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Optimization of Aceh Low-Rank Coal Upgrading Process with Combination of Heating Media to Reduce Water Content through Response Surface Method

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Abstract
This research aims to improve the rank of coal in Aceh, which is known to have a relatively high moisture content of 44-52%. The upgrading process uses hot water and hot oil as media combined with microwaves to remove moisture content in coal. The process was carried out using microwave rotary dryer equipment by varying the coal particle size of 10, 20, and 30 mesh and the time for 20, 40, and 60 minutes. Response surface methodology using the Central Composite Design (CCD) method was used to determine the optimum conditions for low-rank coal, which resulted in 9 runs of low-rank coal experiments. The quadratic polynomial model was validated with the correlation coefficient, R2 value being 0,994. Optimization has been done by maximizing the moisture content of low-rank coal at 4,906 Kg and decreasing it by 20% by setting the particle size and time at 5 Mesh M and 20 minutes, respectively. As a result, this research is projected to be a reference for developing coal upgrading methods in Aceh that can be applied from the laboratory to the commercial scale.

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1. Introduction
Indonesia's coal potential is 85% lignite coal. Aceh is recognized for possessing substantial resources in the mineral extraction sector, particularly coal. According to Wattimena et al. (2019), in West Aceh, approximately 500 million tons of coal reserves are classified as lignite, thus necessitating a process of enhancement or elevation of this lignite coal to medium-rank or high-rank coal (Al Tuffahati, Latif, Arifa, & Mardhatillah, 2023; Umar et al., 2024). This would consequently augment the economic value of coal and optimize its application in the industrial sector (Jannah & Junaidi, 2022).
The upgrading of low-rank coal is typically conducted to diminish moisture content (Sun, Xu, Xin, Xu, & Yang, 2019), volatile matter content, low oxidation temperature, and spontaneous ignition during transit and storage, thereby augmenting the calorific value of the coal (Huang et al., 2022; Mergalimova, Atyaksheva, Ybray, & Seitzhapparov, 2024; Zhang et al., 2022). From examining coal quality parameters in the Meulaboh Basin undertaken by Hanum et al.,

representative low-rank coal data on an as-received (ar) basis for moisture content is acquired in the 44-52% range, constituting a comparatively elevated percentage (Hanum, Hapsauqi, Jamilatun, & Nirmalasari, 2024). Consequently, this investigation was performed to reduce the moisture content of low-rank coal in Aceh and to compute the calorific value of coal after the enhancement process while also contrasting several upgrading methodologies employing diverse techniques so that they can serve as a reference in the advancement of a more cost-effective upgrading process for the sustainability of coal utilization in the future (Ketabdari, 2016; Putri, 2023).

Various methodologies have been devised for the low-rank coal enhancement procedure. Multiple techniques have been developed to increase the calorific value of low-rank coal by eliminating moisture content. Some methods include using heated air, hot oil (Ohm, Chae, Lim, & Moon, 2012), and superheated vapour (Pusat, Akkoyunlu, & Erdem, 2016) as desiccating mediums. Nevertheless, traditional heating techniques are less productive in terms of temporal and energetic demands. This is because the thermal flux in conventional desiccation systems progresses from the particle surface to the core of the particle, whereas the moisture mass flux transitions from the interior to the particle surface. A novel innovation has been implemented among the traditional methods developed, specifically through microwaves. Microwaves emit electromagnetic radiation with a frequency spanning from 300 MHz to 300 GHz or a 1 to 300 mm wavelength. The energy released from microwaves to coal will induce friction within the coal, generating heat that facilitates water evaporation from the coal (Sardi, Uno, Pasila, Altway, & Mahfud, 2023).

In this research, substandard coal from Aceh was employed as feedstocks for pyrolysis, and the operational parameters were refined utilizing Design Expert software. Consequently, the objective of this study was to enhance the pyrolysis of substandard coal from Aceh through the application of response surface methodology (RSM). Design Expert software was utilized to formulate the central composite design (CCD) experimental framework and to explore the optimal conditions. The statistical interactions among various parameters, including sample size and retention duration concerning moisture content, were examined. The moisture content of the substandard coal from Aceh products, influenced by different heating mediums, was significantly contingent upon the sample size and the retention duration.

