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Evaluation of rice husk ash and vermiculite materials in the production of ceramic tiles

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**EVALUATION OF RICE HUSK ASH AND VERMICULITE
MATERIALS IN THE PRODUCTION OF CERAMIC TILES**

Said Abeid

**A Dissertation Submitted in Partial Fulfilment of the Requirements for the Degree of
Master's in Science in Materials Science and Engineering of the Nelson Mandela
African Institution of Science and Technology**

Arusha, Tanzania

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ABSTRACT

The challenging issues in ceramic tiles are low mechanical strength, thermal discomfort and high production costs. Apart from strength the technical functions of floor tiles based on thermal insulation is very important in buildings with human occupation where thermal comfort is required. The objective of this work was to investigate the potential of rice husk ash (RHA) and vermiculite materials in production of ceramic tiles. Initially, a typical ceramic body composed of the mixture of vermiculite and RHA batched with clay, feldspar, quartz and kaolin was prepared. Ceramic bodies were then obtained from this ceramic mixture by pressing samples at a forming pressure of 35 MPa. These bodies were then fired at 1180°C in a laboratory furnace and finally the changes in the physical and mechanical properties caused by the introduction of vermiculite and RHA were tested and evaluated. The chemical composition of the raw samples was analysed by X-ray fluorescence (XRF) while the phase composition was investigated using X-ray diffraction (XRD). The morphology of the powdered samples was studied by using Scanning electron microscopy (SEM). The bulk density and open porosity of the sintered ceramic bodies were evaluated using Archimedes' principle while the flexural rupture strength was determined by the three point bending test method. The major chemical compounds in vermiculite raw sample were SiO₂, Al₂O₃ and Fe₂O₃ while RHA sample was found to contain mainly SiO₂. From the XRD analysis, vermiculite sample had crystalline vermiculite while RHA sample had amorphous silica at low temperature below 900°C and crystallized (tridymite) above 900°C. The results from physical, mechanical and thermal properties tests show that with addition of vermiculite and RHA, the percentage of porosity, water absorption, linear shrinkage and thermal insulation capacity were increasing while the bulk density, bending strength, thermal conductivity and effusivity of the fired ceramic bodies decreased. Among the studied compositions apart from the reference sample, tile bodies made from blend containing 20 wt. % vermiculite, 5 wt. % RHA and fired at 1180°C were found to have the best properties for the production of ceramic tiles. For this combination the values for water absorption, porosity, and linear shrinkage, bulk density, bending strength, thermal conductivity and effusivity were 7.60 %, 12.08 %, 3.29 %, 1.88 g/cm³, 18.54 MPa, 0.82 W/m.K, and 1284.33 W^{1/2}/m²K⁴ respectively. These values were close to the required standards for wall and floor tiles applications. This is an indication that vermiculite and RHA are suitable raw materials for the production of ceramic tiles with a required strength and thermal comfort.

DECLARATION

I, Said Abeid do hereby declare to the Senate of Nelson Mandela African Institution of Science and Technology that this dissertation is my own original work and that it has neither been submitted nor being concurrently submitted for degree award in any other institution.

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The above declaration is confirmed by

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CERTIFICATION

The undersigned certifies that he has read the dissertation, and found it to be in a form acceptable for examination by the Nelson Mandela African Institution of Science and Technology a dissertation entitled “Evaluation of rice husk ash and vermiculite materials in the production of ceramic tiles”, in fulfillment of the requirements for the degree of Master of Science (Materials Science and Engineering) of the NM-AIST.

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Date

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Name and signature of supervisor 2

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Date

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DEDICATION

This dissertation is dedicated to my lovely mother Rachel Daniel for her love in me.

TABLE OF CONTENTS

ABSTRACT.....	i
DECLARATION	ii
COPYRIGHT.....	iii
CERTIFICATION	iv
ACKNOWLEDGEMENT	v
DEDICATION	vi
LIST OF TABLES	ix
LIST OF FIGURES	x
LIST OF APPENDICES.....	xi
LIST OF ABBREVIATIONS AND SYMBOLS	xii
CHAPTER ONE.....	1
INTRODUCTION	1
1.1 Background information	1
1.2 Problem statement.....	3
1.3 Research problem justification	3
1.4 Objectives	3
1.4.1 Main objective	3
1.4.2 Specific objectives.....	3
1.5 Research questions.....	4
1.6 Significance of the study.....	4
CHAPTER TWO	5
LITERATURE REVIEW	5
2.1 Effect of quartz on strength of ceramic tiles	5
2.2 Ceramic tiles forming, firing and sintering.....	6
2.3 Standard chemical composition of raw materials for ceramic tiles production.....	8
2.4 General mineralogical structure of vermiculite and RHA	8
2.5 General microstructure of the final ceramic body	9
2.6 Standard physical, mechanical and thermal properties of ceramic tiles	9
2.7 Vermiculite	10
2.8 Rice husk ash (RHA)	10
CHAPTER THREE	12
MATERIALS AND METHODS.....	12

3.1 Pretreatment of ceramic samples' constituents.....	12
3.2 Proposed batch composition and green tile bodies preparation.....	13
3.3 Firing of tile bodies.....	14
3.4 Chemical, mineralogical and microstructure analyses.....	14
3.5 Determination of physical, mechanical and thermal properties of tile bodies.....	15
CHAPTER FOUR.....	18
RESULTS AND DISCUSSION.....	18
4.1 Chemical composition, phase and microstructural analyses of raw materials.....	18
4.2 Physical and mechanical properties of tile samples.....	21
4.2.1 Influence of vermiculite on the physical and mechanical properties of tile samples.....	22
4.2.2 Influence of RHA on the physical and mechanical properties of tile samples.....	23
4.3 Influence of vermiculite and RHA compositions on thermal properties of tile samples.....	26
CHAPTER FIVE.....	31
CONCLUSION AND RECOMMENDATIONS.....	31
5.1 Conclusion.....	31
5.2 Recommendations.....	31
REFERENCES.....	32
APPENDICES.....	36
RESEARCH OUTPUTS.....	43
Journal paper.....	43
Paper Presentation.....	44
Research poster.....	54

LIST OF TABLES

Table 1: Chemical characteristics of the raw materials used in the porcelain tile body (wt. %) (Dondi <i>et al.</i> , 1999; Vieira and Monteiro, 2007)	8
Table 2: Mix proportion of Raw Materials (Wt. %)	13
Table 3: Chemical composition of raw materials (wt. %)	18
Table 4: Physical-mechanical properties of sintered tile bodies sintered at 1180°C	21
Table 5: Physical and thermal properties of tile samples	27

LIST OF FIGURES

Figure 1: Variety of ceramic tiles	2
Figure 2: Schematic representation of the sintering mechanisms of two particles (De Jonghe <i>et al.</i> , 1986).....	7
Figure 3: Photomicrographs of vermiculite samples from three different mining sites at Kalalani area –Korogwe Tanga	12
Figure 4: Disc- and tile-shaped samples fabricated using vermiculite and RHA for physical and mechanical properties tests (a) green (b) fired (c) fired and labelled (d) samples with visual defects such as cracks, lamination and surface deformation	14
Figure 5: Determination of bulk density, water absorption and porosity using Archimedes principle.....	15
Figure 6: X-ray diffraction of (a) Vermiculite (b) RHA.....	20
Figure 7: SEM micrographs of (a) Vermiculite (b) RHA.....	20
Figure 8: Influence of vermiculite content on the physical and mechanical properties of the fired ceramic bodies (a) bulk density and flexural strength (b) water absorption, porosity and linear shrinkage	23
Figure 9: Influence of vermiculite and RHA content to the physical and mechanical properties of the fired ceramic bodies (a) bulk density and flexural strength (b) water absorption, porosity and linear shrinkage	25
Figure 10: The relationship between porosity, bulk density and flexural strength of the tile samples.....	26
Figure 11: The influence of vermiculite content on thermal conductivity and effusivity of tile samples.....	28
Figure 12: The influence of RHA content on thermal conductivity and effusivity of tile samples	29
Figure 13: Relationship of thermal conductivity and effusivity with percentage of porosity	30

LIST OF APPENDICES

Appendix 1: Batch formulations	36
Appendix 2: Physical and mechanical properties calculations	39
Appendix 3: Thermal properties calculations	41

LIST OF ABBREVIATIONS AND SYMBOLS

SEM	Scanning Electron Microscopy
XRD	X-Ray Diffractometry
XRF	X-Ray Fluorescent Spectroscopy
BD	Bulk Density
AP	Apparent Porosity
WA	Water Absorption
LS	Linear shrinkage
σ_s	Flexural strength

CHAPTER ONE

INTRODUCTION

1.1 Background information

Demand for ceramic tiles is increasing day by day and researchers are becoming interested in searching for new materials in order to develop tiles with good mechanical strength for household uses as well as for decoration purposes. Apart from strength, the technical functions of floor tiles based on thermal insulation is very important in buildings with human occupation where thermal comfort is required (Effting *et al.*, 2006). The technology of obtaining ceramic floor tiles that provide thermal comfort to the contact assists market niches little explored, as hot environments like swimming pools areas and cold environments like bedrooms and bathrooms (Effting *et al.*, 2007). In this study the influence of vermiculite and RHA on the physical, mechanical and thermal properties of ceramic tiles bodies has been tested and evaluated.

Ceramic tiles are composed essentially of clay and other inorganic raw materials such as quartz, feldspar and carbonates, the clay being the most important raw material. In fabrication of ceramic products, selection of raw materials must be taken into consideration because it plays vital role in the final ceramic body (Celik, 2010). A typical ceramic body composition will consist of 50 % clay, 25 % quartz and 25 % feldspar. Clay imparts rigidity to the ware. Feldspar as a flux provides a glassy phase in the microstructure. Quartz on the other hand lowers both drying and firing shrinkage and imparts rigidity to the wares. Mullite and glass constitute the major phases of final ceramic product. Other constituents of the fired ceramic body in minor levels are quartz, cristobalite, tridymite and corundum (Akwilapo and Wiik, 2003). The distinguishing factors in the properties of different ceramic products are brought about by variations in the proportion of these materials, the processing and the firing schedule adopted. Some of the clay based ceramic products are as shown in Fig. 1.



Roof tile



Wall tile



Floor tile

Figure 1: Variety of ceramic tiles

Quartz and kaolin are mostly preferred as a source of silica and alumina respectively and they have a great influence on the mechanical strength of ceramic tiles because when they are mixed and fired at very high temperature close to their melting points they form a very strong alumina-silica phase called mullite which controls the strength of the ceramic body. But quartz grains embedded in the glassy matrix have a deleterious effect on the mechanical strength mainly because of its α - β phase transformation of quartz crystals during cooling (Hassan and Maharaz, 2015) resulting to the development of stresses which initiate fracture (Carty and Senapati, 1998). Therefore, in most efforts to increase strength, emphasis has been placed on minimization of quartz content by partial or full replacement with other silica precursors in the porcelain formula because of its β to α phase inversion.

On the other hand, ceramic floor tiles are widely used in buildings, possessing technical and aesthetic functions; however the technical function becomes very important in constructions with human occupation where the thermal comfort is required. Thermal discomfort is characterized by hot floor surfaces in external environments which are exposed to sun radiation like swimming pools areas or by cold floor surfaces in internal environments like bed rooms and path rooms (Effting *et al.*, 2007). We can generally verify that in a lot of situations the ceramic tiles don't offer appropriate thermal comfort due to their high thermal conductivity, diffusivity and effusivity. Materials with low thermal conductivities are likely to have low density and hence thermal effusivity while their porosity is high. So thermal conductivity of porous materials decreases as porosity increases (Rhee, 1975), although high porosity generally implies low mechanical strength. Depending on materials selection, batch formulations and forming processes (processing techniques) it is possible to obtain porous ceramics with high mechanical strength. In this work, porous ceramic tile bodies were obtained by incorporating ceramic powder and two natural additives which are vermiculite

and rice husk ash (RHA) with various weight fractions; finally physical, mechanical and thermal properties (effusivity and conductivity) were tested and evaluated.

1.2 Problem statement

For ceramic tiles, the challenging issues are low mechanical strength, thermal discomfort and high production costs. Quartz grains embedded in the porcelain glassy matrix have a deleterious effect on the mechanical strength mainly because of its transformation during cooling resulting to the development of stresses which initiate fracture (Carty and Senapati, 1998). In most cases, ceramic tiles don't offer appropriate thermal comfort due to their high thermal conductivity, diffusivity and effusivity because materials with low porosity are likely to have high thermal conductivities. Also most of ceramic products especially tiles are produced at a temperature above 1000°C which is associated with high energy consumptions and hence high production costs.

1.3 Research problem justification

In most efforts to increase strength, emphasis has been placed on minimization of quartz in the porcelain formula because of the β to α phase inversion of quartz which occurs at 573°C during cooling (Hassan and Maharaz, 2015). In this case, partial replacement of quartz by rice husk ash (RHA) was expected to reduce the possibility of β to α phase inversion which may lead to development of cracks in the ceramic body while the addition of vermiculite was expected to increase porosity and hence reducing thermal conductivity of tile bodies. In this way the fired ceramic tile bodies would have the required thermal comfort close to that of the International standards.

1.4 Objectives

1.4.1 Main objective

To investigate the potential of rice husk ash and vermiculite materials in production of ceramic tiles.

1.4.2 Specific objectives

- (i) To characterize the raw materials, formulations and the fabricated ceramic bodies
- (ii) To design ceramic body formulations from vermiculite and rice husk ash with proportions that lead to the maximum formation of mullite phase during sintering.

- (iii) To investigate the influence of vermiculite and rice husk ash content on the physical, mechanical and thermal properties of the fired tile bodies.

1.5 Research questions

- (i) Are the chemical, phase and morphological compositions of raw materials, formulations and fired ceramic bodies similar to that used in traditional ceramic tiles production?
- (ii) What proportions of vermiculite and rice husk ash lead to the maximum formation of mullite phase during sintering?
- (iii) What is the influence of vermiculite and rice husk ash content on the physical, mechanical and thermal properties of the fired tile bodies?

1.6 Significance of the study

The knowledge from this study will help ceramists to produce low-cost lightweight ceramic tiles with a required thermal comfort and good mechanical strength using porous materials which are vermiculite and RHA. The study will lead to localization of ceramic tile manufacturing technology and immediate socio-economic impact of value added product from indigenous raw materials. Regarding environmental point of view, other significant benefits of using RHA can be looked based on avoided landfill disposal of the residue.

CHAPTER TWO

LITERATURE REVIEW

2.1 Effect of quartz on strength of ceramic tiles

Quartz which is among the constituents in the ceramic tiles formula as the major source of silica, it has grains embedded in the porcelain glassy matrix which have a deleterious effect on the mechanical strength. This is mainly because of its transformation during cooling which results in the development of stresses which initiate fracture (Hassan and Maharaz, 2015). Different studies have been done based on the replacement of quartz in ceramic products by other materials of nearly the same chemical composition. Reduction of quartz has been achieved through replacements by RHA (Prasad *et al.*, 2001, Kurama 2008; Haslinawati *et al.*, 2009), fly ash and blast furnace slag (Dana *et al.*, 2004) and silica fume (Prasad *et al.*, 2003). Replacement of quartz by rice husk ash (RHA) reduces the possibility of β to α phase inversion of quartz which occurs at 573°C during cooling which results into decrease of quartz particle volume and may lead to cracks in the ceramic body.

Based on strength of the porcelain systems three major hypotheses describing how the strength properties of porcelain formulations have been developed. These include the mullite hypothesis, the matrix reinforcement hypothesis and the dispersion strengthening hypothesis (Carty and Senapati, 1998). The mullite hypothesis suggests that porcelain strength depends on the interlocking of fine mullite needles. Specifically, the higher the mullite content and the higher the interlocking of the mullite needles, the higher is the strength (Ece and Nakagawa, 2002). Hence, the strength of porcelain depends on the factors that affect the amount and size of mullite needles, like the firing temperature and composition of alumina and silica in the raw materials (Bragança and Bergmann, 2003). On the other hand, the dispersion strengthening hypothesis states that dispersed particles in the vitreous phase of a porcelain body, such as quartz and mullite crystals, in the glassy phase of a porcelain body limit the size of Griffith flaws resulting in increased strength. The concept of the matrix reinforcement hypothesis concerns with the development of compressive stresses in the vitreous phase as a result of the different thermal expansion coefficients of dispersed particles or crystalline phases (usually quartz) and the surrounding vitreous phase (Hassan and Maharaz, 2015). The larger these stresses are the higher is the strength of the porcelain bodies. The phenomenon is known as the pre-stressing effect.

