Physicochem. Probl. Miner. Process., 59(5), 2023, 166262 | Physicochemical Problems of Mineral Processing

http://www.journalssystem.com/ppmp | ISSN 1643-1049

Investigation of the recycling of ceramic sludge waste from wall tile production in ceramic factory

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Abstract: In ceramic production, vast amount of green process waste is created. Green process waste in other words, sludge waste creates a big environmental problem. It can create water pollution as well as create environmental problems having a big physical change of the stored area., Therefore, the use of sludge waste in the process as raw material is vitally important. The research programme was carried out to use the green process wastes as raw materials in the recipes of various products in Kaleseramik Research and Development Center in Turkey. Firstly, the long term samples were taken in order to observe the fluctuation of the created waste. Then, the chemical and mineralogical characterization of sludge wastes was carried out by using XRF and XRD. Different recipes were prepared by using green process waste. The behavior and the phases of fired products were evaluated by using a double-beam optical non-contact dilatometer and XRD. SEM and EDX were carried out for microstructural and micro-chemical analysis. Finally; the physical, mechanical and color properties of waste added recipes; such as water absorption, linear shrinkage, breaking strength and chromatic coordinates were measured. The results showed that it is possible to develop wall tile body with suitable technological properties by using appropriate mixture of process wastes in the body composition. The usage of sludge waste eliminates environmental problems and also provides the cost advantages as a raw material input. The full results of the case study performed at Kaleseramik Factory is illustrated in details.

Keywords: ceramic tile, waste management, recycling, eco-products, green deal

1. Introduction

Significant amounts of sludge waste occur when producing ceramics in ceramic tile factories. These wastes cause adverse effects such as storage and environmental pollution. This is the most important issues that ceramic manufacturers have to deal with the significant problems due to legal restrictions and recycling policies. As of 2020, 370 million m^2 of ceramic tile products have been produced in Turkey, which indicates a raw material consumption of approximately 7,5 million tons. With an optimistic estimation, when it is taken into account that 5% waste material is produced, 370 thousand tons of waste is generated. World ceramic tile production increased by 1.7% in 2020 compared to its production of 15.827 million m^2 in 2019 and increased to 16,093 million m^2 in 2021 (Ceramicworldweb.com, 2021). Considering this upward trend and production capacity, these waste amounts creates a serious environmental problem, while the carbon footprint that emerges during the production, transportation and freight of raw materials poses a significant risk. Ceramic wastes are disposed of in accordance with the legal regulations inspected by the Ministry of Environment, Urbanization and Climate Change of the Republic of Turkey. The disposal of these wastes creates an extra carbon footprint.

There are many studies in the literature on the use of industrial wastes in the ceramics industry. For example, the use of fly ash from thermal power plants (Rajamannan et al., 2013), wastes such as granite, marble, and quartzite released during natural stone production (Torres et al., 2007), and the reuse of fired faulty sanitary ware in the ceramics industry (Tarhan et al., 2017; Tarhan et al., 2019). Additionally, Daniyal and Ahmad (2015) found that using waste ceramic tile as a replacement for natural coarse aggregates in concrete production can enhance the properties of concrete. Wattanasiriwech et al. (2009)

DOI: 10.37190/ppmp/166262

investigated the utilization of waste mud generated during ceramic tile production as a raw material in paving block production and suggested that the use of waste mud in paving block production has the potential to reduce waste and promote sustainability in the ceramic tile industry. El-Fadaly et al. (2010) examined the potential of recycling cyclone dust, sludge, and filter dust generated during ceramic tile production in the preparation of ceramic tiles and found that the addition of cyclone dust improved the properties of the base body, while sludge had a negative effect, and filter dust had little impact, suggesting that the recycling of ceramic industry wastes has the potential to conserve resources, but the type and proportion of waste used must be carefully considered. Amin et al. (2018) conducted a different waste study and demonstrated that municipal wastewater sludge contains many hazardous pathogens and toxic heavy metals, posing a significant ecological threat to the environment. The researchers investigated whether it was possible to use hazardous waste in the ceramic floor tile industry. As a result, they found that tiles produced with a maximum of 7% sludge added (for water absorption <10%) and tiles fired at 1150°C and 1100°C with 10% sludge or 5% sludge (for water absorption >10%) met standards and were recommended due to their economic and environmental benefits.

