Recycling of Ceramic Industry Wastes in Floor Tiles Recipes

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Abstract: The purpose of this work is to assess the possibility of the recycling of some solid wastes of ceramic industry in the preparation of ceramic tiles at the same factory. Cyclone dust, sludge, and filter dust were added to the base body in proportions ranging from 2.5 to 10 % weight content. The mixed powders were pressed at 225 bar, then fired in an industrial kiln at 1190 ºC for 35 minutes cycle. The results showed that addition of cyclone dust improved the physico- mechanical properties of the base body, while sludge additions deteriorated these properties. Filter dust had nearly no effect on properties. The phase analysis via XRD and SEM demonstrated that the very rapid industrial cycle is insufficient for the complete melting of soda feldspar and the formation of any mullite crystals. [Journal of American Science. 2010;6(10):241-247]. (ISSN: 1545-1003).

Key words: cyclone dust, sludge, filter dust, wastes, recycling, ceramic, densification, mechanical properties

1. Introduction

Waste recovery is a very important topic from the public health, the environmental and industrial perspectives. The use of wastes as useful raw materials is strongly recommended since it reduces the negative environmental impact associated with landfill, minimizing manufacturing cost, and preserves non- renewable natural resources.

Many attempts were made to incorporate solid wastes into raw materials mixtures in order to produce different ceramic products. Rice husk ash and silica fume were used in the preparation of whiteware bodies [1, 2]. They lead to a marginal improvement in the body strength. Granite wastes which are produced by the granite processing industry were successfully used in the production of ceramic tiles of water absorption lower than 3 % and fired at 1200 ºC. The addition of this waste up to 35 % resulted in an increase of the modulus of rupture; however a slight increase in water absorption was observed [3]. Wastes from marble working were considered to be potentially beneficial by recycling them in the manufacture of certain ceramic products, in which calcite and dolomite are used as raw materials [4]. Several wastes such as phosphogypsum, fly ash and iron rich waste have been identified for possible mixing with clay in novel ceramic bodies intended for a water filter application. A body of 80% fly ash, 5% phosphogypsum and 10 % iron rich waste and 5 % bentonite has shown a favorable water absorption and shrinkage for employment as a filter [5]. Oily waste obtained directly from the petroleum separation process was added at levels of 5, 10, 20 and 30 % to a ceramic body composition and revealed no change of porosity [6]. Sewage sludge and steel work slag when used to formulate ceramics have improved the mechanical strength of the sintered specimens, but it can be used only where the color of the finished product is not important [7].

In fact, the recycling of waste produced from a given industry as raw materials for the same product is the most beneficial waste management method, and this is the trend of this work. Generally, ceramic tiles production has shown a great rate of increase in recent years. In Egypt, tile production was 20 million m^2 in 1996, while it reached 83 million m^2 in 2004 with an increase of more than four folds (> 400 %). In 2006, the production of Egyptian factories was about 100 million m^2 [8]. This increase in production is accompanied with a large increase of industrial solid wastes. This article aims at making use of solid wastes produced from a ceramic factory in the preparation of ceramic tiles at the same factory conditions, i.e. without changing the process parameters. This will get two targets at once; reduce the pollutants produced from ceramic industry and economical production of ceramic tiles.

2. Experimental

2.1- Preparation of raw materials

A representative sample of 10 Kg of each raw material was provided in the form of lumps. The size of these lumps was reduced through crushing by a jaw crusher, then wet ground in a ball mill. Materials were then thoroughly mixed and quartered. The fine powders were mixed with solid wastes according to the designed proportions. A slip of each batch is prepared by mixing the powders with 40 %

water. The slurry was left to dry at 110 º C. The dried cakes were crushed and dry milled.

2.2- Preparation of the test samples

The dry powders were pressed at 225 bar using 6 % water content as a binder. Rectangular specimens of 10 cm x 5 cm x 0.5 cm were prepared in order to measure the densification parameters; i.e. water absorption, apparent porosity, bulk density and linear shrinkage. Bars of 5 cm x 2.5 cm x 0.5 cm were also pressed in order to measure the modulus of rupture. Formed specimens were dried at 110 ºC for 24 h, and then fired in an industrial kiln at 1190 ºC for 35 minutes cycle.

