Uluslararası Mühendislik Araştırma ve Geliştirme Dergisi
International Journal of Engineering Research and Development
UMAGD, (2021) 13(3), s40-s48.
10.29137/umagd.1036506
Cilt/Volume:13 Sayı/Issue:3 Aralık/December 2021 Özel Sayı / Special Issue
Seçilmiş Konferans Makalesi / Selected Conference Paper

The Effect of Mould Filling on the Mechanical Properties of Wall Tile Production

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***Başvuru/Received**: 07/10/2021* ***Kabul / Accepted**: 25/12/2021* ***Çevrimiçi Basım / Published Online:** 31/12/2021*
***Son Versiyon/Final Version**: 31/12/2021*

Abstract
This study is focused on the development of properties of ceramic wall tile bodies by changing the degree of mould filling under industrial conditions (Usak Seramik A.S., Usak, Turkey). The effects of spray dryer operating conditions and granule size distribution on the degree of mould filling have been investigated. The study measured the ceramic wall tile bodies water absorption, fired shrinkage, fired and dried strength, and porosity. The granule size and distribution were changed by adjusting the pressure and the nozzles' diameter of the spray dryer. This study aimed to reduce grain size and quantity over 500 microns and to increase quantity between 500 microns and 75 microns by increasing the spray dryer pressure and reducing the nozzle disc diameter. It was also aimed to increase the degree of mould filling by decreasing granule size. 200 tone wall tile slips were prepared and 11,000 m2 of wall tile products were produced. The phase and microstructure analysis were investigated using an X-ray diffractometer (XRD) and scanning electron microscope (SEM). As a result of this study, it was found that a lower granule size of above 500 µm improved the technical characteristics of the tiles. Ceramic wall tiles produced with lower granule grain size have high strength values, mainly due to the higher degree of mold filling. The ceramic tile production parameters in the Usak Seramik Company have been rearranged according to the data obtained from this study. This granule size and distribution set is now an industrial production standard in Usak Seramik.

Key Words
"Wall tile, spray dry, sintering, thermal expansion, powder fluidity"

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1. Introduction

Wall tiles are porous ceramic products. They are suitable for indoor applications due to their high open porosity. Wall tiles are composed of clays, carbonates, quartz, and feldspars. They are single or double fired in industrial roller kilns at temperatures between 1050 °C and 1145 °C for 50 min. Fired bodies contain quartz, albite, anorthite, and gehlenite. anorthite and gehlenite are formed as calcium-based crystalline phases in the firing process. The crystalline phases formed during firing are very important due to it affecting the technical properties and tile performance (Çelik et al. 2010, Siqueira FB et al. 2018, Freire MN et al. 2006, Tarhan M. et al. 2019, Tarhan B. 2019). All ceramic tiles are formed using dry pressing to give the desired shape. Dry pressing is also preferred because of the technical and quality characteristics of tiles. Certain parameters such as density, apparent density, compactness, and degree of filling are crucial to the result of an efficient pressing (Vodola L. et al. 2014, Tarhan M. et al. 2016, Aydın T. et al. 2019, Ring TA.1966). To achieve a good pressing result, a uniform distribution of granules in the mold cavity is required (Ring TA.1966, Sacmi, 2002). To achieve this, the powder must have good fluidity, which allows tile manufacturers to produce faster without disturbing the homogeneity of the pressed tiles.

Spray drying is the method of spraying a slip obtained from the grinding process in the slurry into a warm drying medium to produce nearly spherical powder granules with controlled moisture, shape, and particle size distribution. Generally, the diameter of the spray-dried powder grains are between 100 µm and 600 µm. Granule characteristics are determined with spray dryer type, slurry composition, inlet temperature, slip pump pressure, nozzle diameter, the humidity of the air, slip viscosity, and density. Using high pressures brings about a smaller particle size distribution; nozzle diameters determine the particle size distribution. Larger particle size distributions are enabled using larger orifice diameter (Reed JS. 2002). Properties of granules (shape and grain size), moisture, and pressing additives (water, binding and plasticizer agents) are important parameters for the characteristics and microstructures of the pressed tiles. These parameters determine the technological characteristics of powders such as volume density, flowability, compression rate, and friction angle of powders (Beddow JK.1995, Santomaso A.et al. 2003, Soldati R. et al. 2018).