2.2 Ceramic tiles forming, firing and sintering

The fundamental steps used in the process of manufacturing ceramic tiles involve mixing, moulding, glazing and firing. Before firing the ceramic body is said to be green, means not fully processed or treated. This green body lacks hardness and strength, so it is heated to fix its shape and to achieve hardness and strength. Firing is the heat treatment process that sinters the ceramic material it is performed in a furnace called a kiln. In sintering, bonds are developed between the ceramic grains, and this is accompanied by reduction of porosity. Sintering means that the powdered particles start to stick to one another where they touch. The finer the ceramic particles are ground, the earlier the sintering will start and the stronger the bond will become. As the temperature rises above 600°C, the sintering of ceramic powders starts.

Ceramic sintering is part of the firing process used in the manufacture of pottery and other ceramic objects like tiles. Sintering occurs by diffusion of atoms through the microstructure. This diffusion is caused by a gradient of chemical potential, atoms move from an area of higher chemical potential to an area of lower chemical potential (Hassan and Maharaz, 2015). The different paths the atoms take to get from one spot to another are called sintering mechanisms. Figure 2 shows the six common sintering mechanisms:

- (i) Surface diffusion which is a diffusion of atoms along the surface of a particle
- (ii) Lattice diffusion from surface where the atoms from surface diffuse through lattice
- (iii) Vapor transport in which evaporation of atoms condense on a different surface
- (iv) Grain boundary diffusion where the atoms diffuse along grain boundary
- (v) Lattice diffusion from grain boundary where atoms from grain boundary diffuses through lattice
- (vi) Plastic deformation which is a dislocation motion that causes flow of matter (Hassan and Maharaz, 2015).

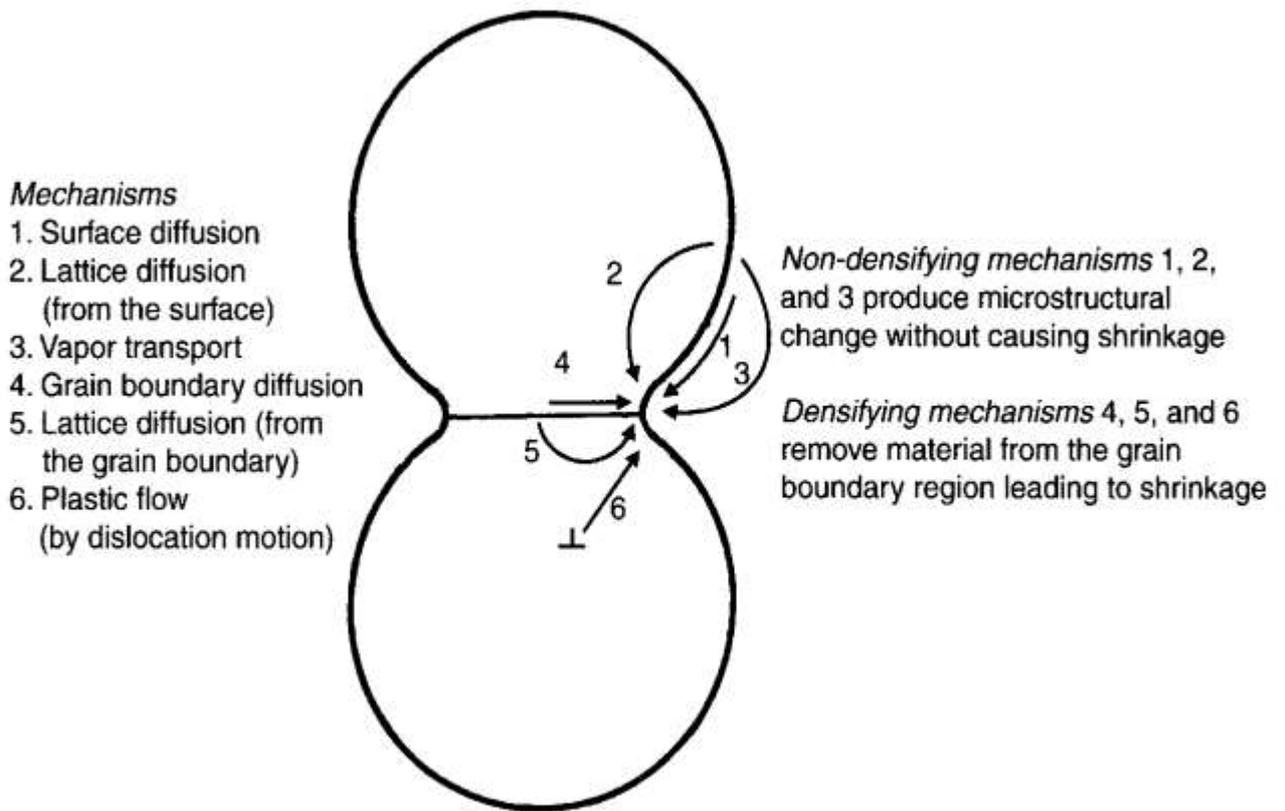
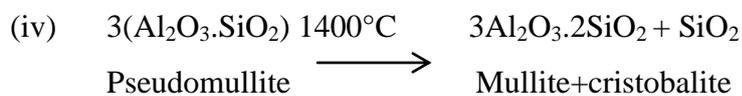
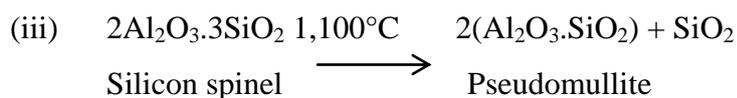
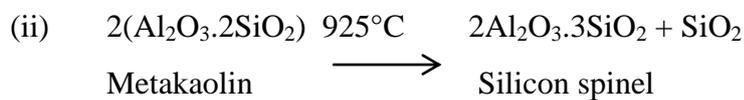
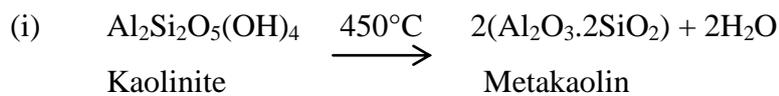


Figure 2: Schematic representation of the sintering mechanisms of two particles (De Jonghe *et al.*, 1986)

The chemical reactions occurring during the sintering process and microstructural development to form mullite phase from kaolinite materials can be outlined as follows (Worrall, 2013):



2.3 Standard chemical composition of raw materials for ceramic tiles production

The required chemical compositions for raw materials required for production of ceramic tiles are summarized in Table 1.

Table 1: Chemical characteristics of the raw materials used in the porcelain tile body (wt. %) (Dondi *et al.*, 1999; Vieira and Monteiro, 2007)

	Plastic clay	Kaolin	Feldspar	Quartz	Talc	Industrial body
SiO ₂	61.2 – 74.9	47.3 – 79.3	66.3 – 79.5	98.0 – 99.1	51.0 – 61.2	66.0 – 73.0
Al ₂ O ₃	19.2 – 31.5	15.0 – 36.0	11.5 – 20.1	0.65 – 0.8	0.3 – 8.0	17.1 – 23.2
Fe ₂ O ₃ + TiO ₂	0.6 – 3.2	0.2 – 1.9	0.05 – 1.0	0.07 – 0.30	0.4 – 0.9	0.6 – 2.1
CaO+ MgO	0.2 – 1.4	0.2 – 0.7	0.30 – 4.2	0.04 – 0.3	30.3 – 31.5	0.5 – 6.6
K ₂ O + Na ₂ O	0.9 – 5.7	0.2 – 2.8	6.1 – 13.3	0.1 – 0.2	0.2	4.0 – 6.8
LOI	4.7 – 10.0	5.0 – 12.8	0.2 – 1.8	0.2	0.57 – 7.0	3.3 – 4.2

Table 1 shows the range of variation for the main chemical compounds of typical raw materials used in the composition of porcelain tile bodies as well as a typical industrial porcelain tile body (Dondi *et al.*, 1999; Vieira and Monteiro, 2007). It is observed that in the industrial body, SiO₂ and Al₂O₃ are the most abundant oxides and the SiO₂ amount is higher than 60 wt. %. The colorant and opacifier oxides, Fe₂O₃ + TiO₂ must be maintained at low amount to avoid undesired color in the ceramic product. The alkaline oxides, which are also fluxing oxides K₂O + Na₂O, are essential to liquid phase formation. The earth alkaline oxides, CaO + MgO, must be maintained in the low amounts due to the possibility of excessive fluxing as well as undesired variation on the ceramic color. On the other hand the loss on ignition (LOI) is mainly associated with the dehydration of clay minerals. Organic matter oxidation as well as carbonates decomposition also contributes to this parameter. The LOI must be maintained in low amounts, normally lower than 6%, because excessive weight loss during the firing stage contributes to increase the porosity and the linear shrinkage.

2.4 General mineralogical structure of vermiculite and RHA

The expanded vermiculite consists mainly of vermiculite, phlogopite mica and hydrobiotites, likely an interstratified. Interstratified structure of vermiculite after milling does not contain a hydrated phase, vermiculite (two layers of water molecules in interlayer) and the mica-type

phase (zero water molecules in the interlayer) still remain but the crystallinity of vermiculite changes after the milling (Sutcu, 2015). Rice husk ash ceramic sintering at 900°C and above consists mainly of crystal phase of two forms, cristobalite and tridymite (Haslinawati *et al.*, 2009) while amorphous phase occur at low temperature 700°C and 800°C (El Wahed, 1991). Tridymite is the silica polymorph with lowest density and it is the only silica phase that able to accommodate interstitial K⁺ cations into its big cavities (Real *et al.*, 1996), where K⁺ cation is noted as the major impurity.

2.5 General microstructure of the final ceramic body

The final microstructure of a porcelain tile is generally formed by 55-65 mass-% amorphous phase, 20-25 mass-% of quartz and 12-16 mass-% of mullite dispersed in a vitreous matrix (Dondi *et al.*, 1999). Mullite is an important constituent in refractories, whitewares and structural clay products for kaolin is frequently used as the raw material. Early studies insist the importance of the extent of mixing during the production of porcelain. A green porcelain body contains micro-regions of varying composition which includes pure clay agglomerates and feldspar-enriched regions (Chen *et al.*, 2000). The size and extent of these regions is controlled by the raw powder particle size, level of agglomeration and degree of mixing. On heating, these regions react to form different types of mullite. Primary mullite is formed from decomposition of pure clay while secondary mullite from reaction of feldspar and clay and feldspar, clay and quartz. Additionally tertiary mullite may precipitate from alumina-rich liquid obtained by dissolution of alumina filler (Lee and Iqbal, 2001). The general microstructural features of commercial porcelains are coarse quartz grains surrounded by solution rims of almost pure silica glass embedded in a much finer matrix system.

2.6 Standard physical, mechanical and thermal properties of ceramic tiles

The optimum vitrification range of ceramic bodies is achieved when the porosity reaches a minimum value nearly to zero while the linear shrinkage is maximum. In fact the apparent porosity decreases with increasing firing temperature due to the formation of a glassy phase that is mainly originated from the feldspar (Márquez *et al.*, 2008). Firing above vitrification range results in a drastic fall of the physical properties due to forced expulsion of the entrapped gases, resulting in blisters and bloating (Márquez *et al.*, 2008). The required values for flexural rupture strength and water absorption are higher than 35 MPa and lower than 0.5 %, respectively (Vieira and Monteiro, 2007). According to German Institute for Standardization based on technical specification for density, DIN 51082; the densification

parameter of the bulk materials for the final fired ceramic body must be greater than 2 gcm^{-3} . Thermal conductivity of porous materials decreases as porosity increases and it is directly correlated with thermal effusivity and material density. Thermal conductivity of porcelain tiles is around 1 W/mK (Stulz and Mukerji, 1988). Another study based on thermal properties of ceramic tiles was done by addition of 75 wt. % of porous industrial wastes and thermal conductivity and effusivity were found to be 0.21 W/mK and $387.5 \text{ mK}^{-1}\text{W}^{1/2}$ respectively (Effting *et al.*, 2007).

2.7 Vermiculite

Vermiculite is a clay mineral produced by the decompositions of mica and occurs as large crystals of mica-like lamellar structure that quickly expands on heating to produce a lightweight material (Önen, 2016). In Tanzania vermiculite can be found in places like Kalalani Tanga and Mikese Morogoro (Marwa *et al.*, 2009) (Fig. 2). Another vermiculite deposit of high quality has been seen at Kwekivu area in Kilindi District but it is not mined yet due to high costs associated with transport to the nearby railway station (Marwa *et al.*, 2009). In its expanded form, vermiculite has a very low density and thermal conductivity, which makes it attractive for use as a lightweight construction aggregate, and thermal insulation filler (Silva *et al.*, 2014). Vermiculite wall and roofing tiles are much stronger and highly heat resistive (Kumar *et al.*, 2015) which provide thermal comfort when used in houses. A low sintering temperature of a ceramic body due to addition of vermiculite is desirable to reduce energy costs; although the temperature has to be high enough in order to achieve an acceptable strength. The people below poverty line will live in single roofed buildings, it is quite uneconomical in usage of air conditioning, and in such condition it is economical in providing them with the vermiculite roofing.

2.8 Rice husk ash (RHA)

Rice husk ash obtained from the incineration of husk is rich in silica (87-97 %) with small amounts of alkali and other trace elements (Prasad *et al.*, 2003). Silica in Rice Husk Ash (RHA) exists in amorphous or crystalline form, depending upon the production conditions. In Asian countries, Rice husk (RH) is considered as a suitable energy and silica resource. By heating at higher temperatures, the unburned carbon can be removed from the ashes, but this leads to the crystallization of the ash from amorphous silica into cristobalite or tridymite (Haslinawati *et al.*, 2009). In Tanzania Rice Husk is abundant and as an agricultural residue it

is mostly used as an alternative source of energy especially in rural areas. Also rice husk used as poultry and cattle feed in urban and rural areas.

Previous studies discovered that, RHA could be used as an insulator in the steel industry as well as pozzolanic material in the cement industry (Andreola *et al.*, 2012). This is because RHA is an excellent insulator having low thermal conductivity, high melting point, low bulk density and high porosity. Also in the manufacture of concrete there are two areas in which RHA is used. Firstly in the manufacture of low cost building blocks and secondly in the production of high quality cement. The addition of RHA to cement enhances the cement properties like early strength of concrete and formation of a calcium silicate hydrate gel around the cement particles, which becomes highly dense and less porous (Andreola *et al.*, 2012). With the addition of RHA weight density of concrete reduces by 72-75%. Thus, RHA concrete can be effectively used as light weight concrete for the construction of structures where the weight of structure is of supreme importance (Nagrle *et al.*, 2012). The use of RHA considerably reduces the water absorption of concrete. Hence concrete containing RHA can be effectively used in places where the concrete can come in contact with water or moisture (Nagrle *et al.*, 2012). The use of RHA in ceramic body improves compressive strength compared to the body without RHA (Hassan and Maharaz, 2015). The reduction in the vitrification temperature range of the mixes with addition of vermiculite and RHA during firing would also contribute significantly to the economical production of ceramic tiles.

Currently no research has been done to test the potential of the combination of both vermiculite and RHA ceramic tiles applications. In this work, the changes in the physical, mechanical and thermal properties of ceramic tile samples caused by the introduction of different weight fraction contents of vermiculite and RHA were studied and evaluated in order to test the suitability of the materials in ceramic tiles production.

CHAPTER THREE

MATERIALS AND METHODS

3.1 Pretreatment of ceramic samples' constituents

Materials that were involved in this study include V₁, V₂ and V₃ vermiculite samples from three mining sites; Red Garmet, Red Safaya, and Pink Safaya respectively all found at Kalalani village in Tanga region eastern part of Tanzania, as shown in Fig. 3. Rice husk samples were taken from Bahi district in Dodoma region central part of Tanzania. Pugu kaolin, Same clay, quartz and feldspar were obtained from Geological Survey of Tanzania Laboratory in Dodoma.



Figure 3: Photomicrographs of vermiculite samples from three different mining sites at Kalalani area –Korogwe Tanga

The procedure for preparing rice husk powder consist of washing RH in distilled water in order to remove clay and rock impurities and subsequently drying in an oven at 120°C for 12 hrs. Then 30g of dry husk was reacted with 2 M HCl at 25°C in a 500 mL under constant agitation for 2 hrs. This was done to remove metallic impurities and organics contained in rice husk before calcinations. The husk was then washed with distilled water till neutral pH was obtained and then dried. The dried RH was later heated at 700°C for 2 hrs at a heating rate of 10°C/min under air atmosphere to obtain carbon free white ash. At 700°C and below, ash rich in amorphous silica is formed which is highly reactive. Above 700°C, crystalline silica which is far less reactive is obtained. On the other hand, Pugu kaolin, vermiculite, quartz, and feldspar samples were separately dried at 130°C in the oven for 24 hrs and then ground by using ball mill for 6 hrs. The powdered samples were sifted through a sieve of 150 µm pore size. Grinding and sieving were repeated until almost all the materials passed through the sieve.

