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Integrative Machine Learning Approaches for Predicting the Rheological Behaviour of Soft Magnetorheological Elastomers

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Abstract

Magnetorheological Elastomers (MREs) are advanced composite materials known for their ability to alter mechanical properties under external magnetic fields, making them highly valuable in adaptive damping systems, vibration control, and smart devices. The accurate prediction of rheological behavior in soft MREs remains a significant challenge due to the complex interplay between material composition and magnetic fields. To address this challenge, this study employs a multi-pronged approach that integrates traditional material science techniques with advanced data analytics. Scanning Electron Microscopy (SEM) and Energy-dispersive X-ray Spectroscopy (EDS) were used to confirm the uniform distribution of iron particles within the MRE samples, ensuring material integrity. Magnetorheological testing data were subsequently collected and utilized for training and validating machine learning algorithms, specifically Random Forest and Gradient Boosting models. These algorithms were rigorously evaluated using established metrics such as R-squared (R^2), Mean Absolute Error (MAE), and Root Mean Square Error (RMSE) to determine their predictive performance. The results demonstrated high accuracy, with R^2 values exceeding 90% for both training and validation datasets. Furthermore, exploratory analysis, including partial dependence plots and feature importance rankings, provided deeper insights into how factors such as Carbonyl Iron Percentage, curing magnetic field, and strain rate influence the complex shear modulus. This research bridges the gap between empirical material science and computational prediction methods, offering a robust framework for predicting the viscoelastic behavior of MREs. The findings contribute to the optimization of MREs for real-world applications, enhancing their potential in smart material systems.

Keywords: Soft magnetorheological elastomers (MREs), machine learning, regression models, rheological behaviour, ferromagnetic particles, curing magnetic field, complex shear modulus

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INTRODUCTION

In the domain of advanced materials, Magnetorheological Elastomers stand as a quintessential exemplar of innovation and tunability [1]. These composite materials, characterized by the dispersion of ferromagnetic particles within an elastomeric medium, have unlocked a realm of possibilities in dynamic systems by offering real-time tunability of mechanical properties under the influence of an external magnetic field [2], [3], [4]. The physics underlying this behavior stems from the interplay between the microstructure of dispersed particles and the encompassing elastomer matrix, a synergy that has been the subject of extensive studies. MREs are reshaping the dynamics of adaptive damping by offering unparalleled control over material responses [5], [6]. introducing innovations in the domain of magnetically actuated medical prosthetics and devices [7], and even redefining boundaries in optoelectronics by facilitating magnetically tunable optical systems [8], [9].

The Complex Shear Modulus (CSM) is a critical rheological parameter for the characterization of MREs. It is an indicator of the material's resistance to shear deformation when a force is applied which capture both its elastic and viscous behaviours [10]. The accurate prediction of CSM in the presence of varying factors such as external magnetic fields, frequencies, and strain rates becomes crucial, especially when optimizing the performance of devices utilizing MREs [11], [12]. Over the years, various modelling techniques have been employed to predict the CSM of MREs. One popular approach was the microstructure-based model [13]. [14]. which delves deep into the material's microscopic realm. These models focus on interactions and arrangements of ferromagnetic particles within the elastomer matrix. While it offers valuable insights into micro-interactions, it sometimes falls short in capturing the overall rheological properties, especially under dynamic conditions. On the other hand, continuum-based models [15–17] looks at MREs from a macroscopic perspective, treating the material as a continuous whole rather than focusing on individual particle interactions.

LITERATURE SURVEY

Traditional viscoelastic models [18]. often seen as the foundation for rheological studies, have also been used extensively. Rooted in classical mechanics, models such as the Kelvin-Voigt [19]. model, Four-parameter viscoelastic model [20]. Bouc-Wen model [21]. Prandtl-Ishlinskii [22]. and Fractional rheological models [23]. hinge on the premise that materials inherently exhibit both elastic and viscous components. Viscoelastic models, often phenomenological, are primarily tailored to force-displacement responses [24]. predominantly emphasizing the external macroscopic behaviour of MREs. They tend to gloss over the microstructural characteristics of MREs. More so, they frequently overlook the magnetic interplay and interactions at the micro-level, leading to potential discrepancies between theoretical musings and experimental realities [25]. Furthermore, several of these models, despite their analytical proficiency, can become computationally hefty, especially when diving deep into mathematical derivations involving higher-order differentiations. Such intricacies can hamper swift predictions of MRE behaviour, especially when new factors or input dimensions, like composition or temperature, are brought into the modelling equation [25, 26].

While certain deterministic models have exhibited proficiency in capturing the intricate dynamics of Magnetorheological Elastomers under specific scenarios, there remains a scarcity of models that empirically predict their viscoelastic responses in varying magnetic conditions. Contrasting these deterministic frameworks, Machine Learning approaches, inherently non-deterministic, offer a more adaptable framework [27–29]. This adaptability facilitates the modelling of Magnetorheological Elastomer behaviours, granting ease in tailoring input-output relationships. ML methodologies have emerged as a transformative force in the computational modelling of complex systems, presenting a paradigm shift from traditional techniques. In myriad fields, from healthcare to finance, machine learning is driving innovations by enabling researchers and professionals to make predictions more accurately and efficiently than ever before. For instance, in the medical arena, ML algorithms sift through enormous datasets to detect the early signs of diseases, sometimes even before symptoms manifest [30]. In the finance sector, predictive models facilitate risk assessment, streamlining investment strategies [31]. In the specific context of MREs, machine learning algorithms offer a novel framework to capture the intricate magneto-mechanical coupling and nonlinear viscoelastic responses that have proven elusive for classical models. Among the domain of machine learning approaches, ensemble methods such as Random Forest and boosting algorithms like Gradient Boosting have shown particular promise [32]. These algorithms leverage the power of multiple decision trees to capture complex, high-dimensional relationships, thus offering a nuanced understanding of the underlying physics of MREs. Notably, ensemble methods have demonstrated the ability to mitigate the risk of overfitting, providing models that not only fit the training data well but also generalize effectively to new, unseen data.