The Hausner ratio is used in a wide variety of industries including the ceramic industry. The Hausner ratio, which is not an absolute property of a material, is a number correlated with the flowability of granules. The Hausner ratio is calculated by the formula; (Beddow JK.1995, Santomaso A.et al. 2003, Soldati R. et al. 2018).

$H=\rho T/\rho B$
ρT: Tapped bulk density
ρB: Freely settled bulk density of the powder

The good flowability of the granules alone are not enough for ideal pressing. The powder should be inappropriate volume weight. Extremely low volume weight causes a large amount of air to escape during pressing or a thicker powder-filling layer. Both cause difficulties in the pressing cycle (Ring TA.1966, Sacmi, 2002, Reed JS. 2002).

There are many studies on the technological properties of ceramic wall tiles. Although it is one of the most important parameters regarding its technological properties, there are no studies in the literature on the degree of mold filling. Therefore, this study focused on the effect of the degree of mould filling on the technical properties of ceramic wall tile bodies under industrial conditions.

2. Materials and Methods

Wall tile slips were prepared under industrial conditions at Usak Seramik A.S. The ceramic wall tile composition is composed of clay, kaolin, sodium feldspar, and marble powder as a source of CaO. A mixture of 200 tons of ceramic wall tile raw materials for the standard body was ground using a rotary mill until the residue obtained was 2.5–3% over 63 µm. The density and viscosity of slip taken from the mill are respectively, 1660 g/lt and 40-45 seconds (Ford cup #4). The slip taken from the mill was sent to the spray dryer. The pressure was 14 bar and the diameters of the spray dryer nozzle discs were 2.5 mm, 2.2 mm, and 2 mm for standard production conditions. In this study, the spray pressure and nozzle disc diameters were changed while the density and viscosity of the slip were kept constant. The spray dryer pressure was increased from 14 bar to 18 bar. The 2.5 mm nozzle disc diameter was reduced to 2.2 mm, the 2.2 mm nozzle disc was reduced to 2 mm and the 2 mm nozzle disc diameter was reduced to 1.8 mm. It was aimed to decrease the granule grain size to be above 500 µm and increase between 500 µm and 75 µm with an increase in spray dryer pressure and a reduction in the nozzle discs diameter. 6-6.5% moist granules obtained from the spray dryer were pressed to 30 x 60 cm in size, to 280 kg/cm^2 of pressure under industrial conditions. After the drying process, the pressed tiles were fired in an industrial roller kiln at 1120ºC for 47 minutes.

In this study, about 11,000 m^2 of ceramic wall tiles were produced under industrial conditions. Technological properties such as water absorption, fired and green density, fired shrinkage, dry and fired bending strength were analyzed. The values of the firing shrinkage were measured by calculating the size of green and fired samples. The values of the dry bending strengths were determined using the flexure test (three-point bending test) (HYK-500B Digital Electricity Anti-fold Instrument) according to ISO 10545-4 standards. The

values of the water absorption were also determined according to ISO 10545-3 standards (HY Ceramics absorbs water rate testing instrument).

Chemical analyses of raw materials were determined by an X-ray fluorescence analyzer (Rigaku Technologies, In, Nex OC+EZ). The crystalline phases in the fired body compositions were evaluated by using an X′ Pert Pro MPD diffractometer with Cu Kα radiation at 40 kV and 30 mA. The samples were scanned from 2θ=10−70° at a scanning rate of 2°/min. The microstructures of the sintered bodies were detected using a SEM (scanning electron microscopy) (QUANTA FEG 250). After the polishing process, all samples were coated with Au and Pd for SEM analysis. In order to determine the sintering behavior of the samples, a non-contact optical dilatometer was used (Netzch 402 EP dilatometer).

3. Result and Discussion

3.1. Chemical analysis of raw materials

Chemical analyses of raw materials are given in Table 1. In the study, two kinds of clay kaolin, and Na Feldspar were used, and marble powder was used as a CaO source.

