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Investigation of the possibility of production of ceramic tile from tailings of the copper concentration plant

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ABSTRACT

Tailings of the copper concentration plants, which mainly consist of fin e aluminusilicate components, have many applications in silicate industry. Their application in the field of ceramic industry is the subject of the present study. Regarding the mentioned target, single soil bodies were created using the waste. The construction was done according to the certain standard procedures. Next, the constructed bodies were compared with the ones made by the main soils of the ceramic industry. Eventually, some ceramic bodies were produced by the combination of the waste and also the main soils of the ceramic industry. Determinations of the chemical and mineralogical compositions were carried out by X-ray fluorescence spectrometry (XRF), X-ray diffraction analysis (XRD) and Scanning electron microscopy (SEM). Thermal analysis was done by Thermo gravimetry (TG) and Differential scanning calorimetry (DSC). Water absorption, bend strength and linear thermal expansion were measured according to the ISO 10545-3, 10545-4 and 104545-8, respectively. The results show that the used waste is a proper raw material for the ceramic industry.

KEYWORDS

Copper waste;
Sintering;
Ceramics.

INTRODUCTION

Sarcheshmeh Copper Complex is one of the largest copper deposits in the world. Annually, more than 140000 tons of copper is extracted from the mines. In addition, 15 million tons of solid wastes are produced in the concentration unit. The produced solid wastes which mainly contain aluminusilicates are collected in the tailings dam of the Copper Complex. The mentioned deposited wastes have the potential of polluting the surrounding environment.

In recent decades, many studies have been done for the recovery and reuse of the industrial wastes around the world. Catarino et.al. (2003) used rock waste (the residue powders left from cutting and machining operations on rocks) to produce building ceramics[1]. The produced ceramics had improved mechanical performance compared to the tiles used in the construction of buildings. In another study, Cheng and Chen (2004) constructed some glass ceramics from the incinerator fly ash[2]. They showed that the optimum incinerator fly ash-based ceramics considering the physical and mechanical properties was the one sintered and heat treated at 950 °C for 2 hours.

Karamanov et.al. (2007) also investigated the usage of copper flotation waste to make glass ceramics[3]. They made two glasses containing 30wt% waste. The glass frit showed high chemical durability which was measured by the TCLP test. Haiying et.al. (2007) worked on production of ceramic tile by use of municipal solid waste incineration (MSWI) fly ash[4]. They showed that heavy metals had been cemented among the solid lattice of the product and might hardly be extracted. They suggested that the MSWI fly ash must be used just as an adding value. Coruh and Nuri Ergun (2007) studied the leaching characteristics of the copper flotation waste before and after vitrification[5]. Their results showed that the copper flotation waste was a toxic one before vitrification while levels of the leached metals after vitrification were within the acceptable limits.

As 15 million tons of solid wastes are produced in the concentration unit of Sarcheshmeh Copper Complex, the possibility of production of ceramic tile from the tailings of the copper concentration plant at Sarcheshmeh Copper Complex is investigated in the present study.

EXPERIMENTAL

Raw materials for the ceramic industry in terms of their role in the ceramic body are classified to plastic clays, fluxes and fillers. In this work A1 and A2 (plastic clays), B (flux) and C1 and C2 (fillers) are used as common raw materials for the ceramic industry. It is notable that the parent body is produced by them. The composition of each of them is given in TABLE 1.

As the tailings are stored at the tailings dam during the past years; they have been subjected to different aging and weathering conditions. Therefore, there is a necessity to characterize the waste. Due to the mentioned fact, several samples (W1, W2 and W3) were collected and were physically characterized by determination of the particle size distribution. They were also analyzed for their chemical compositions. Analyses were performed using XRF (Philips PW 1480). The results are shown in TABLE 2.

Then, equal portions of W1, W2 and W3 were mixed to make W. Mineralogical analysis of the sample (W) was carried out using XRD (PHILIPS- XPERT Anod= Co-Kalpha= 1.789010). Microstructure was examined by SEM (Smart Scan MV2300). The thermal behavior of the waste was evaluated by TG and DSC analyses. It should be noted that DSC and TG analyses were down by NETZCH STA 409 PC/PG at 10 °C/min heating rate in nitrogen atmosphere.

