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Recycling Waste Clay and Rice Husk Ash In The Production of Low-Density Ceramics

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Abstract: Rice husk ash is a residue that is generated when rice husk is burned. The high silica content makes this ash a waste with high economic potential. The waste clay used in this work is obtained from the processes employed at Eti Maden Bigadiç Boron Works. This study aims to recycle the waste clay and rice husk ash in the production of low-density ceramics. Rice husk ash and waste clay were characterized by chemical and X-ray diffraction analyses. The batches containing different ratios of rice husk ash and waste clay were prepared and shaped by cold pressing. The pressed samples were fired at 950, 1000, 1050, and 1100˚C. The physical properties such as water absorption, apparent porosity, linear shrinkage, and bulk density were determined. Microstructural observations were performed by using scanning electron microscopy. Based on the technological characteristics; it was observed that low-density ceramics can be produced by using waste clay and rice husk ash. This study also supports the environmentally friendly recycling of waste clay and rice husk ash, which may have a storage problem.

Keywords : Rice husk ash, waste clay, recycling, low-density ceramic

Düşük Yoğunluklu Seramik Üretiminde Atık Kil ve Pirinç Kabuğu Külünün Geri Dönüştürülmesi

Öz: Pirinç kabuğu külü, pirinç kabuğu yakıldığında oluşan bir kalıntıdır. Yüksek silis içeriği, bu külü yüksek ekonomik potansiyele sahip bir atık haline getirir. Bu çalışmada kullanılan atık kil, Eti Maden Bigadiç Bor fabrikalarında uygulanan prosesler sonucunda oluşmaktadır. Bu çalışma, düşük yoğunluklu seramik üretiminde atık kil ve pirinç kabuğu külünün geri dönüştürülmesini amaçlamaktadır. Bu çalışmada pirinç kabuğu külü ve atık kil, kimyasal ve X-ışını kırınım analizleri ile karakterize edildi. Farklı oranlarda pirinç kabuğu külü ve atık kil içeren partiler soğuk presleme ile hazırlandı ve şekillendirildi. Şekillendirilmiş numuneler 950, 1000, 1050 ve 1100˚C'de pişirildi. Pişen numuneler kullanılarak su emme, görünür gözeneklilik, küçülme ve hacimsel yoğunluk gibi fiziksel özellikleri belirlenmiştir. Mikroyapısal incelemeler, taramalı elektron mikroskobu kullanılarak gerçekleştirildi. Teknolojik özelliklere göre; düşük yoğunluklu seramiklerin atık kil ve pirinç kabuğu külü kullanılarak üretilebileceği görülmüştür. Bu çalışma aynı zamanda depolama problemi olabilecek atık kilin ve pirinç kabuğu külünün çevreci bir şekilde geri dönüşümünü desteklemektedir.

Anahtar Kelimeler: Pirinç kabuğu külü, atık kil, geri dönüşüm, düşük yoğunluklu seramik

1. Introduction

The increase of consumption and the growing industrial production lead to the exhaustion of natural resources. In addition to this, the number of industrial production wastes or sub-products are

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steadily rising and leading to environmental problems [1]. Recently, the researchers tend to study on cleaner production practices in industrial processes, considering the environmental impacts throughout the life cycle of products [2-5]. The objective of solid waste recycling is to reduce raw material consumption, so this will minimize pollution problems and treatment costs [6]. Ceramic materials consist of clays with a very wide-ranging overall composition. Because of this, clay containing wastes in considerable percentages can be used to produce these materials [7,8].

Turkey has huge boron reserves (about 800 million tons of), which consist of nearly 70% of the world's boron reserves. In western Anatolia, south of the Sea of Marmara, within an area roughly 300 km east-west by 150 km north-south, the borate deposits of Turkey occur. The main borate districts are Bigadiç, Kestelek, Sultançayır, Emet, and Kırka. The Bigadiç borate deposits are among the largest colemanite and ulexite deposits in the world. In this deposit, the high–grade colemanite and ulexite ores exist to supply the world's boron needs for many years. During the processing of boron ores, occurred concentrator wastes are stored in tailing dams [9-13].

