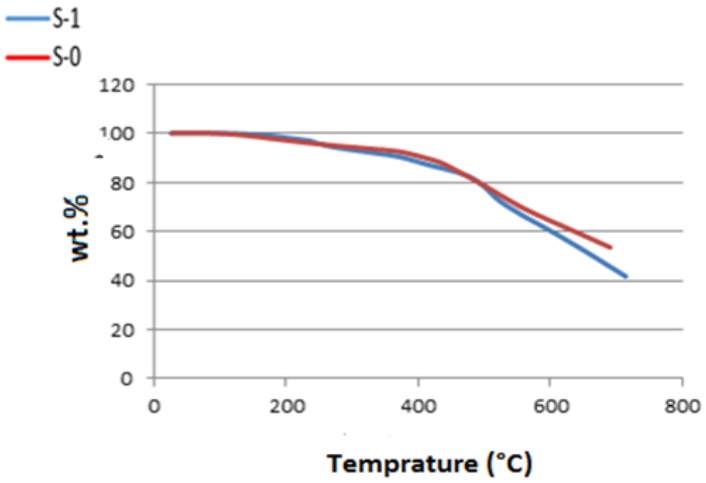


Fig. 3- Comparisional TGA diagrams of S-0 and S-1 samples.

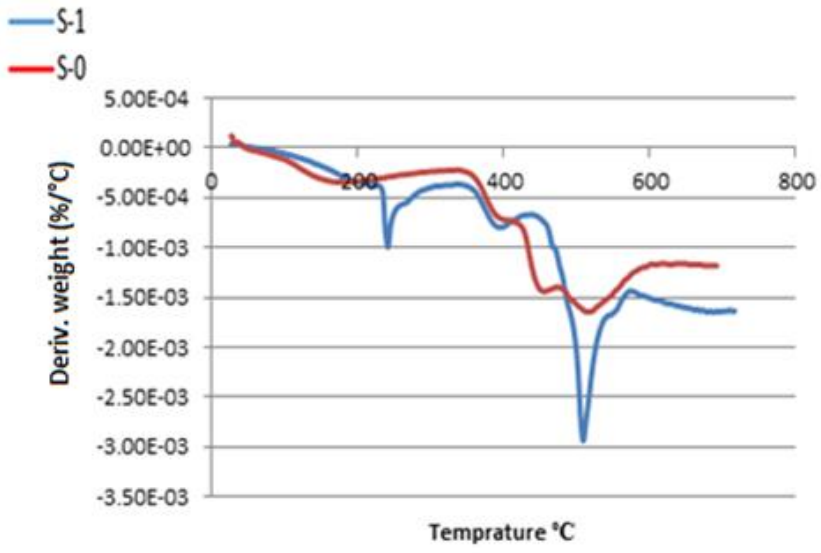


Fig. 4- Comparisional DTGA diagrams of S-0 and S-1 samples.

The second zone is at the range of 350-450 °C. In this area, an almost wide peak is observed at 390 °C. At this temperature, about 7 wt.% loss occurred, which could be the HTBP

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degradation and starting point of the resin destruction, as has been showed in previous studies [21, 22]. The third area is at the range of 450-580 °C. The peak occurred at 570 °C with a 21 wt.% loss which is the largest weight loss that begins with the destruction of all resins and carbon fibers [21, 25]. Finally, 97.8% of the material was completely destroyed and 2.2% of the material remained.

According to Fig. 4, the termogravimetric behavior of the pure phenolic resin sample (S-0), includes of two important areas. The first area has a peak that occurred at 111 °C with a 10% decrease in volume that is related to volatile materials. The secondary region is related to a peak that occurred in the temperature range of 350-500 °C, with a slope drop at 412 °C that shows the initiate of degradation and weight loss of the phenolic resin. Previous studies have shown the same trends [14, 21].

The comparison of TGA diagrams of S-1 and S-0 samples shows that, each two curves start at 100% and are observed with minimal difference up to 290 °C, which can be related to volatile materials. Later that Up to 490 °C, the S-1 diagram is lower than the S-0 diagram, with a steeper slope that may indicate the destruction HTPB [22] and onset of weight loss associated with the phenolic resin [15, 21], and so two diagrams separated from each other at 490 °C. Also, the slope of the S-1 diagram is high steeper that related to degradation and weight loss of carbon fibers in accordance with similar works [17, 25].