Yu et al [33]. investigated the use of a hybrid model that merged Support Vector Regression (SVR) with Particle Swarm Optimization (PSO) to predict the hysteretic behavior of an MRE-based base isolator. The PSO method enhanced the model's convergence capabilities using optimized parameter tuning within the SVR model, and with the integration of an S-curve function-based self-adaptive inertia weight factor, showing better prediction accuracy than traditional methods. Additionally, Yang et al. [34]. revealed the significance of using Ant Colony Optimization (ACO) combined with neural networks to further refine the predictions of shear force in MRE systems by optimizing the model parameters dynamically, thus reducing the prediction error significantly. Another compelling study by Leng et al. [35] employed an Artificial Neural Network (ANN) optimized through fuzzy algorithms to predict the nonlinear relationship between displacement, frequency, current, and shear force in MRE systems. The ANN-Fuzzy hybrid model exhibited remarkable precision in predicting complex behaviors of MREs, particularly under varying magnetic fields.

Similarly, Yu et al. [27] introduced an Extreme Learning Machine (ELM) approach for predicting the dynamic responses of MRE-based devices, demonstrating the potential of ELM to outperform traditional parametric models, especially in the rapid prediction of MRE behavior under different magnetic and mechanical conditions. These studies underscore the flexibility and accuracy of ML models in capturing the complex nonlinearities of MREs, which are often missed by deterministic models. Saharuddin et al. [25], [26] conducted another key study comparing several machine learning architectures to predict the viscoelastic properties of MREs. Their evaluation of ANN models with different hidden neuron configurations and ELM models using various activation functions such as SIGMOID and HARD LIMIT revealed that ELMs provided more accurate predictions of the storage modulus and loss factor under changing magnetic field conditions. In particular, the ELM-HARD LIMIT model demonstrated the best performance for loss factor prediction, while the ANN with a higher number of hidden neurons was more adept at predicting the linear viscoelastic limit.

Following an in-depth exploration of the foundational studies and recent advancements in MRE modeling, this article shifts focus to the pivotal role of machine learning models in enhancing the predictive accuracy of MRE behavior. By integrating experimental techniques like X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM), and Energy-Dispersive X-ray Spectroscopy (EDS), along with rheological data, the subsequent sections aim to provide a comprehensive evaluation of the machine learning frameworks applied to MREs. For the evaluation of machine learning models, the study employs a set of well-established metrics such as Coefficient of Determination (R^2), Mean Absolute Error (MAE), and Root Mean Square Error (RMSE) designed to rigorously validate the predictive capabilities of the algorithms in question. The rigorously validated predictive framework promises to not only elevate the understanding of MREs but also catalyse their optimization for real-world applications.

EXPERIMENTAL APPROACH AND ANALYTICAL TECHNIQUES

Machine Learning Tools

In the domain of predictive analytics within material sciences, machine learning algorithms present a diverse range of modelling capabilities. This study utilizes two such algorithms Gradient Boosting, and Random Forest. Each offers distinct strengths in modelling the complex behaviours inherent in the material. The foundational basis of employing these algorithms is delineated in Figure 1, which visually represents the structured workflow of the study.

Material Fabrication and Data Acquisition

The material fabrication process aims to yield a diverse dataset to capture a wide range of behaviours exhibited by MREs.

Sample fabrication

Four distinct categories of MRE samples were fabricated with varying weight percentages of iron particles (4-8 μm) in Elastosil rubber of 12 Shore hardness [36]. Specifically, samples were made at 25%, 40%, 55%, and 70% weight percentage of iron particles. These were then subjected to curing under magnetic fields of 0, 0.2, 0.35, and 0.45 Tesla, resulting in 4 isotropic samples (cured without a magnetic field at 0 Tesla) and 12 anisotropic samples (cured under varying magnetic fields), for a total of 16 unique samples.

Data acquisition

To check the dispersion of carbonyl iron particles (CIP), advanced characterization techniques were employed to provide a multi-faceted understanding of the material properties. Energy-dispersive X-ray Spectroscopy (EDS) was performed to ascertain the elemental composition and the spatial distribution of these elements in the composite material [37]. Figure 2 provides a compelling EDS map that showcases not only the presence of iron particles but also their dispersion and interaction with the elastomeric matrix, thereby confirming the material's compositional integrity and heterogeneity.

Subsequently, rheological properties were measured using an Anton Paar Rheometer 102e, located in the Fluid Mechanics Lab at IIT Ropar, ensuring precise and controlled testing conditions for the study. The measurements offer invaluable insights into the material's viscoelastic behaviour under varying magnetic field intensities and strain rates. These measurements serve as the foundational dataset for the machine-learning models, providing both training and validation data. Figure 3 supplements this data by presenting the results of the complex shear modulus against strain levels for various CIP weight percentages of 25%, 40%, 55%, and 70%, thus offering a comprehensive overview of the material's rheological behaviour.

Feature Description

The feature selection for studying MREs was strategic, focusing on Carbonyl Iron Percentage for magnetic susceptibility, Curing Magnetic Field for particle alignment, Frequency to assess viscoelastic behaviour, and Strain Rate to understand mechanical stress response. These, alongside the Complex Shear Modulus as the target variable, were crucial in capturing the multifaceted rheological properties of MREs, integrating both elastic and viscous responses [38].

