

Microwave assisted sintering of high voltage porcelain material and its characterization

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The microwave assisted sintering of a commercial grade quartz body porcelain composition was carried out in air at a frequency of 2.45 GHz. The sintered samples were characterized by different analytical techniques. The properties of the microwave- sintered products were compared with those obtained by a commercially- processed method in a partial reducing atmosphere. It was observed that porcelain samples can be fast fired in a microwave furnace to give better mechanical strength. The required phase contents can be developed during fast microwave firing. The electrical properties such as volume resistivity, breakdown voltage, dielectric strength and dissipation factor are reported which are essential for high voltage applications. The comprehensive results obtained in this study indicate that significant savings in energy are possible in processing a complicated triaxial porcelain material.

Key words: Porcelain, Characterization, Microwave processing, Strength, Electrical Properties, Energy savings.

Introduction

Electro -porcelain insulators are critical components and defect sensitive because of their use in high voltage applications. The insulators are generally fired in the range of 1200 °C to 1300°C with a cycle time exceeding 100 h [1]. Nearly 70% of the energy used in ceramics manufacturing is accounted for by the firing process, with oil and gas as the predominant fuels. The strength of electro -porcelain materials is dependent on the homogenous glassy matrix [2] and thus a uniform heating is necessary throughout the component in order to achieve this. The energy efficient microwave processing of these materials can reduce the cycle time drastically by a volumetric heating mechanism resulting in better properties of the material [3-7]. The microwave processing of porcelain is rather complicated since porcelain as such is a complex material and the fast heating rate by microwave processing poses a further challenge in achieving the proper phase formation. The reactions occurring during firing need to be properly controlled in order to achieve the best properties. It was observed that to achieve the same degree of water absorption, the temperature applied in microwave hybrid sintering needs to be slightly higher (approximately 20 °C) than those used in conventional firing [3]. The study by Mizuno *et al.*[4], using 84 GHz microwave heating highlighted that it is very important to consider the temperature difference between the liquid amorphous phase and the solid crystalline phases during microwave

processing of porcelain bodies. However, the effect of microwave radiation at the widely used 2.45 GHz frequency on the sintering of these materials needs to be studied, since the 84 GHz frequency is not used commercially to process materials. Further, in this study, the porcelain after firing was porous and thus can not be used in high voltage applications. The additional need for a reducing atmosphere during processing as practiced by few industries worldwide complicates the overall processing. Further, the earlier experiments have not used commercial compositions, but rather concentrated on laboratory level experiments, which may pose problem in scale-up in future. These limitations have been overcome in our study by employing a 6 kW microwave furnace at the commonly used 2.45 GHz frequency and by obtaining pore-free porcelain bodies which can be subsequently tested for property measurements.

A systematic study has thus been carried out on the fast firing of porcelain components in air. A commercial quartz body porcelain composition was used for microwave-assisted sintering in air. The processed materials have been characterized by different analytical techniques to confirm the retention of properties of porcelain insulators after undergoing a very fast firing cycle. Since the porcelain materials are required to operate in high voltage applications, the results of electrical properties have been measured for the first time for microwave- processed bodies and compared with those obtained in conventionally- processed samples.

Experimental Procedure

A commercial clay-quartz-feldspar system was used in this study. The typical composition is feldspar: 11-13%, washed clay: 20-25%, raw clay: 25-35%, quartz: 20-25%, pyrophyllite: 8-10% and feldspar: 5-8%. All these raw