Fig. 4, shows the comparison of DTGA diagrams of S-1 and S-0 samples. As can be seen at this diagrams, the two curves have a same starting point, and initially the S-0 sample has an initial drop before of 200 ºC which can be due to the high percentage of H and OH. It then proceeds directly, but in the meantime, the S-1 chart gives a peak due to the onset of weight loss and destruction of the HTPB [22]. They then make a simultaneous drop peak at the range of 360-400 ºC that related to begin of weight loss and degradation of resol phenolic resin whitch

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is connected to another drop at range of 430-480 ºC for S-0 related to continue of degradation of phenolic resin [14, 21]. In the following, 490-580 ºC a simultaneous drop peak is observed, but the diagram of S-1 gives a long peak showing carbon fibers degradation [15, 25].

As a final result, given that the degradation of phenolic resin starts from about 360 ºC and carbon fibers showed good thermal resistance up to 500 ºC, so the thermal resistance of phenolic base composite is higher than pure resin and its destruction rate up to 500 ºC is less than pure matrix depend on the wt.% of carbon fibers. In the case of HTPB, Figs. 3 and 4 show that the fully degradation of this elastomer occurs at the temperatures lower than phenolic resin degradation, so it can improve a little the thermal resistance of composite at the temperatures below 390 ºC, but causes decrease it at higher temperatures [21, 22]. So using the higher wt.% of HTPB don't recommended for thermal applications.

### 3.2. Tensile Test Results

The results of the tensile tests and Izod impact resistance of specimens have been presented in Table 4.

Table 4- Tensile and Impact strength test results of phenolic resin composite specimens

| No. | Randomization | HTPB (wt.%) | Carbon fibers (wt.%) | Impact strength ($kJ/m^2$) | Tensile strength (MPa) |
|---|---|---|---|---|---|
| S-0 | - | 0 | 0 | 0.2 | 7 |
| S-1 | 7 | 7.5 | 32.5 | 1.8 | 33 |
| S-2 | 1 | 2.2 | 38.41 | 1.4 | 45 |
| S-3 | 2 | 7.5 | 45 | 1 | 54 |
| S-4 | 9 | 0 | 22.5 | 0.9 | 36 |
| S-5 | 6 | 2.2 | 55.48 | 0.75 | 63 |
| S-6 | 3 | 7.5 | 65 | 0.4 | 14 |
| S-7 | 5 | 2.2 | 6.59 | 0.3 | 18 |
| S-8 | 4 | 12.8 | 38.41 | 2.1 | 32 |
| S-9 | 11 | 12.8 | 55.48 | 0.88 | 39 |
| S-10 | 10 | 7.5 | 0 | 0.5 | 10 |
| S-11 | 8 | 7.5 | 22.5 | 1.45 | 28 |

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According to the Table 4, with increasing carbon fibers up to 45 wt.% in present of 7.5 wt.% of HTPB (S-3), the tensile strength of the composite increases up to 54 MPa, that's about 7 times more than the net resin sample (S-0). Also, for sample reinforced with 55 wt.% of carbon fibers (S-5), the tensile strength increases to about 9 times of S-0. But excess increasing in the percentage of carbon fibers up to 65 wt.% (S-6) with the 7.5 wt.% of HTPB causes to decrease of tensile strength of composite sample to 14 MPa. It is very loose compared to previous examples, showing negative effect of excess amounts of carbon fibers on tensile strength of composite. It is related to the non-wetting of the fibers by the resin that is the most important factor for effective interaction of them.

Fig. 5. shows the changes of the tensile strength of composite samples by varying of wt.% of carbon fibers content. It can be said that, the enhance of the tensile strength of composite by increasing of wt.% of carbon fibers up to 40% is fully linear that, almost similar result with reference [24].

