Recycling of the Mixture Resulted from Roller Kiln Waste and Ceramic Tiles Sludge Waste in the Manufacturing of Ceramic Floor Tiles

M H Roushdy

Abstract*: Ceramic tiles industry produced many types of wastes that are routinely discarded as stock piles. One of the environmental problems associated with roller kilns is the periodic need to grind its rollers which results in a waste powder. The second type of waste is ceramic tiles sludge which produced from a water treatment plant in the ceramic tiles factory. This paper investigates the probability of substituting part of the main body of floor tiles mixture by these two types of waste powder. Many Experiments were done on the raw materials. Rectangular tile specimens were molded, dried and fired. Linear firing shrinkage, loss on ignition, sintering parameters, and mechanical properties were determined. It was found that the samples with the composition (1% Roller Kiln, 35% Ceramic tiles sludge, and 64% Floor tiles mixture) or (2% Roller Kiln, 24% Ceramic tiles sludge, and 74% Floor tiles mixture) have the optimum properties.

***Index Terms*: ceramic sludge; roller kiln waste; ceramic floor tiles; waste recycling.**

I. INTRODUCTION

Ceramic tiles history appeared from a very long time. In ancient Egypt, Greece, China and other parts of the world before thousands of year's ceramics have been traditionally produced at a high temperature using sands, clays and other earthly raw materials. Ceramic tiles, refractory products, sanitary ware, table ware and glasses are an example of the ceramics products. Floor and wall tiles are used for interior and exterior decoration [1].

Roller kilns was considered the main device used in firing of ceramic tiles. Roller Kilns have a lower cost, better efficiency, a uniform heat and temperature and can be used in fast firing process so it has more advantages than tunnel kiln, [2]. The distance between the rollers determines the size of the product that will be fired in it [3].

Roller kiln is a reason for environmental problems due to its operation idea and it needs surface grinding for its rollers from time to time to remove any traces on it. The rollers are made mainly from alumina. The sources of contaminations are either the flow of glaze from tile edges or alkali salts deposition as it condensate from vapor phase in the cooling zone of the kiln. These alkali salts, are added to ceramic tiles to give the ceramic tiles some special properties. This waste has a big hazard to human health because of its storage in outdoor [3, 4].

Ibrahim [6] used the roller kiln waste in the production of alumino–silicates refractories. The results showed this waste is good raw material for production of refractories and the best firing temperature is 1350 °C. On the other hand, Ahmed [7] found that the roller kiln waste can be used to produce refractories when fired at for 6 hours at 1300 °C and can be used as raw material for porcelain production when fired at for 2 hours at 1300 °C. Two researchers found that the roller kiln can be added to wall and floor tiles mixtures without any treatment. They found that adding 2% of this waste gives a wall or floor tiles compatible with the standard ceramic tiles with good properties [8, 9].

Ceramic tiles sludge is also a waste resulted from ceramic tiles industries. It results by washing all equipment used in this industry from time to time. It has a range of 2 weight percent of the products which is considered to be waste. Although, the sludge ceramic which is produced form the treatment plant wastewater of the ceramic tile factory is considered as a waste and rejected but it must take in consideration that the elimination of the waste become an issue for the presence of the amount which is considered to be huge which is generated every year and it also increase the disposal cost [10].

Andreola [11] reuse the ceramic tiles glazing sludge which is produced from washing the phase of the glazing tile. He used it as a raw material for wall and floor tiles and found that it produced a high sintered wall and floor covering tiles. The resulted tiles have a good benefit of energy saving as it was fired at 1000°C instead of 1200°C. Abdel Ghafour, [12] utilized the ceramic sludge as a raw materials and replacement of clay in the production of building bricks. He found that 15% replacement of clay can lead to production of perfect bricks with respect to physical and mechanical properties. Nandi et al. [13] investigated the possibility of mixing glass with ceramic sludge and use this mixture as a raw material for ceramic tiles production. He found that this mixture is good idea for production of ceramic tiles compatible with the standards.

II. EXPERIMENTAL WORK

A. Raw Materials

There are three types of raw materials used in this work as kindly supplied by Ceramica Venus Company, 10th of Ramadan city as follow:

1- Ceramic floor tiles basic mixtures. This type of floor tiles basic mixture was prepared from Egyptian raw materials that have the batch composition shown in Table 1 as stated by the supplying company.

