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Hashimu Hamisi[1,2,*], **Yusufu Abeid Chande Jande**[2,3] **and Askwar Hilonga**[2,3]

[1] Department of Civil Engineering, Mbeya University of Science and Technology, PO Box 131, Mbeya, Tanzania
[2] School of Materials, Energy, Water, and Environmental Sciences (MEWES), The Nelson Mandela African Institution of Science and Technology, PO Box 447, Arusha, Tanzania
[3] Water Infrastructure and Sustainable Energy Futures (WISE-Futures) Centre of Excellence, Nelson Mandela African Institution of Science and Technology, PO Box 447, Arusha, Tanzania
[*] Author to whom any correspondence should be addressed.

E-mail: hashimuh@nm-aist.ac.tz, hashimu.mzava@gmail.com, yusufu.jande@nm-aist.ac.tz **and** askwar.hilonga@nm-aist.ac.tz

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Abstract

Multi-parameter effect consideration during metakaolin conversion gives the best optimum processing conditions. Kaolin deposits have different properties, which makes it vital to establish optimum conditions for a distinctive kaolin deposit. The Response Surface Methodology under the Box-Behnken Design has been adopted in the current study to optimize the processing conditions for kaolin-to-metakaolin conversion. The temperature of 765 °C, the rate of 10 °C min^{-1}, and 6.46 h of soaking were the optimum conditions for yielding the highly reactive (1382.15 mg Ca $(OH)_2$/g) metakaolin. The kaolin sample's distinctive reflections on two theta degrees at 14.32° and 29.03° proved that kaolinite was present. However, x-ray diffraction suggested that the optimized metakaolin was lacking those reflections. The Fourier-transformation infrared spectroscopy confirmed the presence of kaolinite in the sample with wave number range at the bands 3692, 3650, and 3620 $cm^{-1,}$ which disappeared to form a broad band in this region, which validates the formation of reactive amorphous metakaolin.

1. Introduction

Portland cement is one of the most commonly used materials in the construction industry. Its production involves carbon dioxide emissions during raw material transportation and clinker formation. The world's carbon dioxide contribution from cement production is approximately 8%–9% [1, 2]. However, using pozzolan to replace cement in cement-based materials like geopolymer concrete reduces global carbon dioxide emissions and energy demand [3]. Aluminosilicate-rich materials, like metakaolin, are the primary source of geopolymer mortar and concrete. Other aluminosilicate sources include mine tailings, fly ash, blast furnace slag, incinerator bottom ash, ladle slag, metallurgical slag, ceramic waste, high magnesium nickel slag, and demolition and building debris [4]. Due to its inherent silica and alumina, metakaolin has been exploited as a mineral additive in Portland cement concrete to improve mechanical qualities and durability [5, 6]. The ingredients' characteristics will significantly influence the final properties of the end product. In that instance, producing the most effective metakaolin is crucial because it will generate the best metakaolin-based geopolymer. Kaolin and other aluminosilicates have little or no cementitious value but can be modified to become reactive.

A high level of amorphousness, a strong pozzolanic reactivity, and a specific surface area define the ideal metakaolin for geopolymer concrete [7, 8]. The preparation of metakaolin involves the calcination of kaolin, which produces highly reactive metakaolin. Amorphous aluminosilicate ($Al_2O_3.SiO_2$) is left behind after removing structurally bound water in kaolinite ($Al_2O_3.SiO_2.2H_2O$) through calcination [9–11]. The calcination temperature for producing the reactive metakaolin varies from 500 to 900 °C, subject to the kaolinite source [12, 13]. Overheating leads to recrystallization, which reduces the pozzolanic reactivity of metakaolin [9, 14, 15].

Of course, the final properties of the calcined kaolin depend on the processing conditions, including calcination temperature, rate, and holding time [7, 16, 17].

The effect of heating rate on the metakaolin employed in geopolymer concrete production was from 1 °C–20 °C min^{-1} at a constant temperature of 700 °C and duration of 30 min [17], and it was found that the best properties were obtained at a low heating rate. Also, *Elimbi et al* [18] premeditated the effect of calcination temperature at a rate of 5 °C min^{-1} and a holding time of 10 h in a temperature range of 500 °C–800 °C in 50 °C increments. They found 700 °C to give the best metakaolin properties. A study on the influence of the degree of de-hydroxylation on the pozzolanic activity of metakaolin at a temperature range of 500 °C–850 °C at a time interval of 30 min to 15 h concluded that prolonged heating above 5 h resulted in a reduction of the pozzolanic reactivity of the produced metakaolin [19].

