

# Optimization of Mechanical Properties of Rubber/Silica/Epoxy Nanocomposites by RSM

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**ABSTRACT:** In this paper, Young's modulus (E), fracture toughness ( $K_{IC}$ ) and fracture energy ( $G_{IC}$ ) of composites reinforced with rubber and silica nanoparticle was investigated. In composites reinforced with a type of nanoparticle, rubber and silica nanoparticles has been used up to 15 wt% and 20 wt%, respectively. Also the maximum weight fraction of rubber and silica was 10 wt% in hybrid composites. The results have exhibited dramatic increase for hybrid of both nanoparticles in a composite. Eke, each mechanical properties modelled by Response Surface Method and formulas are offered. In addition, in the lack of presence of rubber and 19.8 wt% silica, maximum value for Young's modulus was equal to 3.477 GPa. This method showed that the best quantity of fracture toughness was 4.051 MPa.m<sup>1/2</sup> in 15 wt% of rubber and 20 wt% from silica and the best quantity of fracture energy is 4.974 KJ/m<sup>2</sup> in 15 wt% rubber and 20 wt% silica.

**KEYWORDS:** Mechanical properties, Nanoparticles, Response Surface Method, Modelling

## I. INTRODUCTION

Epoxy matrix loaders as a widely in the field of automotive industry, various, such as for aviation, maritime industry and so have the application And also because of useful features, such as the proper hardness, high strength, low cost can be used. [1]

But some of their properties such as strength, low resistance and low fragility against the growth of cracks, researchers and scientists forced to have been to improve and strengthen the embedded solution they that one of these solutions are add nanoparticles, such as silica, aluminum oxide, rubber, carbon nanotubes. [2-4]

Silica nanoparticles simultaneously increases fracture toughness and elastic modulus. It is also a significant impact on the rubber compound high stiffness of the polymers is capable of. Razavi nuri [5] Mechanical properties of acrylonitrile butadiene rubber on the PE were examined. They used three particles weight fraction rubber; 20, 40 and 60 wt% that the maximum volume Young's modulus obtained where 70% polymer, 20% filler and 5% Organocaly was used that it was improved 32%. Reinholds et al [6] searched about acrylonitrile rubber and Polyethylene. They have the possibility to get some of mechanical properties by using 26 wt% of rubber. Elastic modulus increased till 1200 MPa that improved 20% and also Tensile strength 35 MPa increases that it improved 13%.

Zhang et al [7] have studied the effect of silica on epoxy. Temperature and particle weight fraction were tested. They reported that maximum volume of Young's modulus is 4.47 GPa with 15 wt% and 23 C that it improved 60%. Maximum volume for fracture toughness is 1.26 MPa.m<sup>1/2</sup> where the particle weight fraction was 15 wt% and the temperature was 80 C that the growth was equivalent to 125% and also they reported maximum volume for fracture energy; it was 348 J/m<sup>2</sup> where the particle weight fraction was 15 wt% and the temperature was 80 C that it improved 248%.

Liang & Pearson [8] Influence of silica and rubber compound based on acrylonitrile were examined. Volume fracture matrix and nanoparticles were two parameters were discussed. The results of the experiment show reducing the three mechanical properties; fracture toughness and Young's and fracture energy; so that the value of Young's modulus in 20% of acrylonitrile and with a size of 80 nm nanoparticles; Young's modulus declined from 1.62 GPa to 0.85 GPa and also fracture toughness decreased from 2.49 MPa.m<sup>1/2</sup> to 1.48 MPa.m<sup>1/2</sup>. Fracture energy changed too; it declined 3.25 J/m<sup>2</sup> to 2.18 J/m<sup>2</sup>. The results for 20 nm nanoparticles size also cuts down of the show. Fu et al [9] in a review article, the

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effect of particle size, particle content and the interaction between particles and matrix on mechanical properties of several different manufactures with different reinforcement reviews.

Wan et al [10] the impact of the graphene oxide and silica addition on the epoxy were examined and their results show that with the addition of 0.1 wt% of the composition of fillers, an increase of 39% for fracture toughness and 83% for fracture energy and toughness of failure was pose. They reported  $0.659 \text{ MPa.m}^{1/2}$  for fracture toughness and  $127.5 \text{ J/m}^2$  for fracture energy. Liu et al [11] examined the influence of rubber and silica nanoparticles on the investigated epoxy. They are using different rubber and silica weight fraction. They searched for properties of Young's modulus, fracture toughness and fracture energy. They reported their results as presented in table 1 but they did not state their opinion on the best volume of mechanical properties for two nanoparticles. The experimental results have been modelled through response surface method in this article with regard to the above mentioned explanations. The optimal values of Young's modulus, fracture toughness and energy have then been calculated through genetic algorithm and based on two input parameters of nanoparticles.

## II. EXPERIMENT

### Materials

The resin was prepared diglycidyl ether of bisphenol A (DGEBA). Nano silica particle with an average size of 20 nm and rubber nanoparticles with an average size of 100 nm were used for this experiment.

Preparing the nanocomposites.

