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OPTIMIZATION OF COCONUT HUSK AS PORE FORMING AGENT IN POROUS CERAMIC USING RESPONSE SURFACE METHODOLOGY (RSM) STATISTICAL DESIGN

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DECLARATION

I declare that this thesis entitled tittle of the thesis is the results of my own research except as cited in the references.

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Optimization of Coconut Husk as Pore Forming Agent in Porous Ceramic Using Response Surface Methodology (RSM) Statistical Analysis.

ABSTRACT

Coconut husk is the most well-known fibrous waste product from coconut tree cultivation. Coconut husk contains a high concentration of lignin, which makes it exceptionally elastic, durable, and resistant to rotting. Coconut husk was suitable to be as pore-forming agents in porous ceramic. Coconut husk was sieved to 250μm before mixing with the kaolin clay. Porous ceramic material was developed through the incorporation of kaolin clay and coconut husk, then make the composition of coconut husk with 10 wt.%, 20 wt.% and 30 wt.%. The slurry is dried at 100°C for 24 hours before crush it into powder. Lastly, compacted the powder mixture and sintered at 850°C, 900°C and 950°C before we do characterization. The characterization used in this study were physical properties like water absorption, apparent porosity and bulk density, mechanical properties like compressive strength, FTIR for coconut husk and XRD for kaolin clay and porous ceramic. In this study, response surface methodology (RSM) statistical design was applied to optimize the coconut husk added and sintering temperature for the porous ceramic. Statistical analysis of RSM such as Central Composite Design (CCD), includes model adequacy checking, analysis of variance (ANOVA), main effect, interaction plot and contour plot. The factor (experimental parameters) involved were weight percentage of coconut husk (wt.%), whilst the responses (final properties of ceramics) investigated were water absorption, apparent porosity, bulk density and compressive strength. The optimal sintering temperature and composition in the contour plot where water absorption, apparent porosity must be as minimum and bulk density and compressive is at maximum, which shown to be around 900°C with 10 wt.% coconut husks

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LIST OF AMBREVIATIONS

PFA	Pore Forming Agent
CH	Coconut Husk
KC	Kaolin Clay
FTIR	Fourier Transform Infrared Spectroscopy
XRD	X-ray Diffraction
CCD	Central Composite Design
RSM	Response Surface Methodology
ANOVA	Analysis of Variance

LIST OF SYMBOLS

°C	Degree Celsius
%	Percentage
2θ	Diffraction Angle
MPa	Compress Strength
g/cm^3	Bulk Density
Wt.%	Weight Percentage
μm	Micrometer
mm^2	Square Millimeter
N	Newton

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Porous ceramic or known as cellular ceramics began to be developed in the 1970s. Porous ceramics are classified as those ceramics with a high percentage porosity between 20 and 95%. Their also serving temperature varies from room temperature to 1,600 C. These materials consist of at least two phases solid ceramic phases, and the gas-filled porous phase (German et al., 2009c). The gas content of these pores normally regulated itself to the environment possible through pore channels. Porous ceramics are classified according to the nature of porosity, volume fraction and size. The porosity properties of natural ceramics depend on origin, whereas synthetic ceramics depend on how they are made and can usually be controlled (Al-Naib, 2018)

Pore-forming agents are used as a material that forms due to porosity. Pore-forming agent (PFA) in the ceramic was mixed with ceramic powder to create porous ceramics. Due to their abundance and low cost, the banana stem (Sengphet et al., 2013), rice husk (Dele-Afolabi et al., 2022), and potato starch (Eva et al., 2008) have been studied as organic pore-forming agents. Nowadays, researchers tend to use natural pore-forming agents (PFA) which are derived or made from agricultural waste. Agricultural waste is unwanted or unsalable materials produced entirely in agricultural operations directly related to the growing of crops or raising animals for the primary purpose of generating a profit or making a living. Coconut husk waste is agricultural waste, which may be employed in the production of ceramics, and was the pore-forming substance used in the present study.

Previous conducted preliminary study has concluded that the coconut husk can be used as a pore-forming agent for clay-based porous ceramic (Sifa Aminah, 2023). The coconut husk was dried in an oven at about 110°C for 24 hours. Dry coconut husk was processed in a heavy-duty blend and the ground coconut husk was sieved using a 250μm sieve to ensure that only ground coconut husk with particle sizes equal to or less than 250μm were utilized in the clay-based porous ceramic. From the previous study, the water absorption and apparent porosity of porous ceramic incorporated with 30 wt.% of coconut husk is the highest. The highest bulk density of the porous ceramic incorporated is at 0 wt.%. From the previous study, the highest compressive strength of the porous ceramic incorporated the same bulk density at 0 wt.% of coconut husk. Previous conducted preliminary study has evaluated that coconut husk is suitable as pore-forming agent.

Nevertheless, the material that used as a PFA in the clay product will improve the ceramic porosity, thermal conductivity and water absorption which leading to a decrease in the compressive strength of the clay product. The percentage of porosity and compressive strength should be balanced where is associated with the amount of pore-forming agent added to clay prior to burning (Nasr et al., 2020). Therefore, in this study, the optimization of integrated coconut husk powder will be carried out utilizing one of the Response Surface Methodology's statistical experiment designs known as Central Composite Design. The statistical design will aid in optimizing the wt.% coconut husk and sintering temperature added to porous ceramics.

1.2 Problem Statement

Coconut husk can be used or suitable as a pore-forming agent for clay-based porous ceramic has been done by preliminary study (Sifa Aminah, 2023). As the composition of coconut husk grows, adding it to ceramic has resulted in a decrease in compressive strength. It has to do with the fact that the porous ceramic's density fell as a result of having more pores, which decreased compressive strength. Therefore, the composition needs to be balanced with the application of the porous ceramic in terms of the porosity and strength of the ceramic to obtain the optimum properties. To create porous ceramic with the optimum qualities, particularly compressive strength, this additional study will optimize the weight percentage (wt.%) of coconut husk used and the sintering temperature. Hence, the Response Surface Methodology statistical experiment will be used to optimize the composition which is wt.% coconut husk and sintering temperature added to porous ceramics throughout the study.

1.3 Objectives

This study will concentrate on a few objectives:

1) To prepare and characterize the porous ceramic incorporated with coconut husk as pore-forming agent (PFA)
2) To optimize the weight percentage (wt.%) of coconut husk and sintering temperature to physical and mechanical properties porous ceramic using Response Surface Methodology as statistical experimental design.

1.4 Scope of Study

The purpose of this study is to optimize the % addition of coconut husk as a pore-forming agent while also tabulating data using Response Surface Methodology (RSM). The pore-forming agent is used to make the porous ceramic porous. Meanwhile, a study should be performed to improve the compressive strength of porous ceramics. This study will also optimize the body formation of the porous ceramic utilizing a statistical experiment design known as Response Surface Methodology (RSM), which will aid in the creation of the optimal weight & of coconut husk that will be added to the porous ceramic. Finally, X-Ray Diffraction (XRD), and Fourier Transfer Infrared Spectroscopy (FTIR) will be used to complete these characterizations.

1.5 Significance of Study

The key focus of this work is to incorporate coconut husk waste into the production of porous ceramics. Furthermore, the incorporation of coconut husk into porous ceramic is expected to improve the porous ceramic's characteristics. The optimization of coconut husk into porous ceramic is done in this investigation through the application of statistical design. As a result, the purpose of this analysis is to find the best and optimum porosity and weight % of coconut husk, as well as an exceptional compressive strength value that will affect the porous ceramic final product.

CHAPTER 2

LITERATURE REVIEW

2.1 Porous Ceramic

Porous ceramics have recently become increasingly important in the industry due to their various applications such as filters, dust collectors, thermal insulators, hot gas filters, liquid food production, membrane reactors and electronics components. This is because, they exhibit excellent characteristics including high-temperature and chemical corrosion resistance, low thermal conductivity, high surface area and low density. The porous ceramic has been categorized according to pore size and porosity. Porosity in natural ceramics is dictated by their origin, whereas porosity in synthetic ceramics is determined by their manufacturing process and, in most circumstances, may be altered. Pores in porous ceramic can be formed using a variety of techniques, including partial sintering, sacrificial discoloration, replica templates, and direct foaming. The technique is determined based on the final properties of porous ceramics required, like porosity percentage and pore size distribution.

These pores in this material can be classified into three types based on their diameter: microporous (pore diameter less than 2 nm), mesoporous (pore diameter between 2 and 50 nm), and macroporous (pore diameter greater than 50 nm). Mercury intrusion porosimetry is usually measured for pore size distributions (Al-Naib, 2018). Those parameters are essential factors influencing both end porosity and pore connectivity of the porous ceramic. It has been suggested that increasing processing conditions such as heat, sintering temperature, and time reduce porosity %. In general, the particle size of raw ceramic powder should be 2 or 5 times higher than the pore size so that the resulting pore size is optimal (Ohji & Fukushima, 2012).

Porous ceramics should have several common characteristics which as good stability where choosing the appropriate material species and techniques can make porous products suitable for various corrosive conditions in which the product was expected to function. It also should have great specific strength and rigidity. Porous ceramics are not affected by changes in liquid or gas pressure or by other types of stress loadings on their pore size or shape. Thermal stability comes last. Porous products made of heat-resistant ceramics can filtrate molten steel or high-temperature burning gas (Liu et al., 2017).

2.1.1 Properties of Porous Ceramic

Porous ceramics possess a number of suitable properties, which combine the features of ceramics, and porous materials such as low density, lightweight, low thermal conductivity, low dielectric constant, thermal stability, high specific surface area, high specific strength, high permeability, high resistance to chemical attack and high wear resistance. Pore size and porosity percentage are controlled by the particle size distribution of starting ceramic powders, fabrication techniques, types of binder used, concentration of binder, and sintering conditions. Generally, the particle size of raw ceramic powder should be geometrically in the range of two to five times larger than that of pores in order to provide the desired pore size. The porosity percentage decreases with increased making conditions such as pressure, sintering temperature, and time. Furthermore, fabrication influences such as the amount and type of additives, green densities, and sintering conditions (temperature, pressure, atmosphere, etc.) significantly affect the porous ceramic microstructures.

In comparison to polymeric materials, porous ceramics offer several advantages, including thermal stability, chemical and mechanical resistance. During usage, these materials are usually exposed to thermal and mechanical loading stresses (Salvini et al., 2018). Physical properties include density, porosity, water absorption and surface morphology. Mechanical properties include hardness and compressive strength (Susilawati et al., 2020). The effect of sintering temperature on the ceramic properties such as apparent porosity and mechanical strength was investigated, increasing the sintering temperature yielded porous ceramics with decreased porosity and increased mechanical strength (Mouiya et al., 2019). The amount of pore-forming agent used has a significant impact on the porosity and pore size of porous ceramic membranes. So, the properties of the porous ceramic were determined on the type of pore-forming agent that has been added to produce the porous ceramic.

2.1.2 Formulation of Porous Ceramic

Porous ceramic requires two materials: ceramic powder (kaolin clay) and coconut husk as a sacrificial fugitive material that works as a pore-forming agent (PFA). The combination will then be sintered at a specific temperature, resulting in porous ceramic material.

