

Fabrication of Porous Clay Ceramics Using Kenaf Powder Waste

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Abstract- Porous clay ceramics were fabricated by using kenaf powder waste (KP as a pore-forming agent PFA). The raw clay powder was characterized using XRF, XRD and TGA. Samples were prepared with different kenaf powder wastes (up to 30wt.%) using granulated powder mixtures and followed by hydraulic pressing. After drying, the ceramic green bodies were fired with different temperatures ranging from 1100 to 1175°C for 3 h with a heating rate of 5°C/min. The pore formation and microstructure in the samples were characterized using FE-SEM, while bulk densities, porosities and water absorption were determined using the Archimedes method. Their mechanical properties were also investigated in order to optimize the fabrication process itself. The results obtained showed shrinkages between 11.72-15.90% for the samples. The density values decreased from 1.30 -1.75g/cm³, while porosity was determined to be between 11.86 to 45.64 %. The tensile strength was in the range of 9.06 to 24.05MPa. These results prove that kenaf powder wastes (KP) are potentially capable to produce porous ceramic materials.

Index Terms- Laotian clay; Sintering temperatures; Shrinkage; Porosity; Kenaf powder waste

I. INTRODUCTION

Porous clay ceramic materials are used for a wide range of applications such as insulators, catalyst, supports, filters, etc. It can be produced using various techniques for example by addition of chemicals, polymers and organics pore forming agent [1]. Environmentally friendly materials which can be recycled and conserve energy are very important in many research fields today. On the other hand, as a result of environmental regulations the demand for clay with higher insulation capability has increased [2-4]. It is well known that to increase the insulation capacity of the porous clay, it is important to generate porosity in the body. Typically, organic pore-forming agent additives are used. Sawdust, polystyrene, paper, sludge, coal and coke are some examples of organics which are used as pore forming agent materials. Recently, Perlite, diatomite, calcite, pumice and vermiculite which are examples of inorganic minerals have also been used as pore-forming agents [2; 3]. Organic pore formers are generally cheaper than inorganic ones and also have advantage of ensuring a heat contribution to the firing furnace. However, CO₂ emission is the main drawback of organic pore

formers. Inorganic pore former have less environmental problems but they may change the plasticity of the clay system negatively and increase the amount of water needed to maintain acceptable plasticity. Organic product residues are extensively used as a pore former in the clay industry [5; 6]. Kenaf powder waste on the other hand can be derived from bamboo [4] and sawdust [2]. Malaysia is the world's largest producer of industrial kenafs. The main objective of this study is to investigate the effects of the aforementioned types of organics to create pores, as well as understand the properties of kenaf powder with firing clay processes.

II. EXPERIMENTAL

The raw clay materials used in this study was obtained from bricks and tiles at construction sites in Lao PDR. These areas have traditionally been an important place for the production of building bricks and tiles. The clay behavior during sintering was examined using similar process parameters as that of traditional techniques used in local brick manufacturing operations. The kenaf core waste was selected from Malaysia, then grinded to form the powder. Selected mixtures containing 0, 10, 20 and 30 wt % kenaf powder were prepared (Table1).

Table 1 The proportions of the mixtures for the formulations (wt.%)

Formulation	Clay (BC)	Kenaf powder (KP)
BC	100	0
BCK10	90	10
BCK20	80	20
BCK30	70	30

The mineralogical analyses of the raw clay samples as well as its corresponding clay fraction were carried out utilizing (XRF) and X-ray diffraction techniques (XRD). The XRD patterns were obtained with a Rigaku Rint-2200 (Japan) diffractometer operating at a tube voltage and current of 30 kV and 30 mA respectively using monochromatic Cu-K α 1 radiation ($\lambda=1.5406 \text{ \AA}$). Diffraction patterns were recorded between 2 and 70° 2 θ with scanning rates of 2°/min.

Differential thermal analysis (DTA) of the clay was performed with a ramp rate of 10°C/min from room temperature to 1200°C in air. The presence of possible defects in the sintered

samples as well as their surface morphology was inspected with Scanning Electron Microscopy (SEM SHIMADZU SSX-550).

The raw materials (Table1) were mixed and sieved using a 500 µm screen. The specimens were then hand-pressed in a hydraulic system using stainless steel moulds. The samples had a diameter of 23mm and thickness of 4mm. Similar samples were also prepared and sintered for determining the tensile strength via diametral compressive methods (DMA). The green body samples were dried at 80°C for 24h to attain equilibrium residue moisture content in the clay bodies. Dried samples were then sintered with heating rate 5°C/min at a maximum temperature of 1175°C with soaking time of 3h and allowed to cool down naturally inside the furnace. The shrinkage, density, porosity, water absorption and compressive strengths were determined in accordance with ASTM standards (C326, ASTM C373, 2005).