3.2 Proposed batch composition and green tile bodies preparation

Vermiculite was gradually incorporated into the ceramic samples with a composition range of 20 to 35 wt. % to make the first batch of tile samples with vermiculite, T₂ to T₅. Then the composition range of 5 to 20 wt. % for RHA was added together with 20 to 35 wt. % of vermiculite in order to complete the second batch of tile samples, T₆ to T₉. The first test sample T₁ used as a control sample contained neither vermiculite nor RHA, (Table 2). The compositions were mixed with 5 to 6 wt. % water using ball mill for 90 min.

Table 2: Mix proportion of Raw Materials (Wt. %)

Tile code	Vermiculite	Pugu kaolin	Same clay	RHA	Quartz	Feldspar
T ₁	—	10	60	—	10	20
T ₂	20	10	40	—	10	20
T ₃	25	10	35	—	10	20
T ₄	30	10	30	—	10	20
T ₅	35	10	25	—	10	20
T ₆	20	10	35	5	10	20
T ₇	25	10	30	10	5	20
T ₈	30	10	22.5	15	2.5	20
T ₉	35	10	15	20	—	20

Discs-shape ceramic bodies of 3 cm diameter and 8 mm in thickness were prepared in order to measure the physical properties while square tiles of $140 \times 64 \times 14 \text{ mm}^3$ were prepared for bending strength measurements (Fig. 4a). The tile samples were subsequently oven-dried at 110°C for 24 hrs followed by cooling at room temperature for 1 h. The dimension and weight of each green (dried) body was recorded before sintering.

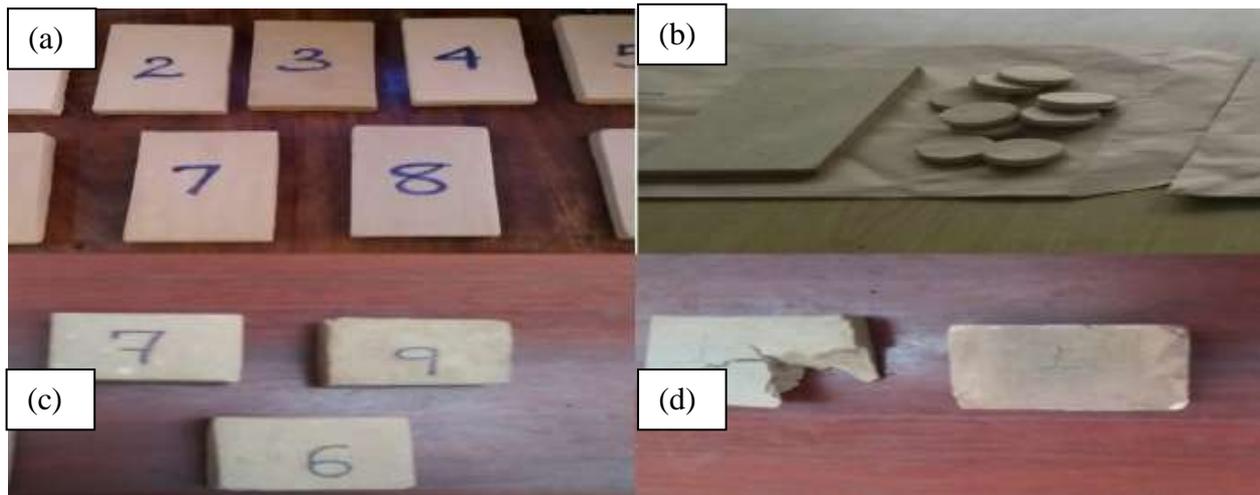


Figure 4: Disc- and tile-shaped samples fabricated using vermiculite and RHA for physical and mechanical properties tests (a) green (b) fired (c) fired and labelled (d) samples with visual defects such as cracks, lamination and surface deformation

3.3 Firing of tile bodies

The ceramic bodies were then fired at 1180°C for 2 hrs soaking time, at a heating rate of 50°C per minute in a Carbolite box furnace (RHF 14/8 Model) manufactured by Keison products Inc, UK. The firing parameters tests were all performed at Tanzania Portland Cement Company Limited (TPCC). After firing, the samples were permitted to cool down to room temperature inside the kiln for 24 hrs and then the dimensions and weights of the fired tile bodies were recorded. Finally, the fired bodies (Fig. 4 b) were labeled (Fig. 4 c) and then stored in airtight containers ready for mechanical tests and further characterizations.

3.4 Chemical, mineralogical and microstructure analyses

The chemical compositions of raw materials were studied at Geological Survey of Tanzania Laboratory using X-Ray Fluorescence (XRF) PANalytical, Minipal4 (PW4030)-Rh X-Ray Tube, operating at 30 kV, 0.002 mA. The amorphous and crystalline structures were identified by using Ultima IV Rigakudiffractometer operating at tube voltage and current at 40 kV and 44 mA, respectively using monochromatic Cu-K α radiation. Diffraction patterns were recorded by scanning from 5° to 75° (2 θ / θ) in steps of 0.02° (2 θ / θ) at a rate of 2 sec/step. The morphological features were studied by using Scanning electron microscopy (FEI Nova Nano SEM 450, 2 kV). The SEM and XRD analyses were all carried out at the University of Connecticut, USA.

3.5 Determination of physical, mechanical and thermal properties of tile bodies

Fired ceramic bodies were inspected for any visual defects such as cracks, lamination and surface deformation. Thus, good quality ceramic bodies were selected and used for determination of the physical and mechanical properties. The physical and mechanical properties tests of the fired tile bodies were all carried out at Tanzania Bureau of Standards (TBS) - Materials testing laboratory.

The bulk density and open porosity of the sintered ceramic bodies were evaluated using Archimedes 'principle which involved drying tiles in an oven at 110°C for 24 hrs and cooling in desiccators. The dry mass of the tiles (W_d) was then measured, followed by water impregnation, which involved boiling tiles in distilled water for 5 hrs and then left soaked in water for an additional 24 hrs at room temperature. After impregnation, the suspended mass (S) of each tile body was recorded. The saturated mass (W) was measured after removing all excess water from the surface by using moistened cotton cloth.



Figure 5: Determination of bulk density, water absorption and porosity using Archimedes principle

The bulk density calculated using the formulae:

$$B. D = \left[\frac{W_d}{W-S} \right] \cdot D \dots\dots\dots (1)$$

While porosity was given by the following expression:

$$P = \left[\frac{W-W_d}{W-S} \right] \cdot 100\% \text{ (Escalera, 2015)} \dots\dots\dots (2)$$

Where W_d is dry weight, W is soaked weight, S is suspended weight, and D is density of water. Experiments were carried out at a room temperature of 18°C, in which the density of

water is 1.00 g/cm³. The water absorption A was calculated as the ratio of the mass of water absorbed to the mass of the dry specimen, given by the expression:

$$A = \left[\frac{W - W_d}{W_d} \right] \cdot 100\% \quad (\text{Olasupo and Borode, 2009}) \dots\dots\dots (3)$$

While linear shrinkage (L.S) was calculated by measuring dimensions of the prepared specimen before and after firing.

$$\text{Generally } L.S = \frac{L_g - L_f}{L_g} \cdot 100\% \quad (\text{Viruthagiri et al., 2009}) \dots\dots\dots (4)$$

Where L_g and L_f are the length (mm) of green and fired tiles respectively. The flexural rupture strength was determined by the three point bending test in a bending strength tester (MEGA 10-200-10 DS) manufactured by Prufsysteme Inc. in Germany, 2001. The tests were performed at Tanzania Bureau of Standards (TBS) - Materials testing laboratory. On the other hand thermal properties, conductivity and effusivity were calculated by using Equation 7 and 8, respectively.

Thermal conductivity was calculated according to ISO 8301 (Standard, 1991) as follows:

$$\frac{K}{K_0} = \frac{1 - P}{1 + nP^2} \dots\dots\dots (5)$$

Where K is the thermal conductivity of a porous ceramic body, K₀ is the thermal conductivity of a porous free ceramic body, P is the volume fraction of porous, and n is a constant. According to Sugawara and Yoshizawa, (1962) measurements on thermal conductivity of a ceramic tile at 70⁰C, n corresponds to 3 and K₀ = 1.65 W/m.K. The equation becomes:

$$K = 1.65 \left(\frac{1 - P}{1 - 3P^2} \right) \dots\dots\dots (6)$$

Good correlation occurs in the porosity interval from 0 to 50% for n = 4.5 and

K₀= 1.6W/mK (Carneane Effting *et al.*, 2007). And the equation becomes:

$$K = 1.60 \left(\frac{1 - P}{1 - 4.5P^2} \right) \dots\dots\dots (7)$$

According to Carneane Effting (2007) thermal effusivity is given by the expression

$$\varepsilon = \sqrt{K\rho c} \dots\dots\dots (8)$$

Where ϵ is thermal effusivity, K is the coefficient of thermal conductivity in W/mK, ρ is density in kg/m³ and C is specific heat capacity in J/kgK. The human foot is considered as having a thermal conductivity of 0.37 W/m.K, density of 1000 kg.m⁻³ and specific heat of 1000 J/kgK. Therefore, the resulting thermal effusivity is 600 Ws^{1/2}/m²K⁴ (Carneane Effting *et al.*, 2007). The ceramic materials (porous or non-porous one) show specific heat values very close to 1 kJ/kgK.

CHAPTER FOUR

RESULTS AND DISCUSSION

4.1 Chemical composition, phase and microstructural analyses of raw materials

The presence of various compounds within the raw materials can be seen in Table 3.

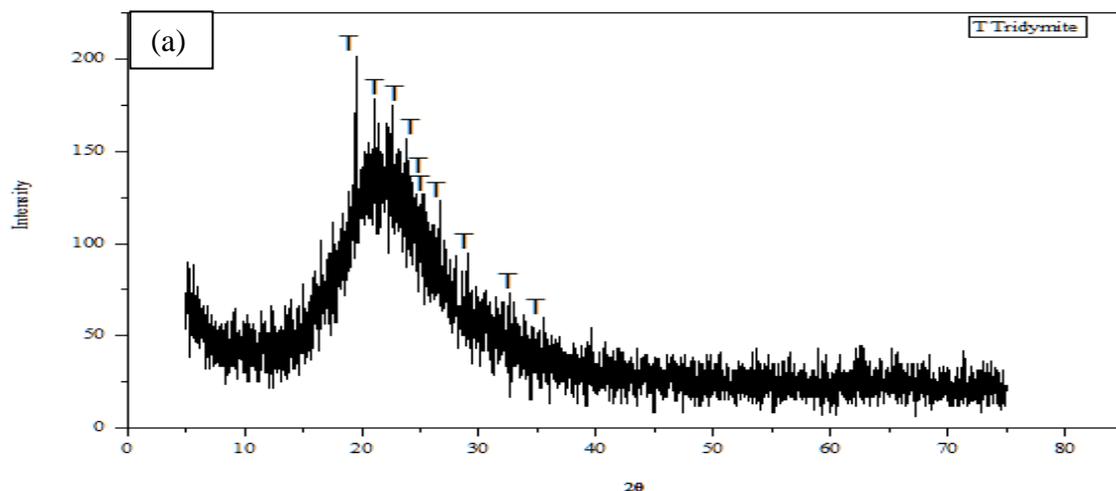
Table 3: Chemical composition of raw materials (wt. %)

	Vermiculite	Pugu kaolin	Same clay	RHA	Feldspar	Quartz
SiO ₂	62.10	60.00	60.40	92.10	57.10	97.70
Al ₂ O ₃	9.20	30.30	13.90	—	14.00	—
Fe ₂ O ₃	18.70	3.95	1.40	2.26	1.05	—
TiO ₂	1.83	1.22	0.14	0.18	—	—
CaO	0.81	0.39	0.02	2.98	—	—
MgO	6.10	—	—	—	—	—
Na ₂ O	0.05	0.04	—	—	—	—
K ₂ O	0.11	2.14	22.60	0.96	23.50	—
LOI	5.76	1.96	1.56	1.52	4.35	2.30

It is evident that SiO₂ is the major composition in the raw materials with 92.1 wt. % in RHA, 97.7 wt. % in quartz, and 62.10 wt. % in vermiculite. Alumina was high in Pugu kaolin with 30.30 wt. % and lowest in vermiculite with 9.20 wt. % while RHA and quartz showed no any indication of alumina. Silica and alumina in the raw materials are to be taken into account as they play a play very important role during sintering. Silica is a glass former while alumina is a refractory which gives strength and durability to a ceramic body. Vermiculite samples showed high percentage of Fe₂O₃ at 18.70wt. %. The flux oxide, K₂O which is essential for liquid phase formation and lowering the melting points of silica in the ceramic body, was high in feldspar than in all other raw materials with 23.5 wt. % although Na₂O was only present in Pugu kaolin and vermiculite in a small amount less than 1 wt. %. The opacifier TiO₂ which makes the ceramic body opaque was high in vermiculite and Pugu kaolin with 1.83 wt. % and 1.22 wt. % respectively, but was too low in the remaining samples with less than 1 wt. %. Another flux oxide, CaO was present in all raw samples except in feldspar and quartz and the highest amount was obtained in RHA with 2.98 wt. % while MgO was present

in vermiculite only with 6.10 wt. %. The fluxing oxides and colorants must be maintained at low amount to avoid undesired color and the possibility of excessive fluxing in the ceramic product (Vieira and Monteiro, 2007). In this study RHA had a chemical composition of 92.10 wt. % silica which is too close to other authors' findings 93.70 wt. % (Hassan and Maharaz, 2015), 93.67 wt. % (Haslinawati *et al.*, 2009), and 94.95 wt. % (Della *et al.*, 2002). Also the amount of silica in Kalalani vermiculite is close to the required standard in production of ceramic tiles 47.3 to 79.3 wt. % as reported by Vieira and Monteiro (2007). Vermiculite raw sample shows a loss on ignition of about 5.76 % which is the highest compared to that of the remaining samples. This value was related to the presence of organic residues where the decomposition of carbonates and sulphates produced a significant weight loss at a temperature above 1000°C (Table 3).

Figure 6 shows the X-ray diffractograms of the vermiculite and RHA. The XRD patterns of raw vermiculite show the presence of vermiculite as the major phase which was identified by peaks around 5° and 31.5° 2θ (Fig. 6 a). These results are close to XRD results of Kalalani vermiculite according to work done by Marwa (2009) which showed vermiculite as the only phases in the sample. In Figure 6 b the evolution of the crystalline phase from the amorphous silica present in the RHA can be seen. The XRD pattern shows the presence of tridymite as the major phase which was identified by peaks around 20.76° and 39.49° 2θ. According to Haslinawati (2009), silica of RHA is amorphous at low temperatures between 700 and 800°C. Firing RHA at 900°C and above produces crystal phases of two forms, cristobalite and tridymite (Haslinawati *et al.*, 2009). The transformation of amorphous silica to a cristobalite phase occurs at a temperature interval between 550 and 950°C (Escalera, 2015).



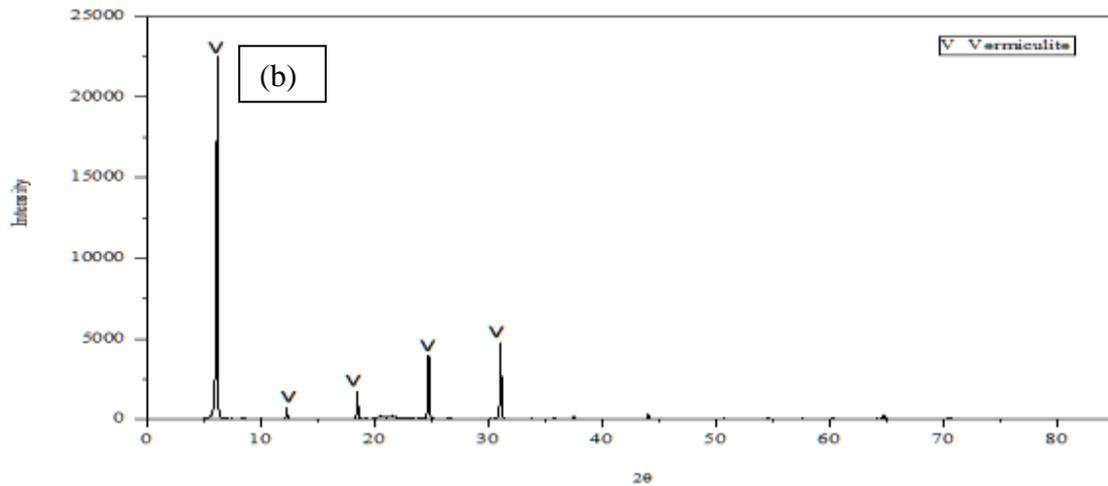


Figure 6: X-ray diffraction of (a) Vermiculite (b) RHA

The SEM micrographs of the raw vermiculite and RHA powder samples (Fig. 7) show the morphology of the vermiculite and RHA particles. Some of the detected phases are identified based on local chemical analysis by EDS.