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materials were sourced from India. The average particle size of the feldspar was in the range of 100-110 μm and that of the quartz in the range of 50-55 μm . The required quantity of powder raw materials was milled in a ball mill for 24 h using river pebbles as a grinding media and water as the solvent. The milled slurry was passed through a filter pressing unit to form a cake. The cake was used in extruding rods of diameter 12 mm and length of 500 mm. The room temperature dried rods were dried in an aluminum channel and cut to lengths of 150 mm. The rods were further dried in a microwave furnace, [8] to achieve a moisture level of $\sim 0.5\%$. The fully-dried rods were suspended through fiber insulation boards into an alumina-silicon carbide casket with additional silicon carbide for susceping microwaves at room temperature. The typical assembly for firing these components is depicted in Fig. 1. The temperature was measured by an infrared pyrometer with an emissivity of 0.6 by focusing on a green disc of diameter 50 mm of the same composition, which was fabricated by extrusion using a different die. The whole assembly with 12 samples was kept in the uniform zone of the 6 kW microwave cavity (M/s Cober Inc., USA) and sintered using a heating cycle as depicted in Fig. 2. In a typical conventional cycle, the samples are loaded into the cars and fired in a tunnel kiln alongwith the regular production samples . However, in microwave firing, the exclusive firing was used in an insulated casket alongwith the required susceptors. The susceptors interact with the microwave radiation initially and increase temperature in the assembly so as to allow the porcelain

samples to interact with the microwaves after the critical temperature is reached. Fig. 2 also compares this cycle with a typical industrial heating cycle for these porcelain materials. A maximum temperature of 1260 $^{\circ}\text{C}$ in air was used in this study, which is similar to that used in the commercial process. Further, a low temperature of 1210 $^{\circ}\text{C}$ was also used to compare the progress in sintering. The sintered samples were characterized by different analytical techniques namely, density and porosity by a water displacement method, phase identification by X-ray diffraction, the microstructure by a scanning electron microscope (LEICA) after polishing and etching with HF,

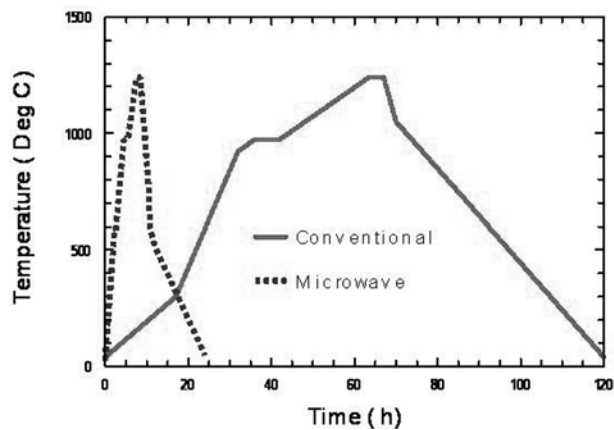


Fig. 2. A comparison of the thermal cycle during the conventional heating and Microwave heating of a quartz based electro porcelain material.

