

ALKALI FELDSPAR GRANITE FROM THE SARIHACILI (YOZGAT) REGION: USE IN WALL TILE CERAMIC BODIES AND EFFECT ON MOISTURE EXPANSION VALUE

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1. ABSTRACT

Yozgat (Central Anatolia, Turkey) is a good place for felsic magmatic rocks, which can be used as a replacement of feldspars in the production of ceramic wall tile. Metamorphic rocks, Palaeozoic to Mesozoic in age, form their basement, and granitoidic batholith of the Late Cretaceous intruded into said basement.

One component of the granitoidic batholith is the "Sarihacılı alkali feldspar granite". Its petrographical composition varies from alkali feldspar granite to quartz diorite. It is geochemically classified from tonalite to granite. The main mineral paragenesis is as follows: *quartz + alkali feldspars + plagioclase (An₈₋₃₂) + biotite ± muscovite ± sphene ± apatite*. Granular, porphyritic and granophyric textures are identified in these rocks.

Around the Sarihacılı, Salmanfakılı and Topçu villages, granitic rocks are represented by the altered leuco-granite, aplogranite/micromonzonites, and quartz-porphyrries as small dikes cutting the other rocks. All these leucogranitic rocks are the main subject of this presentation.

Anorthite is the crystalline phase (CaO.Al₂O₃.2SiO₂) formed in wall-tile structures during the firing process. This occurs due to the reactions of alumina silicate (Al₂O₃.SiO₂)

from clay and kaolines, and the CaO comes from the decomposition of carbonates at temperatures between 1100-1150°C. The given amount of anorthite should be formed so the tile has certain strength after firing. Undesirable moisture expansion develops in three ways: **a)** volumetric expansion resulting from the hydration of remaining CaO without entering into the reaction with the other phases, **b)** expansion caused by the contact of relict amorphous alumina silicate with the moisture, and **c)** the reactions of amorphous glassy phase with water. The high moisture expansion gives rise to tile deformations and even causes cracks and dismantling from paved areas.

In this study, we have focused on the relationships between the moisture expansion and alumina (A) / silica (S) and potassium (K) using by the Al_2O_3 - SiO_2 - K_2O thermodynamic system. Alternative experiments with K-dominated raw materials have been carried out on wall-tile compositions. Then, effects of different compositions on moisture expansion were investigated. In light of these experiments, the effects of recipe compositions, microstructure with the newly formed crystal phases, the development of the anorthite phase, the alkali contents and the free alumina silicate structures on the moisture expansion of the wall tiles are documented. Additionally, microstructure designs providing the minimum moisture expansion values are also defined.

The CaO content was kept constant, and the Sarihacılı granite was added to the recipes in increasing amounts. These processes were continued until the components well reacted with each other and the content of glassy phase was minimum. Consequently, we characterized the best recipe that would guarantee that the moisture expansion value would be below 0.02%. In accordance with decreasing the moisture expansion, thermal expansion values also improved.

2. INTRODUCTION

Ceramic wall tiles are made by mixing certain proportions of raw materials, such as clay, kaolin quartz and calcite. These are porous indoor pavement materials with a water absorption value of over 10%, which is formed with 4-7% moisture and obtained by fast firing (Escardino, 1993; Sandoval and Ibanez, 1999). They are manufactured according to the TS EN 14411 Annex L Group BIII standard. In traditional ceramics production, it is very advantageous, in terms of transportation costs, to supply from the region where the main raw materials are located. Currently, transportation costs of body-forming raw materials are 2-3 times the raw material costs. Therefore, it is important to investigate the local raw material potential in the area where the production plants are located. In this study, as preparation for the production of wall tiles at the Yozgat (Yerköy) factory, the potential use of Sarihacılı granitic rocks located close to the factory and other raw materials of the region has been studied. There is no kaolinite source near the factory for the production of monoporosa in the region.

Wall tile demand and expectations are increasing in the market. Customers request larger-sized wall tiles and thinner products. Therefore, it is necessary to develop appropriate masses suitable for this type of production. One of the most important parameters is the moisture expansion for the large-sized, thinner wall tiles. If the moisture expansion is high, cracks will form in the glaze of the wall tiles after a certain time of usage. Due to this, it is expected that the value of moisture expansion in wall tiles is very low, even none at all.

In this study, by using hard and soft (altered) parts of Sarıhacılı granite together with other regional clays, low-moisture expansion body designs have been carried out in conventional and dry production processes.