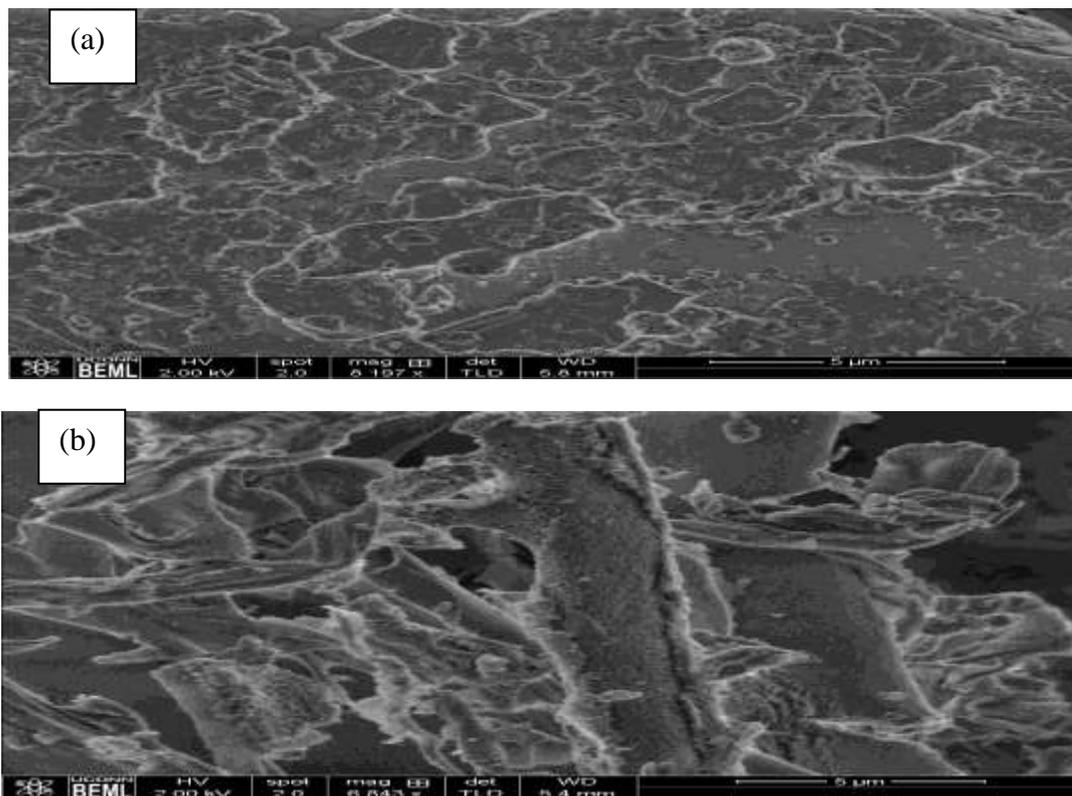


Figure 7: SEM micrographs of (a) Vermiculite (b) RHA

Figure 7 (a) shows the morphology of vermiculite sample containing flakes with individual layer crystals which are very closely spaced. The milled vermiculite flakes have a size range of smaller than 150 μm . The images show that vermiculite is more porous than porcelain and hence increasing in vermiculite content cause an increase in porosity size and amount. These results have a good agreement with bulk density and porosity results of the tile samples.

Figure 7 (b) shows the scanning electron microscopy of RHA powder which indicates that the ash is siliceous in nature with a porous structure and consists of quartz grains of various sizes and irregular shapes; these results agree with other authors as per Hassan and Maharaz (2015), Farooque (2009) and Zhang and Malhotra (1996). The porous nature of RHA and its honeycombed structure is responsible for its high specific surface and this is a good indication for its suitability in ceramic tiles applications (Farooque *et al.*, 2009).

4.2 Physical and mechanical properties of tile samples

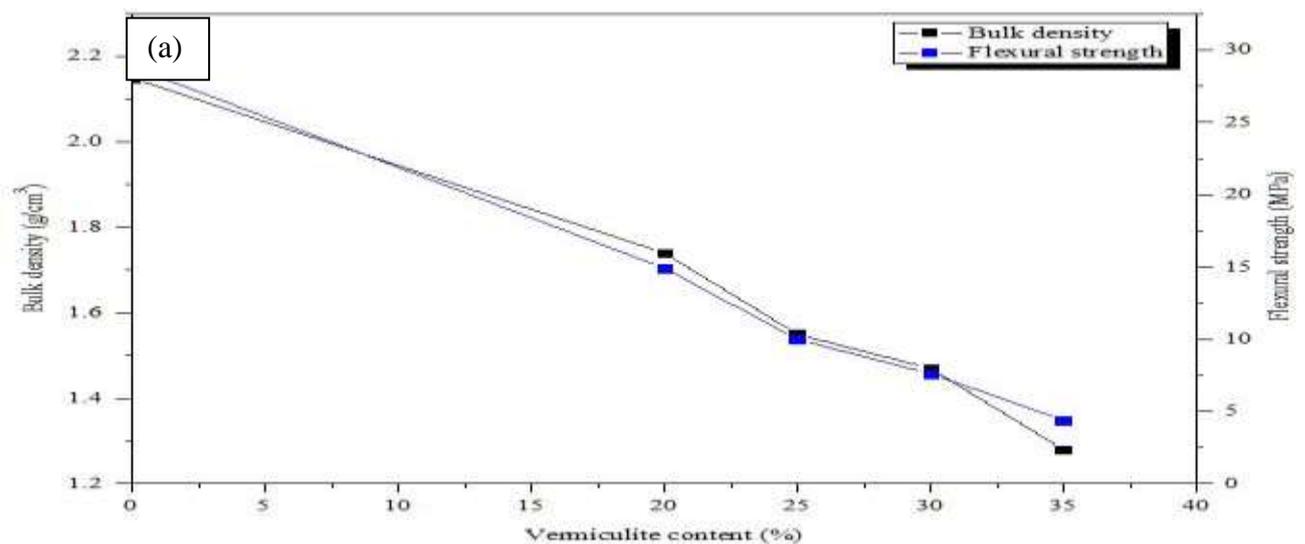
The results for this study on the physical and mechanical properties tests of the ceramic samples fabricated using vermiculite and RHA are presented below and well summarised in Table 4. The symbols WA, P, BD, LS and ∂_{fs} are water absorption, porosity, bulk density, linear shrinkage and flexural strength, respectively.

Table 4: Physical-mechanical properties of sintered tile bodies sintered at 1180°C

Tile code	WA (%)	P (%)	BD (g/cm ³)	LS (%)	∂_{fs} (MPa)
T ₁	2.17	5.72	2.15	1.02	28.86
T ₂	8.21	13.88	1.74	3.85	14.90
T ₃	10.42	15.10	1.55	5.88	9.99
T ₄	13.56	19.26	1.47	9.88	7.61
T ₅	16.81	21.29	1.28	12.00	4.40
T ₆	7.60	12.08	1.88	3.29	18.54
T ₇	9.69	13.95	1.70	5.71	14.59
T ₈	10.87	17.16	1.62	6.07	12.03
T ₉	13.34	19.74	1.46	8.55	8.50

4.2.1 Influence of vermiculite on the physical and mechanical properties of tile samples

The bulk densities of fired tile bodies were decreasing with addition of vermiculite from 20 to 35 wt. % (Fig. 8 a). The maximum bulk density was 1.74 g/cm^3 obtained when vermiculite was 20 wt. % while the minimum was 1.28 g/cm^3 obtained when vermiculite was added up to 35 wt. % The bulk density of the control sample which had 0 wt. % vermiculite was 2.15 g/cm^3 (Table 4). Decrease of bulk density of tile bodies with addition of vermiculite was mainly caused by low unit weight and density of raw vermiculite (Sutcu, 2015). The expanded vermiculite has the density ranging between 0.2 to 0.3 g/cm^3 (Önen *et al.*, 2016). These values are low compared to that of clay and kaolin which their bulk density is about 0.8 g/cm^3 . Hence, increase of vermiculite content in the samples led to decrease in bulk densities. On the other hand results show that, bending strength of fired ceramic bodies decrease with addition of vermiculite content (Fig. 8 a). The reference sample had bending strength of 28.86 MPa when the amount of vermiculite in the body was 0 wt. % and this value decreased up to 4.40 MPa when vermiculite content reached 35 wt. % which was the maximum vermiculite content for this study. The maximum bending strength for tile bodies with vermiculite as the only additive material was 14.90 MPa obtained when the vermiculite composition was 20 wt. %. This tile body had the strength value close to the required standard for wall tiles while the remaining compositions had values less than 14.90 MPa. According to commercial standards the minimum strengths for wall and floor tiles are 16 MPa and 22 MPa respectively ISO 10545 (Matteucci *et al.*, 2002).



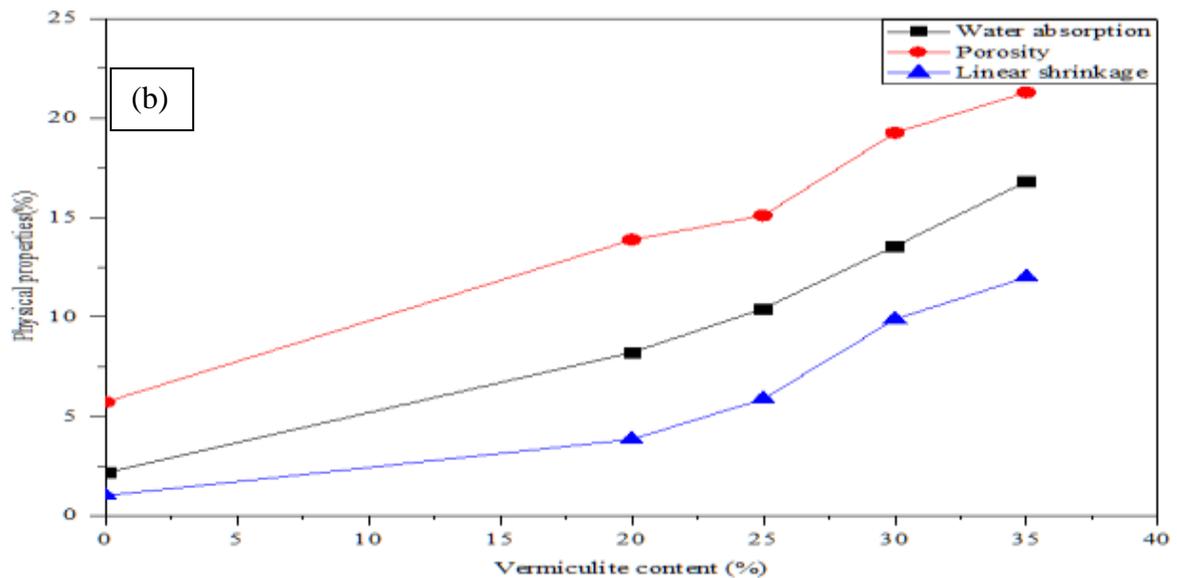


Figure 8: Influence of vermiculite content on the physical and mechanical properties of the fired ceramic bodies (a) bulk density and flexural strength (b) water absorption, porosity and linear shrinkage

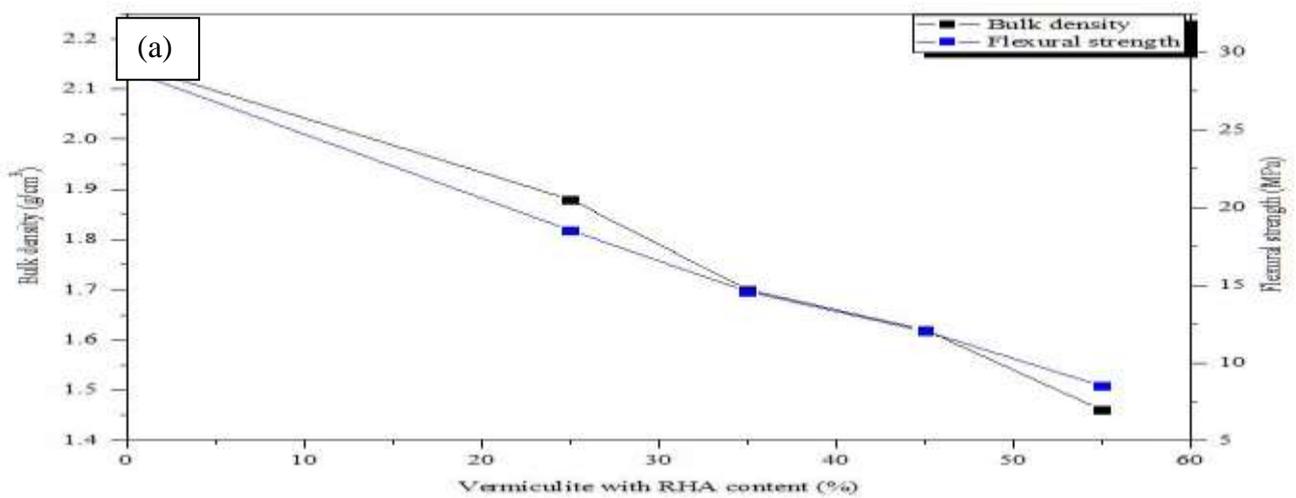
Figure 8 b shows the influence of vermiculite content on water absorption, porosity and linear shrinkage of the fired tile samples. The percentage of porosity of the fired ceramic bodies increase with increase of the vermiculite content. The minimum value was 5.72 % when vermiculite content was 0 wt. % and the highest was 21.29 % obtained when the vermiculite content was 35 wt. % The tile body with 20 wt. % vermiculite had porosity value close to the required international standard for wall and floor tiles applications which is 7 to 12 % of total porosity (Vieira and Monteiro, 2007). Since water absorption is directly related to open porosity, its value also increases with the increase of vermiculite content too. With addition of vermiculite the water absorption increased from 2.17 % of the control tile sample up to 16.81 % which was the highest and was obtained when vermiculite content reached 35 wt. %. The linear shrinkage of the tile samples increased with the increase of vermiculite content with values ranging from 1.02 % of the control sample to 12.00 % when vermiculite was 35 wt. %. This is because during firing vermiculite lose water moisture and other volatile (carbon contents) materials, this leads to reduction in particles size and hence dimension.

4.2.2 Influence of RHA on the physical and mechanical properties of tile samples

The bulk density of the tile samples decreased with increase of RHA content. The control tile sample had the bulk density of 2.15 g/cm³ when RHA was zero and this value decreased up to 1.46 g/cm³ when RHA was added up to 20 wt. % together with 35 wt. % vermiculite (Fig. 9 a). The maximum bulk density of samples with RHA was 1.88 g/cm³ obtained when RHA

was added up to 5 wt. % together with 20 wt. % vermiculite. Bulk densities of ceramic bodies with both vermiculite and RHA are higher than those with vermiculite only because vermiculite is more porous than RHA. The incorporation of RHA in the range of 5 to 20 wt. % in the tile composition had a favorable feature in bulk production in view of the enhancement of the densification process as well as the wide vitrification range of body mixes.

Since the bulk density is directly proportional to strength of a material the addition of both vermiculite and RHA caused the bending strengths of the fired ceramic bodies to decrease from 28.86 MPa when the additives were 0 wt. % to 8.50 MPa when RHA was 20 wt. % and with 35 wt. % vermiculite (Fig. 9 a). From these results, the maximum bending strength (apart from the reference sample value) was 18.54 MPa obtained when RHA was 5 wt. % and with 20 wt. % vermiculite. At least the bending strength for this body was too close to the required standards for wall and floor tiles applications, 16 MPa and 22 MPa, respectively according to ISO 10545-4 (Matteucci *et al.*, 2002).