According to the desired weight fraction and after the composition, they stired by mechanical agitator for 2 hours under vacuum and eventually was added curing agent to the mix. Then the mixture was poured into a preheated mold for curing at 120 C for 16-22 hours.

## III. RSM MODELING

Response Surface Methodology (RSM) is a method for understanding the correlation between multiple input variables and one output variable. In this approach, an approximation to the response of the aluminium columns is assumed a series of the basic functions in a form of,

$$\tilde{y}(x) = \sum_{j=1}^N a_j \phi_j(x) \quad (1)$$

Where N represents number of basis function, x A typical class of basic functions is the polynomials, for instances, whose full quartic form is given as:

$$\tilde{y} = a_0 + a_1x_1 + a_2x_2 + a_3x_3 + \dots \quad \text{Linear terms} \quad (2)$$

Low- order polynomials are usually used to find the best order of the model. The approximation function of the response surface method is called first order method if the response is modelled through a linear function of the independent variables.

## IV. MODEL EVALUATION

There are many methods and equations for evaluating all numerical models. The most popular being the mean squared error method. The mean squared error is calculated in accordance with the following equation and based on the mean square difference between the model value and the experimental value.

$$MSE = \frac{\sum_{i=1}^M (y_i - \bar{y}_i)^2}{M} \quad (3)$$

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There are other methods aside from this criterion. The relative error is for example a very suitable criterion for analysing the validity of the model in case there is no zero in the experimental data.

$$RE = \left| \frac{y_i(x) - \tilde{y}_i(x)}{y_i(x)} \right| \times 100 \quad (4)$$

Other common methods in the field of conformity of models include Pearson's correlation coefficient or Pearson product-moment correlation coefficient which measures the linear correlation between the output value of the model and the experimental output value. The value of this coefficient varied between -1 and 1. "1" means complete correlations, "0" indicates the absence of correlation, and "-1" means complete negative correlation. This coefficient was developed by Carl Pearson based on the initial idea of Francis Galton. Equation 5 shows the mathematical form of this coefficient.

$$\rho(y, \tilde{y}) = \frac{\text{cov}(y, \tilde{y})}{\sigma_y \sigma_{\tilde{y}}} = \frac{E((y - \mu_y)(\tilde{y} - \mu_{\tilde{y}}))}{\sigma_y \sigma_{\tilde{y}}} \quad (5)$$

It is important for the absolute value of this criterion to be as close as possible to zero when matching two models. The farther it is from one the more inefficient and weak the model. They sometimes express this parameter as percentage which will have a value between zero and one hundred.

### V. EXPERIMENTAL RESULTS

#### Young's modulus

Tensile test was carried out based on ASTM D638-99 standard by an Instron 5567. The tests were conducted in the room temperature. 5 samples selected for the test. 50 mm and 25 mm gauge lengths were prescribed, respectively, for the longitudinal and lateral strain signals. The results of Young's modulus showed in table 1; this parameter without nanoparticle was equal 2.68 GPa. When nanoparticle added to composite, this parameter increased. The maximum of Young's modulus was 3.20 GPa where silica added till 20 wt%. This indicates an increase equal to 19percent.

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**Table 1- Young's modulus, fracture energy and toughness of the composites reinforced with different volume percentages of silica nanoparticles**

| Sample | E (GPa) | K <sub>IC</sub> (MPa.m <sup>1/2</sup> ) | G <sub>IC</sub> (Kj/m <sup>2</sup> ) |
|--------|---------|---|--------------------------------------|
| E      | 2.68    | 0.951                                   | 0.277                                |
| S2     | 2.88    | 1.01                                    | 0.308                                |
| S4     | 2.93    | 1.14                                    | 0.390                                |
| S6     | 2.98    | 1.26                                    | 0.465                                |
| S8     | 3.12    | 1.39                                    | 0.546                                |
| S10    | 3.14    | 1.57                                    | 0.690                                |
| S12    | 3.20    | 1.70                                    | 0.791                                |
| S20    | 3.48    | 2.11                                    | 1.12                                 |
| R2     | 2.63    | 1.01                                    | 0.341                                |
| R4     | 2.51    | 1.31                                    | 0.604                                |
| R6     | 2.45    | 1.62                                    | 0.946                                |
| R8     | 2.33    | 1.89                                    | 1.34                                 |
| R10    | 2.30    | 2.25                                    | 1.93                                 |
| R12    | 2.25    | 2.47                                    | 2.38                                 |
| R15    | 2.18    | 2.77                                    | 3.10                                 |
| R6S2   | 2.52    | 1.72                                    | 1.03                                 |
| R6S4   | 2.55    | 1.79                                    | 1.10                                 |
| R6S6   | 2.67    | 1.95                                    | 1.25                                 |
| R6S8   | 2.80    | 2.03                                    | 1.29                                 |
| R4S10  | 2.92    | 1.93                                    | 1.12                                 |
| R6S10  | 2.90    | 2.11                                    | 1.35                                 |
| R10S10 | 2.78    | 2.80                                    | 2.48                                 |

**Young's modulus (E), Fracture toughness (K<sub>IC</sub>), Fracture energy (G<sub>IC</sub>), Silica (S), Rubber (R)**

Fracture toughness and Fracture energy

Fracture test setup and the CT sample and based on ASTM D5045-99. A sharp crack of length a between 0.45W and 0.55W was introduced by tapping a fresh razor blade at the notch tip. Fracture energy obtains by two ways; by load–displacement curve and the formula down:

$$G_{IC} = \frac{K_{IC}^2}{E} (1 - \nu^2) \tag{6}$$

Where E is the tensile Young's modulus of the composite and  $\nu$  is the Poison coefficient of the epoxy which is considered to be equal to 0.35.