3. GEOLOGICAL SETTING

The study area is located near the Yozgat city of Central Anatolia, Turkey (Fig 1). This area is known as the Kırşehir continental block (KCB) or Central Anatolian Crystalline Complex (CACC) (Şengör and Yılmaz, 1981; Göncüoğlu et al, 1991; Okay and Tüysüz, 1999). The basement of the KCB and/or CACC is represented by metamorphic rocks of Paleozoic to Mesozoic age, together with Cretaceous ophiolitic rocks. KCB is bound by the İzmir-Ankara-Erzincan suture belt at the North, and the Inner Tauride suture at the SW, South and SE (Fig 1). The upper "cover" units of the KCB are late Cretaceous igneous and volcanic rocks with a minor amount of sedimentary rocks of the same age, and Middle Eocene volcano-sedimentary rocks together with the Neogene continental sedimentary rocks.

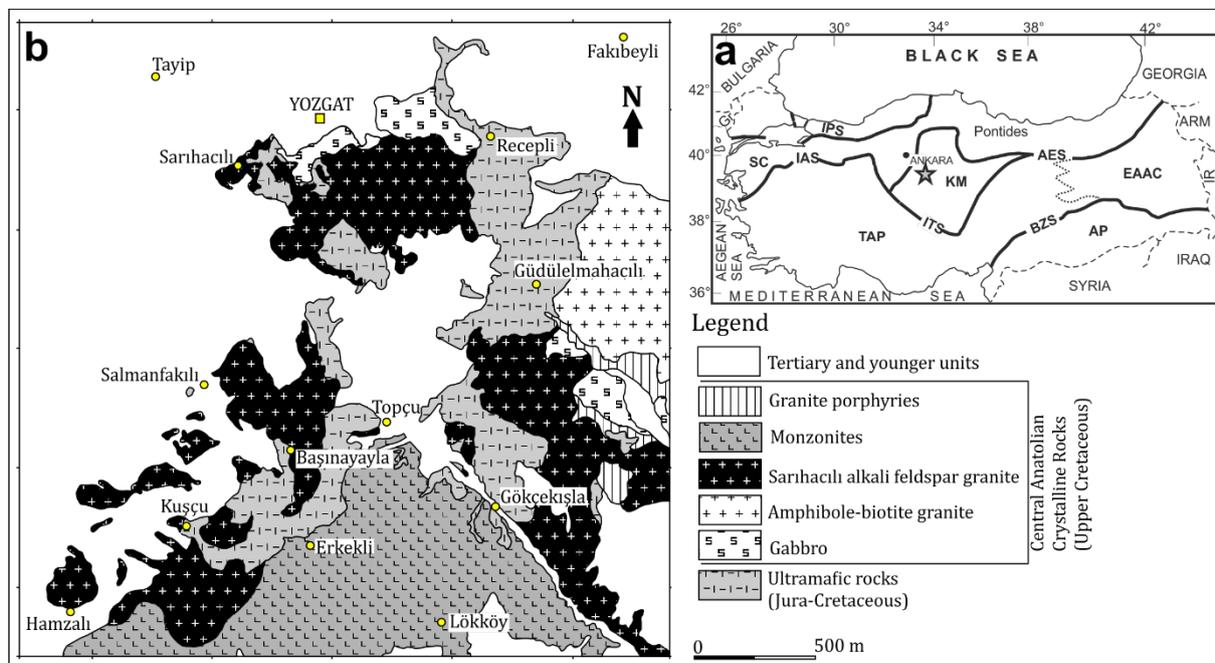


Fig 1a. Main tectonic units and suture belts of Turkey (star indicates the location of the study area), and simplified geological map of the Yozgat-Hamzalı-Lökköy area (b) (after Akçe, 2010 and MTA, 2008; coordinates are UTM European ED 50 zone 36N). Abbreviations: Gr: Greece, IR: Iran, ARM: Armenia; IPS: Intra-Pontide suture, IAS: İzmir-Ankara suture, AES: Ankara-Erzincan suture, ITS: Inner-Tauride suture, BSZ: Bitlis-Zagros suture, SC: Sakarya continent, TAP: Tauride-Anatolide Platform, AP: Arabian Platform, EAAC: Eastern Anatolian Accretionary Complex, KM: Kırşehir massif).