Another study on calcined kaolin from the Perak state of Malaysia found that a calcination temperature of 800 °C for 3 h was enough to convert kaolin to highly reactive metakaolin [16]. Also, a study on four kaolin deposits in the western region of Turkey concluded that the optimum calcination condition was at 850 °C for 3 h [20]. Several previous studies have also reported a 2-hour calcination time as the optimum soaking time to convert kaolin to highly reactive metakaolin at a temperature above 600 °C [11, 21]. This conclusion contradicts the study by *Mehsas et al* [22] that studied two metakaolin samples from Algeria and concluded that at any calcination temperature, a soaking time of 2 h was insufficient to convert the kaolin to the reactive phase. Table 1 summarizes the metakaolin processing condition from the previous studies.

It is well observed that kaolin from different sources would result in various properties, and the optimum processing conditions may also differ. Therefore, it is vital to establish the optimum processing conditions for a specific source of kaolin. The best way to achieve the critical optimum parameter is to use multi-parameter effect consideration rather than one factor at a time, which has been used for most reported works [11, 14, 17, 18, 23, 24]. The Response Surface Method (RSM) is one of the most widely used experimental approaches for optimisation. The RSM optimisation technique associates statistical and mathematical procedures for experimental design, model fitting, and regression analysis, thereby assessing the impacts of various factors and their interactions on one or more response variables [25]. This study aimed to evaluate the optimal processing conditions of metakaolin along Pugu Hill kaolin deposits for producing highly reactive metakaolin for geopolymer concrete production. This process was achieved by simultaneously considering the multi-parametric effect by varying three factors (heating rate, soaking time, and temperature) with the RSM-Box-Behnken-Design (BBD) technique to maximise pozzolanic reactivity.

2. Materials and methods

2.1. Raw materials

The kaolin used in the present study was sampled from a natural deposit in Pugu, Tanzania (6° 55' S; 39° 2' 54' E; and 215m altitude). The raw material was sun-dried for a day before being ground. The raw material was oven-dried at 110 ± 5 °C for 24 h and then sieved by a 45 μm sieve. The chemical composition of the kaolin used in this study was more reflected and characterized by silica, alumina, and iron oxide, as shown in table 2. Other oxides were also found to be present at a deficient percentage. It is noted that silicon dioxide, aluminum oxide, and iron oxide account for more than 70% of the material's composition, indicating that it can be employed as a cementitious starting raw material as ASTM C618 [26].

2.2. Optimization by response surface methodology

The response surface methodology (RSM) is a mathematical and statistical technique used in modelling and analyzing a process to optimize a desired response impacted by several input variables [25, 27, 28]. RSM under Box-Behnken, aided by Design-Expert 13 software, was used to design the experiment for studying the effect of calcination temperature, soaking time, and heating rate on maximizing metakaolin's pozzolanic reactivity. Hence, seventeen (17) experimental works with five replicates of the center point run were designed and, at the same time, randomized to ensure no unexplainable results occurred due to the variance of unrelated variables. The independent variables and related responses from the seventeen designed experiments were recorded, as shown in table 3. The table also indicates the residual values to experimental values and the predicted values of the metakaolin's pozzolanic reactivity.

2.3. Calcination process of kaolin

The Lindberg/Blue M (BF51731BC-1) box furnace was used for calcination. The materials were put in a crucible and placed inside the box furnace. The material was calcined to a 650 °C–850 °C temperature array at a 1 °C–19 °C min^{-1} rate for 1–12h. The calcination temperature was set, and the time to reach the designed temperature was calculated depending on the selected rate. Then, when the calcination temperature reached,

Table 1. Various Kaolin to metakaolin conversion parameters.

Reference	Temperature	Rate	Soaking time	Comment
B.B. Kenne Diffo *et al* [17]	700 °C Constantly	1, 2.5,5,10.15 and 20 °C min^{-1}	Constant duration of 30 min	A low heating rate was found to be ideal.
A. Elimbi *et al* [18]	450 °C–800 °C constant at a 50 °C increment.	5 °C min^{-1} constant	Soaking time of 10h constant	700 °C resulted in the best properties.
C. Bich *et al* [19]	500 °C–850 °C	Not stated	30 min—15h	Calcination duration of 5h and below at a temperature above 650 °C brought the best properties.
N. Shafiq *et al* [16]	600 °C–800 °C at a 100 °C increment.	Not stated	1–5h	800 °C for a time of 3h found to be the optimum
Güneyisi *et al* [20]	550 °C–850 °C at a 50 °C increment.	Not stated	Soaking time of 3h constant	750 °C was found to be the optimum

Table 2. Chemical composition.

Component	SiO_2	Al_2O_3	Fe_2O_3	Na_2O	MgO	CaO	K_2O	P_2O_5	MnO	TiO_2
Kaolin (%)	61.5	33.7	3.1	0.08	0.4	0.2	0.27	0.07	0.24	0.44

Table 3. Box-behnken experimental design coding.