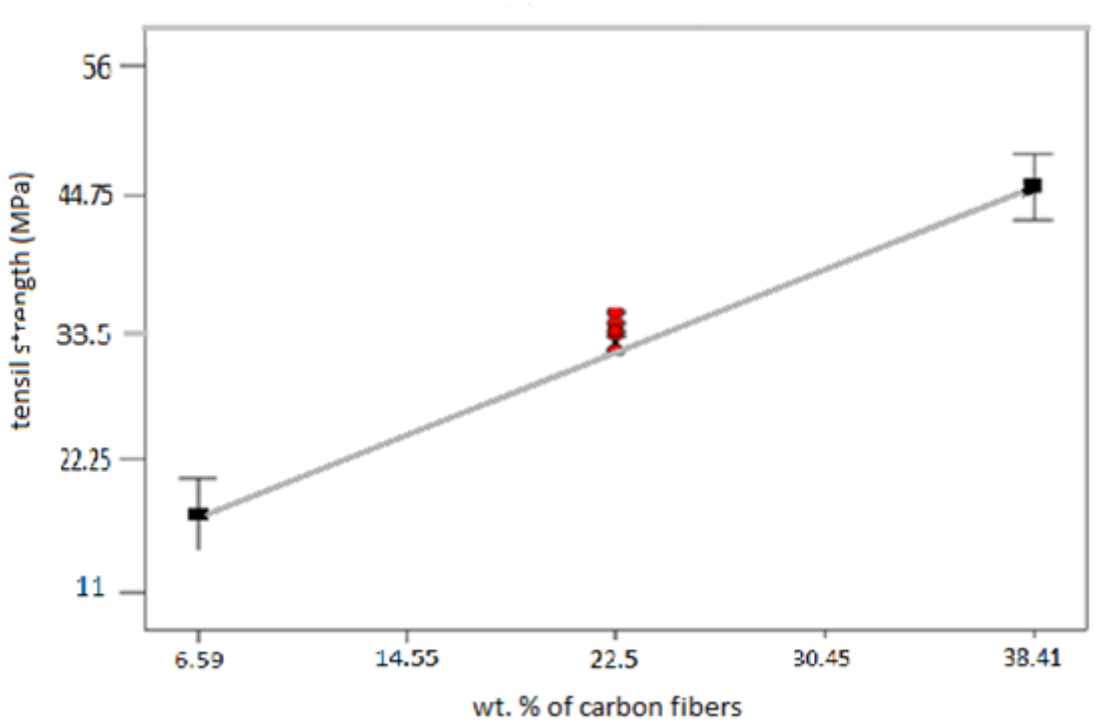


Fig. 5- changes of the tensile strength of composite by varying wt.% of carbon fibers.

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The results of analysis of variance of experimental findings related to tensile strength using DE software are shown in Table 5.

Table 5- The results of analysis of variance of experimental data related to tensile strength

| Source | Sum of squares | Df | Mean square | F-value | P-value | Considering |
|---|---|---|---|---|---|---|
| Model | 1612.88 | 2 | 806.44 | 46.87 | <0.0001 | Significant |
| A-Carbon fibers | 1530.14 | 1 | 1530.14 | 88.94 | <0.0001 | |
| B-HTPB | 82.74 | 1 | 82.74 | 4.81 | 0.0531 | |
| Residual | 172.04 | 10 | 17.20 | | | |
| Lack of fit | 167.04 | 6 | 27.84 | 22.27 | 0.0049 | Significant |
| Pure error | 5.00 | 4 | 1.25 | | | |
| Cor total | 1784.92 | 12 | | | | |

The meanings of the terms listed in the Table 5 are as follows:

Sum of squares- Total effects of variables on the model

Df- Degree of model freedom and variables

Mean square- Normalized effect of the variables

F-Value- The effect of each variable on the model, which indicates the significance of the model

P-Value- Model risk-taking or uncertainty of the effect of variables on the model, which is inversely related to the significance of the model.

A- The coded variable of weight percent of carbon fibers.

B- The coded variable of weight percent of HTPB.

The mathematical model proposed by the software for estimating tensile strength was as follows:

$$\text{Tensile strength} = +31.42 + 13.83\,A - 3.22B \quad (1)$$

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The higher F-Value the lower P-Value, the greater the significant and value of model. The P-Value of the given model was calculated by the variance analysis of software by 0.0001. Considering its critical value in most references as well as experimental design (critical value of P-Value in experimental design, often considered 0.05) indicates the importance and meaningful of the model and its high reliability. Equation (1) indicates a direct relation between tensile strength and the wt.% of fibers used in the preparation of phenolic base composite samples up to a reasonable and logical extent.

