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ScienceDirect

Ceramics International 40 (2014) 14997–15006

CERAMICS INTERNATIONAL

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Rapid starch consolidation of red clay-based ceramic slurry under simultaneous pressure-cooking and microwave irradiation

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Received 11 May 2014; received in revised form 19 June 2014; accepted 19 June 2014
Available online 2 July 2014

Abstract

This study presents a simultaneous application of pressure cooking and microwave irradiation to consolidate ceramic slurry as a rapid shaping technique. The ceramic slurry, composed of red clay, feldspar, quartz and water, was solidified with the gelatinization of starch under varying heating conditions inside a domestic microwave pressure cooker. It was found out that, for a small amount of ceramic slurry, a shortest heating time of 2.52 min was achieved at microwave pressure cooking conditions of 50% power level and target temperature of 60 °C. Based on these conditions, large shaped ceramic prototypes of different shapes were successfully formed using proposed empirical equations to determine a desired heating time at any power levels. The physical characterizations revealed that the physical properties of dried and fired ceramic bodies formed under microwave pressure cooking were better than that of the conventional counterpart. Microstructural investigation into the fired samples revealed larger void spaces in conventional sample than that in microwave pressure-cooked sample.

Keywords: Microwave; Pressure cooking; Red clay; Consolidation; Starch

1. Introduction

The starch consolidation technique has been widely explored in the field of ceramic processing for forming ceramic materials through the use of starch loaded in ceramic slurry. The starch has gained popularity in ceramic processing as a pore former and forming agent owing to the lack of environmental concerns and defect-free burnout. The benefit of using starch is economically cheap for implementation in ceramic processing to produce porous ceramic compacts with diverse applications. This technique has been commonly applied to shaping ceramic powders, such as pure oxide ceramics [1–3], mullite [4], and cordierite ceramics [5]. Interestingly, important applications are gaining renewed interest for clay-based systems involving a ternary mixture of red clay, quartz and feldspar [6–8]. The clay, containing iron impurity, imparts plasticity to the powder mix while the quartz provides filler during compaction. The inclusion of feldspar in the powder mixture reduces the firing temperature of the resulting ceramic compact. This ternary ceramic mixture offers possibility to synthesize an antibacterial ceramic filter containing natural iron and alkalis for water purification and wastewater treatment [7].

The solidification of the ceramic slurry to form a ceramic compact is imparted by the gelatinization of starch at elevated temperature. This thermal effect is a complex process, involving granule swelling and disruption of two polysaccharide molecules of starch: amylose and amylopectin [9]. The amylose is a predominantly linear molecule linked by α-1,4 bonds, and amylopectin is a highly branched polymer with branch points linked by α-1,6 bonds. The ratio of amylose and amylopectin affects the physicochemical properties of starch. In ceramic processing, the starch is commonly mixed with the aqueous ceramic suspension and poured into impermeable mold, which is subsequently heated up to a gelatinization temperature between 55 °C and 80 °C. This gelatinization

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http://dx.doi.org/10.1016/j.ceramint.2014.06.100
0272-8842/

process is usually associated with impairment and rearrangement of the intermolecular bonds in the polymeric constituents [10], while heat is applied on the ceramic slurry. During this heating process, a rapid and irreversible swelling of the starch granules takes place by water uptake that consolidates the ceramic slurry into a solid compact. The alteration of the starch granules by heat leads to increased viscosity of the slurry, which finally locks water causing the solid particles to stick together and consolidate into a rigid body [11]. Therefore, the success of the starch consolidation technique depends upon the heat transfer between the mold material and the ceramic slurry, and the method of heating employed to achieve homogeneous gelatinization of the starch component.

The starch-loaded ceramic slurry is conventionally heated in a convection oven to effect the thermal-assisted consolidation. The applied heat is transferred from the mold material to the ceramic slurry, which subsequently elevates the slurry temperature. This heating method works very well for small amount slurry, but it poses serious problems for a large amount of ceramic slurry due to a long heating time in achieving the gelatinization temperature of the starch. The long heating time results in non-uniform heating of the ceramic slurry and unavoidable sedimentation of ceramic particles. To resolve these heating problems, a microwave heating process has been explored to solidify ceramic slurries being loaded with thermally responsive consolidating agent [12]. This heating method allows uniform heating of ceramic slurry at a relatively short time due to the volumetric heating effect [13]. It means that the heating process starts from the interior of the material to its surface, leading to uniform temperature distribution. Although this method presents some advantages over conventional heating, it poses some challenges with the use of high microwave power and the heating of ceramic slurry contained in a sealed/unsealed mold. A sudden build-up of pressure is developed due to fast heating process that causes spilling and boiling of the ceramic slurry. Moreover, it is only effective for solidifying ceramic slurry at low power lever (about 10%) to avoid the sudden build-up of pressure [12]. Thus, the microwave heating method also presents long heating time for the large amount of slurry, which reduces the production volume.