This study offers a new approach in the low-rank coal upgrading process in Aceh by combining three heating media, namely hot water, hot oil, and microwaves, which have not been widely studied in previous studies. Unlike conventional methods that only use single heating, this study utilizes a combination of heating media to increase efficiency in reducing water content, which is proven through modeling with Response Surface Methodology (RSM) and optimization using Central Composite Design (CCD). In addition, the use of microwave rotary dryer in this process shows an increase in time and energy efficiency compared to traditional heating methods. The results of this study are expected to be a reference in the development of more efficient coal upgrading methods and have the potential to be applied on an industrial scale, especially in the utilization of coal resources in Aceh.

2. Material and Methods

The selection of particle sizes of 10, 20, and 30 mesh in this study was based on technical considerations and relevance to the low-rank coal upgrading process. These sizes were chosen to explore the effect of particle dimension variations on the efficiency of moisture release during the heating process using a combination of hot water, hot oil, and microwave media. Larger particle sizes (>30 mesh) tend to have lower moisture release rates due to diffusion barriers, while sizes that are too small (<10 mesh) can cause changes in the physical properties of coal that affect process stability and heating efficiency. In addition, this size range has been widely used in previous studies as the optimal size in the coal upgrading process because of the balance between the surface area available for heating and the efficiency of heat transfer in the dehydration process. Thus, the selection of these parameters aims to obtain optimal conditions that can be applied on a laboratory scale and become the basis for future industrial-scale development.

This research was undertaken at the Energy and Resources Laboratory, Department of Chemical Engineering, Syiah Kuala University and the Mechanical Engineering Laboratory, Abulyatama University. Proximate Analyses for low-rank coal can be elucidated in **Table 1**. Low-rank coal specimens were triturated utilizing an iron mortar. The specimens were prepared by sifting through sieves of dimensions 10, 20, and 30 mesh. Furthermore, an aggregate of 100 grams of coal that has undergone refinement is placed into the oven at a temperature of 100°C for 4 hours and subsequently weighed to ascertain the moisture content before the enhancement procedure.

Table 1. Characteristics of low-rank coal Aceh

Coal Sample	Moisture Content (%)	Volatile Matter (%)	Ash (%)	Sulfur (%)	Calorific value (Kcal/Kg)
Subbituminous	45	39	2	0,2	3400-3200

Equipment and Material.

The equipment used in this research is a microwave-stirred dryer **Fig.1**, bomb calorimeter, oven, analytical balance, 10 kg bench scale, iron mortar, and mechanical sieve 5, 10, 15 mesh. The materials used are low-rank coal samples, water, oil, and aluminium foil.

