Modelling the electromechanical sensitivity of silicone composites using response surface methodology

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Abstract. Dielectric elastomers are used for sensor and actuator applications. These materials convert voltage applied directly into strain through Maxwell stress. An important parameter in the determination of voltage induced strains is electromechanical sensitivity. This paper presents an approach to establish the model for predicting the electromechanical sensitivity of silicone dielectric elastomer composites using the response surface methodology. Two-factor and two-level, face-centered composite design is used for experimentation. The parameters which affect the electromechanical sensitivity are selected as superconducting carbon black (SCCB) and barium titanate (BT). Response surface methodology is used to derive second-order quadratic model with interactions. Investigation showed that SCCB has more significant effect than parameter BT, on controlling the electromechanical sensitivity of these composites. Maximum electromechanical sensitivity for these composites was found at around ≤ 4 parts per hundred weight fractions for both the parameters.

1. Introduction

Dielectric elastomers find applications as sensors and actuators[1,2]. Electromechanical Sensitivity (β) is regarded as an important value to evaluate deformation induced through voltage. It is evaluated as the ratio of permittivity to elastic modulus ‘as in equation (1)’.

$$\beta = \text{permittivity} \ (\text{elastic modulus})^{-1}$$

Researchers strive to achieve high actuation performance in a low electric field[3–6]. Using novel elastomers that offer high electromechanical sensitivity, a given deflection can be achieved at lower voltages. This can be achieved with higher permittivity at lower elastic modulus.

Researchers have investigated various composites for improving the voltage induced strain values through permittivity improvement of the composites. Dielectric fillers such as barium titanate[7], TiO₂[8], have been added to PDMS matrix[9] materials. Conductive fillers such as carbon black[10], CNT, Al₂O₃[11] have been investigated for their role in improving the actuation performances. Researchers have also investigated combined use of dielectric and conductive fillers towards achieving improvements in actuation strains[12].

Most of the studies on use of dielectric elastomers for actuation applications has been concentrated on room temperature vulcanised PDMS[13], while few studies have been undertaken on high
temperature vulcanised (HTV) silicone rubber[14]. Solid silicone rubber offers advantages such as environmental stability and matured industrial production techniques[15].

The experimental design involving a number of parameters requires a large number of experiments to be performed[16]. The use of response surface methodology for evaluating & modelling the responses of dielectric elastomers have not been observed in the literature.

This study investigates the combined influence of barium titanate and superconducting carbon black on the electromechanical sensitivity of solid silicone rubber composites. Two-factor and two-level, face-centred composite design is used for experimentation. Superconducting carbon black (SCCB) and barium titanate (BT) are selected as parameters that affect the electromechanical sensitivity. A mathematical model is developed using response surface methodology in order to predict the electromechanical sensitivity of HTV Silicone rubber composites. Electromechanical sensitivity is expressed in terms of weight fractions of SCCB & BT. The main effects and interaction effects of these parameters on the electromechanical sensitivity is discussed. Regression model for the same is built using MINITAB 17 software.

2. Materials and Methods
HTV silicone dielectric elastomer composites were fabricated via compression moulding process. Roll mill was used to disperse the fillers and mix them thoroughly. Solid silicone rubber matrix used was 40 Shore A, commercially available HTV silicone rubber from DJ Silicones. Barium titanate of 3μm was sourced from Sigma-Aldrich, while superconducting carbon black used was Ketjenblack 300 J that was sourced from AkzoNobel. Dicumyl peroxide of 98% purity (DCP) was used as curing agent.

Once the solid silicone rubber was thoroughly masticated in the roll mill, BT and SCCB were added in proportions of parts per hundred parts of rubber (phr) as shown in table 1. Then 1 phr of DCP was added to the roll mill while mixing for 30 mins. The mixture is laid out as a precured sheet. Using appropriate dies as required for permittivity and elastic modulus measurement standards, solid silicone rubber composite samples are obtained by compression moulding at 160°C.

The specimens are tested as per ASTM D150-11, standard test methods for AC loss characteristics and permittivity (dielectric constant) of solid electrical insulation, to obtain the permittivity of the specimens at 1kHz frequency using the Agilent Precision LCR meter, model E4980A. Agilent 16451B test fixture for the above equipment was used.

In order to obtain the elastic modulus, the specimens were tested as per ASTM D575-91(2012); standard test methods for rubber properties in compression, using test method A. From the stress strain plots obtained, elastic modulus was evaluated at 10% strain.

In the current study, electromechanical sensitivity was selected as the response for modelling the performance of HTV silicone dielectric elastomer. Weight fractions of SCCB & BT are controlled parameters. Effects of curing agent, compression pressure during moulding, curing temperature, mixing time on the response were neglected after screening tests were conducted. Table 1 shows the parameter and their levels.

The experimental design was based on two-level, two factor, central composite design as is shown in table 2.

| Parameters and their levels |
|----------------------------|
| Independent variables | Coded values |
| Weight fraction of SCCB, phr | 3.5 7.75 12 |
| Weight fraction of BT, phr | 3.5 7.75 12 |

3. Results and Discussions
Electromechanical sensitivity is calculated from the values of permittivity and elastic modulus. These values of the electromechanical sensitivity were utilized to build a mathematical model using response surface methodology. Second-order polynomial was used to model electromechanical sensitivity as a function of weight fractions of SCCB & BT. Quadratic model with the square and interactive terms
are utilised. The regression coefficients for the polynomial were determined using MINITAB software. The analysis was done using uncoded units.