To avoid the boiling of ceramic slurry under the microwave field, a heating method that takes advantage of the elevation of the boiling point of water was successfully explored with the use of domestic pressure cooker [14]. This method assures the attainment of the pasting or gelatinization temperature of starch in the ceramic slurry as compared with the conventional heating process. However, the issue of long heating time was partially solved because it relied on the heat transfer of the materials and the pre-heating of the pressure cooker and the load to cause pressurized steam heating [15]. Thus, the present study demonstrates a simultaneous pressure cooking and microwave irradiation to consolidate red clay-based ceramic suspension loaded with the starch powder. This can be conveniently carried out inside a microwave pressure cooking system that speeds up the gelatinization of starch and shortens the heating time. This new system operates with the same heating principle as the conventional pressure cooker [15,16]. The difference with this new system is that the whole assembly is made up of microwave transparent materials [17]. This means that only the loads that couple microwaves are heated while steam is produced to elevate the internal pressure in a very short time. The sudden boiling of ceramic slurry due to microwave irradiation is possibly avoided by the pressurized environment, owing to the boiling point elevation of the liquid involved [18].

This study aimed to investigate the consolidation of red clay-based ceramic suspension using the gelatinization of starch inside a domestic microwave pressure cooker. Specifically, this study has fourfold aims: (i) determine the optimum power level, microwave exposure time and temperature in terms of the gel strength of a green body, (ii) compare the physical properties of the microwave pressure-cooked consolidation to the conventional method of starch consolidation, (iii) investigate the microstructures of fired ceramic bodies formed under the microwave pressure-cooking and the conventional method of starch consolidation, and (iv) fabricate different shapes of prototypes formed under microwave pressure-cooked consolidation.

2. Materials and experimental procedures

2.1. Preparation of raw materials

The red clay used as plastic material was mined in lumped form from Lama–Lama, Tubod, Lanao del Norte, Philippines. It was dried in a conventional oven for 4 h at 110 °C to completely remove all the moisture present. The dried clay was crushed in a ball mill and passed through 100 mesh sieve with the help of a rotary shaker. The non-plastic materials such as feldspar and quartz were commercially available with average particle size (LS 100Q, Coulter Corporation) of 35.1 μm, and 27.0 μm, respectively. Correspondingly, the chemical composition of the ceramic raw materials is given in Table 1. These powdered materials were dispersed in distilled water with the aid of sodium tripolyphosphate (STPP). The non-plastic materials and dispersant were supplied by Elmar Marketing, Mindanao, Philippines. A commercially available potato starch was used as a binding agent with an average particle size of 40.92 μm. A small cylindrical mold cavities (diameter: 2.94 cm and height: 3.45 cm) for a small amount of slurry were assembled by cutting an acetate film and attaching the cut pieces with double-sided tape and masking tape.

Table 1
Chemical composition of the raw materials.

Materials	Oxide contents							
	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	l.o.i.
Red clay	41.6	25.2	8.2	1.1	0.5	0.5	0.5	22.4
Quartz	95.1	1.7	0.4	1.4	0.5	0.1	0.3	0.6
Feldspar	66.3	21.5	0.3	0.5	2.4	8.5	0.1	0.4

2.2. Preparation of red clay-based ceramic slurry loaded with starch

Aqueous ceramic slurry was prepared with 70% of solid loading, which composed of 46.70 wt% clay, 35.30 wt% feldspar, 18.00 wt% quartz and 30% of water. This ternary formulation was based on the reported stoneware ceramic body [14]. The ceramic slurry was prepared by filling 30% water, based on the desired total batch weight, and the right amount of dispersing agent based on the amount of clay was added into a cylindrical polyethylene container. To facilitate mixing of the solution, alumina balls equivalent to the mass of the ternary powder were added and rolled for 3 min. The powdered clay was then poured into the container and milled for 4 h. When the clay powder was dispersed, the container was then added with the dispersant based on the mass of feldspar and quartz and milled for additional 3 min to achieve a better mixing. Then, the prepared feldspar and quartz were then added gradually and milled for additional 4 h. All milling speed was implemented with a rotation speed of 55 revolutions per minute (rpm).

The prepared ceramic slurry of 70% solid loading was loaded with 12 wt% potato starch based on the mass of ternary powder. An additional milling step was employed to homogenize the ceramic-starch slurry for about 4 h. After this milling step, the ceramic slurry was passed through a 70 mesh screen to remove the milling balls and to remove large agglomerates. Correspondingly, the rheological data of the ceramic slurries loaded with and without the starch powder were then immediately collected using a viscometer (RVDE 230, Brookfield Engineering Laboratories, Inc.). The measurements were conducted at varying spindle speeds of 10 rpm, 20 rpm, 30 rpm, 50 rpm, 60 rpm and 100 rpm. Subsequently, the flow curves of the starch-loaded ceramic slurries were reported in terms of viscosity against varying spindle speed.

2.3. Microwave pressure cooking and conventional cooking of the ceramic slurry

The potato starch-loaded ceramic slurry was then cast to the assembled cylindrical mold until 25 g of slurry were loaded. Five replicates of slurry-loaded cylindrical molds were prepared and set in a domestic microwave pressure cooker (Meyer, Japan, volume: 2.2 l, inside diameter: 18 cm, base thickness: 5.3 mm). The body and cover steaming rack were made of food grade polypropylene plastics that are microwave transparent. The lid was equipped with a silicone rubber that secured the tightness of the chamber. The microwave pressure cooker was initially loaded with tap water of about 300 ml to ensure enough source of steam during heating. It was then locked tightly and placed in a modified domestic microwave oven (Sharp, model: R-390H(S), power: 1100 W, volume: 34 l).

