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Predictive Modelling of Graphene-Based Supercapacitors for Enhanced Energy Storage Applications: A Machine Learning Approach

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Abstract

Despite the growing interest in graphene-based materials for supercapacitors, owing to their high electrical conductivity and huge specific surface area, there are currently no systematic methods for accurately predicting their electrochemical performance. Current research in this area is often hindered by empirical trial–and–error techniques and fragmented datasets, impeding the logical design and optimization of high-performing devices. To address these gaps, the present study leverages ML to forecast the performance of graphene-based supercapacitors, focusing on specific capacitance, power density and energy density. A comprehensive dataset was compiled from existing literature, encompassing physicochemical properties and electrochemical test features. Three ML models, Gradient Boosting Regression (GBR), Random Forest Regression (RFR), and Multiple Linear Regression (MLR) were employed to predict supercapacitor performance. The GBR model achieved the best overall performance with R^2 values of 0.9, 0.7, and 0.8, and MSE values of 1.03 F^2/g^2, 5.05 $(Wh/kg)^2$, and 2.30 $(W/kg)^2$ for specific capacitance, energy density, and power density respectively. The results indicate that GBR outperformed other models, achieving the highest determination coefficient (R^2) values and the lowest mean squared error (MSE) for energy density, power density, and specific capacitance. RFR showed comparable robustness with slightly higher MSE values, while MLR had the lowest accuracy among the three. Correlation analysis revealed that annealing temperature and current density significantly influence specific capacitance and power density, respectively. This study underscores the potential of ML in optimizing graphene-based supercapacitors, providing valuable insights for the advancement of next-generation energy storage technologies.

Keywords*: Graphene, supercapacitors, Machine learning, current density.*

1.0 Introduction

The growing global demand for effective energy storage solutions has accelerated research into advanced materials for supercapacitors, which are known for their substantial power density and quick charge-discharge ability. Among these materials, graphene which is a single layer of carbon atoms arranged in a 2D honeycomb lattice, has become an attractive candidate due to its exceptional electrical conductivity, large specific surface area, and superior mechanical properties (Akhtar & Akhtar, 2021; Du et al., 2023; Huang et al., 2012; Lemine et al., 2018; Liu et al., 2013; Salunkhe et al., 2014; Wang et al., 2021; Worsley et al., 2022). The unique structure of graphene facilitates efficient electron transport and provides an extensive surface area for charge accumulation, making it ideal for improving the supercapacitor performance. Recent advancements have focused on developing graphene-based composites – such as those combined with metal oxides or conducting polymers – to further improve energy storage capabilities (Salunkhe et al., 2014; Wang et al., 2021). For instance, nitrogen-doped graphene combined with metal oxides, including manganese, nickel, and cobalt oxides, has shown promise for supercapacitor applications due to enhanced pseudocapacitive properties (Deshsorn et al., 2023; Yadav & Devi, 2020). Conducting polymers, such as polyaniline, offer environmental stability, low cost, and high pseudocapacitance when combined with graphene (Kumar & Baek, 2014). Despite these advancements, challenges persist in optimizing the synthesis processes to maintain the intrinsic properties of graphene and in achieving scalable production methods. Addressing these issues is essential for the practical application of graphene-based supercapacitors in next-generation energy storage systems (Akhtar & Akhtar, 2021; Du et al., 2023; Worsley et al., 2022). Recently, ML has become a potent tool in materials science, offering predictive insights that can significantly expedite the development of advanced materials. By analyzing vast datasets, ML algorithms can identify patterns and relationships among variables that traditional experimental approaches might overlook.