2.2 Kaolin Clay

Kaolin is widely used in the ceramic industry, where its high fusion temperature and white-burning qualities make it particularly suitable for the production of whiteware (China), porcelain, refractories, and building materials These differ in plasticity, crystal and surface chemistry, particle shape and size, flow properties, permeability, and other characteristics. The absence of iron, alkali, or alkaline earth in kaolinite's molecular structure results in the creation of these desired ceramic properties. The consequence has been a significant concentration on developing low-cost porous ceramics, with clay serving as the porous ceramic material of choice, particularly for construction applications such as bricks and tiles. Kaolin is usually mixed with almost equal amounts of silica and feldspar, as well as a slightly lower proportion of ball clay, a flexible light-burning clay used in the production of porcelain. These elements are essential for vitrification, shrinkage, plasticity, and a variety of other processes. Many applications for kaolin clay have been developed, particularly in ceramic manufacture. Kaolin clay properties significant to industrial uses include particle-size distribution, surface area, structural order disorder or crystallinity, and whiteness (Obada et al., 2016). Furthermore, kaolin clay is one material to produce porous ceramic besides the pore-forming agents (PFA), and it is mandatory material in making porous ceramic. Therefore, a pore-forming agent (PFA) will be discussed in the next section, another material in producing porous ceramics.

2.3 Pore-forming Agent (PFA)

In ceramic production, additives are widely employed to create clay products with adequate physical and mechanical qualities. Choosing additives depends on the required characteristics. Lightweight ceramics with high compressive strength and low water absorption are excellent. One method for increasing ceramic capacity is to introduce pores into the clay body using organic or inorganic pore formers (Karaman et al., 2008). Pore formers are pyrolytic substances that burn out during sintering. Specific pore-forming agents such as kenaf fibre, rice hush ash, and coconut husk can be used to create porous ceramics with controlled microstructure (Beal et al., 2019). Thus, the choice of coconut husk as a pore-forming agent was considered toward the properties of porous ceramic.

Coconut husk was chosen as a pore-forming agent due to its features that will improve the application of porous ceramic. Coconut husk is the most well-known fibrous waste product from coconut tree cultivation. Every year, over 30 million tonnes of coconut are produced worldwide, with coconuts being especially abundant along the coastlines of tropical countries. The coconut husk comprises 30% fibre and 70% pith, with a high concentration of lignin and phenolic compounds. Coconut husk contains a high concentration of lignin, which makes it exceptionally elastic, durable, and resistant to rotting (Gaspar et al., 2020). In addition, coconut husk fibres, particularly fine particles or coir dust, can be incorporated into ceramic mixtures to introduce porosity.

2.4 Utilization of Coconut Husk into Porous Ceramic (Previous Study)

Porous ceramic permeability is greatly influenced by porosity, pores, and pore shape. Using pore-forming agents will increase the number of pores and expansion can be permitted. The use of pore-forming agents in ceramic membrane preparation is capable of growing the number of pores. Instead, the addition of coconut husk in porous ceramic increases thermal conductivity attenuation, which limits energy loss and so improves thermal comfort within homes. The additives' goal was to generate pores inside the ceramic that would improve its qualities for specific use. Furthermore, the water absorption of bricks can affect their longevity when exposed to environmental conditions (Beal et al., 2019). Thus, the weight percentage of pore-forming added to the ceramic must be optimized.

2.5 Physical Properties

Physical properties tested for were apparent porosity; bulk density; apparent density; percentage water absorption. The apparent porosity (the number of voids or pores in a volume of porous solid) is influenced by kaolin. Higher water absorption reduces the properties of porous ceramics (Obada et al., 2016). The sintering temperature also affects the size of the pore distribution, with closed pores increasing and open pores decreasing by the liquid phase, with maximum shrinkage occurring (Vasić et al., 2022).

From the previous study, the water absorption and apparent porosity of porous ceramic incorporated with 30wt.% of coconut husk is the highest. The sintering temperature also increases with the increasing apparent porosity of porous ceramic. The relationship between apparent porosity and water absorption was shown by a similar trend. A higher percentage of water absorption resulted from an increase in apparent porosity. Higher sintering temperatures, however, can impact apparent porosity because optimal viscous flow can cover the porosity. However, the weight percentage of coconut husk increased and the bulk density of porous ceramic is decreased. The highest bulk density of the porous ceramic incorporated is at 0wt.%. Meanwhile, the sintering temperature increased and the bulk density also increased (Sifa Amina, 2023).

2.6 Compressive Strength

Strength is an important parameter. It is typically boosted by decreasing total pore volume, even though this may compromise other functional qualities. Morphological characteristics such as pore size and volume might be critical in optimizing performance while maintaining high strength (Seuba et al., 2016). Additionally, as porosity increased, the compressive strength decreased. This demonstrates once again that porosity is insufficient to describe the effect of pore structure on strength. The proposed pore size effect function quantifies the effect of pores per unit volume on strength based on pore size. The effect can be described as a pore-size power function. In addition to apparent porosity, bulk density, and water absorption, compressive strength plays an important role in porous ceramic performance. The purpose is to find the balance between compressive strength and porosity.

The highest compressive strength of porous ceramics with is at 0 wt.% coconut shell in previous studies. The compressive strength of porous ceramics decreased as the weight ratio of coconut husk increased. On the other hand, increasing temperature works in reverse and increases the compressive strength of porous ceramics. As the apparent porosity % rises, so will the number of open pores (Sifa Aminah, 2023).

2.7 Statistical Analysis for Porous Ceramic

Statistical analysis involves the collection, interpretation and presentation of data. Statistical analysis has been widely used to study the optimization of porous ceramic. In the context of porous ceramic optimization, it is used to study and improve various properties of ceramic through the application of statistical methods. Statistical analysis can be employed to study effects of various processing parameter on the material such as porosity, density and mechanical strength (Papageorgiou, 2020).

This can involve the use of methods like Response Surface Methodology, which helps in optimizing the manufacturing process and the properties of the final ceramic product. For example, optimization of the physical properties of rice husk ash (RHA) in ceramic materials were carried out using Response Surface Methodology (Wisdom et al., 2021). The independent variables, namely the firing temperature and residue content were statistically merged in a Central Composite Design of experiments. It is an efficient statistical technique, to determine the optimum condition like firing temperature and residue content that improve its physical properties.

Another example is optimizing production parameter of ceramic tiles incorporating fly ash also using Response Surface Methodology (Koçkal, 2015) The three variables chosen for the study were namely, sintering temperature, fly ash content and fly ash type designated as A, B, C whereas the predicted responses such as shrinkage, water absorption and flexural strength, were each designated as Y. The mathematical relationship between the variables and the responses were approximated by the second order polynomial. Historical data design under Response Surface Methodology was used to analyze the interactive effect of temperature, fly ash content and fly ash type and to arrive at an optimum.

Response Surface Methodology (RSM) is a valuable optimization technique that recommends optimal parameter settings, particularly for cutting speed, to enhance the mechanical and physical properties of materials in reinforcement applications. By combining statistical and mathematical methods, RSM identifies the best experimental conditions while minimizing the number of experiments required for accurate outcomes. This evolving optimization technique is well-suited for experimental research, emphasizing efficiency and effectiveness. The central composite design, a common RSM approach, is typically executed using software such as Design Expert or Minitab.

Overall, statistical analysis plays a crucial role in the optimization of ceramic by providing a systematic approach to data analysis, process improvement and quality enhancement.

2.8 Summary

Porous ceramics have gained significance in various industries due to their diverse applications, such as filters, dust collectors, thermal insulators, and electronics components. They exhibit desirable properties like high-temperature and chemical corrosion resistance, low thermal conductivity, high surface area, and low density. Porous ceramics can be categorized based on pore size and porosity, which can be controlled through different fabrication techniques and processes.

Coconut husk is utilized as a pore-forming agent in ceramic production to create lightweight ceramics with high compressive strength and low water absorption. The research suggests that the choice of coconut husk as a pore-forming agent is beneficial for enhancing the properties of porous ceramics, making it a promising and sustainable additive for ceramic production. Physical properties and compressive strength of porous ceramics are influenced by factors like apparent porosity, bulk density, and water absorption, with kaolin and coconut husk content playing significant roles.

Statistical analysis plays a crucial role in the optimization of porous ceramic materials, as it provides a systematic approach to data analysis, process improvement, and quality enhancement. In the context of porous ceramic optimization, statistical analysis has been used to study and improve various properties of ceramic materials, such as porosity, density, and mechanical strength, through the application of statistical methods like Response Surface Methodology (RSM). There are examples demonstrate the effectiveness of statistical analysis in optimizing porous ceramic materials, as it helps identify the best conditions for various processing parameters and improves the final properties of the ceramic products.

CHAPTER 3

MATERIALS AND METHODS

3.1 Research Flowchart

This is the research flow for study of optimization of coconut husk as pore forming agent in porous ceramic using response surface methodology (RSM) statistical design.

Preparation of raw material

↓

Coconut Husk	Kaolin Clay
- Dried in an oven at 110°C for 24 hours, then processed in a heavy-duty blend for 6 minutes - powder being sieve through 250 µm.	

↓

Characterization of raw material

- FTIR
- XRD

↓

Sample preparation		
10 CH%	20 CH%	30 CH%

↓

Mixing process

- The CH powder being mix with kaolin clay and 50-60 wt.% water until homogeneity achieved

⬇

Drying process
- The mixture dry in oven at 110ºC for 24 hours

⬇

Crushing process
- the dried mixture being milling using mortar

⬇

Granulation process
- The powder mixture being moistened with 6 wt.% by spraying with water

⬇

Compacting process
- Compact by uniaxial hydraulic press with pressure of 40 MPa
- Compact granule powder about 3g in a 20mm hardened tool steel mould

⬇

Drying process
- Dry the mixture in oven at 110ºC for 24 hours

⬇

Sintering Process		
850℃	900℃	950℃

⬇

Characterization Of Porous Ceramic

⬇

Response Surface Methodology

Figure 3.1: Research Flowchart

3.2 Materials

3.2.1 Coconut Husk

The coconut was collected and the physical sorting was done using a larger sieve to ensure no foreign substance in the sample would be a grind. After that, coconut husks were dried in an oven at about 110℃ for 24 hours. Mass before and after drying was recorded to conduct a moisture content test. Dry coconut husk was processed in a heavy-duty blend for 6 minutes. The ground coconut husk was sieved using a 250 µm sieve to ensure that only ground coconut husk with particle sizes equal to or less than 250 µm were utilized in the clay-based porous ceramic (Sifa Aminah, 2023). Then, the coconut husk powder was inspected by Fourier Transform Infrared Analysis (FTIR).