Table 1. Chemical analysis of raw materials (wt. %)

	LOI*	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	Na_2O	K_2O
Kaolin A	5.00	75.51	16.30	1.17	0.44	0.29	0.78	0.05	0.47
Kaolin B	5.00	74.16	14.83	2.02	0.42	0.53	1.40	0.01	1.65
Clay A	8.00	60.54	21.86	3.73	1.27	0.40	0.75	0.78	2.62
Clay B	7.00	60.95	23.70	2.98	1.12	0.40	0.80	0.82	2.24
Na Feldspar	0.50	71.78	15.82	0.85	0.13	0.29	0.62	4.40	3.89
Marble Powder	32.00	0.01	0.16	0.08	0.01	56.19	1.34	0.01	0.01

3.2. Characterization of ceramic granules used in pressing

Figure 1 indicates the morphology of the STD and D1 granules. As seen in Figure 1, D1 granules have much smaller grains which show that the greater the tendency for a spherical shape, the larger the surface area is. STD granules have much more irregular coarser grains than D1 granules. The moisture content of the granules over 400 micron is higher because they spend less time in the spray drying chamber on account of their volume. These large granules have a tendency towards agglomeration (indicated with arrows in Figure 1) and inclusion of porosity because of their high residual moisture content (donut shape). Donut-shaped and more agglomerated STD spray-dried granules are indicated in Figure 1.

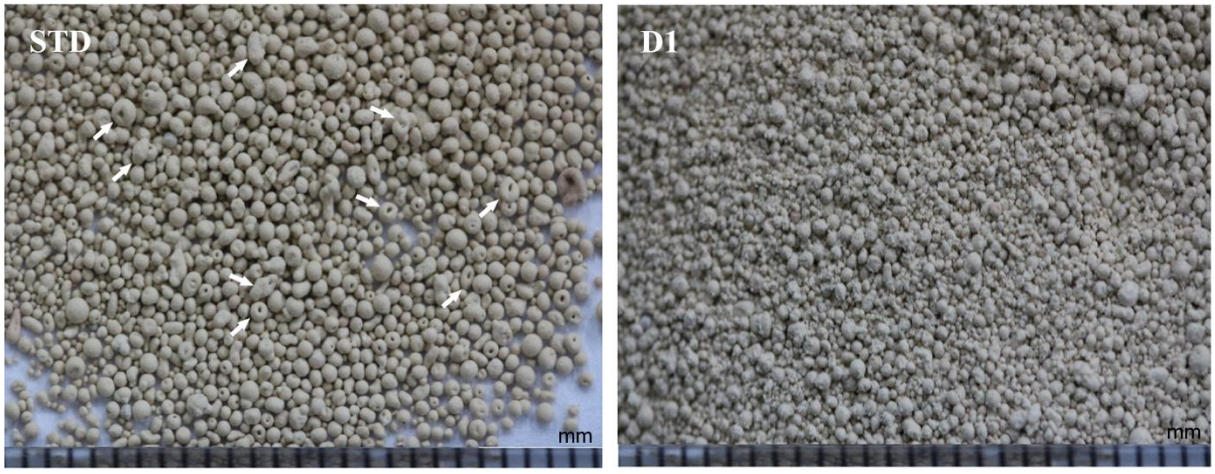

Figure 1. MOD micrographs of the STD and the D1 spray-dried powder

Granule size distributions of samples and particle trapped density and vibrated density values are given in Table 2 and Table 3. As can be seen from Table 2, with the reduction of the nozzle disc diameters and increasing the slip pressure, an increase in the amount of granules between 150-500 microns was achieved. The increase in the amount of granule in this range led to an increase in trapped density and vibrated density and thus the Hausner index increased (Table 3). The Hausner index is the ratio between the tapped density of the powder and the vibrated density (Tarhan M. Et al. 2016). The trapped density is the bulk density of a known volume of powder after controlled vibration. The higher the flowability between the granules, the higher the trapped density and the Hausner index. The

specific diameter for the prepared powders is an important parameter. In both cases, small granule sizes give low fluidity. Especially, particles smaller than 200 microns reduce the flow rate to almost zero. Those between 200 and 500 micron provide optimum flow rates. Grains other than 150-500 micron have the lowest volumetric density values and those in this range have higher density and best packaging characteristics (Sacmi, 2002).