III. RESULTS AND DISCUSSION

a. Analysis of materials

The crystalline phases were in agreement with the results observed via XRF (Table.2), whereby the clay and kenaf powder consisted mainly of SiO₂ and Al₂O₃ which make up about 78% of BC in accordance with the quantitative XRD analysis. Table 2 presents the chemical composition of the raw clay and kenaf powder. The clay minerals and kenaf powder are presented a typical composition, which constitutes mainly of silica, alumina and minor contents of Fe, Mg, Ti, K and Ca oxides. The amount of CaO and MgO was low and indicated the absence for carbonates. Particle size distribution was determined by Laser particle analyzer on 0.1-100µm fractions. The particle size distributions of the clay sample of the arranged particle size is 0.68 µm.

X-ray diffraction (XRD) was used to identify the mineralogical phase of the raw clay where the following compounds were found in the raw clay material: Quartz (SiO₂), Kaolinite (Al₂(Si₂O₅)(OH)₃), Illite (KAl₂(Si₃AlO₁₀)(OH)₂), Gibbsite (Al₂(OH)₃) and Montmorillonite (Na,Ca)_{0.3}(Al,Mg,Fe)₂Si₄O₁₀(OH)₂xH₂O. In the fired samples phases are identified as oxides and hydroxides, as well as silicate in addition to residue quartz as mentioned by [7]. Above 1100°C, new crystallite phases are formed, which are quartz(SiO₂) and mullite (2Al₂O₃ SiO₂) with a dominant presence of quartz [8], phase could be found in presence of a minor amount of MgAl₂O₄ spinel and mullite phases. As the sintering temperature increased from 1100 to 1175°C, the diffraction peaks of the cordierite phase became more intense, suggesting that the crystallinity of the cordierite phase in the membrane improved. As a result the highly crystalline structure composed of mullite in clay [9]. It is important to notice that mullite contents increased at 1175°C as shown in X-ray pattern in Figure 2.

Table.2. Chemical compositions of the black clay and kenaf (BC and KP) (wt.%)

Oxides	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	TiO ₂	K ₂ O	CaO	NaO ₂	L.O.I
BC	55.00	25.00	3.60	0.76	2.60	1.30	0.24	14.00
KP	2.52	0.41	0.24	0.06	1.45	0.06	0.62	92.87

LOI*, Loss on ignition at 1000°C

Figure.2. shows the TG/DTA curves of the raw powder. At temperatures less than 100°C, followed by dehydroxylation ether are endothermic DTA peak and about 6 wt% mass loss, which were caused by the removal of residual physically adsorbed water. There is a small endothermic DTA peak at 167°C and about 4 wt% weight loss because during crystallization water evaporated from basic magnesium carbonate. Because the ceramic powder was pre-heated at 1200°C, during which kaolinite transformed into meta-kaolinite and aluminum hydroxide into alumina, no weight loss or thermal effect is shown for the phase changes in ceramics themselves. Between 400 and 600°C, a large exothermic DTA peak appears which is attributed to the phase transformation from metakaolinite to Al-Si spinel. For temperatures ranging between 900 to 1000°C, the exothermic DTA peak indicates the crystallization of MgAl₂O₄ spinel phase as revealed from TG curve has no weight loss.

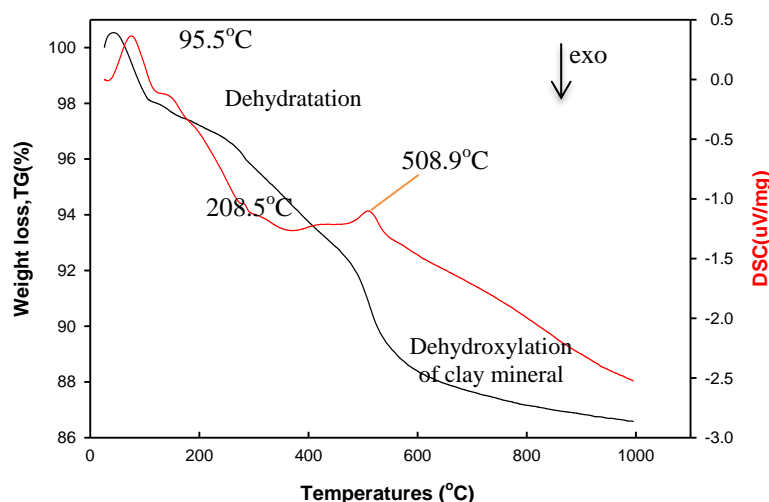


Figure 2. TG/DSC curves of the LC raw clay powder.