The schematic diagram for the microwave pressure cooking experiment is shown in Fig. 1. There were several type of microwave ovens, but in this study we preferred to use the oven with a magnetron (source of microwaves) located at its right side. It was then convenient to drill a small hole at the center top of the oven for inserting a resistance thermometer. The microwave oven was equipped with timer and alarm to signal the completion of heating. A turntable (diameter: 335 mm) also was provided, which was rotated in all experiments to ensure homogeneous transmission of microwaves. To approximately measure the slurry temperature inside the microwave pressure cooker, external test slurry similar to the slurry inside the microwave pressure cooker was penetrated with the temperature sensor until its center. This allowed continuous transmission of microwaves until the preset or target temperature at a desired power level. The temperature was monitored by an interface module, and the microwave power was controlled by a smart controller. This controller was equipped with digital LED display, programmable function, and PID auto tuning capability. The microwave oven had five power levels of 10%, 30%, 50%, 70% and 100%. For the small amount of ceramic slurry (25 g), power levels less than 70% were chosen because they provided a sufficiently long exposure time. The preset or target temperature was based on the pasting temperature of starch between 60 °C and 80 °C [19,20].

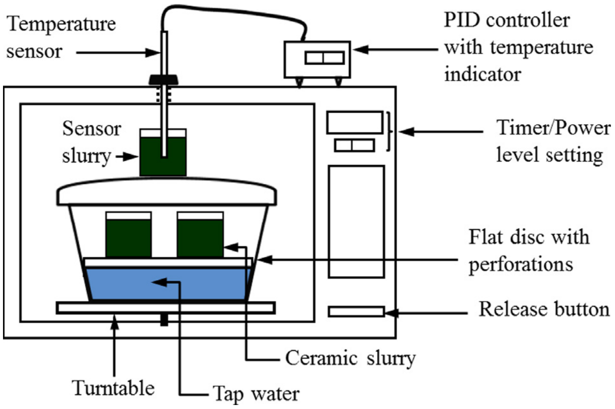

Fig. 1. Schematic diagram for microwave pressure-cooking experiment.

The microwave heating procedure was done at the varying power levels, and the microwave power was manually stopped when the target temperature was reached. The cooking time was then noted, and the pressure cooker was withdrawn from the microwave oven. Before opening the pressure cooker, tap water was consistently run over its outer surface to induce cooling for about 3 min. Subsequently, the cover of the microwave pressure cooker was slowly removed to release the contained pressure, and the solidified samples were withdrawn. The gelled samples were cooled for an additional 1 h, and the gel strengths were immediately measured using an unconfined compression apparatus (Marui & CQ., LTD., Osaka, Japan) equipped with 5 mm circular plunger. The plunger was allowed to penetrate to the gelled sample until a depth of 5 mm and the loading force was noted. The gel strengths were then computed as the force divided by the cross-sectional area of the plunger, and the average of three measurements was reported. Subsequently, the microwave pressure cooking time that provided the highest gel strength was selected as the best heating time inside the microwave pressure cooker. For comparison, similar casted potato starch-loaded ceramic slurry was placed in the conventional oven and heated for 3 h at 80 °C. Five replicates were prepared and solidified in this method.

2.4. Fabrication of large ceramic prototypes

Large ceramic prototypes were made in different shapes such as tile and hollow disk in order to justify the applicability of the present study. For a large amount of slurry, tile cavity (length: 10 cm, width: 10 cm, and thickness: 2 cm) and hollow disk cavity (outside diameter: 15 cm, inside diameter: 12.5 cm, and height: 3 cm) were assembled in similar procedure as the assembly of the small mold cavities described above. The plastic molds were then filled with the potato starch-loaded ceramic slurry and set inside the microwave pressure cooker. Finally, the microwave heating process followed the same procedure as the small sample and was conducted using the best heating conditions. It was expected that, for the large amount of ceramic slurry, the solidification of the ceramic slurry was unlikely. Hence, an appropriate heating condition was calculated by using proposed empirical equations based on the best heating conditions.

2.5. Drying and firing of the gelled ceramic bodies

The gelled ceramic bodies were demolded and dried at room conditions for one week. The naturally dried samples were further oven dried at temperatures between 50 °C and 110 °C until reaching a constant weight. The dried samples were then polished (Leco VP-50, Leco Corporation) into symmetric disks. After this polishing step, the polished samples were fired in an electrical muffle furnace (Vecstar furnaces) to a firing temperature of 1200 °C. The firing schedules started from 100 °C with 1 h soaking and the temperature was then increased up to 300 °C with 2 h soaking. The temperature was further increased to 500 °C, 800 °C, and 1100 °C, with 1 h soaking for each temperature to avoid firing cracks. Finally, upon reaching the temperature of 1200 °C, the soaking was 2 h and the furnace was naturally cooled.

2.6. Physical characterizations

The dried and fired shrinkage was determined by measuring the initial and final heights of the green and fired cylindrical samples with the use of a vernier caliper. The shrinkage was then calculated as initial height minus final height divided by the initial height. The dried bulk porosity was determined on polished disk samples. The dried porosity was then calculated as one minus bulk density of the sample divided by an equivalent powder density of the sample determined by the pycnometer method. Finally, all results were multiplied by 100% and the average of three measurements was reported.

The porosity of fired disk samples was determined by boiling in water-filled container for 4 h. After boiling, the fired samples were naturally cooled for 24 h in water before weighing. The apparent porosity was calculated using Eq. (1).

$$\%\mathrm{AP} = [(I - F)/(I - S)] \times 100\% \tag{1}$$

where %AP is apparent porosity, I is the weight of the immersed sample after boiling, F is the weight of fired sample before immersion, and S is the suspended weight of the sample while it was submerged in water.