The late Cretaceous plutonic rocks are mainly monzonite, granite and syenitic in compositions (Boztuğ, 1995; Eler and Göncüoğlu, 1996; Güleç et al, 1996; Akçe, 2003, 2010; Akçe and Kadioğlu, 2009). Rare gabbroic intrusions are also found in this magmatic series. Sarıhacılı granitic rocks are one of the main bodies within this magmatic suite, which is the main subject of this study. Their petrographic composition varies from alkali feldspar granite to quartz diorite. They geochemically classified as granite using the Cox et al's (1979) total alkali versus silica plot (not shown here). Main

mineral paragenesis is as follows: quartz + alkali feldspar + plagioclase (An₈₋₃₂) + biotite ± muscovite ± sphene ± apatite. Granular, porphyritic and granophyric textures are identified in these rocks.

Around the Sarihacılı, Salmanfakılı and Topçu villages, Sarihacılı granite includes altered leuco-granite, aplogranite/micromonzonites, and quartz-porphyrines as small dikes cutting the other rocks. These rocks do not have mafic minerals, therefore, they are light-coloured and poor in iron, calcium and magnesium elements. Contrary to this, they are rich in alumina, potassium and sodium, as seen the Table 0.

Sample	Coordinates		SiO ₂	Al ₂ O ₃	TiO ₂	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	LOI	SO ₄	Total
	x	y											
Y36	652765	4397412	76.59	13.21	0.16	0.83	0.77	0.17	2.43	5.19	0.62		99.97
Y37	652925	4396825	72.38	14.20	0.22	1.80	0.84	0.54	1.99	4.58	3.03	0.63	99.58
Y36_Oz	652767	4397414	77.26	12.95	0.23	0.72	0.41	0.18	2.00	5.30	0.84		99.89
Y37_Oz	652927	4396827	77.33	13.37	0.25	1.04	0.19	0.40	0.12	4.43	2.66	0.24	99.79
Y39	645978	4394104	76.76	13.83	0.12	0.41	0.08	0.10	1.54	5.71	1.46		100.10
Y45	644302	4390385	74.60	14.77	0.23	0.75	0.21	0.25	2.71	5.16	1.25		99.93
Y45_Oz	644304	4390387	75.25	14.65	0.30	1.21	0.20	0.28	2.03	3.93	2.08		99.93
Y91	652725	4398132	76.17	13.51	0.21	1.44	0.27	0.15	1.05	4.82	2.31	0.05	99.98
Y92	652444	4398383	79.23	12.60	0.12	0.57	0.20	0.29	0.76	4.39	1.77		99.93
Y93	645969	4394098	77.81	13.47	0.05	0.49	0.06	0.12	1.13	5.43	1.42		99.98
Y94	645971	4394099	75.78	13.99	0.13	0.82	0.19	0.17	1.25	6.08	1.53		99.94
Y95	645975	4394100	76.86	13.35	0.15	0.48	0.08	0.09	2.36	5.41	1.21		99.99
Y96	644316	4390416	72.51	15.30	0.33	1.82	1.03	0.35	2.90	4.25	99.84		99.84

Table 0. Geochemical analysis results of Sarihacılı granite (coordinates are in UTM European ED50 zone 36).

3.1. MATERIAL AND METHODS

Wall tiles are clay-based indoor pavement materials with high porosity and water absorption. The firing of the industrial single firing diver arose (monoporosa) materials in a process under temperature conditions of 1135 to 1155°C, covered from 35 to 55 minutes. This material was produced by the mixture of clay, kaoline, calcite and quartz in given compositions. The production process was provided by the wet and dry systems. In the traditional wet system, raw materials were separated into 3 different groups based on their grinding properties. Clays were fed into a mixer called blunger, to eliminate organic particles such as coal. Marble was ground separately, as it is a relatively soft material, to decrease its particle size in the mill; this was referenced as "Hard-1". The other raw materials were ground together in the mill. These were referenced as "Hard-2", as shown in Fig. 2.