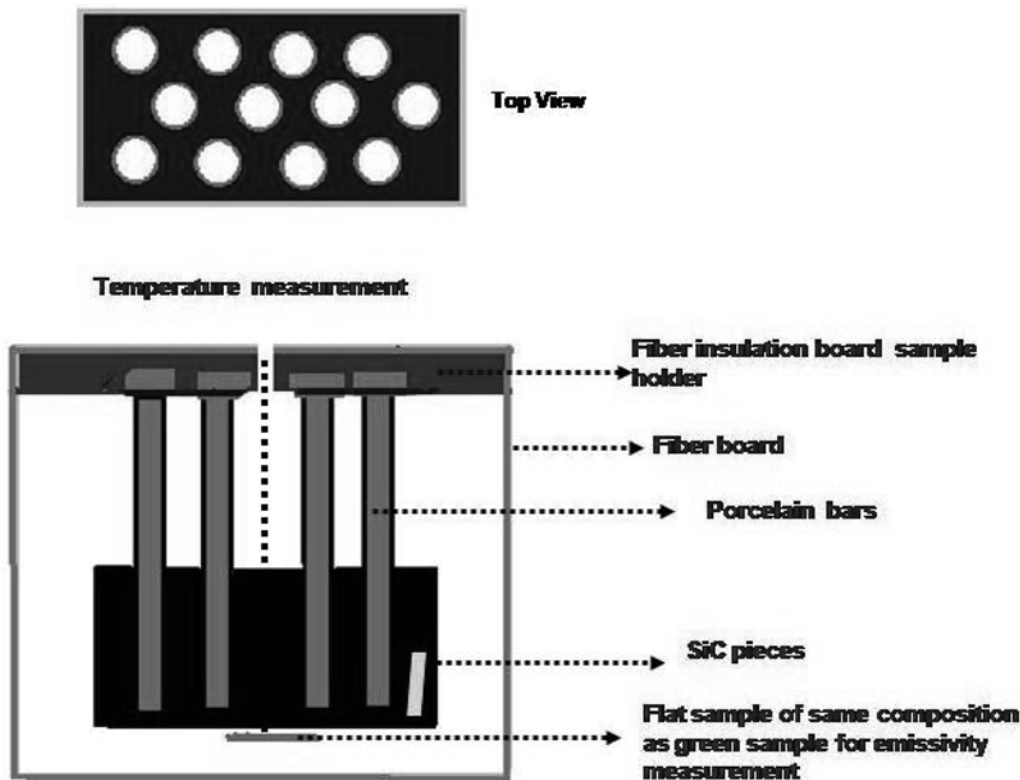


Fig. 1. Typical assembly for microwave-assisted firing of porcelain components in air.

quantitative phase determination by an internal standard method using rutile as an internal standard and analyzing the data by Rietveld analysis. The three point bend strength of at least 20 samples was measured on the sintered bars with a cross head speed of 0.5 mm /minute. The Young's modulus of five samples was measured following the ASTM standard C 1259 (1998) and E1876 (2001) using a dynamic elastic properties analyzer (M/s Jagdish electronics, Bangalore, India). The thermal expansion was measured on a 50 mm rod of 10 mm dia using a Harrop dilatometer from room temperature to 1000 °C. Further, the electrical properties such as dielectric constant, dielectric loss and dielectric strength of the sintered disc of ~50 mm diameter and 3 mm thick (< 0.5% apparent porosity and both sides with flat surfaces) were measured following the IEC-60672 standard. The data were compared with those obtained on conventionally- fired samples. The conventionally sintered samples means that similar shaped articles were sintered using the typical heating cycle depicted in Fig. 2 using a partial reducing atmosphere.

Results and Discussion

The experimental results were analyzed based on the parameters of uniform firing of the samples, the effect of peak temperature, the amount of amorphous phase formation during microwave heating and microstructural and electrical characterization.

It was observed that, the samples fired at 1210 °C with a heating cycle of 6 h and a soaking period of 30 minutes were not fully sintered and the difference in porosity content was very high from top to bottom of the sample (Fig. 3). The cycle time and temperature are the two most important parameters to achieve a uniform fullysintered body. Further, a few more experiments were carried out by varying the heating cycle and the peak temperature keeping the soaking period of 30 minutes constant in all the experiments. It was observed that, a heating cycle of 8 h and a peak temperature of 1240 °C is ideal, reducing the porosity variation throughout the sample and simultaneously densifying the component uniformly (Fig. 3). This series of experiments have confirmed that microwave heating of these compositions does not require a higher temperature [2] than that used in the conventional process. As the figure suggests, the reduction in the processing cycle during microwave treatment from 120 h in conventional to 8 h in microwave translates to a reduction in energy requirement and the cost of processing.

The peak temperature, the duration at the peak temperature and the heating cycle are three important parameters for proper densification of samples. The peak temperature plays a very important role in the final densification of the product and in imparting properties to the final product. In these experiments, the peak temperature was varied from 1220 °C to 1240 °C to 1260 °C keeping the soaking duration constant at 30 minutes. All other experimental conditions were kept constant. It was observed that the bending strength of the samples varied drastically from 85 MPa for firing at