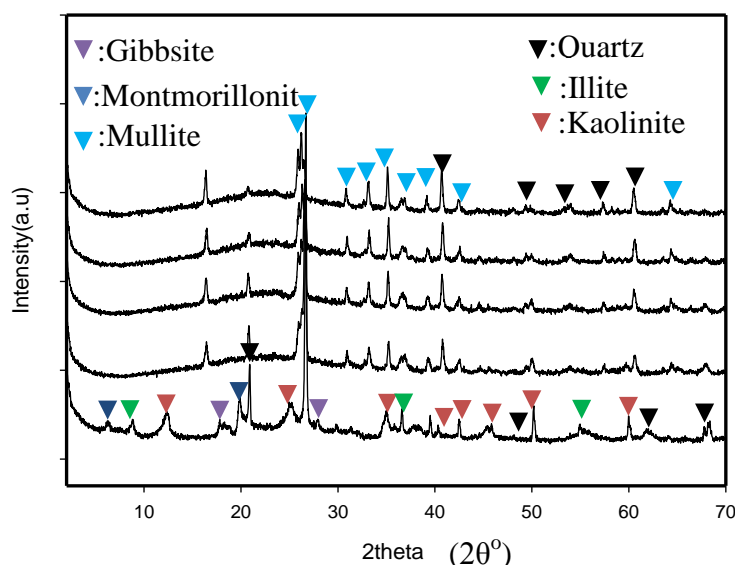


Figure.3. XRD diffraction patterns of the BCKP30 at fired different temperatures (a): raw; (b): 1100; (c): 1125; (d): 1150 and (e): 1175°C

b. Sintering behaviour

The samples were fired from 1100 to 1175°C followed by measurement for shrinkage, density, apparent porosity and water absorption. These properties are shown in Fig.4. The shrinkage, density apparent porosity and water absorption changed depending on sintering temperatures and kenaf powder content. An increase in kenaf powder (KP) waste content from 10 to 30 wt.% showed an increase in shrinkage, porosity and water absorption. The main reason for such a trend is the composition of organic residues during the sintering. [2]. In this case, the effect is related to kenaf powder (KP) composition residues whereby all of organics were burnt off with the firing processes.

In addition, the shrinkage, apparent porosity and water absorption all of values of sample were found that both of properties were increased with increasing KP content addition while the densities were significantly reduced [2-4]. However, the shrinkage and density increased with increasing sintering temperature, while apparent porosity and water absorption decreased with sintering temperatures as depicted in Figure. 4(a-d).

