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Investigation on the Physicochemical Properties of Blended Algae-Diesel Fuel with Nanoparticles Additive Using Mixture Design Method

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ABSTRACT

Biodiesel from algae sources is one of the most promising alternative fuels to substitute fossil fuels in the future as it has relatively high oil content, besides rapid biomass production. However, in terms of exhaust emissions, the nitrogen (NOx) emission stemming from algae-diesel blends is higher due to the presence of oxygen content. As a solution, one of the approaches is by using the addition of fuel additives, such as nanoparticles as it plays a major role in increasing biodiesel performance and improving the properties of the biodiesel blends. Therefore, this study directly focuses on improving algae-diesel blends by adding cerium oxide as the fuel additive. The main phases involved in this research are the extraction of algae oil, blending process of algae-diesel with cerium oxide and data collection of dynamic viscosity with calorific value. Then, the data collected were used as an input for Design of Expert (DOE) software to obtain the optimum blends for algae-diesel blends with cerium oxide addition. Analysis of variance (ANOVA) was applied to evaluate the properties of the blends and provides equations for the model, wherein the equations involved in this research were dynamic viscosity and calorific value. The equations generated through the software were used to obtain the theoretical value of dynamic viscosity and calorific value. Finally, the theoretical value was further validated by comparing it with the experimental data for each of the blends. The results of experimental and theoretical (predicted) for each blend model showed the percentage errors between the experimental and the calculated values was between 0.19% and 9.51%. Hence, numerically derived equations are considered valid and reliable. In conclusion, the results showed that the optimum blend obtained by the software was significant and complied with the biodiesel standard of EN14213.

Keywords: *algae-diesel; cerium-oxide additive; calorific value; dynamic viscosity; Design of Expert (DOE)*

1. INTRODUCTION

The use of traditional fossil fuels has increased due to the rapid development of the world economy. The transportation and industrial sectors, which are growing at a faster rate, use nearly 88 % of the energy derived from fossil fuels to support their growth [1-2]. This trend has led to the depletion and scarcity of fossil fuel resources, including petroleum oil, coal, and natural gas. As the usage of fossil fuels intensifies, fuel reserves will decrease, resulting in an

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energy crisis and a significant increase in fossil fuel demand and cost. Moreover, the combustion of diesel engines produces pollution that has adverse effects on the atmosphere and human safety [3]. The pollutants generated by diesel engines can have significant impacts on human health, the environment, and contribute to ozone depletion and global warming. Previous studies have confirmed that emissions from diesel engines can lead to respiratory and cardiovascular health problems [4]. To mitigate these drawbacks, the production of renewable alternative fuels that are readily available and internationally suitable has become a crucial solution to address the current issues.

Recognising the reach of energy and related pollution problems through the combustion of fossil fuels and their products has prompted many researchers to explore the possibility and prospect of using renewable energy sources as a replacement for conventional fuels [3]. One of the most promising renewable energy sources is biodiesel. It has become an alternative source of fuel to reduce the use of petroleum oil [4]. Biodiesel serves many benefits, such as high performance in engine and combustion [5]. In terms of its production, biodiesel is commonly produced from a great variety of feed stocks comprising vegetable oils, such as palm, sunflower, soybean, cottonseed, peanut and rapeseed, animal fats, as well as waste oils such as used frying oils [6-7]. The use of biodiesel would make it possible to pursue a compromise between cultivation, economic growth and climate since it is biodegradable and non-toxic, thereby reducing pollution profiles relative to conventional diesel fuel [8].

