THE POLARIZING EFFECTS IN SINTERED KAOLIN

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ABSTRACT

The polarizing effects in sintered kaolin samples were analyzed in terms of the sample density (or porosity) using direct current (dc) techniques. More porous samples exhibited higher polarizing effects than the less porous ones. The conduction carriers in kaolin samples at room temperature were found to be mainly electrons, with ionic conduction being enhanced by the moisture content within the pores. The influence of the uniaxial pressure and particle-size on the compacted and sintered density of the ceramic have been studied, and a density – pressure relationship for before- and after-sintering conditions obtained.

INTRODUCTION

Ceramics have been known to mankind for thousands of years, and have been used in construction materials. In many applications, ceramics have properties superior to some competing materials such as metal alloys, polymers, and composites. This is due, in a large part, to modern methods of preparation. With detailed knowledge of microstructure and associated materials properties, ceramics are designed for such diverse uses as high temperature turbines, engines, artificial joints (for implants) and tools. Ceramics are widely used in the electrical industry because of their high strength and resistivity.

The performance of most ceramics is sensitive to preparation conditions, and could be totally or partially responsible for many discrepancies among the reported results (Salib & Vipulanandan 1990). The preparation conditions include the particle size, the compaction pressure, the sintering temperature and time. In the compaction-sintering process, various densities of the compacts have been obtained by varying the uniaxial compaction pressures up to 350 MPa (Salib & Vipulanandan 1990, Hill *et al.* 1995).

Measurements of high resistivities at room temperature is reported to be difficult due to the polarization and re-ordering of defects and impurities which result in an initially high current that takes a long time to fall to a steady state (Moulson & Herbert 1990). Tallan *et al.* (1966) observed these effects under dc conduction measurements as a time-dependence of the current following the application of the field and an "after-effect" current in the opposite direction after the removal of the field. In investigating the charge carriers in a porous ceramic sensor, Yeh *et al.* (1990) plotted charging and discharging curves and concluded that the carriers were both ions and electrons, with the dominant conduction carriers being ions.

The raw materials used in the manufacture of low- and high-tension electrical porcelain are clays, and it is to this purpose that this research is dedicated. The kaolin used in this study was obtained from the Bundle Industries, in Kampala. This industry gets its supply of kaolin from a dig-out in a quarry and processes it. Odegard (1995) measured the dielectric and mechanical properties of samples from this kaolin and proposed that it could be used for the manufacture of ceramic insulators. In the process of measuring the resistivities of our samples, a large initial current was observed which slowly reduced to a stabilized value. The purpose of this research was therefore to investigate this current and its relationship to samples of varying densities.

METHODS

The kaolin powder was wet sieved into different particle size ranges and dried. The dried powders were then compacted into disks of 25 mm diameter and 2-3 mm thickness by uniaxially pressing the powder from both ends in a steel die, using a Hydraulic Laboratory Manual Press. To obtain samples of varying densities, disks were prepared by: (1) varying the uniaxial compaction pressure, and (2) varying the particle-size ranges of the kaolin powder. The disks were sintered in air to 1200 °C. The density of the dry-compacted green and sintered samples was measured by the geometric method. The dimensions of the green samples were taken while the samples were still in the die to avoid damaging the surfaces. The solid density was obtained by using the pyconometer bottle. For the dc measurements, a three terminal electrode system was used with a guard ring as a third electrode to eliminate errors due to surface conduction. The power was provided by the SF-9586 kV Power Supply, and current measurements were made by the Thurlby 1503-HA Digital Multimeter. For a set voltage, the current was measured after short intervals until it reached a constant value.

RESULTS AND DISCUSSION

Density measurements

The relative density, (∂), expressed as a percentage ratio of the ceramic density to solid density, was plotted as a function of compaction pressure for both the green and the sintered samples (Fig. 1). The sintered density was lower than the green density and this may be attributed to the loss of volatile compounds during sintering. Salib & Vipulanandan (1990) showed that the sintered density was higher than the green density for the polymer-impregnated superconducting ceramics. The increase in density, which they observed, could have been caused by swelling of the polymer molecules. They proposed a power law of the form:

$$\partial = \alpha P_c^{\beta} \tag{1}$$

to describe the variation of relative density ∂ and compaction pressure P_C, where α and β are dependent on the method of processing. In the present study, the power law was also investigated for both the green and sintered samples. The values of α and β are shown in Table 1. β appears to be the same for both the green and sintered samples.

