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The effect of heating rate and sintering temperature on the elastic modulus of porcelain tiles

Elif Eren Gültekin

Nevşehir Hacı Bektaş Veli University, Fine Arts Faculty, Department of Ceramic and Glass, Prof. Dr. Rıza Ayhan Street, 50800 Hacibektas-Nevsehir, Turkey

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ABSTRACT

The scope of the present study is to investigate the change of elastic modulus with the physical and mechanical properties of porcelain tiles. In the study, porcelain tiles were sintered at different temperatures and at different heating rates to obtain minimum water absorption (%). After this, the time of flight of longitudinal and shear ultrasonic waves was measured through the tile. The time of flight of ultrasonic waves was measured using contact ultrasonic transducers operating on a pulse-echo mode. Using the time of flight of the ultrasonic waves and the thickness of the tiles, the velocity of the waves was determined. Using the ultrasonic velocities and bulk density, the elastic modulus of the tiles was determined. Helium pycnometry, a bend test, scanning electron microscopy (SEM) and X-ray diffraction (XRD) analyses were also carried out. The results show that the elastic modulus decreased with an increase of total porosity, but increased with bulk density and firing shrinkage. There is a polynomial relationship between elastic modulus and strength.

1. Introduction

Characterization techniques are the basic tool for the quality control and quality assurance of a material or component or product. Destructive, semi-destructive and non-destructive testing (DT & NDT) techniques are available for complete characterization. Quantities, ultrasonic velocity and attenuation are important parameters which are required for the ultrasonic non-destructive technique of material characterization. The ultrasonic velocity is related to the elastic constants and density of a material. Hence, it gives information about the mechanical, anisotropic and elastic properties of the medium it passes through [1]. Determination of the change of physical and mechanical properties, depending on the measurement of the wave transmission time, detection of defects and ultrasonic signal measurement were performed on ceramic tiles using the application of the ultrasonic method [2–14]. The apparent density and bulk density distribution of green ceramic tiles can be measured using calibration curves determined using the conversion factor between ultrasonic velocities and densities [2,3]. Ultrasonic velocity was also found to be a sufficiently sensitive and reliable technique for the detection of delamination in green and sintered ceramic tiles [4]. While the measurement of ultrasonic velocity is utilized to predict strength and the dynamic elastic modulus of porcelain tiles [5], it can also be used to characterize porosity, water absorption, bulk and apparent density and to identify defects (cracks, a piece of paper and aluminum foil, silicon nitride, zirconia and steel balls of different diameter, rubber of different thicknesses, carbon black and polymethylmethacrylate) [6–12]. In addition, the effects of the phases on ultrasonic properties have been investigated by Eren Gültekin et al. and Aydın et al. [13,14].

In this study, the aim is to measure ultrasonically the differentiating elastic modulus of porcelain tiles with low water absorption and low open porosity. For this purpose, porcelain tiles were sintered at different combinations of sintering temperatures and heating rates. Helium pycnometry, a bend test, scanning electron microscopy (SEM) and X-ray diffraction (XRD) were also performed to support the results of the study.

2. Experimental

Standard porcelain tile granules were prepared by the uniaxial pressing technique in a 50 mm × 100 mm rectangular die at 450 kgf/cm^2 (Gabrielli Laboratory Press). In Ref. [15], the determination of factors (heating rate and sintering temperature) and levels were previously reported [15]. Sintering was carried out in a Nabertherm LS 25/26 furnace. The sintering temperatures of the porcelain tiles were 1210, 1220, 1230 °C, the heating rates were

E-mail address: eeren@nevsehir.edu.tr

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40, 50, 60 °C/min. and the soaking time at sintering temperature was constant (6 min). The cooling rate of all the tiles was also kept constant at 60 °C/min. Two tiles were sintered for each temperature and heating rate combination. All the sintered tiles were characterized through the determination of firing shrinkage (ASTM C326).

Ultrasonic measurements were performed at room temperature using a 5 MHz longitudinal wave transducer and a 2.25 MHz shear wave transducer. The transducers were excited by timed pulses from an Olympus Panametrics-NDT Model 5800 Computer Controlled Pulse/Receiver through the thickness of the samples. The time of flight of the ultrasonic signals was performed using the pulse-echo method with a digital oscilloscope (Tektronix, TDS 1012 Two Channel Digital Storage Oscilloscope). Transit time was determined to within an accuracy of ± 40 nsec. Ultrasonic wave velocity was determined by measuring the tile thickness with a micrometer (0.01 mm resolution Mitutoyo M110-25 DS micrometer) and dividing the path length (twice the measured thickness) by the time of flight [16]. The time of flight measurements for each of the porcelain tiles were repeated three times.

The bulk density, open porosity and water absorption of the tiles were measured using the Archimedes technique. Ultrasonic velocities and densities were used to calculate the elastic modulus. Assuming that the samples used in this analysis are isotropic, standard velocity-elasticity relationships can be used to calculate the elastic modulus. These relationships are:

$$E = \frac{v_l^2 d_b (1+\sigma)(1-2\sigma)}{(1-\sigma)} \quad (1)$$

$$\sigma = \frac{(1-2b^2)}{(2-2b^2)} \quad (2)$$

where v_l is the longitudinal wave velocity (m/s), v_s is the shear wave velocity (m/s), d_b is the bulk density (kg/m^3), E is the elastic modulus (GPa), σ is the Poisson's ratio and b is equal v_s/v_l [17].