2.3- Investigation techniques

The chemical analysis of raw materials and wastes was carried out by x- ray fluorescence using Philips equipment type (Spectrometer PW 2404 X-ray, Holland). The particle size distribution of raw materials and wastes were measured using laser particle size instrument (HORIBA LB-500). The plasticity of the base body and batches containing wastes was studied according to Pffefrkorn method. The phases of the fired specimens and their relative percentage were determined using a Philips 1730 X – ray diffractometer with Ni- filtered Cu-Kα radiation at a scanning speed of 1 /min. Scanning electron microscopy was performed on specimens that were polished and etched for 4 minutes in 2 % HF solution. Water absorption, apparent porosity and bulk density were measured according to Archimedes method. Dimensions of green and fired specimens were measured in order to calculate the linear shrinkage. Ten specimens of each batch were measured by 3- points bending test at room temperature in order to determine the green and fired modulus of rupture (Strumento MOD. MOR/ 3- E, Scalo 0+60 Kg, Sassuolo, Italy).

3- Raw Materials

3.1- Batch formulations and chemical analysis

In the present work, local Egyptian raw materials were used in the preparation of the base body; which is composed of 50 % ball clay, 20 % K-feldspar, 25 % Na- feldspar, and 5 % talc. Three solid wastes of the ceramic industry (cyclone dust, sludge, and filter dust) were added to the base body in different proportions ranging from 2.5 % to 10 %. The compositions formed after these additions are given in Table 1. The chemical analysis of the raw materials and wastes are given in Table 2. Table 3 represents the chemical analysis of the prepared batches.

Table (1): Composition of the prepared samples, (weight %).

Batch	Base Body	Cyclone dust (C)	Sludge (S)	Filter dust (F)
C0	100	0.0	0.0	0.0
C1	97.5	2.5	0.0	0.0
C2	95.0	5.0	0.0	0.0
C3	92.5	7.5	0.0	0.0
C4	90	10	0.0	0.0
S1	97.5	0.0	2.5	0.0
S2	95.0	0.0	5.0	0.0
S3	92.5	0.0	7.5	0.0
S4	90	0.0	10	0.0
F1	97.5	0.0	0.0	2.5
F2	95.0	0.0	0.0	5.0
F3	92.5	0.0	0.0	7.5
F4	90	0.0	0.0	10

Table (2): Chemical analysis of raw materials and wastes.

Constituent	Clay	Na-Feldspar	Talc	K-Feldspar	Cyclone dust	Filter dust	Sludge
L.O.I	7.99	0.97	7.20	0.48	4.24	4.47	4.0
SiO_2	66.1	66.4	47.2	71.29	64.45	63.3	65.4
Al_2O_3	19.5	19.0	7.22	15.51	19.25	20.2	18.8
Fe_2O_3	1.66	0.75	7.13	0.54	3.07	2.76	2.10
TiO_2	1.95	0.41	0.15	0.07	1.16	1.04	0.74
CaO	0.06	0.60	0.35	0.47	0.81	0.74	2.80
MgO	0.05	0.12	30.2	0.00	1.87	2.19	2.99
K_2O	0.52	0.32	0.13	7.71	1.28	1.24	1.57
Na_2O	0.03	10.9	0.03	3.41	2.70	2.97	2.57
MnO_2	0.03	0.03	0.11	0.03	0.04	0.04	0.03
P_2O_5	0.00	0.03	0.00	0.00	0.57	0.47	0.46
Cl^-	0.16	0.06	0.05	0.08	0.00	0.00	0.00
SO_3^{--}	1.44	0.18	0.09	0.08	0.00	0.00	0.00
Total	99.7	99.8	99.9	99.67	99.44	99.5	99.58

Table (3) : Chemical analysis of the base body and prepared batches.