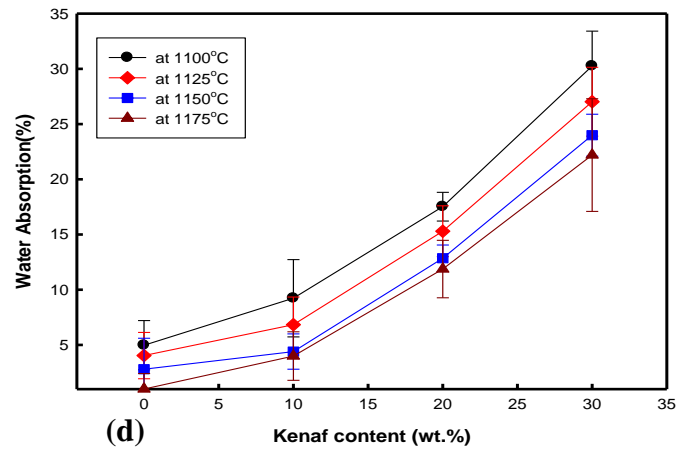
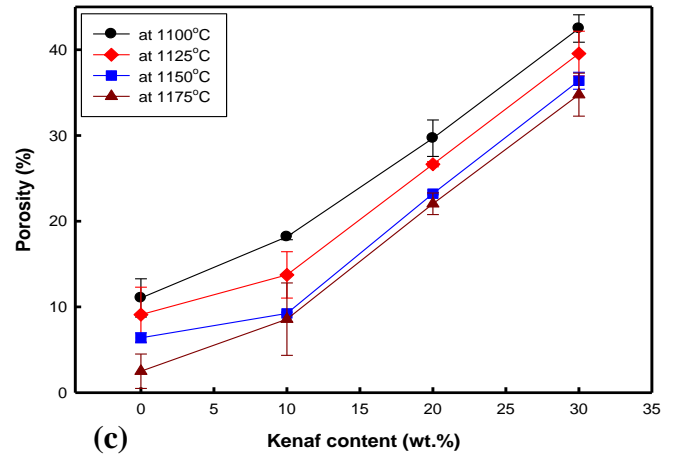
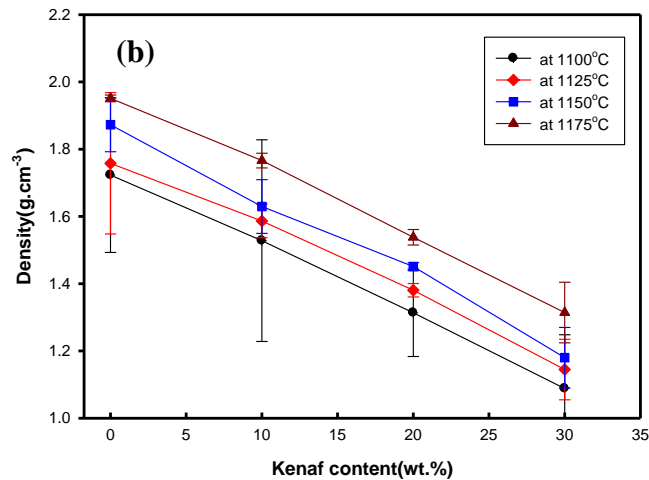
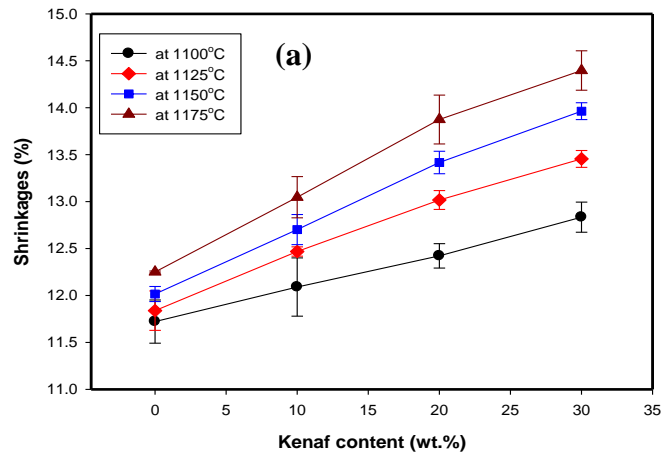


Figure 4. Shrinkage, density, porosity and water absorption of the samples BCKP with kenaf contents sintered at different temperatures.

c. Mechanical properties and Structural morphology observations

The mechanical properties of the samples were determined in terms of diametral tensile strength (DTS). This is performed to obtain the mechanical properties of the sintered porous clay samples. It can be seen that the tensile strength decreased as KP content increased. The DTS of the porous clay ceramic samples having increasing KP content decreased especially when the ratio increased between 10 to 30 wt.%. Such a behavior is mainly related to factors such as decomposition of organic matter from the KP waste which generates pores in the fired structure [3; 4; 10].