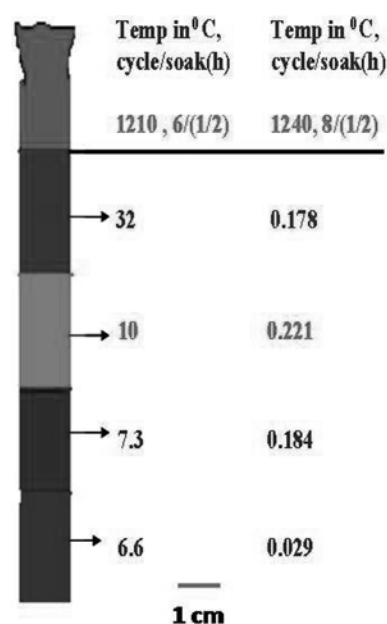


Fig. 3. A comparison of the porosity content along the length of a high L/D sample which was sintered at two different temperatures in a microwave furnace.

1220 °C to 118 MPa for firing at 1240 °C to 107 MPa for firing at 1260 °C. This result indicated that the peak temperature of 1240 °C is ideal in imparting maximum strength in this porcelain composition. It is pertinent to mention that the same peak temperature is commonly used in the conventional commercial process; however, the strength of the microwave sintered body is 15 % higher than that of its conventional counterpart of 103 MPa. Further, this result has demonstrated that a higher sintering temperature than that used in commercial process is not suitable for achieving better properties in these materials.

The amorphous phase content in porcelain plays an important role in optimizing the properties. The higher peak temperature and higher heating time are directly proportional to greater glass phase formation which translates to lower mechanical properties. Besides the amorphous content, the amount of crystalline phases such as quartz, mullite and cristobalite influence the properties of porcelain material to some extent. It is always desirable to possess a lower amorphous content and higher crystalline content to obtain better properties in quartz- based porcelain materials. This is limited in conventional processing due to the long heating time. However, the present method of microwave processing is a very fast process and it is expected to contribute to the phase formation significantly.

The qualitative X-ray diffraction analysis is compared in Fig. 4. The figure indicate that microwave sintered samples possess the same XRD pattern as that obtained by the conventional process. A quantitative X-ray diffraction analysis was carried out on the porcelain powder samples which were obtained from grinding the sintered rods using both the processing methods. The powders were mixed with 10% titanium oxide (rutile) as an internal

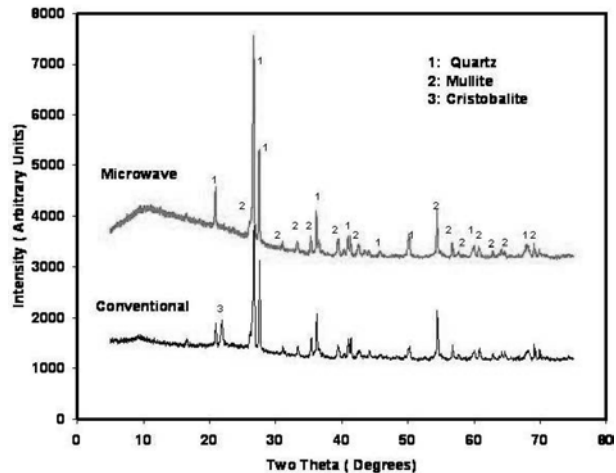


Fig. 4. A comparison of the phase formation during conventional heating and microwave heating of a quartz body porcelain material.

standard and the quantification of each phase was analyzed using the standard Rietveld method. The results are shown in the Table 1. The quantified XRD results on microwave sintered rods were compared with those of commercial-processed rod samples. It was observed that the amorphous phase content was lower and the crystalline phase content was higher in the present microwave-sintered samples in contrast to that processed by the commercial process. This result is supportive of higher mechanical properties

Table 1. compositionPhase content of Porcelain body after processing both by conventional and microwave methods (as calculated by Rietveld analysis of X-ray diffraction patterns in vol. %)

rocessing condition	Quartz	Mullite	Cristobalite	Amorphous Phase (%)
Microwave heating, heating time 7.5 hr, peak temp. :1240 °C, soaking time: 0.5 h	43.8	1.3	0	54.9
Typical commercial process with heating time of ~70 h, peak temp.:1240 °C and soaking time: ~2 h	18.4	12.4	8.6	61.6

in these materials as described earlier. The higher quartz content in microwave- processed samples contributes significantly to the mechanical properties. The significant amount of the cristobalite phase as present in the conventionally- processed samples was not found in microwave-processed samples. The results indicated that the microwave process is ideal for the processing of porcelain materials.