To determine the mechanical property of the consolidated samples, both dried and fired disk samples (diameter = 25 mm, thickness = 15 mm) were prepared and compressed on a versa-loader machine (Model G-900-4, ELE International, SOILTEST Products Division). The breaking force, y in unit kgf, was determined using an Eq. (2).

$$y = 4.466171 \times -25.061 \tag{2}$$

where x is the dial reading on the machine multiplied by 10 divisions. A diametral compressive strength was then calculated using the equation:

$$\mathrm{DS} = \frac{2P}{(\pi D t)} \tag{3}$$

where DS is the diametral compressive strength in mega pascal (MPa), P is the breaking force (N), D is the diameter (mm), and t is the thickness (mm). Subsequently, the average of three measurements was reported.

The microstructural features of the fired ceramic samples shaped inside the pressure cooker were compared to ceramic samples shaped in the conventional oven. Small fractured fragments were taken from fired samples and observed on a scanning electron microscopes (JSM-6100, JEOL, Japan). The fractured surface of the specimen was platinum sputtered to aid viewing at ×250 magnifications.

3. Results and discussion

3.1. Rheological behavior of the starch-loaded ceramic slurry at room conditions

The concentrated suspension of pure starch in water [21] usually exhibits an increasing viscosity with increasing applied shear stress, which is typical of shear thickening behavior. This slurry behavior is a consequence of its dependence on particle configuration accompanied by an order to disorder transition in the starch particle configuration [22]. When the starch powder is added in a small amount to ceramic slurry, the characteristic behavior of the ceramic slurry is expected to be retained. Indeed, this can be observed on the viscosity profile of the ceramic slurries loaded with and without starch as shown in Fig. 2. It can be seen that the viscosities of both slurries are decreasing with the increasing spindle speed (rpm). This is due to the induced shearing effects that break down particle networks, resulting in a shear thinning behavior [23]. This slurry behavior is a requisite for a direct casting process. The shear thinning behavior can be illustrated according to the arrangement of ceramic particles in suspension. During near equilibrium of ceramic suspension, random collisions among particles make them naturally resistant to flow leading to a

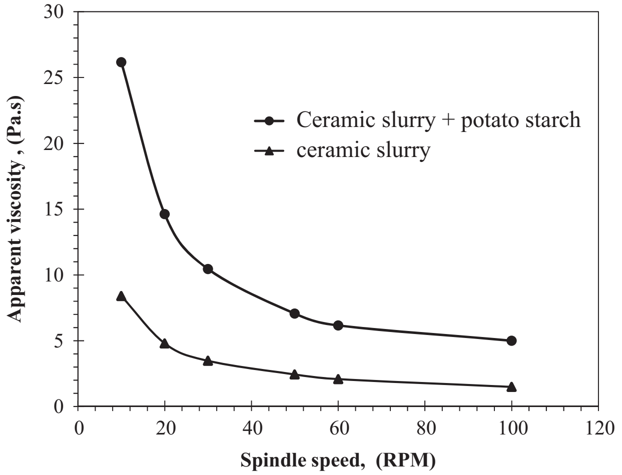

Fig. 2. Comparison of the viscosity against spindle speed of the ceramic slurries loaded with/without potato starch.

high observed viscosity. But as the shear stress (or, equivalents, the shear rate) increases, particles become organized in streamlined configuration with the flow, which ultimately lowers viscosity [24].

Although the shear thinning behavior is observed for the starch-loaded ceramic slurry, its viscosity increases when it is compared with the ceramic slurry without starch. Such behavior can be attributed to a possible adhesion of ceramic particles on the potato starch granules leading to a particle clustering [25]. Also, the increased viscosity may be due to a reduction of the available liquid for dispersion due to the addition of starch granules and the associated liquid wetting on the surface of starch granules [26]. Nevertheless, the observed viscosity of the potato starch-loaded ceramic slurry remained pourable into a desired mold. The specific gravity of the ceramic slurry without starch was found to be about 1.786, which was increased to about 1.794 after the addition of starch. These measured specific gravities fall in the desirable slip-casting range between 1.7 and 1.8 for clay-based mixture.

3.2. Gel strength and heating time of starch-loaded ceramic green body

The gel strength of consolidated slurry should be determined on defect-free samples. In the present study, a trial and error method was initially conducted to determine the best microwave pressure heating conditions that present no boiling of ceramic slurry as the major causes of defects. To determine the heating temperature, the reported gelatinization temperature between 50 °C and 80 °C was the basis [19,20]. However, a microwave pressure heating at 80 °C and power level of 30% causes severe drying and cracking of samples, and thus the heating temperature was lowered. It was determined that any heating above 60 °C at the same power level caused undesirable drying and cracking of the samples. These defects could be attributed to the rapid conversion of water to steam within the sample that built up internal pressure leading to unwanted cracks. Further lowering of heating temperature below 50 °C resulted in ungelled slurry at which the characteristics of fluid slurry remained. Hence, a desirable heating temperature was determined to be between 50 °C and 60 °C, which is high enough to solidify the slurry into the desirable green body.

