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Ceramic Tiles Production from Cullet and Agricultural Wastes Obtained from Wheat and Sugarcane Cultivation

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Abstract: - Agricultural wastes are a big source for environmental pollution so it's a good choice to recycle them so as to get an environmental and economic benefit. The purpose of this research is to determine the possibility of recycling two types of agriculture wastes obtained from wheat and sugarcane cultivation in addition to broken glass (cullet) as raw materials for ceramic wall tiles production. The used agriculture wastes are the agricultural residue obtained from wheat and sugarcane cultivation. The experimental work starts with performing XRF and sieve analysis for all raw materials. Ceramic wall tiles specimens were made with dimensions $110.4 \times 55.4 \times 8 \text{ mm}^3$ using 27 MPa dry pressing then dried at 120 °C overnight, then fired using firing temperatures equal to 1100 °C or 1150 °C during 15 min soaking time. Water absorption, apparent porosity, and mechanical properties were done to compare them with ISO standards. According to the previous experimental work, it was found that the samples with the composition (10% Cullet, 15% Wheat ash straw, 20% sugarcane) for samples that fired at 1150 °C or (10% Cullet, 13% Wheat ash straw, 18% sugarcane) for samples that fired at 1100 °C has the optimum properties.

Key-Words: Wheat Straw; Sugarcane; Cullet; Wall tiles; ISO standard; Optimization

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1 Introduction

Ceramics are known as inorganic and non-metallic materials, which are the main materials in our daily lifestyle. The material engineers are the designers for every process in for these products formation. They are there to create new products of ceramics and to find out many different usages for the ceramic products in our daily life [1].

The main raw material for the ceramics products are clays which are the main body of the ceramic products, feldspars which is used as fluxing components for their ability to melt at low temperature, and sand which is used as the main glass forming raw material. Some other raw materials might be used such as the dolomite and the limestone [2].

White wares, refractory products, and glasses are examples of traditional ceramics. Wall and floor tiles which are known as white wares are used for interior and exterior decoration. Recently, however, some ceramic products could be prepared from non-silicate raw materials and sometimes even without firing. That is why the traditional view towards ceramics has changed so that nowadays a ceramic material is defined as being an inorganic non-metallic solid [3].

Egyptians, the Babylonians, and the Assyrians are the first producers for ceramic tiles. The Step Pyramid for the Pharaoh Zoser that was built by ancient Egyptian around 2600 B.C., contained colorful glazed tiles. European country and the United States manufactured ceramic tiles later. Today, tile manufacture is highly automated [4]. Amin et al. re-used Waste sludge in the floor and wall tiles production. It was found it is possible to produce floor tiles contains 20% sludge and wall tiles contains 10 % sludge [5]. Roushdy (2019a) investigated using mixture of roller kilns waste and ceramic tiles sludge as a raw materials for floor tiles production. The results shows that the optimum samples have composition of 64% Floor tiles mixture, 35% Ceramic tiles sludge, and 1% Roller Kiln or 74% Floor tiles mixture, 24% Ceramic tiles sludge, and 2% Roller Kiln [6]. Roushdy (2019 b) examined the possibility of using a mixture of roller kilns, ceramic tiles sludge, and sanitary ware waste powder as raw materials for floor tiles production. The results shows that the optimum samples have composition of 44% Floor tiles mixture, 39% Ceramic tiles sludge, 15% sanitary ware waste, and 2% Roller Kiln [7]. Allam et al. investigate producing roof tile samples from ordinary Portland cement (OPC) with Marble Waste Slurry with composition in range of 5-25%, and

roof tiles from sand with Marble Waste Slurry with composition in range of 5-40%. All mechanical tests should that roof tile samples containing MWS as a partial replacement for sand and cement were complied with the ASTM standard limits till 20% replacement ratio [8].

The main substrates of human diet are sugar. China, Brazil, Australia, Thailand and India are the top five countries in the world in sugar producing. At the year 710 AD Egypt started sugar production [9]. Sugar cane was a source of sugar in sugar production since 1981. Some areas of Upper Egypt are famous with cane plantations as the amount of cultivated cane was 16 million tons in 2009 [10; 11]. On the other hand, Egypt is famous with wheat cultivation as it occupies about 32.6% of the total winter land area. Wheat straw ash is considered as the most important agricultural residues. Straw consists mainly of lignin, hemicelluloses, and cellulose [12].