The Scanning electron micrographs of the microwave fired samples from the present process were compared with that in commercial process. Undissolved quartz particles can be seen in the microstructures (Fig. 5(a) & (b)). The

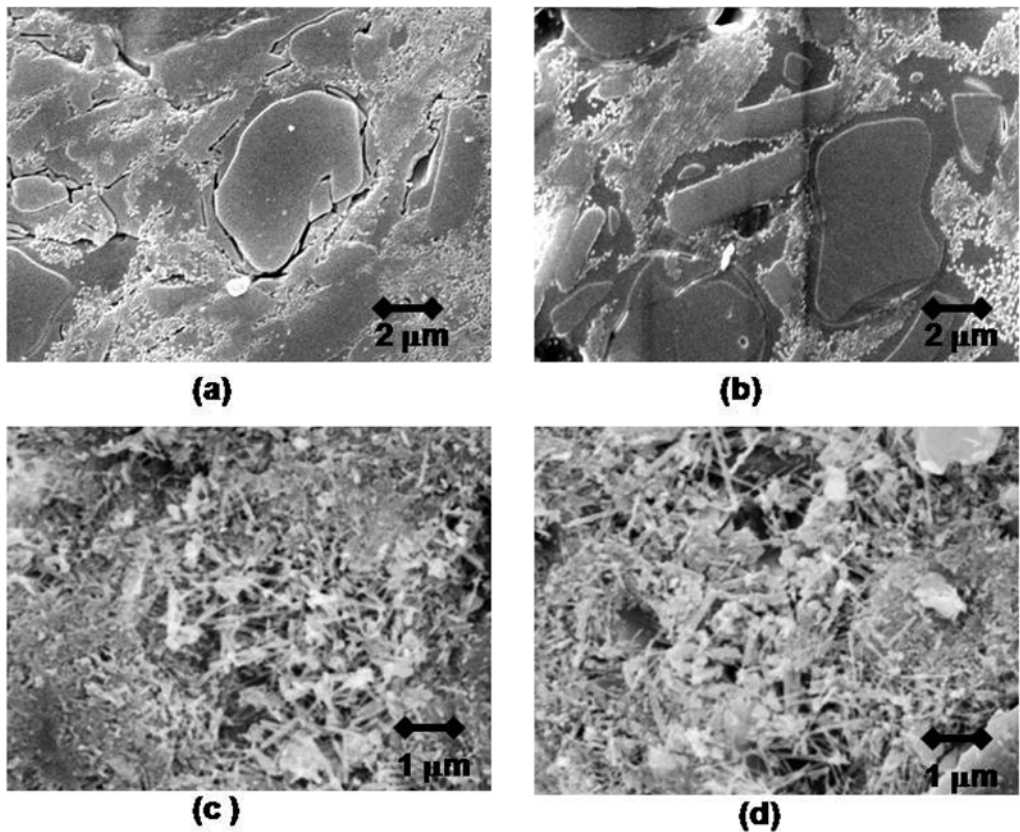


Fig. 5. Scanning electron micrographs of quartz based porcelain bodies : a & c : conventional sintered and b & d : microwave sintered, Marker size : 2 µm for 'a' and 'b' and 1 µm for 'c' and 'd'.

most important observation is that a stress concentration around quartz particles was not observed in the microwave-processed sample compared to the resulting peripheral cracks in conventionally -processed samples. This is a significant observation which may explain the higher strength in the microwave- processed materials. This result is significant, since in an earlier recent report, [2] cracks were reported around quartz particles in the microwave-processed porcelain material. Further, there were no cracks or defects noticed in the microstructure. Secondary mullite needles can be seen in both microstructures (Fig. 5(c) & (d)). The size of the needles has not varied drastically in spite of the large difference in heating cycle. A similar microstructure in both the processes indicates that a reducing atmosphere in the conventional processing may not be essential in developing the right microstructure during fast fired microwave processing. This observation has a serious implication in modifying the processing cycle of quartz-based porcelain on a commercial scale.