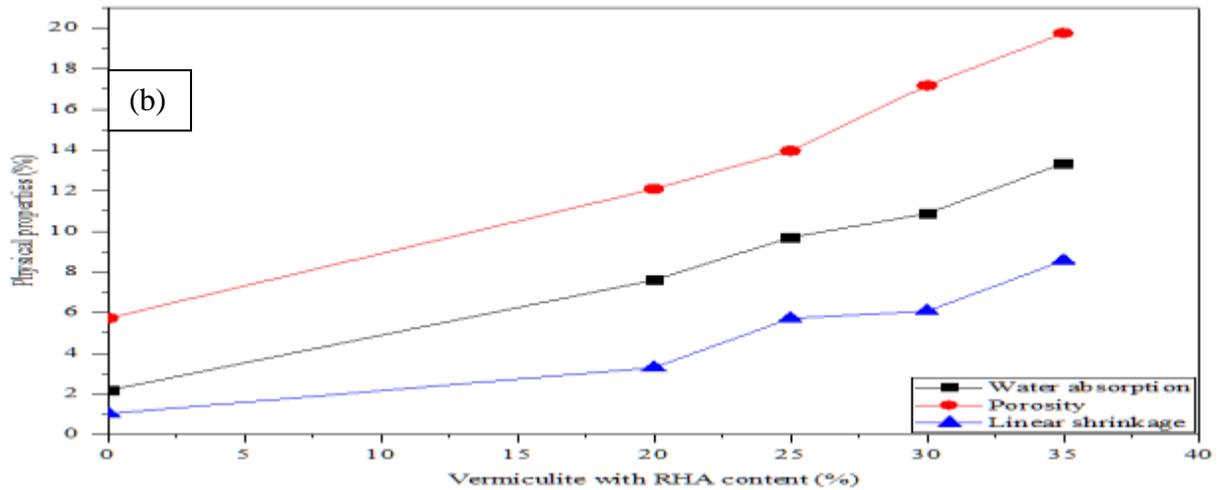


Figure 9: Influence of vermiculite and RHA content to the physical and mechanical properties of the fired ceramic bodies (a) bulk density and flexural strength (b) water absorption, porosity and linear shrinkage

It is clearly observed that tile samples with both vermiculite and RHA have high bending strength compared to the tile bodies without RHA because more glass was formed due to the introduction of RHA which filled the pores, in this way the porosity and water absorption decreased, hence bulk density and finally the strength of the tile bodies' increased. Partial replacement of quartz by rice husk ash (RHA) was expected to reduce the possibility of β to α phase inversion of quartz which occurs at 573°C during cooling which results into decrease of quartz particle volume and may lead to cracks in the ceramic body (Martín-Márquez *et al.*, 2008). According to literature information the progressive substitution of quartz by RHA in a conventional ceramic body composition resulted in an early vitrification of the mixes, while complete replacement of quartz by RHA drastically reduced both the maturing temperature and the percentage of thermal expansion, and increased the strength marginally (Prasad *et al.*, 2001). Figure 9 b shows the relationship between percentage of porosity, water absorption and linear shrinkage with addition of both vermiculite and RHA. With addition of both vermiculite and RHA percentage of porosity, water absorption capacity and linear shrinkage increased from 5.72 to 19.74 %, 2.17 to 13.34 % and 1.02 to 8.55 % respectively. But these values are low compared to that of tile bodies without RHA. The linear shrinkage of tile bodies with both vermiculite and RHA is low due to the addition of RHA. When RHA was added, the quartz starts to dissolve rapidly and produces more silica content to assist feldspar in dissolving the particles in the tile bodies (Prasad *et al.*, 2003), this reduced the pores and hence lowering of the shrinkage. Generally it can be revealed from the graphs that; with

addition of vermiculite and RHA the porosity, water absorption and linear shrinkage were increased this caused a decrease in bending strength and bulk density. This is because vermiculite and RHA are porous materials with low density compared to clay and kaolin. Also vermiculite is a light material which expands at higher temperature and this result into the development of cracks due to formation of vacancy defects usually created when the body cools after firing. However, ceramic bodies with both vermiculite and RHA had better physical and mechanical properties compared to those with vermiculite only. Theoretically, when the porosity increases, flexural strength and bulk density was expected to decrease and that was exactly what happened (Fig. 10).

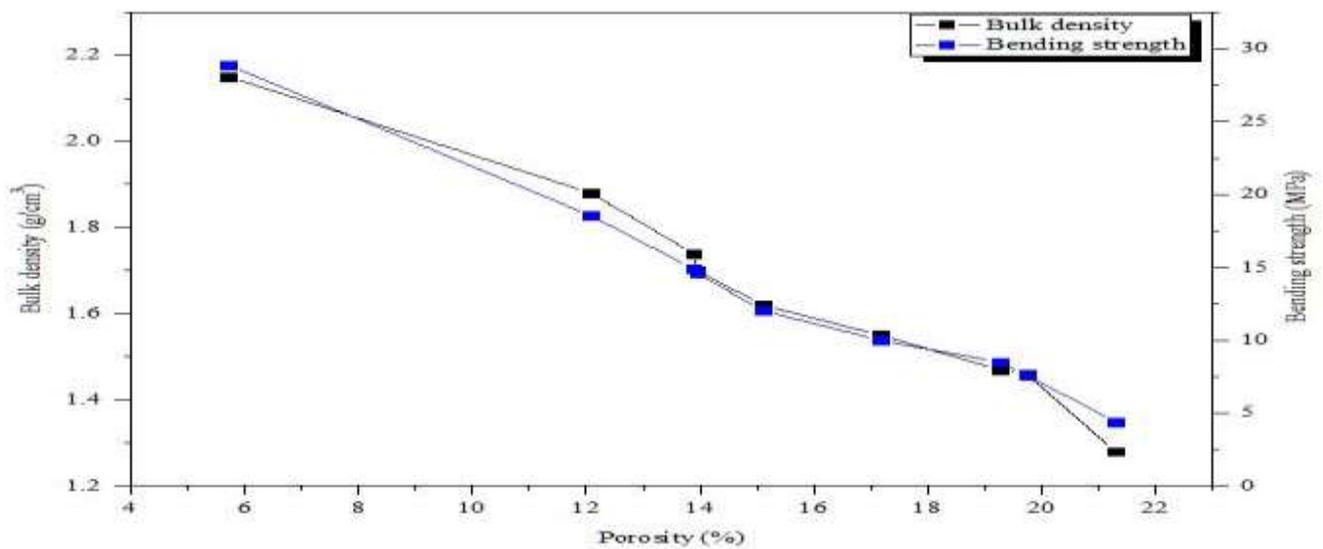


Figure 10: The relationship between porosity, bulk density and flexural strength of the tile samples

4.3 Influence of vermiculite and RHA compositions on thermal properties of tile samples

Table 5 shows the summary of thermal properties of test samples T₂-T₅ obtained when vermiculite was added and T₆-T₉ samples with both vermiculite and RHA while T₁ was the reference sample which had no any of the additives. The aim was to evaluate the influence of the two additives on thermal conductivity and effusivity. These values were calculated using Equation 7 and 8 while bulk density and porosity of ceramic bodies were obtained by using Equation 1 and 2 based on Archimedes principle.

Table 5: Physical and thermal properties of tile samples

Tile code	Porosity (%)	Bulk density (g/cm ³)	Thermal conductivity (W/mK)	Thermal effusivity (W ^{1/2} /m ² K ⁴)
T ₁	5.72	2.15	0.93	1462.69
T ₂	13.88	1.74	0.79	1212.77
T ₃	15.10	1.55	0.77	1130.06
T ₄	19.26	1.47	0.69	1041.78
T ₅	21.29	1.28	0.65	943.53
T ₆	12.08	1.88	0.82	1284.33
T ₇	13.95	1.70	0.79	1198.75
T ₈	17.16	1.62	0.73	1124.89
T ₉	19.74	1.46	0.68	1030.68

The contact temperature can be correlated by a property named “thermal effusivity” which defines the interface temperature when two solids are putted in contact. As lower is the thermal effusivity more comfortable is the ceramic floor tile (i.e. the lower the flooring thermal effusivity and the closer is the contact temperature to the human body, the temperature resulting provides the best comfort). The thermal effusivity is correlated directly with thermal conductivity and material density (Effting *et al.*, 2007). Materials with low conductivities and densities can be obtained by porous inclusion. Generally, thermal conductivity of porous materials decreases as porosity increases (Rhee, 1975).

Figure 11 shows the dependency of thermal conductivity and effusivity of tile samples on vermiculite content. It is clearly seen that, both thermal conductivity and effusivity were increasing with addition of vermiculite. The minimum thermal conductivity and effusivity were 0.65 W/mK and 943.53 W^{1/2}/m²K⁴ respectively with 35 wt. % vermiculite addition. With 20 wt. % vermiculite addition the maximum thermal conductivity and effusivity were 0.79 W/mK and 1212.77 W^{1/2}/m²K⁴ respectively.

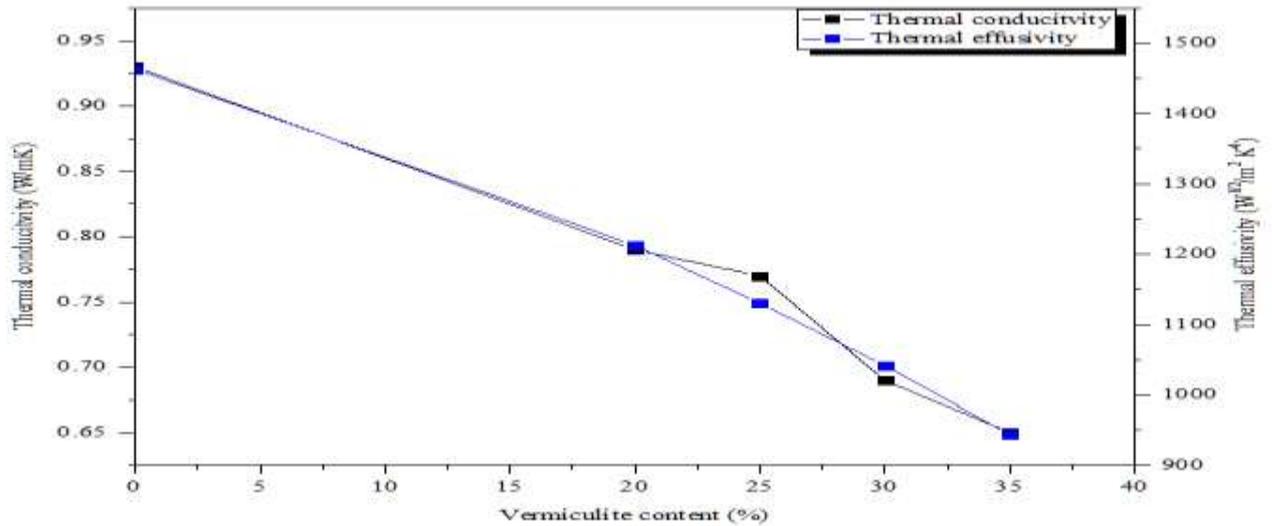


Figure 11: The influence of vermiculite content on thermal conductivity and effusivity of tile samples

Figure 12 shows the influence of RHA content on thermal conductivity and effusivity of tile samples. Both thermal conductivity and effusivity were increasing with addition of RHA. The minimum thermal conductivity and effusivity were 0.68 W/mK and 1030.68 W^{1/2}/m²K⁴ respectively with 20 wt. % RHA addition. With 5 wt. % RHA addition the maximum thermal conductivity and effusivity were 0.82 W/mK and 1284.33 W^{1/2}/m²K⁴ respectively. In both cases of the two additives the calculated thermal conductivities of the tile samples were close to the required standard of 1.05 W/mK but most of the test samples had no sufficient strength to make them useful for wall or floor applications. At least tile codes T₂, T₆, and T₇ with thermal conductivity 0.79, 0.82 and 0.79 W/m.K had bending strengths 14.90, 18.54, and 14.59 MPa, respectively which are too close to the required standard for both wall and floor applications.

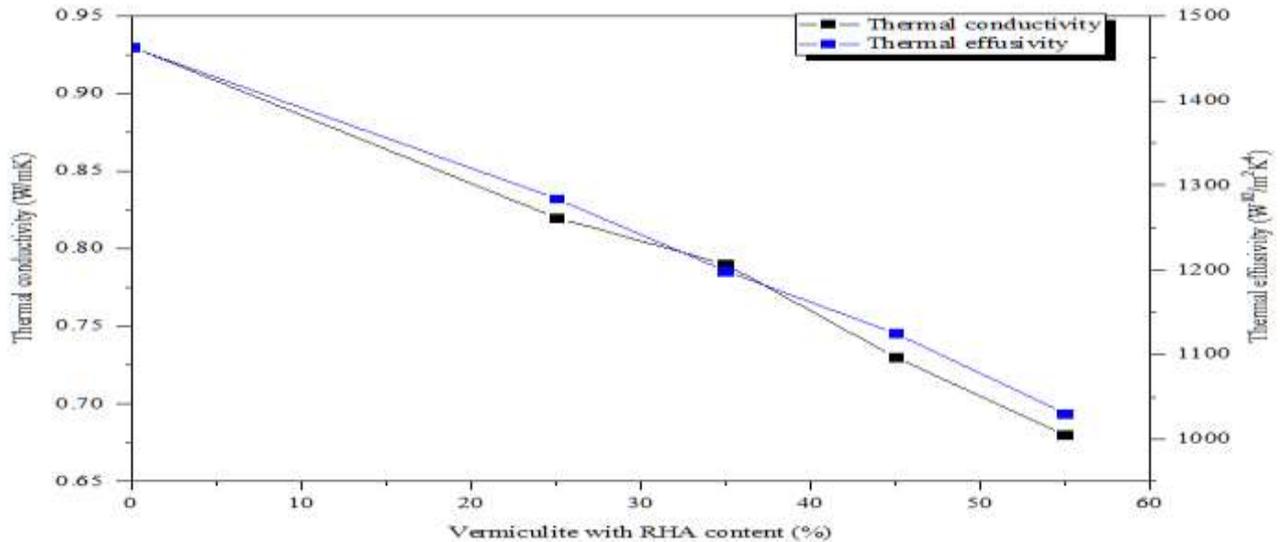


Figure 12: The influence of RHA content on thermal conductivity and effusivity of tile samples

Figure 13 shows the relationship between thermal conductivity and effusivity with porosity. It is observed that the calculated values of thermal conductivity of the samples are in a satisfactory agreement with the relation of open porosity, i.e. the thermal conductivity and effusivity decreases as the open porosity increases in the samples. For instance the sample with 35 wt. % vermiculite showed a thermal conductivity of 0.65 W/mK which is lowest value as compared to the remaining samples; this is due to high porosity of 21.29%. This value for thermal conductivity is in agreement with 0.65 W/mK obtained by Effting (2007) with addition of 20 wt. % industrial residue although the porosity was 35 %. The values for thermal conductivity obtained in this study are in agreement with the literature information and the standard reference values. The standard thermal conductivity for porcelain tile is 1.05 W/m K (Stulz and Mukerji, 1988). The same observation can be seen in terms of thermal effusivity which was decreasing with addition of vermiculite and RHA. Samples with vermiculite only had low thermal effusivity between 943.53 and 1212.77 Ws^{1/2}/m²K⁴ compared to samples with both vermiculite and RHA which their thermal effusivity was between 1030.68 and 1284.33 Ws^{1/2}/m²K⁴ (Fig. 13). It has been pointed that vermiculite is a porous material. Literature information show that, with addition of 75 wt. % of residue from porcelainized stoneware tile polishing; thermal conductivity and effusivity were 0.21 W/m K and 387.5 mK⁻¹W^{1/2} respectively according to Effting (2007), and these values were obtained when the fractional porosity was 0.65.