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2- Sludge waste obtained from the water treatment unit.

3- Roller kiln waste obtained on surface grinding of the kiln rollers

Table 1. Batch Composition of Floor Tiles Mixture

Raw Materials	Weight %
Kaolin Clay	1
Ball Clay	41
Bentonite	2
Feldspars	40.5
Sand	5.5
Green Tiles Scrap	7
Talc	3

B. Raw Materials Characteristics

The chemical analysis of fine ceramic waste was assessed using XRF. The equipment used was Wavelength Dispersive (WD–XRF) Sequential Spectrometer, installed at the central laboratories sector at the Egyptian mineral resources authority, the ministry of petroleum Center, Cairo.

The phase composition was obtained by XRD analysis using X–Ray Diffractometer apparatus (the central laboratories sector at the Egyptian mineral resources authority, the ministry of petroleum Center, Cairo).

On the other hand, thermal analysis was used to follow up possible thermal changes in the prepared mixtures (TGA and DTA), installed at the Center of accurate analysis, Cairo University, Cairo.

BT–2001 Laser Particle Size Analyzer was used to determine the particle size distribution of the fine waste and raw materials mixture according to ASTM D 422 [14]. It is present at the central laboratories sector at the Egyptian mineral resources authority, the ministry of petroleum Center, Cairo.

Finally, the density bottle method was used to determine the powder density was determined according to ASTM B 311 [15].

C. Preparation of Ceramic Tiles

The ceramic tiles sludge which produced from ceramic tiles factory is first dried at 110 ^{o}C for 24 hours to remove its water content then crushed in jaw crusher and finally grinded in ball mill to get a final fine waste powder. Prepared mixtures of waste powder were prepared using a roller kiln waste with percent 1% or 2% mixed with a ceramic sludge waste with percent in range from 10% to 50%. The floor tiles formation from the prepared mixtures was carried out as follow:

1. Rectangular ceramic tile specimens with dimensions 110.4 × 55.4 × 8 mm^3, were molded using dry pressing under pressure of 27 MPa.
2. Tile specimens were dried using a laboratory dryer on two steps. It dried for four hours at 70 °C, then for one hour at 110 °C.
3. Tile specimens were fired using the laboratory furnace, Protherm–electrical furnace model PLF 14015 at 1180 °C, for 15 min soaking time. Heating rates were chosen to be as close as possible to industrial conditions.

The single fast firing technique is used. The following steps show the firing schedule in a roller kiln:

1. The temperature is increased from room temperature to 600 °C quickly.
2. The temperature is increased gradually 600 °C to 700 °C, increasing 20 °C every 10 min, in order to provide slowly escape for combined water and prevent crack formation.
3. Fast increase the temperature from 700 °C to the required firing temperature and soaked for 15 min then stop the furnace.

D. Testing of Ceramic Tiles

According to ASTM C 326 [16] the percent linear firing shrinkage was determined, apparent porosity and percent water absorption were determined according to ASTM C 373 [17] and breaking strength and modulus of rupture according to ISO 10545 – 4 [18]. The micrographs of the optimum ceramic tiles were resulted using a scanning electron microscope (SEM). The SEM apparatus used is JEOL–JSM 6510 apparatus.

III. Results and Discussion

A. Chemical composition of ceramic waste

The details of XRF analysis of the raw mix are shown in Table 2. The following conclusions the obtained results:

1. For ceramic sludge waste, silica is the main component rather than alumina. The loss on ignition has a reasonable value due to presence of limestone and organic matter in it.
2. For roller kiln waste, alumina is the main component rather than silica as the rollers in the kiln is high alumina type. Also an almost zero loss on ignition was observed.

able 2. Chemical Analysis of Raw Materials

Main constituents	Sludge waste	Roller kiln waste	Floor mix
SiO_2	54.35	10.69	61.21
Al_2O_3	21.32	83.75	0.83
Fe_2O_3	4.67	1.61	20.19
TiO_2	0.85	0.21	4.78
ZrO_2	< 0.01	2.35	0.99
MnO	0.12	< 0.01	1.21
SO_3	0.12	0.07	2.72
MgO	0.11	0.22	1.21
CaO	5.99	0.25	0.21
Na_2O	2.88	0.02	0.28
K_2O	1.4	0.03	0.10
Cl	0.05	0.06	0.286
P_2O_5	0.17	0.03	5.99
LOI	7.7	0.42	0