The results of these two parameters are showed in table 1. As is specified in the table, the best value of fracture toughness was obtained with 10 wt% for each nanoparticles that according to the pure epoxy, along with the equivalent of 194% growth. Also for fracture energy, the maximum value was showed in 15 wt% rubber and improved 101%.

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## VI. MODELLING THE EXPERIMENTAL RESULTS

As noted in part two, the method of Response Surface has been used for model Young's modulus, Fracture toughness and fracture energy. Based on the results of the simulation, the first order response surface for young's modulus and the second order response surface for two others parameters had suitable accuracy.

In all similar, 22 samples was assigned for train (70%) and 7 samples for test. Selection of samples for train and test was random. The formula obtained from 1-order response surface method based on information education for young's modulus is as follows:

$$E(X_1, X_2) = 2.7583 - 0.0444X_1 + 0.0363X_2 \quad (7)$$

$X_1$  represents particle weight fraction of rubber and  $X_2$  represents particle weight fraction of silica. In this case, in the lack of presence for each nanoparticles, zero number are assign to percent. In the figures (1) and (2) the results of the model and experimental results has been brought. Also there are error figures for train and test samples.

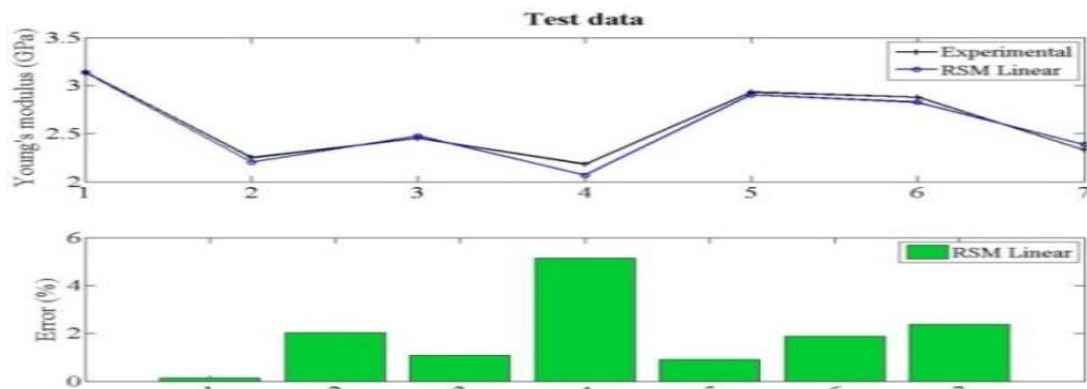


Fig. 1. Compare the result of model and experiment for Young's modulus with different samples by linear RSM for test and error

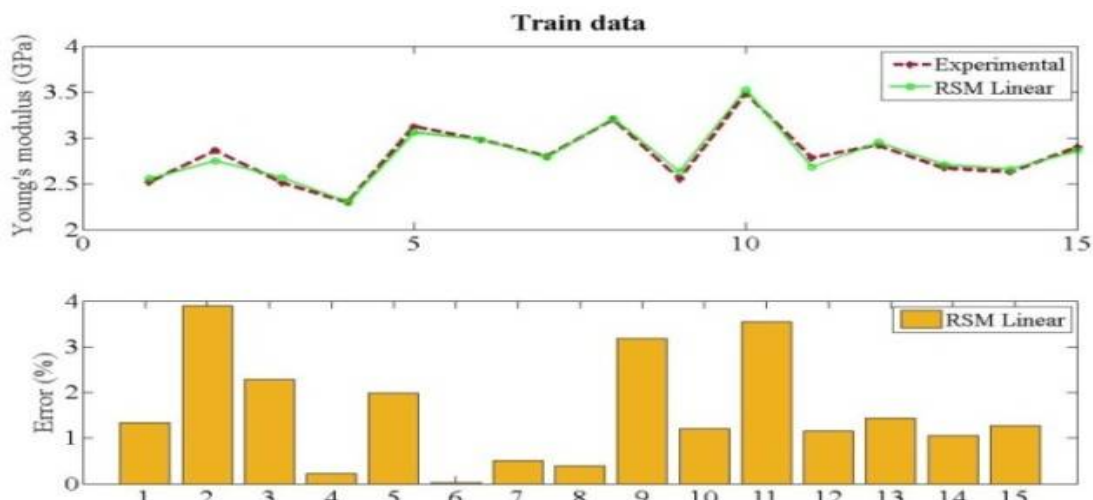


Fig. 2. Compare the result of model and experiment for Young's modulus with different samples by linear RSM for train and error

The equations obtained for fracture toughness from the first and second order response surface method under these conditions are as follows:

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$$K_{IC}(X_1, X_2) = 0.8748 + 0.1256X_1 + 0.0623X_2 \quad (8)$$

$$K_{IC}(X_1, X_2) = 0.8907 + 0.1007X_1 + 0.0665X_2 + 0.002X_1^2 - 0.0003X_2^2 \quad (9)$$

As same as Young's modulus's formula,  $X_1$  represents particle weight fraction of rubber and  $X_2$  represents particle weight fraction of silica. On the action of size of nanoparticles in the second order considered to be fitted positive. Although power two of both variables had negative effect on the fracture toughness and decreased it. At last the second order on the basis of higher precision selected and the value obtained by the model, the real value and the relative error in percent has been brought in figures (3), (4), (5) and (6).