Constituent	Base Body	C1	C2	C3	C4	S1	S2	S3	S4	F1	F2	F3	F4
L.O.I	4.70	4.69	4.68	4.67	4.65	4.68	4.67	4.65	4.63	4.69	4.69	4.68	4.68
SiO_2	66.27	66.23	66.18	66.13	66.09	66.25	66.23	66.21	66.18	66.2	66.12	66.05	65.97
Al_2O_3	18.38	18.40	18.42	18.44	18.47	18.39	18.40	18.41	18.42	18.43	18.47	18.52	18.56
Fe_2O_3	1.48	1.52	1.56	1.60	1.64	1.50	1.51	1.53	1.54	1.5	1.54	1.58	1.61
TiO_2	1.099	1.10	1.10	1.10	1.11	1.09	1.08	1.07	1.06	1.10	1.10	1.10	1.09
CaO	0.29	0.30	0.32	0.33	0.34	0.35	0.42	0.48	0.54	0.30	0.31	0.32	0.34
MgO	1.57	1.58	1.59	1.59	1.60	1.61	1.64	1.68	1.71	1.59	1.60	1.62	1.63
K_2O	1.89	1.88	1.86	1.84	1.83	1.88	1.87	1.87	1.86	1.87	1.86	1.84	1.83
Na_2O	3.42	3.4	3.38	3.37	3.35	3.40	3.38	3.36	3.34	3.41	3.40	3.39	3.38
MnO_2	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03
P_2O_5	0.01	0.02	0.04	0.05	0.07	0.01	0.03	0.04	0.06	0.01	0.03	0.05	0.06
Cl^-	0.11	0.11	0.11	0.10	0.10	0.11	0.11	0.10	0.10	0.11	0.11	0.10	0.10
SO_3^{--}	0.79	0.77	0.75	0.73	0.71	0.77	0.75	0.73	0.71	0.77	0.75	0.73	0.71
Total	100.0	100.0	100.0	99.98	99.99	100.1	100.1	100.2	100.2	100.0	100.0	100.0	99.99

3.2- Particle size distribution

Tables 4, 5, and 6 represent the particle size distribution of the cyclone dust, sludge and filter dust respectively. It is observed that cyclone dust has the finest grain size. About 30 % of the cyclone dust sample is less than 3.4 micron while this fraction size is lowered to 5 and 3 % for sludge and filter dust respectively. The mean particle size of the base body was measured to be 25 micron. Generally, the particle size distributions of these wastes are suitable for participating in ceramic floor tiles recipes. The very small grain size permits direct incorporation in ceramic formulations.

Table (4): The particle size distribution of cyclone dust.

No	Diameter, µm	Fraction size, (q) %	Cumulative weight, (Q) %
1	3.4090	29.975	29.975
2	3.9045	37.022	66.997
3	4.4721	24.216	91.213
4	5.1223	7.738	98.951
5	6. 0000	1.049	100.000

Table (5): The particle size distribution of sludge.

No	Diameter, µm	Fraction size, (q) %	Cumulative weight, (Q) %
1	3.4090	4.448	4.448
2	3.9045	13.573	18.021
3	4.4721	27.213	45.234
4	5.1223	33.160	78.395
5	6. 0000	21.605	100.000

Table (6): The particle size distribution of filter dust.

No	Diameter, µm	Fraction size, (q) %	Cumulative weight, (Q) %
1	3.4090	3.278	3.278
2	3.9045	10.926	14.204
3	4.4721	24.869	39.073
4	5.1223	34.951	74.024
5	6. 0000	25.976	100.000

Table (7): Water workability of different batches (at a = h/ho = 3.3).

Batch	Base Body	C1	C2	C3	C4	S1	S2	S3	S4	F1	F2	F3	F4
% H_2O	21	19	18.5	18	18	17	17.5	18	18.5	18	18.5	19	20

3.3- Water Workability

Table 7 shows the plasticity of the base body and the batches of different wastes content. It is shown that the addition of wastes decreases the plasticity of all batches compared to the base body. This is expected since the waste additions lead to the decrease of the loss on ignitions (L.O.I) of all batches, as shown in Table 3. Lower L.O.I means lower organic content which is responsible for high plasticity. Further additions of sludge and filter dust causes an increase of the plasticity but still lower than the base body. This increase of plasticity may be referred to the lower particle size of wastes compared to the base body. It is well known that the finer the particles, the greater the plasticity [9]. On the other hand, although the cyclone dust has the lowest particle size of these wastes, the plasticity of its batches decreases as the cyclone dust proportion increases. The high Fe_2O_3 content of cyclone dust, as shown in Table 2, seems to play a role in decreasing the plasticity.