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B. Mineralogical of ceramic waste

The XRD pattern of floor tiles mixture as shown in Fig 1, showed that it contains the following phases: Quartz (SiO_2), Albite Na (Al Si_3 O_8), and Kaolinite (Al_2 Si_2 O_5 $(OH)_4$). The main phase is quartz [19]. The XRD pattern of ceramic sludge waste as shown in Fig 2, showed that it contains the following phases: Quartz (SiO_2), Calcite ($CaCO_3$), Albite ($Na_{0.98}Ca_{0.02}$) ($Al_{1.02}Si_{2.98}O_8$), and Kaolinite ($Al_2Si_2O_5(OH)_4$). The main phase is quartz. As expected, ceramic sludge is composed of a mixture of all ingredients constituting the raw mixes for wall and floor tiles. The XRD pattern of roller kiln waste as shown in Fig 3., showed that it contains the following phases: Mullite ($Al_2.3Si_7O_4.8_5$), Corundum (Al_2 O_3), Anorthite (Ca $Al_2Si_3O_8$), and Gehlenite ($Ca_2Al((Al$ $Si)$ $O_7)$. The main phase is Corundum [9].

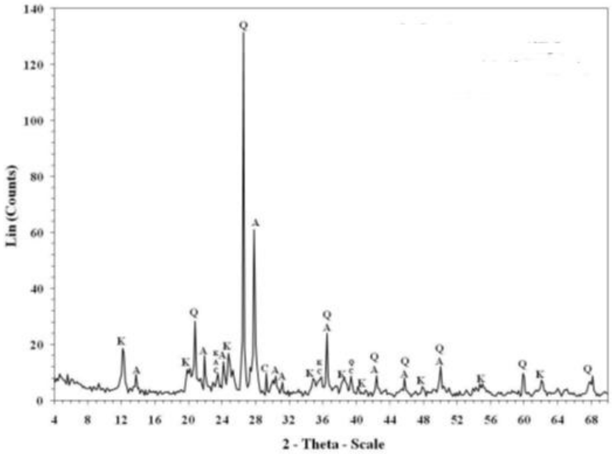

Fig. 1. XRD Pattern of Floor Mix

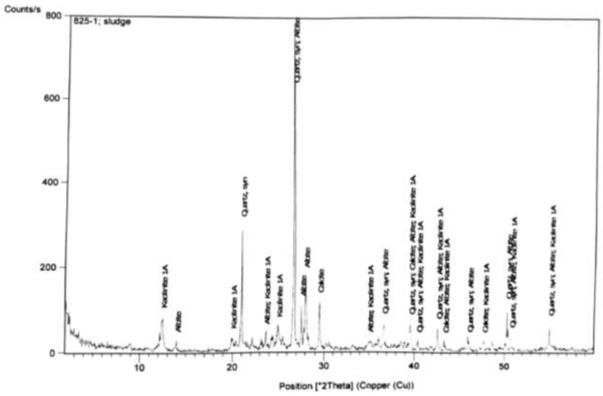

Fig. 2. XRD Pattern of Ceramic Sludge Waste

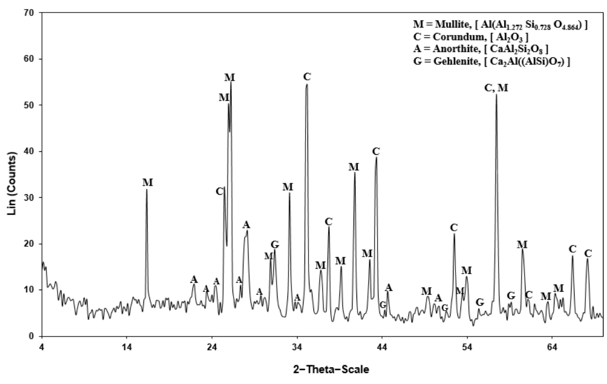

Fig. 3. XRD Pattern of Roller kiln waste

C. Thermal analysis of ceramic waste

Combined TGA – DTA chart for floor tile mixes is shown in Fig 4 and 5. The first weight loss is because of physical water removal. Then there is an exothermic because of the organic impurities oxidation that ends at about 400^{o}C. Then after that there is an endothermic peak because of lattice water of clays that ends at about 525^{o}C and practically completed at about 650^{o}C on TGA. Also owing to the presence of limestone another peak can be observed at about 720^{o}C [19].