It was then informative to study the heating of the small amount of ceramic slurry at target temperatures of 50 °C and 60 °C under different microwave power levels of 10%, 30%, and 50%. The results can be observed on Fig. 3 showing the trend of microwave heating that is reasonably linear for any power level used. The increasing power level introduced an increasing amount of microwave energy available to increase the rate of heating as evidence of increasing slopes of 3.10 °C/min, 6.92 °C/min and 13.21 °C/min. At the same target temperature of 60 °C, these heating rates correspond to the total heating time of 10.35 min, 4.50 min and 2.52 min, respectively. The effect of varying microwave power level and temperature on gel strength of the consolidated green bodies is shown in Fig. 4. It can be observed that, for both target temperatures, the gel strength of the consolidated green body increases with increasing percentage of power level and temperature. This phenomenon could be attributed to the effective disruption of the intermolecular bonding of the crystalline structure of starch under the increasing power level of microwave irradiation. The increasing power level at short exposure time might provide an increasing degree of transformation of the crystalline to an amorphous structure of starch, leading to a better gelatinization of starch granules. However, the reduction of the target temperature to 50 °C at any microwave power levels presented the weaker gel strength due to insufficient microwave energy available to effect gelatinization of the starch granules. It is, therefore, reasonable to decide that the heating time of 2.52 min and target temperature of 60 °C at 50% power level demonstrated the best characteristics of the solidified green body.

A comparative evaluation for the gel strengths of the green bodies was made between samples heated by both heating methods as presented in Table 2. As listed, the gel strength of conventionally heated sample exhibited higher value than that of the sample heated in the microwave pressure cooker.

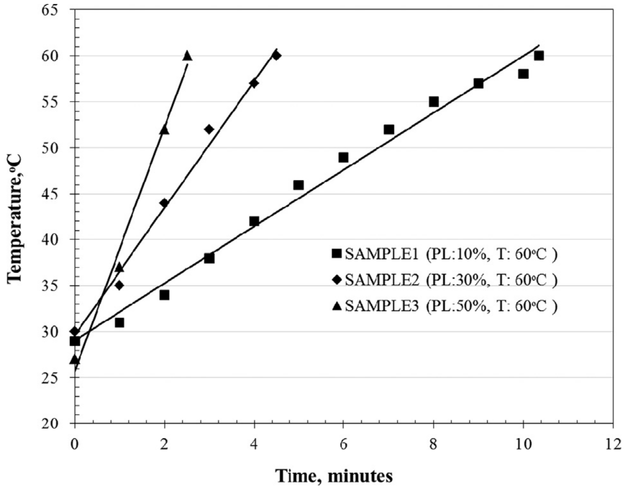

Fig. 3. Heating profile of a small mass of starch-loaded ceramic slurry (about 25 g) at different power levels.

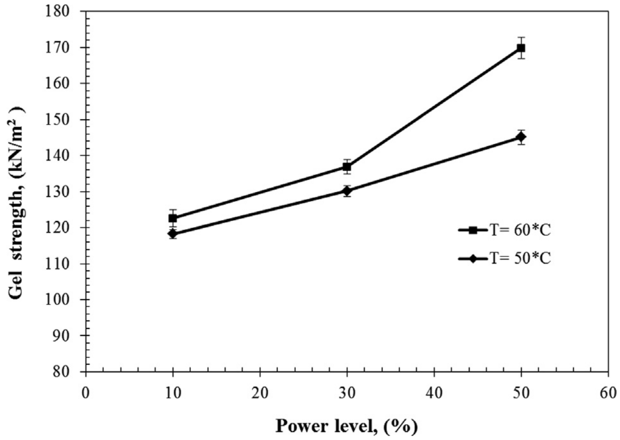

Fig. 4. Trend of gel strength of the consolidated ceramic slurry loaded with potato starch subjected to different power level and temperature.

Table 2
The average gel strength of solidified slurry loaded with potato starch subjected to both heating conditions.

Conventional heating (in the Oven)			**Microwave pressure cooking at 50% power level**		
Temperature (°C)	Gel strength (kN/m^2)	Heating time (min)	Temperature (°C)	Gel strength (kN/m^2)	Heating time (min)
80	207.35 ± 8.94	180	60	169.82 ± 6.03	2.52

A possible reason for this is due to the long heating time that allows proper swelling of starch particles, leading to higher swelling volume for better starch gelatinization. The large swollen granules result in polymeric networks that contribute to firm starch gel [27]. In the case of microwave pressure-cooked sample, the lower gel strength may be due to the simultaneous action of pressurized steam and microwave energy. The gelatinization of starch takes place in a very short time as it is facilitated by the microwave vibration of absorbed water molecules that are trapped within the polymeric constituents [28]. The friction of the vibrating molecules as they couple with microwaves collapses the large molecular units of starch into smaller units that are randomly disoriented or amorphous [29]. Under the pressurized environment, the disrupted polymer units become more mobile and facilitate rearrangement or formation of new bonds among molecules at lower temperatures. These results in the formation of an array of new molecular rearrangements connected with bonds having different stabilities during the structural re-ordering process [30]. This mechanism results in a smaller swelling volume of starch granules, which leads to the reduction of gel strength due to hampered network formation of swollen granules. Nevertheless, the strength of the microwave pressure cooked sample is tolerable and strong enough for handling purposes. This reduction in the gel strength of microwave pressure cooked sample is commensurable with the very short heating time for consolidating ceramic slurry.