4. Results and Discussion

The measured values of water absorption (W.A), bulk density (B.D), apparent porosity, and linear shrinkage (L.S) of the fired specimens are shown in Figures (1-3). Figures (4-6) represent the variation of modulus of rupture of the dried and fired specimens with wastes content. The addition of 2.5 % cyclone dust leads to a remarkable decrease of water absorption and apparent porosity which is accompanied by an increase of bulk density and linear shrinkage. This may be attributed to the presence of relatively high percentage of Fe_2O_3 in the cyclone dust, which is known as a highly fluxing mineral that enhances vitrification by reducing the viscosity of liquid phase [10, 11]. The fine particles of the cyclone dust are considered as another factor that helps in the densification via improving the packing density [12]. Higher proportions of finer particles in the raw material favor the vitrification due to the good compaction of samples during molding [13]. The same enhancement of physical properties is observed at further additions of cyclone dust up to 7.5 %, while adding of 10 % cyclone dust results in less vitrified specimen. However, it is still better than the base body.

Tendency for a general degradation of properties with sludge additions may be attributed to its chemical composition. As given in Table 2, sludge has the highest CaO content compared to other wastes. It was observed that the presence of lime in ceramic recipes generally increase the final firing temperature [14], which means that specimens S1 to S4 may need a higher maturing temperature. The degradation of physical properties in filter dust batches is a little bit lower than that in sludge batches. Addition of wastes has shown a slight effect on the modulus of rupture of the dried specimens, while modulus of rupture of fired specimens has affected noticeably by waste additions. The variation of modulus of rupture with waste additions has exhibited the same trend of physical properties variations. The porosity seems to play the main role regarding the mechanical properties of the studied specimens. The lower the apparent porosity, the higher the modulus of rupture in all fired specimens. In spite of the observed degradation of physical and mechanical properties at certain wastes additions, all the measured properties are still in the accepted range for tiles fired at these industrial conditions [15].

In addition to the base body, C3, F3 and S3 specimens have been selected to be examined by XRD since they revealed the best physical properties. Fig.7 reveals that all specimens contain quartz (Q) and albite (A) in addition to a little portion of hematite (H) (about 6 %) in C3. This is coincides with the relatively significant Fe_2O_3 content in cyclone dust as shown in Table 2. Although the specimens were fired at 1190 ˚C, no mullite crystals were detected. This can be realized since the firing was carried out according to the short industrial firing cycle at which the thermodynamic equilibrium can not be achieved. The existence of a notable amount of albite after firing at 1190 ˚C is also referred to the very rapid firing given that the eutectic point is expected at 1070 ˚C in the presence of orthoclase, quartz and albite [16].

Depending on XRD results, the base body and C3 specimens were selected to be examined by SEM. Fig.8.A,B reflect an irregular structure of both of the base body and C3 as a result of the short firing cycle. As shown in Fig.8.C, aggregates of hematite (H) particles are identified by their characteristic morphology [17] and that is confirmed by the XRD results of C3 (Fig.7). Fig.8.D shows elongated grains of albite (A) in addition to quartz (Q) in the base body.

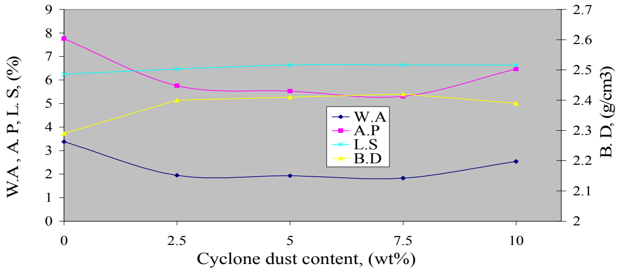

Fig.1 Effect of cyclone dust additions on the densification parameters.

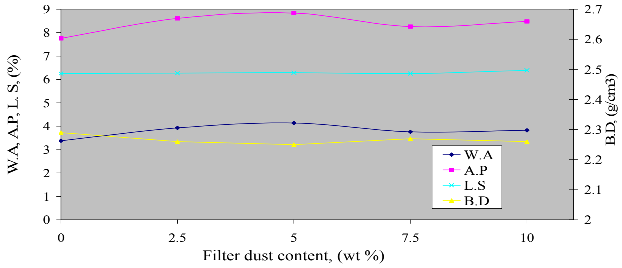

Fig.2 Effect of filter dust additions on the densification parameters.

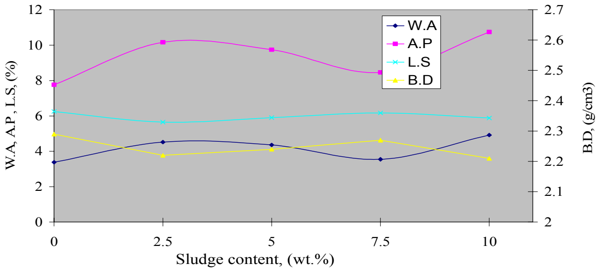

Fig.3 Effect of sludge additions on the densification parameters.