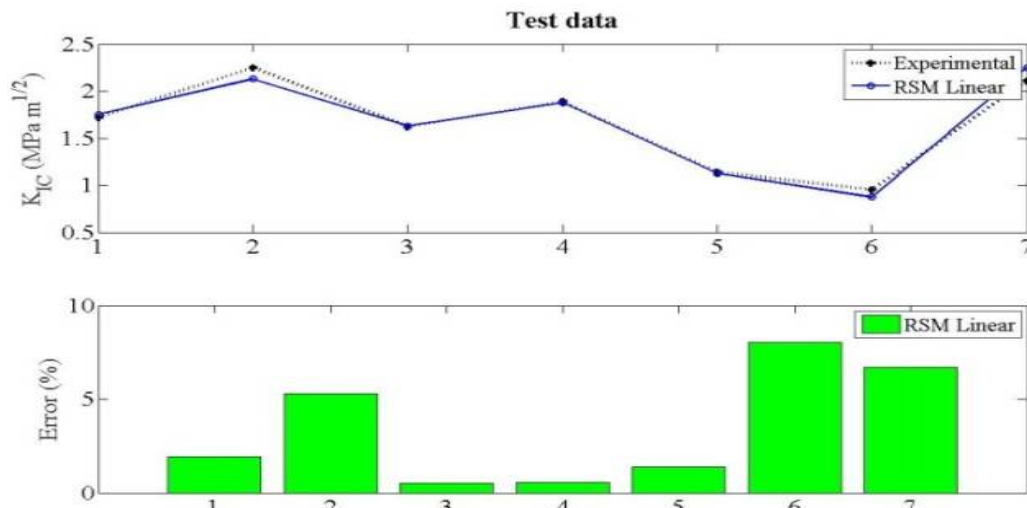


Fig. 3. Compare the result of model and experiment for fracture toughness with different samples by linear RSM for test and error

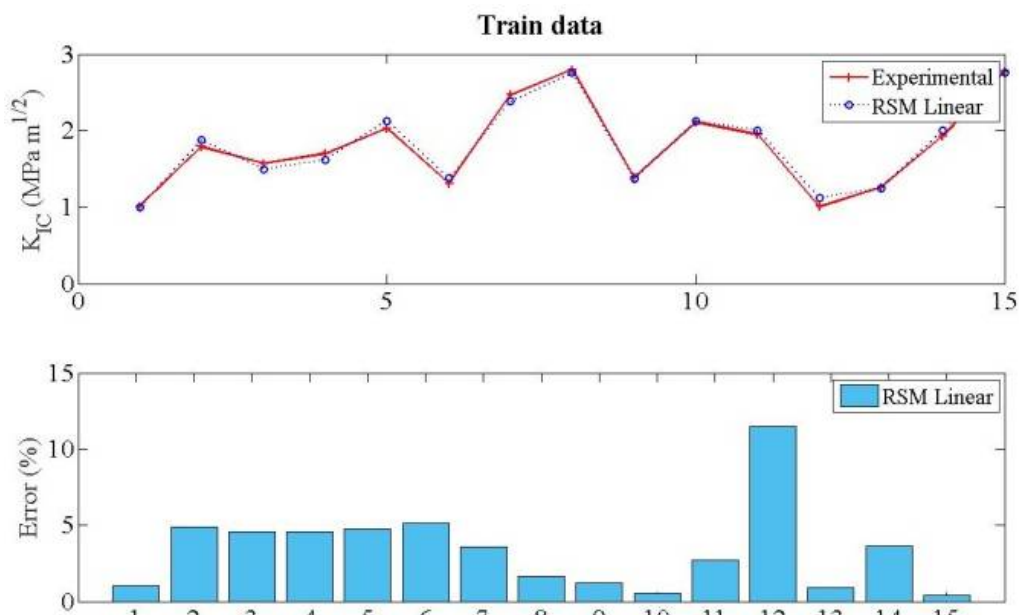


Fig. 4. Compare the result of model and experiment for fracture toughness with different samples by linear RSM for train and error

