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Payne Effect Prediction of Magnetorheological Elastomer by Krauss Model Comparing with Machine Learning

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ABSTRACT

Magnetorheological elastomer (MRE) is having a complex viscoelastic behaviour especially when facing loading conditions such as different magnetic flux density and strain amplitude. Simple regression model such as least square regression and low order polynomial regression is unable to simulate such behaviour since the behaviour consists of linear and nonlinear portion. Hence, a parametric model namely Krauss model able to perform well on such behaviour specifically for viscoelastic materials. Thus, this work aimed to simulate the viscoelastic properties of MRE by applying Krauss model given four fitting parameters for each property where in this work, they are storage modulus and loss modulus, respectively. By following the trend of current research, a few machine learning models were also developed to simulate the behaviour to compare its performance with parametric model. The mentioned machine learning model were adopted including Guassian Process Regression (GPR), Support Vector Regression (SVR), Extreme Learning Machine (ELM) and Backpropagation Neural Network (BPNN). For both type of model structure consists of two inputs which were strain amplitude and magnetic flux density while two outputs which were storage and loss modulus. The results were analysed practically on the accuracy of prediction at linear region and nonlinear region of the viscoelastic properties. As the result, Krauss model exhibits 0.04171MPa of RMSE while among machine learning, ELM produced 0.00185MPa of RMSE for the highest magnetic field at linear region which achieved the smallest error among other machine learning. As the conclusion, it is noted that Krauss model as parametric model is computational burden when dealing with more variables. Meanwhile, machine learning model performance will keep improved when dealing with more variables. This can be one of the reasons on why machine learning kept utilizing in material properties prediction where it is significantly contributes to fasten the new material developments.

Keywords:
Magnetorheological elastomer; machine learning; neural network; viscoelastic behaviour; parametric model

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1. Introduction

Magnetoactive elastomer or Magnetorheological elastomer (MRE) is known as a smart material which its dynamic viscoelastic properties such as storage modulus, loss modulus and loss factor can externally control by applying external magnetic fields [1] .The mixture of matrix-based polymer and magnetic particles allowed this smart rubber to be applicable for various applications such as vibration absorber [2], prosthetic devices [3] and soft robotic [4] . The presence of magnetic field is significantly changing the stiffness of MRE typically observed in nonlinear viscoelastic properties [5]. The decrease in storage modulus and increase in loss modulus in filled elastomer happened at low shear strain amplitude or as the shear strain increases is described as Payne effect phenomenon [6].

This phenomenon occurred usually at low strain amplitude and saturated at large deformation [7]. This Payne effect occurs due to deformation changing within the microstructure of MRE. Variety of the fabrication process such as matrix elasticity, amount of filler content and filler alignment will contribute to the Payne effect [8]. In addition, the Payne effect is significantly increased with the presence of external magnetic field and increment of filler concentration. At first, this filler concentration contributes to stiffness of MRE. Nevertheless, as strain increases, this filler network starts to breakdown, leading to energy dissipation. However, there are a lot of studies have been conducted to improve the microstructure of MRE by reduced the Payne effect such as utilized flower-like cobalt particles [8], introduced graphene-coated iron (G-Fe) [9] and addition of nano-sized maghemite with carbonyl iron [10].

The condition of MRE is stable before the breakdown of the filler network leading the storage modulus linearly aligned with increasing of shear strain amplitude. After the breakdown of the filler, storage modulus is independent of strain amplitude and leads to nonlinear viscoelastic properties. Thus, the viscoelastic properties of MRE can be divided into two parts, linear viscoelastic region (LVE) and nonlinear viscoelastic region (NLVE). As Payne effect significantly occurred in the presence of magnetic fields, different applied magnetic fields have different LVE limits. This means that increasing the applied magnetic field will reduce the LVE limit.