Factor code	Factor	Units	Coded minimum	Coded maximum	Center point
A	Temperature	(°C)	$-1 \leftrightarrow 650.00$	$+1 \leftrightarrow 850.00$	750.00
B	Rate	(°C/min)	$-1 \leftrightarrow 1.00$	$+1 \leftrightarrow 19.00$	10.00
C	Soaking time	h	$-1 \leftrightarrow 1.00$	$+1 \leftrightarrow 12.00$	6.50

samples were held for the scheduled soaking time isothermally before letting the furnace cool gradually to ambient temperature. The specimens were subsequently removed from the box furnace and prepared for the necessary examination.

2.4. Pozzolanic reactivity determination

The pozzolanic reactivity of metakaolin was determined following the modified Chappelle test procedure [29]. The 1 g of metakaolin sample and 2 g of calcium oxide were added to a clean, dry Elmer flask. A Teflon stirrer bar and 250 ml of carbon dioxide-free water were introduced to the flask to homogenize its contents. The flask was attached to the reflux apparatus, set on a heating mantle at 90 °C for 16 h, and continuously stirred. After heating, the flask was cooled to room temperature, after which 250 ml of 0.7 M sucrose was added. The flask was then stirred for 30 min, then collected 25 ml aliquots of the sample. The collected aliquot was titrated with 0.1 M HCl using an auto-titrator, and then the fixed mg Ca $(OH)_2$ was calculated.

2.5. Characterization of the optimized Metakaolin

X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FT-IR) examinations were done for the raw and calcined kaolin to confirm the transformation of kaolin to metakaolin. Material for the XRD analysis was prepared using the back-loading technique. Before scanning, the material was micronized with a McCrone micronizing mill. The addition of 20% silicon was done to quantify the amorphous content in the material quickly. A Malvern Panalytical Aeris diffractometer equipped with a PIXcel detector, fixed slits, and Fe-filtered Co-K radiation was used to produce the diffractograms. The phases were recognized with the aid of X'Pert Highscore Plus software. The Rietveld method calculates the relative phase quantities by weight percent. Then, the sample was scanned for FT-IR using the Shimadzu IRSpirit series within the 400–4000 cm^{-1} wave number range to acquire the FT-IR spectrum.

3. Results and discussion

3.1. Response surface analysis

Design expert software generated a quadratic regression model with the experimental results obtained in the kaolin to metakaolin conversion. Equation (1) describes the relationship between the independent variables under study (temperature, rate, and soaking time) and the pozzolanic reactivity, considered the response during the study.

$$\begin{gathered} R = 1,\ 375.19 + 74.5175{*}A + -76.8675{*}B + \\ -35.6925{*}C + 273.797{*}AB + 170.063{*}AC + \\ -104.412{*}BC + -211.688{*}A^2 + -623.893{*}B^2 + -493.218C^2 \end{gathered} \tag{1}$$

Where R is pozzolanic reactivity (mg Ca $(OH)_2$/g), A is Temperature (°C), B is the rate (°C/min), and C is the soaking time (h).

3.2. ANOVA Analysis and model fitting

ANOVA statistically evaluated and validated the derived model fit by testing the significance of the regression model, individual model coefficients, lack of fit, and pure error. The higher F-values and the smaller probability values ($p < 0.05$) indicate more significance for the corresponding coefficients. From table 5 and equation (1), the obtained F-value of 804.67 had a very small p-value ($p < 0.0001$), implying that the model is significant and that there is only a 0.01% chance that an F-value this large could occur due to noise. The terms A, B, C, AB, AC,

Table 4. Independent variables and response.

	Process variables			Response (Pozzolanic Reactivity (mg $Ca(OH)_2/g$))		
Run order	Temperature (°C)	Heating rate (°C/min)	Soaking time (h)	Actual value	Predicted value	Residual
1	650	19	6.5	120.32	114.43	5.89
2	750	19	12	43.12	41.11	2.01
3	750	1	1	264.21	266.22	−2.01
4	750	19	1	332.62	321.31	11.31
5	750	1	12	392.36	403.66	−11.30
6	750	10	6.5	1359.82	1375.19	−15.37
7	650	10	12	382.10	390.01	−7.91
8	850	1	6.5	411.30	417.19	−5.89
9	750	10	6.5	1397.06	1375.19	21.87
10	750	10	6.5	1359.82	1375.19	−15.37
11	850	10	1	618.34	610.43	7.91
12	850	19	6.5	791.84	811.05	−19.21
13	750	10	6.5	1399.42	1375.19	24.23
14	650	10	1	784.32	801.52	−17.20
15	650	1	6.5	834.97	815.75	19.22
16	750	10	6.5	1359.82	1375.19	−15.37
17	850	10	12	896.37	879.17	17.20

Table 5. Analysis of Variance for the response surface reduced to quadratic model.