Amongst the promising feedstock options that could offer feasible options as alternative fuel, algae are known to be the third-generation biofuel feedstock due to high yields and relatively low land demands for growth [9]. Due to the relatively high oil content of algae and rapid biomass production, algae have been known as a potential source of biofuel production, as well as being one of the best alternative fuels for future use in the transportation sector. Algae-derived biofuels have lower environmental impact than crop-derived biofuels and do not compete with food supplies [9-10]. Hence, the production of biodiesel from algae is very promising to replace biofuels derived from land-based crops owing to their rapid growth, which is approximately 7–10 times that of land crops [11]. This proves that biodiesel from algae appears to be a renewable biodiesel capable of completely replacing diesel fuel without adversely affecting the supply of food and other crops [10]. In addition, the use of algae biodiesel as an alternative fuel is gaining high interest from scientists, and researchers. A research found that for all load conditions, the brake specific fuel consumption for biodiesel and algae oil was higher as compared to diesel [12]. Other researchers discovered that the brake thermal efficiency (BTE) for algae biodiesel was slightly lower than the pure diesel due to its higher viscosity and lower vitality stemming from insufficient blending of air with fuel [13]. A study revealed that amongst biodiesel blends, the algae biodiesel blends had higher viscosity, density and normal boiling point than waste cooking oil and cottonseed oil with the same blending ratio. Despite these variations, all related properties of algae biodiesel were found to be within the range of biodiesel specifications ASTM 6751-12 and EN 14214 [9].

Based on the previous explanation, it is shown that algae biodiesel blends have been introduced widely as one of the most promising alternative fuels, providing high efficiency in engine performance and combustion. However, the production of nitrogen oxide (NOx) is higher in biodiesel compared to diesel, thus resulting in adverse impact towards the environment. This problem is due to the high oxygen contents in the biodiesel [14]. To overcome this disadvantages, previous study has reported that the inclusion of fuel additives, such as cerium

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oxide, graphite oxide and aluminium oxide, in the biodiesel helps to alter the fuel properties, including density, sulphur content and volatility which will improve the emissions and combustion of the biodiesel [15]. However, compared to all these fuel additives, cerium oxide is more beneficial as it is known to be the most abundant element in the rare earth family with good thermal stability and redox cycling performance between oxidation conditions [16]. Cerium oxide has indicated a dramatic reduction of carbonaceous particulate matter (PM) pollution and improvement in fuel efficiency by lowering the light–off temperature of carbon soot and encouraging more complete (efficient) fuel combustion [17]. However, very limited research is being done on the physicochemical properties of the algae diesel blends by inclusion of cerium oxides which also included in determining the optimum blend formulation for algae diesel blends with addition of cerium oxide.

Therefore, this study directly focuses on improving algae diesel by adding cerium oxide as the fuel additive. The main phases involved in this research are the extraction of algae oil, blending process of algae-diesel with cerium oxide and data collection of dynamic viscosity with calorific value. Then, the data collected will be used as an input for Design of Expert (DOE) software to obtain the optimum blends for algae-diesel blends with cerium oxide addition. Analysis of variance (ANOVA) was used to evaluate the properties of the blends and provide equations for the model, wherein the equations involved in this research were dynamic viscosity and calorific value. The equations generated through the DOE software will be used to obtain the theoretical value of dynamic viscosity and calorific value. Finally, the theoretical value was further validated by comparing it with the experimental data for each of the blends. The results obtained were also compared to biodiesel standards, which are ASTM D 6751 and EN 14214 as it is crucial to ensure the fuels comply with the available standards and it is a critical factor for the fulfilment of engine requirement.

2. METHODOLOGY

2.1 Algae Oil Extraction

In this research, Algae was collected from a water pond located at the Sekolah Menengah Tun Syed Shah Shahabudin (SOKSEK). The extraction of algae oil was carried out at the Faculty of Chemical Engineering UiTM Penang by using the Soxhlet extractor. The soxhlet extraction method was employed to extract oil from the algae plant as it is the most efficient method used to extract oil from plant-based material. Figure 1 shows the setup used for the soxhlet extraction method. The solvent used for this extraction method was ethanol with 99.9% purity.

After extracting oil from the algae, residual solvent contained in the oil was removed by using a rotary evaporator as shown in Figure 2. A collecting flask was clamped onto the condenser to collect the remaining ethanol in the algae oil. The mixture of algae oil and ethanol after soxhlet extraction was poured into the rotating flask and later connected to the motor to rotate. The heating water bath was filled with water until it reached above the level of the mixture in the rotating flask. The temperature for the water bath was set at 60°C, while rotating speed and vacuum pump pressure was set at 135 rpm and 427 mbar, respectively, for 3 h. Then, the algae oil was obtained after undergoing this rotary evaporator process.