3.2.2 Kaolin Clay

The kaolin clay powder was sieved using a 250 µm test sieve to ensure that only the same particle kaolin clay sizes equal to or less than 250 µm were used in the coconut husk powder. After that, kaolin clay was combined with coconut husk powder to create porous ceramic. For characterization technique was performed by using X-Ray Diffraction analysis (XRD) to determine the crystallographic structure of kaolin clay (Sifa Aminah, 2023)

3.3 Fabrication of Porous Ceramics

3.3.1 Response Surface Methodology (RSM) Statistical Design

Central Composite Design (CCD) was used to optimize the porous ceramic, identify a correlation between independent variables (factors) and dependent variables (response), and create the model equation describing the design of the experiment. Numerous sources emphasized the significance of optimizing the addition of coconut husk to porous ceramics to improve qualities such as RSM and CCD (Ayan et al., 2020). Based on statistical software and laboratory experimental methodologies, this Response Surface Methodology (RSM) statistically applies combined parameters to achieve higher optimization output, which forecasts better modifications that enhance the wt.% of coconut husk added.

3.3.2 Experimental Design Mental

Central Composite Design (CCD) was used to improve the weight percentage of coconut husk and the sintering temperature. A single factor experiment was used to determine the central values of the two factor conditions. In order to anticipate the ideal circumstances for preparation and the interaction between the preparation conditions, the RSM approach is used. In this study, a 2-factor (wt.% coconut husk and sintering temperature 3-levels (-1, 0 and 1) CCD with a small number of experimental runs is used to assess and optimize the addition of coconut husk into porous ceramic and achieve the best composition to produce the best porosity and strength. Also, coding was used to denote the level or range of each evaluated factors on scale where for the wt.% coconut husk, '-1for 10%, 0for 20% and '1' for 30%. While for the sintering temperature, '-1for 850°C, '0for 900 °C and 1' for 950 °C.

Table 3.1: Factors and their Respective number of levels investigated in Central Composite Design (CCD)

Factors	Notation	Unit	Levels (in coded)		
			-1	0	1
Wt.% CH	A	Wt.%	10	20	30
Sintering temperature	B	℃	850	900	950

Table 3.2: Experimental Design Matrix for Central Composite Design (no value include for all response)

Factor			Response			
Run Order	**Wt.% Coconut Husk**	**Sintering Temperature**	**Water Absorption (%)**	**Apparent Porosity (%)**	**Bulk Density (g/cm^3)**	**Compressive Strength (MPa)**
1	0	0				
2	-1	-1				
3	-1	-1				
4	1	0				
5	-1	0				
6	0	0				
7	0	-1				
8	0	0				
9	1	0				
10	1	-1				
11	0	0				
12	0	-1				
13	-1	0				
14	-1	1				
15	1	-1				
16	0	0				
17	0	1				
18	-1	1				
19	0	1				
20	0	0				
21	0	0				
22	0	0				
23	1	1				
24	0	0				
25	0	0				
26	1	1				

3.4 Preparation of Porous Ceramic Incorporated with Coconut Husk

The process then began by combining the coconut husk and kaolin clay, followed by 50-60 wt.% of water and 1 hour in the hand mixer. The sludge was well blended. After mixing, the slurry was dried for 24 hours in an electric oven set to 100 °C. It is high density component sediment appears to occur during the drying phase of the slurry, the dried combination is milled using mortar for 1 hour to generate powder mixture and re-homogenize components in the mixture. Following that, the granulation procedure on the powder mixture is carried out. First, the powder combination was moistened with distilled water using a water sprayer. The amount of moisture introduced is approximately 5 to 6 wt.% (2.5 to 3.0 g) per powder mixture (50 g). A uniaxial hydraulic press at a pressure of 40 MPa compacted the granule powder about 3g in a 20mm hardened tool steel mold. The compressed body is then dried for 24 hours in an electric oven at 100°C before burning for 1 hour in a muffle furnace. The material is prepared for sintering once it has completely dried. Three temperatures are usually used for the sintering process: 850, 900, and 950 °C. The WS particles are consumed during the sintering process, leaving holes in the ceramic matrix. The ceramic particles will fuse together and go through sintering to create a solid structure. To avoid thermal stress, the porous ceramic is cooled gradually after the sintering process. Its physical qualities can then be investigated. We will examine the water absorption, bulk density, apparent porosity, and compressive strength of porous ceramic samples.

3.5 Characterization of Porous Ceramic

The characterization of water absorption test, apparent porosity, bulk density and compressive strength was done for all sample and then the data were analyzed using CCD by RSM. Based on the results of water absorption, apparent porosity, and bulk density, a specific sample was subjected to compressive strength characterization, microstructural investigation, and XRD. The mechanical characteristics of the porous ceramic samples were determined using this test.

3.5.1 Water Absorption, Apparent Porosity and Bulk Density

MS ISO 10545-3: 1995 was utilized in this investigation to characterize water absorption. This characterization will evaluate the water absorption, apparent porosity, and bulk density of the sample. In the present study, the vacuum method is utilized to extract the dry mass of the sample before weighing it to measure the water absorption. This procedure will extract air from a chamber containing samples. The samples were then submerged in water for at least 30 minutes at room temperature. The sample is then removed from the bath, patted dry, and reweighed to determine the saturated mass of cold water in the sample. Finally, the sample is measured while immersed in water. The volume, V, of each sample is calculated using the water displacement method. The samples are then tested before and after immersion in water for determining water absorption (Equation 3.1), apparent porosity (Equation 3.2), and bulk density (Equation 3.3) applying the equations below.

Equation 3.1:

$$\textbf{E (b,v)} = \frac{m^2(b,v)}{m^1} - \frac{m^1}{m^1}\ x\ 100$$

∴ Where,

m^1: *mass of dried porous ceramic*

m^2: *mass of wet porous ceramic*

Equation 3.2:

$$\textbf{P} = \frac{m^2 v}{V} - \frac{m^1}{V}\ x\ 100$$

∴ Where,

m^1: *mass of dried porous ceramic*

m^2: *mass of wet porous ceramic*

V: volume

Equation 3.3:

$$\textbf{B} = \frac{m^1}{V}$$

∴ Where,

m^1: *mass of dried porous ceramic*

V: volume

3.5.2 Compressive Strength

The compressive strength of a concrete is a very important property. This test adheres to ASTM C109 in order to accomplish this characterization. The test method allows you to determine the compressive strength of hydraulic cement and other mortars. The tests may be applied to determine compliance with requirements. This test examines the effect of adding coconut husk to the porous ceramic strength and its capacity to sustain compressive stress. Testometric Materials Testing Machine was used to measure the compressive strength. The final product was compressed using a compression machine, and the strength was calculated using Equation 3.4 below.

Equation 3.4:

$$\mathbf{F} = \frac{P}{A}$$

∴ Where,

F: compressive strength (N/mm^2)

P: applied load (N)

A: surface area (mm^2)

3.5.3 FTIR Analysis

The infrared (IR) spectra of coconut husk were obtained using a Fourier transform IR spectrometer (FT-IR) model (Thermo Fisher Nicolet iS20 FTIR Spectrometer). Functional groups and compound contain has been observed and data was analyzed based on absorption coconut husk.

3.5.4 XRD Analysis

To investigate the structure and phases present in the porous ceramic, the measurements were recorded under Bragg-Brentano geometry in the 10°-90° at 2θ range.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Characterization of Raw Materials

The characterization of raw materials involves understanding the properties and behavior of the materials during fabrication and sintering processes. Raw material was used in the experiment are coconut husk and kaolin clay. The raw material which is coconut husk were characterized using FTIR to investigate their functional group and kaolin clay were characterized using XRD are to investigate phases present in the materials.

4.1.1 FTIR Analysis of Coconut Husk

Fourier Transform Infrared Analysis is required to identify the functional groups in the coconut husk. This technique detects functional groups such as vibrational bands N-H, O-H, C-H, C = O, C = C, C = N, and C = N (Younis et al., 2021). These are few chemical compounds or functional groups are found in coconut husk, it is determined by different color at the peak to present functional groups. The absorption spectrum on the infrared region of coconut husk can be observed in the Figure 4.1.

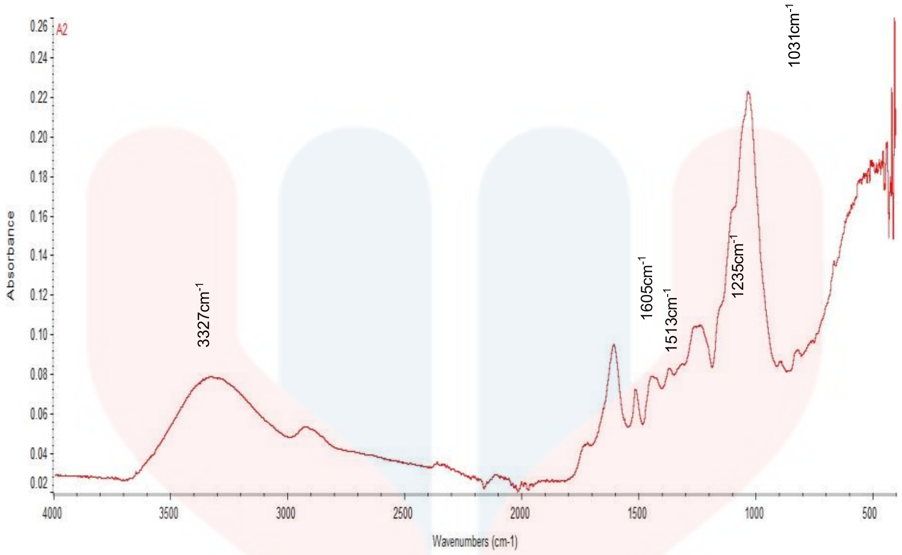


Figure 4.1: FTIR peaks of coconut husk.

Based on Figure 4.1, the FTIR analysis has discovered several peaks that are indicative of different functional groups. The peak at around 3327 cm^{-1} attributed to hydroxyl (OH) group. The broad peak at 3000 - 3800 cm^{-1} corresponds to the hydroxyl (OH) groups from cellulose, hemicellulose and lignin (Koay et al., 2013). The large band is attributed to the axial deformation of the O-H group (Mothé & De Miranda, 2009).

The peak at around 1605 cm^{-1} in the FTIR analysis corresponds to the aromatic ring of lignin due to the C=C stretching vibrations. This peak is due to the carbonyl stretching conjugated with the aromatic ring skeleton (Rashid et al., 2016). Next, the peak at around 1513 cm^{-1} associated to the C-N stretching and N-H bending vibrations. The C-N stretching observed in the region of 1400 to 1000 cm^{-1} meanwhile N-H bending observed in the region of 1650 to 1580 cm^{-1} (IR Spectrum Table, n.d.) (Mallamace et al., 2015). The peaks around 1232 cm^{-1} indicate C-O stretching absorption within the alkyl aryl ether compound group.

Furthermore, the peak at about 1031 cm^{-1} is characteristic of primary aliphatic groups, demonstrating strong C-O stretching, typical of primary alcohol in alcohol functional groups. C-O stretching give rise to an intense stretching peak normally found between 1300 and 1000 cm^{-1} (Smith, 2020). The carbonyl group of the hemicelluloses gives signal at 1620 cm^{-1}, while the band at 1235 cm^{-1} is associated to the presence of C-O-C in cellulose chain. The strong absorption band at 1208 cm^{-1} is related to the C-OH stretching vibration (Mothé & De Miranda, 2009).