In the context of graphene-based supercapacitors, techniques involving ML have been used to predict specific capacitance and life expectancy, thereby guiding the design and optimization of electrode materials (Deshsorn et al., 2023; Jamaluddin et al., 2023; Mishra et al., 2023; Saad et al., 2022; Sawant et al., 2023; Yogesh et al., 2025; Zhu et al., 2018). For instance, research has shown that artificial neural networks and other ML

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models are effective in predicting the graphene-based electrode's electrochemical performance, considering factors like surface area, pore structure, and doping levels (Deshsorn et al., 2023; Saad et al., 2022). Shariq et al. (2024) utilized various ML models, including support vector regression, random forest, and multiple linear regression, to predict electrochemical properties such as electrical conductivity and sheet resistance (Saad et al., 2022). The integration of ML techniques into the study of graphene-based supercapacitors has opened new avenues for optimizing energy storage devices. For example, Saad et al., (2022) formulated various ML models, including artificial neural networks (ANN), Bayesian ridge regression (BRR), decision tree regression (DTR), and k-nearest neighbors 'regression (KNN) to predict the graphene-based electrode's specific capacitance. By extracting experimental data from over 200 published studies, they included physicochemical features including the percentages of nitrogen, oxygen, and carbon atoms; electrode configuration; specific surface area (SSA); pore volume; pore size; and the ID/IG ratio. Additionally, electrochemical test properties from electrochemical impedance spectroscopy analyses and galvanostatic charge-discharge tests were included. With a coefficient of determination (R^2) of 0.88 and a root mean square error (RMSE) of 60.42, the ANN model demonstrated exceptional accuracy. Further analysis revealed that doping levels of nitrogen and oxygen significantly influenced the model's predictions. Similarly, Mishra et al. (2022) used ML algorithms to evaluate the impact of various carbon-based material's physicochemical properties on the capacitive performance of electric double-layer capacitors. A total of 4,899 entries were extracted from 147 references and considered features like current density, pore volume, pore size, defect presence, potential window, SSA, and oxygen and nitrogen content. Categorical variables like testing methods, electrolytes, and carbon structures were also analyzed. SSA, nitrogen doping, and potential window were found to be important descriptors for particular capacitance by the extreme gradient boosting model, which had the best association among the five regression models. In addition, Jamaluddin et al. (2023b) investigated the key factors of graphene properties influencing supercapacitor performance using ML models. Four algorithms were evaluated: Random Forest (RF), Decision Table (DT), lazy IBK, and Linear Regression (LR). The RF model exhibited the highest correlation value of 0.745, indicating its effectiveness in predicting capacitance. The study also highlighted that graphene's porous structure and large specific surface area lead to enhanced capacitance values. Recent studies have continued to advance this frontier. For example, Liao et al. (2024) employed ML to optimize N-doped biochar, while Tawfik et al. (2024) implemented a deep learning framework to predict specific capacitance from raw electrochemical parameters. Similarly, Bi et al. (2024) explored feature importance ranking in supercapacitor datasets, improving interpretability and material selection strategies.

This study aims to use ML to predict graphene-based supercapacitors effectiveness, with emphasis on how various physicochemical properties influence specific capacitance. By integrating experimental data with ML models, we seek to uncover critical factors that enhance energy storage capabilities, thereby contributing to the development of more efficient and durable supercapacitors.

2.0 Materials and Methods

Figure 1. present the flowchart that outlines the methodologies employed in the development and evaluation of predictive models for the graphene-based supercapacitors.

2.1 Data Collection

Three ML algorithms were implemented to forecast graphene-based supercapacitors' specific capacitance, energy, and power densities: A multiple independent variable's linear relation with a dependent variable is modeled using multiple linear regression (MLR). To increase prediction accuracy and reduce overfitting, random forest regression (RFR) was used. RFR is an ensemble technique that constructs multiple decision trees. With Gradient Boosting Regression (GBR), complex datasets are handled effectively by building models one after the other, each of which corrects errors made by previous ones. Each model was trained using the collected dataset, with hyperparameters optimized through cross-validation techniques to ensure robust performance.

A total of 510 data entries were compiled from peer-reviewed literature, covering both graphene-only and graphene-composite electrode materials. The dataset includes physicochemical properties (e.g., annealing temperature, surface area, pore volume) and electrochemical performance metrics (e.g., specific capacitance, energy density, and power density). All data processing and model development were carried out using Python 3.9, utilizing libraries such as Scikit-learn for machine learning, Pandas and NumPy for data manipulation, Excel and Matplotlib for visualization.