In the next step, some single soil ceramic bodies were made with each of A1, B, C1 and W. Their properties were measured according to the relevant standards and the results were discussed. Due to the performed analyses and tests, different percentages of the wastes were used in the final body.

To create the ceramic body, the following steps were performed. Slurry was prepared in the first step. Then it was dried at the temperature of 105Ë%C and was granulated with 5-7% of moisture. After three times of deaeration, the bodies were pressed in the 10 mm ×50 mm ×100 mm and 7 mm ×50 mm × 100 mm molds under the pressure of 8.5 MPas. To remove the added moisture, the bodies were dried at the temperatures

TABLE 1 : Chemical composition of common raw materials in the ceramics industry (A1, A2, B, C1 and C2)

	SiO_2	Al_2O_3	Fe_2O_3	S	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	MnO
A1	51.78	26.22	10.05	0.203	0.35	0.28	0.63	1.83	1.167	0.309	0.036
A2	65.92	17.59	3.79	0.713	0.85	0.61	2.26	2.74	0.486	0.467	0.087
B	77.85	12.39	9.67	0.003	0.71	0.63	3.81	1.15	0.200	0.003	0.012
C1	62.05	18.55	6.32	0.475	1.27	1.38	0.68	3.39	0.798	0.282	0.109
C2	64.76	18.02	6.21	0.002	0.46	1.04	0.96	3.12	0.798	0.188	0.088

TABLE 2 : Chemical composition of the waste samples

	SiO_2	Al_2O_3	Fe_2O_3	S	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	MnO	Mo	Cu
W1	61.41	15.98	5.61	4.11	0.98	2.58	0.73	4.20	0.25	0.10	1.10	0.023	0.80
W2	61.11	16.21	7.53	2.55	1.03	2.88	0.81	4.92	0.26	0.11	0.27	0.016	0.22
W3	59.26	15.88	6.87	1.54	1.23	3.09	1.19	4.87	0.25	0.10	0.24	0.034	0.18

of 105°C. At the final step, some of the bodies were glazed while the others were left without glazing. Then they were sintered by heating under the proper temperature regime[6-8].

The characterization of the produced ceramics was carried out according to ISO 10545. Water absorption, raw, dry and fired bend strength and linear thermal expansion measurements were done according to ISO 10545-3, 10545-4 and, 10545-8, respectively[9].

RESULTS AND DISCUSSION

The properties of the single soil bodies are given in TABLE 3. As can be seen, the waste has a high dry strength which is an important feature of the ceramic clay which increases the body capabilities for shaping and printing. Due to the obtained results, different percentages of the waste (TABLE 4) were replaced in the parent body composition.

Particle size distribution

To determine the particle size, samples were separated using standard screens. Particle size distribution curves are shown in Figure 1. The grain size of the 30 to 70 percent of the samples is below 44 microns. This would lead to the efficient pressing and sintering of the samples which cause high bend strength and low water absorption.

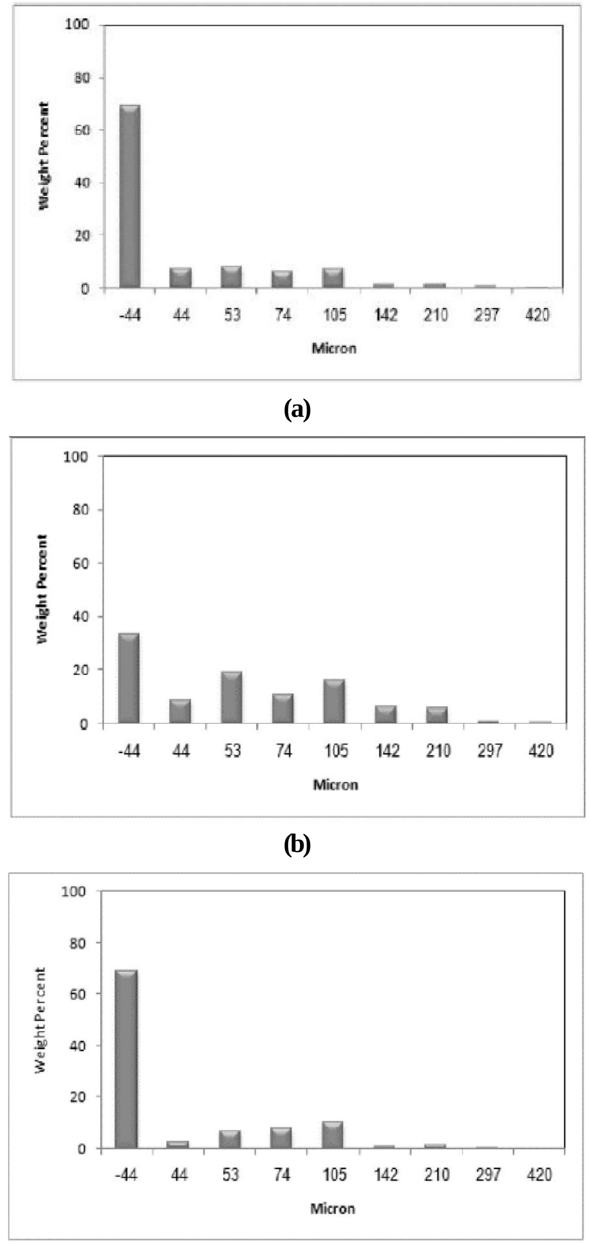
X-ray fluorescence spectrometry analysis