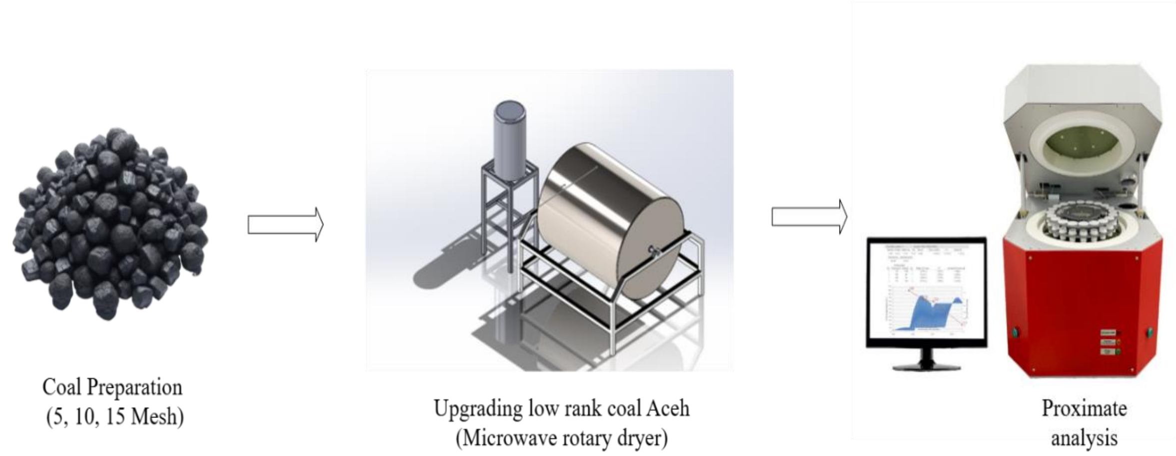


Fig. 1. Experimental Procedure

Experimental Design

RSM was utilized to enhance the optimum value of moisture content pyrolysis of low-rank coal Aceh. It ensured that the correlation between the responses (moisture content) and the quantitative experimental variables (sample mass and residence duration) were scrutinized (Gani et al., 2024; Gani, Adisalamun, et al., 2023; Piepho, Herndl, Pötsch, & Bahn, 2017). The primary objective of employing RSM was to ascertain the factor combinations that would produce optimal responses and illustrate curvatures in the response surfaces (Chen et al., 2022; Mohammed, Khed, & Nuruddin, 2018; Pereira, Milan, & Tapia-Blácido, 2021). Design Expert (Version 13) provided two response surface configurations: Central Composite Designs (CCDs) and Box-Behnken designs, of which CCD was deemed the most appropriate. CCD could fit first-order (linear) and second-degree models with the assistance of the axial and central points. Likewise, design attributes such as orthogonality, curvature and rotatability were evaluated through CCD.

A central composite design (CCD) was employed to optimize the process. This design included a two-level factorial design, centre point experiments, and axial points. The factorial design consisted of nine experimental runs (32), while six centre points (m=6) were replicated three times, and six axial points were added. The axial points were located at a distance α from the centre, where α is greater than 1. The CCD was conducted in multiple blocks to ensure orthogonality and minimize variation in the regression coefficients. This allowed for the independent estimation of factor and block effects. The total number of experiments, N, was calculated using (Eq 1), where n is the number of factors. In this case, with two factors, the total number of experiments was 9 with two different heating media.

$$N = 2^n + 2n + m = 2^2 + 2.2 + 1 = 9 \qquad (1)$$

Low-rank coal Aceh was pyrolyzed under various conditions to decrease moisture content—the sample size from 5 to 15 mesh and the residence time from 20 to 60 minutes. A central composite design (CCD) was developed using Design Expert software. The CCD included five levels for each factor: high (+1), low (-1), centre point (0), and two outer points (±1.414). Table 2 outlines the coded and actual levels of the experimental variables, and Table S1 summarises the coded and experimental variables for the CCD. The response data set, consisting of the experimental results for banana peel waste pyrolysis, is presented in **Table 3**.

Table 2. Coded and Experimental Variables for CCD

Coded Variable	Experiment Variable	Coded level and actual Level				
Hot Water and Hot Oil		-α	-1	0	+1	+α
A	Particle size (Mesh)	2.928	5	10	15	17.071
B	Time (minute)	11.716	20	40	60	68.284