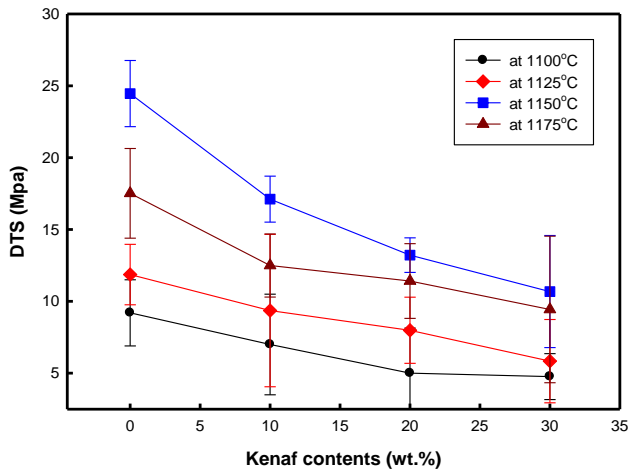
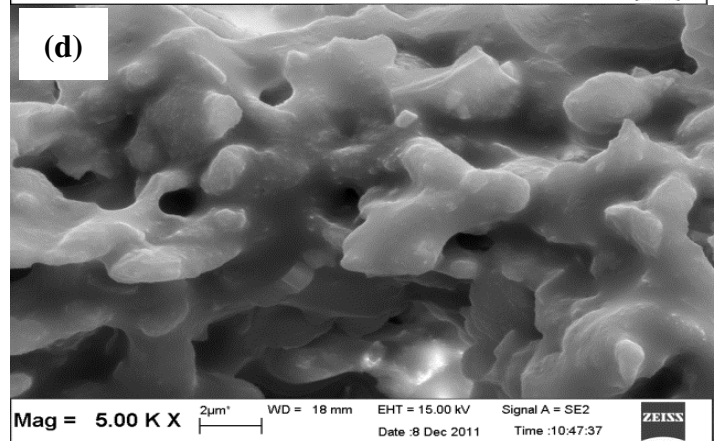
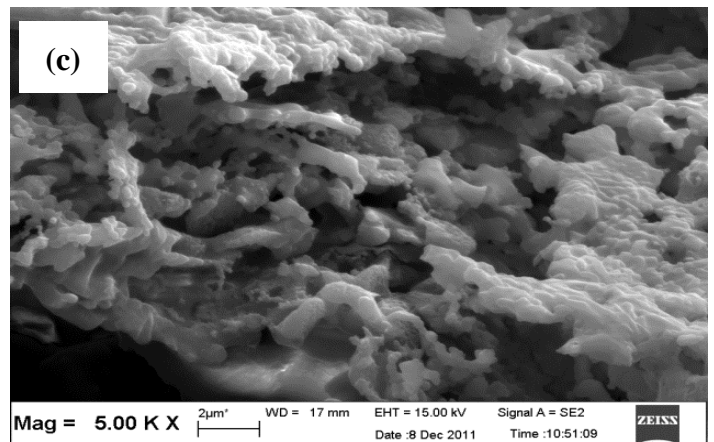
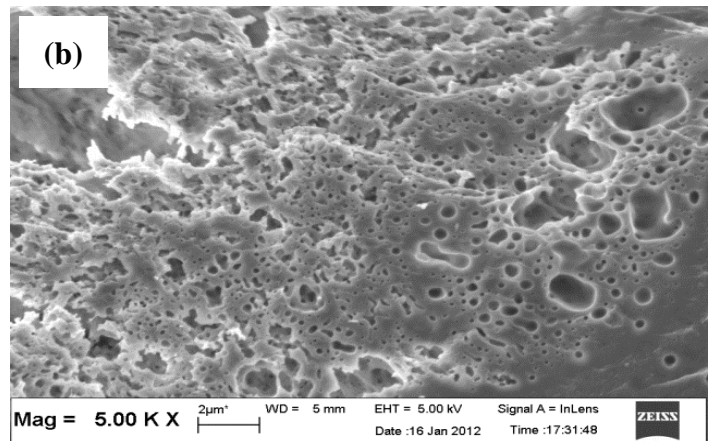
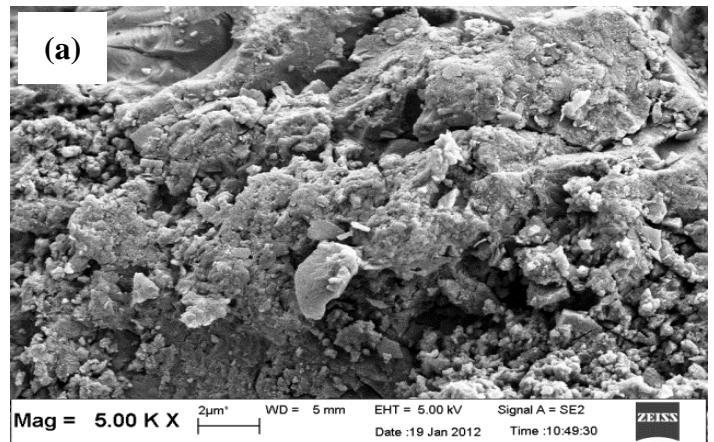


Figure 5. Diametral tensile strength (DTS) of the samples BCKP sintered at different temperatures.