Table 2. Granule size distributions of samples

	STD / %	D1 / %
+500 µm	35.11	20.31
250-500 µm	39.82	53.59
150-250 µm	15.16	18.88
75-150 µm	7.41	4.50
-75 µm	2.50	2.72

Table 3. Particle trapped density and vibrated density values

Sample	Tapped Density (g/lt)	Vibrated Density (g/lt)	Hausner Index
STD	0.872	0.950	0.917
D1	0.987	1.068	0.924

3.3.Technological properties

Table 1 shows the technological properties of the samples. As mentioned before, increasing the spray dryer slurry pressure from 14 bar to 18 bar and reducing the spray dryer nozzle diameters of 2.5, to 2.2, 2.2 mm to 2.0, and 2.0 to 1.8 mm led to an increase in trapped density and vibrated density. Properly formulated and well-controlled feed material is a key to successful pressing operations. Pressing problems are reduced when the bulk density of the feed in the die is high. A high bulk density reduces both the content of the air in the powder and the punch travel (Reed JS, 2002)]. Increase in trapped density and vibrated density resulted in an increase in bulk density. With this increase, dry and fired strength increased while water absorption and total porosity slightly decreased. This behavior can be explained by the fact that the particles are broad close together, thus reducing voids between particles. There is also increased inter-particle contact, forming bridges and links between them (Sacmi, 2002). Mechanical strength depends on two factors: intrinsic material characteristics (including material microstructure) and the presence of microcracks. Pores obviously decrease the cross-sectional area on which the load is applied but also act as stress concentrators. The strength of porous ceramics is decreased in a way that is nearly exponential with porosity (Kingery WD, 1960). Mechanical strength drops due to the microcrack size growth on working with larger granules. In addition, increase in trapped density and vibrated density contributed to decreasing in firing shrinkage (Sacmi, 2002). In terms of production stability of ceramic wall tiles, low shrinkage and low porosity are important in firing temperatures ranges (Amoros et al. 2000).

Table 4. Technological properties

Sample	Bulk Density (g/cm^3)	Water Absorption (%)	Total Porosity (%)	Dry Strength (kg/cm^2)	Fired Strength (kg/cm^2)	Fired Shrinkage (%)
STD	1.662	17.24	33.17	20.86	196.7	0.06
D1	1.853	16.72	30.01	24.53	235.2	0.04

3.4.Phase analyses (XRD)

Phase analyses of STD and D1 bodies are given in Figure 2. STD and D1 bodies contain mainly quartz anorthite and gehlenite crystalline phases. Gehlenite and anorthite are formed in a mixture of clay and calcite during firing. The first crystallized phase is gehlenite from metakaolinite and calcium oxide (Souse SGJ, 2005). Calcium oxide is formed from the decomposition of $CaCO_3$ (marble powder). The reaction between calcium oxide and the low liquid phase formed by the decomposition of kaolinite leads to high porosity and formation of anorthite and gehlenite crystalline phases in the ceramic bodies (TarhanM.et al.2016, Aydın T. et al., 2019). Then, anorthite is crystallized by gehlenite, metakaolinite, and quartz. As it can be seen from Figure 2, both ceramic tile bodies have similar crystal phases and the peak intensities of these phases are quite close.