To ensure data suitability for modelling, the pre-processing steps undertaken include data cleaning, where units were standardized, and missing values for parameters like annealing temperature, energy density, and power density were imputed using mean or median values, as appropriate. Secondly, categorical encoding was used, where hot encoding was used to transform categorical variables (electrolyte type and composite

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material) into numerical representations. To aid in model validation, datasets were split into training (80%) and testing (20%) subsets during the train-test split.

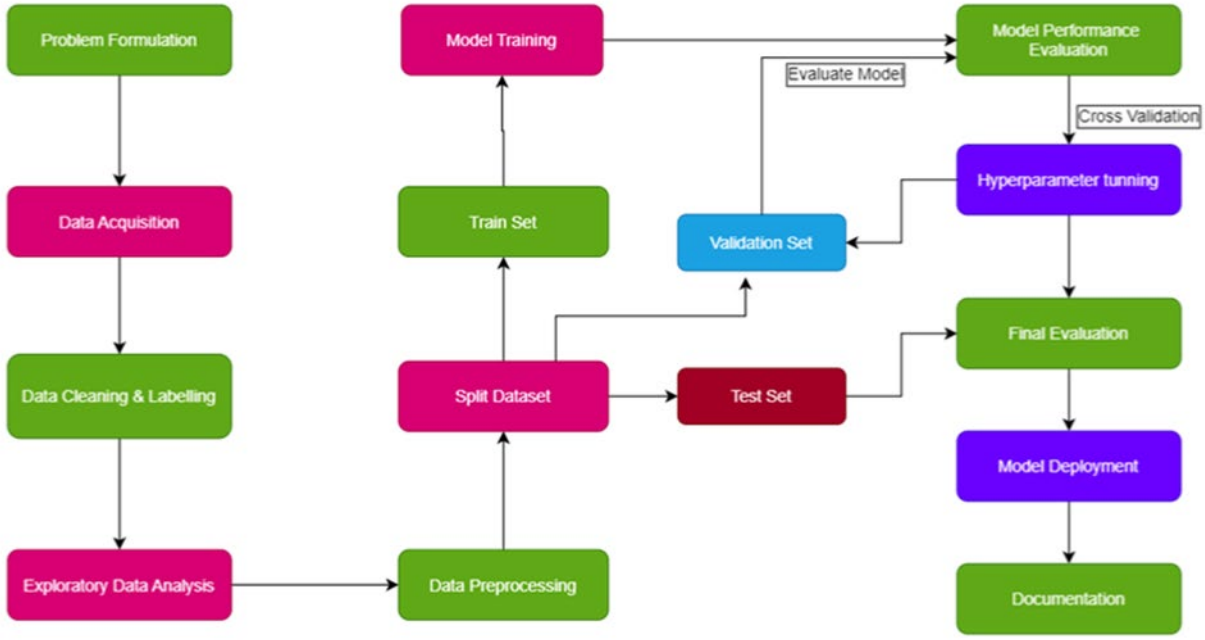

Figure 1: Flowchart summarizing the method

2.2 Model Training and Validation

Each of the three regression models MLR, RFR, GBR, was trained using the pre-processed dataset. MLR assumes a linear relationship between input features and target output. RFR constructs an ensemble of decision trees using bootstrap aggregation and outputs the mean of individual predictions. The model was configured with 100 trees and a maximum depth of 10, determined via grid search cross-validation. GBR builds an ensemble in a stage-wise fashion, where each successive tree corrects errors made by the previous one. Learning rate and number of estimators were optimized using a 5-fold cross-validation.

The dataset was randomly split into 80% for training and 20% for testing. A 5-fold cross-validation strategy was used for model selection and hyperparameter optimization to avoid overfitting. For further validation, model predictions were compared against experimental values from the literature not included in the training set. To validate the generalizability of the developed models, they were evaluated on a different validation dataset that was not utilized during training. The performance of the models on this dataset was compared to their training performance to detect any signs of overfitting. Furthermore, the model's predictions were bench marked against experimental results from recent studies to evaluate their practical applicability.