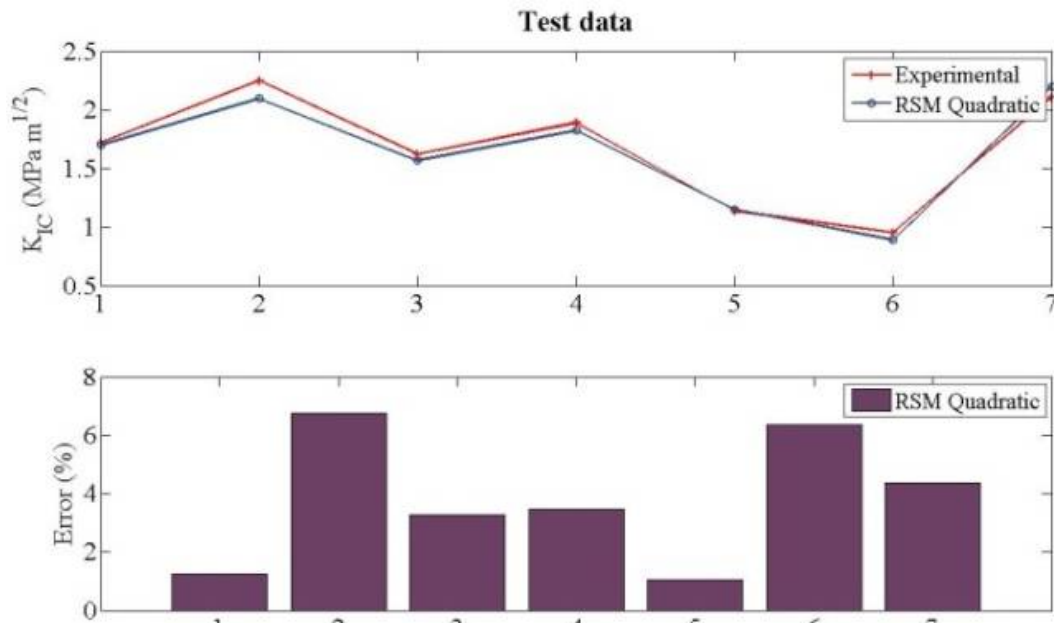


Fig. 5. Compare the result of model and experiment for fracture toughness with different samples by tow order RSM for test and error

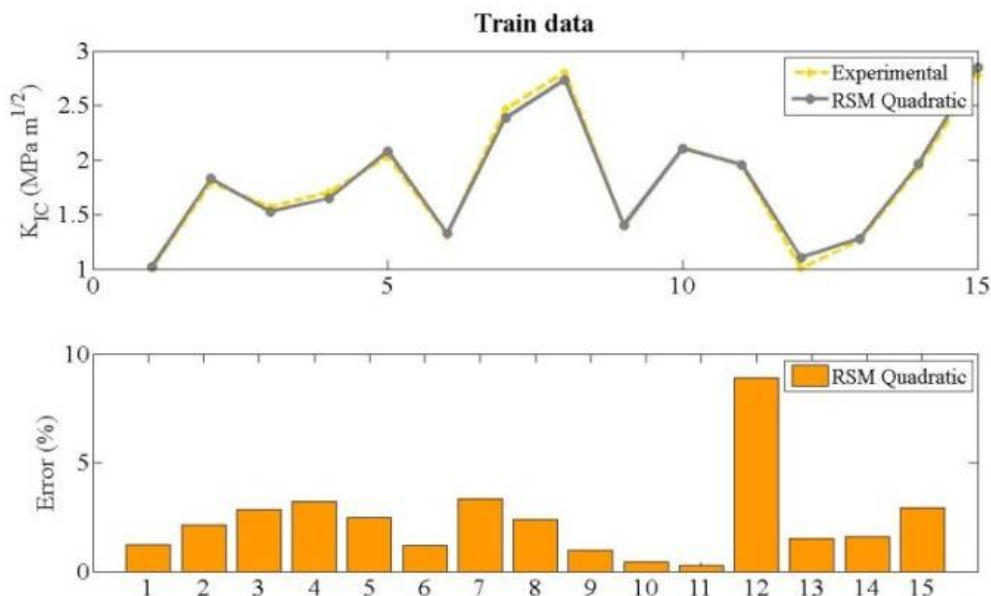


Fig. 6. Compare the result of model and experiment for fracture toughness with different samples by tow order RSM for train and error

Based on the formula 8 and 9, clearly specify that the effect of two nanoparticles on fracture toughness is positive. with this difference that the effect rubber is almost double the impact of silica. Because the coefficient of weight fraction rubber is nearly double the weight fraction silica in the formula 8. Because it is that in some cases the error was too high, two order response surface method used to raise the accuracy. The answers showed in formula 9. Can be easily compared with the formula coefficients of fracture toughness, understand the fixed number coefficients  $X_1$  and  $X_2$  in two formula resembles a lot to each other that represent the linear tendency of two nanoparticles. But about power two of nanoparticles, although the effect of power two of these two nanoparticles is bit in response but the effect of power

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two of rubber is positive and it is negative for silica. In can be concluded that the impact of two nanoparticles is positive; recalling that the impact of rubber is much more powerful that silica. The proposed model compared to the previous state from the perspective of error, little suffered a drop and we set in the train and test arrived to 9% error that with regard to the engineering system, it's a suitable error.

One of the reasons for rise of error is less than the value of fracture toughness compared to the Young's modulus. That error in the calculation of the percentage, the more the value of the parameter is to know less, with a slight change it leads to error on the percentage will be higher.