In the research on recycling of ceramic Sludge waste to produce alternative Eco-Products, some amount of waste was used in the developed recipes (Kayaci et al., 2014). Research conducted by Kayacı and his colleagues in 2019 stated that ceramic waste cake and fired shards of Ceramic Sanitary Ware can be used as a melting agent in porcelain tile bodies.

As a result, recycling and reusing ceramic wastes as raw materials in order to save on natural raw materials used in production, reduce production costs and have a more environmentally friendly policy makes a great contribution to the environment and economy. The subject of this study is to evaluate the residual filter press waste known as "cake" in the industry and to investigate its usability in wall tile recipes.

2. Materials and methods

Clay and kaolin used in wall tile bodies were obtained from Ukraine, $CaCO_3$ sources and feldspar was obtained from Çanakkale Region and the ceramic wastes subject to our research were obtained from Kaleseramik A.Ş. Chemical and mineralogical analyses of the wastes used are shown in Tables 1 and 2. Different wall tile body compositions were fed to a laboratory type jet mill and ground below 63 micrometers such that 3.0-3.6 % oversize material was allowed. Then, the ground product was dried in an oven at 110°C and the dried powder was screened through 1 mm aperture screen and humidified to 5-6%. Round shaped samples with a diameter of 50 mm were prepared in a laboratory press with a compression pressure of 340 kg/cm^2. Finally, the samples were fired at 1150°C for 50 minutes in a wall tile roller kiln at Kaleseramik. Technical properties of fired wall tile bodies are given in Table 3.

For raw material characterization, XRD PANalytical X'Pert Pro MPD diffractometer (with CuKα radiation) and XRF analysis were performed on pelleted samples with Panalytical Axios brand device. The sintering behaviour analysis of the bodies was made with the Misura Hsm ODHT 140.80 model branded non-contact dilatometer.

After firing, % shrinkage, % water absorption, bulk density and strength tests were carried out on the wall tile bodies. Colour L, a, b values were measured with a Minolta 3600 d Colorimeter device.

In order to determine the microstructures, SEM and EDX analysis were performed using the scanning electron microscope device in the Ceramic Research centre, in Eskisehir, Turkey. The samples were coated with a gold (Au)-palladium (Pd) alloy to ensure conductivity for the test.

3. Results and discussion

3.1. Waste materials characterization

Chemical analyses results revealed that cake wastes taken at different time intervals were similar in character. The results of the chemical analysis revealed that the cake residues taken at different time intervals had similar properties. For the studies, the sampling interval of the wastes was planned daily at the beginning of the study for almost six months and continues to be taken weekly throughout the year for control purposes. Chemical, mineralogical, and ceramic technical properties of 5 waste products (WS-1, WS-2, WS-3, WS-4, WS-5) collected at different times for this study are provided below. Among the major oxides, the SiO_2 components of the cakes were determined as between 64.2% and 66.00%, and the Al_2O_3 components of the cakes were around 20%. Iron oxide and titanium dioxide components of cake waste were found to be less than 1%. When the alkali oxides (K_2O, Na_2O) are examined, it is seen that they can be used in ceramic bodies. Sodium oxide and potassium oxide are commonly derived from minerals known as feldspar. These alkali oxides lower the melting point of ceramic products, reducing the temperature at which they become glassy and speeding up the formation of the glass phase. A low amount of alkali oxides in the raw materials increases water absorption and reduces shrinkage in ceramic bodies, which is undesired (Ryshchenko et al. 2009). The amount of ZrO_2 and ZnO oxides in the wastes are not beyond detrimental levels. They are assumed to originate from the glaze and engobes used in the wall tiles.

As a result of XRD analysis, it has been determined that these raw materials contain Kaolinite, Albite, Muscovite, Zircon, Quartz and Orthoclase minerals at certain rates. It is thought that kaolinite minerals come from Kaolin and clays, minerals such as albite, muscovite and orthoses generally come from feldspar sources and zircon comes from glaze preparation facilities. When the analysis was examined, it was determined that the only mineral that would not be desired in ceramic body recipes was muscovite type mica. However, since the amount of that mineral does not exceed 3%, no mica related problem is aroused.