Source	Sum of squares	Df	Mean square	F-value	p-value	
Model	3.686E + 06	9	4.095E + 05	804.67	<0.0001	significant
A-Temperature	44422.86	1	44422.86	87.29	<0.0001	
B-Rate	47268.90	1	47268.90	92.88	<0.0001	
C-Soaking time	10191.64	1	10191.64	20.03	0.0029	
AB	2.999E + 05	1	2.999E + 05	589.19	<0.0001	
AC	1.157E + 05	1	1.157E + 05	227.31	<0.0001	
BC	43607.88	1	43607.88	85.68	<0.0001	
A^2	1.887E + 05	1	1.887E + 05	370.73	<0.0001	
B^2	1.639E + 06	1	1.639E + 06	3220.25	<0.0001	
C^2	1.024E + 06	1	1.024E + 06	2012.55	<0.0001	
Residual	3562.58	7	508.94			
Lack of Fit	1788.48	3	596.16	1.34	0.3786	not significant
Pure Error	1774.10	4	443.53			
Cor Total	3.689E + 06	16				

Model fit statistics						
R^2	Adjusted R^2	Predicted R^2	Adeq. Precision	Std. Dev.	Mean	CV %
0.9990	0.9978	0.9915	77.1035	22.56	749.87	3.01

BC, A^2, B^2, and C^2 are significant coefficients for the pozzolanic reactivity model in kaolin to metakaolin conversion. The F-value for the lack of fit, which is 1.34, indicates that the lack of fit is not significant compared to the pure error. An enormous lack of fit in the F-value has a 37.86% chance of being caused by noise. A non-significant lack of fit is advantageous because we want the model to fit [30]. The model fit statistics showed that the estimated R-squared is nearly equal to one, which suggests the accuracy of stimulated data [31].

Additionally, the difference between the projected R-squared and the adjusted R-squared was less than 0.2, demonstrating that the regression model suits the data that was generated and will be able to forecast future observations well. Adequacy of precision was used to assess the signal-to-noise ratio, which was found to be bigger than four and was considered desirable [32]. The results imply that using the derived regression model, one can navigate the design space.

In objective optimization, the final purpose is to maximize desirability. As shown in figure 3, the optimization constraints for temperature, rate, and soaking time were used to maximize the produced metakaolin's pozzolanic reactivity. It was found that a temperature of 765 °C, a rate of 10 °C min^{-1}, and a soaking time of 6.46 h resulted in the production of metakaolin with a pozzolanic reactivity of 1382.15 mg Ca

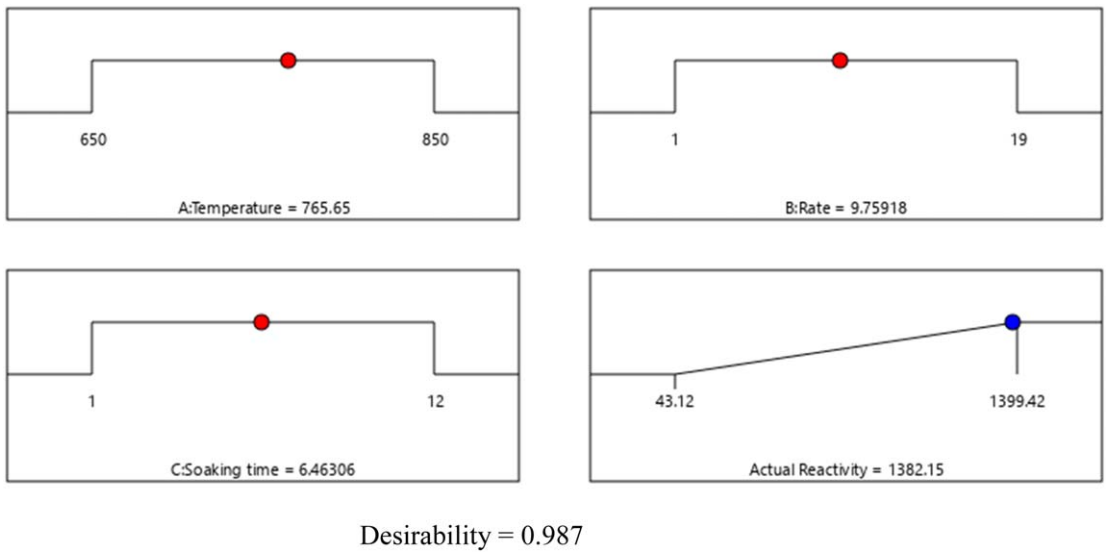

Figure 1. Optimization ramps.