2.3 Performance Evaluation

The models' predictive accuracy was evaluated using the coefficient of determination (R^2), which indicates the proportion of variance in the target variable that can be predicted from the input features, and the root mean square error (RMSE), which measures the average size of the prediction errors. The R^2 score is defined as:

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}{\sum_{i=1}^{n}(y_i - \bar{y}_i)^2}$$

Where y_i is the actual value, $\hat{y}_i$ is the predicted value, and $\bar{y}_i$ is the mean of actual values.
The RMSE is computed as:

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}$$

These metrics were computed for each target variable, specific capacitance, energy density, and power density on both training and test datasets to evaluate prediction accuracy and generalizability.

3.0 Results and Discussion

The results include dataset distribution analysis, model performance evaluation, correlation between input and output parameters, and a comparative analysis of predicted and actual values.The dataset was

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categorized into two groups: graphene-only supercapacitors and graphene-composite supercapacitors. The annealing temperature for graphene-only supercapacitors varied between 80°C and 637°C. Current density ranged from 0.05 A/g to 1.84 A/g, while the specific capacitance values spanned 67.9 F/g to 226.28 F/g. The highest recorded energy and power densities were 53.06 Wh/kg and 4078.14 W/kg, respectively. For graphene-composite supercapacitors, the annealing temperature ranged from 60°C to 200°C. Current density had a broader range, from 0.10 A/g to 1.98 A/g, while the specific capacitance values varied from 36.25 F/g to 649.73 F/g. The energy density peaked at 55.00 Wh/kg, whereas the maximum power density reached 3245.27 W/kg. These variations highlight the influence of composite materials in enhancing supercapacitor performance.

Table 1. Summary of the performance evaluation for graphene-only and graphene composite samples, for each model.

Models	R^2			MSE		
	Specific Capacitance	Energy Density	Power Density	Specific Capacitance	Energy Density	Power Density
Graphene-Only						
MLR	0.7	0.90	0.5	5.10	1.29	2.36
RFR	0.6	0.40	0.6	1.53	4.09	2.19
GBR	0.9	0.7	0.8	1.03	5.05	2.30
Graphene Composite						
MLR	0.6	0.7	0.5	73.9	21.1	33.0
RFR	0.8	0.6	0.6	59.1	25.2	28.2
GBR	0.8	0.8	0.7	56.8	24.4	30.7

The mean squared error (MSE) and coefficient of determination (R^2) were utilized to evaluated the predictive capabilities of gradient boosting regression (GBR), multiple linear regression (MLR), and random forest regression (RFR). The results for both datasets are summarized in Table 1. Among the models tested, GBR exhibited the best predictive performance with the highest R^2 values (0.9, 0.7, and 0.8) and the lowest