Table 1. Chemical analysis of wastes sampled at different time intervals

Sample Code	Loi	SiO_2	Al_2O_3	TiO_2	Fe_2O_3	CaO	MgO	Na_2O	K_2O	ZrO_2	ZnO
WS-1	4.31	65.03	19.82	0.74	0.93	2.02	1	3.86	1.03	0.54	0.42
WS-2	4.1	64.2	20.55	0.8	0.99	2.36	0.91	3.58	1.23	0.5	0.33
WS-3	3.74	66.00	20.48	0.78	0.85	1.12	0.81	4.27	0.93	0.57	0.26
WS -4	4.11	65.03	20.72	0.82	0.79	1.35	0.84	4.12	1.01	0.36	0.18
WS -5	4.59	64.04	20.09	0.77	0.97	2.29	0.98	3.68	1.1	0.41	0.21

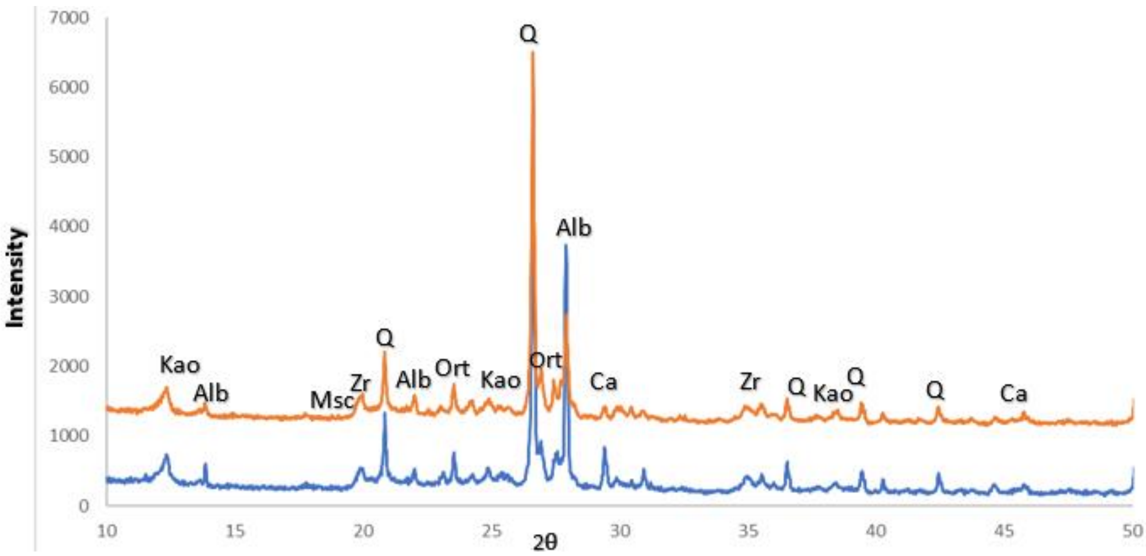

Fig. 1. XRD analysis of wastes sampled at different times. Orange Line: WS-1, Blue Line: WS-3 (Kao : Kaolinite, Alb: Albite, Msc: Muscovite, Zr: Zircon, Q: Quartz, Ort: Orhoclase)

After the characterization studies, it was determined that these waste cakes can be used as raw material in wall tiles. However, it is clearly seen that raw materials such as clay, kaolin, feldspar, calcite, which are used for different purposes in the bodies, are not exactly alike with waste. Waste cakes can be defined as a blended material obtained by mixing clay and feldspar in certain proportions due to their formation. Therefore, when defining the wall tile body recipes, it is designed not only instead of a raw material, but by mixing it with the whole body recipe at certain proportions.