Table 6. Model verification for pozzolanic reactivity maximization.

Temperature (°C)	Rate (°C/min)	Soaking time (h)	Predicted value (mg $Ca(OH)_2$/g)	Experimental value (mg $Ca(OH)_2$/g)	Error (%)
765	10	6.46	1381.65	1351.05	2.22

Table 7. Physical properties.

	Density(g/cm^3)	Blaine Specific Surface area(cm^2/g)	R45 micron (%)
Kaolin	2.632	7208	8.0
Metakaolin	2.747	8909	4.4

$(OH)_2$/g and a desirability of 0.987. The higher the desirability, the greater the reliability of the obtained solution [33]. The optimized parameters are plotted in optimization ramps, as shown in figure 1.

To confirm the model, a confirmation location point was chosen with parameters (temperature of 765 °C, rate of 10 °C min^{-1}, and soaking time of 6.46 h), which gave the predicted mean value for the pozzolanic reactivity of 1381.65 mg Ca $(OH)_2$/g. The recorded pozzolanic reactivity at a prediction point parameter was 1351.05 Ca $(OH)_2$/g. As seen from table 6, the prediction error is less than ten percent, which suggests the high accuracy of the model [34]. The value of pozzolanic reactivity is above 700mg $Ca(OH)_2$/g, which indicates that the produced metakaolin is highly reactive [29].

The interaction effect of the independent factors was analyzed using the contour and three-dimensional plots in figure 2. The 3D response surface plots make it easy to understand how two combined variables impact the measured response. As response surface curvature increases, the significance of the factor relationship becomes more apparent [25]. It is observed that the pozzolanic reactivity increased with an increase in temperature from 650 °C to 765.61 °C and declined beyond that temperature. Considering the soaking time, it is observed that the holding time beyond 6.46h reduced the pozzolanic reactivity of the metakaolin. The decrease in pozzolanic reactivity at a higher temperature may be due to the crystalization of metakaolin [9].

Figure 3 shows a normal probability plot of the residual for the pozzolanic reactivity: the predicted value versus the experimental value of pozzolanic reactivity. The normal distribution of the data for all residual responses is demonstrated by the nearly perfectly straight distribution of the points for all dependent variables. A model's fitness is also analyzed graphically on a plot of actual versus predicted. It is depicted that every point in the plot is quite close to a straight line. The smooth fitting of the points to a straight path shows that the actual versus projected tends to follow established models pretty well and match the data nicely [35].