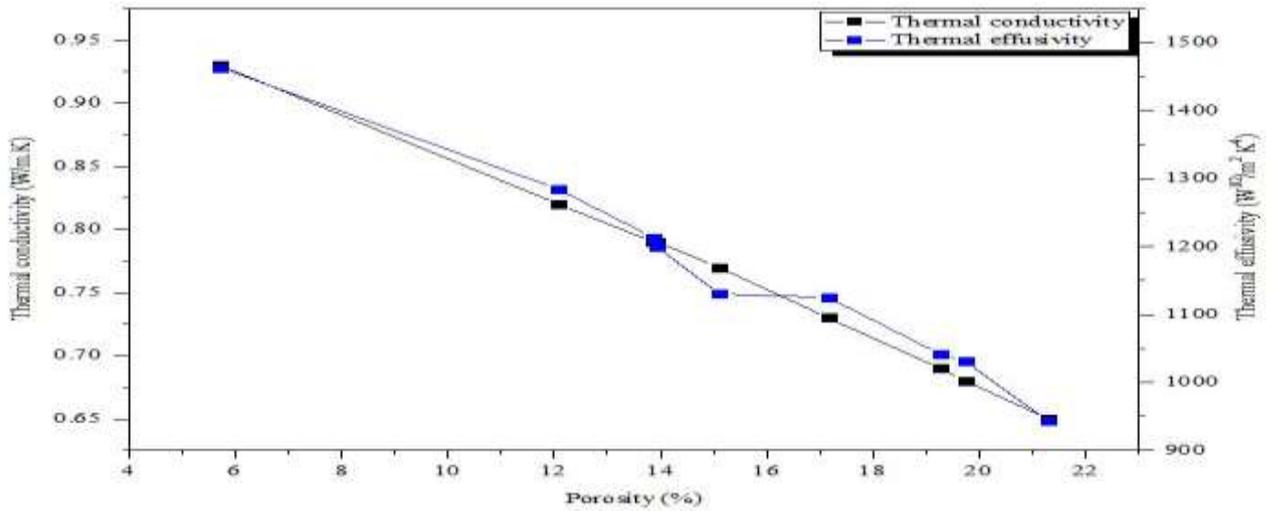


Figure 13: Relationship of thermal conductivity and effusivity with percentage of porosity

The human foot can be considered as having a thermal conductivity of 0.37 W/mK, density of 1000 kgm⁻³ and specific heat of 1000 J/kgK and hence the resulting thermal effusivity is 600 Ws^{1/2}/m²K⁴ while ceramic materials (porous or non-porous one) show specific heat values very close to 1kJ/kgK (Effting *et al.*, 2007). In order to produce tiles with improved thermal comfort; considerations must be made on how to minimize thermal effusivity as it depends on the thermal conductivity and material's density. Materials with low thermal conductivities are likely to have low density and thermal effusivity while their porosity is high although high porosity generally implies low mechanical strength. Depending on materials selection, batch formulations and forming processes (processing techniques) it is possible to obtain porous ceramics with high mechanical strength.

CHAPTER FIVE

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

In this study, the potential of vermiculite and rice husk ash (RHA) in the production of ceramic tiles of good mechanical strength and thermal comfort was investigated. Porous ceramic tiles were obtained by incorporating ceramic powder and two natural additives which are vermiculite and RHA with various weight fractions. Finally physical, mechanical and thermal properties (effusivity and conductivity) of tile samples fired at 1180°C were tested and evaluated. The major chemical compounds in vermiculite raw sample were SiO₂, Al₂O₃ and Fe₂O₃ while RHA sample was found to contain mainly SiO₂. From the XRD analysis, vermiculite sample had crystalline vermiculite while RHA sample had amorphous silica at low temperature below 900°C and crystallized (tridymite) above 900°C. The results from physical, mechanical and thermal properties tests show that; with addition of vermiculite and RHA, the percentage of porosity, water absorption, linear shrinkage and thermal insulation were increasing while the bulk density, bending strength, thermal conductivity and effusivity of test samples decreased. Among the studied compositions apart from the reference sample, tile bodies made from blend containing 20 wt. % vermiculite, 5 wt. % RHA were found to have the best properties for the production of ceramic tiles. For this combination, the bending strength and thermal conductivity were 18.84 MPa and 0.82 W/mK respectively. These values were close to the required standards for wall and floor tiles applications. This is an indication that vermiculite and RHA are suitable raw materials for the production of ceramic tiles with a required strength and thermal comfort.

5.2 Recommendations

Further studies on the strength and insulating properties of ceramic tiles should be done by analyzing the effects of addition of other agricultural wastes on ceramic tiles as well as reducing the iron content in vermiculite samples. Vermiculite and RHA are porous materials hence when added in excess to ceramic tile batches would reduce the strength of the final body. While the presence of high amount of Iron in vermiculite increases thermal conductivity of the final fired body which also affects thermal comfort of the tiles. Depending on materials selection, batch formulations and forming processes (processing techniques); it is possible to obtain porous ceramics with high mechanical strength.

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APPENDICES

Appendix 1: Batch formulations

Identity of raw materials involved

- i) Pugu Kaolin: $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$.
- ii) Vermiculite : $22\text{MgO} \cdot 5\text{Al}_2\text{O}_3 \cdot \text{Fe}_2\text{O}_3 \cdot 22\text{SiO}_2 \cdot 4\text{H}_2\text{O}$
- iii) Quartz : SiO_2
- iv) RHA : SiO_2
- v) Feldspar : KAlSi_3O_8
- vi) Same clay: $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$.

Appendix 1 a: Ratios of the original samples before formulations

Raw materials	Raw materials' components of interest				Al: Si
	Al_2O_3		SiO_2		
	%	mass(g)	%	mass(g)	
Pugu Kaolin	30.30	7.58	60.00	15.00	1: 2
Same Clay	23.50	5.875	62.10	15.525	1: 2.6
Vermiculite	13.00	3.25	35.20	8.80	1: 2
RHA	—	—	92.10	23.02	—
Quartz	—	—	97.70	24.40	—
Feldspar	14.00	3.50	57.10	14.28	1: 4

XRF Analysis: 25g of each sample was used ie. $W_s = 25\text{g}$

Appendix 1 b: Tile formulation by Wt. %

Tile code	Vermiculite	Pugu Kaolin	Same Clay	RHA	Quartz	Feldspar
T ₁	—	10	60	—	10	20
T ₂	20	10	40	—	10	20
T ₃	25	10	35	—	10	20
T ₄	30	10	30	—	10	20
T ₅	35	10	25	—	10	20
T ₆	20	10	35	5	10	20
T ₇	25	10	30	10	5	20
T ₈	30	10	22.5	15	2.5	20
T ₉	35	10	15	20	—	20

Appendix 1 c: Tile formulation by mass (g)

	Vermiculite	Pugu Kaolin	Same Clay	RHA	Quartz	Feldspar
T ₁	—	40	200	—	40	120
T ₂	80	40	160	—	40	80
T ₃	100	40	140	—	40	80
T ₄	120	40	240	—	40	80
T ₅	140	—	140	—	40	80
T ₆	80	20	180	20	20	80
T ₇	100	20	140	40	20	80
T ₈	120	20	100	60	20	80
T ₉	140	20	60	80	20	80

Appendix 1 d: Alumina and Silica ratios for the final formulations

Tile code	Al ₂ O ₃ (g)	%	SiO ₂ (g)	%	Total(g)	%	Other(g)	%	Al: Si
T ₁	75.93	18.98	230.74	57.69	306.67	76.67	93.33	23.33	1: 3.00
T ₂	71.33	17.83	236.26	59.07	307.59	76.90	92.41	23.100	1: 3.10
T ₃	69.23	17.31	230.78	57.70	300.01	75.00	99.99	25.00	1: 3.33
T ₄	54.81	13.70	225.50	56.38	280.31	70.08	119.69	29.92	1: 4.00
T ₅	62.30	15.58	220.96	55.24	283.26	70.82	116.74	19.19	1: 3.55
T ₆	69.96	17.49	235.57	58.89	305.53	76.38	94.47	23.62	1: 3.37
T ₇	63.16	15.79	236.19	59.05	299.35	74.84	100.65	25.16	1: 3.74
T ₈	56.36	14.09	236.80	59.20	293.16	73.29	106.84	26.71	1: 4.20
T ₉	49.56	12.39	191.38	47.85	240.94	60.24	159.06	39.77	1: 3.86

Appendix 2: Physical and mechanical properties calculations

Bulk density

$$\text{Bulk density} = \left[\frac{W_d}{W - S} \right] \cdot D$$

Where W_d is dry weight, W is soaked weight, S is suspended weight, and D is density of water. Experiments will be carried out at a room temperature of 22 °C, in which the density of water is 1.00g/cm³

Appendix 2 a: Evaluation of Archimedes principle

Tile code	W_d (g)	S (g)	W (g)	$W - S$ (g)	$W - W_d$ (g)	L_g (cm)	L_f (cm)	$L_g - L_f$ (cm)
T ₁	395.50	222.07	406.02	183.95	10.52	14.00	13.86	0.14
T ₂	377.25	190.53	407.34	216.81	30.09	14.00	13.46	0.54
T ₃	352.19	159.28	386.50	227.22	34.31	14.00	13.18	0.82
T ₄	338.76	152.69	383.14	230.45	44.38	14.00	12.62	1.11
T ₅	316.53	121.89	369.18	247.29	52.65	14.00	12.32	1.68
T ₆	382.11	203.41	406.66	203.25	24.55	14.00	13.54	0.46
T ₇	369.22	181.69	399.52	217.19	30.30	14.00	13.20	0.80
T ₈	344.79	168.48	381.31	212.83	36.52	14.00	13.15	0.85
T ₉	325.37	146.50	369.36	222.86	43.99	14.00	12.80	1.20

Appendix 2 b: Physical-mechanical properties

Tile code	%	%	g/cm ³	%	σ_{fs}
	W. A	P	B. D	L. S	(MPa)
T ₁	2.17	5.72	2.15	1.02	28.86
T ₂	8.21	13.88	1.74	3.85	14.90
T ₃	10.42	15.10	1.55	5.88	9.99
T ₄	13.56	19.26	1.47	9.88	7.61
T ₅	16.81	21.29	1.28	12.00	4.40
T ₆	7.60	12.08	1.88	3.29	18.54
T ₇	9.69	13.95	1.70	5.71	14.59
T ₈	10.87	17.16	1.62	6.07	12.03
T ₉	13.34	19.74	1.46	8.55	8.50

Appendix 3: Thermal properties calculations

Appendix 3 a: Thermal conductivity theory

Thermal conductivity measurement was conducted according to ISO 8301 (Standard, 1991).

$$\frac{K}{K_0} = \frac{1-P}{1+nP^2} \dots\dots\dots (i)$$

Where K is the thermal conductivity of a porous ceramic body, K₀ is the thermal conductivity of a porous free ceramic body, P is the volume fraction of porous, and n is a constant. According to (Sugawara & Yoshizawa, 1962) measurements on thermal conductivity of a ceramic tile at 70⁰C, n corresponds to 3 and K₀ = 1.65 W/m.K. The equation becomes:

$$K = 1.65 \left(\frac{1-P}{1-3P^2} \right) \dots\dots\dots (ii)$$

Good correlation occurs in the porosity interval from 0 to 50% for n = 4.5 and K₀= 1.6 W/m.K and the equation becomes:

$$K = 1.60 \left(\frac{1-P}{1-4.5P^2} \right) \dots\dots\dots (iii)$$

$$K = 1.60 \left(\frac{1 - P}{1 + 4.5P^2} \right)$$

Appendix 3 b: Thermal effusivity theory

According to Carmeane Effting (2007) thermal effusivity is given by the expression $\epsilon = \sqrt{K\rho c}$ where ϵ is thermal effusivity, K is coefficient of thermal conductivity (W/mK), ρ is density (Kg/m³) and C is specific heat capacity (J/KgK).

$$\epsilon = \sqrt{K\rho c}, c=1070J/Kg.K$$

Appendix 3 c: Thermal properties of test samples fired at 1180⁰C

Sample code	Porosity (%)	Bulk density g/cm ³	Thermal conductivity W/mK	Thermal effusivity W ^{1/2} /m ² K ⁴
T ₁	5.72	2.15	0.93	1462.69
T ₂	13.88	1.74	0.79	1212.77
T ₃	15.10	1.55	0.77	1130.06
T ₄	19.26	1.47	0.69	1041.78
T ₅	21.29	1.28	0.65	943.53
T ₆	12.08	1.88	0.82	1284.33
T ₇	13.95	1.70	0.79	1198.75
T ₈	17.16	1.62	0.73	1124.89
T ₉	19.74	1.46	0.68	1030.68

RESEARCH OUTPUTS

Journal paper

Abeid, S. and Park, S. E. (2018). Suitability of Vermiculite and Rice Husk Ash as Raw Materials for Production of Ceramic Tiles. *International Journal of Materials Science and Applications*, 7(2): 39.

Suitability of Vermiculite and Rice Husk Ash as Raw Materials for Production of Ceramic Tiles

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Abstract: The challenging issues in ceramic tiles are low mechanical strength, thermal discomfort and high production costs. And in most efforts to improve strength, emphasis has been placed on minimization of quartz content in the ceramic tiles formula. This is due to β - α phase inversion of quartz which occurs at 573°C during cooling resulting to the development of stresses which initiate fracture and affects the strength of the final body. The objective of this work was to evaluate the possibility of using vermiculite and rice husk ash (RHA) in the composition of ceramic tile body. Initially, a typical ceramic body composed of the mixture of vermiculite and RHA batched with clay, feldspar, quartz and kaolin was prepared. Ceramic bodies were then obtained from this ceramic mixture by pressing samples at a forming pressure of 35MPa. These bodies were then fired at 1180°C in a laboratory furnace and finally the changes in the physical and mechanical properties caused by the introduction of vermiculite and RHA were tested and evaluated. The chemical composition of the raw samples was analyzed by X-ray fluorescence (XRF) while the phase composition was investigated using X-ray diffraction (XRD). The morphology of the powdered samples was studied by using Scanning electron microscopy (SEM). The bulk density and open porosity of the sintered ceramic bodies were evaluated using Archimedes' principle while the flexural rupture strength was determined by the three point bending test method. The major chemical compounds in vermiculite raw sample were SiO₂, Al₂O₃ and Fe₂O₃, while RHA sample was found to contain mainly SiO₂. From the XRD analysis, vermiculite sample had crystalline vermiculite while RHA sample had amorphous silica at low temperature below 900°C and crystallized (tridymite) above 900°C. The results from physical and mechanical properties tests show that with addition of vermiculite and RHA, the percentage of porosity, water absorption and linear shrinkage were increasing while the bulk density and bending strength of the fired ceramic bodies decreased. Among the studied compositions tile bodies made from a blend containing 20% wt. vermiculite and 5% wt. RHA were found to have the best properties for ceramic tiles applications. For this combination the percentage of porosity, water absorption and linear shrinkage were 12.08%, 7.60% and 3.29% while the bulk density and bending strength were 1.88 g/cm³ and 18.84 MPa respectively. These values were close to the required standards of wall and floor tiles.

Keywords: Vermiculite, Tridymite, Amorphous, Crystalline, Flexural Strength

1. Introduction

A typical ceramic tile composition will consist of 50% clay which imparts rigidity to the ceramic body, 25% quartz which lowers both drying and firing shrinkage and 25% feldspar which serves as a flux and also provides a glassy phase in the microstructure. Mullite and glass constitute the major phases of final ceramic product. Other constituents of the fired ceramic body in minor levels are quartz, cristobalite, tridymite and corundum [1]. Quartz and kaolin are mostly

preferred as a source of silica and alumina respectively and they have a great influence on the mechanical strength of ceramic tiles because when they are mixed and fired at very high temperature close to their melting points they form a very strong alumina-silica phase called mullite which controls the strength of the ceramic body. But quartz grains embedded in the glassy matrix have a deleterious effect on the mechanical strength mainly because of its α - β phase transformation during cooling resulting to the development of stress which initiate fracture [2, 3]. Also, most of ceramic

products especially tiles are produced at a temperature above 1000°C which is associated with high energy consumptions and raises the production costs. For these reasons this study was based on testing and evaluating the suitability of vermiculite and RHA as additives in the production of ceramic tiles by considering strength as the central objective. Vermiculite is a clay minerals produced by the decompositions of mica and occurs as large crystals of mica-like lamellar structure that quickly expands on heating to produce a lightweight material [4]. In Tanzania vermiculite can be found in places like Kalalani Tanga and Mikese Morogoro. In its expanded form, vermiculite has a very low density and thermal conductivity, which makes it attractive for use as a lightweight construction aggregate, and thermal insulation filler [5]. Partial replacement of quartz by RHA is expected to reduce the possibility of β to α phase inversion of quartz which occurs at 573°C during cooling which leads to decrease in quartz particles volume and thus cracks in the ceramic body. The use of RHA in ceramic body have been proven to improve the strength compared to the body without RHA [3]. The reduction in the vitrification temperature of the mixes with addition of both vermiculite and RHA during

firing would also contribute significantly to the economical production of ceramic tiles due to low energy consumptions although the temperature has to be high enough in order to achieve an acceptable strength. Currently no research has been done to test the potential of the combination of both vermiculite and RHA in ceramic tiles applications. In this study the suitability of vermiculite and rice husk ash as raw materials for production of ceramic tiles was tested and evaluated.

2. Materials and Methods

2.1. Raw Materials Collection

Materials that were involved in this study include vermiculite samples V_{K1} , V_{K2} and V_{K3} (Figure 1) taken from three mining sites; Red Garnet, Red Safaya, and Pink Safaya respectively all found at Kalalani village in Tanga region eastern part of Tanzania. Rice husk samples were taken from Bahi district in Dodoma region central part of Tanzania. Pugu kaolin, Same clay, quartz and feldspar were obtained from Geological Survey of Tanzania Laboratory in Dodoma.