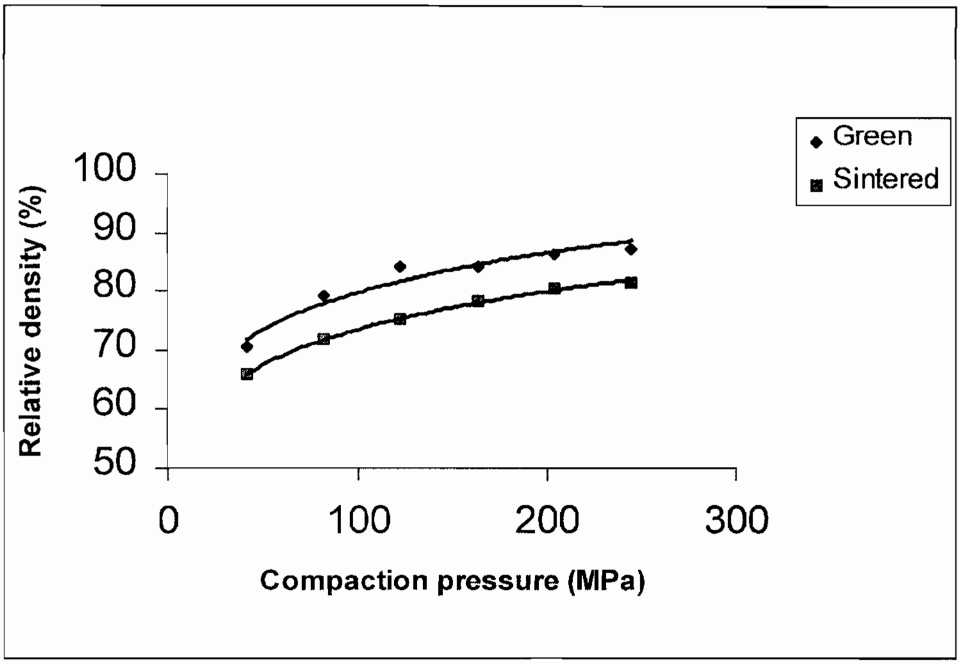

Fig. 1: Relative Density – Compaction Pressure relationship for kaolin powder obtained in Kampala, Uganda

Table 1: Correlation between green and sintered samples' results

Sample	α	β	Correlation, r
Green	46.4	0.12	0.948
Sintered	42.1	0.12	0.998

There was an initial increase in the density, which then fell off as the particle sizes increased (Fig. 2). The initial increase in the density is expected since the compaction curve is known to shift to higher densities when there are larger particles in the pressing powder (Salib & Vipulanandan 1990). As the particle sizes increased, the shape of the powder could have had an influence on the packing of the powder, possibly allowing for a less dense structure (D'ujanga 1998).