4.1.2 XRD Analysis of Coconut Husk

XRD stands for X-ray diffraction is used to analyze physical properties such as phase composition, crystal structure and chemical composition of materials. The resulting diffraction pattern provides information about the arrangement of atoms in the sample, which can be used to identify the crystal structure of the material. The XRD peak of the kaolin clay is show in Figure 4.2.

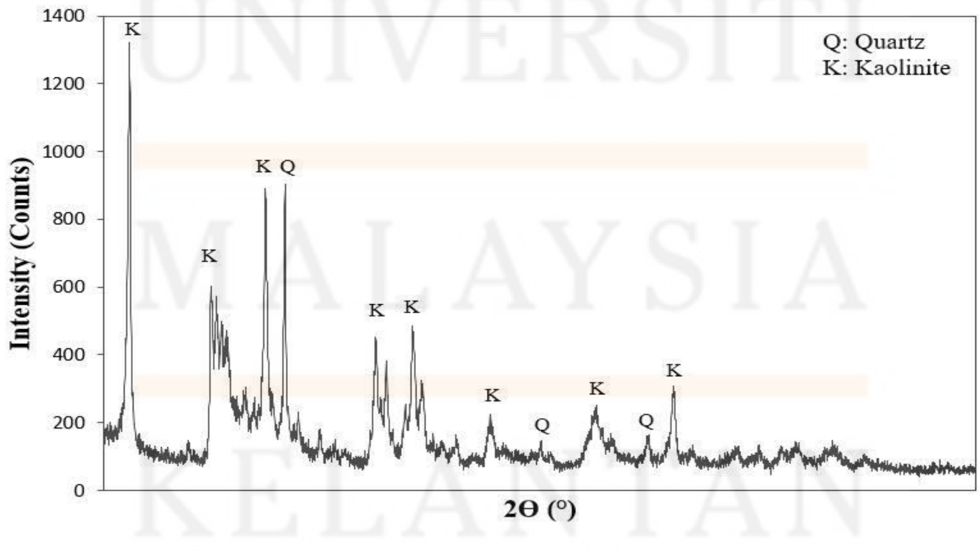


Figure 4.2: XRD peaks of kaolin clay.

According to Figure 4.2, it can be inferred that the XRD analysis observed consisted of kaolinite (COD 9009230) and quartz (COD 9010144). Kaolinite is a layered silicate clay mineral with the chemical formula Al2Si2O5(OH)4 while quartz is a silica mineral with the formula SiO2.

The XRD pattern of kaolinite typically shows characteristic peaks at specific 2θ values, which are indicative of its crystalline structure. The absorption peaks appear at 2θ (°) is in around 11.68°; 21.22°; 34.94°; 38.35° meanwhile quartz is in around at 26.57°. These peaks are associated with the crystallographic structure of kaolinite and can be used to identify and characterize the mineral. The presence of crystalline quartz in kaolin raw material can impact its properties, such as plasticity and refractoriness.

4.2 Response Surface Methodology (RSM) Statistical Design Analysis

The statistical design aimed to optimize the composition of porous ceramic using kaolin clay and coconut husk. The Central Composite Design (CCD) under RSM was used as a convenient design optimization approach. The Central Composite Design is a statistical design that allows for the optimization of a response variable by varying the levels of two or more factors. The optimization of the porous ceramic was carried out using the Minitab Analysis Output design. The optimization process helps to identify the optimal combination of factors, such as the composition of coconut husk to kaolin clay to achieve the desired properties for the porous ceramic.

The wt.% of coconut husk and the sintering temperature were chosen as the factors to be varied because they are known to have a significant impact on the properties of porous ceramic such as water absorption, apparent porosity, bulk density and compressive strength. The experimental design involved 26 runs in random, with each run representing a combination of the wt.% of coconut husk and the sintering temperature. The runs were carried out in order with 2 replicates are shown in Table 4.1.

The design allows for the calculation of the adequacy of the design, which helps to ensure that the design is capable of accurately estimating the response variables. The analysis of variance (ANOVA) helps to identify the significant factors that affect the response variables, while the main effect and interaction plots provide a visual representation of the relationship between the factors and the response variables.

4.3 Experimental Design Matrix

In experimental design, design matrix is a tool used to represent all the combination of factors being assessed in an experiment. Every column of the matrix corresponds to a factor, and the entries within a column correspond to levels of the factor. It is based on a two-level factorial design with the addition of 2^k star points, allowing the exploration of quadratic effects and the construction of a second-order polynomial equation (Wagner et al., 2014). In a two-level factorial design, factors are manipulated at two levels usually labeled as -1 and +1. This experiment to study the effects of different factors at low and high levels.

In the context of this study, the Central Composite Design was used to optimize the composition of the system by varying the wt.% coconut husk added and sintering temperature to determine the water absorption, apparent porosity, bulk density and compressive strength. The experimental design matrix and response for the water absorption of coconut husk which was obtained from the total 26 experimental runs in random order 2 replication are shown in Table 4.1.

Based on the experimental design matrix result, the lowest water absorption is at 20 wt.% of coconut husks at 850°C, where the value is 16.89%, and the highest water absorption is at 30 wt.% of coconut husks at 850°C, where the value is 53.73%. Meanwhile, the lowest compressive strength is at 30 wt.% of coconut husks at 900°C, where the value is 11.09 MPa, and the highest compressive strength is at 10 wt.% of coconut husks at 950°C, where the value is 55.59 MPa.

Table 4.1: Experimental Design Matrix for Central Composite Design

Factor			Response			
Run Order	**Wt.% Coconut Husk**	**Sintering Temperature**	**Water Absorption (%)**	**Apparent Porosity (%)**	**Bulk Density (g/cm^3)**	**Compressive Strength (MPa)**
1	0	0	35.31	41.59	1.17	24.17
2	-1	-1	25.09	36.41	1.45	37.12
3	-1	-1	25.13	35.29	1.40	36.51
4	1	0	52.11	50.78	0.97	12.53
5	-1	0	25.71	35.06	1.36	28.32
6	0	0	30.65	38.69	1.26	22.96
7	0	-1	28.03	41.93	1.49	20.58
8	0	0	34.95	42.31	1.21	33.01
9	1	0	45.14	47.02	1.04	11.08
10	1	-1	34.78	38.70	1.11	12.51
11	0	0	32.06	39.58	1.23	25.64
12	0	-1	16.88	23.33	1.38	22.33
13	-1	0	26.43	36.05	1.36	29.05
14	-1	1	24.76	35.04	1.41	55.59
15	1	-1	53.72	54.02	1.00	11.35
16	0	0	33.52	40.98	1.22	32.36
17	0	1	28.46	37.32	1.31	39.18
18	-1	1	25.46	35.58	1.39	48.71
19	0	1	32.97	40.88	1.24	40.72
20	0	0	33.25	36.67	1.10	35.36
21	0	0	35.65	49.39	1.38	14.66
22	0	0	34.29	41.19	1.20	15.34
23	1	1	45.00	47.88	1.06	20.63
24	0	0	34.29	40.93	1.19	32.10
25	0	0	33.74	40.96	1.21	26.18
26	1	1	50.12	51.10	1.01	22.05

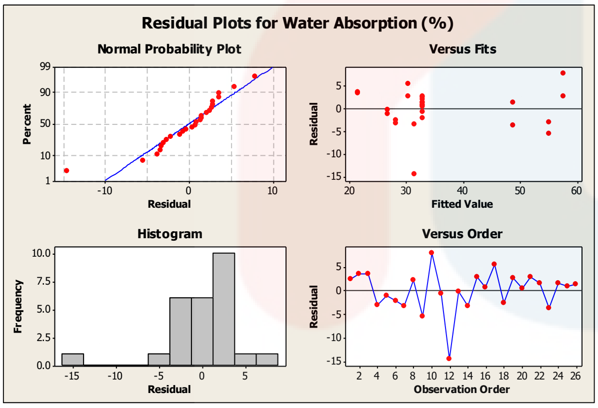
4.4 Model Adequacy Checking

Model Adequacy Checking is the process of statistical modelling whether the chosen statistical model is appropriate for the data at hand and whether the assumptions underlying the model are reasonable. The assumption that the relationship between the dependent variable and the explanatory variables is linear. The aim is to ensure that the model accurately represents the underlying patterns in the data and provides reliable and meaningful results. It may involve refining the model or choosing a different modeling approach based on the findings. It is an essential to ensure the reliability of the statistical inferences drawn from the model.

Residual analysis is a technique used to assess the validity of a regression model by examining the differences between observed values and the values predicted by the model. It helps in identifying patterns, trends or outliers in the residuals, which are the differences between the observed values and the predicted values from the regression model. Various types of graphics can be examined for different assumptions, and these graphics are generated by regression software. The normal probability plot used to determine whether a data set is normally distribution or not. It is approximately linear supporting the condition that the error terms are normally distributed. The normal probability plot is a useful graphical technique for determining whether a data set is approximately normally distributed, it also provide insights into the normality of the error terms in a statistical model.

The residual plots indicate a light-tailed distribution, suggesting a reduced rate of outliers compared to a normal distribution where the observations are close to a distribution boundary. The first few points show departure from fitted line below the line for long runs where the final few points show an increasing departure from the fitted line above the line. Despite the departure in the first and last few points, it is inferred that water absorption, apparent porosity and bulk density of porous ceramic incorporated coconut husk. The properties satisfy the first requirements of the adequacy checking model were distributed normally, which fundamental for detecting model misspecifications.

The residual plots for water absorption, apparent porosity, bulk density and compressive strength of the porous ceramic incorporated with coconut husk are presented in Figure 4.4 (a), (b), (c), and (d) which contain normal probability plot, histogram of frequency versus residual, residual versus fits and residual versus observation order of data.



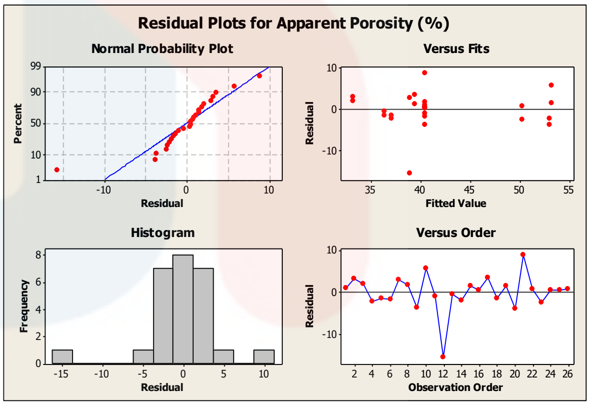
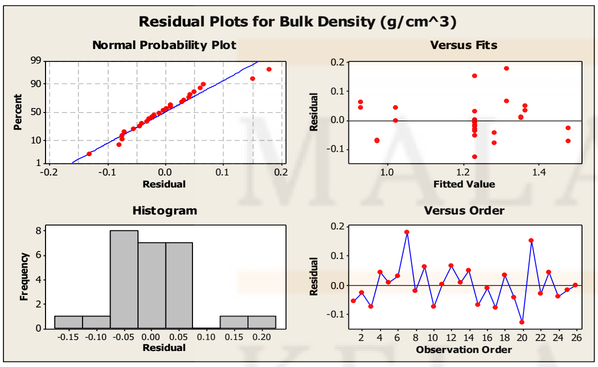


Figure 4.4 (a) **Figure 4.4 (b)**



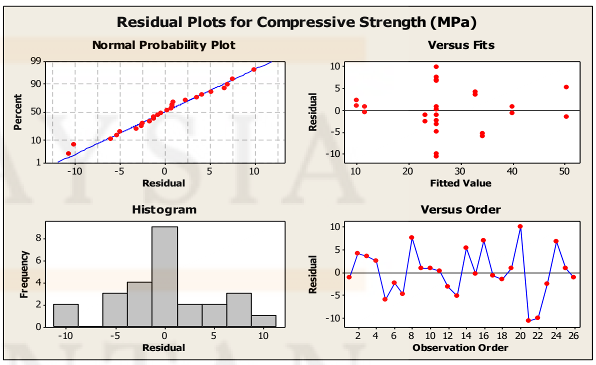


Figure 4.4 (c) **Figure 4.4 (d)**

The pattern observed in histogram images at Figure 4.4 (a), (b), (c), and (d) which is shaped like a dumbbell suggest that the data is normal distribution. The residual against fitted value plots demonstrate a random allocation of data points indicates that the model residuals are evenly distributed across the range of predicted values, which supports the assumptions of constant residual variance, also known as homoscedasticity. The independence of residual against observation order meets the assumption of independent observations, which ensures that the error terms are uncorrelated with each other and with the independent variables in the model.