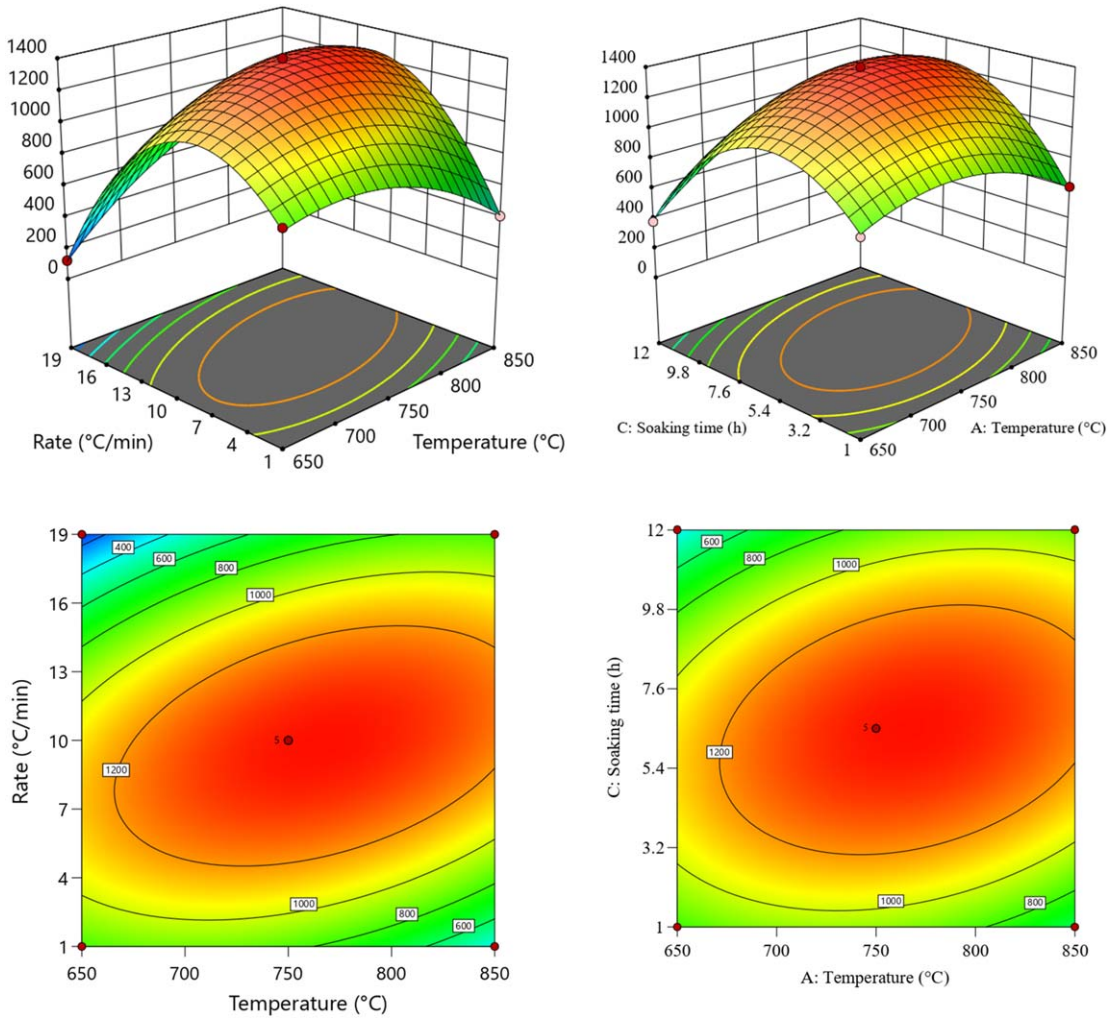

Figure 2. Contour and 3D of the combined effect of temperature and rate, temperature, and soaking time on the pozzolanic reactivity of metakaolin.

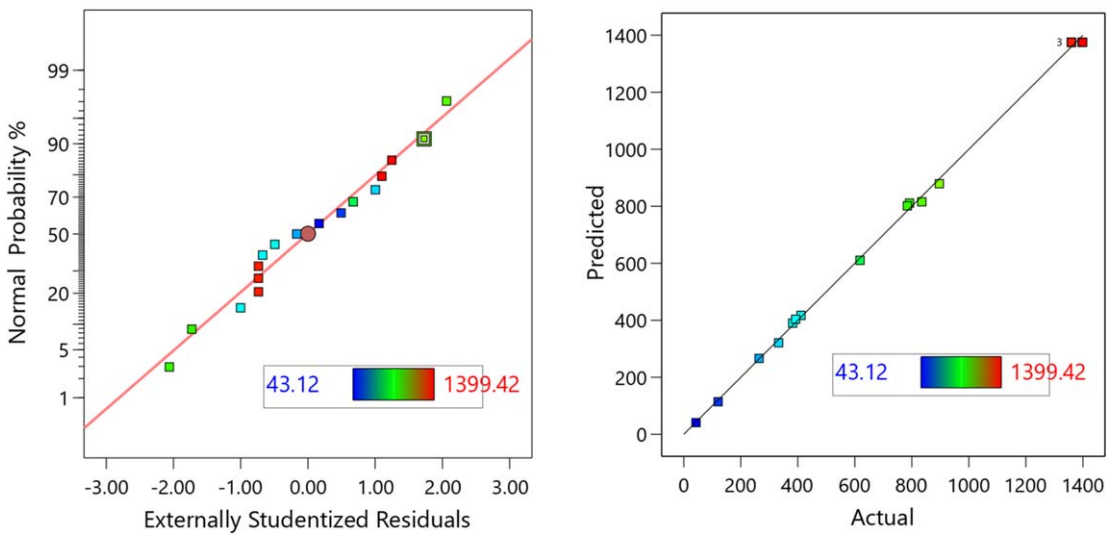

Figure 3. Normal probability plot residual for the pozzolanic reactivity; predicted value versus the experimental value of pozzolanic reactivity.

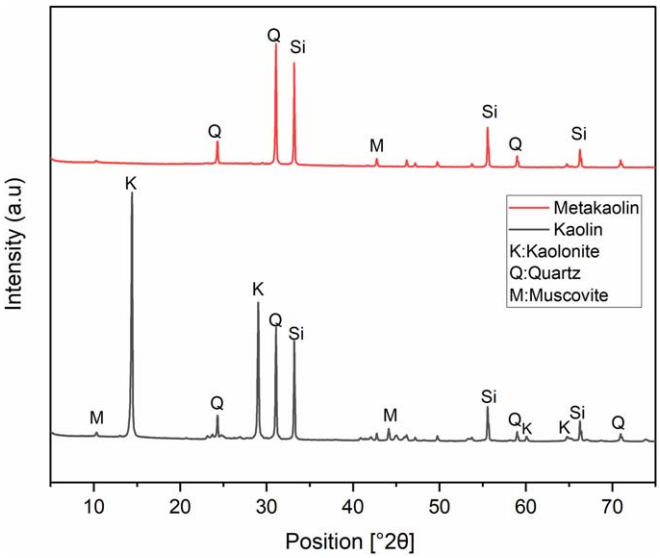

Figure 4. XRD pattern for the raw and calcined kaolin.