Figure 1. Photomicrographs of vermiculite samples.

2.2. Sample Preparations and Pretreatment of Ceramic Constituents

The procedure for preparing rice husk powder consist of washing RH in distilled water in order to remove clay and rock impurities and subsequently drying in an oven at 120°C for 12 h. Then 30 g of dry husk was reacted with 2 M HCl at 25°C in a 500 mL under constant agitation for 2h. This was done to remove metallic impurities and organics contained in rice husk before calcination. The husk was then washed with distilled water till neutral pH was obtained and then dried. The dried RH was later heated at 700°C for 2 h to at a heating rate of 10°C/min under air atmosphere to obtain carbon free white ash. At 700°C and below, ash rich in amorphous silica is formed which is highly reactive. Above 700°C, crystalline silica which is far less reactive is obtained. On the other hand, Pugu kaolin, vermiculite, quartz, and feldspar samples were separately dried at 130°C in the oven for 24 h and then ground by using ball mill for 6 h. The powdered samples were sifted through a sieve of 150 μ m pore size. Grinding and sieving were repeated until almost all the materials passed through the sieve.

2.3. Chemical, Mineralogical and Microstructural Analyses of Raw Materials

The chemical compositions of raw materials were studied at Geological Survey of Tanzania (GST) Laboratory using X-Ray Fluorescence (XRF) PANalytical, Minipal4 (PW4030)-Rh X-Ray Tube, which was operating at 30 kV, 0.002 mA. The amorphous and crystalline structure of vermiculite and RHA samples were identified by using Ultima IV Rigaku diffractometer operating at tube voltage and current at 40 kV and 44 mA, respectively using monochromatic Cu-K α radiation. Diffraction patterns were recorded by scanning from 5° to 75° (2 θ) in steps of 0.02° (2 θ) at a rate of 2sec/step. The morphological features were studied by using Scanning electron microscopy (FEI Nova NanoSEM 450, 2kV). The SEM and XRD analyses were all carried out at the University of Connecticut, USA.

2.4. Batch Composition

Vermiculite was gradually incorporated into the ceramic samples with a composition range of 20 to 35% wt. to make

the first batch of tile samples with vermiculite, T₂ to T₅. Then 5 to 20% wt. of RHA was added together with 20 to 35% wt. of vermiculite to complete the second batch of tile samples, T₆ to T₈. The first test sample T₁ used as a control sample contained neither vermiculite nor RHA (Table 1). The compositions were mixed with 5 to 6% wt. water using ball mill for 90 min.

Table 1. Mix proportion of raw materials (% wt.).

Tile code	Vermiculite	Pugu Ka	Same Cl	RHA	Quartz	Feldsp
T ₁	—	10	60	—	10	20
T ₂	20	10	40	—	10	20
T ₃	25	10	35	—	10	20
T ₄	30	10	30	—	10	20
T ₅	35	10	25	—	10	20
T ₆	20	10	35	5	10	20
T ₇	25	10	30	10	5	20
T ₈	30	10	22.5	15	2.5	20
T ₉	35	10	15	20	—	20

2.5. Preparation of Ceramic Green Bodies

Discs-shape ceramic bodies of 3 cm diameter and 8 mm in thickness were prepared by pressing samples at a forming pressure of 35MPa in order to measure the physical properties while square tiles of 140 × 64 × 14 mm³ were prepared for bending strength measurements (Figure 2a). The tile samples were subsequently oven-dried at 110°C for 24 h followed by cooling at room temperature for 1 h. The dimension and weight of each green (dried) body was recorded before sintering.



Figure 2. Disc- and tile-shaped bodies fabricated using vermiculite and RHA for physical and mechanical properties tests (a) green bodies (b) fired bodies (c) fired and labelled bodies (d) fired bodies with visual defects such as cracks, lamination and surface deformation.

2.6. Firing of Ceramic Green Bodies

The ceramic bodies were then fired at 1180°C for 2 h

soaking time, at a heating rate of 50°C per minute in a Carbolite box furnace (RHF 14/8 Model) manufactured by Keison products Inc, UK. The firing parameters tests were all performed at Tanzania Portland Cement Company Limited (TPCC). After firing, the samples were permitted to cool down to room temperature inside the kiln for 24 h and then the dimensions and weights of the fired tile bodies were recorded. Finally the fired bodies (Figure 2b) were labeled (Figure 2c) and then stored in airtight containers ready for mechanical tests and further characterizations.

2.7. Determination of Physical and Mechanical Properties of Tile Bodies

Fired ceramic bodies were inspected for any visual defects such as cracks, lamination and surface deformation. Thus, good quality ceramic bodies were selected and used for determination of the physical and mechanical properties. The physical and mechanical properties tests of the fired tile bodies were all carried out at Tanzania Bureau of Standards (TBS) - Materials testing laboratory.

The bulk density and open porosity of the sintered ceramic bodies were evaluated using Archimedes' principle which involved drying tiles in an oven at 110°C for 24 h and cooling in desiccators. The dry mass of the tiles (W_d) was then measured, followed by water impregnation, which involved boiling tiles in distilled water for 5 h and then left soaked in water for an additional 24 h at room temperature. After impregnation, the suspended mass (S) of each tile body was recorded. The saturated mass (W) was measured after removing all excess water from the surface by using moistened cotton cloth. The bulk density was calculated using the formula;

$$B. D = \left[\frac{W_d}{W - S} \right] \cdot D \quad [6] \quad (1)$$

While porosity was given by the following expression;

$$P = \left[\frac{W - W_d}{W - S} \right] \cdot 100\% \quad [6] \quad (2)$$

Where W_d was dry weight, W was soaked weight, S was suspended weight, and D was density of water. Experiments were carried out at a room temperature of 18 °C, in which the density of water was 1.00 g/cm³. The water absorption A was calculated as the ratio of the mass of water absorbed to the mass of the dry specimen and given by the expression

$$A = \left[\frac{W - W_d}{W_d} \right] \cdot 100\% \quad [7] \quad (3)$$

While linear shrinkage (L. S) was calculated by measuring dimensions of the prepared specimen before and after firing hence;

$$L. S = \left[\frac{l_g - l_f}{l_g} \right] \cdot 100\% \quad [8] \quad (4)$$

Where L_g and L_f were the lengths (mm) of green and fired tile bodies, respectively. The flexural rupture strength was

determined by the three point bending test in a bending strength tester (MEGA 10-200-10 DS) manufactured by Prufsysteme Inc. in Germany, 2001.

3. Results and Discussion

3.1. Chemical Composition and Phase Analysis of the Raw Materials

The presence of various compounds within the raw materials can be seen in Table 2. It is evident that SiO₂ was the major composition in the raw materials with 92.1 wt.% in RHA, 97.7 wt.% in quartz, and 62.10 wt.% in vermiculite. Alumina was high in Pugu kaolin with 30.30 wt.% and lowest in vermiculite with 9.20 wt.% while RHA and quartz showed no any indication of alumina. Silica and alumina in the raw materials are to be taken into account as they play a very important role during sintering. Silica is a glass former while alumina is a refractory which gives strength and durability to a ceramic body. Vermiculite samples showed high percentage of Fe₂O₃ at 18.70 wt.%. High content of Iron affects thermal comfort of the final ceramic body due to increase of thermal conductivity and effusivity. The flux oxide, K₂O which is essential for liquid phase formation and lowering the melting points of silica in the ceramic body, was high in feldspar than in any other raw materials with 23.5 wt.% although Na₂O was

only present in Pugu kaolin and vermiculite in a small amount less than 1 wt.%. The opacifier TiO₂ which makes the ceramic body opaque was high in vermiculite and Pugu kaolin with 1.83 wt.% and 1.22 wt.% respectively, but was too low in the remaining samples with less than 1 wt.%. The alkaline earth oxide, CaO was present in all raw samples except in feldspar and quartz; and the highest amount was obtained in RHA with 2.98 wt.% while MgO was present in vermiculite only with 6.10 wt.%. The low earth-alkaline oxides (CaO and MgO) content for Pugu kaolin and Same clay indicates that the studied clays are poor in carbonates [7]. The fluxing oxides and colorants must be maintained at low amount to avoid undesired color and the possibility of excessive fluxing in the ceramic product [8]. In this study RHA had a chemical composition of 92.10 wt.% silica which was too close to other authors' findings 93.70 wt.% [3], 93.67wt.% [9], and 94.95 wt.% [10]. Also the amount of silica in Kalalani vermiculite was close to the required standard for production of ceramic tiles with 47.3 to 79.3 wt.% according to [8]. Vermiculite raw sample shows a loss on ignition of about 5.76% which was the highest compared to that of the remaining samples. This value was related to the presence of organic residues where the decomposition of carbonates and sulphates produced a significant weight loss at a temperature above 1000°C (Table 2).

Table 2. Chemical composition of raw materials (wt.%).

	Vermiculite	Pugu kaolin	Same Clay	RHA	Feldspar	Quartz
SiO ₂	62.10	60.00	60.40	92.10	57.10	97.70
Al ₂ O ₃	9.20	30.30	13.90	–	14.00	–
Fe ₂ O ₃	18.70	3.95	1.40	2.26	1.05	–
TiO ₂	1.83	1.22	0.14	0.18	–	–
CaO	0.91	0.39	0.02	2.98	–	–
MgO	6.10	–	–	–	–	–
Na ₂ O	0.05	0.04	–	–	–	–
K ₂ O	0.11	2.14	22.60	0.96	23.50	–
LOI	5.76	1.96	1.56	1.52	4.35	2.30

Figure 3 shows the X-ray diffractograms of the vermiculite and RHA samples. The XRD patterns of raw vermiculite show the presence of vermiculite as the major phase which was identified by peaks around 5° and 31.5° 2θ (Figure 3a). These results are close to XRD results of Kalalani vermiculite according to work done by [11] which showed vermiculite as the only phases in the sample. In Figure 3b the evolution of the crystalline phase from the amorphous silica present in the RHA can be seen. The XRD pattern shows the presence of tridymite as the major phase which was identified by peaks around 20.76° and 39.49° 2θ. According to [9], silica of RHA is amorphous at low temperatures between 700 and 800°C. Firing RHA at 900°C and above produces crystal phases of two forms, cristobalite and tridymite which are less reactive. The transformation of amorphous silica to a cristobalite phase occurs at a temperature interval between 550 and 950°C [12].

The SEM micrographs of raw vermiculite and RHA samples

(Figure 4) show the morphology of the vermiculite and RHA particles. Some of the detected phases are identified based on local chemical analysis by EDS. Figure 4(a) shows the morphology of vermiculite sample containing flakes with individual layer crystals which were very closely spaced. The milled vermiculite flakes had a size range of smaller than 150 μm. The images show that vermiculite was more porous than porcelain and hence increasing in vermiculite content cause an increase in porosity size and amount. These results had a good agreement with bulk density and porosity results of the tile samples described in Part 3.2 of this article. Figure 4(b) shows the scanning electron microscopy of RHA powder which indicates that the ash was siliceous in nature with a porous structure and consists of quartz grains of various sizes and irregular shapes; these results agree with other authors as per [3], [13] and [14]. The porous nature of RHA and its honeycombed structure is responsible for its high specific surface [13] and this is a good indication for its suitability in ceramic tiles applications.

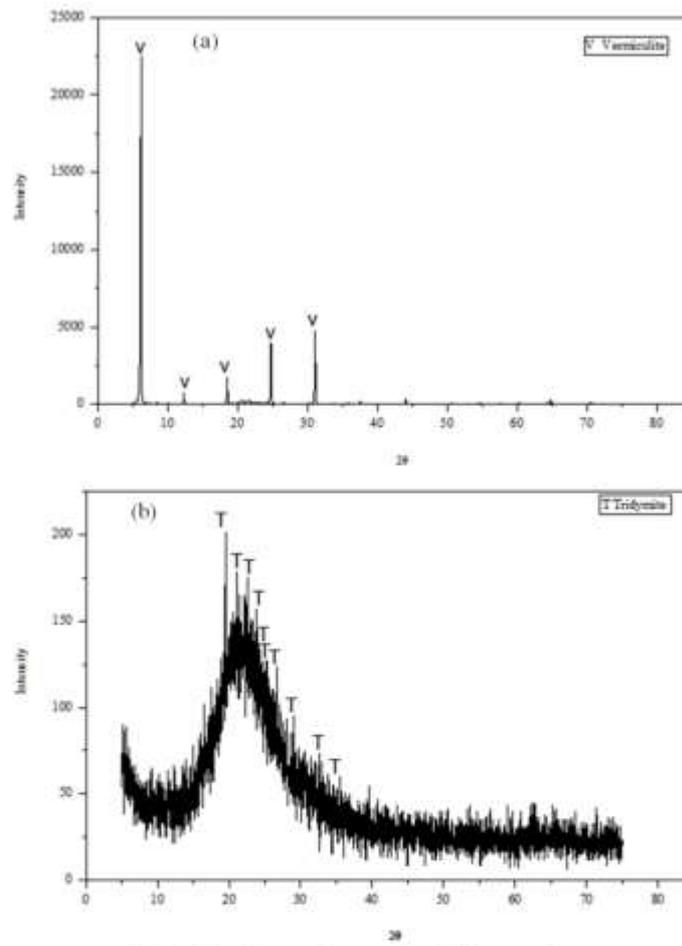
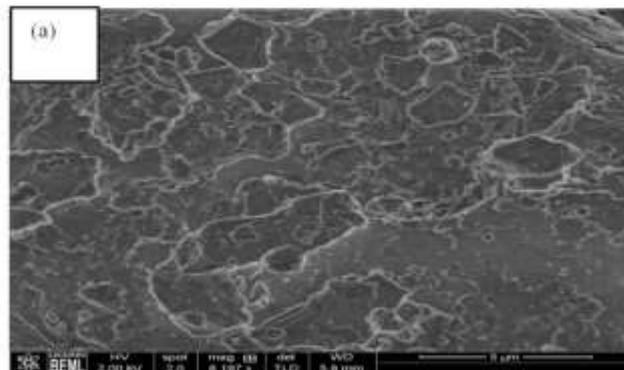


Figure 3. X-ray diffraction of (a) vermiculite (b) RHA raw samples.



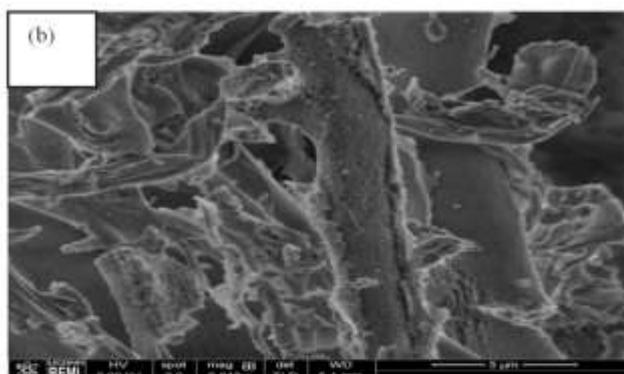


Figure 4. SEM micrographs of (a) vermiculite (b) RHA raw samples.

3.2. Physical and Mechanical Properties of the Fired Tile Bodies

Results for the physical and mechanical properties tests of the ceramic bodies were presented and summarised in Table 3. The symbols WA, P, BD, LS and σ_b are water absorption, porosity, bulk density, linear shrinkage and flexural strength respectively.

Table 3. Physical and mechanical properties of tile bodies sintered at 1180°C.

Tile code	WA (%)	P (%)	BD (g/cm ³)	LS (%)	σ_b (MPa)
T ₁	2.17	5.72	2.15	1.02	28.86
T ₂	9.21	13.88	1.74	3.05	14.90
T ₃	10.42	15.10	1.55	5.88	9.99
T ₄	13.56	19.26	1.47	9.88	7.61
T ₅	16.81	21.29	1.28	12.00	4.40
T ₆	7.60	12.08	1.88	3.29	18.54
T ₇	9.69	13.95	1.70	5.71	14.59
T ₈	10.87	17.16	1.62	6.071	12.03
T ₉	13.34	19.74	1.46	8.551	8.50

3.2.1. Influence of Vermiculite on the Physical and Mechanical Properties of Tile Samples

The bulk densities of fired tile bodies were decreasing with addition of vermiculite from 20 to 35% wt. (Figure 5a). The maximum bulk density was 1.74 g/cm³ obtained when vermiculite was 20% wt. while the minimum was 1.28 g/cm³ obtained when vermiculite was added up to 35% wt. The bulk density of the control sample which had 0% wt. vermiculite was 2.15 g/cm. Tile code T₂ had the highest bulk density due to low vermiculite content compared to T₃, T₄ and T₅ among the bodies with vermiculite as the only additive (Table 3). Decrease of bulk density of tile bodies with addition of vermiculite was mainly caused by low unit weight and density of raw vermiculite [15]. The expanded vermiculite has the density ranging between 0.2 to 0.3 g/cm³ [16]. These values are low compared to that of clay and kaolin which their bulk density is about 0.8g/cm³. Hence, increase of vermiculite content in the samples led to decrease in bulk densities. On the other hand results show that, bending strength of fired ceramic bodies decrease with addition of vermiculite content (Figure 5a). The reference sample had bending strength of 28.86MPa when the amount of vermiculite in the body was 0% wt. and this value

decreased up to 4.40 MPa when vermiculite content reached 35% wt. which was the maximum vermiculite content for this study. The maximum bending strength for tile bodies with vermiculite as the only additive material was 14.90 MPa obtained when the vermiculite composition was 20% wt. This tile body had the strength value close to the required standard for wall tiles while the remaining tile bodies had values ranging between 4.40 and 9.99 MPa which were too far from the required standards (Table 3).