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ESTEEM Academic Journal
Vol. 19, March 2023, 79-90

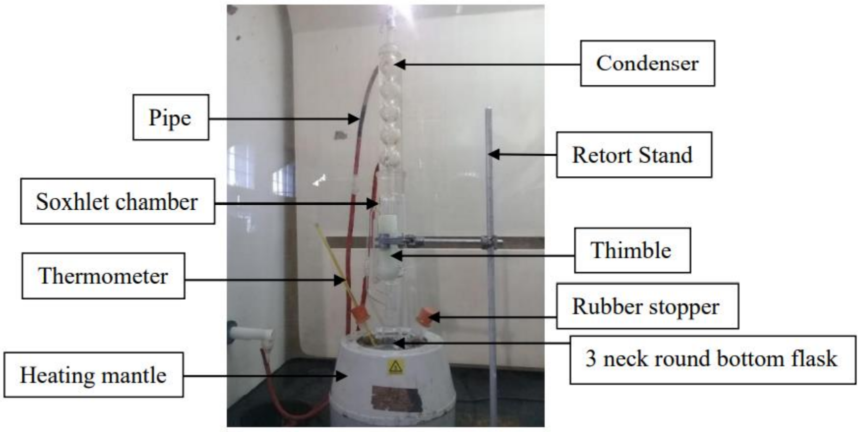

Figure 1**:** The setup equipment for Soxhlet Extraction Method

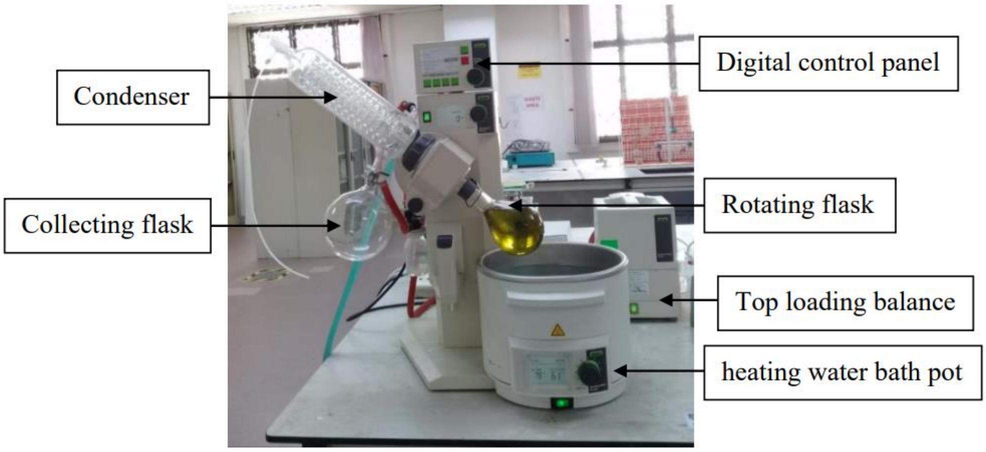

Figure 2: Rotary evaporator

2.2 Formulation of Algae-diesel Fuel with Cerium Oxide Using Design of Expert (DOE) software

The Design of Expert (DOE) software was applied for this research to obtain the optimum algae diesel blends with cerium additive. Generally, this software offers valuable resources to optimise the experiment based on method, mixture or combination of factors and components, besides visualising the results and graph analysis [20].

Firstly, the algae biodiesel fuel blends were prepared according to American Society for Testing Material (ASTM) standards which are ASTM D6751 and ASTM D7467. The low limit and high limit for both algae and diesel were fixed according to the range specified in the Design of Expert (DOE) software. The high limits for algae and diesel were set to 20 % and 100 %, respectively, hence the software adjusted 0 % for algae and 80 % for diesel automatically as the low limits. Next, the experimental run was carried out to determine the dynamic viscosity and calorific value based on different percentages of fuel blends designed by the DOE software. This step was necessary to seek the best and most suitable blend to be mixed with cerium oxide.