3.3. Physical characteristics of the dried and fired ceramic bodies

The physical properties of the ceramic green bodies dried in the convection oven at 110 °C are listed in Table 3. The dried shrinkage of conventionally heated body is lower than the microwave pressure-cooked body. This is because of the fact that the conventionally heated ceramic body was solidified with highly swollen granules that leave large voids while drying, which counteracted the drying shrinkage. For microwave pressure-cooked body, the higher drying shrinkage is attributed to more compact particles bound by less swollen starch granules. While drying, there were only fewer voids that counteracted the shrinking body, resulting in the increase of the drying shrinkage. The measured bulk porosities corroborated with the observed drying shrinkage of the samples heated by both methods. The dried bulk porosity of the conventionally heated sample is significantly higher than that of the sample heated in the microwave pressure cooker. Moreover, the diametral compression strength is also lower for conventionally formed sample as compared with the sample heated in the microwave pressure cooker. This is primarily attributed to higher void spaces as stress concentrators in the conventionally formed sample.

The dried ceramic samples consolidated by both heating methods were fired at 1200 °C, and the measured physical properties were shown in Table 4. It can be observed that the fired shrinkage of microwave pressure-cooked ceramic sample remains higher as compared with the fired ceramic sample heated conventionally. This is because of the fact that more compact particles provide high contact area for particle sintering [31], which results in higher fired shrinkage. This finding is supported with the lower fired bulk porosity of the microwave pressure-cooked sample as compared with the conventional counterpart. Correspondingly, the diametral compression strength of the fired sample consolidated inside the microwave pressure-cooked is higher than that of the conventional counterpart. The weaker strength for the conventional sample is attributed to the higher bulk porosity that provides a higher concentration of weak points.

3.4. Microstructure of fired red clay-based ceramics

It was shown above that the potato-starch loaded ceramic samples shaped inside the microwave pressure cooker

Table 3
Physical characteristics of the oven dried ceramic bodies at 110 °C.

Physical properties	**Heating conditions**	
	Conventionally formed	**Microwave pressure cooked**
Dried shrinkage (%)	11.63 ± 0.45	13.27 ± 0.65
Apparent porosity (%)	25.28 ± 2.13	20.85 ± 1.61
Compressive strength (MPa)	5.28 ± 0.55	6.40 ± 0.22

Table 4
Physical properties of fired ceramic bodies sintered at 1200 °C.

Physical properties	**Fired ceramic bodies**	
	Conventionally formed	**Microwave pressure cooked**
Fired shrinkage (%)	19.28 ± 0.61	20.38 ± 0.10
Apparent porosity (%)	33.79 ± 1.60	29.88 ± 0.31
Compressive strength (MPa)	52.00 ± 3.23	70.74 ± 11.30

exhibited better physical properties than that of the conventional sample. This can be related to the difference in porous characteristics of the two samples consolidated by the two heating methods as shown in Fig. 5. Both micrographs show irregular voids with different sizes due to empty spaces left by the starch particles during burnout [4,32]. It can also be observed on the micrograph the interconnecting solid networks known as the cell struts. These solid networks contribute to the mechanical strength of the ceramic bodies, whereas the void spaces account for the observed bulk porosity. Comparing Fig. 5a and b, it is clearly evident that the void spaces are larger and interconnected in the conventional sample as compared with ceramic sample consolidated inside the microwave pressure cooker. This observed microstructure with large pore networks supports the mechanism of network formation for highly swollen starch granules in the conventionally formed body. Moreover, the presence of large pores in the conventional sample explains the reduction of the mechanical strength as the weak points are larger. The smaller void spaces in the microwave pressure-cooked sample confirm the hindered network formation due to smaller swollen starch granules.

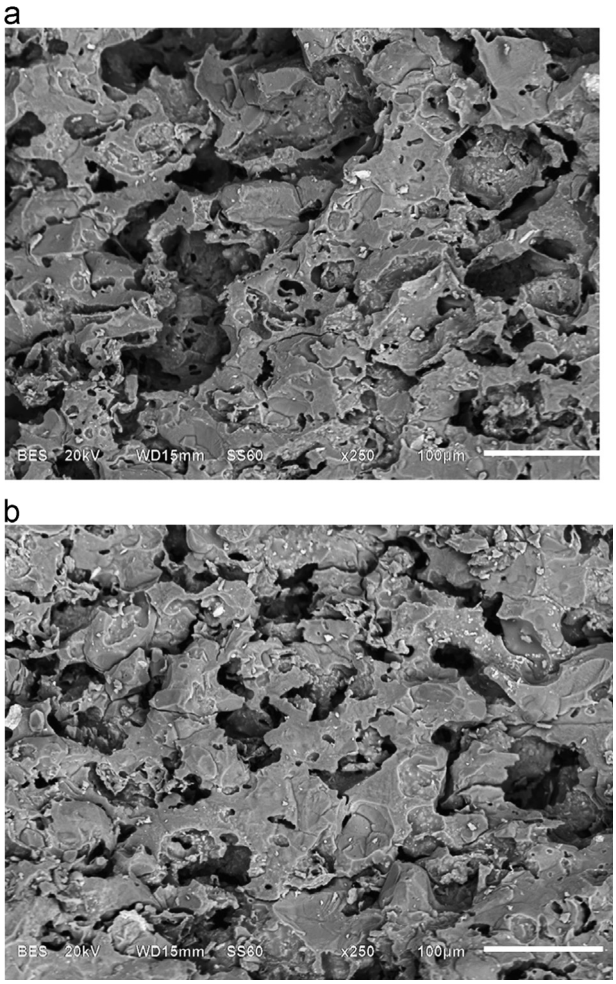

Fig. 5. Scanning electron micrographs of potato starch-loaded ceramic body fired at 1200 °C: (a) micrograph of fired body shaped conventionally, (b) micrograph of fired body under microwave pressure-cooking.