XRF was performed to determine the chemical composition of the tailings. Considering the percentages of the SiO_2 and Al_2O_3 and also the reasonable amount of the Fe_2O_3 and the other oxides, it seems that the waste is suitable for the manufacturing of the ceramic tiles. However, the existence of sulfur in the tail causes some restrictions for the high percentages usage of the waste in the body composition.

X-ray diffractio.0n and scanning electron microscopy analyses

TABLE 4 : Weight percentage of the body composition

	W%	A1%	A2%	B%	C1%	C2%
X	30	28	12.6	15.4	8.4	5.6
Y	50	20	9	11	6	4
Z	70	12	5.4	6.6	3.6	2.4



(a)

(b)

(c)

Figure 1 : Particle size distributions of a) W1, b) W2 and c) W3

TABLE 3 : Mechanical properties of single soil sample

	A	B	C	W
Water abs.%	6.52	18	5.07	3.29
Raw bend strength (kg/cm^2)	7.90	1<	4.96	6.60
Dry bend strength (kg/cm^2)	33.34	11.97	12.16	43.37
Fired bend strength (kg/cm^2)	525.64	115.44	521.59	412.75
linear thermal expansion (kg/cm^2)	3.27	1.66	4.61	7.06

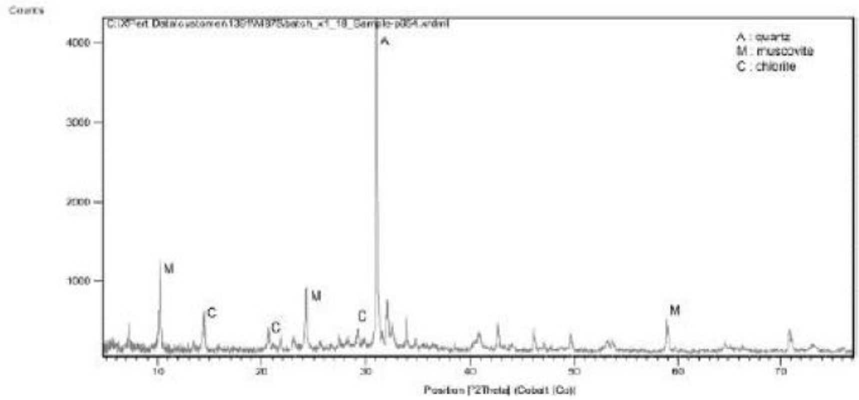


Figure 2 : The XRD image of the waste

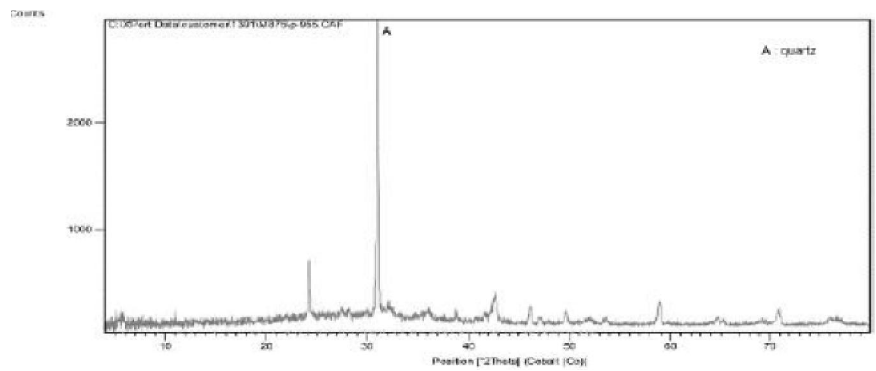
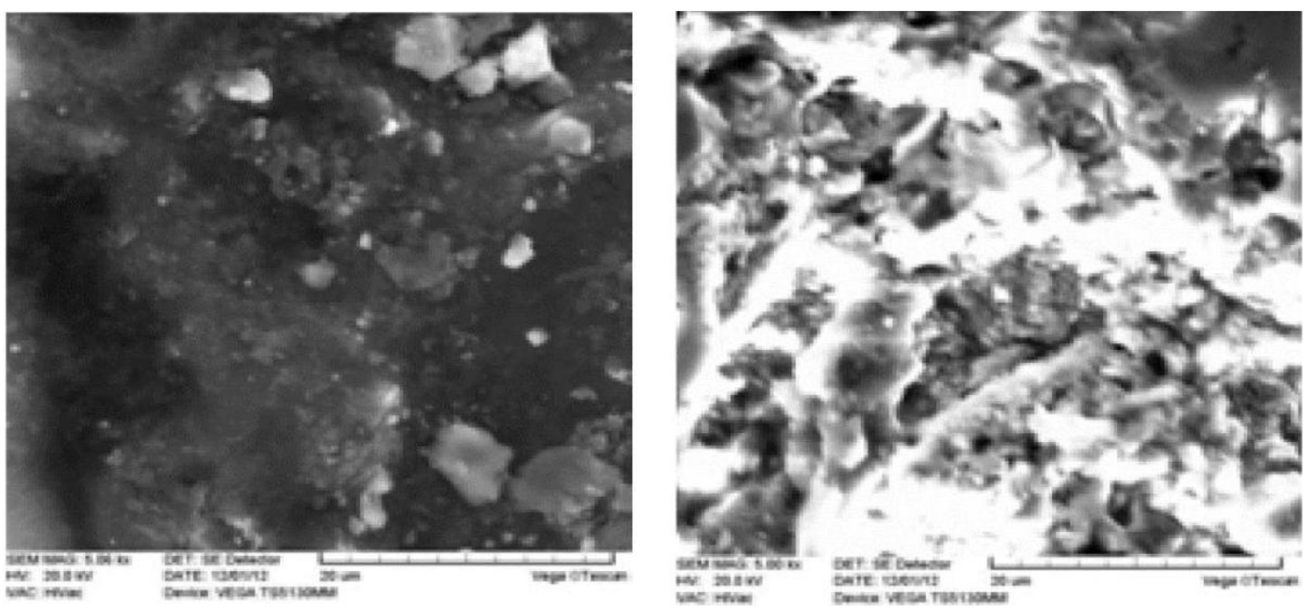


Figure 3 : The XRD image of the sintered single soil sample of W



(a) **(b)**

Figure 4 : The SEM images of a) the body made by the waste (100%W) and b) the parent ceramic body (40%A1, 18%A2, 22%B, 12%C1, 8%C2)

To determine the crystalline phases present in the tailings and also in the waste-produced fired samples, the XRD analysis was performed. The results are shown in Figures 2 and 3. The results indicate the existence of quartz, muscovite, chlorite and orthoclase, gypsum, dickite, albite and illite phases in the tailings. It should be mentioned that the existence of quartz, orthoclase and albite is one of the reasons of the high bend strength of the dry waste-produced sample.