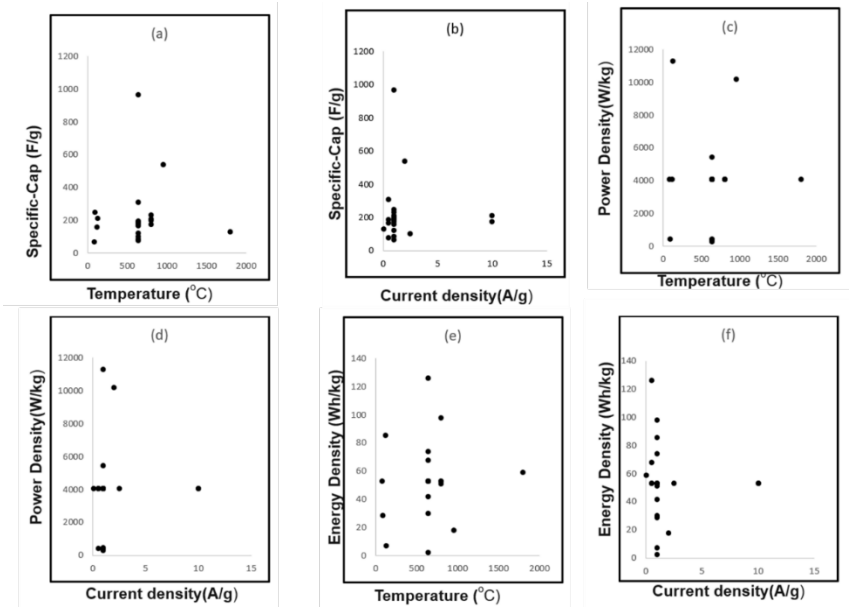

Figure 2. The correlation between input and output parameters for Graphene Supercapacitor; (a)Temperature vs Specific-cap (b)Current density vs specific-cap (c) Temperature vs power density (d) current density vs power density (e) Temperature vs energy density (f) current density vs energy density

MSE (1.03, 5.05, and 2.30) for specific capacitance, energy density, and power density, respectively. MLR showed moderate performance, whereas RFR had lower accuracy in this dataset. In the composite dataset, GBR and RFR exhibited comparable performances, with R^2 values above 0.7 for most parameters. The MSE values of GBR (56.8, 24.4, and 30.7) were slightly lower than those of RFR (59.1, 25.2, and 28.2), confirming that GBR provided more stable predictions. The random forest regression model emerged as the most reliable

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predictor across both datasets, demonstrating high R^2 values and the lowest MSE values. The results affirm the robustness of ensemble learning techniques for predicting complex electrochemical properties.

A correlation analysis was performed to evaluate the connections between the output parameters, including energy density, power density, and specific capacitance, and the important input factors, such as annealing temperature and current density. The correlations are visualized in Figure 2 and Figure 3. A positive correlation was observed between annealing temperature and specific capacitance, particularly in the graphene-only dataset. This suggests that controlled thermal treatment enhances electrochemical performance. Conversely, no clear trend was observed between annealing temperature and power density, indicating that power density may be more influenced by other factors such as electrolyte composition. An inverse correlation was found between current density and specific capacitance, meaning higher current densities tend to reduce specific capacitance. Power density, however, showed a positive correlation with current density, confirming that higher charge/discharge rates lead to enhanced power delivery.

While the GBR model achieved the highest accuracy in terms of R^2 and MSE during training and testing, the RFR model showed greater robustness in generalization, especially when predicting experimental values not included in the training set. This difference can be attributed to the learning strategies of the two models: GBR builds sequential learners where each model attempts to correct the residuals of the previous one, making it highly optimized for the training set, but potentially sensitive to noise or overfitting when faced with unseen data. In contrast, RFR, as a parallel ensemble method, averages predictions across multiple uncorrelated trees, which leads to better generalization, especially when the dataset contains outliers or noisy features.