Agirre-Olabide *et al.,* [11] suggested that the limit of LVE region is defined as point where the storage modulus and loss factor deviate 10% from the approximates straight line. This is a common practice by a few studies to determine the linear region of MRE to make sure that the investigation of properties happened in linear region [7,12,13]. The LVE and NLVE region change according to the changing of applied magnetic field contributed to highly nonlinear behaviour. To simulate this nonlinear behaviour, simple regression models such as least square regression and low order polynomial are not capable of being done due to unable to mimic specifically at the NLVE part.

A parametric model known as Krauss model was commonly used to depict viscoelastic behaviour primarily to investigate the Payne effect. A few studies were found to have Krauss model to predict amplitude-dependent Payne effect [14,15]. Walter *et al.,* [6] described that Krauss model considers the dynamical breakup (collapse) of the particle network to be related to a deagglomeration and (re)agglomeration process (also referred to destruction and formation). Furthermore, parameters included inside the Krauss model bring the physical meaning such as m parameter where it represents the sharpness of transition between low strain and high-strain behaviour [16]. In terms of magnetic field, m value is expected to have large value at higher magnetic field due to filler interaction is easily break at this point.

Nowadays, the trends to consider machine learning model in the current research is increase. This is due to the ability of this machine learning can simulate and mimic the pattern and trend of complex and nonlinear material properties. Implementation of machine learning in predicting the MRE properties had been done and shows excellent results [13,17-19]. In addition, easily implement multiple inputs into the model structure without a need to do complex derivations is kind of advantages. Meanwhile, this data-driven based model will generalize enough as more data is applied during the training process. Furthermore, current trends showing that many new materials have applied machine learning model to simulate its nonlinear behaviour [20].

Hence, this study will compare the model performance between Krauss model and machine learning models. Four types of algorithms will be introduced to predict the Payne effect of MRE which are Back-propagation neural network (BP-ANN), Extreme Learning Machine (ELM), Gaussian Process Regression (GPR) and Support Vector Regression (SVR). The model structure is made up of two inputs which are shear strain amplitude and magnetic flux density and storage and loss modulus as output models. The best models that can predict the Payne effect of MRE can be labelled as potential simulation tools that contribute to fasten the development of new MR materials both in material characterization and its applications.

2. Methodology

In this section, the Krauss model parameters used to simulate the Payne effect will be introduced. Furthermore, four machine learning algorithms will be introduced and lastly, the fabrication process of MRE is briefly described as the process is taken from Saharuddin *et al.,* [13].

2.1 Krauss Model Parameters

From Eq. (1) and (2), γ_C, G'_0, G'_∞, G''_∞ and m were the characteristic value of strain amplitude at which the loss modulus reaches its maximum G''_m, storage modulus for small strain amplitude, asymptotic plateau values for storage modulus at large strain amplitude, asymptotic values for loss modulus at large strain amplitude and non-negative phenomenological exponent, respectively.

$$G'(\gamma) = G'_\infty + \frac{G'_0 - G'_\infty}{1+(\frac{\gamma}{\gamma_C})^{2m}} \tag{1}$$

$$G''(\gamma) = G''_\infty + \frac{2(G''_m - G''_\infty)(\frac{\gamma}{\gamma_C})^{m}}{1+(\frac{\gamma}{\gamma_C})^{2m}} \tag{2}$$

2.2 Machine Learning Algorithm

2.2.1 Support vector regression (SVR)

Support vector machine (SVM) is a well-known method for classification problems in the machine learning algorithm. However, to use for a regression problem, the SVM for the regression problem is introduced as SVR. It concerns reducing the error on a certain degree. It does not care how significant the error is nor falls within an acceptable range. In other words, SVR give the flexibility to define how much error is acceptable in the model and fine an appropriate line, namely hyperplane in a higher dimension, to fit the data. The objective of SVR is to minimize the coefficient vector. The error is handled in a constrains, where the absolute error less than or equal to the specific margin, called maximum error (epsilon, ε). The epsilon can be tuned to the desired accuracy for the model. The objective function and constraints are as follow in Eq. (3) and (4):

$$F = min\ \frac{1}{2}\|w\|^2 \tag{3}$$

$$|y_i - w_i x_i| \leq \varepsilon \tag{4}$$

Where, y_i is the target, w_i is the coefficient and x_i is the predictor. In this work, target refer to storage and loss modulus while predictor refer to shear strain amplitude and magnetic flux density.

