Optimisation of Pre-structuring Process of Magnetorheological Elastomer Performance

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Abstract. In this study, MREs based on natural rubber and carbonyl iron were prepared. The Taguchi method was utilised to study the effect of a number of dominating factors during the fabrication process, namely, pre-curing time, pre-curing temperature and applied magnetic field on the microstructure and tensile properties of MREs. Tensile properties were measured with a universal tensile testing machine. It was found that the pre-curing temperature had the greatest influence on ultimate tensile strength and elongation at break. Morphology characteristics of MREs were examined using scanning electron microscopy. The results revealed that isotropic MREs cured without an applied magnetic field had a homogenous carbonyl iron particle distribution in the rubber matrix, and that applying a magnetic field at elevated temperature allowed the particles to organise into chain-like columnar structure in anisotropic MREs.

1. Introduction

Rubber is one of the most commonly used materials for energy absorption and vibration suppression. However, rubber itself has low stiffness [1]. Many efforts have been put into modifying available rubber to improve its mechanical properties, such as inclusion of particle or fibre fillers [2-4]. In more recent research, magnetic particles have been included in rubber to create a magnetic rubber composite known as magnetorheological elastomers (MREs). MREs can be classified into two types: isotropic and anisotropic. Isotropic MREs have a uniform dispersion of magnetic particles in the rubber matrix. In anisotropic MREs, the particles form columnar structures, achievable by applying a magnetic field during curing of the matrix [5-8]. The addition of magnetic particles in a rubbery matrix allows more damping through magnetic interactions between neighbouring particles and particles-matrix interfacial damping. Moreover, MREs’ modulus and stiffness can be changed during fabrication or in service by applying an external magnetic field. Most commonly used matrices for MREs include natural rubber, silicone rubber, polybutadiene, polyisobutylene, polyisoprene, and polyurethane rubber [5,9-13]. The magnetic particle choice for MREs is carbonyl iron [5-8].

The curing condition plays an important role in MRE performance. Pre-curing stage is essential for formation of magnetic particle alignment in the rubber matrix. In this process the magnetic particles are driven by the magnetic force and chain-like structures are formed in the rubber matrix. As far as the authors are aware, there is limited published work that study factors effecting the pre-structuring process. This work aims to study the effects of three predominant factors during pre-structure process on MREs, namely pre-curing time, pre-curing temperature and applied magnetic field [6,10] on the tensile strength...
of MREs. In this study, the Taguchi method is utilised to determine the optimum processing condition for MREs.

2. Taguchi Method

The Taguchi method, developed by Dr. Genichi Taguchi, proposes a simplified, standardised, consistent and cost-effective approach in design of experiments to study the influence of multiple factors on a product or a process.

In general, the Taguchi method includes the following steps: identification of the factors and their respective levels, design of experiments using an appropriate orthogonal array (OA), conducting of experiments, analysis of data and selection of the optimum level for each factor, confirmatory tests of the optimum condition.

The key steps in the Taguchi method are to identify the factors and levels, and to assign them to the appropriate OA. The Taguchi method allows for 18 standard orthogonal arrays, as mentioned in reference [14]. After the conditions for individual experiments, called trials, are specified using the OA, the trials are carried out, preferably in random order [14]. The S/N ratio for larger is better is then calculated for each factor, using the following equations:

\[
\frac{S}{N} = -10 \log \left( \frac{1}{n} \sum_{i=0}^{n} \left( \frac{1}{Y_{ai}} \right)^2 \right)
\]

in which \(Y_{ai}\) is the value of factor \(A\) at level \(a\) at the \(i^{th}\) observation and \(n\) is the number of observations for factor \(A\) at level \(a\). Another advantage of the S/N ratio is the determination of the trend of influence of the factors by calculating the average influence of a factor and plotting it against the corresponding level. The average factor effect is given by equation (2):

\[
A_F = \frac{\sum_{i=0}^{n} \left( \frac{S}{N} \right)_{i}}{n}
\]

in which \(A_F\) is the mean of the S/N ratios for factor \(F\) at level \(i\), \((S/N)_i\) represents the S/N ratio and \(n\) the number of observations for factor \(F\) at level \(i\). Moreover, the significance of each processing parameter is the difference between the maximum and minimum S/N ratio:

\[
\text{Significance of factor} = \left( \frac{S}{N} \right)_{\text{max}} - \left( \frac{S}{N} \right)_{\text{min}}
\]

\(S/N_{\text{max}}\) and \(S/N_{\text{min}}\) represents the maximum and minimum average S/N ratios, respectively, for a factor.

By using the Taguchi method, it is also possible to predict the optimum condition for the process. The optimum level for each factor is identified as the level at which the S/N ratio has the maximum value. However, the Taguchi method does not provide other statistically vital information, which can instead be obtained by performing an analysis of variance (ANOVA). Using ANOVA in the Taguchi method allows determination of sums of squares, degrees of freedom, percentage confidence level, and percentage contribution. Equations to calculate ANOVA parameters are discussed in reference [11].