Rice is the second largest grain produced in the world. While, in the early 1990s, rice production was around 350 million tons (annually), it has reached 661 million tons in 2008. After the rice harvest process, the milling takes place. Milling generates a by-product known as husk (a low-density residue of the process) that surrounds the paddy grain. During this process, about 80% of the weight is received as rice, broken rice, and bran while the remaining 20% is obtained as husk. Consequently, in 2008 the world rice production has also generated 132 million tons of rice husks. Disposal of rice husk is, therefore, an important issue in those countries that cultivate large quantities of rice [14]. Rice husk is organic waste and it is a major byproduct of the rice milling process. Rice husk contains nearly 20% silica in the hydrated amorphous form [15]. The production of rice husk is estimated to be 80 million tons annually. When rice husk is burned a new waste called rice husk ash (RHA) is generated. The high silica content (>92%) makes this ash a residue with high economic potential. This waste presents great potential use on large scales, such as in ceramic, cement, construction, electronics, and composite industries [16-18].

The present study was carried out: (i) to determine the characteristics of the Bigadiç Boron Works waste clay and rice husk ash and (ii) to investigate their usage in the production of low-density ceramics. This study also will minimize the environmental impact of these wastes.

2. Experimental Methods

The waste clay (BWC) investigated in this work was obtained from Eti Maden Bigadiç Boron Works, which is located in Balıkesir, Turkey. RHA was commercially obtained from Erdoğanlar Food Company. The samples were characterized by chemical and X-ray diffraction (XRD) analyses. The chemical composition of the waste clay was analyzed by Atomic Absorption Spectroscopy (GBC). The B_2O_3 component was determined by the titrimetric method at Eti Maden laboratories. The chemical composition of the RHA sample was analyzed by inductively coupled plasma mass spectrometry (ICP-MS). The phases present in the sample were identified by X-ray diffraction (XRD) under monochromatic CuKα radiation.

Preparation and testing of the samples were done on a laboratory scale. The compositions were formulated by using waste clay and RHA at varying proportions (Table 1).

Table 1. Weight percent compositions of the samples R1-R4.

Raw Materials (%)	R1 (%)	R2 (%)	R3 (%)	R4 (%)
Waste Clay	90	80	70	60
RHA	10	20	30	40

To enlighten the physical properties pellet samples were used. To produce pellet samples, each composition was dried and ground separately. The ground agglomerates were then humidified up to 6 wt. % of water. The humid powders were pressed under 150 kg/cm^2 pressure to obtain 100×50×6 mm prismatic samples. The shaped samples were dried at 120°C for 24 h and fired at 950, 1000, 1050, and 1100˚C by using a laboratory kiln (Nabertherm LH 15/14). Fired samples were used to characterize the physical properties of the materials according to TS EN ISO 10545-3 and TS EN ISO 10545-4 norms. The microstructures of the fired samples were analyzed by SEM analyses.

3. Results and Discussion

Table 2 lists the chemical compositions of the waste clay and RHA. The main characteristic of the waste clay is its high MgO, CaO, and B_2O_3 content and its low Fe_2O_3 content. RHA sample has high SiO_2 content and it also has 3.85% K_2O. XRD analysis showed that the BWC powder sample contains ulexite, smectite, illite, quartz, calcite, and magnesite phases (Figure 1). As expected from the chemical analysis results, the XRD spectrum of RHA showed cristobalite as the major crystalline phase (Figure 2).

Table 2. Chemical analysis results of the samples.

Oxides (wt%)	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	TiO_2	B_2O_3	LOI
Waste clay	29.05	2.63	0.26	14.57	17.16	1.02	0.12	0.03	9,61	25.20
RHA	90.55	0.49	0.34	0.60	0.88	0.23	3.85	0.11	-	2.10

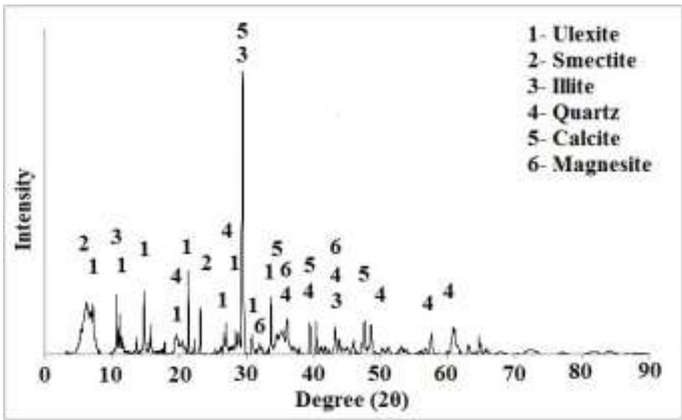

Figure 1. XRD analysis of the waste clay.