3.2. Technical specifications of compositions

For this study, a combined sample was created to reflect the generality of the sampled waste cakes. After the wall tile bodies prepared with standard and waste cakes were shaped, they were fired at 1150° C for 50 minutes in a roller kiln. The % shrinkage, % water absorption, bending strength (N/mm^2), bulk density (gr/cm^3) and colour of L, a, b values of the fired bodies were determined. A combined sample of waste cakes were created by adding 1, 2, 3, 4 and 5% to the standard body recipe (W-1, W-2, W-3, W-4,and W-5). The chosen %5 of maximum usage is due to the fact that the amount of created cake allows fot the tile production capacity. (Table 2).

Table 2. Body Formulations created with waste cakes from Kaleseramik factories

Body Formulations	**STD (%)**	**W-1 (%)**	**W-2 (%)**	**W-3 (%)**	**W-4 (%)**	**W-5 (%)**
Clays	35	35	35	35	35	35
Feldspar	24	24	24	24	24	24
Calcite	11	11	11	11	11	11
Kaolin	20	20	20	20	20	20
Fired Waste	10	10	10	10	10	10
Total	**100**	**100**	**100**	**100**	**100**	**100**
Waste Added (%)	**0**	**1**	**2**	**3**	**4**	**5**

Table 3. Ceramic technical properties fired bodies

Sample Name	Shrinkage (%)	Water Absorption (%)	Bulk Densıty (gr/cm^3)	Raw Strength (kg/cm^2)	Fired Strength (kg/cm^2)	Colour		
						L	a	b
STD.	0.12	17.15	1.786	5.32	236.5	72.41	8.92	21.5
W-1	0.13	17.17	1.780	5.24	235.8	71.64	9.06	21.67
W-2	0.13	17.16	1.792	5.27	232.6	72.68	8.91	21.61
W-3	0.14	17.18	1.787	4.89	227.5	71.58	8.68	21.72
W-4	0.13	17.19	1.766	5.31	237.1	72.58	8.86	21.53
W-5	0.14	17.17	1.774	5.15	230.4	71.46	8.90	21.65

It was observed that there was no difference between the standard and waste added samples in terms of shrinkage, water absorption, bulk density, raw and fired strength tests performed in Kaleseramik R&D laboratories. Waste recipes with water absorption ranging from 17,17% to 17,19% are in compliance with the standard. Likewise, while the standard shrinkage value is 0,12%, the maximum shrinkage value observed for the waste added samples was 0.14%. As for the colour values, the L value decreased by 1 point (the colour became darker). In the case of sample W-5 prepared with the highest amount of waste those values observed; L: 71,46, a: 8,90, b: 21,65 which are quite acceptable in the ceramic use. Comparing the results of this study with previous research, El-Fadaly et al. (2010) concluded that adding similar waste to floor tile recipes has negative effects on the tile body. However, our findings suggest that there are no issues with using this waste in wall tile production. This is due to the difference in firing temperatures and regimes between floor and wall tiles.

3.3. Phase analysis of the bodies

The XRD analysis of the waste added tile bodies that are prepared conforming to the wall tile production standards are seen in Fig. 2. When the phases in these bodies are examined in detail, inert quartz from kaolin and feldspar and a small amount of orthoclase and anorthite, one of the plagioclase minerals desired to be formed in wall tile bodies, were determined. Anorthite minerals are formed as a result of the reaction of calcite minerals and clay minerals. According to the phase analysis, no undesirable phase was observed.