The equations obtained through the first and second order response surface method are as follows for the fracture energy.

$$G_{IC}(X_1, X_2) = 0.1767 + 0.1656X_1 + 0.0455X_2 \quad (10)$$

$$G_{IC}(X_1, X_2) = 0.2562 + 0.0425X_1 + 0.0359X_2 + 0.0117X_1^2 + 0.0004X_2^2 + 0.0019X_1X_2 \quad (11)$$

Like before,  $X_1$  particle weight fraction of rubber and  $X_2$  represents particle weight fraction of silica.

In the case of fracture energy the situation is quite different. By comparing the response surface of linear and two order we can see that the ratio of the coefficient are different.

The principle of this difference shows yours in power two rubber in formula 11.

In addition to that, fracture energy is only quale that multiplied by two nanoparticles has shown as same as a number. The two times can be observed that the effect of power two silica is very small and indicates that the nanoparticle silica also tend to retain its linear being in fracture energy. With this description, it is evident that the linear model has a high error that we have been exhibited in figures (7), (8), (9) and (10). In the case of power two, there is much more suitable adaptation and of course the error is also less.

The greatest error in results of test is 14% and it is less that 14% for train that represent the relative accuracy of two order model.

According to the results achieved can be found it is argued that both nanoparticles have a positive impact on fracture energy. But clearly that coefficient of rubber in both models is stronger than silica nanoparticles.

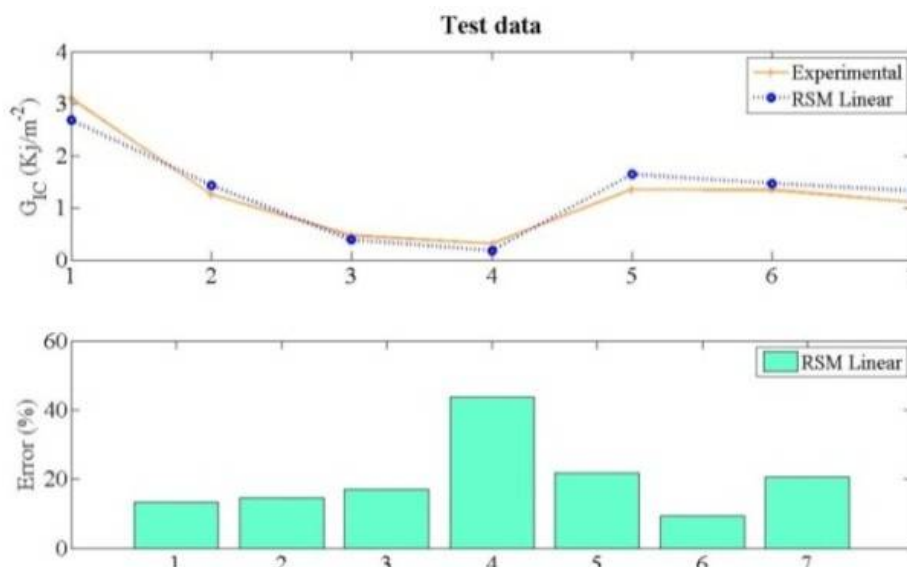


Fig. 7. Compare the result of model and experiment for fracture energy with different samples by linear RSM for test and error



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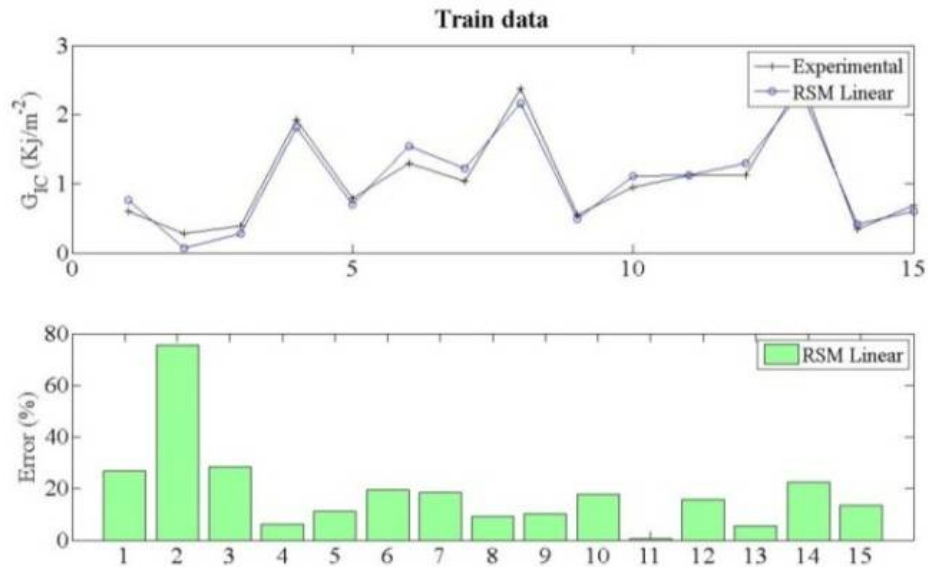


Fig. 8. Compare the result of model and experiment for fracture energy with different samples by linear RSM for train and error