### Table 2. Experimental design in uncoded units

| BT, phr | SCCB, phr | β (10^{-3}) (kPa)^4 |
|---------|-----------|----------------------|
| 7.75    | 7.75      | 1.12                 |
| 12      | 12        | 0.93                 |
| 3.5     | 7.75      | 1.40                 |
| 3.5     | 3.5       | 1.79                 |
| 12      | 7.75      | 1.16                 |
| 12      | 3.5       | 1.79                 |
| 7.75    | 12        | 1.05                 |
| 7.75    | 3.5       | 1.10                 |
| 3.5     | 12        | 1.16                 |

It is seen that electromechanical sensitivity varies from 0.93 (10^{-3}) (kPa)^{-1} to 1.79 (10^{-3}) (kPa)^{-1} for the given range of filler loadings.

### Table 3. Estimated regression coefficients (coded units) for electromechanical sensitivity

| Terms                  | Regression Coefficients (coded units) | SE Coefficients | T      | P      |
|------------------------|--------------------------------------|-----------------|--------|--------|
| Constant               | 1.0828                               | 0.0700          | 15.46  | 0.000  |
| SCCB phr               | -0.2500                              | 0.0689          | -3.63  | 0.008  |
| BT phr                 | -0.0833                              | 0.0689          | -1.21  | 0.265  |
| SCCB phr x SCCB phr    | 0.060                                | 0.101           | 0.59   | 0.571  |
| BT phr x BT phr        | 0.260                                | 0.101           | 2.57   | 0.037  |
| SCCB phr x BT phr      | -0.0750                              | 0.0843          | -0.89  | 0.403  |

SE- Standard Error, T- Student T test, P-Probability.

From table 3, it is observed that P value is low for superconducting carbon black filler, thus it would be contributing significantly to the model. Dielectric filler’s contribution would be insignificant to the electromechanical sensitivity response.

### Table 4. Analysis of variance for electromechanical sensitivity

| Source                | DF | Adj SS (10^{-1}) | Adj MS (10^{-1}) | F-Value | P-Value |
|-----------------------|----|------------------|------------------|---------|---------|
| Regression            | 5  | 7.08583          | 1.41717          | 4.98    | 0.029   |
| Linear                | 2  | 4.16667          | 2.08333          | 7.32    | 0.019   |
| Square                | 2  | 2.69416          | 1.34708          | 4.74    | 0.050   |
| Interactions          | 1  | 0.22500          | 0.22500          | 0.79    | 0.403   |
| Residual Error        | 7  | 1.99109          | 0.28444          |         |         |
| Lack-of-Fit           | 3  | 1.99109          | 0.66370          |         |         |
| Pure Error            | 4  | 0                | 0                |         |         |
| Total                 | 12 | 9.07692          |                   |         |         |

DF-Degree of Freedom, Adj SS- Adjusted sum of squares, Adj MS- Adjusted mean squares, F-Fischer, P-Probability.
Table 4 shows the analysis of variance results. The significance of regression was evaluated by P and F values. The larger the F value, the better the fit of the regression model with the experimental data. Linear term contributes more significantly than square and interaction terms.

For solid silicone rubber dielectric composites, the electromechanical sensitivity is expressed in terms of superconducting carbon weight fractions & barium titanate weight fractions ‘as in equation (2)’.

Electromechanical sensitivity = 2.437 - 0.1999 BT - 0.0640 SCCB + 0.01334 BT *BT + 0.00188 SCCB *SCCB - 0.00324 BT *SCCB

(2)

![Residual Plots for Electromechanical Sensitivity](image)

**Figure 1.** Residual Plots for electromechanical sensitivity

From the Figure 1, it is observed that the data is not skewed. The residuals are randomly distributed, with no recognizable patterns seen in the residual plots.
Figure 2. Main effects plot for electromechanical sensitivity

Main effects plot as shown in Figure 2, indicates the significance of the factors on electromechanical sensitivity of solid silicone rubber composites. It initially decreases with increase in dielectric filler loading till 7.75 phr. Beyond 7.75 phr, electromechanical sensitivity increases with dielectric filler loading till 12 phr. Electromechanical sensitivity however continues to decrease with increasing conductive filler loading.

Interactions among the conductive and dielectric behaviours is evident from Figure 3. For dielectric filler loadings at 3.5 phr and 12 phr, decrease of conductive filler loading improves electromechanical sensitivity. However, with dielectric filler loading at 7.75 phr, electromechanical sensitivity first registers an increase and then a decrease with increasing conductive filler loadings.
Figure 4 shows the contour plot of the electromechanical sensitivity of the composites. The maximum electromechanical sensitivity for these composites lies in the region of less than 4 phr weight fractions of BT & SCCB and less than 4 phr SCCB & greater than 11.8 phr BT.

4. Conclusions
From this study it is seen that response surface methodology was used to create a mathematical model for the electromechanical sensitivity of solid silicone rubber composites filled with barium titanate and superconducting carbon black. Model shows that electromechanical sensitivity is affected by the SCCB weight fractions and the square term involving BT weight fractions. The maximum electromechanical sensitivity was found to be in the range of around 4 phr for both fillers.

5. References
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