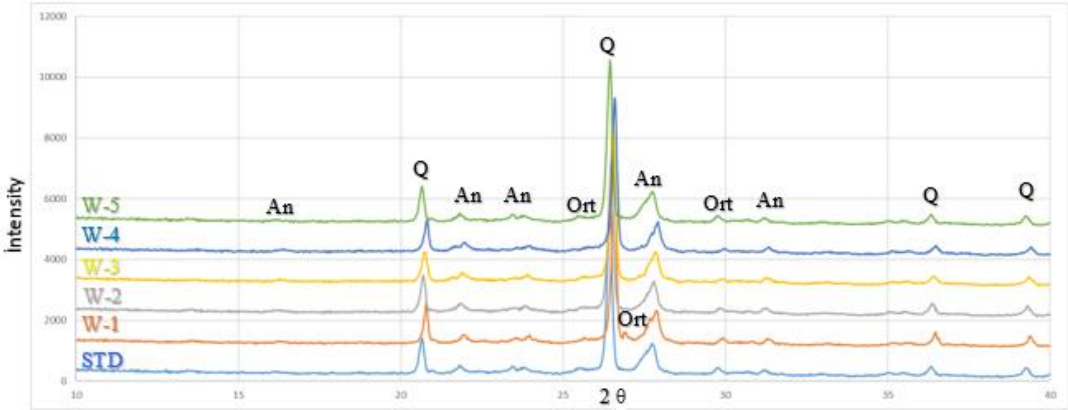

Fig. 2. XRD analysis of the fired bodys

3.4. Sintering behaviour

Comparative non-contact dilatometer analysis was performed on the waste samples whose technical properties were in compliance with those of the standard sample. The test results revealed that the flex points of the standard and waste cake including recipes (W1, W2, W3, W4, W5) were similar. In addition, it was determined that the shrinkage value did not increase with the increase at the ratio of cake (Table 3).

Table 3. Sintering behaviours of the recipes (Non-contact dilatometer)

Recipes	Flex (°C)	Shrinkage (%)
STD	1202	0.40
W-1	1202	0.41
W-2	1203	0.40
W-3	1203	0.39
W-4	1202	0.38
W-5	1202	0.39

3.5. Microstructural analysis (SEM)

SEM and EDX analysis were conducted on the STD and W-5 bodies in order to examine the microstructures in the bodies whose recipes were prepared and fired according to wall tile production conditions. The results of analyses showed that no difference was observed when the microstructures in the standard sample body visualized by SEM was compared to those in the W-5 recipe. Inert quartz and feldspars and newly formed anorthite were observed in EDX analysis to determine the chemistry of the phases (Fig. 3.)

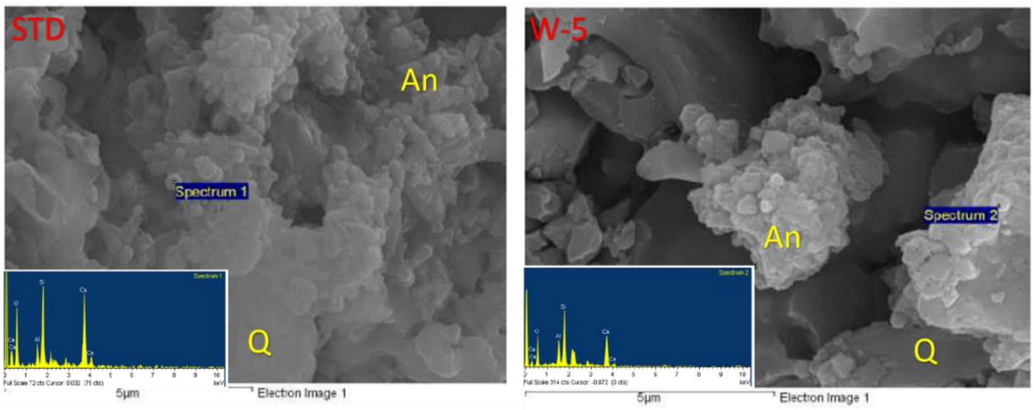

Fig. 3. Microstructure of the fired bodys (STD and W-5) and newly formed mineral phases anorthite phase and inert quartz minerals seen under SEM

When the cross-section images taken by secondary electrons (SE) are examined (200x), there is no difference in the pore distributions between the standard body and the waste added bodies. The large pores measured on the STD tile are 27-53 µm in diameter. It was determined that the W-5 coded bodies containing 5% waste cake had pore diameters between 29-75 µm (Fig. 4).