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The formulation for algae-diesel fuel blend which was obtained from the software then added with cerium oxide. The high limit set for cerium oxide was 10%, while algae-diesel was 90%. After going through the experimental run, the blending percentage of algae-diesel and cerium oxide designed by the Design of Expert (DOE) software was obtained as shown in Table 1. Total of 8 blends has been suggested by the DOE software. In addition, the data of dynamic viscosity and calorific value for the fuel will be employed as the input for DOE software to optimise the blend of algae-diesel and cerium oxide.

Table 1: Blending Percentage of Algae-diesel with Cerium Oxide

Blend	Algae-diesel (%)	Cerium Oxide (%)
1	100.0	0
2	100.0	0
3	97.5	2.5
4	95.0	5.0
5	95.0	5.0
6	92.5	7.5
7	90.0	10.0
8	90.0	10.0

2.3 Blending process of algae-diesel with cerium oxide

The mixing process was carried out according to the experimental design by the Design Expert (DOE) software. Table 2 shows the percentage blends of algae-diesel and cerium oxide suggested by the software with its nomenclature. The procedure on blending is first by mixing the diesel and algae oil blend into a beaker together with a magnet stirrer and blend continuously for about 15 min at room temperature. Then, cerium oxide was added into the fuel blends and mixed for about 3 h in the fume chamber with the magnetic stirrer with the temperature set to 90 °C as suggested by the lab instructor. It is also crucial for the cerium oxide to be weighed prior to adding into the fuel blends.

Table 2: Volume Percentage Blends of Algae-diesel and Cerium Oxide with Its Nomenclature

Fuel Blends (vol. %)		Nomenclature
Algae-Diesel	Cerium Oxide	
100	0	B100
100	0	B100
97.5	2.5	B97.5C2.5
95	5	B95C5
95	5	B95C5
92.5	7.5	B92.5C7.5
90	10	B90C10
90	10	B90C10

2.4 Fuel Properties Characterization

The fuel properties of B100, B97.5C2.5, B92.5C7.5, B95C5, and B90C10 were determined according to the American Society for Testing Material (ASTM) standard which are ASTM D6751 and ASTM D7467. All the experiments were conducted in the lab facilities of Faculty of Chemical Engineering UiTM Shah Alam and similar equipment were also used by other

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researchers to determine the fuel properties [18]. The calorific value was determined by using the IKA bomb calorimeter C200 which manufactured by IKA, UK, and dynamic viscosity was determined by Physica MCR 301 Rheometer (Anton Paar, UK).

2.5 Analysis of variance (ANOVA)

The DOE software provided tools for conducting statistical analysis, including analysis of variance (ANOVA) by using linear models, summary statistics, coefficients, and equations. In this study, both dynamic viscosity and calorific value were analysed by using ANOVA. ANOVA was used to identify the most significant model for the research, based on F-value and p-value, and described model variances. The fuel properties were evaluated by using these values as the input data into the software. The summary statistics provided descriptive statistics, such as standard deviation, mean, coefficient of variation (CV %), predicted R-squared, adjusted R-squared, and adequate precision values, to determine the effectiveness of the model. The coefficients section identified the confidence interval for estimated model coefficients, which helps to identify and address issues within the analysis. The ANOVA also provided equations for the model, which in this research include dynamic viscosity and calorific value. These equations generated through the DOE software will be used to obtain the theoretical values of dynamic viscosity and calorific value. Finally, the theoretical values will be compared with the experimental data for each blend to validate them.

3. RESULTS AND DISCUSSIONS

3.1 Formulation and Development of Algae-diesel with Cerium Oxide

3.1.1 Physicochemical properties of blend: algae-diesel

The physicochemical properties of algae-diesel (B100) and pure diesel (D100) are recorded in Table 3. Based on the data obtained from the experiment, the dynamic viscosity of B100 was lower than that of D100. This showed a positive side in terms of the quality of fuel atomisation. The lower the fuel viscosity, the higher the volatility. Thus, this will also affect the spray characteristics of the fuel in the engine. As for the calorific value, B100 had a lower value than the pure diesel but higher than EN 14241 standard, of which the minimum value for this standard is 35000 J/g.