The total porosity (open and closed pores) of the tiles was measured by pycnometry (Quantachrome, Model No: MVP-1 Multipycnometer) using Eq. (3):

$$P\% = \frac{(d_t - d_b)}{d_b} \times 100 \quad (3)$$

where d_t represents the true density and d_b represents the bulk density [18].

The bend strength of the tiles was measured using the three-point bend test (Gabrielli CR5) according to ISO-EN 10545-4 standards. For scanning electron microscopy (SEM) observations, a Carl-Zeiss EVO-50 was used in secondary electron (SE) imaging mode. The crystalline phases in the sintered porcelain tiles were determined by X-ray diffraction (XRD) analyses. For XRD analyses, sintered tiles were scanned from 2θ = 5–70°, at a scanning speed of 2°/min., using a Rigaku Miniflex 600 diffractometer with $Cu_{K\alpha}$ radiation at 40 kV and 15 mA. The crystalline phase composition was quantitatively analysed with Material Analysis Using Diffraction (MAUD) software based on the Rietveld method.

3. Results and discussion

Porcelain tiles are very strongly sintered ceramic materials with water absorption and open porosity close to zero [19]. All the porcelain tiles sintered at each sintering temperature and heating rate combinations have water absorption values lower than 0.2% and open porosity values lower than 0.45%, as shown in Table 1. Kingery [20] establishes two different steps during sintering. The first shows a decrease in open porosity, which is coincidental with an increase in the sample shrinkage. The sintering process is not finished when open porosity completely disappears. In the second step, the ceramic body could be represented as a conjunction of small and closed pores. The surface energy forces inside each pore give rise to a negative pressure, which tends to densify the ceramic body. Table 1 lists the physical and mechanical properties of porcelain tiles. Open porosity decreases to 0.03% and firing shrinkage increases to 8.29% at a sintering temperature of 1220 °C and a 40 °C/min. heating rate, establishing the optimum firing temperature and the firing range at which open porosity reaches a minimum, which usually corresponds to higher values of mechanical strength [21].

Minimum total porosity was obtained at a sintering temperature of 1220 °C and a 40 °C/min. heating rate. In addition, the highest firing shrinkage was observed at this sintering temperature and heating rate combination. After the minimum porosity was reached, if the temperature was increased, the pressure of the gas inside the closed pores tended to expand the pores and therefore, the linear shrinkage was decreased and the total and closed porosity was increased. Furthermore, as the closed porosity increased, the bulk density decreased [22]. At 1230 °C, and for all heating rates, linear shrinkage decreased and total porosity increased. The lowest firing shrinkage was obtained at a sintering temperature of 1230 °C and a 40 °C/min. heating rate. An SEM analysis indicated differences between the morphology of the porosities in sintered porcelain samples which contained maximum and minimum total porosities. Whereas the sample sintered at 1220 °C and a 40 °C/min. heating rate (Fig. 1a) contained isolated pores in large numbers, the sample sintered at 1230 °C and a 40 °C/min. heating rate (Fig. 1b) contained isolated large pores due to overfiring and expansion of the gas inside the pores. The overfiring effect of the highest sintering temperature with the lowest heating rate was confirmed by SEM observations.

Eren et al. investigated the phase development of porcelain tiles with temperature and found that the amount of glassy phase and mullite increases with temperature as a result of quartz and albite dissolution [23]. Analysis of the phases by XRD shown in Fig. 2–4. Quantitative X-ray analysis was also carried out on the specimens (Table 2). It was found that there was a maximum of 1% difference in the amount of mullite depending on the composition. The difference was ~6% in albite, ~4% in quartz and ~10.5% in the glassy phase. The amount of glassy phase increased from 60.83% to 65.83%, depending on the increase in heating rate at the sintering temperature of 1210 °C, whereas the amount of glassy phase is independent of the heating rate when the sintering temperature is 1220 °C. For a sintering temperature of 1230 °C, the highest amount of glassy phase (71.22%) was obtained at a low heating rate (40 °C/min). With a decrease of heating rate, whereas the glassy phase content increased, the quartz content decreased at this sintering temperature. Due to the high sintering temperatures, the products are rich in glassy phase and, for the same composition, the density of the glass is lower than that of the crystalline phases [22]. The highest glassy phase amount was obtained for sintering at 1230 °C and a 40 °C/min. heating rate with a total porosity of 13.86% so the lowest bulk density was obtained for this combination. In contrast, the combination sintered at 1210 °C and a 40 °C/min. heating rate had the lowest glassy phase amount and the highest density value, although total porosity was not the lowest.

The ultrasonic velocity in powder compacts is directly dependent of density and elastic modulus. An increased understanding of the relationship between ultrasonic velocity and the state of powder compact is needed to realize the full potential of ultrasonic velocity as a parameter for the characterization of sintering materials [16]. The ultrasonic velocity and elastic modulus values dependent on heating rate and sintering temperature are shown in Table 3. Both ultrasonic longitudinal and shear waves' velocities

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