3.5. Shaping of large red clay-based ceramic prototypes

To validate the applicability of microwave pressure cooking to ceramic processing, large ceramic prototypes were consolidated from ceramic slurry loaded with potato starch. The ceramic prototypes were of hollow disk and tile as listed in the first column of Table 5. However, the consolidation of the ceramic slurry in the large amount under the best microwave pressure-heating conditions did not occur leaving behind the slurry in its fluid state inside a tile mold. This suggests that the microwave pressure-cooking time used was insufficient to gelatinize the starch particles for solidifying any large mass of ceramic slurry. This problem might suggest that the exposure time should be increased depending upon the mass of the slurry. The idea of finding the desired exposure time was to employ a ratio and proportion based on the best heating conditions as determined from the small amount of ceramic slurry. Hence, an empirical equation was proposed as the following,

$$t_l = \frac{M_l}{m_o} \times t_o \tag{4}$$

Table 5
Forming conditions for large ceramic prototypes inside the microwave pressure cooker.

Large shapes	**Mass of cast slurry (gm.)**	**Power levels (%)**	**Calculated heating time (min)**	**Time to reach 60 °C, t_a (min)**	**Soaking time, t_b (min)**
Hollow disk	513.36	50	51.11	8.54	42.57
Tile A	353.81	50	35.33	7.45	27.88
Tile B	355.95	70	25.18	5.37	19.81
Tile C	356.34	100	17.64	3.00	14.64

where t_l is the microwave pressure cooking time (minutes) at constant power level of 50% and temperature (60 °C) for large mass of slurry, m_o is the mass of the small sample (25 g), M_l is any large mass of slurry greater than m_o, and t_o is the corresponding microwave pressure cooking time of small sample (2.52 min). Since t_l is always greater than t_o, this suggests that the heating process is expected to involve two steps: (a) heating time (t_a) to reach the target temperature of

60 °C, and (b) soaking time (t_b) that depends on how much time consumed for heating to the target temperature. Therefore, the microwave pressure cooking time, t_l, is the sum of t_a plus t_b.

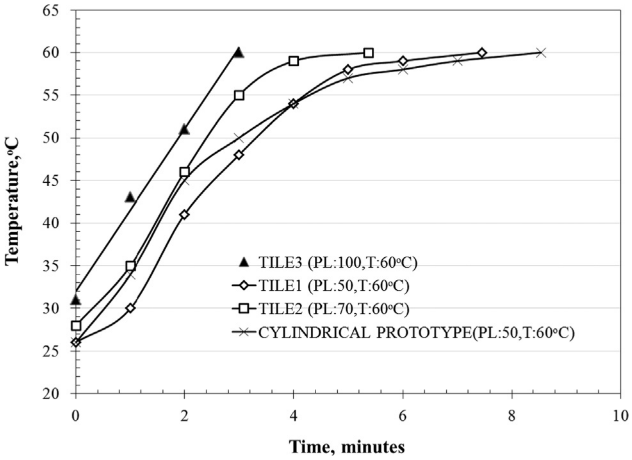

Fig. 6. Heating profile for large masses of ceramic slurry subjected to increasing power levels.

To find t_a, an external sensor slurry of the same mass and composition as the large sample inside the microwave pressure cooker was casted into microwavable plastic (height = 40 mm, and diameter = 40 mm) equipped with a cover to minimize the water evaporation during heating. In this experiment, regardless of the different geometries of the samples inside the microwave pressure cooker, the shape of the container for the external sensor slurry was the same. The temperature rise and time were monitored until the target temperature was reached as shown in Fig. 6. It can be observed that the heating profile is nonlinear for power level less than 100%. This can be related to the penetrating and decaying nature of microwave energy. At 100% power level, the amount of microwave energy penetrated and absorbed by the sample was the highest to allow uniform temperature distribution, which resulted in the linear heating profile. In contrast, the decrease in power level reduced the amount of available microwave energy transmitted. It was then possible to have a greater tendency towards nonuniform temperature distribution from the center of the sample. In other words, the temperature is decreasing towards the surface [33] and contributing to the nonlinear heating. Nevertheless, it is worthwhile to point out that the heating