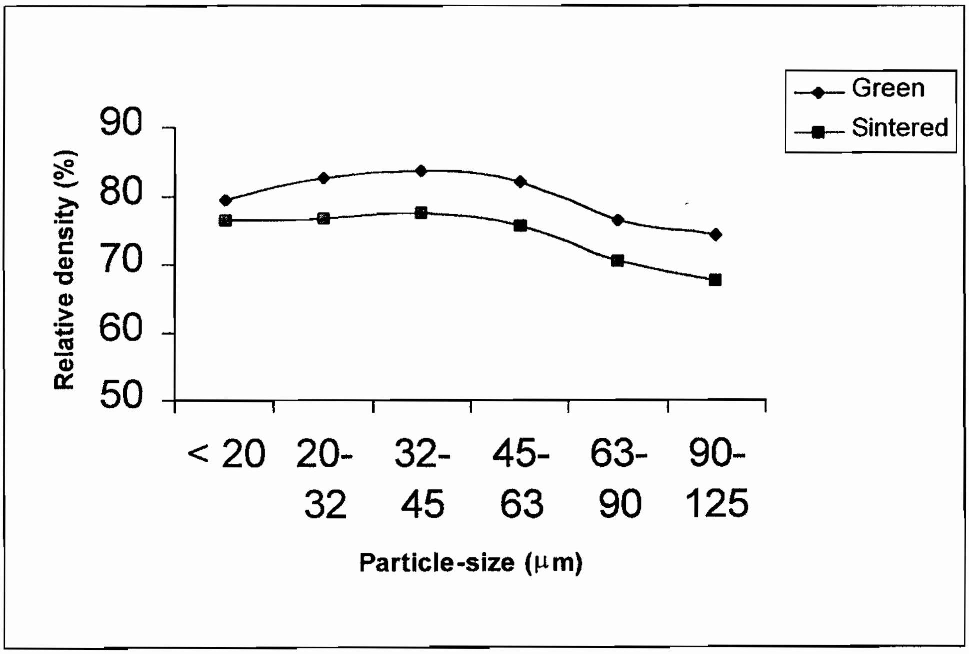

Fig. 2: Relative Density – Particle-size Relationship for kaolin obtained in Kampala, Uganda

dc measurements

When a dc voltage is applied to the sample, one of the following may be observed (Yeh *et al.* 1990) (1) if the conduction carriers are ionic, then the sample is polarized, and the charging current decays exponentially from an initial value $i_c(0)$ to zero in a finite time (2) if the carriers are electrons, the charging current is a constant value; and (3) if the carriers are ions and electrons, then the charging current will decay exponentially from a positive maximum $i_C(0)$ to a steady value i_C. Therefore the current – time characteristics for charging and discharging processes can be used to identify whether the conduction carriers are ions, electrons or both. Fig. 3 shows the charging curves for disks compacted at different compaction pressures, and Fig. 4 shows the charging curves for disks with different particle sizes. It is observed that less polarization occurs in samples in the low particle-size range, and in samples compacted at higher pressures, that is those samples with a high density. When the voltage was removed, the discharging current remained zero, indicating that the conduction carriers were mainly electrons. Hence, unlike in the Yeh *et al.*'s (1990) case where the carriers were mainly ions, our samples exhibited mainly electronic conduction, with a contribution from ionic conduction resulting from the moisture content within the pores.

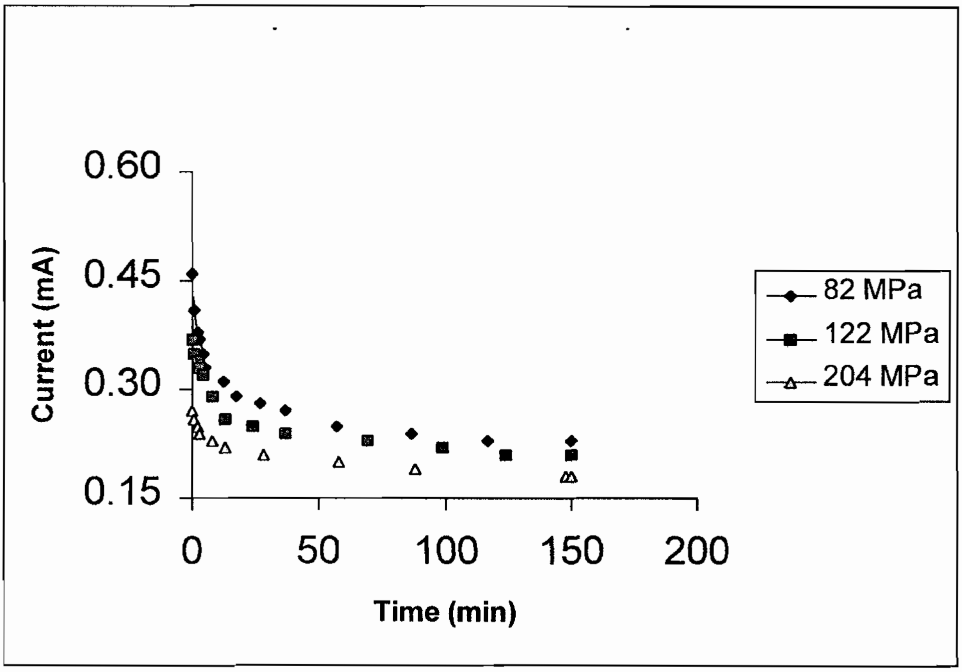

Fig. 3: Charging curves for Kaolin samples with different compaction pressures. Samples collected in Kampala, Uganda