2.2.2 Gaussian process regression (GPR)

GPR is non-parametric probabilistic regression model that is based on kernel function. GPR models the distribution over functions directly, assuming any finites set of function values have a joint Gaussian distribution. The kernel functions are crucial to define the functions that can mimic the data. Among the best kernel function including Radial Basis Function (RBF), Matern and Exponential [21]. Training the GPR model involves predicting the hyperparameters of kernel function by applying training data. The general equation for GPR is as follows in Eq. (5):

$$Y(X_i) = f(X_i) + b(X_i) + \varepsilon_i \tag{5}$$

Where, $Y(X_i)$ is observational data (storage and loss modulus) at parametric input X_i where this work refers to shear strain amplitude and magnetic flux density. $f(X_i)$ represent the basis function and ε_i refer to stochastic error during the training process and is assumed to follow a normal distribution with zero mean and variance $V(\varepsilon_i) = \sigma^2$ [21].

2.2.3 Back-propagation artificial neural network (BP-ANN)

BP-ANN is a common and easy understanding method which commonly used by many researchers to model the material behaviour. The common BP-ANN algorithm used such as Multilayer Perceptron and Radial Basis Function. The backpropagation algorithm usually be the main training algorithm where it can be found in Levenberg-Marquardt learning algorithm. The most common BP-ANN algorithm is feed-forward neural network (FNN) with at least one single hidden layer where the connection between nodes/neurons do not form a loop, unlike the Recurrent Neural Network (RNN). BP-ANN algorithms update the weight and bias that connected to each neuron by using gradient decent method, which accommodate with learning rule as shows in Eq. (6):

$$\mathbf{W}_k = \mathbf{W}_{k-1} - \eta \frac{\partial E(\mathbf{W})}{\partial \mathbf{W}} \tag{6}$$

Where, $\mathbf{W}_k, \mathbf{W}_{k-1}, \eta, \frac{\partial E(\mathbf{W})}{\partial \mathbf{W}}$ are updated weight, previous weight, learning rate (*eta*) and the gradient of error with respect to weight, respectively.

2.2.4 Extreme learning machine (ELM)

ELM training algorithms perform similarly to a common neural network, such as FNN. However, the ELM algorithm assigned the input weight and hidden bias randomly (e.g. normal distribution) if only the activation function is infinitely differentiable. The activation function commonly used are Sigmoid, Sine and Relu. From that, the hidden layer output in the form of the matrix, **H** can remain unchanged once these parameters are defined. Then, a method named Moore Penrose generalized inverse $(\mathbf{H}^{\dagger})$ is used to determine the output weight, β applying Eq. (7) and (8):

$$\beta = \mathbf{H}^{\dagger}\mathbf{T} \tag{7}$$

$$\mathbf{H}^{\dagger} = (\mathbf{H}^{\mathbf{T}}\mathbf{H})^{-1}\mathbf{H}^{\mathbf{T}} \tag{8}$$

After gaining the output weight, the model then can predict the output, $\mathbf{T}$ following Eq. (9):

$$\sum_{i=1}^{N} \beta_i g\left(w_i . x_j + b_i\right) = \mathbf{T}_j \tag{9}$$

Where, g, w, x and b refer to activation function, input weight, input data and hidden layer bias, respectively.

2.3 Fabrication Process of Magnetorheological Elastomer

The preparation of MRE and fabrication process was based on the published works in [13]. Two main materials were prepared in room temperature which were vulcanization silicon rubber (RTV-SR) as matrix and carbonyl iron particle (CIP) as magnetic particle. RTV-SR and CIP were measured by using weighing balance where the CIP concentration used in this work was 30 wt%, homogenously distribute (isotropic). The mixture was vigorously stirring using a mixer about 10 minutes. The compound was then cured for about 2 hours in a mould with a diameter of 40 cm and 1 mm thickness at room temperature. To stiffen the compound, a curing agent from Nippon Steel was used as cross-linking agent. The MRE sample then undergo oscillation testing which was shear strain amplitude sweep test using rotational MCR 302 rheometers from Anton Paar. The operating frequency was set to 1 Hz with 28℃ temperature condition.