Feature Engineering

In this study, dataset normalization and outlier correction ensured balanced feature impact on the model, with imputation enhancing data quality. 'CIP Percentage × Curing Magnetic Field' interaction terms, informed by correlation analysis in Figure 4, and further explored through advanced feature engineering, revealed nuanced MRE behaviours. Partial dependence plots in Figure 5 highlighted how CIP percentage directly increases the shear modulus, with a notable effect of the curing magnetic field observed at 0.25T to 0.5T. In contrast, frequency impact. remained minimal, and strain rate significantly influenced the modulus, illustrating the complex interplay of factors affecting MRE rheological properties.

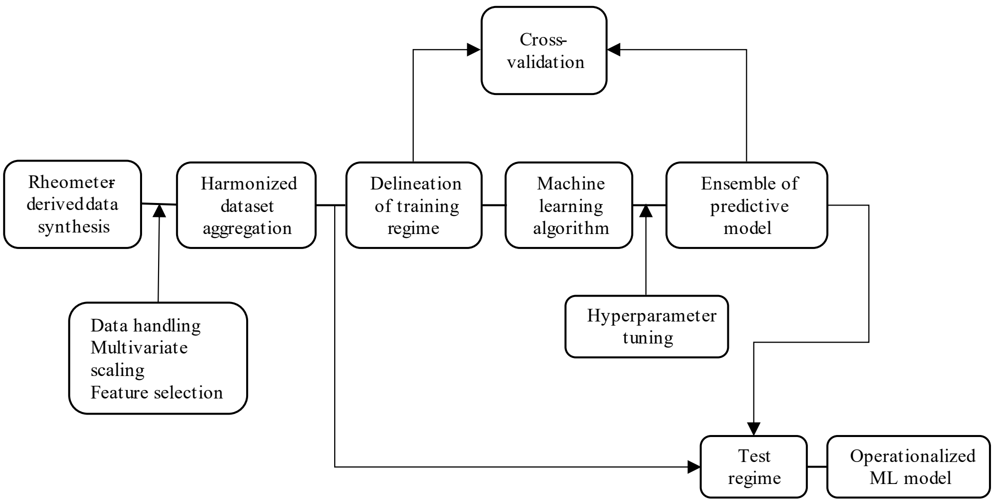


Figure 1. Structured workflow diagram detailing the sequential implementation of machine learning algorithms in the study.

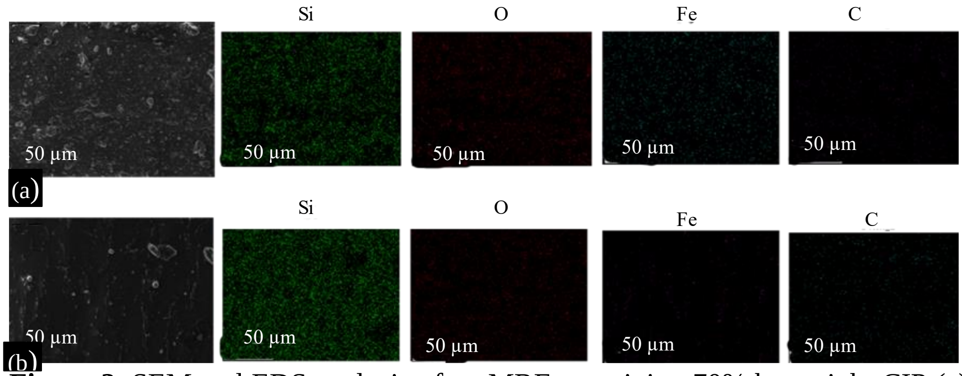
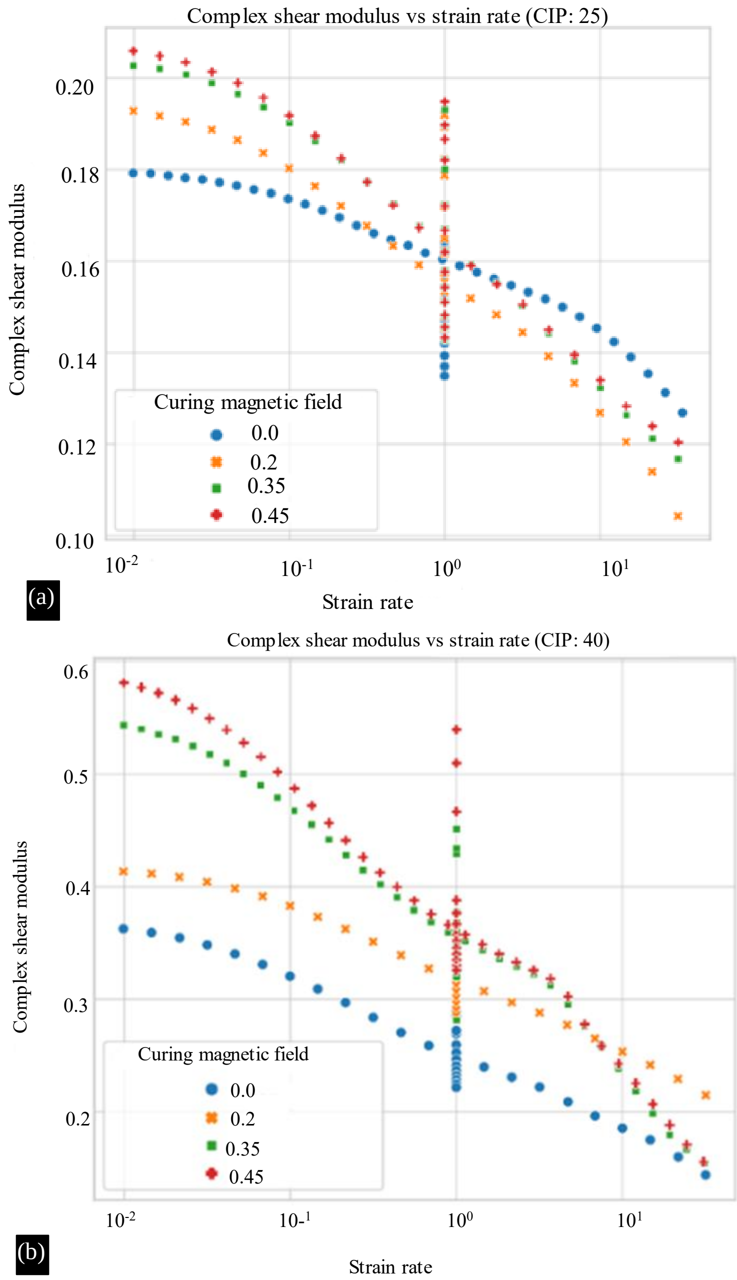


Figure 2. SEM and EDS analysis of an MRE containing 70% by weight CIP (a) isotropic MREs, (b) anisotropic MREs.