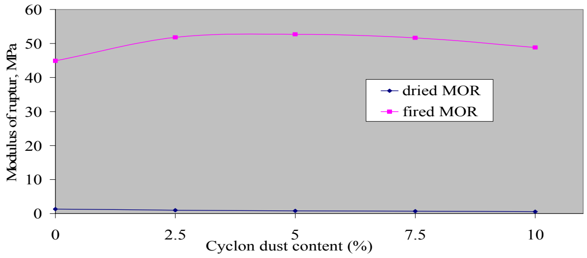

Fig. 4 Effect of cyclone dust addition on the modulus of rupture of the dried and fired specimens.

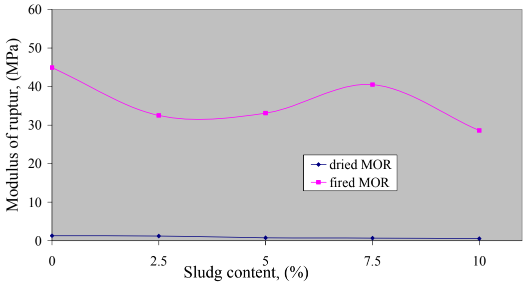

Fig. 5 Effect of sludge additions on modulus of rupture of dried and fired specimens.

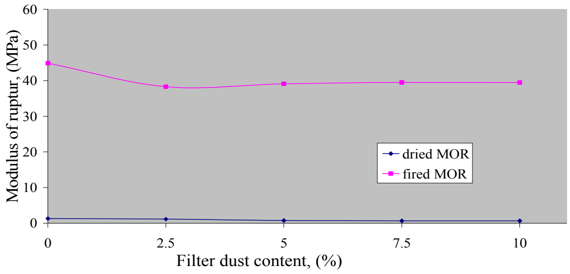

Fig. 6 Effect of filter dust additions on modulus of ruptur of dried and fired specimens

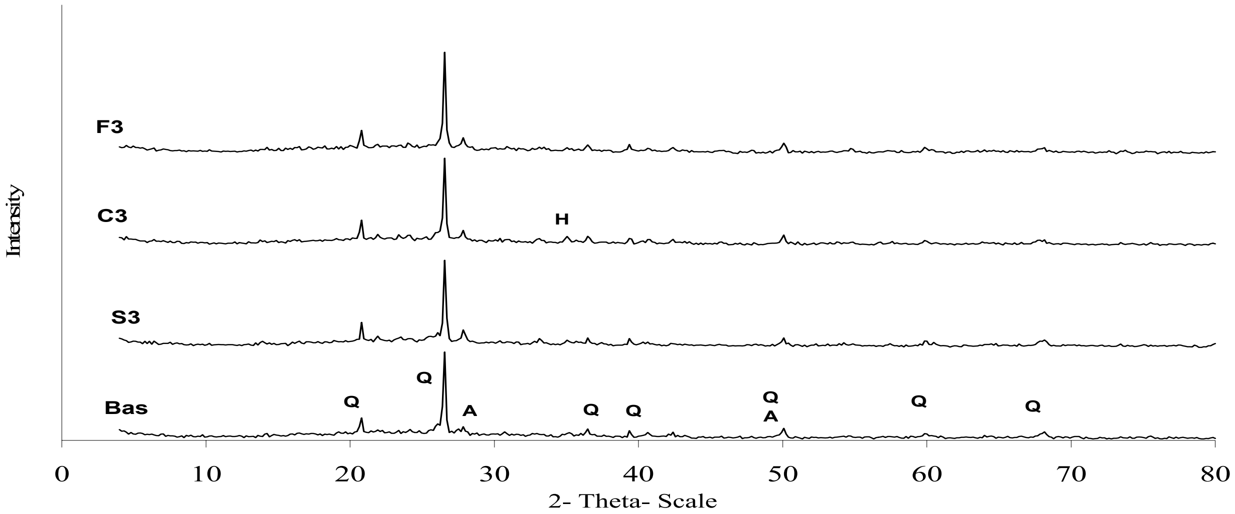

Fig.7 XRD patterns of the selected specimens.