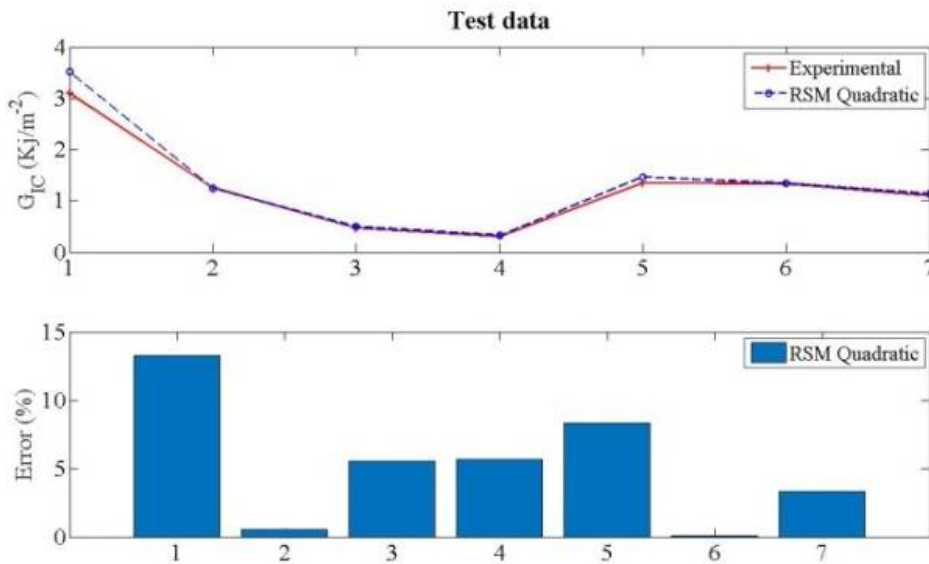


Fig. 9. Compare the result of model and experiment for fracture energy with different samples by tow order RSM for test and error

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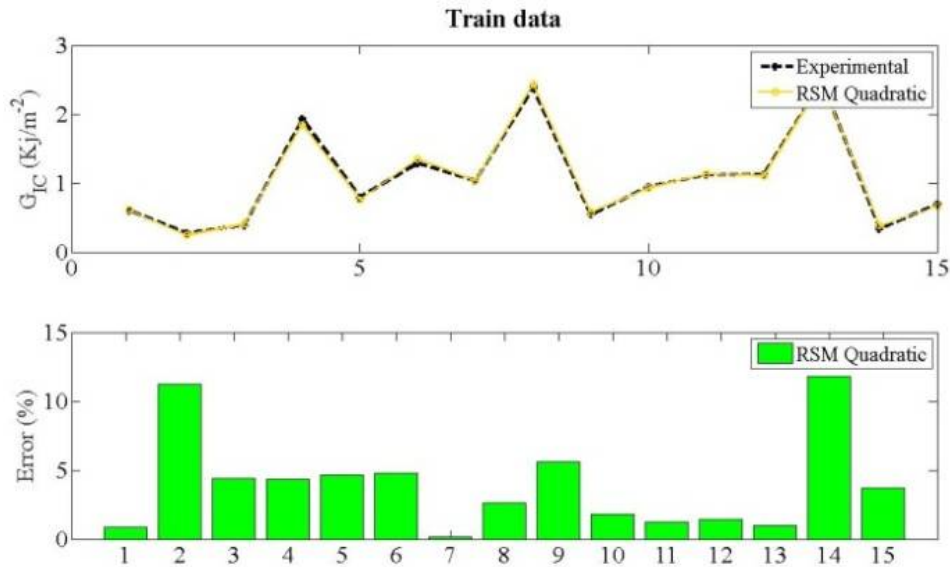


Fig. 10. Compare the result of model and experiment for fracture energy with different samples by low order RSM for train and error

In the final step to complete the process, a process of optimization using by genetic algorithm was used on the models. The algorithm is binary type and for each 10-bit variable were assigned. In this way the number of bits of each chromosome is 20 bits. The basic starting points have been chosen randomly. (The choice of the starting points of a regular at this stage to completely clear the system decreases accuracy). The number of points of overlap with respect to the length of the chromosome 20 bit 5 points and the possibility of mutations against the 1.0 is assumed. Finally for the models and assuming the maximum three parameters, the following answers were obtained: The maximum value for the Young's modulus is 3.477 GPa without rubber nanoparticle and 19.8 wt% silica. For fracture toughness, 15 wt% rubber and 20 wt% are the best content; the maximum value is 4.061 MPa.m<sup>1/2</sup>. The maximum value for the fracture energy is 4.974 Kj/m<sup>2</sup> where rubber has 15 wt% and silica has 20 wt%. The results of the form referenced in figures (11), (12) and (13) are displayed.