2.4 Data Sets and Performance Index

Data sets were collected after finished the oscillation testing on the MRE sample. The data were filtered and undergo normalization process before it was used for training for machine learning model. There were 180×4 of raw data set gained from the testing. From this total data set, the data division was done for training and testing purposes prior to the modelling process. For machine learning models, two magnetic flux densities were chosen as unlearned data (580mT, 850mT) while another four as learned data (0mT, 180mT, 360mT, 701mT). To analyze the performance of models, two performance indexes were selected which were root mean square error (RMSE) and coefficient of determination (R^2) as in Eq. (10):

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(Y_i - \hat{Y}_i)^2}{\sum_{i=1}^{n}(Y_i - \bar{Y}_{output})^2} \tag{10}$$

Where, Y_i, $\hat{Y}_i$, $\bar{Y}_{output}$ and n are target, predicted output, mean of target and number of data set respectively.

3. Results

3.1 Pattern Recognition of Storage and Loss Modulus

The input for this model were shear strain and magnetic flux density. In general, the storage modulus was maintained at low shear strain region. Then, it starts to decrease towards increasing shear strain due to Payne effect. Table 1 presents the fitted model parameters of the Krauss model for all magnetic flux densities. There were four coefficients for each storage and loss modulus as shown in Eq. (1) and Eq. (2), respectively. It noticed that the simulation schemes of the Krauss model have at least six fitting processes to complete for all simulations results. In each stage, a curve fitting process must be calculated to obtain the optimum accuracy. In this case, the burden on the computational process seems un-noticeable. However, it is significant when dealing with more critical parameters of MRE such as varies temperature and other operation conditions.

Table 1
The principal parameters of Krauss model

Magnetic flux density (mT)	Storage modulus (MPa)				Loss modulus (MPa)			
	G'_{∞}	G'_{0}	γ_c	m	G''_{∞}	G''_{m}	γ_c	m
0	0.06	5	0.14	0.14	0.06	0.035	9	0.006
180	0.06	6	0.15	0.15	0.055	0.033	11	0.008
360	0.05	7	0.15	0.16	0.056	0.034	12	0.01
580	0.06	7	0.155	0.18	0.058	0.035	11	0.009
701	0.06	6	0.162	0.20	0.0585	0.036	10	0.007
850	0.06	6	0.163	0.23	0.061	0.037	10	0.0075

The comparison between the experimental and various models for 580mT were shown in Figure 1 and Figure 2 for storage and loss modulus, respectively. From Figure 1, GPR, SVR and Krauss models have similar trends at low amplitude shear strain. Meanwhile, even though there was a slight error from ELM and BP-ANN observed from 0.0014% until 0.00813% of shear strain, the pattern from both simulations was then followed well by the experimental data towards the maximum shear strain, specifically from the outstanding performances of ELM model. Concurrently, GPR, SVR and Krauss models were then able to follow the trend starting from 4.36% towards the maximum shear strain. Despite that, GPR, SVR and Krauss model had difficulties simulating the phase changes from linear to the nonlinear region in which the error was relatively high. It was noted in Figure 1, where changes like the curve formed significantly at 2.67% until 3.08% of the shear strain.

In the meantime, the SVR model performs poorly in predicting loss modulus, as shown in Figure 2. Furthermore, loss modulus should be maintained from 0.00141% until 0.267% of shear strain. Nevertheless, the simulated Krauss model failed to follow this trend. For real, it rises faster than expected. Meanwhile, BP-ANN, ELM and GPR show favourable performances. However, BP-ANN reveals an unusual trend at 0.379% until 3.08% of shear strain leading to higher RMSE value. This pattern which resembles the ‘overshoot phenomenon’ could be caused by overfitting occurred at critical transition position from linear to nonlinear region that was intended to be avoided in the prediction model. Nevertheless, ELM and GPR show excellent performances along with the given shear strain amplitude. However, the GPR model cannot maintain its performance in predicting the storage modulus, while the ELM model maintains good accuracy on both moduli. In the next paragraph, a detailed discussion on the linear and nonlinear region from the various models has been conducted.