Table 3. CCD order and response for hot oil and hot water of low-rank coal

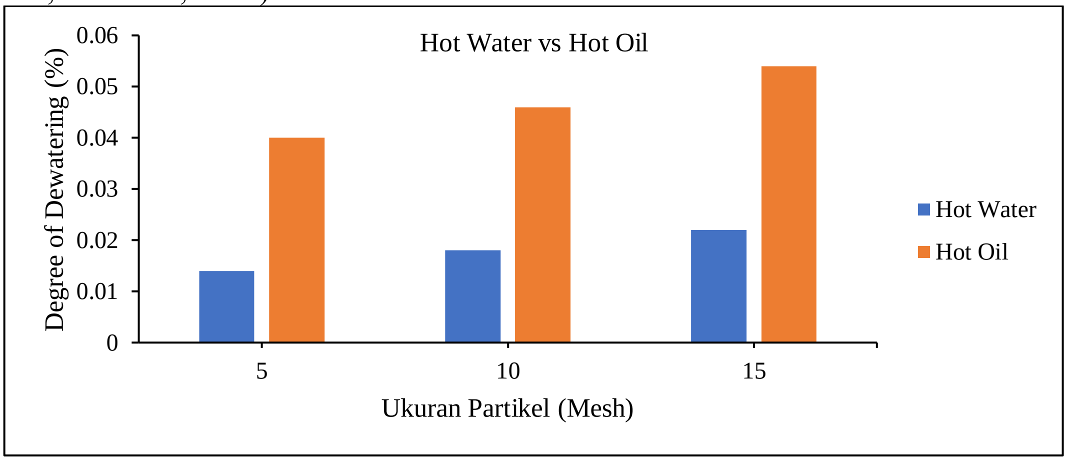
Std	Run	Particle Size (Mesh)	Time (min)	Moisture Content/weight (Kg) Hot Water	Hot Oil
2	1	5	20	5.22	5.19
1	2	5	40	5.15	4,93
3	3	5	60	4.93	4,8
6	4	10	20	5.22	5,17
7	5	10	40	5.1	4,86
8	6	10	60	4.91	4,77
4	7	15	20	5.24	5,12
5	8	15	40	5.07	4,84
9	9	15	60	4.89	4,73

Statistical analysis, including regression modelling, ANOVA, and the generation of response surfaces and contour plots, was conducted using Design Expert 13 software. Significant differences between treatments were considered statistically significant at a p-value of 0.0001 or lower. The optimal conditions for the three variables, particle size (A) and time (B), were determined based on the statistical analysis. Design expert software was also used to fit the developed equations and create the corresponding response surfaces and contour plots.

3. Result & Discussion

Experimental Result

We appreciate the feedback regarding the need for clearer captions in the tables and figures, particularly in **Fig. 2** and **Fig. 4**. To improve clarity, we have updated the caption in **Fig. 2** to more explicitly explain the comparison of the effectiveness of the hot water and hot oil methods in reducing the moisture content of low-rank coal, where the hot oil method shows higher efficiency in dehydration due to its hydrophobic nature that prevents water reabsorption. In addition, **Fig. 4** has been supplemented with a more detailed explanation of the relationship between particle size and heating time on moisture reduction, where the smaller the particle size and the longer the heating time, the greater the moisture reduction that can be achieved. These changes are intended to make it easier for readers to understand the implications of the data presented in the context of coal upgrading process optimization. The comparison between the hot water and hot oil methods can be seen from the percentage of a degree of dewatering, which states the rate of water that can be removed during the process. The degree of dewatering can be calculated using the following formula (Prakoso & Sano, 2022; Yuliansyah, Kumagai, Hirajima, & Sasaki, 2020).



$$Degree\ of\ Dewatering = \frac{M_{Raw\ Coal} - M_{Upgraded\ Coal}}{M_{Raw\ Coal}}\ x\ 100\% \quad (2)$$

Fig. 2. Comparison of Coal Upgrading Results from Variations of Hot Water and Hot Oil Methods

In **Fig. 2**, the hot oil method gives a higher percentage than the hot water method. This is because the oil can press and coat the coal part, which causes the coal to become hydrophobic, thus preventing water from reabsorbing (Gani, Erdiwansyah, et al., 2023). The percentage of a degree of dewatering obtained in the hot water method with a particle size of 5 <10 <15 mesh is 0.014, 0.018, and 0.022%, respectively. In the hot oil method with the same particle size, respectively, it is 0.04, 0.046, and 0.054%.