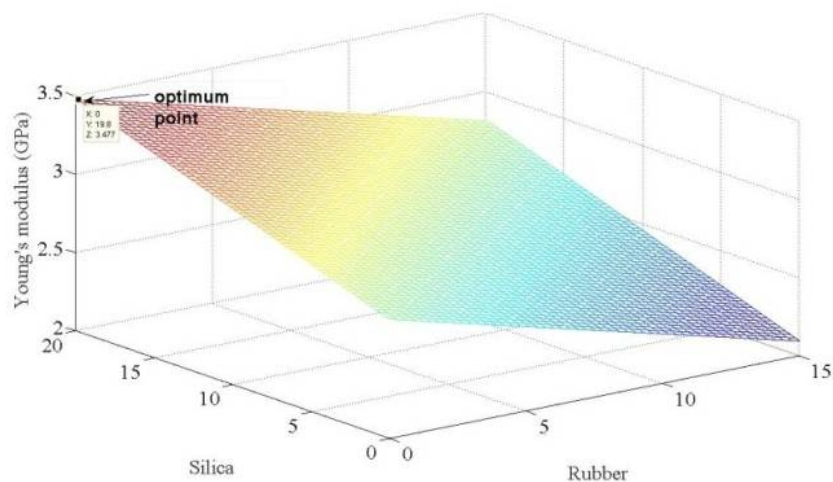


Fig. 11. Three- dimensional graph of Young's modulus in terms of different content percent rubber & silica

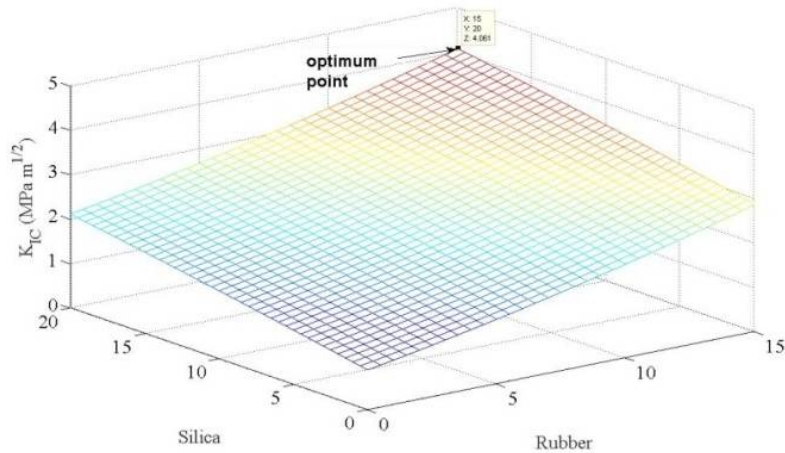


Fig. 12. Three-dimensional graph of fracture toughness in terms of different content percent rubber & silica

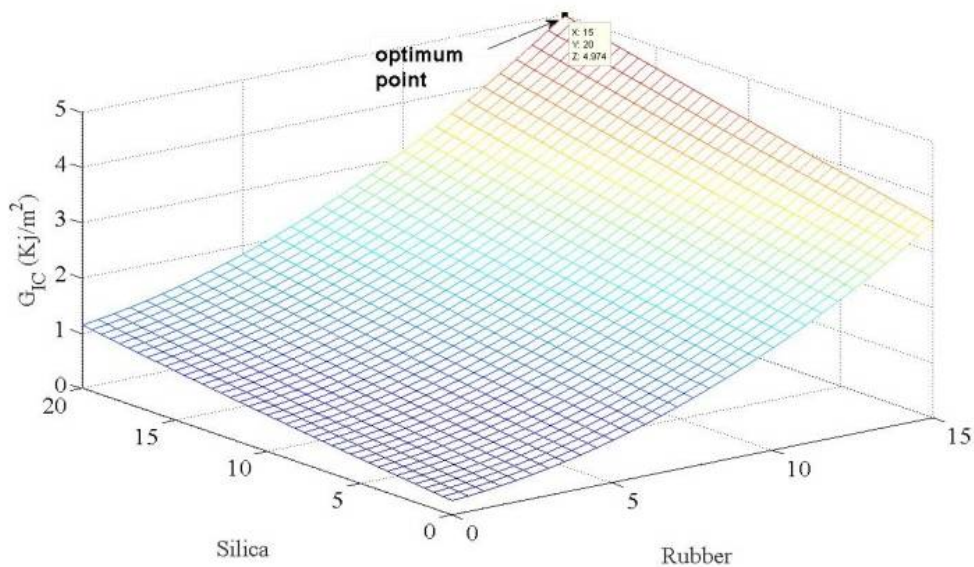


Fig. 13. Three-dimensional graph of fracture energy in terms of different content percent rubber & silica

## VII. RESULT

In this paper, a numerical study has taken place on Young's modulus (E), fracture toughness (K<sub>IC</sub>) and fracture energy (G<sub>IC</sub>) of epoxy reinforced rubber and silica nanoparticles with different contents percent. According to the experimental results, add the amount of Nano-particle depending on the nanoparticles might increase or decrease the mechanical properties. The size of the nanoparticles, along with the type of the input parameter as nanoparticles for modeling response surface method in the results intended. The results of modeling the value decrease or increase depending on the Nano-particle and the impact of the two interactions concomitant use of nanoparticles in each property we fitted logo. Finally the results of the modeling level response manner, with 70% of the samples for training and the rest for the compliance testing of the super well with laboratory results on the mechanical properties of the three was discussed.

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