The electrical properties of the porcelain body both fired by conventional processing and microwave processing were measured following the IEC 60672-2 (1999) standard. The properties measured and compared in this study include tan delta, dielectric constant, volume resistivity, and break down voltage (Table 2). It was noted that the tan delta, dielectric constant and volume resistivity were increased with an increase in the temperature. Similar behaviour in the results of conventionally- sintered samples was also observed. The results are also compared with those of equivalent grade imported porcelain sample from Siemens, Germany [9] (Table 3). It was observed from Table 3 that, the electrical properties did not deteriorate due to very fast microwave heating. Further, the data obtained by the new improved method are either comparable or in some cases better than those obtained by the international manufacturer. This result is new ,not previously reported in the literature and confirms that microwave firing of porcelain is ideal on an industrial scale to achieve the correct electrical properties required for the product. Most importantly, the fast microwave processing of porcelain is not detrimental for its use in high voltage applications.

Table 4 summarizes the best results obtained on the

Table 3. Comparison of the electrical properties of MW-processed porcelain insulator components with those of conventionally and commercially processed typical Indian insulators of the same composition and an equivalent commercial grade of porcelain insulator body

Electrical Properties	Microwave processed (this study)	Conventionally processed (this study)	C112 of Siemens ⁹
Tan Delta @ 100 °C	0.116	0.093	0.093
Dielectric Constant @ RT	6.0	5.5	5-6
Volume Resistivity (Ω -m @ 200 °C)	3.4×10^{09}	1.34×10^{09}	$\sim 10^{06}$
Dielectric strength (kV/mm) @ RT	20	18	20

Table 4. Properties of conventional- and microwave-sintered cylindrical rods of quartz- based porcelain insulator components

Properties	Conventional	Microwave (this study)
Sintering Temperature (°C)	1240	1240
Heating cycle Time (h)	65-70	8-10
Bulk Density (g/cc)	2.35	2.38
Three Point bend strength (MPa)	103	118
Modulus of elasticity (GPa)	79	82
Thermal expansion coeff. ($/K \times 10^{-6}$) (RT- 1000 °C)	5.25	5.83
Amorphous phase content (%)	61.6	54.9
Dielectric Constant @ RT	5.5	6.0
Dielectric strength at RT (kV/mm)	18	20

microwave-sintered quartz -based electro-porcelain components and compares them with the same obtained from the samples sintered by the conventional method. Although the peak sintering temperature was kept constant in both cases, the cycle time was drastically reduced in the microwave sintering. It was observed that, fast microwave sintering is suitable for porcelain components as revealed by the higher mechanical properties, higher bulk density and higher dielectric strength.

Conclusions

Microwave-assisted sintering of a quartz body porcelain composition was carried out in air. It was observed that the cycle time could be reduced drastically by the use of microwave energy. The peak sintering temperature can be made the same as used in conventional processing. A better crack free microstructure could be developed with a low amorphous phase content resulting in a higher strength of the fired components. The phase formation was not affected by the fast sintering process as revealed from the X-ray diffraction patterns. The electrical characterization of the materials was carried out and it was noted that there was no negative effect of microwave

Table 2. Electrical properties of quartz- based porcelain body processed by microwave heating.

Electrical Properties	Temperature (°C)				
	30	60	100	150	200
Tan Delta	0.038	0.047	0.116	NA	NA
Dielectric constant	6.0	6.5	7.3	NA	NA
Volume Resistivity (Ω -m)	8.0×10^{13}	1.2×10^{13}	2.9×10^{11}	3.6×10^{10}	3.4×10^9
Dielectric Strength (kV/mm)	20	NA	NA	NA	NA

processing on the electrical properties of porcelain materials. The process can be used in the large scale manufacturing of these components.

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