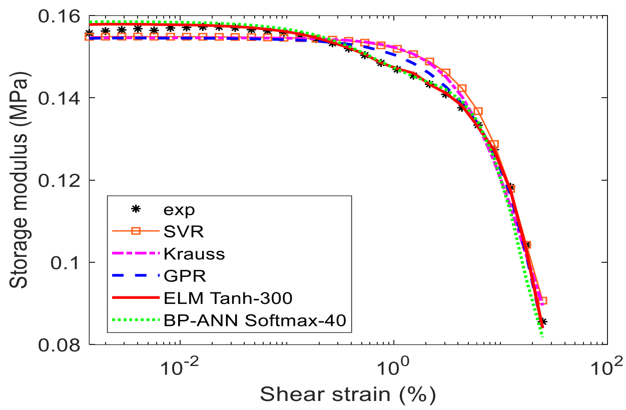

Fig. 1. The comparison of storage modulus between experimental data and simulation model

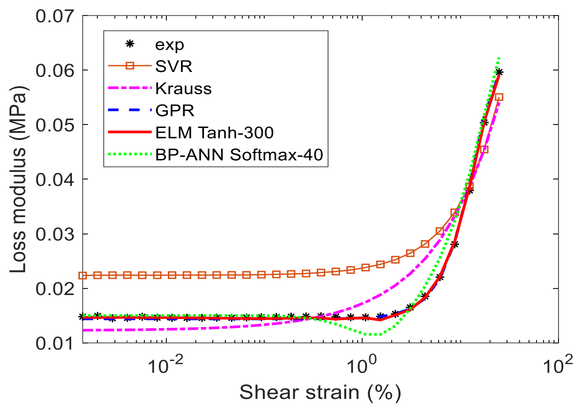

Fig. 2. The comparison of loss modulus between experimental data and simulation model

3.2 Prediction on LVE and NLVE of Storage Modulus

In general, given different loading conditions such as shear strain, the higher the magnetic field, the higher the storage modulus. This was due to chains from the magnetic field strength where adjacent CIP attracted to the magnetic field to increase along the field direction [22]. In addition, the storage modulus keeps stable at low shear strain amplitude before reaching a critical point where the storage modulus starts to decrease. The critical points refer to the LVE region limit in Eq. (11):

$$LVE\ limit\ value = Plateau\ value - (10\% \times plateau\ value) \tag{11}$$

In the LVE region, the microstructure of MRE can be described as stable. In contrast, the microstructure is weak in the NLVE region, which is beyond the LVE limit, mostly at 1% shear strain, contributing to the Payne effect [23]. Furthermore, the higher magnetic field may make the particle-to-particle interaction more utter than matrix interaction, which contributes to the lower LVE region. Agirre-Olabide *et al.,* [24] have explained how the LVE response can be determined. One of the ways is finding the LVE limit where it is located at 10% from the plateau of storage modulus. Hence, the LVE region is placed before that LVE limit. Meanwhile, the NLVE region is located after the LVE limit. Figure 3 presents the illustration of the response.