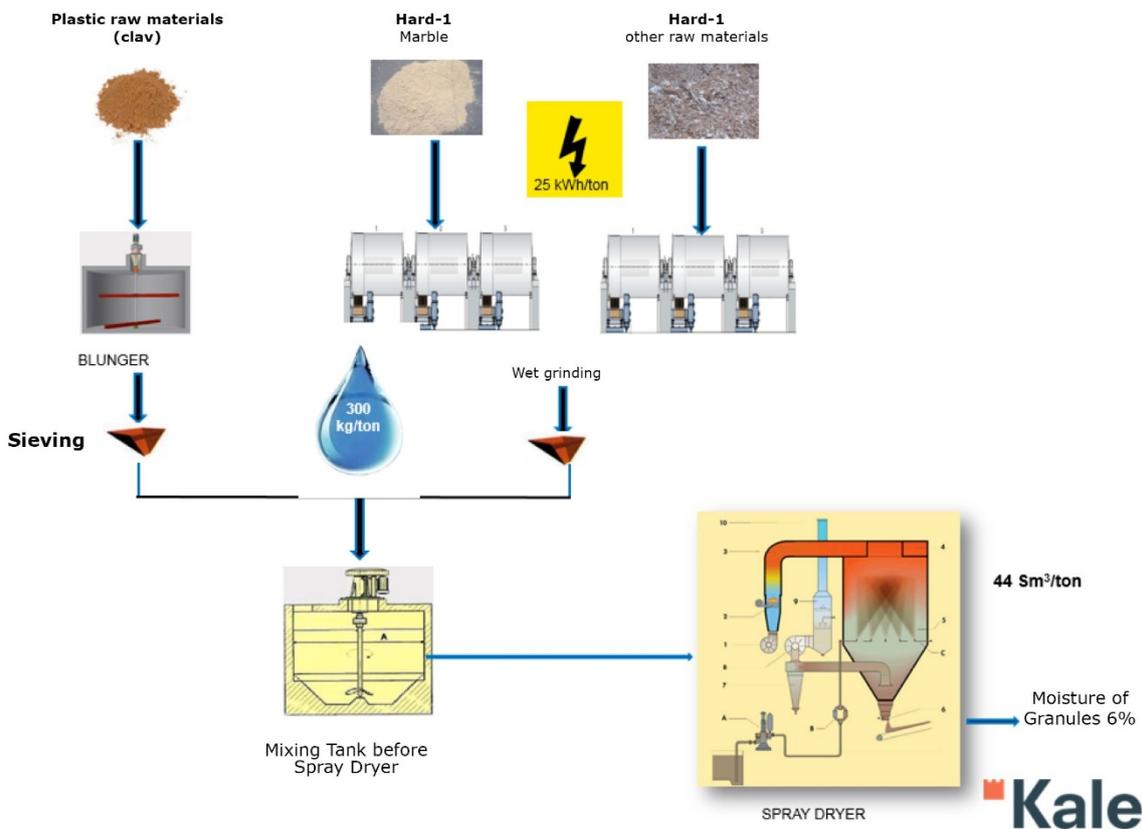


Fig 2. Flowchart for the conventional wet granule preparation process.

The raw materials for monoporosa production are the following: Clay-1, 2: Istanbul-Şile, Kaoline-1: Elmahacılı kaoline; Kaoline-2: Kırkpınar kaoline; Granite-1, 2: Sarıhacılı granite; Clay-3: limy clay, Clay-4: illitic clay; Calcite-1: Yozgat. These raw materials, except for Clay-1, 2 (Istanbul, Şile), are supplied from the Yozgat-Yerköy area.

The recipes for the monoporosa production process are prepared as shown in Figure 2. This process is formed from the stages of mud preparation (grinding, blunger, mixture and sieving), granulation, pressing, drying, glazing and firing. Raw materials weighed at given composition ratios were milled at a density of about 1650 g/lit and a rate of 3.5-4% above 45 µm. The prepared mud mixture was dried at 110°C in etuve, and then turned to dust by grinder. This dust-size material was moistured at a 6.5-7%

proportion and granulated to be ready for pressing. Granulated material was formed under pressure conditions of 325 kg/cm², and then fired under monoporous kiln conditions. The dry strength of the tablets is measured by the Gabrielli instrument. Chemical analyses of used raw materials and mixed recipes are carried out by XRF method. The Netzch 402 EP dilatometer instrument is used to determine the thermal expansion coefficient.

3.2. RESULT AND DISCUSSION

3.2.1 RAW MATERIAL CHARACTERISATION

In order to fill this gap, considering the geological features of this area, some recipe experimentations have been carried out for the Al₂O₃-SiO₂-K₂O system. The raw materials characterisations for these recipes are given in Tables 1, 2, 3 and 4.

