EFFECT OF QUARTZ PARTICLE SIZE ON THE THERMAL EXPANSION OF POROUS WHITEWARE BODIES

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Three compositions were prepared based on a typical porous whiteware body composition (Table 1). The compositions only differed in the quartz particle-size distribution used, the coarsest being S-6 and the finest S-300 (mean particle sizes of 42, 23 and 8 μm for S-6, S-10 and S-300, respectively).

Sample	Clays	Potassium feldspar	Calcite	Quartz S-6	Quartz S-10	Quartz S-300
A	55	5	15	25	-	-
B	55	5	15	-	25	-
C	55	5	15	-	-	25

Table 1. *Compositions used (percentages by weight).*

Two series of test specimens were formed by pressing at a moisture content of 5.5 kg water/kg dry solid with each composition. In one series, the pressing pressure was kept constant, yielding specimens with different green bulk densities according to the quartz particle size involved. In the other series, pressing pressure was modified to produce specimens with the same green bulk density in the three compositions.

The specimens were fired with a typical wall tile thermal cycle, using three different peak temperatures (1050, 1100 and 1150°C). The following determinations were run on the fired specimens: bulk density by immersion in mercury, percentage of quartz by X-ray diffraction, apparent porosity, as water absorption (W.A.) and thermal expansion curve in a dilatometer, using a heating and cooling cycle of 5°C/min. The numerically derived thermal expansion curve was used to calculate the mean coefficient of expansion between 100 and 650°C in the heating stretch ($\alpha_{100-650}$) and the difference between the maximum coefficient of expansion values in the heating stretch and in the cooling stretch ($\Delta\alpha$). The results are detailed in Table 2.

The most significant characteristic of the thermal expansion curves was the hysteresis between the heating and cooling stretches, which yielded a noticeable difference in the coefficients of expansion in these segments in the region approaching 573°C (Figure 1).

The origin of the hysteresis appears to be related to microcrack formation in the specimen during cooling in the kiln, as the quartz coefficient of expansion is larger than that of the other existing phases (glassy phases, anorthite, gehlenite, etc.) [1,2]. Hence, during cooling of the specimen fired in the dilatometer, only a small fraction of the quartz present contributes to overall specimen expansion, owing to the fact that many quartz particles are separated from the matrix by microcracks, which accommodate their expansion. On reaching peak test temperature, which is higher than the transformation temperature of the glassy phase contained in the matrix, the quartz particles are bonded to the matrix.

Pressure (kg/cm^2)	Type of quartz	$\rho_{ap\ dry}$ (g/cm^3)	T_{firing} (°C)	$\rho_{ap\ fired}$ (g/cm^3)	% free quartz	W.A. (%)	$\alpha_{100-650}$ (°C^{-1}·10^7)	$\Delta\alpha$ (°C^{-1}·10^7)
220	S-6	1.99	1050	1.79	45	16.9	103	170
			1100	1.78	42	16.8	100	153
			1150	1.79	36	16.6	87	125
220	S-10	1.96	1050	1.77	45	17.5	106	170
			1100	1.76	43	17.8	99	162
			1150	1.77	35	16.9	89	114
220	S-300	1.87	1050	1.70	42	19.8	108	119
			1100	1.70	38	19.9	106	95
			1150	1.72	30	18.5	92	65
170	S-6	1.96	1050	1.75	44	17.6	100	181
			1100	1.75	42	18.5	97	157
			1150	1.76	36	17.9	85	133
400	S-300	1.96	1050	1.78	41	16.0	109	112
			1100	1.78	38	15.6	105	105
			1150	1.79	29	15.3	92	69

Table 2. *Results of the experiments conducted.*

During specimen cooling in the dilatometer, the existing phases remain bonded to a temperature approaching 573°C, as in this temperature range the coefficients of thermal expansion of all the phases are similar and the cooling rate is slow. When the quartz shrinks abruptly at 573°C, most of the quartz is joined to the matrix and contributes to overall specimen shrinkage. Consequently, at this temperature, the coefficient of thermal expansion found on cooling the specimen must be greater than the one found on heating it. According to this assumption, the fired specimen thermal expansion values on heating, as well as the magnitude of the hysteresis (measured as $\Delta\alpha$), must be directly related to the concentration of microcracks.