The sintering process involving ceramic materials has been widely studied and is known to have a pronounced impact on the resulting microstructure [7]. The microstructure of sintered samples was studied with a scanning electron microscope (SEM) to determine the size, shape morphology and distribution of the pores in the microstructure. The SEM images in Figure 6 show the evolution of the microstructure of the sintered samples. The fracture surface of bodies sintered at 1100°C is characterized by the presence of interconnected pores essentially, distributed homogeneously in the ring fracture surface whose size are significantly larger than those sintered at 1175°C. Spherical pores were observed in mature microstructure where a sort of equilibrium between the gas pressure and capillary pressure was reached. At 1150°C some of the pores were closed and the homogeneity of the ceramic ring decreased.

Figures 6(c - h) showed that the pore size and shape increased when kenaf powder increased from 10 to 30 wt.%. The results show that all clay minerals were transformed to mullite phases. This was also seen via X-ray diffraction studies seen (Fig.3), thus the formation of mullite phases may have started at this temperature [11]. However, when the sintering temperature increased, the pores and grain sizes decreased because of an increase in density of the material itself.

It is reported that mullite has a special crystallographic structure, which demonstrates strong- bond chain links [9; 12]. This could explain the grain growth great with increasing KP content as shown in Fig.6. It is very clear that the densification was due to enhanced grain growth along the grain boundaries.



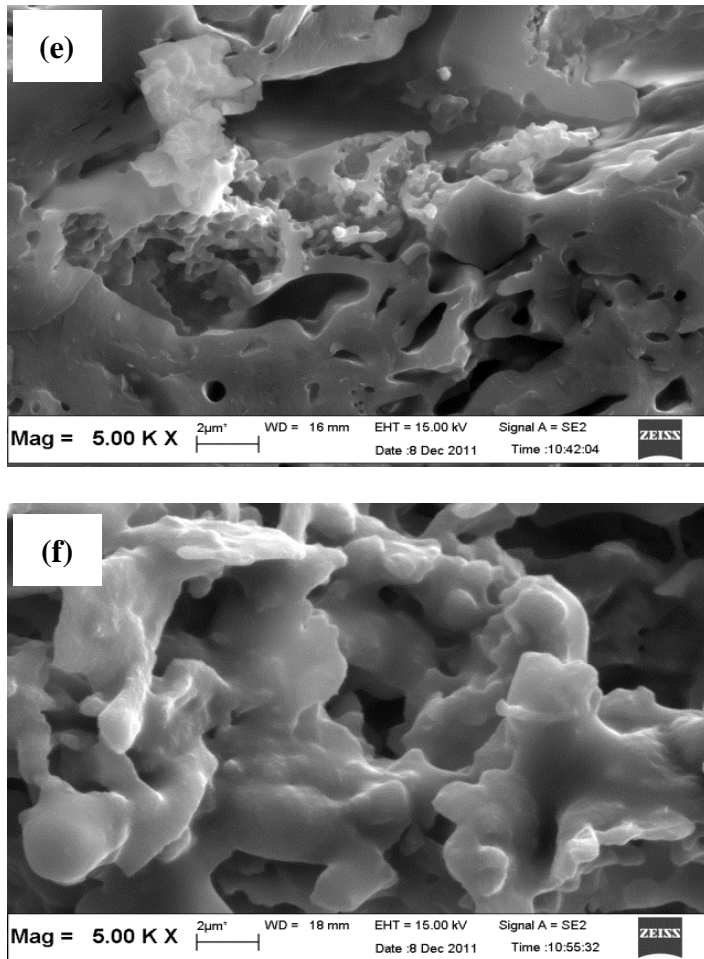


Figure.6. the SEM micrographs of BCKP fracture surface samples with (10; 20wt.%) of kenaf powder fired at different temperatures: (a-b) 1100°C; (c-d) 1125; (e-f) 1150 and (g-h) 1175°C (10 and 20 wt.%).

IV. CONCLUSION

In this literature work, the focus is to develop porous clay ceramic materials which were fabricated using kenaf powder waste as a pore forming agent. Based on the experimental investigations reported in this paper, the following conclusion can be made:

- (1). By increasing the organic content in the clay body, an increase in shrinkage, porosity and water absorption was seen, which also reflected in a decrease of density and tensile strength.
- (2). The organic residues are easily burnt off from the clay body during firing.
- (3). Organic residues can be effectively used for pore-forming for up to 30wt.% residue addition by weight. It has been found that further addition of organic residues is not very effective for decreasing the density of the clay body. The residues increased the porosity and decreased density of the material.
- (4). The organics residues can be used as a pore-forming

additive in clay bodies without any damaging effect on the clay product's manufacturing process itself.

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