Combined TGA – DTA chart for roller kiln waste is shown in Fig 6 and 7. There is some very small losses in temperature range between the 110–170°C because of moisture content losses, after that there is a exothermic peak because of organic impurities oxidation. The weight increase on TGA can be neglected because it rises from 4.1 mg at 300°C to 4.35 mg at 1000°C because of a misshaping of the base line. This results are accepted because of the inert nature of (quartz +mullite) present the sample [9].

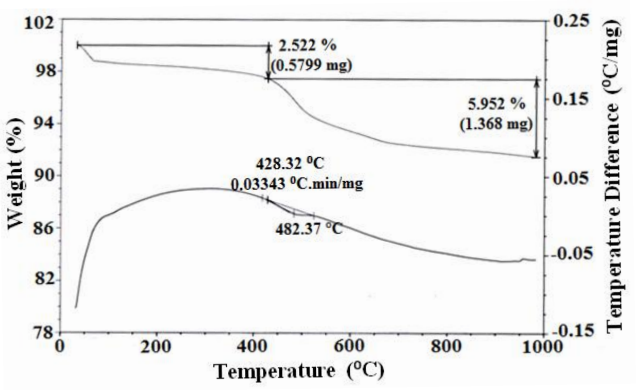

Fig. 4. DTA and TGA Pattern of Floor Mix

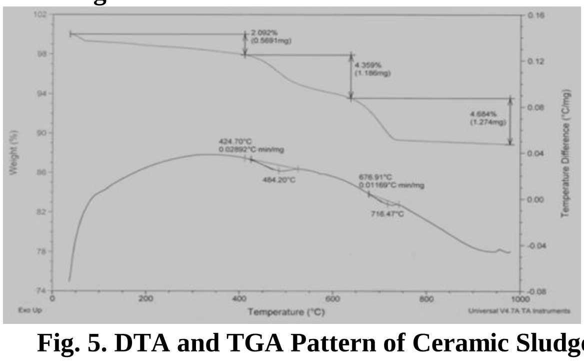
Fig. 5. DTA and TGA Pattern of Ceramic Sludge

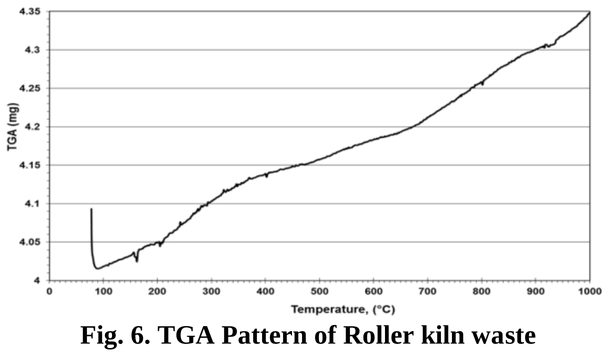

Fig. 6. TGA Pattern of Roller kiln waste

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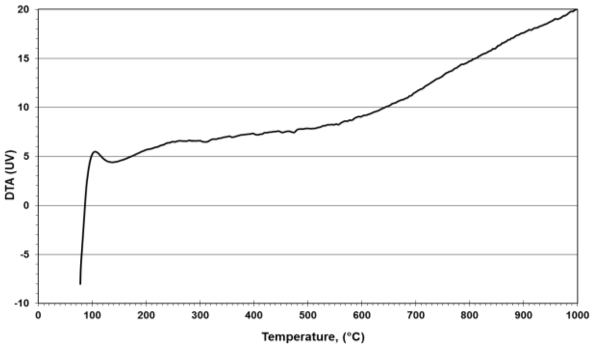


Fig. 7. DTA Pattern of Roller kiln waste

D. Screen analysis of raw materials

Fig. 8 shows the cumulative screen analysis of ceramic roller kiln waste, and ceramic sludge waste compared with ceramic floor tiles. The mean particle size of any type of waste powder is much lower than that of floor tiles mix.