Table 3: Physicochemical Properties of Algae-diesel (B100) and Pure Diesel (D100)

No	Parameter	B100	D100	EN14241	ASTM D6751	EN 14213
1	Dynamic Viscosity (Pa.s)	0.122	2.9785	-	-	-
2	Calorific Value (J/g)	42061	42232	35000	-	Min 35000

3.1.2 Physicochemical properties of algae-diesel with cerium oxide blend

Table 4 shows the data on dynamic viscosity and calorific value obtained from eight different blending ratios of algae-diesel alone and algae-diesel with cerium. The blend ratios generated by the Design of Expert (DOE) software v11 were B100, B97.5C2.5, B95C5, B92.5C7.5, and B90C10 as listed in Table 4; B represents algae-diesel, while C represents cerium oxide. From the experimental run, it was found that the dynamic viscosity of fuel blend increased after adding cerium oxide into the algae-diesel. This proved that the addition of nanoparticles into biodiesel increases the resistance of the fluid layer, hence increases the dynamic viscosity. The

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ESTEEM Academic Journal
Vol. 19, March 2023, 79-90

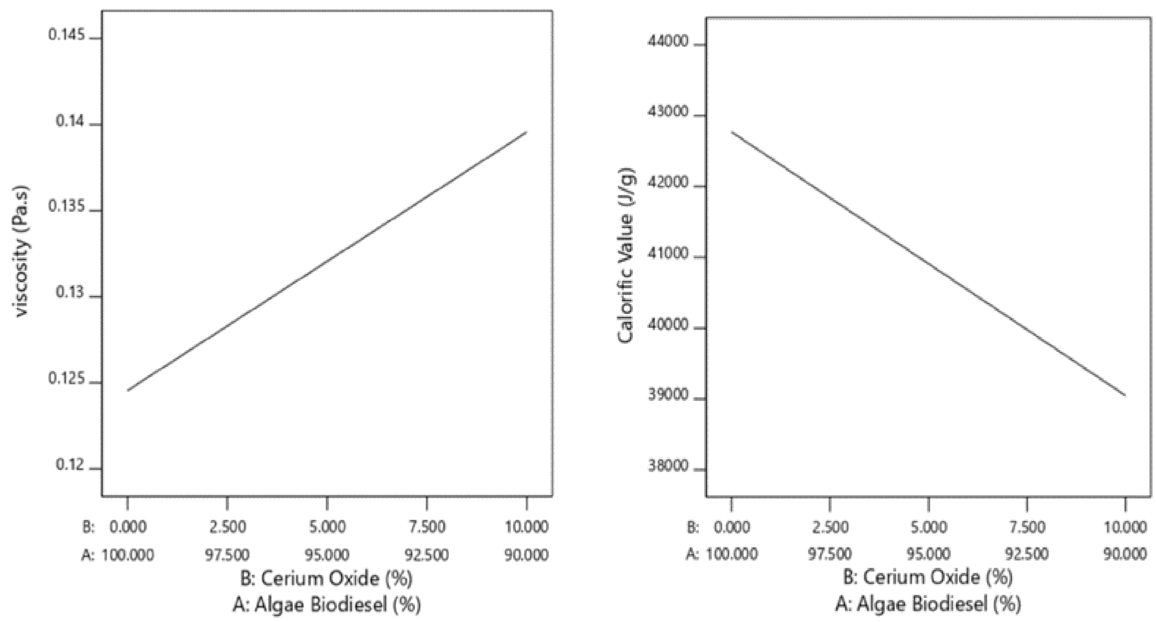
fuel viscosity affects the hydrocarbon emissions as a result of insufficient lubrication of fuel injection pumps, thereby reducing the combustion performance of the fuel [14].