According to commercial standards the minimum strengths for wall and floor tiles are 16 MPa and 22 MPa respectively ISO 10545 [17].

Figure 5b shows the influence of vermiculite content on water absorption, porosity and linear shrinkage of the fired tile samples. The percentage of porosity of the fired ceramic bodies increase with increase of the vermiculite content. The minimum value was 5.72% when vermiculite content was 0% wt. and the highest was 21.29% obtained when the vermiculite content was 35% wt. The tile body with 20% wt. vermiculite had porosity value close to the required international standard for wall and floor tiles applications which is 7 to 12% of total porosity [8]. Since water absorption is directly related to open porosity, its value also increases with the increase of vermiculite content too. With

addition of vermiculite the water absorption increased from 2.17% of the control tile sample up to 16.81% which was the highest and was obtained when vermiculite content reached 35% wt. The linear shrinkage of the tile samples increased with the increase of vermiculite content with values ranging

from 1.02% of the control sample (0% wt. vermiculite) to 12.00% when vermiculite was 35% wt. This is because during firing vermiculite lose water moisture and other volatile (carbon contents) materials, this leads to reduction in particles size and hence dimension.

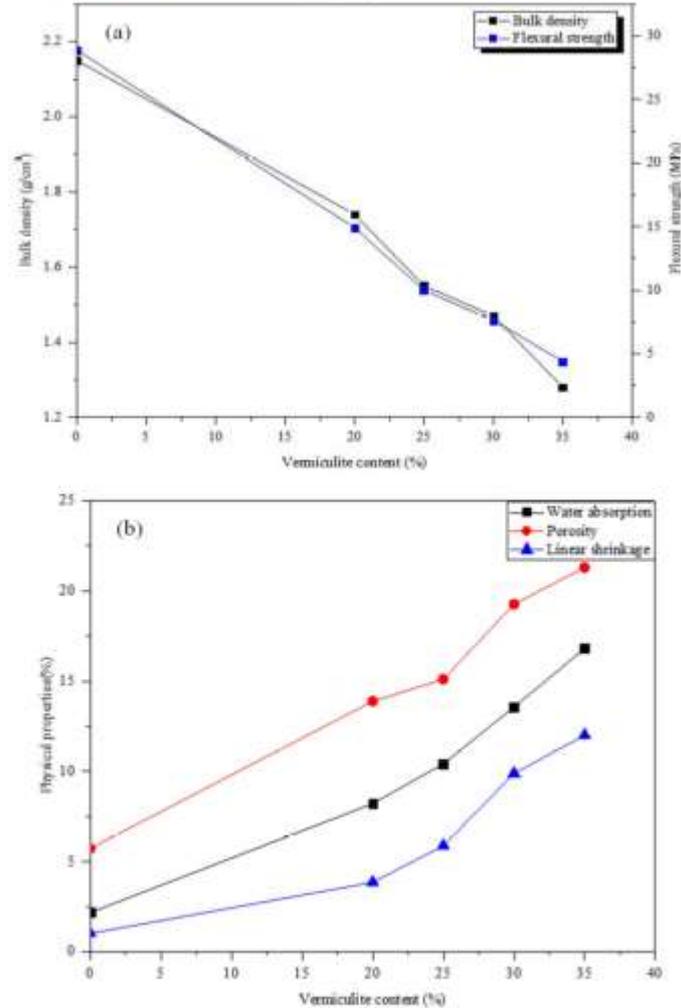


Figure 5. Influence of vermiculite content on the physical and mechanical properties of the fired ceramic bodies (a) bulk density and flexural strength (b) water absorption, porosity and linear shrinkage.

3.2.2. Influence of RHA on the Physical and Mechanical Properties of Tile Samples

The physical and mechanical properties of tile bodies (T_w , T_p , T_s and T_l) with both vermiculite and RHA are shown in Table 3 and Figure 6. Similar correlation was observed on the bulk density of the tile bodies which was decreasing with

increase of RHA content. The control tile sample had the bulk density of 2.15 g/cm³ when RHA was zero and this value decreased up to 1.46 g/cm³ when RHA was added up to 20% wt. together with 35% wt. vermiculite (Figure 6a). The maximum bulk density of samples with RHA was 1.88 g/cm³ obtained when RHA was added up to 5% wt. together with

20% wt. vermiculite. Bulk densities of ceramic bodies with both vermiculite and RHA are higher than those with vermiculite only because vermiculite is more porous than RHA. The incorporation of RHA in the range of 5 to 20% wt. in the tile composition had a favorable feature in bulk production in view of the enhancement of the densification process as well as the wide vitrification range of body mixes. According to [18] the solid density of the clay samples showed a steady reduction from 2.9 to 2.6g/cm³ as the RHA addition increased which correlates with the results obtained in this study. Since the bulk density is directly proportional to

strength of a material the addition of both vermiculite and RHA caused the bending strengths of the fired ceramic bodies to decrease from 28.86 MPa when the additives were 0% wt. to 8.50 MPa when RHA was 20% wt. and with 35% wt. vermiculite (Figure 6a). From these results, the maximum bending strength of the bodies with RHA and vermiculite additives was 18.54 MPa obtained when RHA was 5% wt. and with 20% wt. vermiculite. At least the bending strength for this body was too close to the required standards for wall and floor tiles applications, 16 MPa and 22 MPa respectively according to ISO 10545-4 [17].

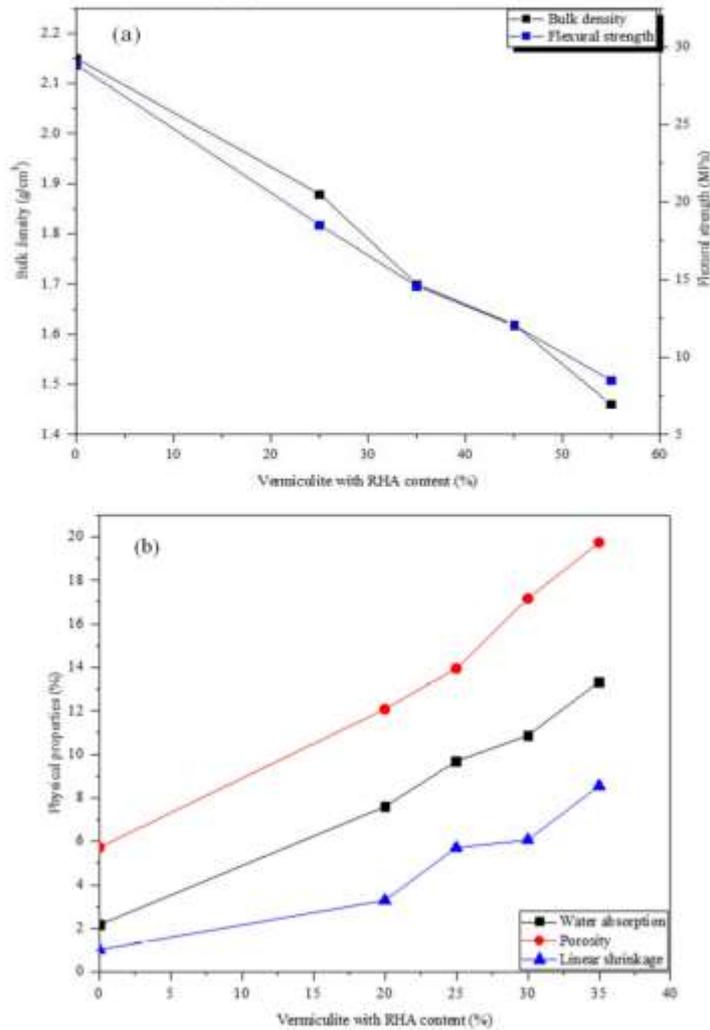


Figure 6. Influence of vermiculite and RHA content on the physical and mechanical properties of the fired ceramic bodies (a) bulk density and flexural strength (b) water absorption, porosity and linear shrinkage.

It is clearly observed that tile samples with both vermiculite and RHA have high bending strength compared to the tile bodies without RHA because more glass was formed due to the introduction of RHA which filled the pores, in this way the porosity and water absorption decreased, hence bulk density and finally the strength of the tile bodies' increased. Partial replacement of quartz by rice husk ash (RHA) was expected to reduce the possibility of β to α phase inversion of quartz which occurs at 573°C during cooling which results into decrease of quartz particle volume and may lead to cracks in the ceramic body [19]. According to literature information the progressive substitution of quartz by RHA in a conventional ceramic body composition resulted in an early vitrification of the mixes, while complete replacement of quartz by RHA drastically reduced both the maturing temperature and the percentage of thermal expansion, and increased the strength marginally [20]. Figure 6b shows the relationship between percentage of porosity, water absorption and linear shrinkage with addition of both vermiculite and RHA. Results show that; with addition of both vermiculite and RHA percentage of porosity, water absorption capacity and linear shrinkage increased from 5.72 to 19.74%, 2.17 to 13.34% and 1.02 to 8.55% respectively. But these values are low compared to that of tile bodies

without RHA. The linear shrinkage of tile bodies with both vermiculite and RHA is low due to the addition of RHA. When RHA was added, the quartz starts to dissolve rapidly and produces more silica content to assist feldspar in dissolving the particles in the tile bodies [21], this reduced the pores and hence lowering of the shrinkage. According to [18], the use of RHA in ceramic samples led to a steady reduction in linear shrinkage from 8.7 to 8.4% as the quantity of rice husks addition increased from 0 to 40% wt. Generally it can be revealed from the graphs that; with addition of vermiculite and RHA the porosity, water absorption and linear shrinkage were increased this caused a decrease in bending strength and bulk density. This is because vermiculite and RHA are porous materials with low density compared to clay and kaolin. Also vermiculite is a light material which expands at higher temperature and this result into the development of cracks due to formation of vacancy defects usually created when the body cools after firing. However ceramic bodies with both vermiculite and RHA had better physical and mechanical properties compared to those with vermiculite only. Theoretically, when the porosity increases, flexural strength and bulk density was expected to decrease and that was exactly what happened (Figure 7).

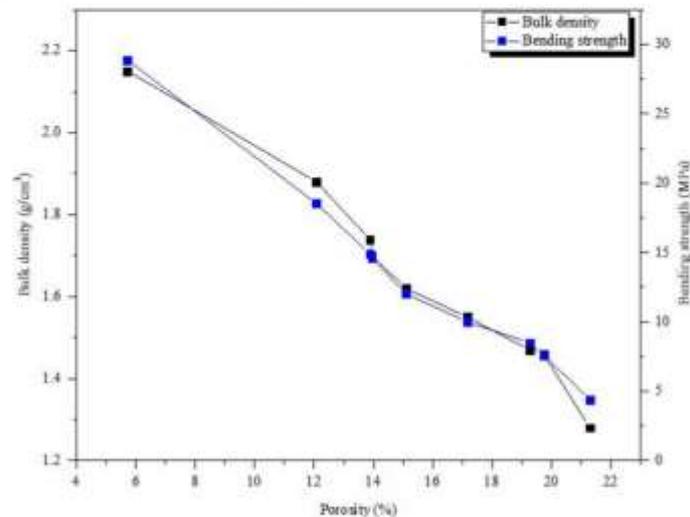


Figure 7. The effects of porosity on bulk density and flexural strength of the tile sample.

4. Conclusion

The following conclusion can be derived according to test results. With addition of vermiculite and RHA the porosity, water absorption and linear shrinkage of the fired bodies increased while the bending strength and bulk density decreased. Ceramic bodies with both vermiculite and RHA

had better physical and mechanical properties compared to those with vermiculite only. Among the studied compositions apart from the reference sample, tile bodies made from blend containing 20% wt. vermiculite and 5% wt. RHA were found to have the best properties for the production of ceramic tiles. For this combination the percentage of porosity, water absorption and linear shrinkage were 12.08%, 7.60% and

3.29% while the bulk density and bending strength were 1.88 g/cm³ and 18.84 MPa. These values were close to the required standards of wall and floor tiles which indicate that vermiculite and RHA are suitable and promising raw materials for production of ceramic tiles. Further studies on the strength and insulating properties of ceramic tiles should be done by analyzing the effects of addition of other agricultural wastes on ceramic tiles as well as reducing the iron content in vermiculite samples. The presence of high amount of iron in vermiculite increases thermal conductivity of the final fired body which also affects thermal comfort of the tiles. Depending on materials selection, batch formulations and forming processes (processing techniques) it is possible to obtain porous ceramics with high mechanical strength.

Acknowledgements

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Research poster

Evaluation of rice husk ash and vermiculite materials in the production of ceramic tiles

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Introduction

A typical ceramic tile composition will consist of 50% clay which imparts rigidity to the ceramic body, 25% quartz which lowers both drying and firing shrinkage and 25% feldspar which serves as a flux and also provides a glassy phase in the microstructure [1].

The challenging issues in ceramic tiles are low mechanical strength, thermal discomfort and high production costs. And in most efforts to improve strength, emphasis has been placed on minimization of quartz content in the ceramic tiles formula. This is due to β - α phase inversion of quartz which occurs at 573°C during cooling resulting to the development of stresses which initiate fracture and affects the strength of the final body [2].

Materials & Methods

Initially, a typical ceramic body composed of the mixture of vermiculite and RHA batched with Same' clay, Pugu kaolin, feldspar and quartz was prepared. Ceramic bodies were then obtained from this ceramic mixture by pressing samples at a forming pressure of 35MPa. These bodies were then fired at 1180°C in a laboratory furnace and finally the changes in the physical and mechanical properties caused by the introduction of vermiculite and RHA were tested and evaluated. The chemical composition of the raw samples was analyzed by X-ray fluorescence (XRF) while the phase composition was investigated using X-ray diffraction (XRD). The bulk density and open porosity of the sintered ceramic bodies were evaluated using Archimedes' principle while the flexural rupture strength was determined by the three point bending test method.

Results

The major chemical compounds in vermiculite raw sample were SiO₂, Al₂O₃ and Fe₂O₃ while RHA sample was found to contain mainly SiO₂. From the XRD analysis, vermiculite sample had crystalline vermiculite while RHA sample had amorphous silica at low temperature below 800°C and crystallized (tridymite) above 900°C. With addition of vermiculite and RHA the percentage of porosity, water absorption and linear shrinkage range were 12.06-21.29%, 8.21-16.81% and 3.85-12.00% while the bulk density and bending strength were 1.46-1.86g/cm³ and 4.40-18.54MPa respectively.

Figure 4: XRD analyses of (a) vermiculite and (b) RHA

Objectives

The objective of this work was to evaluate the possibility of using vermiculite and rice husk ash (RHA) in the composition of ceramic tile body

Figure 1: (a) Vermiculite and (b) rice husk samples

Figure 2: Simplified geological map of Mozambique belt showing vermiculite and kaolin deposits in Tanzania [3]

Figure 3: Ceramic products fabricated using Tanzania raw materials

Table 1: Chemical composition of raw materials

Raw material	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	Total
Vermiculite	52.00	18.00	1.00	0.00	0.00	0.00	0.00	71.00
RHA	88.00	0.00	0.00	0.00	0.00	0.00	0.00	88.00
Same' clay	45.00	15.00	1.00	0.00	0.00	0.00	0.00	61.00
Pugu kaolin	55.00	18.00	1.00	0.00	0.00	0.00	0.00	74.00
Feldspar	65.00	15.00	1.00	0.00	0.00	0.00	0.00	81.00
Quartz	100.00	0.00	0.00	0.00	0.00	0.00	0.00	100.00

Conclusion

The values of the physical, mechanical and thermal properties of the obtained ceramic bodies were close to the required standards of wall and floor tiles. This shows that vermiculite and rice husk are promising raw materials in ceramic tiles industry.

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