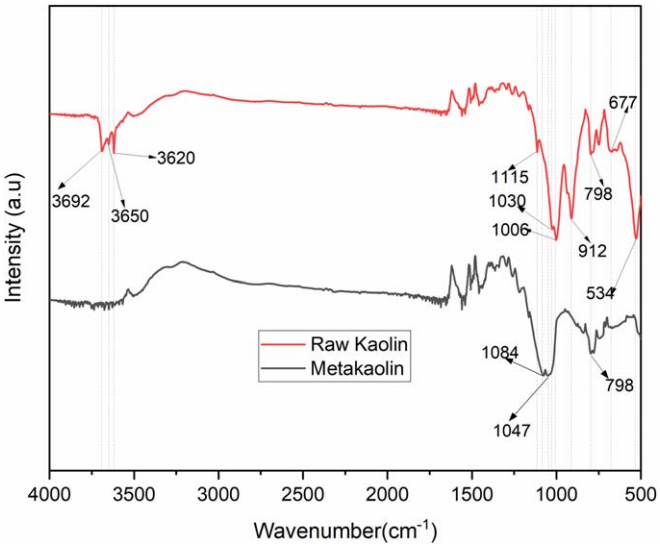

Figure 5. FT-IR spectra for raw kaolin and metakaolin.

3.3. Characterization of the optimized Metakaolin

Figures 4 and 5 show the XRD pattern for the raw and calcined kaolin and the FT-IR spectra for the kaolin and calcined kaolin, respectively. They are all meant to illustrate how kaolinite turns into metakaolin.

By contrasting the XRD patterns for raw and calcined kaolin, as shown in figure 4, the transformation of kaolin to metakaolin was confirmed. The kaolin sample's distinctive reflections on two theta degrees at 14.32° and 29.03° proved that kaolinite was present [19, 36]. X'Pert Highscore Plus software was used to perform Rietveld analysis and quantify the phase composition, and the ICSD-26818 database was used for the matching process. The corresponding identification card for each peak is; Anatase -ICSD: 98-009-6946, Hematite -ICSD: 98-015-4191, kaolinite-ICSD: 98-006-8698, muscovite-ICSD: 98-002-6818, quartz-ICSD: 98-008-3849, and silicon -ICSD: 98-006-0387. The analysis revealed 69.4% kaolinite, 27.4% quartz, and 0.9% muscovite. The observed Si reflections are due to the 20% Si addition added to the sample for amorphousness quantification. The absence of kaolinite-specific reflections in the metakaolin indicates that kaolin has been transformed into metakaolin [36, 37].

The FT-IR spectra in figure 5 were used to confirm the conversion of kaolin to metakaolin. The observed bands at 3692 cm^{-1} and 3650 cm^{-1} are allied with the interlayer hydroxyl group, and the band at 3620 cm^{-1} has an inner OH stretching frequency [36, 38, 39]. The three bands observed are associated with hydroxyl stretching,

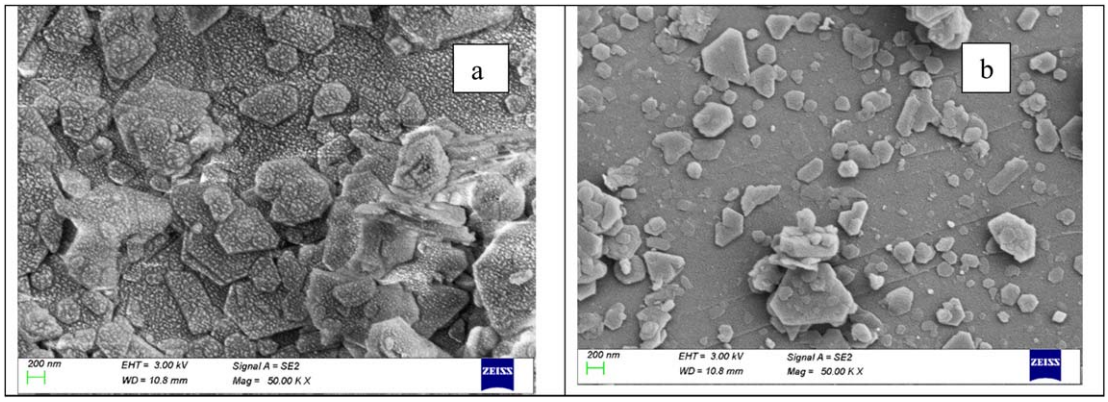

Figure 6. SEM images of (a) Kaolin and (b) Metakaolin.