Table 4: Blend Ratios of Algae-diesel Fuel with Cerium

Parameter	B100	B100	B97.5C2.5	B95C5	B95C5	B92.5C7.5	B90C10	B90C10
Dynamic Viscosity (Pa.s)	0.122	0.124	0.141	0.126	0.127	0.134	0.143	0.140
Calorific Value (J/g)	42061	42060	43594	40832	40829	40720	38950	38594

The fuel testing data on calorific value showed an increasing trend with the increasing of cerium oxide percentage added into the algae-diesel. B100 had a calorific value of 42061 J/g, which was higher than that of B90C10 (38594 J/g). Although there is no standard of calorific value stipulated in ASTM D6751 and EN 14214 requirements, a minimum value of 35 MJ/kg has been recommended by EN 14213 (for biodiesel heating purpose) [19]. The calorific value affects the conversion of thermal energy in the fuel, whereby good conversion of thermal energy delivers excellent brake thermal efficiency, hence leading to a better combustion system of the fuel within the engine.

Figure 3 and Figure 4 show the correlation of percentage blends with dynamic viscosity and calorific value respectively in graph form generated by the software. Based on Figure 3, the statistical analysis showed that the addition of cerium oxide into the algae-diesel increased the dynamic viscosity. Meanwhile in Figure 4, the addition of cerium oxide reduced the calorific value of the algae-diesel. However, the value obtained for these two parameters were within the acceptable range and confirmed by the analysis of variance (ANOVA).



(a) (b)

Figure 3: The Model Graph of Percentage Blends Correlates to (a) Dynamic Viscosity, and (b) Calorific Value

p-ISSN 1675-7939; e-ISSN 2289-4934

3.2 Analysis of Variance (ANOVA) of Blending Ratios

Based on the blending ratios, the results of dynamic viscosity and calorific value were evaluated by analysis of variance (ANOVA) through the Design Expert software version 11. The ANOVA for dynamic viscosity and calorific value are shown in Table 5.

Table 5: Summary of ANOVA for Dynamic Viscosity and Calorific Value

Source	Dynamic Viscosity	Calorific Value
Model	Significant (Linear)	Significant (Linear)
F value	6.5	18.47
Prob > F	< 0.0435	< 0.0051
R^2	0.5199	0.7548
Adjusted R^2	0.4399	0.7139
Predicted R^2	0.2754	0.5756
Adequate Precision	4.8068	8.1030
Standard Deviation	0.0062	918.61

Based on Table 5, linear models were significant for the dynamic viscosity and calorific value of algae-diesel with cerium oxide blend. The F values for dynamic viscosity and calorific value were 6.5 and 18.47, respectively. On the other hand, the prob > F or p-value obtained from the ANOVA of algae-diesel with cerium oxide blend for the dynamic viscosity was < 0.0435, while the calorific value had p-value lower than 0.0051. Since both models had values of prob > F less than 0.0500, thus the data obtained for algae-diesel with cerium oxide were significant [20].

From Table 5, the R^2 for dynamic viscosity and calorific value were 0.5199 and 0.7548, respectively. High R^2 values indicated a good relationship between the developed models and the experimental data [20]. The difference between adjusted R^2 and predicted R^2 should be within 0.20 to ensure the accuracy of the data. Based on the analysis, the adjusted R^2 and predicted R^2 for viscosity generated by the software were 0.4399 and 0.2754, respectively. Meanwhile, the calorific value indicated adjusted R^2 and predicted R^2 of 0.7139 and 0.5756, respectively. Both predicted R^2 and adjusted R^2 values for dynamic viscosity and calorific value were in reasonable agreement since their differences were less than 0.2 [20].

Adequate precision measures the signal to noise ratio, whereby it compares the range of predicted values at the design points to the average prediction error. In this case, the ratio greater than 4 is desirable for the model. The adequate precision values for dynamic viscosity and calorific value were 4.8068 and 8.103, respectively; thus, both models can be used to navigate the design space. In summary, ANOVA concluded the most significant and best fit model for this research. The effectiveness of the model based on standard deviation, mean, coefficient of variation (CV%), predicted R-squared, adjusted R-squared and adequate precision were acceptable and within the range.

