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Temperature dependence of electrical conductivity of a green porcelain mixture

W. Lerdprom^{a,*}, C. Li^a, D.D. Jayaseelan^a, S.J. Skinner^a, W.E. Lee^{a,b}

^a Department of Materials, Royal School of Mines, Imperial College London, SW7 2AZ, UK

^b Centre for Nuclear Engineering, Department of Materials, Imperial College London, SW7 2AZ, UK

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ABSTRACT

AC conductivity of a green porcelain body was investigated using impedance spectroscopy over a temperature range of 100–950 °C. The results showed that during the heating, conductivity at 100–200 °C increased mainly arising from H⁺ and OH⁻ ions generated from adsorbing physical water. The activation energy increased below the dehydroxylation of clay resulting from movement of monovalent ions. At the dehydroxylation of clay, a combination of H⁺, OH⁻ and monovalent ions dominated the conductivity. The activation energy rose to 1.14 eV (600–950 °C) controlled by diffusion of Na⁺, and K⁺ ions. During the cooling, conductivity showed single activation energy with 0.86 eV resulting from denser microstructure and change in mineralogical constituents and the heat treated porcelain sample showed higher electrical conductivity at the same temperature. Understanding conduction behaviour of the green porcelain enabled more accurate control of furnace temperature in flash sintering, a process which relies on electrical conductivity at high temperatures.

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1. Introduction

Porcelains are often referred to as triaxial compositions of clay, feldspar and quartz [1–3]. Clay provides plasticity in the forming process and behaves like a binder for the other constituents in a green sample. Feldspar promotes densification in a porcelain body as it melts at a low temperature (potash feldspar, the eutectic melt forms at ~990 °C whereas the eutectic melt of soda feldspar forms at ~1050 °C), thereby producing a liquid glass phase leading to viscous flow sintering. Quartz is a glass forming agent and a filler which is used to control the final properties of a fired porcelain body [2,3]. There are several types of porcelain such as hard porcelain, vitreous whiteware, vitreous tile, vitreous sanitaryware, and electrical insulator, (porcelains can also be classified as quartz or alumina porcelain) depending on their mineral composition and firing schedules [4]. They are also commonly glazed, but sometimes unglazed depending on the application [1].

A green porcelain body is heated to transform it to a strong body composed of mullite, glass and undissolved quartz [2]. Mullite is significantly formed above 1100 °C by decomposition of metakaolin which is produced from dehydroxylation of kaolinite

at ~450–600 °C. And, the amount of mullite remains constant in the temperature range 1200–1400 °C [5–7]. Glass is produced by melting of the feldspars around the eutectic point of the triaxial composition and the amount of glass increases as a function of temperature and time by further melting of feldspar and dissolution of quartz [8–10]. Quartz starts to dissolve at 1200 °C and its dissolution scales linearly with log-time [5]. However, other minerals can be introduced into a porcelain body to enhance specific properties such as alumina which is introduced for increasing elastic modulus [3]. It has been reported that the phase composition of a porcelain body is heating rate independent whereas the densification process depends on heating rate. Therefore, if a porcelain body is fired at different heating rates with similar mineralogical constituents, the density is lower at a fast heating rate [5].

The ceramic industry has introduced fast-firing processes to the production of ceramic tiles and dinnerware in order to improve energy usage efficiency. These firing schedules are remarkably short, ranging from 40 to 60 min [8]. Recently, field assisted sintering technology (FAST) has been introduced to produce ceramics using energy more efficiently as well as increasing production rate [9]. Flash sintering relies on the Joule heating mechanism that allows heat to be rapidly generated [10]. The basic premise of flash sintering is that an electrical field is applied directly to a sample under a given furnace temperature via a pair of electrodes [10]. As a result, many successful studies of flash sintering on ceramic mate-

* Corresponding author at: Department of Materials, Royal School of Mines, Imperial College London, SW7 2AZ, UK.

E-mail address: w.lerdprom14@imperial.ac.uk (W. Lerdprom).