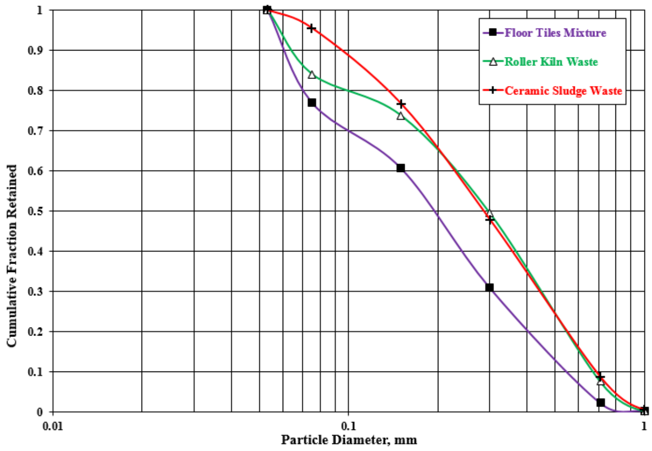


Fig. 8: Cumulative analysis of floor tiles mixture and all types of waste powder

E. Powder density

Table 3 displays the powder density of the raw materials and the waste powder. The roller kiln waste has a higher powder density than the other materials as it has a higher amount of alumina in its constituent. The density of pure alumina equals 3.69 g/cm^3 while the density of pure silica equals 2.65 g/cm^3.

Table 3. Powder Density

Raw Material	Powder Density, (g/cm^3)
Ceramic Sludge Waste	2.72
Roller kiln Waste	3.14
Floor Mix	2.69

F. Green Breaking strength

The green strength of tiles is not a standard requirement. However, a high MOR will ensure less broken green tiles on conveying. The values of MOR are illustrated in Table 4 for different samples. Values of MOR must be higher than the recommended figure of 1 MPa and that was proved by the following table.

Table 4. Values of Green MOR for Different Samples

Percentage			Modules of Rupture, MOR (N/mm^2)
Main Body	Ceramic Tiles Sludge	Roller Kiln Waste	
100	0	0	3.579473119
89	10	1	4.294731193
79	20	1	3.967515651
69	30	1	3.454045737
59	40	1	2.991432755
49	50	1	2.56095172
88	10	2	4.000161901
78	20	2	3.665214683
68	30	2	3.000375106
58	40	2	2.654800784
48	50	2	2.241616212

G. Linear firing shrinkage

Fig. 9. Summarizes the effect of wastes addition on the linear firing shrinkage results. These results show that shrinkage decreases upon adding the roller kiln waste while an increase in ceramic sludge amount will decrease this shrinkage. Floor tiles contain large amount of feldspar which is the source of liquid phase sintering that is responsible for linear firing shrinkage. The addition of ceramic sludge waste will replace the feldspar which will lead to decreasing the firing shrinkage. The increase in the amount of roller kiln waste added has an effect to decrease the firing shrinkage. It is believed that the addition of the high alumina refractory waste would have effect to limit or even suppress the formation of a liquid phase. This would explain the drop in firing shrinkage observed on adding such waste.

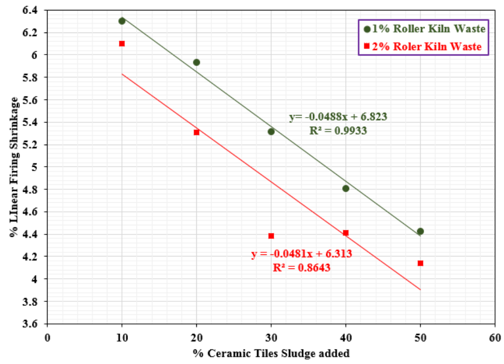


Fig. 9. The Relation between the % Linear Firing Shrinkage and the Percent of Wastes Added

H. Loss on ignition

Fig. 10. summarizes the effect of wastes addition on loss on ignition results. As can be expected the addition of the roller kiln waste decrease the total loss on ignition since the waste does not contain any decomposable material. The results showed that the addition of the ceramic sludge waste increase the total loss on ignition. The loss on ignition has a reasonable value because of the presence of limestone and organic matter in it.