The result of analysis of variance indicates that, the tensile strength of the specimens depends not only on the amount of fibers but also the amount of HTPB elastomer on the tensile strength. In the proposed model, the width of the source indicates the tensile strength of the pure phenolic (without fibers and elastomers) sample.

The model given by the software shows the effect of each material on increasing or decreasing of the tensile strength. In the case of HTPB, after an initial positive effect, it decreases the tensile strength of composite samples, which is a reasonable result because of its softening and flexural effect as an elastomer.

As it is known from Table 5, effect of carbon fibers on tensile strength (total effect: 1530) very more than HTPB effect (total effect: 82.8) by about 19 times, and also HTPB, after an initial positive effect plays negative role on tensile strength of composite. So the highest amount of tensile strength occurs in areas where carbon fibers have the highest and liquid elastomers have the least amounts. It is also evident that the lowest tensile strength is related to the pure phenolic resin sample (S-0). It can be said that by increasing the amount of carbon fibers up to about 40%, the tensile strength of the composite samples increases with an almost constant slope (Fig. 5). However, the simultaneous application of carbon fibers and liquid elastomers resulted in the highest tensile strength regarding optimal impact strength. It is

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clearly evident that both carbon fibers and elastomer addition increased the tensile and impact strength compared to the S-0 sample in the logical extent.

According to the experimental data (Table 4), the highest tensile strength is 63 MPa related to the S-6 sample, which has 55 wt.% carbon fibers and 2.2 wt.% HTPB. But in order to achieve desirable tensile strength and impact strength, optimization is needed. This was done by software and optimized points were suggested for maximum tensile strength with considering optimal impact resistance. The integration results presented as a three-dimensional graph of the tensile strength changes based on two variables shown as Fig. 6 and normalized in Table 6.

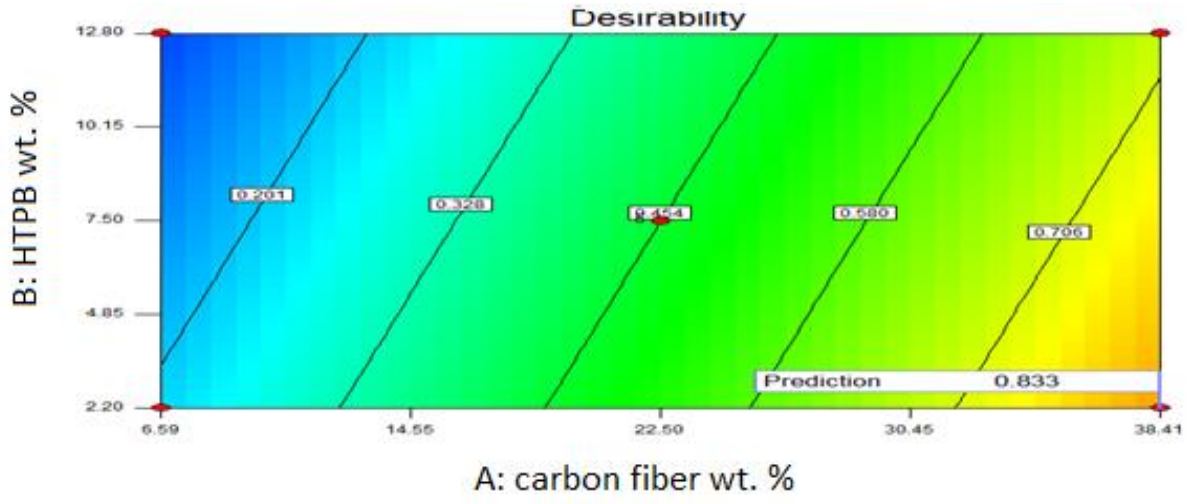


Fig. 6- Three-dimensional graph of the changes of the tensile strength of composite based on two variables, including carbon fibers and HTPB elastomer weight percentages.