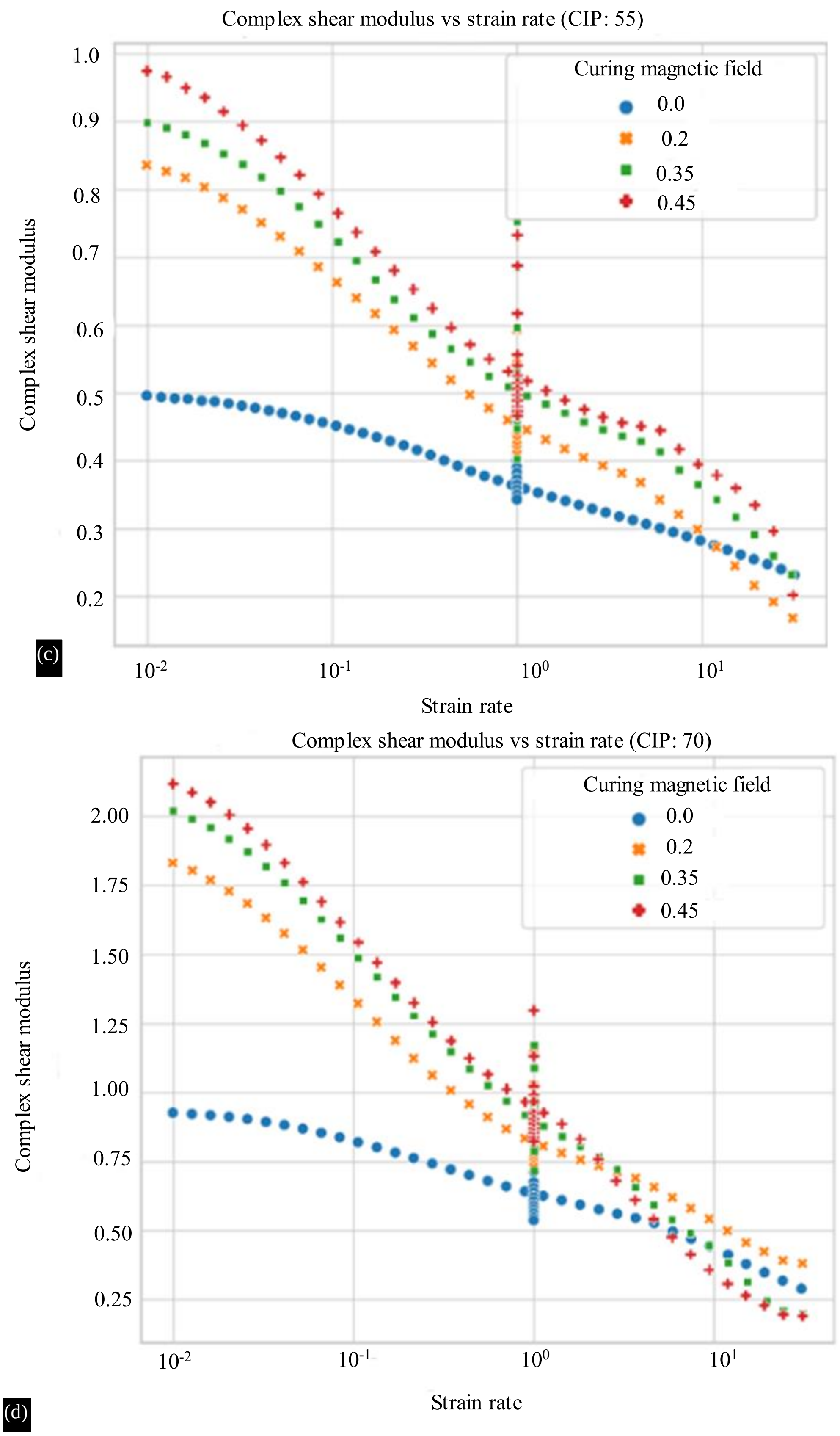


Figure 3. Plots illustrating the relationship between complex shear modulus and strain rate for magnetorheological elastomer samples cured at varying magnetic fields.

RESULTS AND DISCUSSIONS

The evaluation metrics employed include R^2 (R-squared), Mean Absolute Error (MAE), and Root Mean Square Error (RMSE). These metrics collectively offer a comprehensive assessment of each model's predictive performance and robustness [39], [40]. The ensuing discussion also delves into the feature importance analysis and evaluates the robustness of the models by comparing training and validation metrics. For the application of Linear Regression in predicting the Complex Shear Modulus of MREs, the model presented an R^2 value of 0.67 for the training set and 0.64 for the validation set as tabulated in Table 1.

Table 1. Evaluation Metrics Across Algorithms for MRE Shear Modulus Prediction.