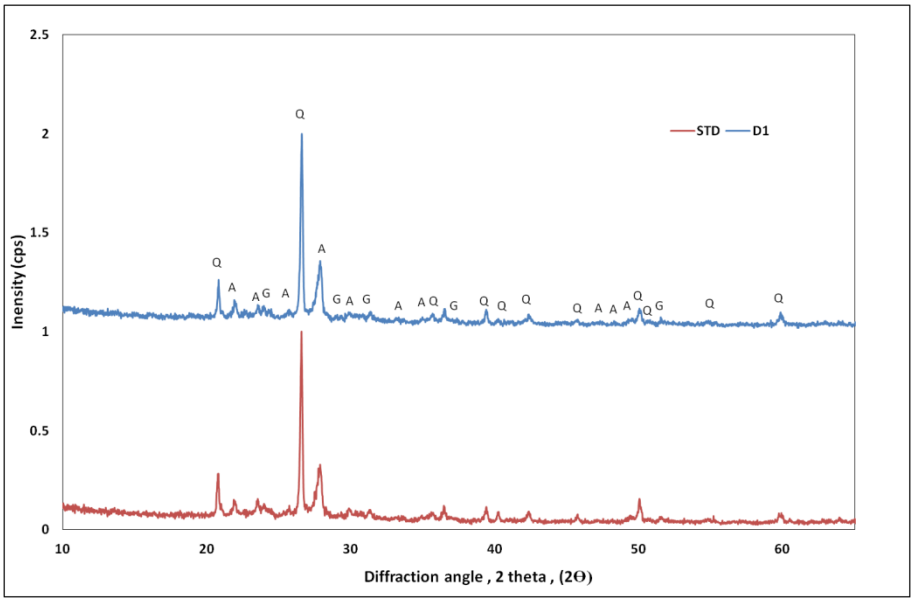

Figure 2. XRD analyses of STD and D1 bodies, Q: quartz, A: anorthite, G: gehlenite

3.5. Thermal Analysis

3.5.1.*Thermal expansion coefficient*

The thermal expansion coefficient and moisture expansions are given in Figure 3 and Table 5. In the study, α, the mean coefficient of linear thermal expansion, was measured between 100 and 600 °C. The free quartz content resulted in a high mean coefficient of linear thermal expansion (α) in ceramic bodies. It was observed that the density of the tiles increased with the decrease in granule grain size. Therefore, the coefficients of thermal expansion of the bodies increased with decreasing granule size. This contradiction can be explained by the microcracking phenomenon and porosity(Amorós JL, 2010, Tarhan M, 2010). Other parameters affecting the thermal expansion coefficient values of the bodies are bulk density and microstructure (Tarhan M, 2010. As can be seen in SEM images, the decrease in the bulk density and the increase in the microcracks in the microstructure reduced the thermal expansion coefficient value in the STD body.

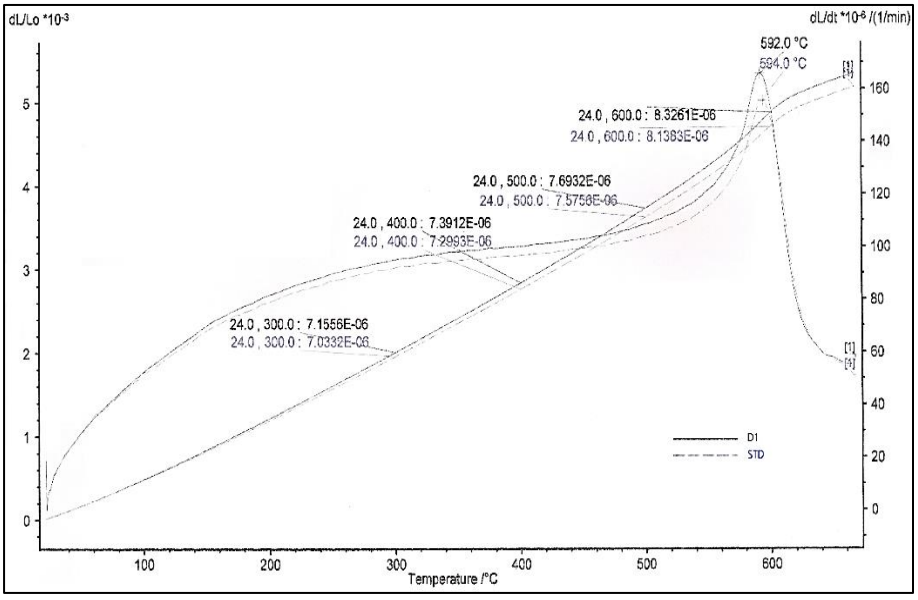

Figure 3. The thermal expansion coefficient of STD and D1

In the study, moisture expansions of the STD and D1 bodies were measured using the dilatometric method. In this method, firstly, the first thermal dilatometric cycles were measured and the samples were heated to 105 °C. Then, the temperature was increased to 700 °C min-1. After that, STD and D1 bodies were kept under a 6-atu steam treatment in an autoclave between 3 and 5h (Aydın T. et al., 2019).