3. Experimental

3.1 Materials

Compounding formulation used in this study is shown in Table 1. Natural rubber and all other chemicals in Table 1 were supplied by Zarm Scientific & Supplies Sdn. Bhd, Malaysia. Carbonyl iron particles (CIP) were purchased from Shanghai YY International Co, Ltd, China, and have a spherical shape and an average particle size of 4µm.
3.2 Design of experiments

In this study, three factors were selected to study: pre-curing time (t), pre-curing temperature (T) and applied magnetic field during pre-curing and curing (M), each with four levels as presented in Table 2. The smallest appropriate OA for three factors and four levels, L’16, was selected for the Taguchi method. The experimental and sample preparation was carried out according to L’16 OA as reported elsewhere [14]. The analysis of S/N ratios was subsequently used to evaluate the results. In this work, larger-is-better target for S/N ratios was chosen. ANOVA was performed with Statistica software to evaluate the percentage contribution of each factor.

Table 1. Compounding formulation

| Materials                  | Functions     | Loadings (phr)* |
|----------------------------|---------------|-----------------|
| Natural rubber (SMR L)     | Matrix        | 100             |
| Zinc oxide                 | Activator     | 5               |
| Stearic acid               | Activator     | 1               |
| Paraffin oil               | Plasticizer   | 5               |
| Carbonyl iron powder       | Magnetic particles | 70          |
| IPPD                       | Antioxidant   | 2               |
| CBS                        | Accelerator   | 2               |
| TMTD                       | Accelerator   | 1               |
| Sulphur                    | Crosslinking agent | 1.5         |

*part per hundred rubber

Table 2. Experimental factors and their levels

| Factors                      | Unit | Level 1 | Level 2 | Level 3 | Level 4 |
|------------------------------|------|---------|---------|---------|---------|
| Pre-curing time (t)          | min  | 3       | 5       | 10      | 15      |
| Pre-curing temperature (T)   | °C   | 60      | 80      | 100     | 110     |
| Magnetic field (M)           | mT   | 0       | 100     | 150     | 165     |

3.3 Fabrication of MREs

The ingredients of the formulation in according to the OA and were compounded using a conventional laboratory two-roll mill according to ASTM D3184-07. The cure time at 150°C was determined by an Alpha MRD200 rotational die rheometer according to D6204 - 15. Compounded rubber samples were placed in a 120mm × 65mm × 3mm mould and undergo a pre-structure process with their respective pre-curing parameters in Table 3. Isotropic samples were cured in a compression moulder at 150°C under a pressure of 10MPa. Anisotropic MREs were cured in a specially developed magnetic mould as shown in Figure 1. Each trial was repeated three times.

3.4 Characterisation

3.4.1 Morphology. The microstructures of MREs were observed using a Hitachi TM3000 scanning electron microscope. The surfaces were coated with a thin layer of gold prior to observation at accelerating voltage of 15kV.

3.4.2 Tensile test. Tensile test was carried out on dumbbell-shaped samples using an Instron universal tensile testing machine at a crosshead speed of 500mm/min according to ASTM D412-16.

![Figure 1. Schematic diagram of pre-structure process](image)
4. Results and discussion

4.1 Morphology

Figure 2 shows SEM images of isotropic and anisotropic MRE samples. From the SEM images, it can be seen that isotropic MREs have a uniform distribution and dispersion of CIP with no obvious aggregation in the rubber matrix. For anisotropic MREs, clearly, as expected, the application of a magnetic field at elevated temperature allowed the CIP to form chain-like structures.

![Figure 2. SEM images of (a) isotropic MRE and (b) anisotropic MRE](image)

4.2 Tensile testing

Table 3 shows the values of tensile strength and elongation at break, which were subsequently used to calculate the S/N ratios and ANOVA, each value representing an average from five samples. The highest value for each measurement is shown in bold.

| Sample | Tensile strength (MPa) | Elongation at break (%) |
|--------|------------------------|-------------------------|
| 1      | 12.60                  | 808.89                  |
| 2      | 10.37                  | 642.22                  |
| 3      | 12.53                  | 710.56                  |
| 4      | 11.02                  | 619.44                  |
| 5      | 2.44                   | 285.56                  |
| 6      | 12.03                  | 752.22                  |
| 7      | 12.56                  | 710.56                  |
| 8      | 11.10                  | 643.89                  |
| 9      | 2.95                   | 345.56                  |
| 10     | 8.96                   | 643.33                  |
| 11     | 11.09                  | 766.11                  |
| 12     | 9.13                   | 582.78                  |
| 13     | 12.54                  | 742.22                  |
| 14     | 11.16                  | 623.89                  |
| 15     | 10.45                  | 681.11                  |
| 16     | **12.92**              | 726.11                  |