Algorithm	**Linear regression**		**Gradient boosting**		**Random forest**	
Metric↓	***Training***	***Validation***	***Training***	***Validation***	***Training***	***Validation***
R^2	0.67	0.64	0.96	0.93	0.98	0.94
MAE	0.145	0.169	0.02	0.029	0.0065	0.0156
RMSE	0.224	0.263	0.027	0.038	0.0105	0.0269

Gradient Boosting Model Evaluation

Hyperparameter tuning of gradient boosting model

Hyperparameter tuning optimized the Gradient Boosting model, focusing on learning rate, estimators, and tree depth to accurately model data without overfitting [41]. A three-fold cross-validation identified optimal parameters: a Learning Rate of 0.2, Max Depth of 5, and 150 Estimators, achieving R^2 values of 0.96 (training) and 0.93 (validation), indicating superior predictive accuracy over Linear Regression. Figure 6 portrays the actual versus predicted values of the Complex Shear Modulus for the Gradient Boosting model. The high degree of overlap between the actual and predicted values reinforces the high R^2 values observed, validating the Gradient Boosting model's efficacy in capturing the complex rheological behavior of MREs.

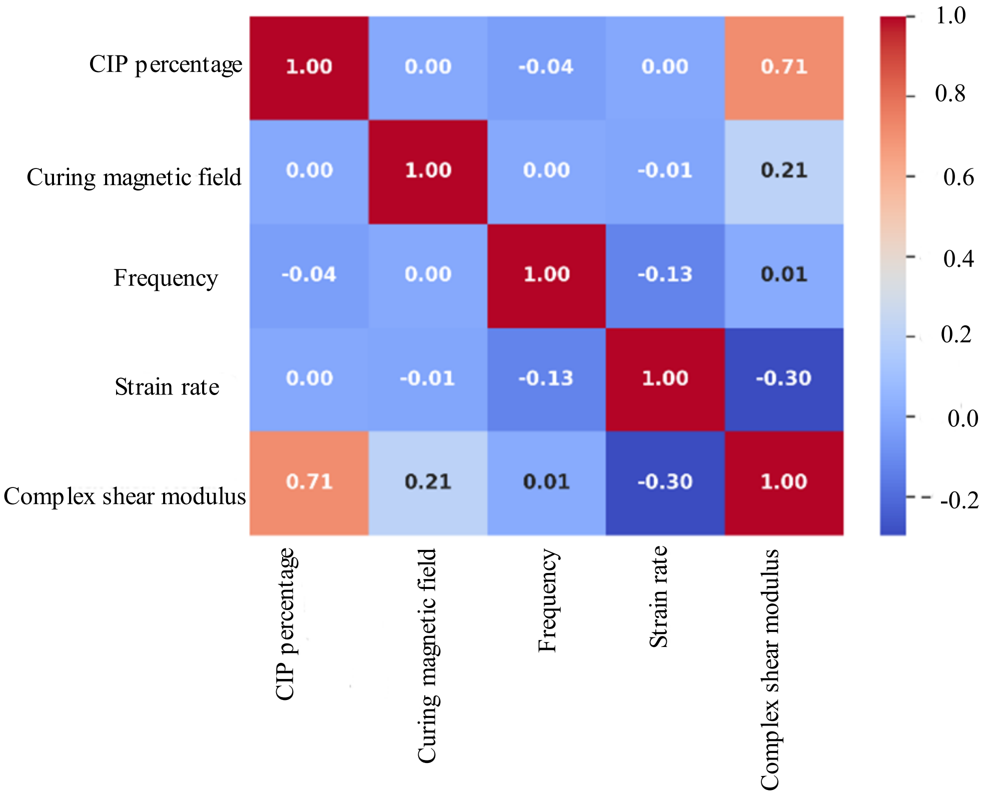


Figure 4. Correlation matrix of the features and target variable in the study.