The second thermal dilatometric analysis was measured after the autoclave process. The differences in dilatometric curves showed moisture expansion of ST and D1 bodies.

There are several parameters affecting moisture expansion. These parameters are surface energy, amorphous and crystal phases, and water-reactive phases such as CaO and MgO. Crystal phases such as anorthite are inert to moisture due to low surface energies. In contrast, amorphous phases are very reactive to moisture due to high surface energies (Aydın T. et al., 2019). CaO and MgO, water-reactive phases, must be bonded to the crystalline phases, such as anorthite and gehlenite. As can be seen in the XRD graph, there is no significant difference in crystal phase amounts in this study. This situation caused the moisture expansion values to be very close to each other.

Table 5. Thermal expansion coefficient and moisture expansions

Sample	Thermal expansion coefficient (α) /10^{-7} K^{-1}				Moisture expansion (%)
	300 ºC	400 ºC	500 ºC	600 ºC	
STD	70.33	72.99	75.75	81.38	0.030
D1	71.55	73.91	76.93	83.26	0.031

3.5.2.Sintering analysis

Sintering analyses of the sample STD and D1 are given in Figure 4. As seen in the graphic, it was determined that the sintering behaviors of the STD body and the body studied on grain size, are very close to each other. The ceramic wall tile bodies were heated at 1150 °C. The soaking time of the samples is 6 minutes. The thermal expansion was observed from the initial temperature to 900 °C. Especially, the increase in expansion resulted from quartz inversion in 573°C. After quartz inversion, shrinkage gradually increased and sintering started between 900 °C and 1000 °C. As seen graph, there was a peak at about 1050°C. This peak shows the formation of anorthite crystals (Aydın T. et al., 2019).