Figure 3 shows the main effect plots of the S/N ratios for the ultimate tensile strength. The highest S/N ratios are observed at 15 min, 100°C and 0mT magnetic field during pre-curing. Table 4 shows ANOVA results for the ultimate tensile strength. It can be seen that the variability for each factor was tested at 99.9% confidence level. From the parameter significance, it can be seen that the most influential factor to form alignment of magnetic particles in the rubber matrix is temperature, followed by magnetic field and time. However, the percentage contribution is approximately the same for each factor. The minimal influence of all tested parameters on percentage contribution would appear to be due to tensile mode of loading during testing. This suggests the interaction between magnetic particles alignment is less efficient in a tensile mode, which is not surprising, given that in tension, it is largely only increasing the spacing between chains and the spacing within chain and breakdown of magnetic
particles interaction between the neighboring particles is limited (see Figure 4), suggesting formation of magnetic particle alignment is less important in determining the ultimate tensile strength of MREs.

![Figure 3](image)

**Figure 3.** Main effect plots for S/N ratios of ultimate tensile strength: (a) effect of pre-curing time, (b) effect of pre-curing temperature and (c) effect of magnetic field during curing

| Table 4. ANOVA Table and factor influence for ultimate tensile strength |
|--------------------------|--------------------------|--------------------------|--------------------------|--------------------------|
| Factor                  | Sum of Squares           | Degree of freedom        | Confidence Level (%)     | Percentage Contribution (%) |
| Time (min)              | 115.522                  | 3                        | 99.99                    | 21.31                     |
| Temperature (°C)        | 115.222                  | 3                        | 99.99                    | 21.26                     |
| Magnetic field (mT)     | 119.963                  | 3                        | 99.99                    | 22.13                     |

![Figure 4](image)

**Figure 4.** Influence of loading types on particle separation in tensile mode[11]

The main effect plots of the S/N ratios for elongation at break are presented in Figure 5. The highest S/N ratios are observed at 15 min, 100°C and 0 mT magnetic field during pre-curing. It is also noted that the suggested optimum conditions to obtain highest elongation at break are similar to those for the optimum conditions when assessing the ultimate tensile strength.

![Figure 5](image)

**Figure 5.** Main effect plots for S/N ratios of elongation at break: (a) effect of pre-curing time, (b) effect of pre-curing temperature and (c) effect of magnetic field during curing
Table 5. ANOVA Table and factor influence for elongation at break

| Factor               | Sum of Squares | Degree of Freedom | Confidence Level (%) | Percentage Contribution (%) | Parameter Significance (S/N_{max}−S/N_{min}) |
|----------------------|----------------|-------------------|----------------------|-----------------------------|---------------------------------------------|
| Time (min)           | 128579.265     | 3                 | 99.99                | 12.97                       | 0.648                                       |
| Temperature (°C)     | 185832.806     | 3                 | 99.99                | 18.74                       | 3.333                                       |
| Magnetic field (mT)  | 343874.089     | 3                 | 99.99                | 34.68                       | 3.330                                       |

Table 5 shows ANOVA results for the elongation at break. From the parameter significance, it can also be seen that the most influential factor to form alignment of magnetic particles in the rubber matrix is temperature, followed by magnetic field and time. From the levels of contribution, it can be seen that the most contributed factor to elongation at break value is magnetic field followed by temperature (34.68% and 18.74%, respectively). The influence of time was much less at 12.97%. Isotropic MREs was found to have better elongation at break when compared to anisotropic MREs. This can be explained by the better interfacial interaction in isotropic MREs, in which the particles are dispersed uniformly and the rubber chains coiled around the particles, thus allowed them to uncoil and better align along the stress direction. On the other hand, when anisotropic MREs underwent stretching, the force was in a perpendicular direction to the particle chains, and the rubber molecule chains were restrained by the particle chains and ruptured by the applied force. This is in agreement with similar finding reported by Wang et al[15].

5. Conclusion

In this study, MREs were fabricated based on natural rubber and carbonyl iron particles, and experiments were designed using the Taguchi method to evaluate the influence of three pre-curing time, pre-curing temperature and magnetic field during curing on the tensile properties of MREs. SEM images revealed that application of a magnetic field during the pre-structure and curing process produce columnar structures of magnetic particles in the rubber matrix, while samples fabricated without a magnetic field had a random dispersion of particles. Calculation of the S/N ratios for each factor implied that temperature had the greatest influence on both ultimate tensile strength and elongation at break. It was also found that both tensile strength and elongation at break for isotropic MREs are higher than those of anisotropic MREs, which could be due to the tensile loading mode. ANOVA results suggested that particle alignment caused little difference on tensile properties of MREs.

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