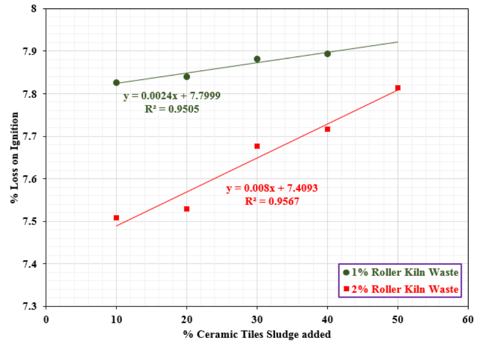


Fig. 10. The Relation between the % Loss on Ignition and the Percent of Wastes Added

I. Water absorption

Fig. 11. summarizes the effect of wastes addition on water absorption results. The resulted tiles can be categorized using the values of water absorption according to ISO13006, (2012) into two categories: tiles of low water absorption, 0.5% ・ E ・ 3 % and tiles of medium water absorption, 3 % ・ E ・ 10 %. These curves show that the percent water absorption behavior is in harmony with that of linear firing shrinkage. It increases with the increase in both ceramic sludge waste and roller kiln waste percent. The addition of ceramic sludge waste on the expense of feldspars will tend to decreasing firing shrinkage so as to increase the porosity to increase the values of water absorption.The increase in the amount of roller kiln waste added has an effect to decrease the firing shrinkage. It is believed that the addition of the high alumina refractory waste would have effect to limit or even suppress the formation of a liquid phase so as to increase the porosity to increase the values of water absorption

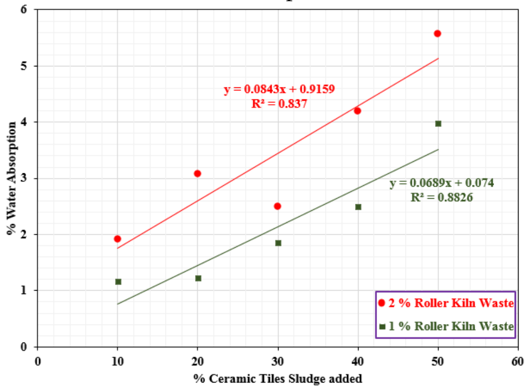


Fig. 11. The Relation between the % Water Absorption and the Percent of Wastes Added

J. Apparent porosity

Fig. 12. summarizes the effect of wastes addition on porosity results. Porosity is not a standard requirement for floor tiles. However, it is often necessary to ensure that floor tiles in particular have limited porosity to reduce the possibility of liquid infiltration.

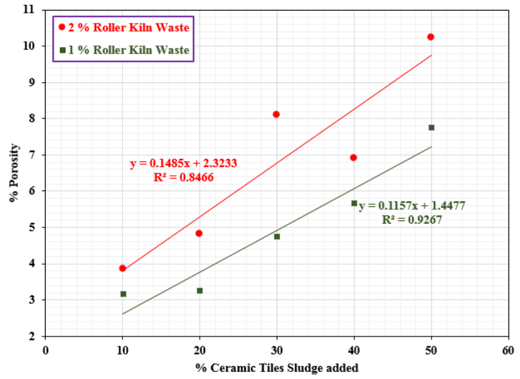


Fig. 12. The Relation between the % Porosity and the Percent of Wastes Added

K. Fired Breaking Strength

Fig. 13. shows the behavior of the firing bending strength. The values of bending strength decreases with the increase in ceramic tiles sludge and roller kiln waste. The behavior of breaking strength has the same behavior and reasons as the linear firing shrinkage. When the linear shrinkage increases the porosity decreases and then the strength increases. According to ISO 13006, (2012) Standard the minimum breaking must exceed the minimum value of 700 N for tiles of category with 0.5% < %WA < 3%. The optimum samples with the maximum waste compositions that satisfying the ISO Standard for breaking Strength [ISO 13006/2012] has a composition of (1% Roller Kiln, 35% Ceramic tiles sludge, and 64% Floor tiles mixture) or (2% Roller Kiln, 24% Ceramic tiles sludge, and 74% Floor tiles mixture).

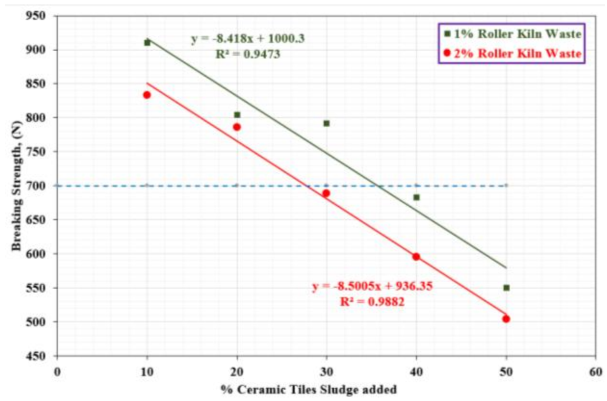


Fig. 13. The Relation between the Breaking Strength and the Percent of Wastes Added