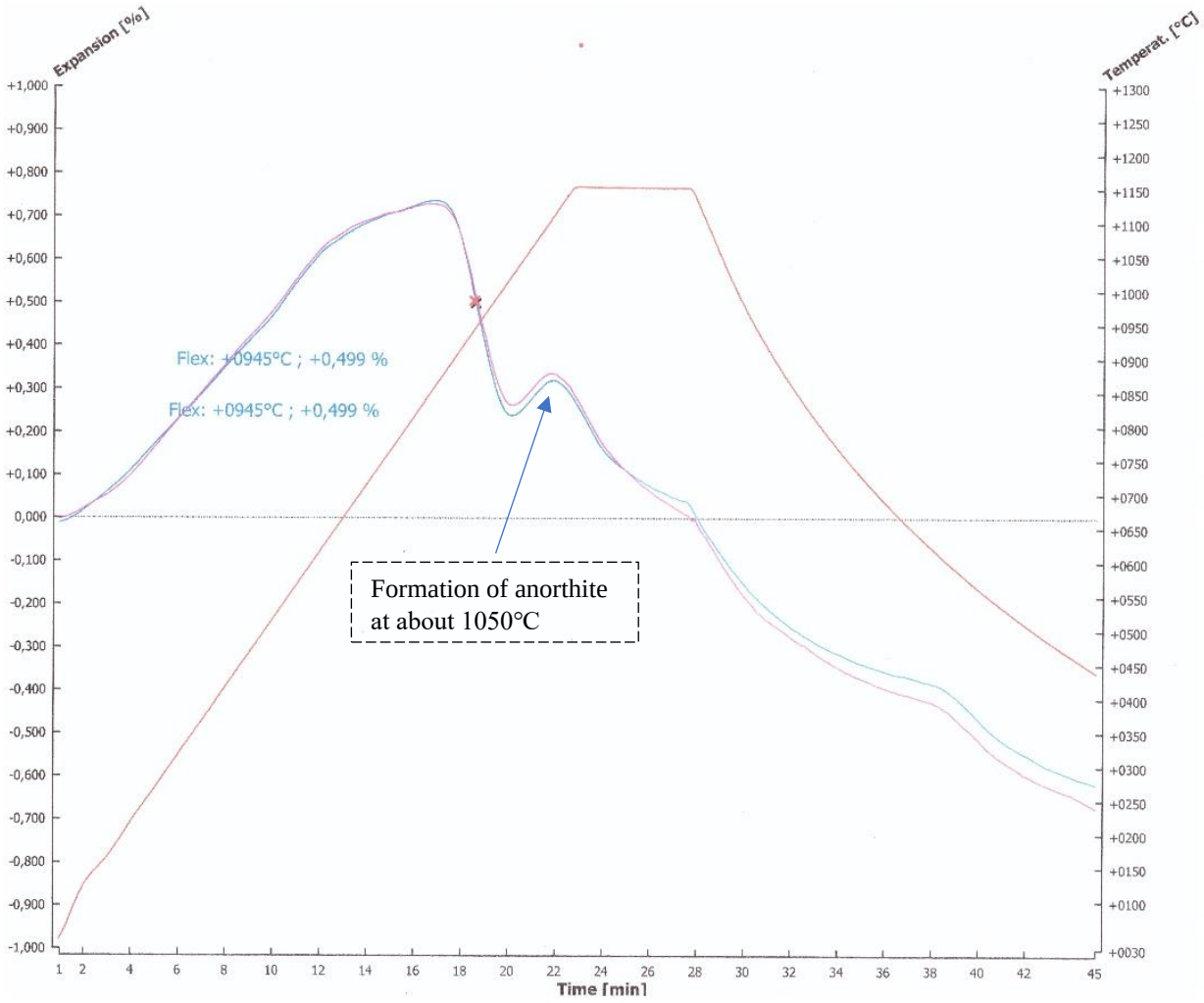

Figure 4. Sintering curves of the samples

3.6. Microstructural analyses (SEM)

The microstructure analysis was performed using the backscattered electrons in order to be able to compare between the matrix and the phases and to determine the phase distribution. Figure 4 shows microstructural images of polished fired STD and D1 bodies. Small spheroidal crystals which contain calcium oxide, aluminum oxide, and silicate are anorthite and gehlenite crystals (Tarhan M.et al.2016, Aydın T. et al., 2019). Dispersed residual particles are quartz crystals, which are characterized by the presence of peripheral cracks around them. Cracks resulted from microscopic stresses between the glassy phase and quartz crystals. As seen in Fig. 4, the open porosities resulted from $CaCO_3$ decomposition and pressing (Tarhan M.et al.2016, Aydın T. et al., 2019). With the reduction of the nozzle disc diameters, increase in slip pressure, and increase in the amount of grains between 125 and 400 µm, contributed to an increase in the degree of mold filling. Therefore, an increase in the degree of mold filling contributed to an increase in trapped density and vibrated density. As can be seen from the microstructural images, an increase in trapped density and vibrated density provided a higher bulk density for the D1 body than the STD body.