Hassan et al. (2018) investigate the use of sugarcane bagasse and wheat straw in the fireclay bricks production. Results proved that adding 5% bagasse with 0.5% polystyrene beads results and firing for 2 hours at 1250°C produced bricks abiding by ASTM standards for C-32 type insulating fireclay bricks. On the other hand, the addition of 5% wheat straw, despite fulfilling the density and thermal conductivity requirements, resulted in marginal values for cold crushing strength [13]. Micheal and Moussa integrated sugarcane bagasse with cement in the production brick. The results showed that replacing cement with 1.5% sugarcane bagasse will give the bricks with a good mechanical properties and higher strength [14]. Dawoud et al. (2020) summarized the techniques used by many researcher's to replace cement with sugarcane bagasse during building bricks production and showed the best procedures for partially replacing cement with sugarcane bagasse which give the best properties of cement bricks [15].

The main objective of the present study is to investigate the possibility of recycling two types of agriculture wastes obtained from wheat and sugarcane cultivation in addition to broken glass (cullet) as raw materials for ceramic wall tiles production.

2 Experimental Work

2.1 Raw Materials

The four types of raw materials that used in this research are the following:

- [1] Mixed raw materials provided from ceramic tiles factory. This type of ceramic wall tiles basic mixture was prepared from Egyptian raw materials as shown in Table 1.
- [2] Cullet which is waste broken glass.
- [3] Agricultural residue obtained from wheat cultivation.
- [4] Agricultural residue obtained from sugarcane cultivation.

Table 1.wall Tiles composition

Raw Materials	Wt. %
Aswan Clay	25
Ball Clay	35
Glass Sand	15
Potash Feldspar	14
Limestone	9
Bentonite	2

2.2. Characterization of Raw Materials

XRF method was used to determine the chemical composition of all raw materials by Wavelength Dispersive (WD-XRF) Sequential Spectrometer. The distribution of particle size for all the raw materials was determined using BT-2001 Laser Particle Size Analyzer according to ASTM D 422 (2007) [16].

2.3. Ceramic Tiles Preparation

The formation of wall tiles was done by the following steps:

- [1] Molding the samples of ceramic tiles by a mold with dimensions $11.04 \times 5.54 \times 0.8 \text{ cm}^3$ pressed under 27 MPa using automatically hydraulic press.
- [2] Drying the tile samples using a laboratory dryer on two steps. Mild Drying at 70 °C for 4 hours, then drastic drying at 110 °C for one hour.

[3] Firing the tile samples using heating rates so close to industrial conditions by Protherm–electrical furnace model PLF 14015. The firing technique used for the preparation of wall tiles samples using the single fast firing. The firing schedule can be described as follows that simulates conditions in a roller kiln:

- Fast temperature increasing to 550 °C.
- Slow temperature increasing from 550 °C to 750 °C in order to prevent crack formation by slow combined water escape.
- Fast temperature increasing from 750 °C to 900 °C.
- Slow temperature increasing from 900 °C to 1000 °C, in order to prevent crack formation by provide suitable time for calcination of calcium carbonate.
- Fast temperature increasing to the target firing temperature (1100 °C or 1150 °C) during soaking time of 15 Minutes.

2.4. Ceramic Tiles Testing

ASTM C 373 (2017) was used to determine the percent water absorption and open porosity while ISO 10545 – 4 (2014) was used to determine breaking strength and modulus of rupture (MOR) [17; 18].

3 Results and Discussion

3.1. Ceramic Raw Materials Chemical Composition

The Chemical analysis results for the raw materials are shown in tables 2, 3, and 4. The following conclusions can be drawn:

Table 2 shows the chemical analysis of a standard wall tile mix. The loss on ignition happened because of chemical water of clays loss and carbon dioxide loss from limestone. As can be seen from this table, silica, alumina, calcium oxide and iron oxide constitute more than 96 percent of the mix (on loss on ignition – free basis). The loss on ignition is due to calcination of limestone and loss of chemical water from clay.

For cullet, silica is the main component as shown in Table 2.

Sugarcane bagasse waste is mainly composed of lignin, cellulose, hemicelluloses, fats and silica. Its ultimate composition was established as shown in Table 3.

Wheat straw samples are mainly composed of lignin, cellulose, hemicelluloses, proteins and

sugars. Its ultimate composition was established as shown in Table 4.