3.3 Blend Model

3.3.1 Final equation in terms of actual components from design of expert (DOE) software

Equation 1 and Equation 2 were utilised to compute the dynamic viscosity and calorific value generated using the DOE software [20]. These equations were employed to validate the theoretical (predicted) values against the experimental values of dynamic viscosity and calorific

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value obtained from the fuel testing. By utilising these equations, all the predicted values for each blend of algae-diesel with the additives can be generated, saving both cost and time in achieving the optimal blend of the algae-diesel.

$$\text{Viscosity} = 0.001246\ (\text{algae-diesel}) + 0.002745\ (\text{Cerium}) \quad (1)$$

$$\text{Calorific Value} = 427.70889\ (\text{algae-diesel}) + 55.53111\ (\text{Cerium}) \quad (2)$$

3.3.2 Validation of blend model

Table 6 shows the results of experimental and predicted dynamic viscosity and calorific value for each blend model. The experimental results were obtained through fuel testing, while the predicted results were calculated from the equations given by the software. There were slight differences in dynamic viscosity and calorific values obtained from the experimental and the predicted data. Table 6 depicts the percentage errors of 0.1 % - 2.5 % between the experimental and the calculated values. Hence, numerically the derived models were considered valid and reliable [20]. The comparison and percentage error of the viscosity and calorific values between the experimental and predicted data were represented by the graphs shown in Figure 5 and Figure 6.

Table 6: Results of Experimental and Predicted Dynamic Viscosity and Calorific Value for Each Blend Model

Run	Blend Ratio (%)	Experimental		Predicted		Percentage Error (%)	
		Viscosity (Pa.s)	Calorific Value (J/g)	Viscosity (Pa.s)	Calorific Value (J/g)	Viscosity	Calorific Value
1	B100	0.1220	42061	0.1246	42770.9	2.09	1.66
2	B100	0.1240	42060	0.1246	42770.9	0.48	1.66
3	B97.5C2.5	0.1405	43594	0.1283	41840.4	9.51	4.19
4	B95C5	0.1264	40832	0.1321	40910.0	4.31	0.19
5	B95C5	0.1265	40829	0.1321	40910.0	4.24	0.20
6	B92.5C7.5	0.1340	40720	0.1358	39979.6	1.33	1.85
7	B90C10	0.1430	38590	0.1396	39049.1	2.44	1.18
8	B90C10	0.1400	38594	0.1396	39049.1	2.09	1.66

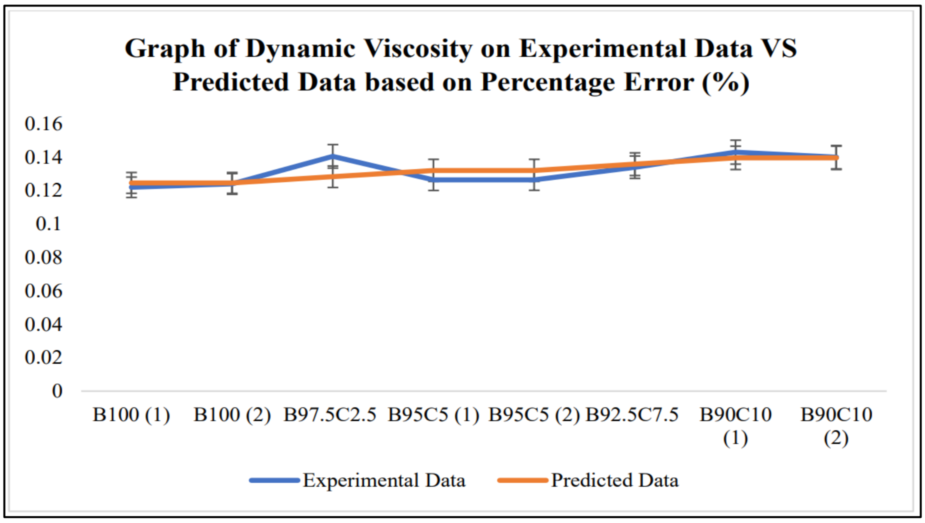

Figure. 5: Graph of Dynamic Viscosity on Experimental Data VS Predicted Data based on Percentage Error (%)