4.5 Analysis of Variance (ANOVA)

ANOVA, which stands for Analysis of Variance is a statistical method used to analyzing the significance of factors and their interactions in a quadratic model. It is a technique used to compare methods from different samples and test the effect of one or more variables. The experimental scheme and data were analyzed using the Minitab 16 Statistical Software. The p-value, or probability value is a measure used in statistical hypothesis testing to determine the significance of a result. The p-value represents the probability of obtaining the observed results. The p-value should be ≤ 0.05 to considered statistically significant. A quadratic model obtained from ANOVA, which can be used to determine the significance of the quadratic terms and their interactions thereby assessing the overall significance of the model.

Table 4.2, Table 4.3, Table 4.4 and Table 4.5 shows the ANOVA for Water Absorption, Apparent Porosity, Bulk Density and Compressive Strength of porous ceramic incorporated coconut husk. The p-value for the wt.% of coconut husk linear factor for water absorption, apparent porosity, bulk density, compressive strength and the p-value for the sintering temperature for compressive strength were reported to be zero (0.000) respectively. This suggests that the wt.% of coconut husk linear factor has significant effect on water absorption, apparent porosity, bulk density and compressive strength. The p-value for wt.% coconut husk * sintering temperature for water absorption is 0.033 which statistically significant less than 0.05. A lower p-value generally indicates a stronger relationship between two variables. In this case, the statistical relevance implies that it is statistically significant, as the different wt.% of coconut husk added give an effect on the water absorption, apparent porosity, bulk density and compressive strength.

Table 4.2: ANOVA for Water Absorption of porous ceramic incorporated coconut husk

SOURCES	P-value
Wt.% Coconut Husk	0.000
Sintering Temperature	0.668
Wt.% Coconut Husk * Sintering Temperature	0.033

Table 4.3: ANOVA for Apparent Porosity of porous ceramic incorporated coconut husk

SOURCES	P-value
Wt.% Coconut Husk	0.000
Sintering Temperature	0.871
Wt.% Coconut Husk * Sintering Temperature	0.321

Table 4.4: ANOVA for Bulk Density of porous ceramic incorporated coconut husk

SOURCES	P-value
Wt.% Coconut Husk	0.000
Sintering Temperature	0.471
Wt.% Coconut Husk * Sintering Temperature	0.157

Table 4.5: ANOVA for Compressive Strength of porous ceramic incorporated coconut husk

SOURCES	P-value
Wt.% Coconut Husk	0.000
Sintering Temperature	0.000
Wt.% Coconut Husk * Sintering Temperature	0.471

However, the higher p-values (≥ 0.05) for the sintering temperature linear factor for water absorption (0.668), apparent porosity (0.871) and bulk density (0.471) meanwhile the higher p-values (≥ 0.05) for the wt.% coconut husk * sintering temperature for apparent porosity (0.321), bulk density (0.157) and compressive strength (0.471). This means that these factors may not have a significant effect on the responses in the study. This result indicates to the sintering temperature factor, where lead to changes in microstructure like reduction in pore number and size, grain growth and increased density. The porosity of ceramic decreases with an increase in sintering temperature.

4.5.1 Main Effect

A main effect plot in statistical analysis is a graphical tool used to examine the differences between the mean values of a response variable for different levels of one or more factors. It is commonly used in the analysis of variance (ANOVA) and experimental design to determine whether the main effect of a factor is present. The plot displays the means for each group within a categorial variable, allowing for a visual comparison of the effects of the independent variable.

If the line is horizontal, then there is no main effect and the response mean is the same across all factor levels. If the line is not horizontal, then the main effect exists and shows whether the average response value increases and decreases. The main purpose of a main effect is to compare the changes in the means to identify the most significant factor. In the analysis of variance, main effect will test whether there is evidence of an effect of different treatment (Hessing & Pv, 2023). In statistics, a main effect is the effect of just one of the independent variables on the dependent variables.

The main effect for water absorption, apparent porosity, bulk density and compressive strength of the porous ceramic incorporated with coconut husk are presented in Figure 4.5.1 (a), (b), (c), and (d)

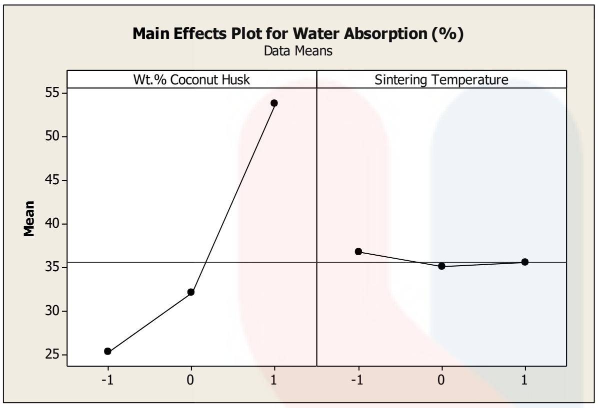


Figure 4.5.1 (a)

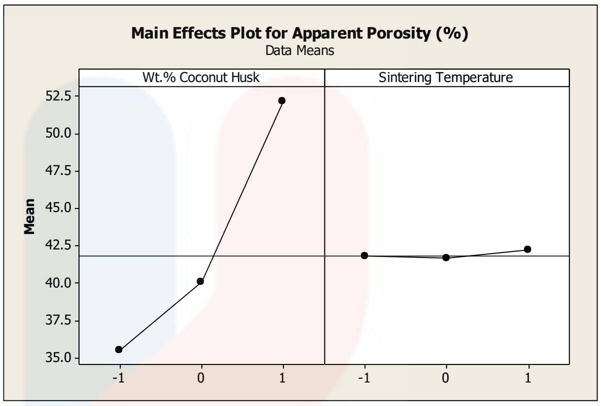


Figure 4.5.1 (b)

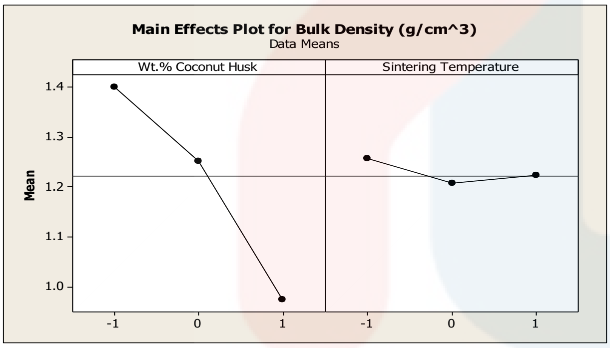


Figure 4.5.1 (c)

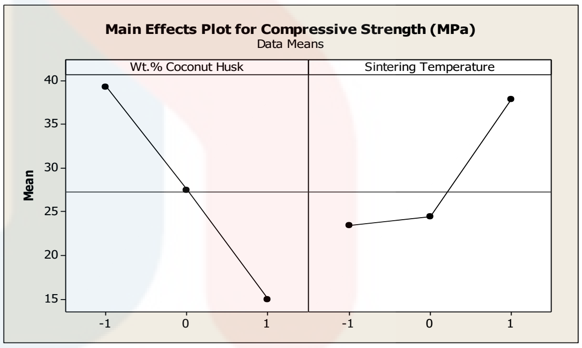


Figure 4.5.1 (d)

Figure 4.5.1 (a) and Figure 4.5.1 (b) present main effect plot for water absorption (%) and apparent porosity (%) in porous ceramic. It is showed an increasing trend to both water absorption and apparent porosity with the weight percentage of coconut husk from 10 wt.% (coded as '-1') to 30 wt.% (coded as '1'). As for the sintering temperature for water absorption, the trend was decreasing from 850°C (coded as '-1') to 900°C (coded as '0'), then increase at 950°C (coded as '1') where impact the porosity and density of the material. Meanwhile, the effect of sintering temperature on apparent porosity shows a decreasing trend from 850°C (coded as '-1') to 900°C (coded as '0'), but then the trend starts to increase at 950°C (coded as '1'). The sintering temperature can significantly impact the porosity of the coconut husk leads to formation of more interconnected pores, which can increase the apparent porosity of the porous ceramic.

Furthermore, figure 4.5.1 (c) and figure 4.5.1 (d) present main effect plot for bulk density (g/cm^3) and compressive strength (MPa) in porous ceramic. Bulk density (g/cm^3) and compressive strength (MPa) showed a decreasing trend to both bulk density and compressive strength with the weight percentage of coconut husk from 10 wt.% (coded as '-1') to 30 wt.% (coded as '1'). The effect on bulk density for sintering temperature show a decreasing pattern from 850°C (coded as '-1') to 900°C (coded as '0'), and the trend started to increase to 950°C (coded as '1'). As the sintering temperature increase, the bulk density of the porous ceramic initially decreases but then start to increase at higher temperature. This is due to the fact where at higher temperature, the porous ceramic become compact and dense, leading to an increase in bulk density. As the sintering temperature for compressive strength, it shows an increasing trend from 850°C (coded as '-1') to 950°C (coded as '1'). The higher percentage of coconut husk has increased the porosity, leading to lower compressive strength while higher sintering temperatures result in higher compressive strength

4.5.2 Interaction Plots

An interaction plot in statistical analysis is a visual representation of the interaction between two factors in a model. It is used to show how the relationship between one categorical factor and a continuous response variable depends on the value of a second categorical factor. The interpretation of an interaction plot is important because if the interaction effects are significant, the main effects cannot be interpreted without considering the interaction effects.

Parallel lines indicate the absence of an interaction effect, suggesting that the relationship between the factors is consistent across different levels. Conversely, the non-parallel lines in an interaction plot indicate the presence of an interaction effect, suggesting that the relationship between the two factors is not consistent across different levels (Frost, 2022).