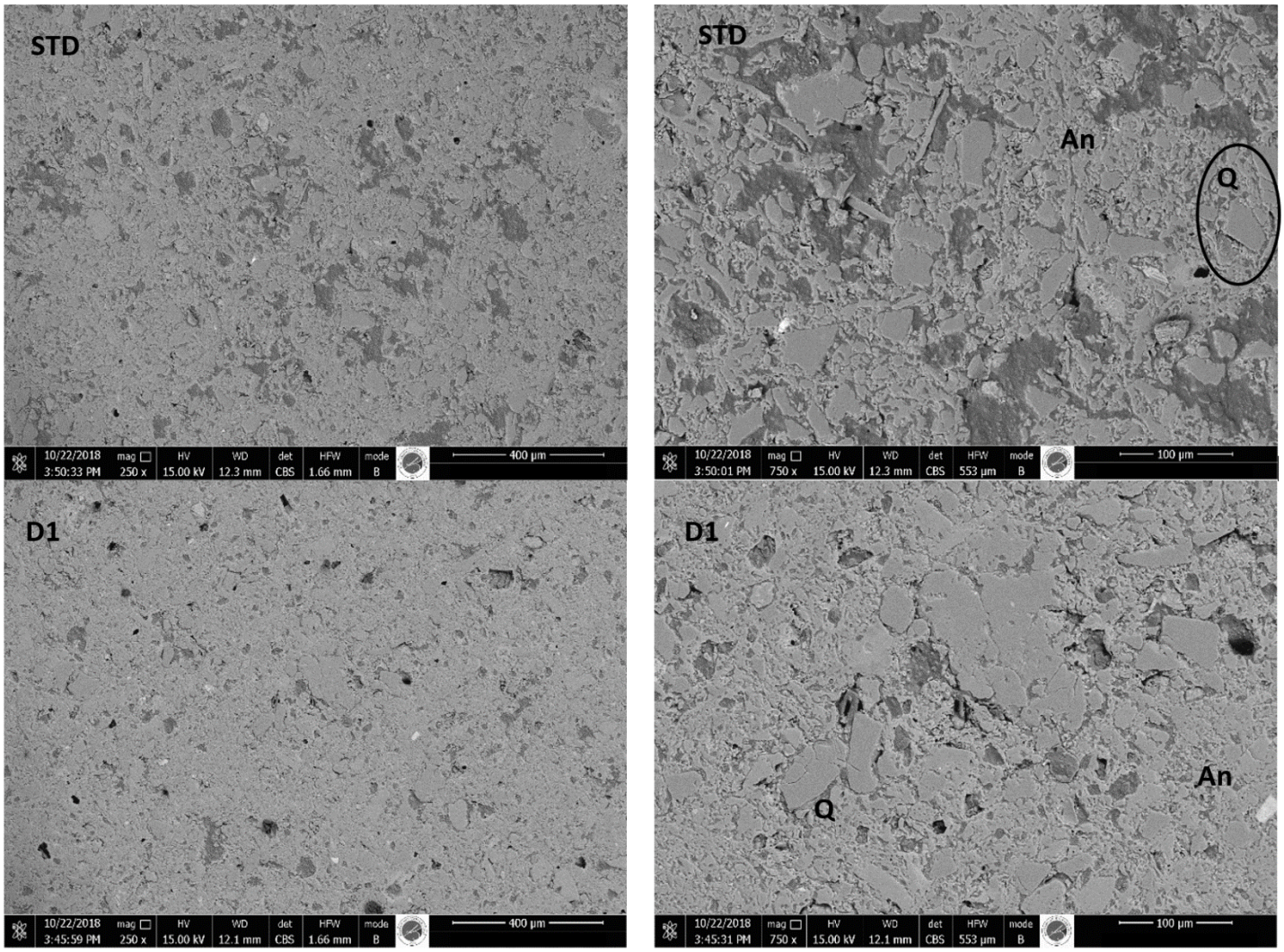

Figure 4. Microstructural images of fired STD and D1 ceramic wall tile bodies. Q: quartz, An: anortithe

4. CONCLUSION

The study investigated the effect of granule size and distribution on the degree of mould filling and on the properties of ceramic wall tile bodies under industrial conditions. In order to increase the degree of mold filling, the slurry pressure was increased from 14 bar to 18 bar and the nozzle diameters of 2.5, 2.2, and 2.0 mm were reduced to 2.2, 2.0, and 1.8 mm respectively. These changes led to an increase in trapped density and vibrated density. An increase in trapped density and vibrated density resulted in an increase in bulk density, dry strength, and fired strength. Water absorption, total porosity, and firing shrinkage also slightly decreased. These changes

improve the production efficiency of ceramic wall tile. The ceramic tile production parameters in Usak Seramik Company have been reorganized according to the data obtained from this study and production still continues in this way.

ACKNOWLEDGMENTS

The authors would like to thank Usak Seramik A.S.

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