L. Fired Modulus of rupture (MOR)

Fig. 14. shows the behavior of MOR. The values of MOR of the tested samples were naturally related to their strengths so that there was a strong similarity between the two cases. According to ISO 13006, (2012) Standard the minimum MOR must exceed the minimum value of 30 MPa for tiles of category with 0.5% < %WA < 3%. The optimum samples with the maximum waste compositions that satisfying the ISO Standard for MOR [20] has a composition of (1% Roller Kiln, 35% Ceramic tiles sludge, and 64% Floor tiles mixture) or (2% Roller Kiln, 24% Ceramic tiles sludge, and 74% Floor tiles mixture).

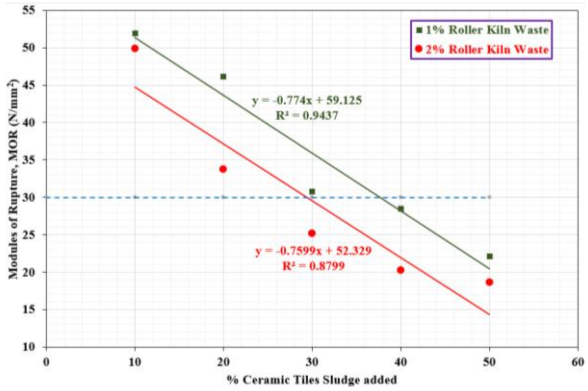


Fig. 14. The Relation between MOR and the Percent of Wastes Added

IV. Possibility of Waste Recycling into Floor Tile Bodies

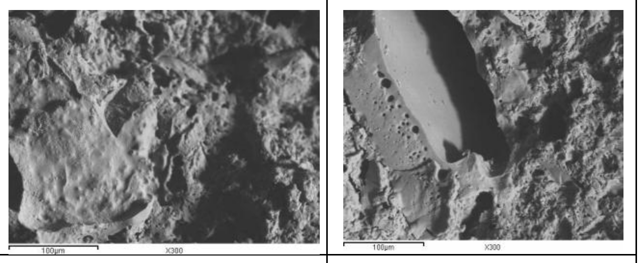
According to the previous properties that were measured, it was found that the sample with the composition (1% Roller Kiln, 35% Ceramic tiles sludge, and 64% Floor tiles mixture) or (2% Roller Kiln, 24% Ceramic tiles sludge, and 74% Floor tiles mixture) has the optimum properties with respect to all experiments.

3 specimens from each optimum ceramic tile composition (Sample 1, Sample 2) were prepared and fired then tested for percent water absorption, breaking strength and MOR. The results of sample 1 and sample 2 are shown in Table 5. The breaking strength must be more than 700 N and MOR must be more than 30 MPa according to (ISO13006/2012). As shown in Table 5 the results of both sample 1 and sample 2 satisfy the requirements of (ISO13006/2012).

Table 5. The results of sample 1 and sample 2

Property	Sample 1	Sample 2	Standard
Composition of Optimum Ceramic Tiles	1% Roller Kiln	2% Roller Kiln	
	35% Ceramic tiles sludge	24% Ceramic tiles sludge	
	64% Ceramic tiles mixture	74% Ceramic tiles mixture	
% Water Absorption	2.4855	2.89	0.5%-3%
Breaking Strength N	705.67	706.8365	> 700
MOR MPa	32.035	31.8117	> 30

In order to prove the previously obtained results, specimens represent sample 1 and sample 2 were examined by SEM at magnification 300×. The following micrographs show the high level of liquid phase formation which results in the low water absorption observed.