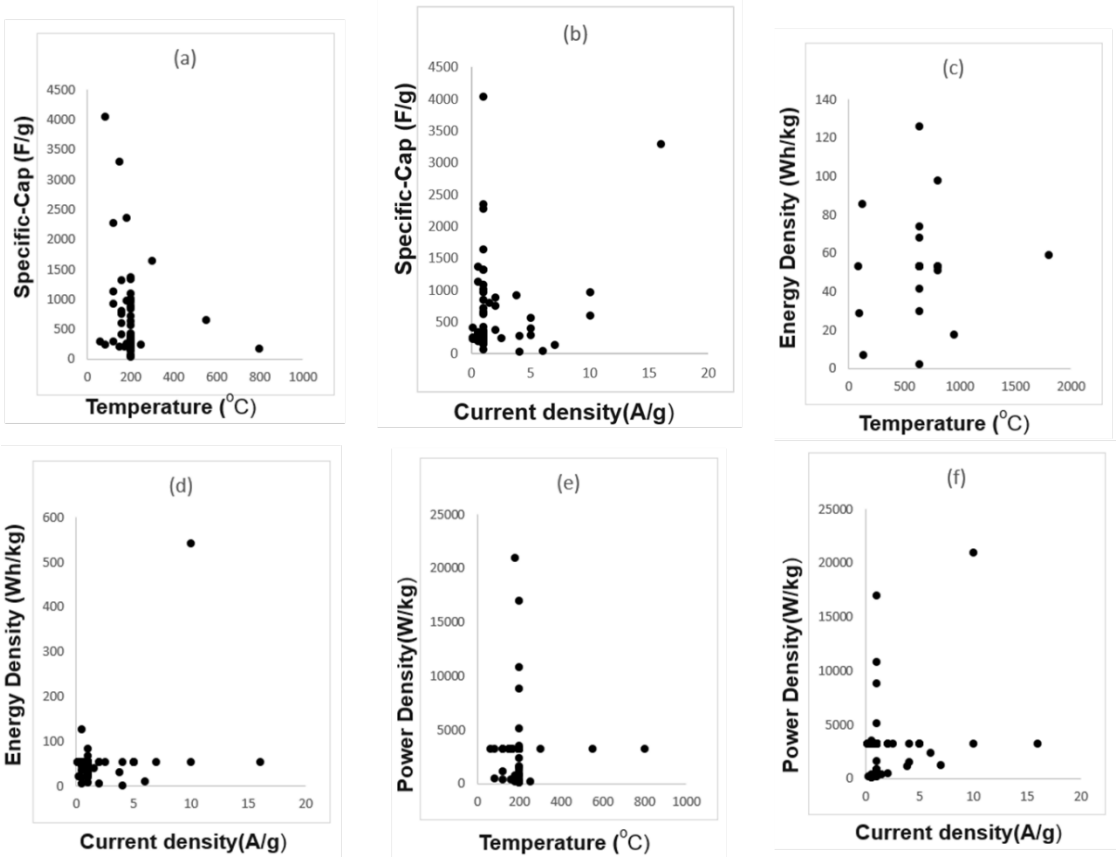

Figure 3. The correlation between input and output parameters for Graphene with Composite Supercapacitor; (a)Temperature vs Specific-cap (b)Current density vs specific-cap (c) Temperature vs energy density (d) current density vs energy density (e) Temperature vs power density (f) current density vs power density

The observed correlation trends are consistent with established electrochemical principles. For instance, the positive correlation between annealing temperature and specific capacitance aligns with the fact that higher thermal treatment can improve material crystallinity, reduce structural defects, and enhance electron transport, all of which improve double-layer formation and pseudocapacitive behavior. Likewise, the inverse relationship between current density and specific capacitance is well-documented in supercapacitor literature: at high current densities, there is insufficient time for ion diffusion into micropores, leading to reduced charge storage efficiency. On the other hand, the positive correlation between current density and power density

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reflects the definition of power density, which favors faster charge-discharge kinetics. These trends confirm that the ML models not only learn patterns from data but also reflect physically meaningful relationships inherent in the electrochemical behavior of graphene-based systems. This concordance enhances the interpretability and credibility of the ML predictions.

To determine the effectiveness of the ML models, predicted output values were compared to the actual experimental results for both graphene-only and graphene-composite supercapacitors. The Random Forest Regression (RFR) model, which has been found to be the most accurate predictor, was used for comparison. Figures 4 present bar charts illustrating the discrepancies between the actual and predicted values for energy density, power density, and specific capacitance. The predicted specific capacitance closely follows the real values, though minor deviations occur, particularly for Sample 3 and Sample 5. The energy density predictions show slight underestimations in the lower range but align well at higher values. Power density predictions remain consistent, with minimal deviations across the dataset, confirming the robustness of the RFR model. The specific capacitance predictions for composite supercapacitors exhibit a strong correlation with experimental values, particularly in the mid-range samples. Energy density predictions demonstrate a slight overestimation at lower values, likely due to non-linearity in material behavior. The power density predictions exhibit higher precision, reinforcing the effectiveness of the RFR model for composite materials. Overall, the results show that the model can generalize across a variety of material compositions while highlighting the need for further optimization for extreme values.