Statistical Regression Analysis

The models delineating the influence of various parameters on the moisture content indices of low-rank coal from Aceh are illustrated in **Table 4**. Equation (3-6) quantitatively depicts the fluctuations in the moisture content of the low-rank coal from Aceh as influenced by the alterations in particle size (A) and time (B). The Model F-value of 81.39 implies the model is significant. There is only a 0.01% chance that an F-value this large could occur due to noise. P-values less than 0.0500 indicate model terms are essential. In this case, tim (B) is a significant model term. Values greater than 0.1000 indicate the model terms are not significant. If there are many insignificant model terms (not counting those required to support hierarchy), model

reduction may improve your model. The Predicted R^2 of 0.9170 is in reasonable agreement with the Adjusted R^2 of 0.9526; i.e. the difference is less than 0.2.
The ANOVA findings, consequently, statistically validate the framework to depict the influence of various factors on the moisture content of the low-rank coal in Aceh. Likewise, the ANOVA evaluations of the models formulated to quantitatively elucidate the impacts of particle dimensions and duration on moisture content are statistically significant with acceptable regression coefficients and sufficient precision values, as demonstrated in **Table 4**.
The mathematical representation of encoded variables can be utilized to formulate forecasts regarding the outcome for specified magnitudes of each variable. By convention, the elevated magnitudes of the variables are encoded as +1, and the diminished magnitudes are encoded as -1. The encoded mathematical representation is advantageous for discerning the comparative influence of the variables through an analysis of the variable coefficients (Sen & Sen, 2023). The mathematical representation of actual variables can be employed to formulate forecasts regarding the outcome for the specified magnitudes of each variable. Herein, the magnitudes should be delineated in each variable's original units. This representation should not be employed to ascertain the comparative influence of each variable due to the coefficients being normalized to accommodate the units of each variable and the intercept not being positioned at the centroid of the design space (Putra et al., 2024). **Fig. 3** illustrates the distribution of experimental vs model-predicted moisture content distributed around the perfect prediction line (dotted line, RSM = experimental).

Table 4. Analysis of variance

Source	SS	DF	MS	F-Value	Prob>F
			Hot Water		
Model	0.1521	2	0.0760	81.39	<0.0001 (Significant)
A-Particle Size	0.0017	1	0.0017	1.78	0.2301
B-Time	0.1504	1	0.00154	161.00	<0.0001
Residual	0.0056	6	0.0009		
			Hot Oil		
Model	0.2559	5	0.0512	215.93	0.0005 (Significant)
A-Particle Size	0.0088	1	0.0088	37.20	0.0089
B-Time	0.2321	1	0.2321	979.03	<0.0001
AB	0.0000	1	0.0000	0.0000	1.000
A^2	5.556×10^{-6}	1	5.556×10^{-6}	0.0234	0.8880
B^2	0.0150	1	0.0150	63.37	0.0041
Residual	0.0007	3	0.0002		

We recognize the importance of providing a more detailed explanation of how regression analysis and ANOVA were used to validate the experimental results. Therefore, we have added a more in-depth discussion regarding the role of these two methods in ensuring the reliability of the predictive model used in the study. Regression analysis was used to identify the relationship between the independent variables (particle size and heating time) and the dependent variable (water content reduction), which were then visualized in a quadratic equation model. Meanwhile, ANOVA was used to test the significance of these factors by comparing the variation between groups and within groups, where the high F value and very small p value (<0.0001) indicate that the model used has high accuracy in predicting the experimental results. In addition, the R^2 value close to 1 (0.994) confirms that the resulting regression model has a very good fit to the experimental data. Thus, this statistical approach ensures that the optimization results obtained are not only accurate but also applicable in a wider range of scenarios, including industrial scale development.

Table 5. Models Representing Moisture Content Analysis of Low-Grade Coal

Response	Model	Coded/Actual	Eq.
Moisture Content Hot Water	$5.08 – 0.0167A – 0.1583B$	Coded	(3)
	$5.4311 – 0.00333A – 0.007917B$	Actual	(4)
Moisture Content Hot Oil	$4.88 – 0.0383A – 0.1967B + 0.0017A^2 + 0.0867B^2$	Coded	(5)
	$5.69 – 0.009A – 0.0217B + 2.51 \times 10^{-8} + 0.000067A^2 + 0.000217B^2$	Actual	(6)