The XRD analysis of the waste-produced fired samples shows the formation of the glassy phase and amorphous silica. Probably, the following phase transformations have occurred during the heating process[10].

$$2FeO.SiO_2+1/2O_2 \rightarrow \alpha Fe_2O_3+SiO_2$$

$$Fe_3O_4 \rightarrow \gamma Fe_2O_3 \alpha Fe_2O_3$$

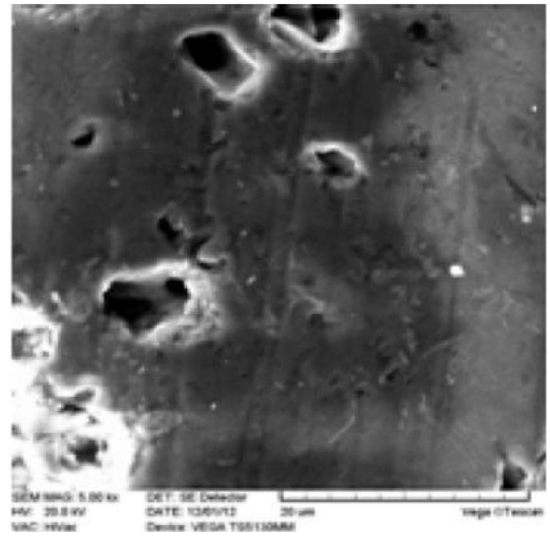
The peaks of hematite and amorphous silica, due to this transformation, have been detected in the product.

The morphology of the parent ceramics and waste-produced bodies are shown in Figure 4. The test was complemented by SEM. As can be seen in Figure 4) a, the fine particle size distribution of the waste leads to the formation of a more homogenous body. The gray background is mainly glassy phase while the white areas are detected as SiO_2. The dark zones are Fe, Al and Mn phases also.

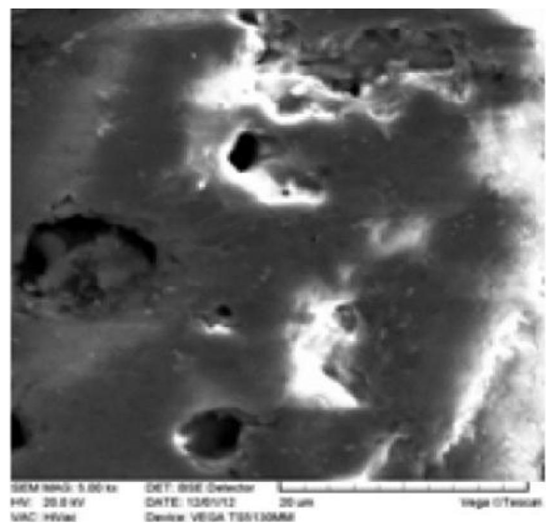
The microstructure of the X, Y and Z sintered bodies are shown in Figure 5. The formation of the high glassy phase observable in the figures is due to the efficient sintering of the waste fine particles. The mentioned sintering leads to the high bend strength and low water absorption in these ceramic bodies.

Differential scanning calorimetry and Thermo Gravimetry analyses

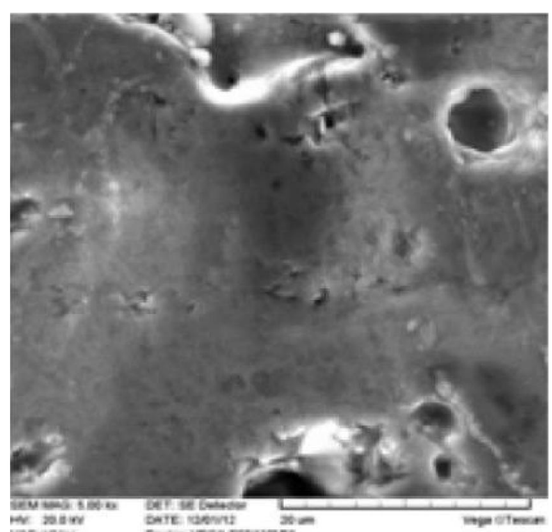
To evaluate the thermal behavior of the waste, DSC and TG analyses were done. The results are shown in Figure 6. At first, there are two stages of weight loss due to the loss of free and bound water. The mentioned losses correspond to the two endothermic peaks at 95°C and 114°C. Then there is a subtle weight gain in the temperature range of 300°C to 400°C due to the oxidation. Next, a sudden weight loss as a result of the exit of the combined water as well as the destruction of the crystal lattice is observable. Quartz deformation occurs in the same temperature range too. These changes correspond to the two endothermic peaks at 517°C and 577°C in the DSC curve. The release of SO_2 and the other gases occur up to the temperature of 1180°C which corresponds to the change of the slope of the TG curve. Above 1200°C, the melting and vitrification occur. These conversions cause no change in the TG curve.