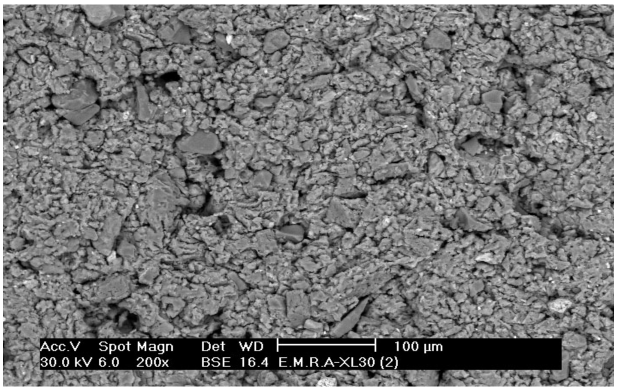

Fig. 8.A: SEM Photomicrograph of Base Body.

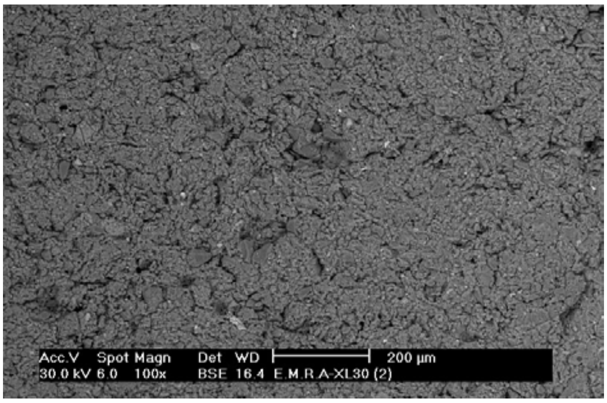

Fig. 8.B: SEM Photomicrograph of C3.

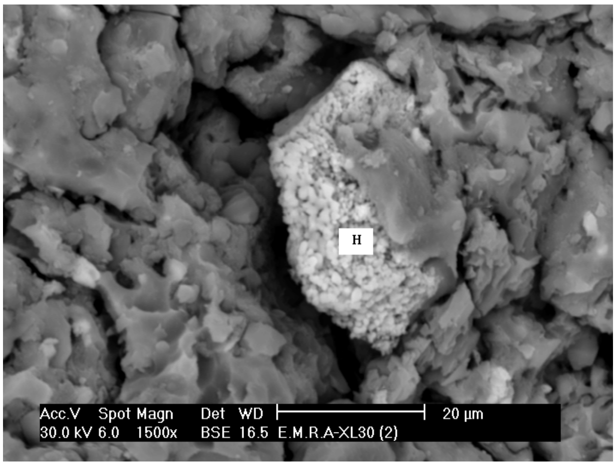

Fig. 8.C: SEM Photomicrograph of C3.

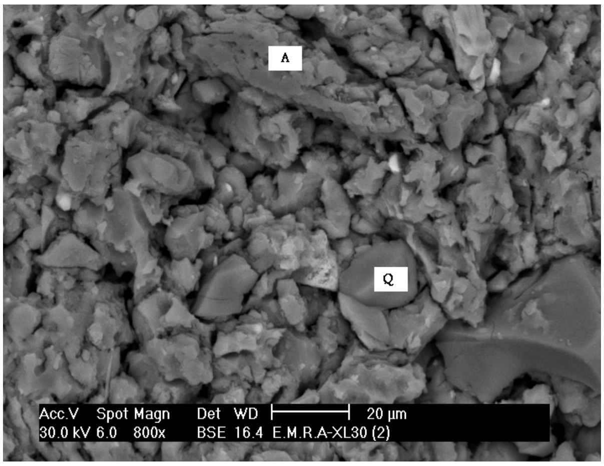

Fig. 8.D: SEM Photomicrograph of Base Body.

5- Conclusion

It is possible to use some wastes of a ceramic factory such as cyclone dust, fly dust, and sludge in the preparation of ceramic tiles at the same factory. The Physico-mechanical properties are enhanced by the addition of cyclone dust and deteriorated by the addition of sludge although they are still within the acceptable range of such products. On the other hand, properties of the product are nearly not affected by the addition of filter dust. The best properties are achieved at 7.5 % wt content of each waste. The XRD and SEM analysis have not shown any substantial differences among the investigated specimens except for cyclone dust batches, which reveal the presence of a small portion of hematite. The followed industrial cycle seems to be too short to lead to the formation of mullite crystals or even the complete melting of soda feldspar.

6. References

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