confirming kaolinite's presence in the raw sample [19, 37, 38]. The observed three peaks define the kaolinite material under study as low-order kaolinite (LOK) [37, 40]. The bands detected at 1115, 1030, and 1006 cm^{-1} are related to the Si-O stretching vibrations band, and the one at 534 cm^{-1} depicts the presence of the Al-Si–O bond within the kaolinite under study. Another reflection at 912 cm^{-1} was attributed to Al-OH bending vibration [37, 39]. Also, the other peaks are found at 798 and 677 cm^{-1}, which are associated with the Si–O–Si stretching of the kaolinite materials [39]. It is well observed that after calcination, the bands at 3692, 3650, and 3620 cm^{-1} disappeared to form a broad band in this region, which validates the formation of reactive amorphous metakaolin [11, 37]. The calcination process leads to the replacement of peaks at 1115, 1030, and 1006 cm^{-1} by broad doublet peaks at 1084 and 1047 $cm^{-1,}$ signifying the transformation of kaolinite to metakaolin [38, 41].

3.4. Morphology and physical properties of produced metakaolin

The physical properties of the produced metakaolin are compared with those of untreated kaolin. Blaine air surface area, which measures the total surface area of the particles per unit mass, is a measure of powder fineness. It is an essential physical property and may impact various application variables. When used in geopolymer concrete, it may affect properties such as pozzolanic reactivity, strength, and durability. A higher surface area can lead to a higher cation degree of reaction between metakaolin and the activating solution and a compact structure, resulting in a more robust and durable geopolymer [42, 43]. As reported in table 7, the optimized metakaolin's Blaine-specific surface area is 8908 cm^2/g. The increased Blaine-specific surface area may be due to reduced particle size after thermal treatment. The decrease in particle size is evidenced in figure 6.

Density is an important property that can affect the behavior and performance of metakaolin in various applications. The higher density recorded in metakaolin (2.747 g cm^{-3}) may be due to the thermal treatment process of kaolin. Dehydroxylation drives the chemically bound water off, decreasing volume and contributing to density increase. The increase in density may result in better particle packing, improving densification, and reduced porosity in the microstructure of metakaolin-based materials.

The R45 microns of the produced metakaolin was found to be 4.4%. The results imply that 95.6% of particles were below 45 microns, and only 4.4% were retained. Generally, a lower R45 microns fraction may indicate a finer particle size distribution, which is desirable for geopolymer mortar application [44].

The morphological characteristic is shown in figure 6. The plate-like in nature images are observed in kaolin (figure 6(a)), which may be due to the layered crystal nature of the kaolinite structure. It is well witnessed (figure 6(b)) that particles lost their plate-like to irregular shape, and some spherical particles, reduced and more uniform particles, are also depicted. These changes in morphology can be attributed to the de-hydroxylation of kaolinite during the calcination process.

4. Conclusions

The influence of calcination temperature, heating rate, and soaking time on maximizing the pozzolanic reactivity of metakaolin has been studied in kaolin-to-metakaolin conversion. The significant findings are summarized as follows:

- Pozzolanic reactivity increases as the temperature rises from 650 °C to 765 °C and then declines as the temperature rises higher.

- The temperature of 765 °C, at a rate of 10 °C min^{-1}, and 6.46 h of soaking were the optimum conditions to yield the highly reactive metakaolin. The pozzolanic reactivity at this optimum condition was 1382.15mg Ca $(OH)_2$/g.
- A chosen confirmation point (temperature 765 °C, rate of 10 °C min^{-1}, and soaking time 6.46 h) resulted in the predicted mean value for the pozzolanic reactivity being 1381.65 mg $Ca(OH)_2$/g. The experimental value was recorded as 1351.05 mg $Ca(OH)_2$/g, which gives an error of 2.22%.
- The quadratic model obtained fits well to predict the response, with a desirability of 0.987.
- SEM images confirmed the particle size reduction after the thermal treatment of kaolin, leading to increased Blaine-specific surface area and pozzolanic reactivity.

Acknowledgments

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Statement of the author

Hashimu Hamisi: Conceptualising, the Method, and Writing: Preparing the First Draft. Yusufu Abeid Chande Jande: Supervision, Writing- Reviewing and Editing. Askwar Hilonga: Supervision, Writing- Reviewing and Editing.

Declaration

There is no competing interest associated with the study.

ORCID iDs

Hashimu Hamisi https://orcid.org/0000-0002-0298-6921

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