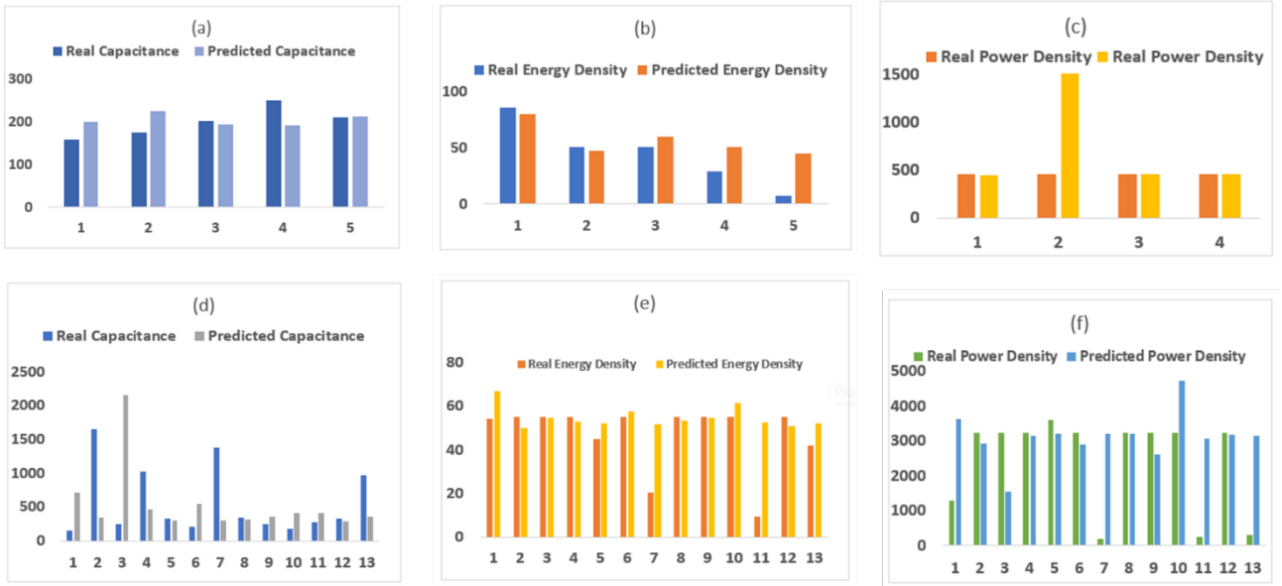

Figure 4. The comparison between predicted and real output parameters for Graphene Supercapacitor using Random Forest Regression; (a) Capacitance (b) energy density (c) Power density. The comparison between predicted and real output parameters for Graphene with composite Supercapacitor using Random Forest Regression; (a) Capacitance (b) energy density (c) Power density

To further validate the reliability of the random forest regression model, a cross-validation analysis was conducted by comparing predicted values with published experimental data. The ability of the model to predict specific capacitance, power density, and energy density was examined across multiple test samples. A radar chart (Figure 5a) was used to compare predicted and experimental specific capacitance values for ten sample points at different current densities. The Random Forest model accurately captured the general trend across the dataset, with minor deviations at Sample 2 and Sample 8. Slight overestimation and underestimation at specific points indicate that feature refinement could further improve predictions. A bar chart (Figure 5b) comparing experimental and predicted power density values at different current densities confirms that predictions align closely with experimental data at moderate current densities. At higher current densities (10 A/g), the model slightly overestimates power density, suggesting a need for further training on extreme cases. A 3D bar chart (Figure 5c) comparing experimental and predicted energy density values across different current densities reveals a good degree of agreement between predicted and actual energy density values throughout the majority of the data points. Underestimations at lower current densities, but improved accuracy at higher current densities, which reinforces the model's predictive strength. The cross-validation results confirm that the random forest regression model predicts supercapacitor performance quite well with minimal errors. However, some discrepancies at extreme values suggest a need for feature engineering improvements, larger datasets, and hybrid ML approaches.