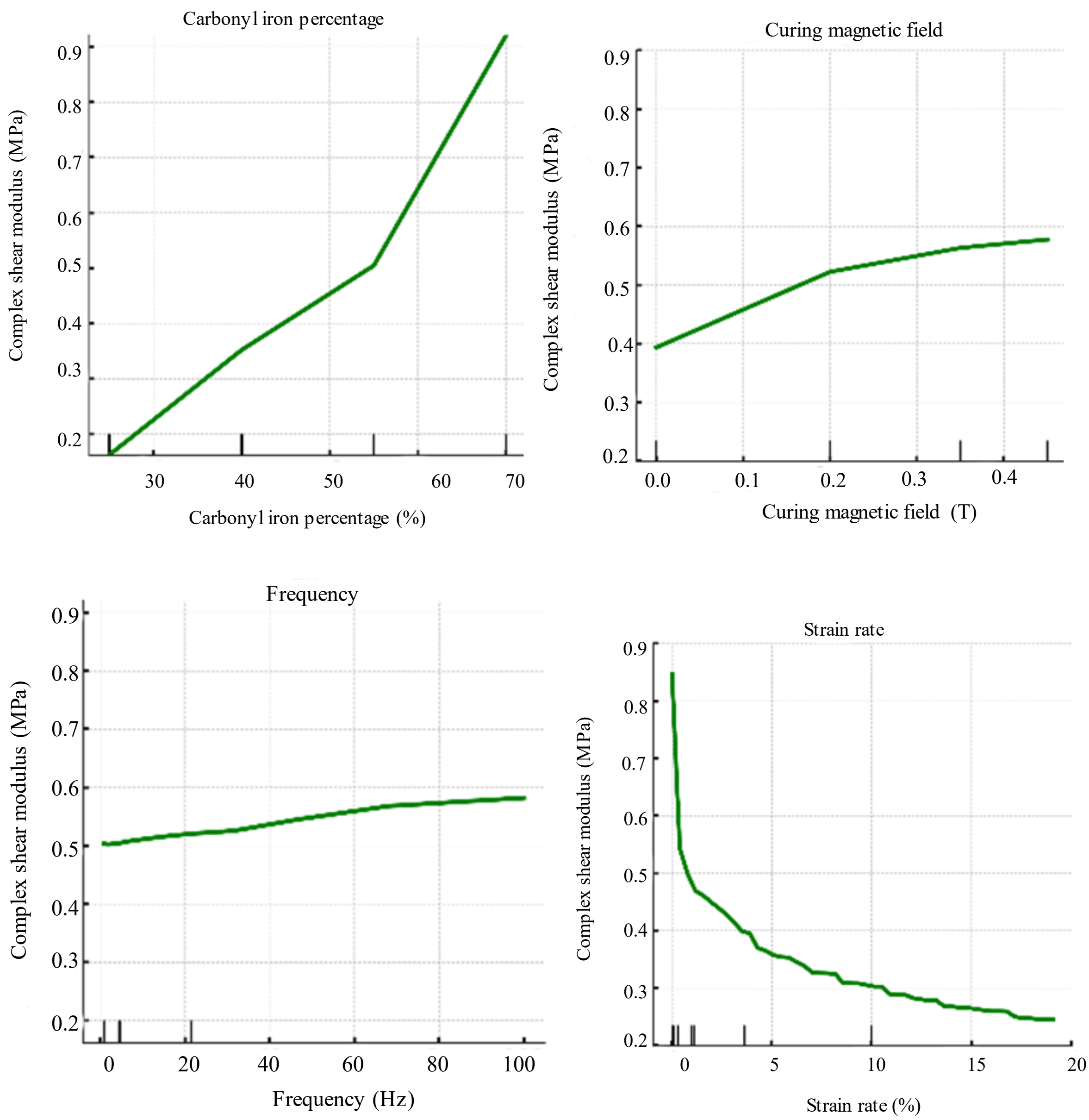


Figure 5. Partial dependence plots showcasing the influence of individual features, namely, (a) carbonyl iron particle (%), (b) curing magnetic field (T), (c) frequency (Hz), and (d) strain rate (%), on the predicted complex shear modulus (MPa).

Feature importance analysis

Figure 7 reveals the variables that significantly influence the Complex Shear Modulus in the context of MREs. The Carbonyl Iron Particle (CIP) emerges as the most influential feature, with an importance value of approximately 0.535, highlighting its pivotal role in determining the material's resistance to shear deformation. Strain Rate follows closely, with an importance of around 0.350, indicating its significant impact on the rheological behavior of MREs. The Curing Magnetic Field and Frequency have lower importance scores, but they are not negligible and contribute to the intricate relationships in the rheological properties of MREs.

Error distribution plots

Figure 8 reveals that over 90% of the training set residuals from the Gradient Boosting model fall within ±0.025 error margin, indicating the model's precise capture of MRE rheology complexities, influenced by factors such as CIP content and curing magnetic field strength. For the validation set, a broader error distribution is noted, with around 85% of predictions within range of ±0.035.

Random Forest Model

Hyperparameter tuning in random forest model

Hyperparameter tuning for the Random Forest model [42], involving n_estimators, max_depth, min_samples_split, and min_samples_leaf, through a three-fold cross-validation grid search, identified an optimal set in Table 2, achieving 98% training and 94% validation variance explanation, detailed in Figure 9.

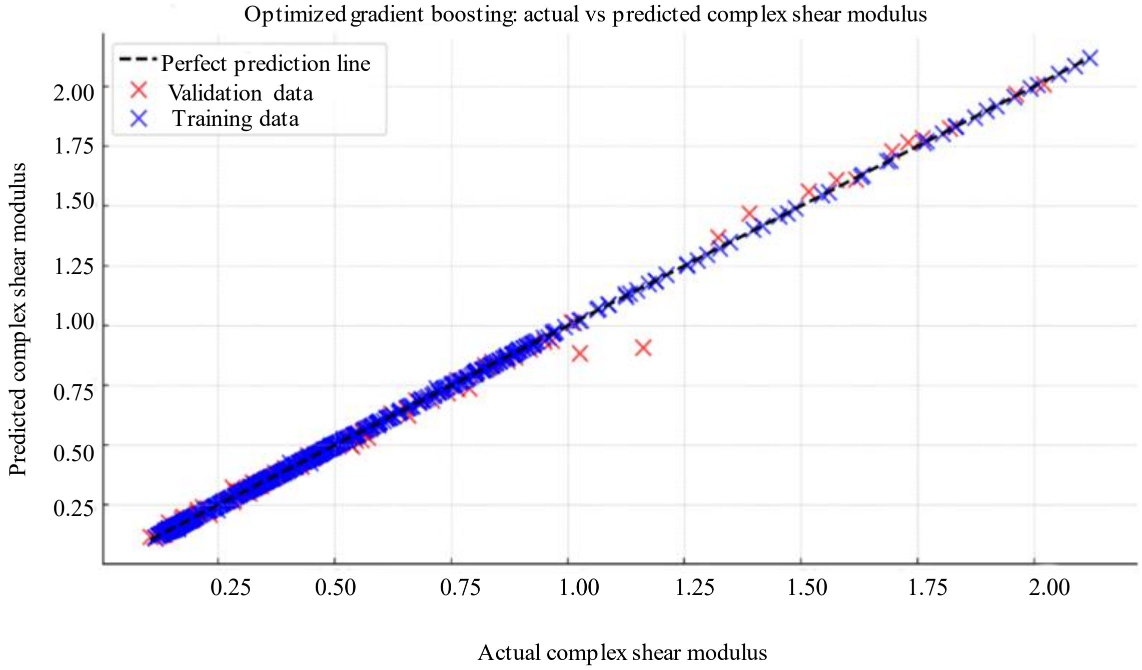


Figure 6. Gradient boosting's predictive performance on complex shear modulus of MREs: actual vs. predicted.