The interaction plots for water absorption, apparent porosity, bulk density and compressive strength of the porous ceramic incorporated with coconut husk are presented in Figure 4.5.2 (a), (b), (c), and (d)

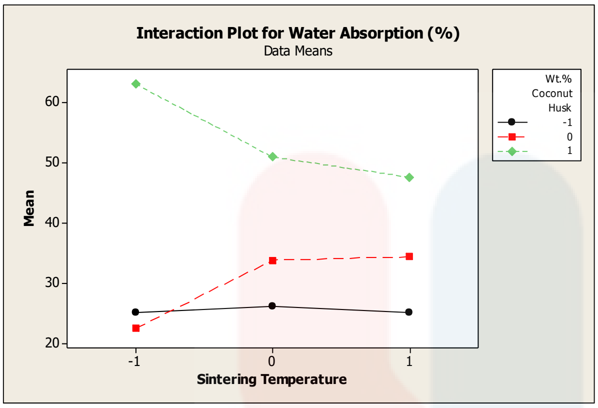


Figure 4.5.2 (a)

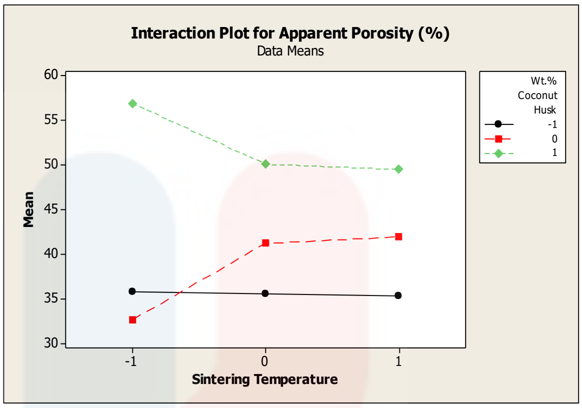


Figure 4.5.2 (b)

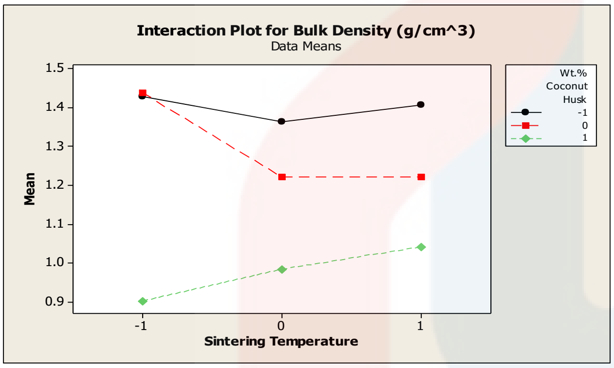


Figure 4.5.2 (c)

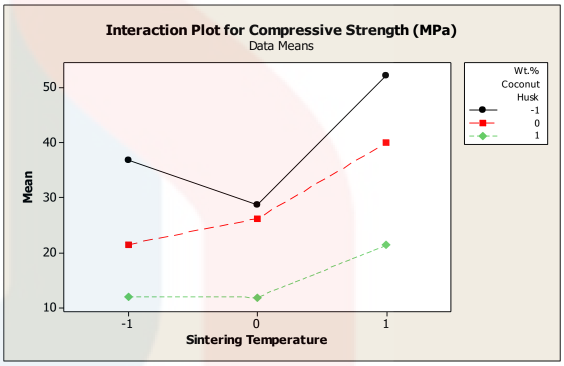


Figure 4.5.2 (d)

Figure 4.5.2 (a), (b), (c), and (d) represent interaction plot for water absorption (%), apparent porosity (%), bulk density (g/cm^3) and compressive strength (MPa) were non-parallel indicates a strong interaction between the weight percentage of coconut husk and sintering temperature. The highest water absorption and apparent porosity in figure 4.5.2 (a) and figure 4.5.2 (b) were observed in porous ceramic with 30 wt.% of coconut husks (coded as '1') at 850°C (coded as '-1'). The higher weight incorporation of coconut husk in porous ceramic is likely to increase water absorption and apparent porosity due to the porous nature of the material created by the coconut husk fibers (Ahmad et al., 2022).

Furthermore, figure 4.5.2 (c) and figure 4.5.2 (d) represent interaction plot for bulk density (g/cm^3) and compressive strength (MPa) in porous ceramic. The highest bulk density in figure 4.5.2 (c) was observed in porous ceramic with 20 wt.% of coconut husks (coded as '0') at 850°C (coded as '-1'). This indicates that the addition of coconut husk as pore-forming agent in the porous ceramic decreases the bulk density. Moreover, the highest compressive strength in figure 4.5.2 (d) was observed in porous ceramic with 10 wt.% of coconut husks (coded as '-1') at 950°C (coded as '1'). The compressive strength of porous ceramic was found to be exponentially correlated with porosity volume (Seuba et al., 2016b). This can be related with the result where 10 wt.% of coconut husks had highest compressive strength due to their pore size distribution.

4.5.3 Contour Plot

A contour plot is graphical technique used to represent a 3-dimesional surface by plotting constant z slices, called contours, on a 2-dimensional format. The graph shows values of the Z variable for combination of the X and Y variables. The Wt.% of Coconut Husk and Sintering Temperature displayed along the X and Y-axes, while contour lines and bands represent the Z-value.

All point that has the same response are connected to produce contour lines of constants responses which useful for investigating desirable response values and operating conditions. Contour plot also enhanced by adding color or shading to the contour lines, which help to highlight the areas of high or low values of high or low values of the variable. Contour plots are valuable tool for optimizing experimental factors and responses, identify optimal operating conditions and assess the impact of multiple factors on the outcomes of interest.

The contour plot for water absorption, apparent porosity, bulk density and compressive strength were shown in figure 4.5.3 (a), (b), (c) and (d). The darkest green colors showing highest of the responses which water absorption, apparent porosity, bulk density and compressive strength in the factors which weight percentage of coconut husks (X-axis) and sintering temperature (Y-axis).

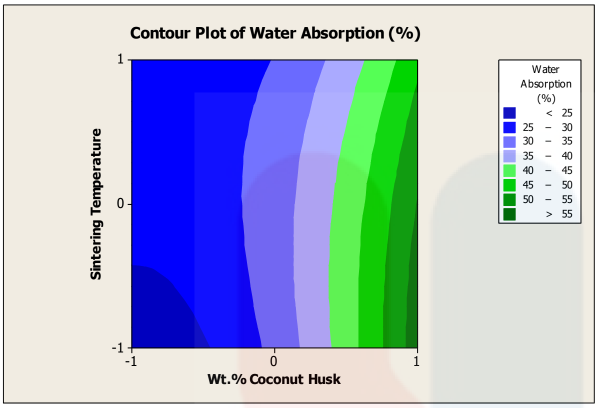


Figure 4.5.3 (a)

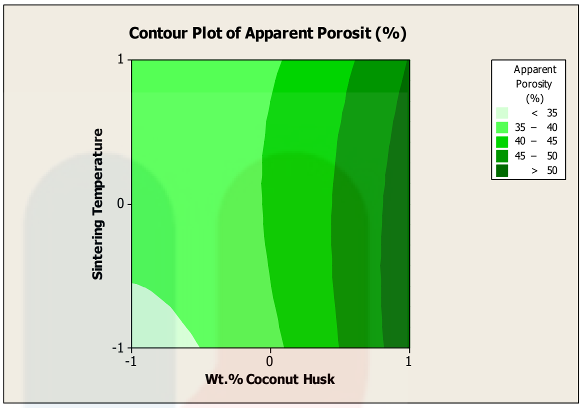


Figure 4.5.3 (b)

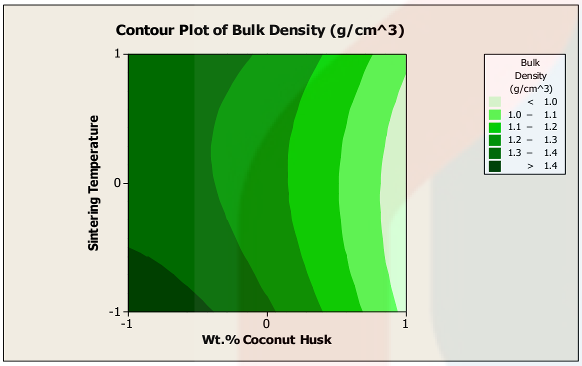


Figure 4.5.3 (c)

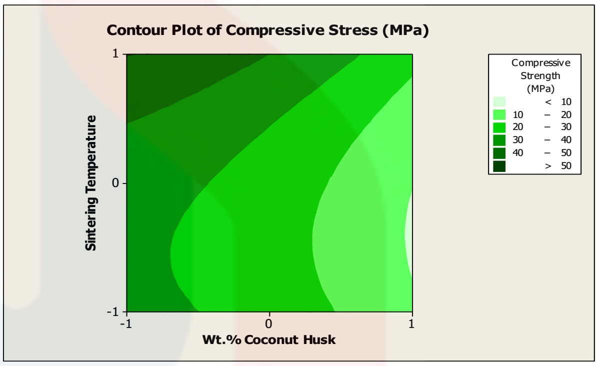


Figure 4.5.3 (d)

The contour plot in Figure 4.5.3 (a) and (b) illustrates the water absorption (%) and apparent porosity (%), where wt.% coconut husk is plotted on the X-axis and sintering temperature is plotted on the Y-axis. The contour areas represent constant responses which correspond to water absorption and apparent porosity of 25, 30, 35, 40, 45, 50 and 55. The contour with the darkest green color in the lower right corner where water absorption and apparent porosity is the highest which is 55% and 50%. The increase in water absorption and apparent porosity moves from lower left to lower right corner, indicating that water absorption and apparent porosity increase as the wt.% of coconut husk increases and sintering temperature decreases. However, the main of porous ceramic in water absorption and apparent porosity need to be as minimum as shown in the contour plot. In order to achieve that, the plot suggests that the optimal conditions for the lowest water absorption and apparent porosity are at sintering temperature of 850°C (coded as '-1') and a 10 wt.% of coconut husk (coded as '-1').

The contour plot in Figure 4.5.3 (c) and (d) represents the bulk density (g/cm^3) and compressive strength (MPa), with the X-axis representing the weight percentage of coconut husk and the Y-axis representing the sintering temperature. The contour areas represent constant responses, which correspond to bulk density of 1.0, 1.1, 1.2, 1.3, and 1.4. The contour areas represent constants responses corresponding to compressive strength of 10, 20, 30, 40 and 50. The contour with the darkest green color in the lower left, where highest bulk density is 1.4 g/cm^3 and compressive strength is 40Mpa. The decrease in bulk density moves from right to lower left corner, indicating that bulk density decrease as the wt.% of coconut husk and sintering temperature decreases. The plot suggests that the optimal conditions for the highest bulk density are a sintering temperature 850°C (coded as '-1') and a 10 wt.% of coconut husk (coded as '-1'). Meanwhile, the decrease of compressive strength in compressive strength moves from lower right to upper left corner, indicating that compressive strength decrease as the wt.% of coconut husk and sintering temperature increases. The contour with the darkest green color, indicating the highest compressive strength (40), is located at a sintering temperature of 950°C (coded as '1') and 10 wt.% of coconut husks (coded as '-1')

4.6 XRD Analysis of Porous Ceramic

Porous ceramic incorporated with coconut husk has been characterized using X-ray diffraction (XRD) analysis, where used to determine the crystalline composition of material. The selection was to determine the crystalline phase on different compositions (wt.% Coconut Husk) and sintering temperature (850°C, 900°C, 950°C). In this analysis, 10 wt.% CH and 30 wt.% CH with 850°C sintering temperature at Figure 4.6 (a) and (b) also 10 wt.% CH, 20 wt.% CH and 30 wt.% CH with 950°C shown at Figure 4.6 (c) to (e) were selected.