Table 6 - Software proposed optimal points for maximum tensile strength of composite

| No. | Carbon fibers (wt.%) | HTPB (wt.%) | Tensile strength (MPa) | Desirability | Considers |
|---|---|---|---|---|---|
| 1 | 38.41 | 2.20 | 48.469 | 0.833 | Selected |
| 2 | 38.29 | 2.20 | 48.3631 | 0.830 | |
| 3 | 37.97 | 2.20 | 48.0878 | 0.824 | |
| 4 | 38.41 | 3.96 | 47.3973 | 0.809 | |

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### 3.3. Impact Strength Results

According to Table 4, the samples with the most carbon fibers wt.% (S-3; 45%, S-5; 55.48%, and S-9; 55.48%) despite having high tensile strength, they have low impact strength. So the result of combination analyses of DE software considering the optimal state of both traction and impact parameters, gives 4 optimum point that have shown in Table 6. So the best optimum point selected by software consist having 38.41 wt.% of carbon fibers and 2.2 wt.% HTPB with calculated tensile strength of 48.5 MPa that according to Fig. 6 and Table 6, have a good desirable.

Based on the results obtained due Izod impact tests (Table 4), it can be concluded that the samples without carbon fibers and elastomer have the least impact strength (S-0; 0.2 $kJ/m^2$). Comparing the samples with equal weight percentage of carbon fibers, it is clear that the samples containing higher percentages of liquid elastomer have higher impact resistance. Sample of S-8 (38.41 wt.% of carbon fibers and 12.8 wt.% of HTPB), having 2.1 $kJ/m^2$ impact strength compared with sample of S-2 (same amount of carbon fibers and 2.2 wt.% of HTPB) that has given 1.4 $kJ/m^2$ impact strength because of softening and flexible properties of HTPB as an elastomer as mentioned in references [12, 13].

As stated, amount of carbon fibers higher than 40 wt.% have negative effect on impact strength of composite samples. For example; S-8 (38.41 wt.% carbon fibers) and S-9 (55.48 wt.% of carbon fibers) with equal amount of HTPB have 2.1 and 0.88 $kJ/m^2$ of impact strength, respectively. At the same way other cases such as comparing of S-1 with S-5, and also S-1 and S-3 could be exampled comparisional. The reason of such behavior of composite samples is the brittleness property of carbon fibers that causes decline of impact strength of composite in case of excess presence. The results of analysis of variance of experimental findings related to impact strength using DE software are shown in Table 7.

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Table 7- The results of analysis of variance of experimental data related to impact strength

| Source | Sum of squares | Df | Mean square | F-value | P-value | Considers |
|---|---|---|---|---|---|---|
| Model | 2.53 | 3 | 0.84 | 20.98 | 0.0002 | Significant |
| A-Carbon fibers | 0.92 | 1 | 0.92 | 22.82 | 0.0010 | |
| B-HTPB | 0.93 | 1 | 0.93 | 23.23 | 0.0009 | |
| $A^2$ | 0.68 | 1 | 0.68 | 16.89 | 0.0026 | |
| Residual | 0.36 | 9 | 0.040 | | | |
| Lack of fit | 0.36 | 5 | 0.072 | 107.06 | 0.0002 | Significant |
| Pure error | 2.680E-003 | 4 | 6.700E-004 | | | |
| Cor total | 2.89 | 12 | | | | |

So, the mathematical model proposed by the software to estimate the impact resistance of the Izod test method was as follows:

$$\text{Impact Izod Strength} = +1.46 + 0.34A + 0.34B - 0.31A^2 \quad (2)$$

As shown in the Table 7, the P-Value of the given analytical model is 0.0002, indicating that the model is significant. Considering the equation (2), it can be seen that the both factors of carbon fibers and HTPB weight percentages affect impact strength of the composite almost the same size (A-carbon fibers; 0.92 and B- HTPB; 0.93). So, both variables increase the impact property simultaneously at the certain ranges of weight percentage and therefor the equation (2) is a degree 2 equation. These effects are called interaction effects.

Fig. 7 shows the interaction effects zone of two variables. The points of intersection of two curves, indicate the simultaneous effects and interaction of two variables consist zone of simultaneously improving of impact strengths by wt.% increasing of both variables.