The concentration of microcracks, which will be proportional to the quartz that does not contribute to the thermal expansion of the fired specimen during heating, will, amongst other variables, depend on the particle size of the quartz used, since the coarser the quartz, the greater will the arising stresses between the existing phases be. [3,4,5]

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[1] Manning, W.R., Hunter, J.R., Calderwood, F.W. y Stacy, D.W.: *Thermal expansion of Nb_2O_5* . J. Amer. Ceram. Soc. 55 (7) 342-347 (1972).

[2] Kuszyk, J.A. y Bradt, R.C.: *Influence of grain size on effects thermal expansion anisotropy in $MgTi_2O_5$* . J. Amer. Ceram. Soc. 56 (8) 420-423 (1973).

[3] Parker, F.J. y Rice, R.W.: *Correlation between grain size and thermal expansion for aluminium titanate materials.* J. Amer. Ceram. Soc. 72 (12) 2364-2366 (1989).

[4] Warshaw, S.I. y Seider, R.: *Comparison of strength of triaxial porcelains containing alumina and silica.* J. Amer. Ceram. Soc. 50 (7) 337-343 (1967).

[5] Amorós, J.L., Belda, A., Orts, M.J. y Escardino, A.: *Expansión térmica de las piezas de pavimento cerámico gresificado: influencia de las variables de prensado y la temperatura de cocción.* Bol. Soc. Esp. Ceram. Vidr. 31 (2) 109-114 (1992).

It can be observed in Table 2 that as quartz particle size increases, keeping firing temperature steady, the values of $\alpha_{100-650}$ drop, independently of whether specimens made at an identical pressing pressure or having identical green bulk density are considered. The hysteresis rose with quartz particle size, the effect being more marked in the specimens with identical green bulk density than in those formed at the same pressure. These data suggest that the larger the starting quartz particle, the more abundant are the microcracks in the fired specimen, as the stresses arising between the quartz and the glassy phase are greater. The fact that the tendencies are clearer in the specimens with identical green bulk density is because these specimens have a similar pore volume. As pores are stress release points [6], their effect will be very similar in this series of specimens. However, in the specimens with different bulk densities, the effect of porosity on stress release was different for each composition, partially disguising the expected tendency.

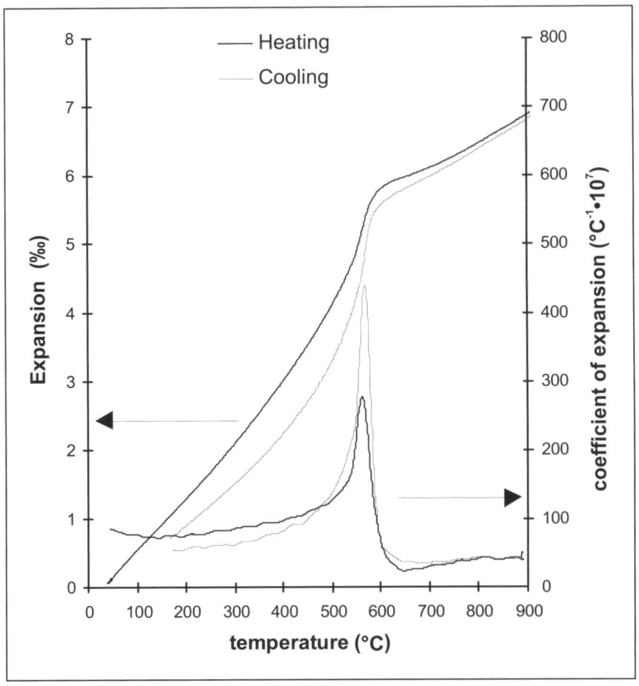

Figure 1. Thermal expansion curve.

The rise in firing temperature entailed a drop in the value of $\alpha_{100-650}$ and $\Delta\alpha$, regardless of the series of specimens considered. This evolution appears to be due to progressive enrichment with silica of the glassy phase and the formation of anorthite and gehlenite, mainly at the expense of the finest quartz fraction, as indicated by the quartz measurements. The formation of these phases, which were richer in silica, with a lower coefficient of thermal expansion than quartz, and the incorporation of SiO_2 into the glassy phase lowered specimen overall thermal expansion.

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[6] LACHMAN, J.M. Y EVERHART, J.O.: *Development of safe cooling schedules for structural clay products.* J. Amer. Ceram. Soc. 49, 30-38 (1956).