Raw Materials	LOI	SiO ₂	Al ₂ O ₃	TiO ₂	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	SO ₄
Kaoline 1	5.34	64.48	23.48	0.57	4.04	0.19	0.06	0.27	1.08	0.2
Kaoline 2	2.92	77.61	13.62	0.15	1.17	0.42	0.28	0.11	3.65	
Clay 1	6.63	64.57	20.97	1.12	2.7	0.14	1.01	0.15	2.54	
Clay 2	6.48	65.99	20.49	1.17	2.02	0.14	0.79	0.13	2.67	
Clay 3	13.56	44.2	14.33	0.59	6.74	13.79	2.9	1.16	2.69	0.85
Clay 4	9.38	59.73	23.33	1.07	4.3	0.52	0.43	0.29	0.8	
Sarihacılı Granite 1	0.45	75.91	13.8	0.14	0.7	0.69	0.16	3.54	4.51	
Sarihacılı Granite 2	1.77	79.23	12.60	0.12	0.57	0.20	0.29	0.76	4.39	
Calcite	41.38	2.69	0.85	0.11	0.32	53.66	0.31	0.07	0.14	

Table 1. Chemical compositions (wt%) of used raw materials (LOI: Loss on ignition).

Raw Materials	Firing shrinkage %	Water absorption (%)	L	a	b
Kaoline 1	-2.67	19.41	72.37	8.25	11.37
Kaoline 2	0.16	15.25	77.25	8.58	14.58
Clay 1	5.80	7.70	70.48	9.12	21.92
Clay 2	5.21	8.96	82.58	2.14	15.85
Clay 3	1.4	20.89	61.11	7.67	24.44
Clay 4	11.89	14.10	64.48	6.34	31.53
Sarıhacılı Granite 1	0	21.94	74.73	8.68	17.28
Sarıhacılı Granite 2	-0.1	17.55	88.11	2.79	13.58

Table 2. Firing shrinkage, water absorption, firing colour values of raw materials (Firing conditions: 40 minutes at 1150°C).

Raw Materials	Minerals
Kaoline 1	Quartz, Pirophyllite, Kaolinite, Alunite, Illite, Hematite
Kaoline 2	Quartz, Illite 1, Illite 2, Albite, Kaolinite
Clay 1	Quartz, Illite, Kaolinite, Anatas
Clay 2	Quartz, Illite, Kaolinite, Anatas
Clay 3	Calcite, Quartz low, Albite, ordered Illite-2\ITM\RG#1[NR Montmorillonite-15A, Gypsum, Microcline, Kaolinite 1Md
Clay 4	Kaolinite, Tridymite, Sanidine, Cristobalite
Sarıhacılı Granite 1	Quartz, Microcline, Muscovite, Albite, Saponite, Al-Chabazite
Sarıhacılı Granite 2	Quartz low, Orthoclase, Albite (calcian), ordered Monmorillonite (bentonite), Muscovite-3\ITT\RG, Kaolinite 1Md
Calcite	Kalsit

Table 3. Mineralogical composition of raw materials.

3.3.2 RECIPE EXPERIMENTATION AND CHARACTERISATION

Alternative experiments with Sarihacılı granite as a K-Feldspar source in wall tile mass compositions are found under this heading. The effects on the moisture expansion of each recipe composition are studied. In this context, the effects on the moisture expansion of the crystal phase occurrence, the development of the anorthite phase, and the amount of alkalis and free alumina silicate textures depending on recipe composition are obtained.

The first step was the study of the available monoporosa recipe. The following stage was forming the new recipe composition by using the alternative clay source and the Sarihacılı granite (as K-Feldspar source), according to the K-Al-Si system. Compositions formed are listed in Tables 4 and 5. The physical aspects of the recipes are shown in Tables 6 and 7, and the thermal expansion values are presented in Tables 8 and 9. In addition to these, microstructural and chemical analyses of the crystalline phases have also been carried out; however, they are not shown here.

Recipe name-Information			Unit	D1	D2	D3	D4	D5	D6	D7	D8
Raw Materials	1	Kaoline 1	(%)	11.5	11.5	11.5	11.5	11.5	10	10	10
	2	Kaoline 2	(%)								10
	3	Marble	(%)	11.5	10	10	10	10	10	13	13
	4	Granite 1	(%)	22	22	22	22	22	22	22	
	5	Granite 2	(%)	20	21.5	21.5	21.5	21.5	23	20	32
	6	Clay 1	(%)	20	20	20	20	20	20	20	20
	7	Clay 2	(%)	10	5	-	10	5	-	15	15
	8	Clay 3	(%)	5	10	15	5	10	15		
	9	Clay 4	(%)								
TOTAL				100	100	100	100	100	100	100	100

Table 4. Monoporosa recipe compositions.