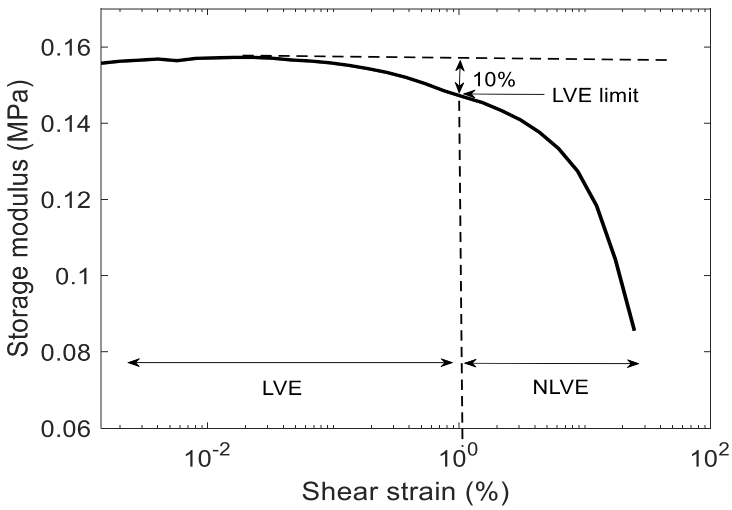

Fig. 3. The illustration of LVE and NLVE region [17]

The LVE region has been determined on experimental data and various models on all learned and unlearned magnetic fields. LVE limits were then found to be compared. Table 2 shows the LVE limit by experimental value and various models. From Table 2, the majority of LVE limits exhibited by SVR, Krauss and BP-ANN models differ much from experimental data except on learned 360mT predicted by SVR and on learned 170mT predicted by BP-ANN. In comparison, none from the Krauss model can predict the exact or the closest one with experimental data. On the other hand, most LVE limits produced by the GPR model from the available magnetic field have the same as experiment data, clearly on the learned magnetic field.

Table 2
The LVE limit for learned and unlearned magnetic flux densities

Magnetic flux density (mT)	Origin	SVR	Krauss	GPR	ELM	BP-ANN
0	0.1293	0.1286	0.1284	0.1268	0.1293	0.1287
180	0.1336	0.1369	0.1309	0.1336	0.1336	0.1332
360	0.1381	0.1381	0.1351	0.1381	0.1380	0.1371
580	0.1433	0.1368	0.1335	0.1386	0.1411	0.1421
701	0.1477	0.1402	0.1441	0.1477	0.1477	0.1458
850	0.1495	0.1423	0.1441	0.1423	0.1502	0.1512

Similar with the ELM model, where it may have the closed one on producing LVE limit at off-state (0mT) compared to the GPR model in which GPR model has a difference of about 0.0025 MPa. As for the unlearned magnetic field, ELM predicted more accurately than the GPR model on both interpolation (580mT) and extrapolation (850mT) estimation. The difference from the GPR model on extrapolation estimation was 0.0072 MPa, while the ELM model produced only 0.0007 MPa. After finding the LVE limit for each model, the accuracy of two regions can easily be measured as presented in Figure 4 and Figure 5 for LVE and NLVE regions, respectively.