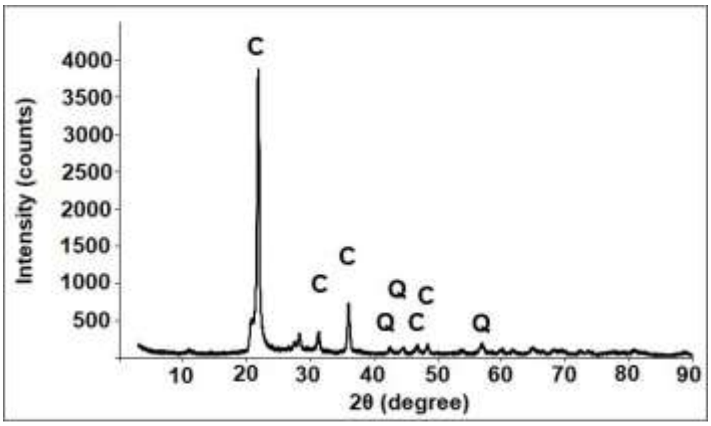

Figure 2. XRD analysis of RHA (C: cristobalite, Q: quartz).

The variations in physical properties of the samples fired at different temperatures are listed in Table 3. The results show that bulk density values decrease while water absorption, apparent porosity, and linear shrinkage values increase with increasing content of rice husk ash at the same firing temperature. As expected, an increase in firing temperature causes enhanced densification and reduced porosity. Above 1000°C one can see the decrease in the porosity and the change of structure due to the beginning of vitrification. As rice husk ash content increases, there becomes void formations that increase the volume of the open pores. The volume of the open pores affects water absorption values. The water absorption is closely related to densification.

Table 3. Physical properties of the samples fired at different temperatures. A — water absorption [%], B — apparent porosity [%], C — bulk density [g/cm3], D — linear shrinkage [%], E — bending strength [kg/cm^2].

	Temperature (°C)	A	B	C	D	E
R1	950	34,69	46,42	1,34	0,50	5,69
	1000	24,35	39,99	1,64	-3,20	13,26
	1050	5,17	12,13	2,34	-12,50	37,84
	1100	2,19	5,36	2,45	-14,80	45,32
R2	950	38,15	47,59	1,25	1,38	3,88
	1000	31,76	42,17	1,33	-1,01	10,53
	1050	15,08	27,91	1,85	-7,76	22,36
	1100	6,12	11,78	1,93	-11,61	24,82
R3	950	41,05	48,03	1,17	1,48	1,13
	1000	37,78	47,80	1,27	0,70	5,18
	1050	28,86	42,08	1,46	-2,04	6,27
	1100	15,96	26,67	1,67	-5,71	11,94
R4	950	43,87	49,48	1,13	1,60	2,13
	1000	42,91	48,75	1,14	1,00	3,05
	1050	39,89	46,52	1,17	0,70	4,81
	1100	36,01	45,22	1,26	0,10	6,68

Shrinkage is an important property for ceramics because even a small alteration in this property can lead to related changes in the mechanical and the physical properties such as effect on cracking as well as dimension quality when firing [19]. With increasing rice husk ash content the shrinkage values had higher values. It is reasonable to think that rice husk ash restrains the clay particles to coalesce. This promotes higher linear shrinkage values and also lower bulk density values. Besides, an increase in rice husk ash content enables the production of low-density ceramic products.