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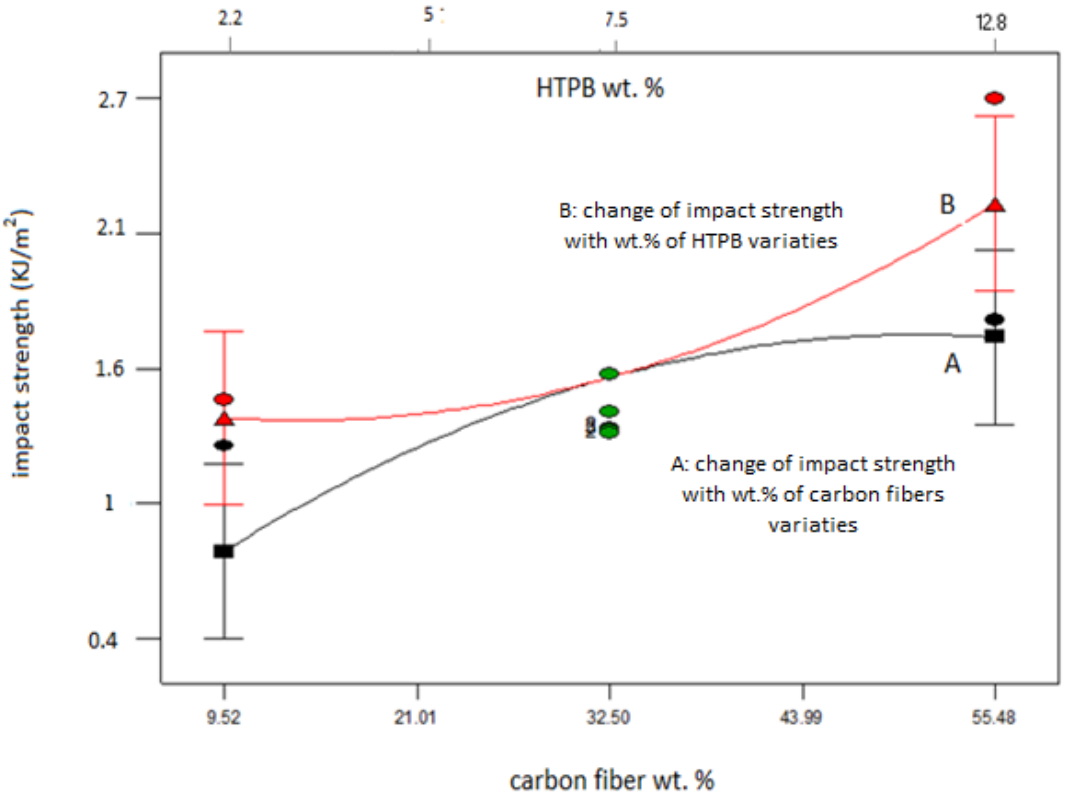


Fig. 7- Interaction effects of carbon fibers and HTPB weight percentages on impact resistance of composite specimens.

The graph A is related to the effect of the weight percentage of carbon fibers and the graph B shows to the effect of HTPB on impact strength of composite. It is evident that increasing of wt.% of carbon fibers up to 38.5 causes enhance of impact strength, thereafter the slope of the increase becomes smoother and eventually becomes negative.

Fig. 8. Shows a three-dimensional graph of the Izod impact resistance changes based on the two variables of carbon fibers and HTPB elastomer weight percentages. As shown in Fig.8, unlike the tensile strength curve, the highest impact resistance of the specimens occurs when the both carbon fibers and HTPB content are maximum. It is well known in the Figure that the shape is a bell and has optimum points. It shows that not only the elastomer but also the carbon fibers up to a certain percentage increase the impact strength and decrease it afterwards. There

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is a huge increase in the impact of the specimens. It is clear that the pure phenolic resin sample showed the least impact resistance. It is evident that the use of carbon fibers with higher intensity and the use of liquid elastomer with close intensity improve the impact resistance of the specimens.