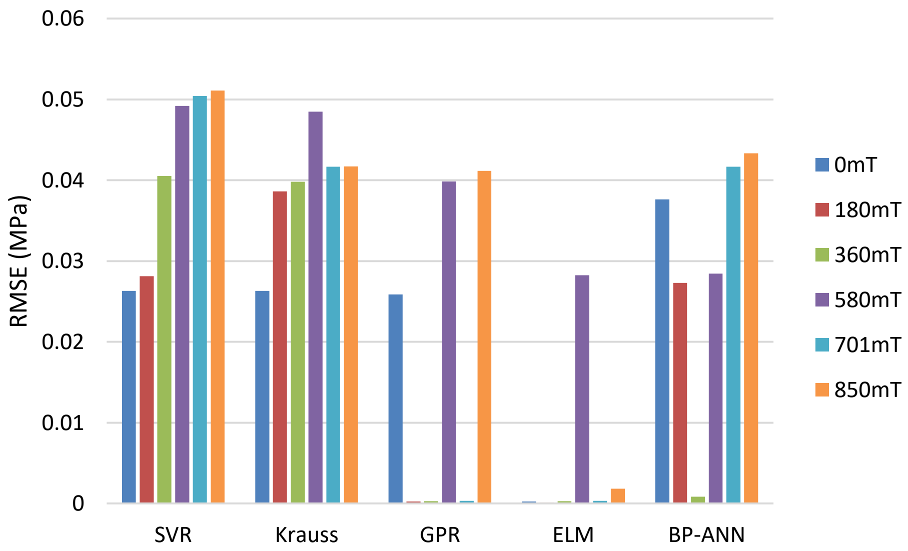

Fig. 4. The RMSE value on LVE region from various models

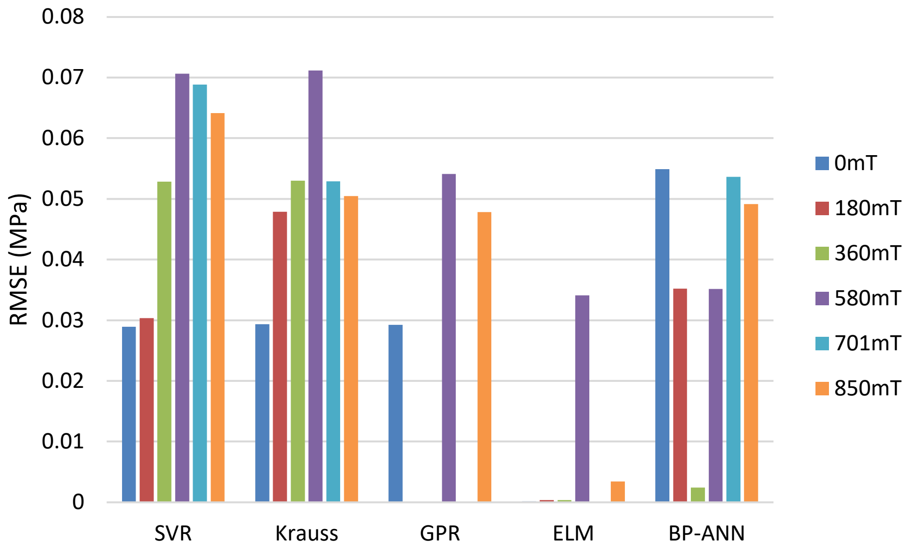

Fig. 5. The RMSE value on NLVE region from various models

The performance of the SVR model drops by increasing the magnetic field, where the error keeps increasing with the increment of the magnetic field. Moreover, error in the NLVE region exhibits higher than LVE region. Even though SVR can predict the exact LVE limit on 360mT, it does not mean that it has high performance. This might be due to a crossing point between simulated SVR and experimental data points. In addition, SVR comes as the worst model among other models in both regions. The Krauss model shows the second poor performance, with a similar error to the SVR model on both LVE and NLVE regions. Furthermore, better performances show from the BP-ANN model at 360mT in both regions compared to SVR and Krauss models. However, it cannot be compared with great simulation from GPR and ELM model in which the error was low obviously on all learned magnetic field happened specifically at NLVE region. In addition, the GPR model exhibits minimal error practically at 180, 360 and 701mT compared to the ELM model in the NLVE region.

Nevertheless, even though the error from the ELM model on the NLVE region was more significant than the LVE region, it was smaller than the error measured from the GPR model on the unlearned magnetic field on interpolation and extrapolation estimation. The error from LVE and NLVE measured from the ELM model was generally considerable and acceptable. By this, ELM has shown its best performance in predicting the nonlinear viscoelastic properties and comes as the best model among other machine learning models.

4. Conclusions

As the conclusion, Payne effect of MRE has been simulated by two kinds of model which were Krauss model and four machine learning models. From the simulated result, it can be concluded that both types of models able to predict both linear and nonlinear MRE viscoelastic properties with the given inputs variables. As the results, Krauss model as parametric model seems to have shortcomings when dealing with more input variables as the models need to undergo complex derivation to consider new input variable while it also contributes to computational burden. Meanwhile, machine learning models was mentioned to be more accurate when dealing with more input data as it learns the pattern from the training data. However, a few limitations can be found such as finding the optimized hyperparameters seems to be difficult and presence of noise data can contribute to higher error. Thus, method to improve such as using the appropriate optimization method and filtering method to remove noise data can be done in future. Currently, MR materials keep progressed with establishment of new material to improve its properties so that it can fulfil the need for application purposes. Thus, machine learning model has a potential to be a simulation tools for early development process.

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