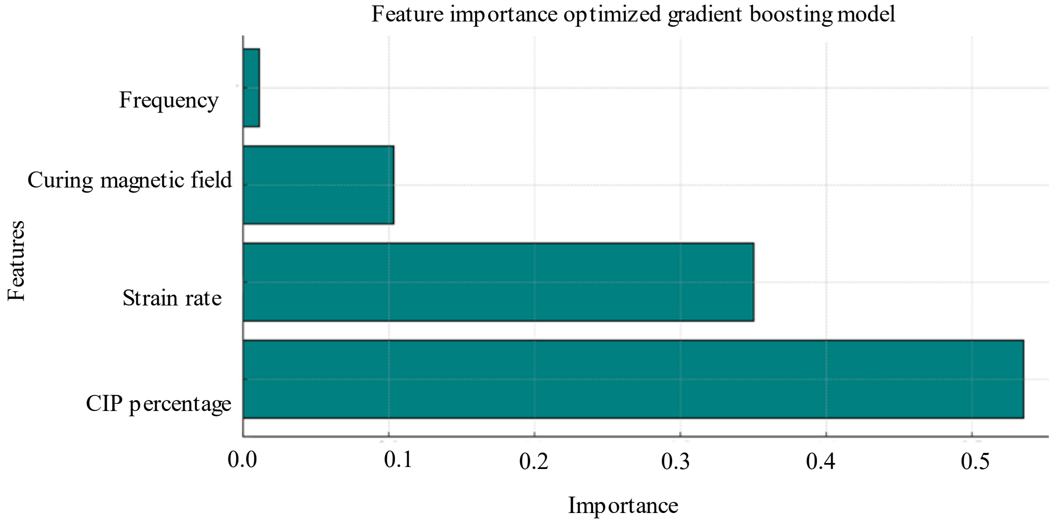


Figure 7. Feature importance distribution derived from gradient boosting analysis for MRE behaviour prediction.

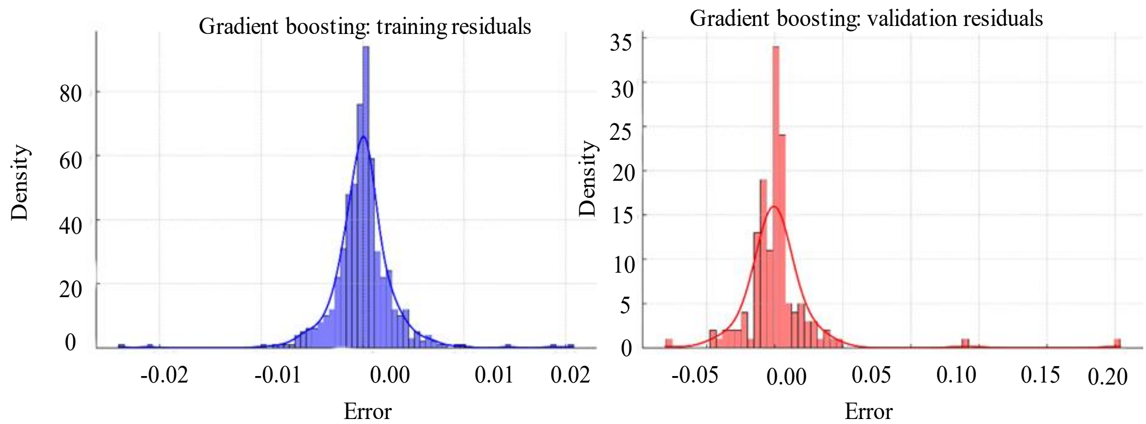


Figure 8. Distribution of errors for the gradient boosting model on both training and validation datasets.

Table 2. Optimized Hyperparameters for Random Forest Model Deduced from Grid Search Tuning.

Parameter	**Optimized**
Number of Estimators (n_estimators):	100
Maximum Depth (max_depth):	None (i.e., nodes are expanded until they contain fewer than min_samples_split samples)
Minimum Samples Split (min_samples_split):	2
Minimum Samples Leaf (min_samples_leaf):	1

Feature importance analysis

The CIP Percentage is identified as the most critical variable, influencing 53.5% of feature importance, followed by Strain Rate at 36.3%, with Curing Magnetic Field and Frequency at 9.2% and 1.1% respectively, illustrating their roles in MRE properties, as detailed in Figure 10.