Recipe name-Information			Unit	D9	D10	D11	D12	D13	D14
Raw Materials	1	Kaoline 1	(%)	11.5	11.5	11.5	11.5	11.5	11.5
	2	Kaoline 2	(%)						
	3	Marble	(%)	10	10	10	10	11	11
	4	Granite 1	(%)	22	22	22	22	19.5	24.5
	5	Granite 2	(%)	21.5	21.5	21.5	21.5	23	23
	6	Clay 1	(%)					20	15
	7	Clay 2	(%)						
	8	Clay 3	(%)	5	10	15			
	9	Clay 4	(%)	30	25	20	35	15	15
TOTAL				100	100	100	100	100	100

Table 5. Monoporosa recipe compositions.

Recipe name-Information		Unit	STD	D1	D2	D3	D4	D5	D6	D7	D8
Rheology Value	Na ₂ SiO ₃	(%)	2.30	2.30	2.30	2.30	2.30	2.30	2.30	2.30	2.30
	NaTPP	(%)	0.80	0.80	0.80	0.80	0.80	0.80	0.80	0.80	0.80
	Coarse sand (+ 63 μ)	(%)	2.08	2.11	2.07	2.07	1.91	2.17	1.93	2.05	2.12
	Density	(gr/lt)	1718	1685	1674	1643	1699	1690	1651	1719	1716
	Flow time	(sec.)	39	26	25	23	26	24	24	26	25
	Grinding period	(min.)		27	27	27	28	27	30	26	27
Granule moisture		(%)	6	6	6	6	6	6	6	6	6
Pres no. – Pressure		kg/cm ²	325	325	325	325	325	325	325	325	325
Firing Aspects KS 5 ALT Wall tile	Kiln Max. temp.	C°	1155	1155	1155	1155	1150	1150	1150	1155	1155
	Firing period	Min.	52	52	52	52	52	52	52	52	52
	Tablet biscuit size	mm	49.87	50.32	50.26	50.26	50.20	50.20	50.12	50.26	50.14
	Shrinkage	%	0.64	0.32	0.24	0.20	0.30	0.44	0.26	0.52	0.58
	Biscuit water absorption	%	19.00	17.7	16.8	17.90	16.40	16.50	16.30	16.10	15.30
	Colour L	L	71.13	71.53	68.67	68.58	69.96	67.71	65.82	72.13	70.42
	Colour a	a	7.62	8.48	9.82	9.69	8.63	9.94	10.63	6.45	7.02
	Colour b	b	17.56	18.30	19.78	18.82	18.90	20.08	20.58	17.13	17.53
Dry Strength	1		15.59	33.9	44.7	38.6	26.6	34.6	38.8	37.80	35.30
	2		18.35	36.2	40.3	34.3	29.5	33.1	47	33.90	38.00
	3		-	35.5	38.3	35.8	36	37.9	43.9	27.80	34.70
	4		20.85	30.1	40.7	31.6	34.2	36.7	44.7	35.30	34.30
	5		17.32	36.2	33.5	-	-	36	38	36.80	33.90
	Av.		18.03	34.4	39.5	35.1	31.6	35.7	42.5	34.32	35.24

Table 6. Physical analysis results for monoporosa recipe compositions (STD: Standard).