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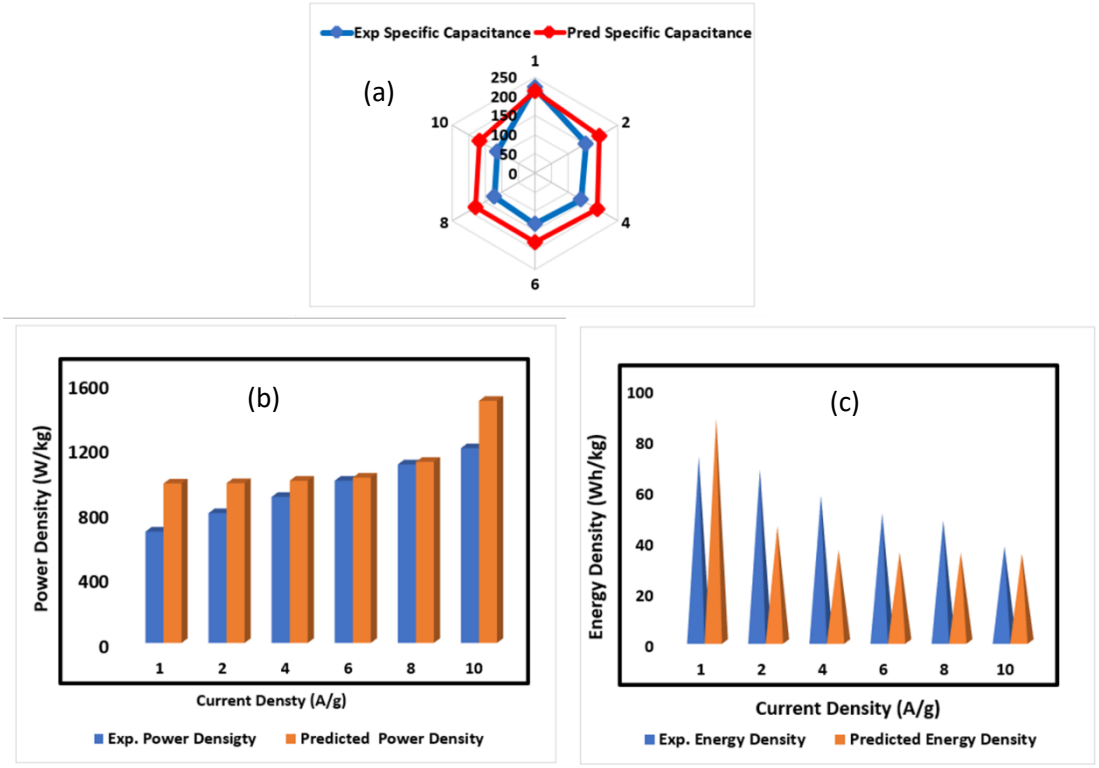

Figure 5. Comparison of the experimental and predicted (a) specific capacitance (b) power density (c) energy density, at different current densities.

4.0 Conclusion

This study demonstrates the capability of ML techniques to predict the electrochemical performance of graphene-based supercapacitors using a dataset of experimentally derived physicochemical and test parameters. Among the models tested, Gradient Boosting Regression (GBR) achieved the highest predictive accuracy, while Random Forest Regression (RFR) showed robust generalizability across material types. Correlation analyses further identified annealing temperature and current density as critical parameters influencing performance metrics such as specific capacitance and power density. Beyond predictive accuracy, the practical implications of this work are significant. The ML models can serve as computational tools for pre-screening materials and synthesis conditions, reducing the need for resource-intensive trial-and-error experiments. This enables a more efficient design of graphene-based composites for real-world energy storage applications. However, the study is not without limitations. The dataset size, while sufficient for training basic ML models, may not capture the full diversity of material compositions and synthesis conditions. In addition, class imbalance between graphene-only and composite materials, and between low and high current densities, could affect model generalizability at extreme conditions. To strengthen the applicability of this work, future studies should integrate real-time experimental feedback into model refinement, validate predictions through targeted laboratory synthesis, and expand the dataset with automated data mining from the literature. Furthermore, coupling ML models with first-principles simulations (e.g., DFT) could offer a hybrid approach to uncover mechanistic insights while maintaining predictive power. Overall, this study highlights the transformative role of ML in accelerating the discovery and optimization of graphene-based supercapacitor materials and encourages deeper integration of data-driven methods into experimental workflows for next-generation energy storage systems.

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