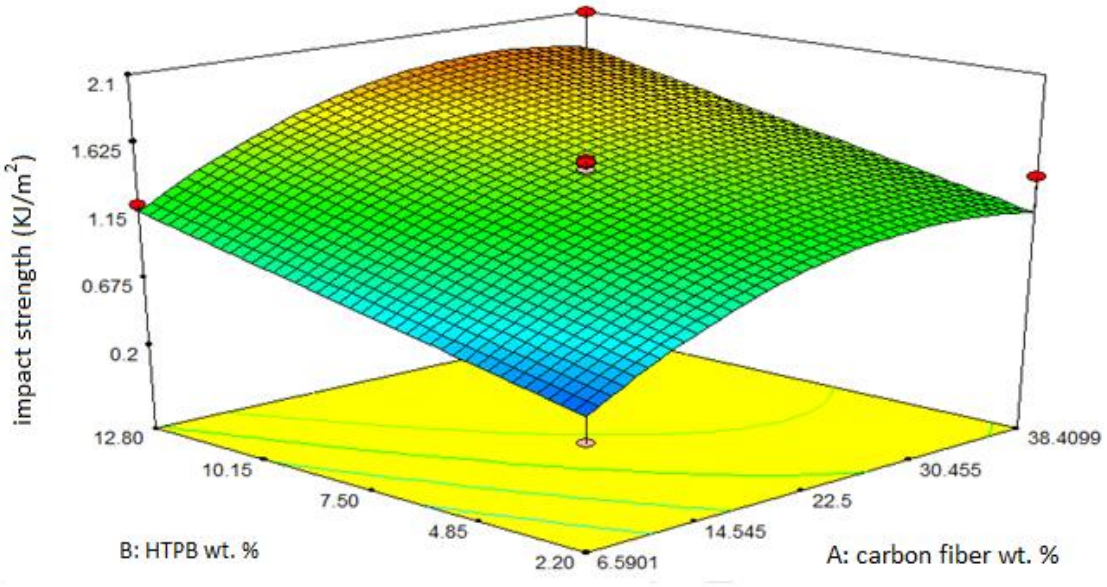


Fig. 8- Three-dimensional diagram of the Izod impact resistance changes based on two variables including carbon fibers and HTPB elastomer weight percentages.

## 4. Conclusion

In this study, effect of simultaneously using of chapped carbon fibers and an elastomer as hydroxyl terminated poly butadiene (HTPB) elastomer on tensile and impact strengths of phenolic matrix compound was investigated and optimized experimentally. This optimization was done by designing of combination of composite with optimum weight percentages of carbon fibers and HTPB elastomer using design expert software. Also TGA and DTGA of a composite sample and a pure phenolic resin molded sample were performed comparisional for study of thermal behavior of them. The final results were as following:

1. The results of thermal analyses showed that the thermal resistance of phenolic base composite is higher than pure phenolic and its destruction rate up to 500 ºC is less. It is a function of the weight percentage of carbon fibers, but the effect of HTPB on thermal resistance of phenolic matrix is negligible and a little negative.
2. Effect of carbon fibers on tensile strength is very more than HTPB effect (about 19 times). Also, the HTPB after an initial positive effect, decreases the tensile strength of composite samples, because of its softening and flexural effect as an elastomer.
3. The p-Value (risk taking) of the given model was calculated by the variance analysis of software by 0.0001, related to great significant and value of the mathematical model proposed by the software for estimating tensile strength and its high reliability.
4. With increasing carbon fibers up to 55.48 wt.%, the tensile strength of the composite sample increases up to about 9 times of net phenolic sample. But excess increasing in the percentage of carbon fibers up to 65 wt.% with the 7.5 wt.% of HTPB causes to decrease of tensile strength of composite sample to 14 MPa, because of the noncomplet-wetting and impregnation of the fibers by the matrix.
5. The samples with equal weight percentage of carbon fibers, containing higher percentages of HTPB elastomer have higher impact resistance. The sample of content 38.41 wt.% of carbon fibers and 12.8 wt.% of HTPB, having 2.1 $kJ/m^2$ impact strength compared with sample of contains same amount of carbon fibers and 2.2 wt.% of HTPB that has given 1.4 $kJ/m^2$ impact strength.
6. So, the amount of carbon fiber was a significant effect on increasing of the tensile strength of the specimens, and HTPB elastomer effected on the increase of the vulnerability and impact resistance of the samples.

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**7.** The samples with the most carbon fibers wt.% (45 and 55.48) despite having high tensile strength, they have low impact strength. So the result of combination analyses of DE software considering the optimal state of both traction and impact parameters, gives the desirable optimum point consist having 38.41 wt.% of carbon fibers and 2.2% HTPB with calculated tensile strength of 48.5 MPa and impact strength about 1.45 kJ/m$^2$.

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