(a)



(b)



(c)

Figure 5 : The SEM images of a) X, b) Y and c) Z

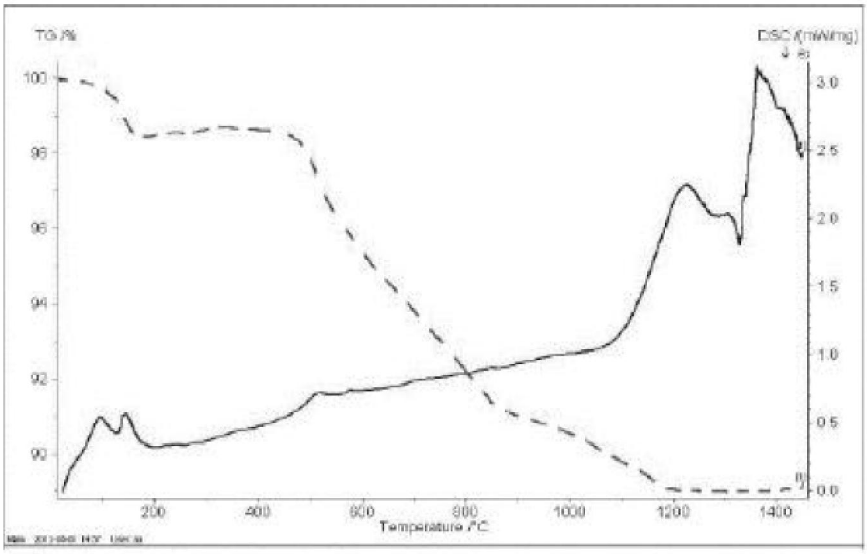


Figure 6 : TG and DSC curves of the waste

TABLE 5 : Mechanical properties of the final body

	X	Y	Z
Water abs.%	4.43	3.61	2.65
Raw bend strength (kg/cm^2)	6.45	5.59	5.93
Dry bend strength (kg/cm^2)	20.90	20.95	28.45
Fired bend strength (kg/cm^2)	504.07	544.31	484.86
linear thermal expansion	5.28	5.48	6.06
Shrinkage%	5.48	6.41	6.60

Mechanical properties

Water absorption, raw, dried and fired bend strength of the body and linear thermal expansion were measured in accordance with the ISO standards. The shrinkage of the samples was measured too. The results are given in TABLE 5. Because of the fine particle size distribution of the waste, increasing the waste percentage in the parent bodies' composition decreases the water absorption in the ceramic bodies. High percentage usage of the waste, due to the hardness of the raw waste and also its fine body pressing, leads to the high dry bend strength of the ceramic bodies. Bend strength of the fired samples are acceptable in accordance to standards. In addition, Shrinkage of the samples is in accordance with the weight loss of the waste during the firing and melting as seen in the TG curve. It should be mentioned that the obtained shrinkage is a little high. Therefore the waste should be combined with a low thermal loss and shrinkage soil to form the ceramic bodies.

CONCLUSIONS

Production of ceramic tiles from the tailings of the copper concentration plant is studied. Due to the existence of quartz, orthoclase and albite crystal compounds in the waste of the tailings dam, it is a suitable raw material for the ceramic industry. The constructed ceramic bodies have adequate strength in dry conditions. Also the fine particle size causes efficient pressing and uniform bodies. This leads to a body with proficient sintering, low water absorption and high dry and fired strength. Glazed sample have an acceptable appearance which shows the emission of SO2 before glassy temperature of the glaze. Therefore, removal of this gas does not harm the glaze. According to the characteristics of the waste, its usage reduces a large percentage of the plastic soils in the body's composition which has many benefits for the ceramic manufacturers. Therefore, by the use of this waste in the construction of the ceramics, in addition to the reduction of the accumulated wastes, a product with the acceptable properties would be achieved.

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