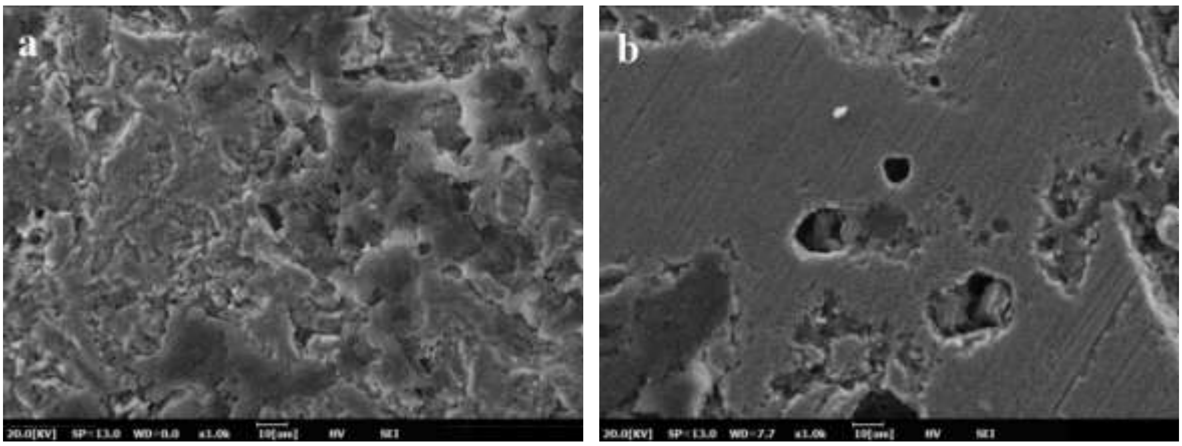

Figure 3. SEM images of R1 fired at (a) 1000°C and (b) 1050°C.

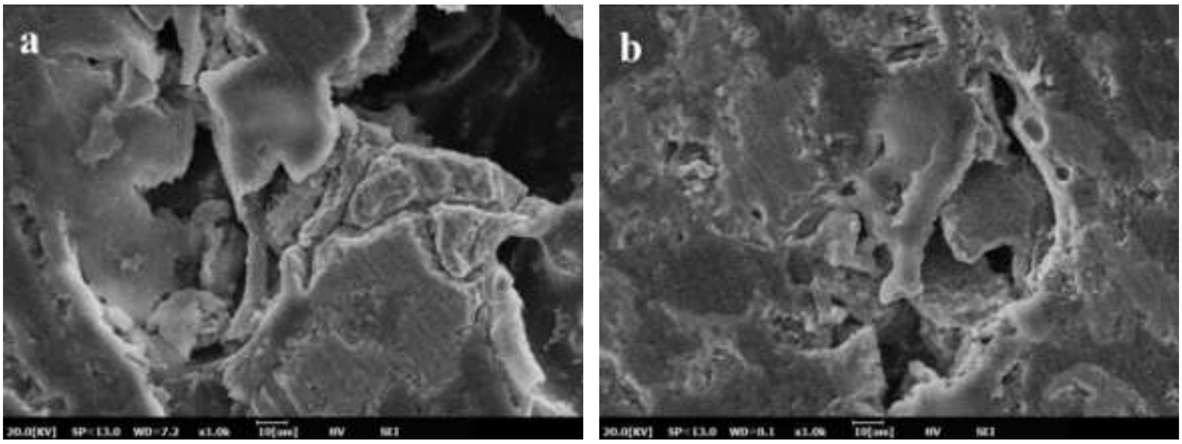

Figure 4. SEM images of R4 fired at (a) 1000°C and (b) 1050°C.

Figures 3 and 4 show the examination of microstructures of the fired R1 and R4 samples fired at 1000°C and 1050°C. SEM micrographs, taken at increasing firing temperatures, show the typical sequence of enhanced densification with increasing temperature. According to Table 3, it is reasonable to think that microstructural features remain nearly unchanged up to 1000°C. At 1000°C, the surface is rough and one can see the porosities and voids. Above 1000°C the grains of the clay minerals coalesce and porosity starts to reduce. It can be thought that a glassy phase starts to emerge above 1000°C and the porosity on the sample surface become lesser due to the liquid phase. Additionally, some pores still can be seen. A comparison of R1 and R4 samples shows that increasing rice husk ash content promotes voids and pores. This explains the water absorption, apparent porosity, and bulk density values of the samples listed in Table 3.

4. Conclusions

In this study, recycling the waste clay and rice husk ash in the production of low-density ceramics was investigated. From the results obtained, the following conclusions can be drawn:

The predominant oxides in waste clay are SiO_2, MgO, CaO, and B_2O_3. Ulexite, smectite, illite, quartz, calcite, and magnesite are found as the main phases. The analyses showed that the predominant oxide in rice husk ash is SiO_2 and it is composed of cristobalite and quartz phases.

The results revealed that increasing rice husk ash content caused higher water absorption and apparent porosity values. On the other hand bulk density and linear shrinkage values decreased with increasing rice husk ash content. An increase in the firing temperature leads to denser samples due to the improved sintering. The changes in the physical properties were slight up to 1000°C. Especially above 1000°C significant change in the physical properties was observed. SEM micrographs, taken at increasing firing temperatures, show the reduction of porosity and the progression of enhanced densification with increasing temperature. In view of these evaluations, these materials are suitable for the production of low-density ceramics.

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