Recipe name-Information		Unit	STD	D9	D10	D11	D12	D13	D14
Rheology Value	Na ₂ SiO ₃	(%)	2.30	2.30	2.30	2.30	2.30	2.32	2.32
	NaTPP	(%)	0.80	0.80	0.80	0.80	0.80	0.80	0.80
	Kaba Kum (+ 63 μ)	(%)	2.08	2.15	2.14	1.91	2.09	2.06	2.04
	Density	(gr/lt)	1718	1445	1423	1446	1491	1632	1625
	Flow time	(sec.)	39	28	20	23	24	26	24
	Grinding period	Min.		28	29	29	30	27	29
Granule moisture		(%)	6	6	6	6	6	6	6
Pres no. – Pressure		kg/cm ²	325	325	325	325	325	325	325
Firing Aspects KS 5 ALT Wall tile	Kiln Max. temp.	C°	1155	1155	1155	1155	1155	1145	1145
	Firing period	Min.	52	40	40	40	40	42	42
	Tablet biscuit size	mm	49.87	49.82	49.97	49.97	49.72	49.94	50.08
	Shrinkage	%	0.64	0.74	0.44	0.44	0.94	0.50	0.22
	Biscuit water absorption	%	19.00	14.50	15.50	14.70	16.10	13.90	14.80
	Colour L	L	71.13	64.91	63.33	63.21	66.90	69.57	70.32
	Colour a	a	7.62	13.10	13.15	12.15	12.23	8.99	9.07
	Colour b	b	17.56	25.54	23.90	21.91	26.13	21.97	21.44
Dry Strength	1		15.59	42.30	28.20	42.3	29.80	51.90	61.60
	2		18.35	41.40	40.00	26.2	24.40	47.90	54.80
	3		-	41.40	-	30.1	28.00	31.30	48.90
	4		20.85	-	21.00	28	33.80	42.30	52.80
	5		17.32	-	29.10	-	-	35.20	48.90
	ORT.		18.03	41.70	29.58	31.65	29.00	41.70	53.40

Table 7. Physical analysis results for monoporosa recipe compositions.

Recipe name-Information	Unit	STD	D1	D2	D3	D4	D5	D6	D7	D8
Dilatometer	300	64.86	64.87	64.07	62.07	65.07	64.37	62.53	64.56	67.07
	400	66.84	66.43	66.44	65.67	67.67	67.17	65.76	66.97	69.19
	500	69.67	68.89	69.12	69.47	70.07	70.58	69.21	69.64	72.02
	600	79.08	77.42	78.88	79.51	79.51	80.99	79.25	78.89	81.10
Moisture Expansion	%	0.049	0.021	0.024	0.025	0.022	0.026	0.030	0.027	0.051

Table 8. Contact dilatometer measurements for monoporosa recipe compositions.

Recipe name-Information	Unit	STD	D9	D10	D11	D12	D13	D14
Dilatometer	300	64.86	61.60	61.56	61.24	62.83	62.07	60.77
	400	66.84	62.77	62.55	62.82	63.44	64.42	63.19
	500	69.67	64.29	64.08	65.07	64.64	67.11	66.05
	600	79.08	71.59	71.17	73.10	71.89	75.93	74.72
Moisture Expansion	%	0.049	0.0077	0.0057	0.00185	0.0034	0.027	0.024

Table 9. Contact dilatometer measurements for monoporosa recipe compositions.

4. CONCLUSION

Presence of micro pores and free CaO, which are un-joint reactions that with time cause expansion, cracking and deformation of the glaze layer due to water absorption and expansion. Wall tiles are expected to be thinner and lighter in the future, as well as larger in size – composition designs that do not exhibit moisture expansion, even when moisture expansion is less than 0.020%, are being tried. The composition called “standard” is produced in a clay-kaolin-calcite (Ca-Al-Si) system. The purpose of this system is to both create more crystalline phases and to not release free alumina silicate and calcium oxide into the environment. In this study, in order to design wall tiles for the Yozgat factory of Kaleseramik AŞ, the two facieses of the local Sarihacılı granite were used. The first facies is the “hard-unaltered to weakly-altered” part, and the other is the “soft-altered” part of the same granitic body. The chemical compositions of these two facieses are the same. Different compositions were designed by combining the hard and soft parts of the Sarihacılı granite and different clays. These are Clay1 to 4. The first two (Clay1, 2) are from the Istanbul region. Clay-3 is the limy clay from the Yozgat region, and the last, Clay-4, is the coal underclay of the Çankırı region. It has been taken into consideration that the rheology is suitable because the masse granule can be produced in conventional (wet) system and dry system. All the recipes have also

been tested in the dry system. When the results were examined, the recipes containing Istanbul clay and limy clay combinations (in order to reduce costs) showed lowered moisture expansion to a 0.025% average. Moisture expansion values (in low density) were found to be very low in the compositions with Sarıhacılı granite, local limy clay and Çankırı underclays. Since rheology is not important in dry grinding granulation systems, the recipes will work successfully in these systems. The other technical features of all the recipes are suitable, and they may be included for wall tile production in certain arrangements. Considering the geological aspects of the region where the production centre is installed, the new recipes were designed using low-cost local raw materials (in terms of transportation costs), such as Sarıhacılı granite and limy clays.

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