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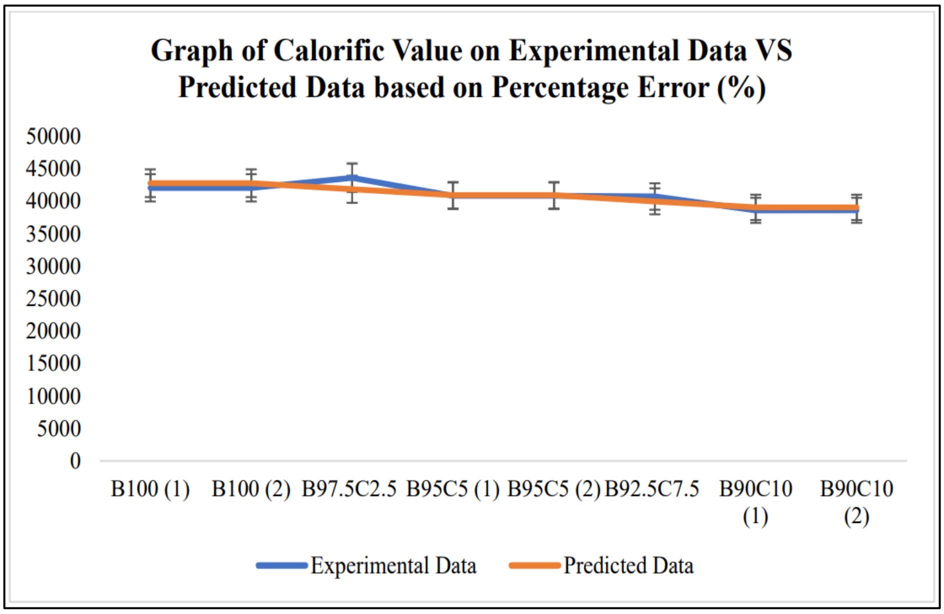

Figure 6: Graph of Calorific Value on Experimental Data VS Predicted Data based on Percentage Error (%)

4. CONCLUSION

From this research, it can be concluded that cerium oxide is one of the most promising fuel additives that can be used to improve the performance of biodiesel. Firstly, for the dynamic viscosity, the addition of cerium oxide as a fuel additive eventually improved the algae-diesel, which reduced the dynamic viscosity as compared to pure diesel. Viscosity generally gives impact on hydrocarbon emissions; hence, offers sufficient lubrication of fuel injection pumps, which improves the combustion performance of the fuel. The analysis of variance (ANOVA) on model blends, as well as the optimum blend generated through the Design of Expert (DOE) software, thus showed that the dynamic viscosity obtained through this research is significant.

Secondly, for the calorific value, the application of cerium oxide into algae-diesel blends had contributed to major reductions in calorific value with the value of 38950 J/g for B90C10 as compared to pure diesel (42232 J/g) and 100% biodiesel (42061 J/g). In general, calorific value affects the thermal energy of the fuel. Effective transfer of thermal energy guarantees good thermal efficiency of the brakes, thereby increasing the combustion of the fuel in the engine. The analysis of variance (ANOVA) on model blends as well as the optimum blend produced by the Design of Expert (DOE) software showed that the calorific value obtained through this research is significant and complied with the biodiesel standard of EN 14213.

The results of experimental and theoretical (predicted) for each blend model showed that there were only slight differences in dynamic viscosity and calorific value. The percentage errors between the experimental and the calculated values was from 0.19% until 9.51% hence, numerically derived equations were considered valid and reliable. In conclusion, the results obtained through the Design of Expert (DOE) software on the analysis of dynamic viscosity and calorific value showed that the addition of cerium oxide as fuel additive had improved the fuel properties of the algae-diesel blends which conformed to the biodiesel standard and within the acceptable range.

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CONFLICT OF INTEREST

We declare that there is no conflict of interest regarding the publication of this paper.

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