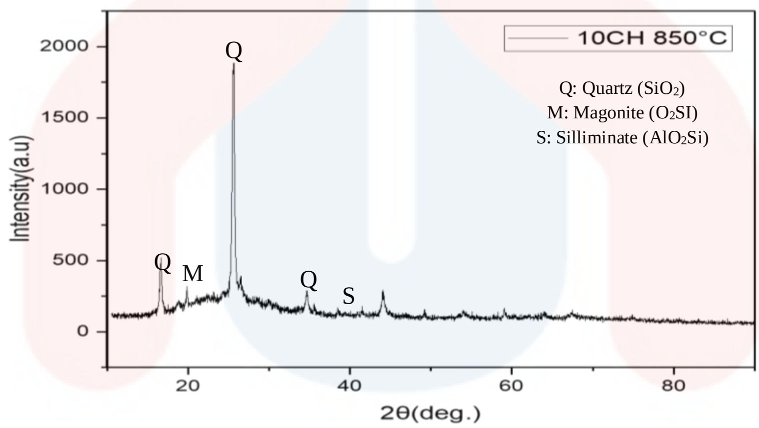


Figure 4.6 (a)

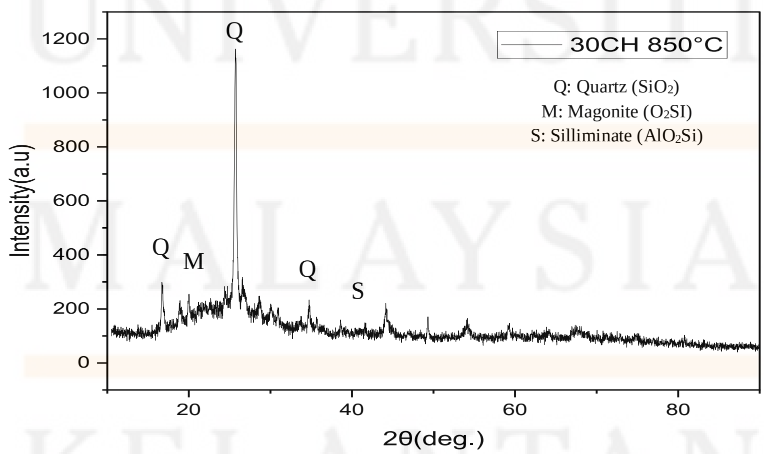


Figure 4.6 (b)

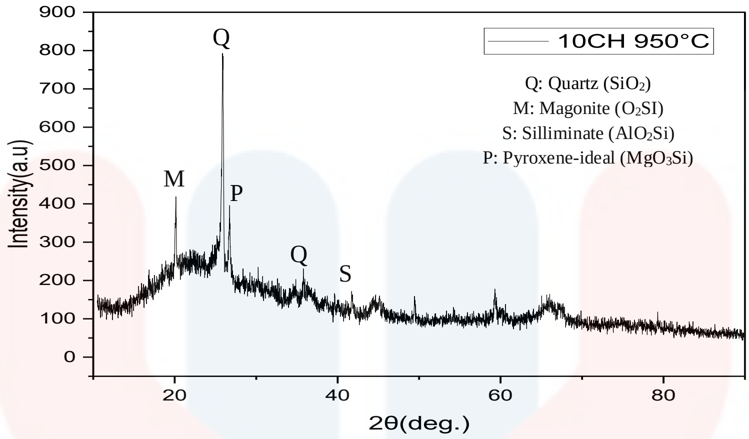


Figure 4.6 (c)

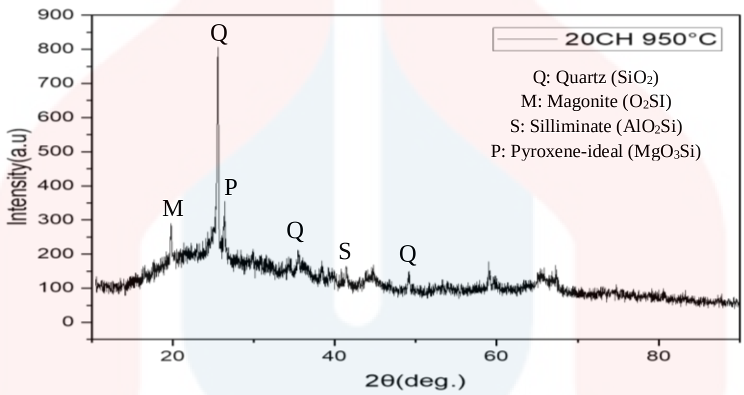


Figure 4.6 (d)

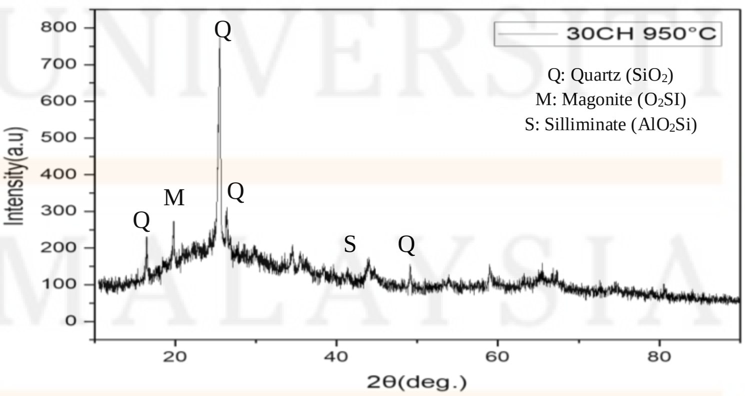


Figure 4.6 (e)

The different crystalline phases found in porous ceramic samples have a substantial impact on their mechanical qualities, including compressive strength. The presence of different crystalline phases affects the sensitivity of the ceramic and its compressive strength. According to the Mohs hardness scale, each crystalline phase has its hardness value, which correlates to the compressive strength. For instance, quartz, moganite, sillimanite, and pyroxene are the crystalline phases observed in the porous ceramic samples, and their respective hardness values contribute to the compressive strength of the material.

The higher compressive strength of the porous ceramic is due to reduced porosity, which is impacted by the crystalline phases present. Higher densification causes increased pore closure, which increases bulk density and, as a result, compression strength. As a result, changes in crystalline phases caused by differences in CH weight percentage and sintering temperature have a direct impact on the mechanical properties of porous ceramic samples. The relation between crystalline phases, compressive strength, and hardness is critical to understanding the behavior of porous ceramic materials and their applicability for particular applications.

The Mohs hardness scale provides a qualitative comparison of the hardness of minerals and crystalline phases, with diamond being the hardest at 10 and talc being the softest at 1. The hardness values of the crystalline phases present in the porous ceramic samples can be compared to those of known minerals to assess their impact on the material's mechanical properties. The study of crystalline phases and their influence on the properties of porous ceramic materials is essential for the development of tailored materials with desired mechanical characteristics. The specific hardness values of the crystalline phases observed in the porous ceramic samples can be determined using the Mohs hardness scale, and this information is valuable for understanding the variations in compressive strength and hardness observed in the different samples.

The relationship between crystalline phases, compressive strength, and hardness in porous ceramic materials is a complex and multifaceted aspect of material science, and it requires careful analysis and characterization to fully comprehend its implications for various applications.

CHAPTER 5

CONSLUSION AND RECOMMENDATIONS

5.1 Conclusion

The provided study concludes that the general central composite design (CCD) statistical experimental design approach can be significantly used to optimize coconut husk added and sintering temperature for the development of porous ceramic incorporated with coconut husk. The study addresses several conclusions based on the CCD statistical analysis:

a) The weight percentage of coconut husk added and sintering temperature was statistically proven to significantly influence the final properties (water absorption, apparent porosity, and bulk density) of the porous ceramic incorporated with coconut husk.

b) The results of statistical analysis, including model adequacy checking, analysis of variance (ANOVA), main effects and interaction plots, and regression model, were highly significant and proven for the porous ceramic incorporated with coconut husk.

c) The optimized properties (minimum water absorption, apparent porosity, and bulk density) of the porous ceramic incorporated with coconut husk were attained at 10 wt.% of coconut added and sintering temperature of 900 °C

The study's findings are in line with the use of coconut husk in porous ceramic composites, as seen in the search results. The search results support the potential of coconut husk to influence the properties of porous ceramics, as indicated in the study's conclusions.

5.2 Recommendations

In future research, it may be recommended to manufacture more uniform particle size to increase porosity in porous ceramic application:

a) The suggestion is to use an atomizer in a spray dryer to achieve more consistent particle sizes. Atomizer spray dryer application disperses the liquid or slurry into a regulated drop size spray, the coconut husk particles will be moist and the granule size more uniform. The atomizer spray dryer will accurately manage the particle size and amount of water injected throughout the moistening and granulation process.

b) Added silica and Pels (PHA) in the formulation for further improve the properties.

c) Optimize the range of wt.% coconut husk beyond 30% and range of sintering temperature beyond 950ºC.

d) The porous ceramic's characteristics will be improved, making its application more efficient.

REFERENCES

Ahmad, J., Majdi, A., Al-Fakih, A., Deifalla, A. F., Althoey, F., Ouni, M. H. E., & El-Shorbagy, M. A. (2022). Mechanical and durability performance of coconut fiber reinforced concrete: A State-of-the-Art Review. *Materials, 15*(10), 3601. https://doi.org/10.3390/ma15103601

Al-Naib, U. M. B. (2018). Recent advances in porous ceramics. In *InTech eBooks*. https://doi.org/10.5772/68104

Ayan, E., Baylan, N., & Çehreli, S. (2020). Optimization of reactive extraction of propionic acid with ionic liquids using central composite design. *Chemical Engineering Research and Design, 153*, 666–676. https://doi.org/10.1016/j.cherd.2019.11.015

Beal, B., Selby, A., Atwater, C. G., James, C., Viens, C., & Almquist, C. B. (2019). A Comparison of Thermal and Mechanical Properties of Clay Bricks Prepared with Three Different Pore-Forming Additives: Vermiculite, Wood Ash, and Sawdust. *Environmental Progress & Sustainable Energy, 38*(6). https://doi.org/10.1002/ep.13150

De Freitas Rosa, M., Medeiros, E. S., Malmonge, J. A., Gregorski, K. S., Wood, D. F., Mattoso, L. H. C., Glenn, G. M., Orts, W. J., & Imam, S. H. (2010). Cellulose nanowhiskers from coconut husk fibers: Effect of preparation conditions on their thermal and morphological behavior. *Carbohydrate Polymers, 81*(1), 83–92. https://doi.org/10.1016/j.carbpol.2010.01.059

Dele-Afolabi, T., Hanim, M. a. A., Jung, D., Ilyas, R. A., Calin, R., & Rahman, N. I. A. (2022). Rice husk as a Pore-Forming Agent: Impact of particle size on the porosity and diametral tensile strength of porous alumina ceramics. *Coatings, 12*(9), 1259. https://doi.org/10.3390/coatings12091259

Dewi, R., Agusnar, H., Alfian, Z., & Tamrin. (2018). Characterization of technical kaolin using XRF, SEM, XRD, FTIR and its potentials as industrial raw materials. *Journal of Physics*, *1116*, 042010. https://doi.org/10.1088/1742-6596/1116/4/042010

Eva, G., Zuzana, Z., & Willi, P. (2008). Porous Ceramic Made Using Potato Starch as a Pore-forming Agent. *Fruit, Vegetable and Cereal Science and Biotechnology, 3*(Special Issue 1).