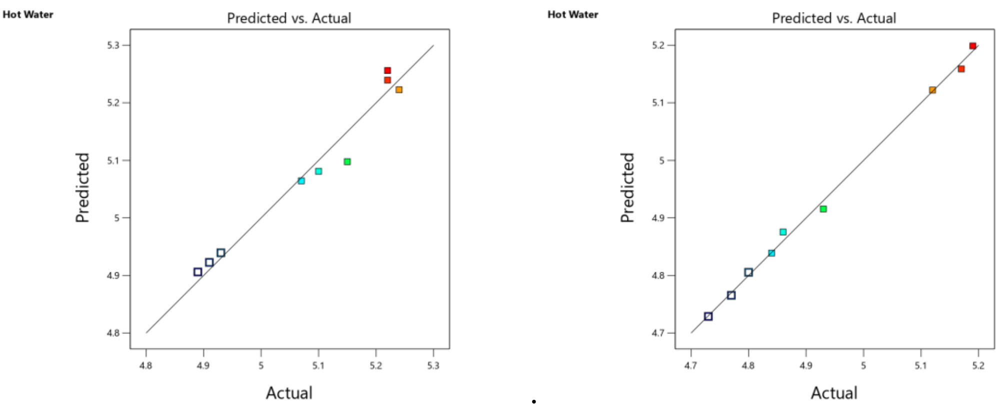


Fig. 3. Experimental vs RSM model predicted values of moisture content. The dotted lines (RSM = Experimental) pass from the origin and represent the perfect prediction.

Factor Variations and Effect on Low-Rank Coal Aceh Model

The contour and response surface plots showed how the different factors of the slow pyrolysis process interact. The 3D surface and contour plots helped categorize the surface projections for the studied variables. The correlation between the particle size and moisture level while maintaining the retention duration constant was investigated, as well as the interplay between moisture level and retention duration while sustaining the specimen at a constant partcle size (Deng, Wang, Zhang, & Luo, 2021), as larger particles facilitate heat transfer and drying. Fig. 4 shows that an augmentation in particle size over a reduced duration for low-quality coal Aceh facilitates the extraction of moisture level (Rego et al., 2022) and that extending the duration at minimal particle size yields analogous outcomes. The modelling of these interactions suggests that optimizing these parameters can significantly improve the efficiency of the pyrolysis process (Wang et al., 2024).

In the context of this study, "hot oil" refers to a heating method using hot oil as a heat transfer medium, which functions to increase the efficiency of water evaporation by creating a hydrophobic layer on the coal surface, thereby reducing the possibility of water reabsorption after the heating process. Meanwhile, "hot water" refers to heating with hot water that works through a convection mechanism to evaporate water in coal. Both methods are used in combination with microwaves to increase the effectiveness of the dehydration process. With this explanation, we hope that the terminology used in the article will be more consistent and clearer in various discussion contexts.