Sample 1 | Sample 2

Fig. 15. SEM micrographs (300x) for sample 1 and sample 2

V. Conclusion

Ceramic tiles industry produced many types of wastes that are routinely discarded and accumulates within the plant premises as stock piles. This represents an extremely high ecological hazard as such powder if inhaled for long periods can lead to serious lung problems such as silicosis. The roller kilns are periodically need to grind its rollers which results in a big environmental problem. The second type of waste is ceramic tiles sludge which produced from a water treatment plant in the ceramic factory. The main aim of this paper was to investigate the possibility of substituting part of the main body mix of floor tiles by these two types of waste powder. The experimental program includes performing XRF, XRD, DTA and TGA of raw materials. Rectangular tile specimens of dimensions 110.4 × 55.4 × 8 mm^3 were molded by dry pressing under uniaxial pressure of 27 MPa then dried overnight at 120 ^oC. Tile samples were fired at for 15 min at temperature of 1180 ^oC. Linear firing shrinkage, loss on ignition, water absorption, mechanical properties, and apparent porosity were determined and compared to ISO standards. According to the previous properties that were measured, it was found that the sample with the composition (1% Roller Kiln, 35% Ceramic tiles sludge, and 64% Floor tiles mixture) or (2% Roller Kiln, 24% Ceramic tiles sludge, and 74% Floor tiles mixture) has the optimum properties with respect to all experiments.

3 specimens from each optimum ceramic tile composition (Sample 1, Sample 2) were prepared and fired then tested for percent water absorption, breaking strength and MOR. The results of both samples satisfy the requirements of (ISO13006/2012).

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REFERENCES

1. Abadir. (2011). "*Elements of Ceramic Technology*", 6th Ed. Cairo University, Faculty of Engineering.
2. Berenshtein P.I. (Nov. 1971), "*Operation of roller kilns for firing ceramic tiles*", Glass and Ceramics, 28(11), p.p 679–681.
3. Mark. (2009). "*A International Company Catalogue*",. Gujarat, India.
4. Association, A. L. (2010). *Occupational Lung Disease*.
5. Gulati, C. (2008). *Occupational Lung Diseases*. International Encyclopedia of Public Health.
6. Ibrahim O.A. (March 2009), "*The use of the product obtained on grinding kiln rollers in then production of refractory bodies*", 24th Int. Conf. Solid Waste Management., Pa, US.
7. Ahmed M.M. (January, 2011), "*Characterization, Properties and Uses of the Powder Obtained on Grinding Kiln Rollers*", Cairo University, Faculty of Engineering, Ph.D. thesis.
8. Youssef, G. Gazal H. (2011). "*Characterization and Reuse of Kiln Rollers Waste in the Manufacture of Ceramic Floor Tiles*". Journal of American Science.
9. Roushdy. M.H., Amin. Sh. K., Ahmed M.M., Abadir M.F., (2014). "*Reuse of the product obtained on grinding kiln rollers in the manufacture of ceramic wall tiles*". Ceramic Technical
10. Wattanasiriwech. (2009). "*Paving blocks from ceramic tile production waste*". Journal of Cleaner Production.
11. Andreola. (2010). "*Recovery of Glazing Ceramic Sludge in Construction*". 2nd International Conference on Sustainable Construction Materials and Technologies.
12. Abdel-Ghafour. (2010). "*Making Green Building Units by Using Some Wastes of Ceramic Industry*". Proceedings of the 10th International Conference for Enhanced Building Operations.
13. Nandi. (2015)." *The Use of Ceramic Sludge and Recycled Glass to Obtain Engobes for Manufacturing Ceramic Tiles*". Journal of Cleaner Production.
14. ASTM D 422 / 1963 (Reapproved 2007), "*Method for particle–size analysis of soils*", ASTM Annual book, U.S.A., 4 (8).
15. ASTM B 311 / 2017, "*Standard test method for density of powder metallurgy (pm) materials containing less than two percent porosity*", ASTM Annual book, U.S.A., 2 (5).
16. ASTM C 326 / 2009 (Reapproved 2014), "*Standard test method for drying and firing shrinkages of ceramic white–ware clays*", ASTM Annual book, U.S.A., 15 (2).
17. ASTM C 373 / 2017, "*Standard test methods for determination of water absorption and associated properties by vacuum method for pressed ceramic tiles and glass tiles and boil method for extruded ceramic tiles and non-tile fired ceramic white-ware products*", ASTM Annual book, U.S.A., 15 (2).
18. ISO 10545 – 4 / 2014, "*Ceramic tiles – Part 4: Determination of modulus of rupture and breaking strength*", International Organization for Standardization (ISO), Geneva.
19. Amin. Sh. K., Abdel Hamid E.M., El-Sherbiny S.A., Abadir M.F (2017). "*The use of sewage sludge in the production of ceramic floor tiles*". Housing and Building National Research Center Journal.
20. ISO 13006 / 2012, "*Ceramic Tiles – Definitions, classification, characteristics and marking, (Annex L)*", International Organization for Standardization (ISO), Geneva, p.p 41–43.

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