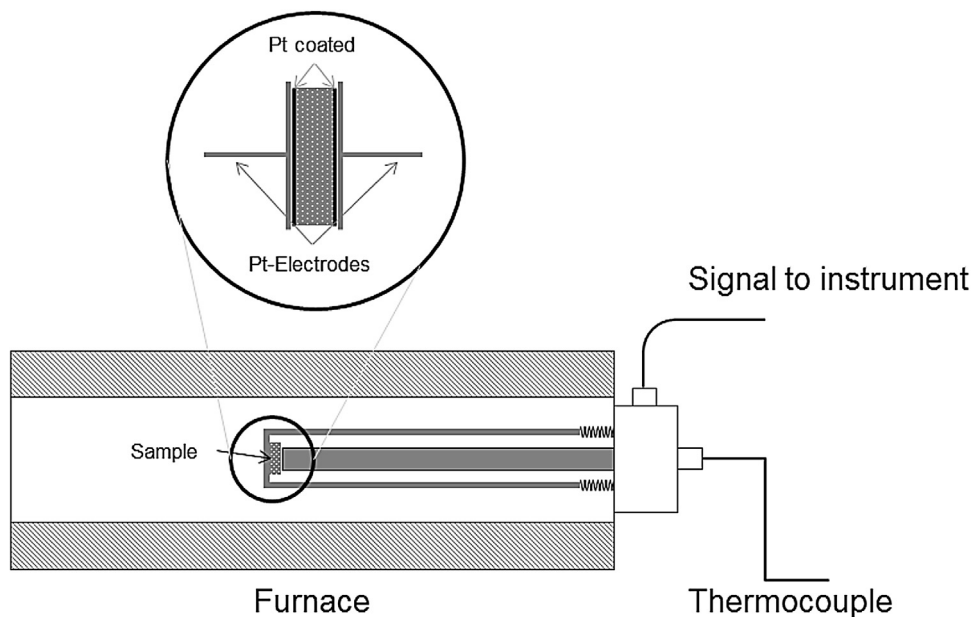


Fig. 1. Schematic diagram of the impedance spectroscopy apparatus. The inset shows the coated sample and two electrodes which were used.

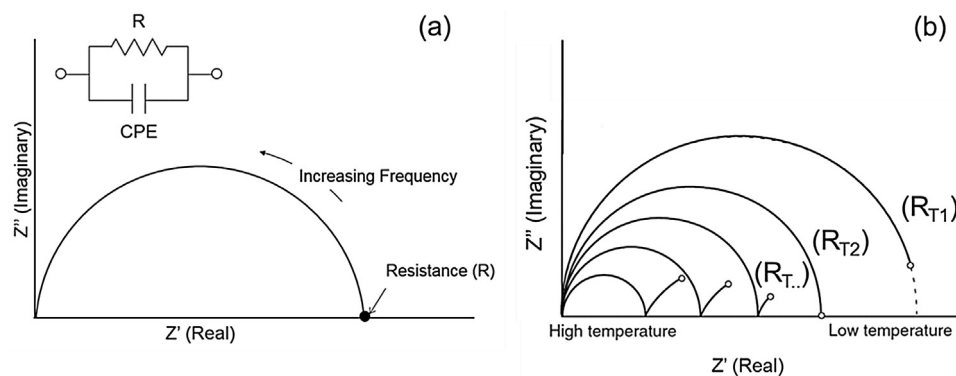


Fig. 2. Schematic diagram showing equivalent circuit and impedance spectra (a). The semicircular responses for bulk (grain interior) conduction mechanism only. Solid circle indicates real and imaginary parts of impedance at a certain frequency which is the obtained resistance. Resistance as a function of temperature (b).

rials have been reported; for example, zirconia [11], yttria stabilized zirconia [12], 3YSZ [13], magnesia doped alumina [14], silicon carbide [15] as well as porcelain [16]. However, it is well recognized that porcelain mixture is a great insulator which exhibits a low conductivity of $10^{-14} \text{ Scm}^{-1}$ at room temperature [17]. This question raises as to the why a porcelain mixture can be sintered by the field assisted technique.

Silicate minerals such as clay, feldspar and quartz are electrical insulators at room temperature but they can behave like semiconductors when temperature is increased due to ionic conduction [18]. Generally, the electrical conductivity of silicate minerals increases exponentially with increasing temperature because the mobility of the charge carrier is a thermally activated process [4,18]. At high temperature, the conductivity increases proportionally with increasing temperature but does not change much at low temperature [19]. Moreover, electrical conductivity of minerals is affected by chemical composition, impurity content, crystal structure, porosity and water content [18–21]. A study on electrical conductivity of quartz and feldspar minerals (anorthite) as a function of temperature reported that electrical conductivity was classified into low and high temperature ranges which corresponded to ionic conduction. At low temperature (below 500°C), the conductivity of both quartz and feldspar was steady whereas at

high temperature (above 500°C) the conductivity rapidly increased [19]. Another study on clay reported that electrical conductivity of kaolinite was caused by protonic conduction at low temperature and free ion conduction at high temperature [22].

The aim of the present work is to determine how electrical conductivity of a green porcelain body changes as a function of temperature in order to establish the lowest possible furnace temperature to achieve flash sintering.

2. Experimental

Industrially-processed spray dried powder obtained from a porcelain tile manufacturer was used in this experiment which provides a better overall uniformity than might be normally expected in a laboratory preparation. The powder was also used in the as-received form without additional processing. Chemical composition (wt.%) of the powder was determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES) (Natural History Museum, London, UK) composing of 72.3% SiO_2 , 19.2% Al_2O_3 , 1.2% Fe_2O_3 , 1.1% CaO , 1.1% K_2O and 3.7% Na_2O . Thermal behaviour of the powder (after dried overnight at 110°C) was characterized using thermogravimetric and differential thermal analysis in static flow mode with alumina as reference mate-

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