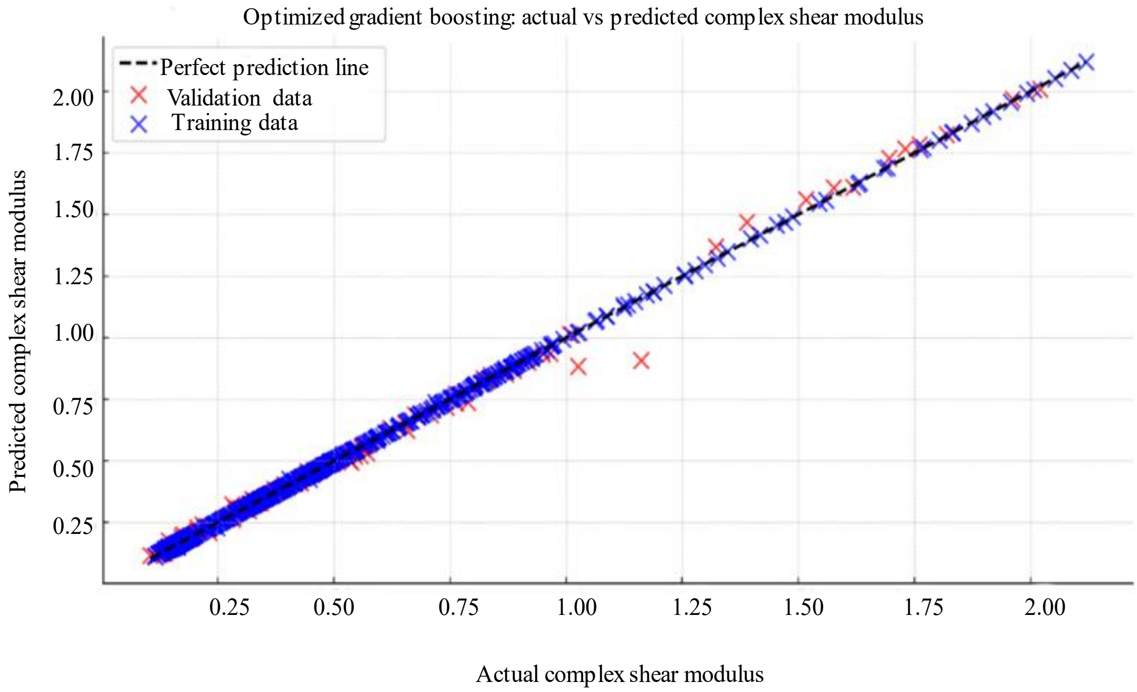


Figure 9. Random forest's predictive performance on complex shear modulus of MREs: actual vs. predicted.

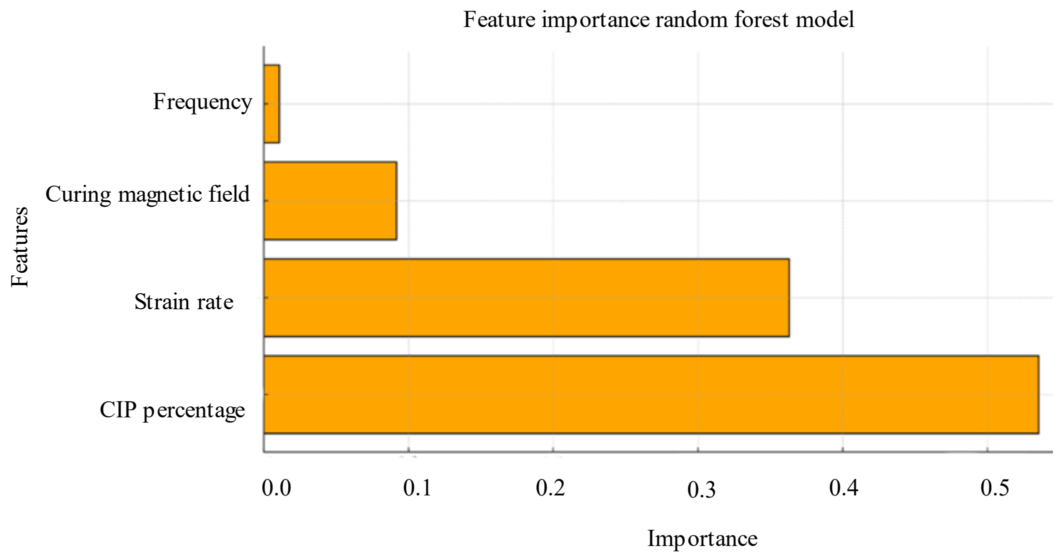


Figure 10. Feature significance assessed through random forest algorithm for MRE rheological analysis.

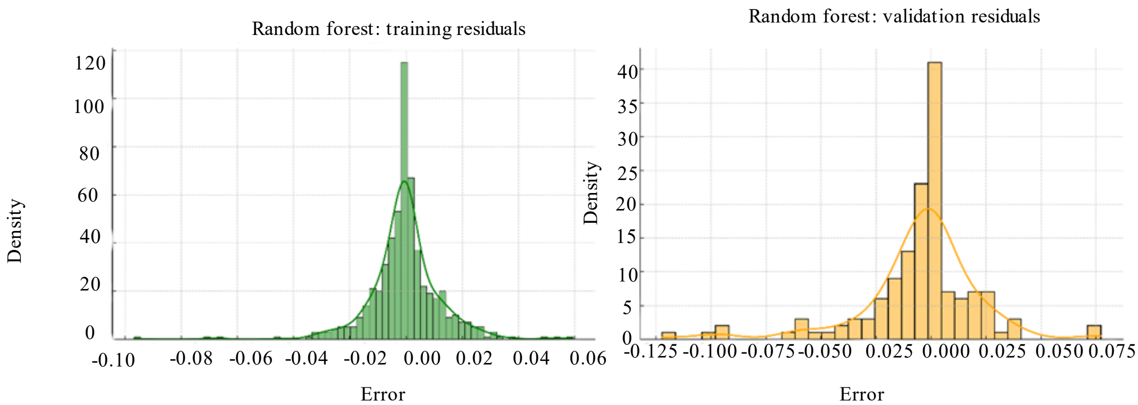


Figure 11. Distribution of residuals for the random forest model on both training and validation datasets.

Error distribution

The Random Forest model demonstrates exceptional precision, with 95% of training predictions within ±0.015 error, highlighting its capability to capture MRE behaviour nuances, as shown in Figure 11. Despite a broader validation error spread (90% within ±0.025), the model's performance indicates strong adaptability and understanding of complex MRE dynamics in unseen datasets.

CONCLUSIONS

Predicting MREs behavior, particularly the Complex Shear Modulus, is of paramount importance for optimizing real-world applications. Traditional modeling approaches, although insightful, sometimes fall short of capturing the intricate dynamism of MREs. In this study, machine learning techniques, specifically Gradient Boosting, and Random Forest models were employed to address this challenge. After rigorous hyper parameter tuning, the Gradient Boosting model demonstrated superior performance with R^2 values of 0.96 and 0.93 for training and validation sets, respectively. Furthermore, exhibiting robustness, the Random Forest model achieved an impressive R^2 value of 0.98 for training and 0.94 for validation. The feature importance analysis underscores Carbonyl Iron Percentage and Strain Rate as significant influencers on the target variable. Error distribution analysis further validated the precision of the employed machine learning models, particularly the Gradient Boosting and Random

Forest algorithms. The predictions were closely aligned with the actual rheological behaviors of the MREs, showcasing their potential as reliable predictive tools.

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