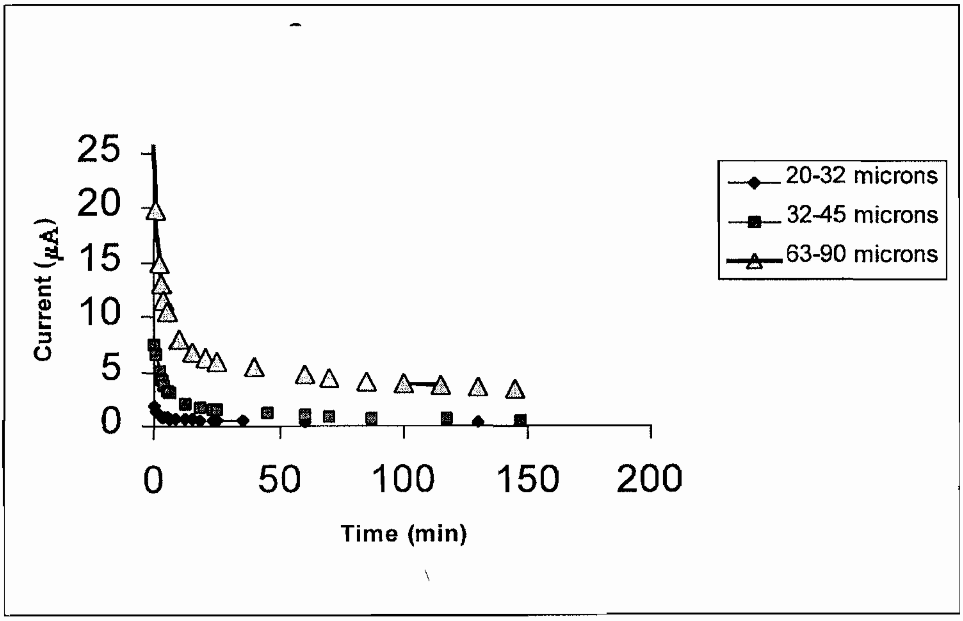

Fig. 4: Charging curves for Kaolin samples with different compaction pressures

The time dependent charging current $i_c(t)$ may be represented by a time varying component $i_i(t)$ contributed by mobile ions and a constant component, i_e, contributed by conduction electrons:

$$i_c(t) = i_i(t) + i_e = i_i(t) + V/R \tag{2}$$

where V is the applied voltage and R, the sample resistance. V/R is a measure of the electron conductivity of the sample and corresponds to i_c when determining the resistivity of the samples. Resistivities for the different samples increased with increasing sample density (i.e. with decreasing porosity) (Tables 2 and 3). The dc results imply that: (1) the more porous samples absorb moisture than the less porous ones; (2) when a voltage is applied, there exists water molecular polarization effects, which is indicated by a higher initial charging current in porous samples; (3)i_c is a measure of the conductivity of the samples, showing that the less porous samples are more resistive; and (4) the curves indicate that the charge carriers are mainly electrons, but ionic conduction is enhanced by the moisture absorbed.

Table 2: Time-dependent charging current in sintered Kaolin compacts for varying powder particle-size

Powder particle-size (μm)	$I_C(0)$ (μA)	$i_C(\infty)$ (μA)	Resistivity, ρ (Ω-m)
20-32	1.79	0.30	1.71×10^8
32-45	7.40	0.59	0.86×10^8
63-90	31.4	3.44	0.14×10^8

Table 3: Time-dependent charging current in sintered kaolin compacts for varying compaction pressures

Compaction pressure (MPa)	$I_C(0)$ (μA)	$i_C(\infty)$ (μA)	Resistivity, ρ (Ω-m)
82	0.46	0.23	1.12×10^8
122	0.37	0.21	1.22×10^8
204	0.27	0.18	1.43×10^8

CONCLUSION

Various densities of the kaolin samples were obtained by varying the uniaxial pressure, and also by using powders of different particle-size ranges. A density-pressure relationship for before- and after-sintering conditions was found to obey a power law proposed by Salib and Vipulanandan (1990). The dc measurements have indicated that the conduction carriers in kaolin samples at room temperature are mainly electrons, with some ionic conduction due to the moisture content within the pores. High density samples (i.e. with low porosity) exhibited less ionic polarization, and a higher electrical resistivity.

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