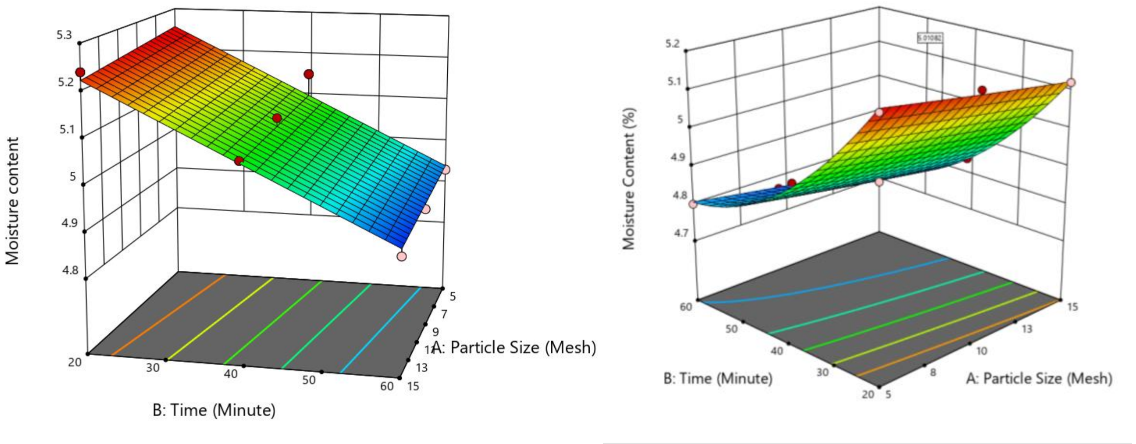
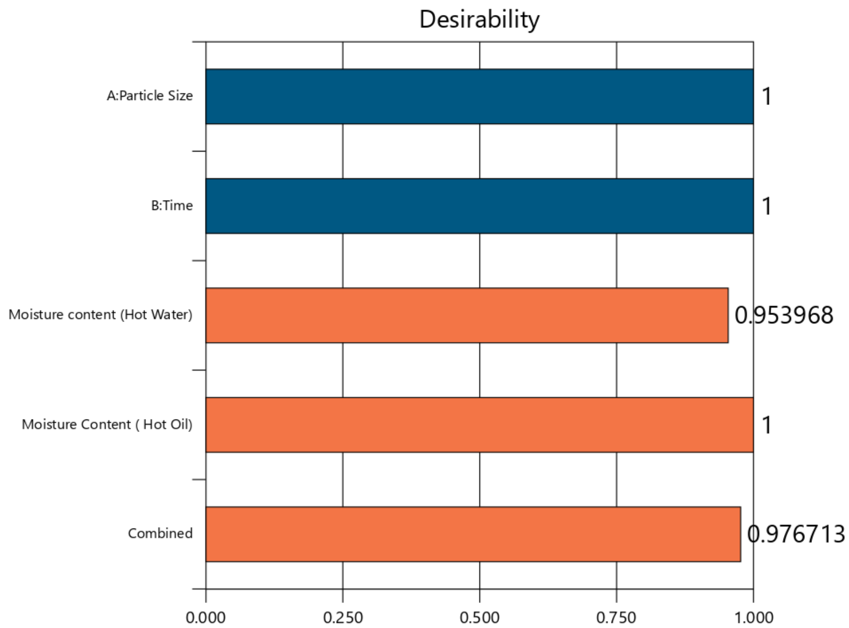


Fig. 4. The moisture content response surface contour and 3-D plots for the effect size particle (mesh) and time (min) on low-rank coal Aceh

Optimization Analysis

A comprehensive resolution of the optimal variable configurations that would produce the minimal response, their collective desirability, and a response forecast was derived from Design Expert software. **Fig. 5** delineates the optimal global solutions for the low-rank coal Aceh specimens, signifying disparate responses from the targeted optimisation of the targeted variable. Utilizing the global solution variables, confirmatory pyrolysis experiments were executed. For example, the derived optimal solutions from the design expert software indicated the minimal response of moisture content at 4.906 in hot water and 4.729 in hot oil to transpire at a particle size of 15 mesh and a duration of 60 minutes. This was substantiated by the confirmation response yields, which demonstrated the optimal conditions for diminishing moisture content at the global solution variables as 4.906 and 4.729 in distinct heating media. Consequently, these findings corroborated that the predicted values from the optimal global



solutions are robust and viable models for optimization, as evidenced by minimal error margins (lower standard deviations) between the predicted and the actual values.

Fig. 5. Response Optimization Parameters for low-rank coal Aceh

4. Conclusion

Response surface methodology (RSM) constitutes an advantageous instrument for modelling, upscaling, and optimizing the alteration in the moisture content of low-rank coal from Aceh, affecting particle size and duration variations. It was noted that the moisture content of the low-grade coal from Aceh exhibits significant fluctuations concerning the diverse variables, including particle size and duration. The moisture content of the low-rank coal increased from 5 to 4.906 in hot water and 4.729 in hot oil (%). The upgrading process has been developed and has reduced the water content in low-rank coal, although not significantly. However, this study is the basis for determining further variables for further research.

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