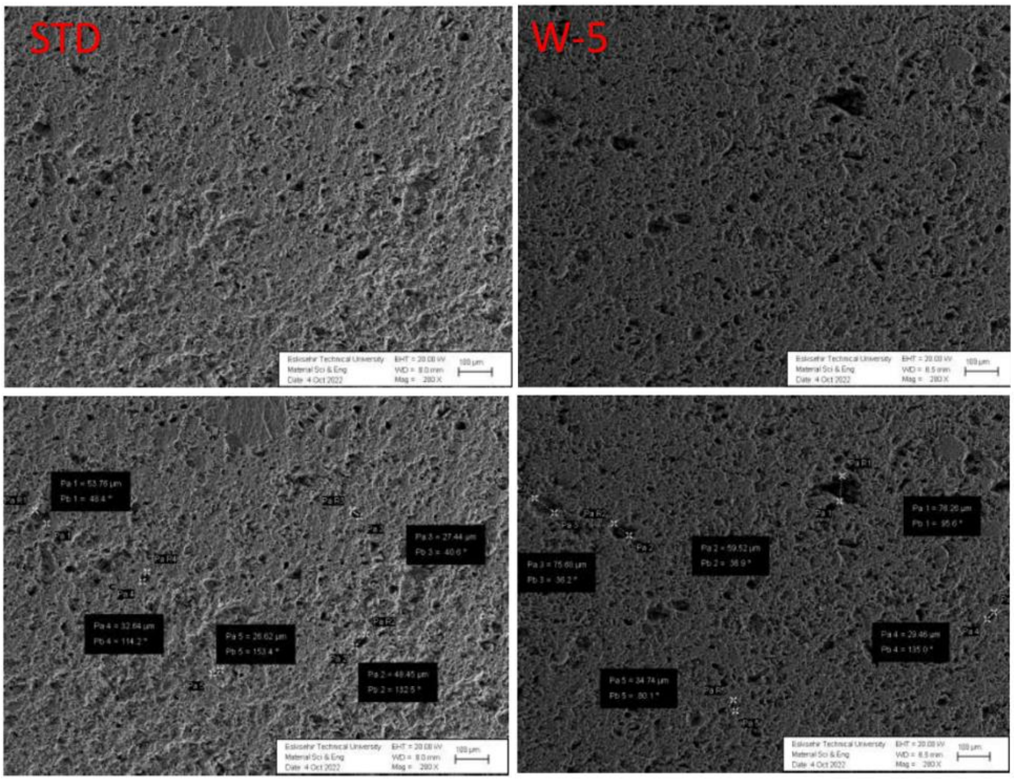

Fig. 4. Micro photos showing pore components

4. Conclusions

The study aimed to investigate the potential of using cake waste as a raw material in the production of wall tiles. The cake waste was collected at different time intervals and analyzed for chemical, mineralogical, and ceramic technical properties. The chemical analysis revealed that the cake waste contains high levels of SiO_2 and Al_2O_3, and low levels of Fe_2O_3, TiO_2, CaO, and MgO. The alkali oxides (Na_2O and K_2O) in the waste could be used in ceramic bodies, but their low amounts could result in increased water absorption and reduced shrinkage, which is undesirable. The XRD analysis showed that the waste contains kaolinite, albite, muscovite, zircon, quartz, and orthoclase minerals. The only mineral that would not be desired in ceramic body recipes was muscovite type mica, but since the amount of that mineral does not exceed 3%, no mica-related problem is aroused.

The results showed that the addition of up to 5% waste to the standard body recipe resulted in acceptable technical specifications for the wall tiles. Therefore, the study concluded that the cake waste could be used as a raw material in wall tile production, and the addition of up to 5% waste to the standard body recipe could lead to sustainable and cost-effective production. The findings of this investigation prove that, in the process of ceramic tile production, ceramic cake wastes that emerge during body preparation, glaze preparation and glazing applications, granulation studies, ceramic squaring and polishing, and treatment of wastewater are suitable to be used in wall tile bodies to some limited amounts. Ceramic cake wastes could also be used as inert materials that provide plasticity to the body. It has been demonstrated that when mineralogical and physical factors are considered, these cake wastes are a mixture of raw materials such as clay, kaolin, feldspar, and calcite used in the bodies, although their properties are not always similar. It has been found that the content of waste cakes did not change in long-term operations. The recycling of these cake raw materials will reduce the use of raw materials transported over long distances, therefore reducing CO_2 emissions and making a positive impact on the green deal, contributing to the solution of today's biggest issue.

Acknowledgments

This research has been performed at Kaleseramik Tile Factories and Kaleseramik Research and Development Center that was supported by Ministry of Science, Industry and Technology, Republic of Turkey.

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