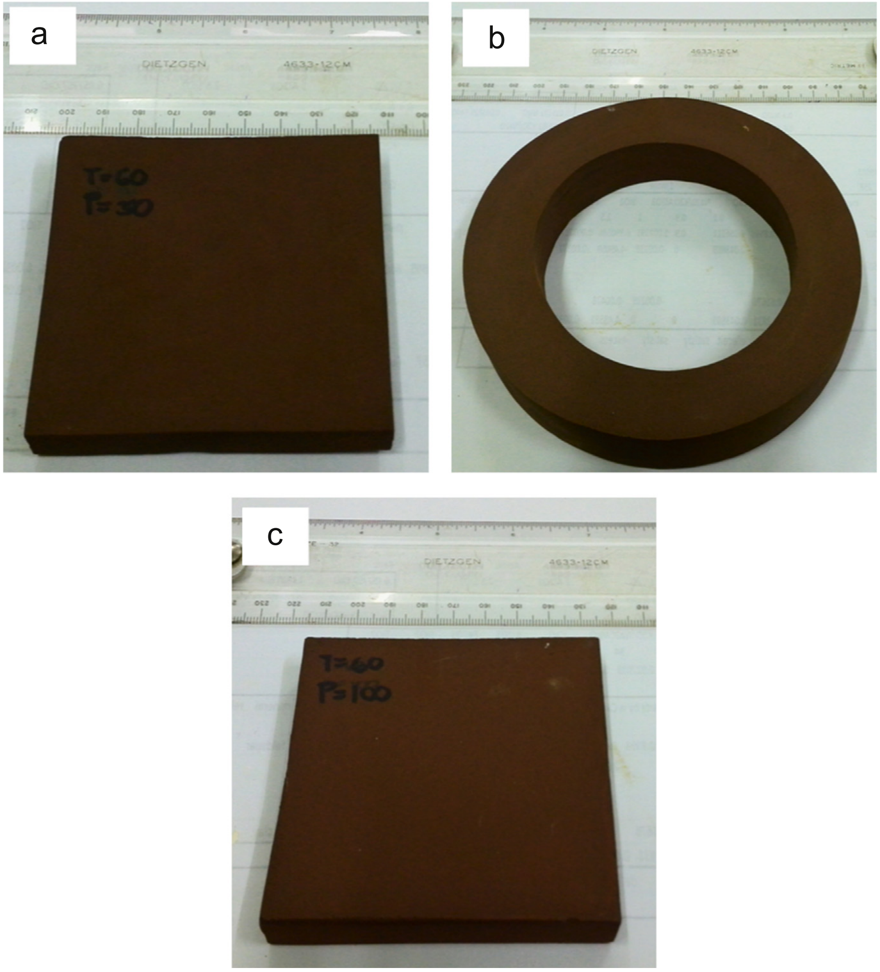

Fig. 7. Large red clay-based ceramic prototypes of different shapes fired at 1200 °C: (a) Tile consolidated at 50% power level and 60 °C, (b) hollow disk consolidated at 70% power level and 60 °C, and (c) Tile consolidated at 100% power level and 60 °C. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

profile did not alter the solidification performance of the ceramic slurry since the heating is dependent only on the total time to reach the target temperature. The soaking or holding time at the target temperature could then be determined as $t_b = t_l - t_a$. This formula worked very well for a large amount of ceramic slurry solidified in different shapes such as hollow disk and tile shown in Table 5 and Fig. 7a and b.

It was observed that relying only on one microwave power level made the microwave exposure time to relatively increase when using a very large mass of ceramic slurry. This will probably result in a reduction of the production volume and undesirable formation of forming defects. To address these issues, the power level should be increased above 50% for the same large mass of the ceramic slurry. The microwave exposure time could then be determined from the power concept in Physics. The power is defined as the rate of doing work upon an object at which the power equals work over time. To determine the microwave exposure time, the work done at any power levels was assumed to be equal. Hence, the following empirical equation has been proposed as

$$t_2 = \frac{P_1}{P_2} \times t_1 \tag{5}$$

where t_2 is the microwave pressure cooking time at desired power level, P_2, to reach the target temperature (60 °C), P_1 is the known power level to reach the same target temperature and mass of slurry at the known microwave exposure time of t_1. The computed microwave pressure cooking time is presented in Table 5 for increasing percentage of microwave power level. Since the computed heating time was expected to be longer than the heating time to reach the target temperature, the heating time was also involved two steps as described above. It can also be observed that, at the same large mass of slurry, the microwave pressure-cooking time significantly decreases. The reduction of the cooking time indicated a significant time saving during shaping. It is, therefore, deduced that the microwave pressure cooking time is directly proportional to the mass of the ceramic slurry and inversely proportional to the desired power level.

The calculated microwave pressure-cooking time was successfully applied to the actual ceramic slurry consolidation for the large ceramic prototypes. The large shaped green ceramic bodies were free of cracks while drying at room conditions without sensitive drying control. This observation suggested that a very uniform starch gelatinization occurred within the solidified ceramic slurry. As physical evidence, the dried prototypes survived without discernable sintering cracks after firing at 1200 °C as shown in Fig. 7. Consequently, these findings suggest that the microwave pressure-cooking technology is highly applicable to the actual fabrication of ceramic materials with different shapes and geometries.

4. Conclusion

A simultaneous application of pressure cooking and microwave irradiation to consolidate starch-loaded ceramic slurry presented a rapid shaping technique. The small amount of ceramic slurry was successfully solidified with microwave pressure cooking conditions of 50% power level and target temperature of 60 °C at a very short heating time of 2.52 min. Based on these conditions, large shaped ceramic prototypes of different shapes were successfully formed using proposed empirical equations to determine a desired heating time at any power levels. The dried and fired physical properties of the microwave pressure-cooked sample exhibited better results as compared with the conventional counterpart. The poor physical properties of the conventional sample were due to the presence of larger void spaces as confirmed by microstructural investigation.

Acknowledgements

The MSU-Iligan Institute of Technology (MSU-IIT) is gratefully acknowledged for allowing us to publish the content of this study. Thanks are also due to Mr. Carl Rhamel M. Adavan our laboratory technician, who was very patient and committed in helping us during our laboratory activities.

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