Frost, J. (2022, July 22). *Understanding interaction effects in statistics*. Statistics by Jim. https://statisticsbyjim.com/regression/interaction-effects/

Gaspar, F., Бакатович, A. A., Давыденко, H. B., & Joshi, A. (2020). Building insulation materials based on agricultural wastes. In *Elsevier eBooks* (pp. 149–170). https://doi.org/10.1016/b978-0-12-819481-2.00008-8

German, R. M., Suri, P., & Park, S. J. (n.d.). *Review: liquid phase sintering* (Vol. 44). https://doi.org/10.1007/s10853-008-3008-0

Goel, G., & Kalamdhad, A. S. (2017). An investigation on use of paper mill sludge in brick manufacturing. *Construction and Building Materials*, *148*, 334–343. https://doi.org/10.1016/j.conbuildmat.2017.05.087

Hessing, T., & Pv, R. (2023, April 11). *Main Effects plot*. Six Sigma Study Guide. https://sixsigmastudyguide.com/main-effects-plot/

Hozman-Manrique, A. S., Garcia-Brand, A. J., Hernández-Carrión, M., & Porras, A. (2023). Isolation and Characterization of Cellulose Microfibers from Colombian Cocoa Pod Husk via Chemical Treatment with Pressure Effects. *Polymers*, *15*(3), 664. https://doi.org/10.3390/polym15030664

Hu, Y., Xiao, Z., Wang, H., Ye, C., Wu, Y., & Xu, S. (2019). Fabrication and characterization of porous CaSiO3 ceramics. *Ceramics International*, *45*(3), 3710–3714. https://doi.org/10.1016/j.ceramint.2018.11.034

IR Spectrum Table. (n.d.). https://www.sigmaaldrich.com/MY/en/technical-documents/technical-article/analytical-chemistry/photometry-and-reflectometry/ir-spectrum-table

Karaman, S., Günal, H., & Erşahin, S. (2008). Quantitative analysis of pumice effect on some physical and mechanical properties of clay bricks. *Journal of Applied Sciences*, *8*(7), 1340–1345. https://doi.org/10.3923/jas.2008.1340.1345

Kechik, A. A., Tharmizi, N. a. M., Ali, A., Taib, M. a. A., Mohamad, M., Boon, J. G., Masri, M. N., Mohamed, J. J., Zakaria, S., & Ter Teo, P. (2022). Optimization of wood saw dust (WSD) as pore forming agent for porous ceramic using response surface methodology (RSM) statistical design. *AIP Conference Proceedings*. https://doi.org/10.1063/5.0078327

Koay, S. C., Husseinsyah, S., & Azizi, F. N. (2013). Characterization and properties of recycled Polypropylene/Coconut shell powder composites: Effect of sodium dodecyl sulfate modification. *Polymer-plastics Technology and Engineering*, *52*(3), 287–294. https://doi.org/10.1080/03602559.2012.749282

Koçkal, N. U. (2015). Optimizing production parameters of ceramic tiles incorporating fly ash using response surface methodology. *Ceramics International*, *41*(10), 14529–14536. https://doi.org/10.1016/j.ceramint.2015.07.168

Kramer, D. G., Da Rocha, B. G., De Souza Pereira, M. C., Souza, R. S., Alves, C. R., Júnior, G. B., Nascimento, J. H. O. D., Quina, M. J., Ferreira, L. G., & Ladchumananandasivam, R. (2014). Determination of the biosorption of CD(II) by coconut fiber. *Journal of Agricultural Science and Technology: B*, *4*(4). https://doi.org/10.17265/2161-6264/2014.04b.007

Liu, T., Tang, Y., Han, L., Song, J., Luo, Z., & Lu, A. (2017). Recycling of harmful waste lead-zinc mine tailings and fly ash for preparation of inorganic porous ceramics. *Ceramics International*, *43*(6), 4910–4918. https://doi.org/10.1016/j.ceramint.2016.12.142

Mallamace, F., Corsaro, C., Mallamace, D., Vasi, S., Vasi, C., & Dugo, G. (2015). The role of water in protein's behavior: The two dynamical crossovers studied by NMR and FTIR techniques. *Computational and Structural Biotechnology Journal*, *13*, 33–37. https://doi.org/10.1016/j.csbj.2014.11.007

Mothé, C. G., & De Miranda, I. C. (2009a). Characterization of sugarcane and coconut fibers by thermal analysis and FTIR. *Journal of Thermal Analysis and Calorimetry*, *97*(2), 661–665. https://doi.org/10.1007/s10973-009-0346-3

Mothé, C. G., & De Miranda, I. C. (2009b). Characterization of sugarcane and coconut fibers by thermal analysis and FTIR. *Journal of Thermal Analysis and Calorimetry*, *97*(2), 661–665. https://doi.org/10.1007/s10973-009-0346-3

Mouiya, M., Bouazizi, A., Abourriche, A., Khessaimi, Y. E., Benhammou, A., Hafiane, Y. E., Taha, Y., Oumam, M., Abouliatim, Y., Smith, A., & Hannache, H. (2019). Effect of sintering temperature on the microstructure and mechanical behavior of porous ceramics made from clay and banana peel powder. *Results in Materials*, *4*, 100028. https://doi.org/10.1016/j.rinma.2019.100028

Murphy, C. (2021, February 26). *Efficient Experimental Design – Conor Murphy*. https://www.lancaster.ac.uk/stor-i-student-sites/conor-murphy/2021/02/26/efficient-experimental-design/

Nasr, M. S., Shubbar, A., Abed, Z. a. R., & Ibrahim, M. S. (2020a). Properties of eco-friendly cement mortar contained recycled materials from different sources. *Journal of Building Engineering*, *31*, 101444. https://doi.org/10.1016/j.jobe.2020.101444

Nasr, M. S., Shubbar, A., Abed, Z. a. R., & Ibrahim, M. S. (2020b). Properties of eco-friendly cement mortar contained recycled materials from different sources. *Journal of Building Engineering*, *31*, 101444. https://doi.org/10.1016/j.jobe.2020.101444

Obada, D. O., Dodoo-Arhin, D., Dauda, M., Anafi, F. O., Ahmed, A. S., & Ajayi, O. (2016). Potentials of fabricating porous ceramic bodies from kaolin for catalytic substrate applications. *Applied Clay Science*, *132–133*, 194–204. https://doi.org/10.1016/j.clay.2016.06.006

Ohji, T., & Fukushima, M. (2012). Macro-porous ceramics: processing and properties. *International Materials Reviews*, *57*(2), 115–131. https://doi.org/10.1179/1743280411y.0000000006

Papageorgiou, I. (2020). Ceramic investigation: how to perform statistical analyses. *Archaeological and Anthropological Sciences*, *12*(9). https://doi.org/10.1007/s12520-020-01142-x

Phonphuak, N., Saengthong, C., & Srisuwan, A. (2019). Physical and mechanical properties of fired clay bricks with rice husk waste addition as construction materials. *Materials Today: Proceedings*, *17*, 1668–1674. https://doi.org/10.1016/j.matpr.2019.06.197

Rashid, T., Kait, C. F., & Murugesan, T. (2016). A “Fourier Transformed Infrared” Compound Study of Lignin Recovered from a Formic Acid Process. *Procedia Engineering*, *148*, 1312–1319. https://doi.org/10.1016/j.proeng.2016.06.547

Salvini, V. R., Pandolfelli, V. C., & Spinelli, D. (2018). Mechanical properties of porous ceramics. In *InTech eBooks*. https://doi.org/10.5772/intechopen.71612

Sengphet, K., Satô, T., Fauzi, M. A., & Othman, R. (2013). Porous ceramic bodies using banana stem waste as a Pore-Forming agent. *Advanced Materials Research*, *858*, 131–136. https://doi.org/10.4028/www.scientific.net/amr.858.131

Seuba, J., Deville, S., Guizard, C., & Stevenson, A. J. (2016). Mechanical properties and failure behavior of unidirectional porous ceramics. *Scientific Reports*, *6*(1). https://doi.org/10.1038/srep24326

Smith, B. C. (2020, December 20). The C-O Bond, Part I: Introduction and the infrared spectroscopy of alcohols. *Spectroscopy Online*. https://www.spectroscopyonline.com/view/c-o-bond-i-introduction-and-infrared-spectra-alcohols-0

Susilawati, Siburian, J., Sihombing, Y. A., Ferdiansyah, B., & Pakpahan, S. N. Y. (2020). Fabrication and characterization of physical and mechanical properties based on clay and cacao rind porous ceramics. *AIP Conference Proceedings*. https://doi.org/10.1063/5.0003174

Vasić, M., Pezo, L., Vasić, M., Mijatović, N., Mitrić, M., & Radojević, Z. (2022). What is the most relevant method for water absorption determination in ceramic tiles produced by illitic-kaolinitic clays? The mystery behind the gresification diagram. *Boletín De La Sociedad Española De Cerámica Y Vidrio*, *61*(3), 241–251. https://doi.org/10.1016/j.bsecv.2020.11.006

Wagner, J. R., Mount, E. M., & Giles, H. F. (2014). Design of experiments. In *Elsevier eBooks* (pp. 291–308). https://doi.org/10.1016/b978-1-4377-3481-2.00025-9

Wisdom, A., Mark, U., Zeberu, E., & Adeleke, A. (2021). Optimisation of the physical properties of rice husk ash in ceramic materials using the response surface methodology. *Hungarian Journal of Industrial Chemistry*, *49*(1), 23–30. https://doi.org/10.33927/hjic-2021-04

Younis, U., Rahi, A. A., Danish, S., Ali, M. A., Ahmed, N., Datta, R., Fahad, S., Holátko, J., Hammerschmiedt, T., Brtnický, M., Zarei, T., Baazeem, A., Sabagh, A. E., & Glick, B. R. (2021). Fourier Transform Infrared Spectroscopy vibrational bands study of Spinacia oleracea and Trigonella corniculata under biochar amendment in naturally contaminated soil. *PLOS ONE*, *16*(6), e0253390. https://doi.org/10.1371/journal.pone.0253390