Table 2: Chemical composition of Wall Mix and Cullet

Main constituents	Cullet	Wall mix
SiO ₂	73.58	60.25
Al ₂ O ₃	1.24	15.69
Fe ₂ O ₃	0.23	3.59
TiO ₂	0.04	1.4
MnO	0.007	-----
SO ₃	0.42	0.04
MgO	0.17	0.58
CaO	7.42	5.89
Na ₂ O	15.84	0.53
K ₂ O	0.05	0.67
Cl	0.06	-----
P ₂ O ₅	0.02	0.13
LOI	-----	11.18

Table 3: Chemical Analysis of Sugarcane Bagasse Waste

Main constituents	Sugarcane Bagasse
Carbon	48.7
Silica	44
Nitrogen	1.3
Hydrogen	4.9
Phosphorous	1.1

Table 4: Chemical Analysis of wheat straw

Main constituents	Wheat Straw Ash
Carbon	49
Hydrogen	5.3
Oxygen	37
Silica	1.6
Sulfur	0.66
Potassium	0.52

3.2. Particle Size Distribution for Raw Materials

Figure 1 shows the cumulative screen analysis of all raw materials. The mean particle size of wall tiles mixture is much higher than any type of waste powder so it was expected that the addition of wastes to the main raw materials will decrease the porosity of the mixture.

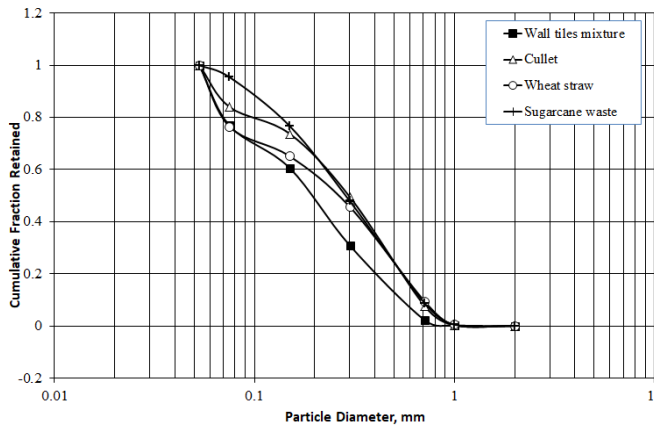


Figure 1. Cumulative Analysis of Raw Materials

3.3. Green Breaking Strength and Modulus of Rupture (MOR)

The values of green MOR are higher than 1 MPa so the tiles will face less broken on conveying.

3.4. Water Absorption and Apparent Porosity

Cullet (C) or waste glass addition to the samples increase the liquid phase formation then decreases the porosity. On other hand, there was more liquid phase formation accompanied with dilation in closed pores and expansion due to the entrapment of released gases, mainly CO₂, which in turn increase the porosity and hence increase the water absorption. The thermal treatment of waste glass at high temperature released volatile matter which promoted porous structure. Addition of cullet more than 10% results in samples warping.

Addition of wheat straw ash (W) increases the porosity and hence the water absorption. According to the chemical analysis done, the results showed that it contains mainly carbon and oxygen so its addition to the samples increases the amount of gases produced and then increases the porosity and water absorption.

Addition of sugarcane bagasse waste ash (S) increases the porosity but with little effect than wheat straw ash and hence the water absorption. According to the chemical analysis done, the results showed that it contains mainly carbon and silica so its addition to the samples increases the amount of gases produced and then increases the porosity and water absorption.

According to Egyptian standards (ISO13006/2012) [18] the values of obtained water absorption was found to be in the category of tiles with high water absorption, E > 10 %. These results are presented in following tables.

The equations related each waste percent and percent open porosity (P) using Excel program are the following

$$P, 1100^{\circ}C = 27.12326 + 0.04999 C + 0.06947 W + 0.06892 S \quad (1)$$

$$P, 1150^{\circ}C = 26.05626 + 0.04803 C + 0.06674 W + 0.06621 S \quad (2)$$

Table 5: Effect of wastes addition on percent open porosity

Main Body	Percentage			Firing Temperature, °C	Percent Open Porosity
	(C)	(W)	(S)		
100	0	0	0	1100	27.123
90	10	0	0	1100	27.812
80	0	20	0	1100	28.512
70	10	20	0	1100	29.201
80	0	0	20	1100	28.123
70	10	0	20	1100	28.812
60	0	20	20	1100	29.512
50	10	20	20	1100	30.201
100	0	0	0	1150	26.056
90	10	0	0	1150	26.718
80	0	20	0	1150	27.391
70	10	20	0	1150	28.053
80	0	0	20	1150	27.016
70	10	0	20	1150	27.678
60	0	20	20	1150	28.351
50	10	20	20	1150	29.013

The equations related each waste percent and water absorption (WA) using Excel program are the following

$$WA, 1100^{\circ}C = 14.42581 + 0.04032 C + 0.05936 W + 0.04886 S \quad (3)$$

$$WA, 1150^{\circ}C = 13.94170 + 0.03896 C + 0.05737 W + 0.04722 S \quad (4)$$

From the previous equations it was found than the addition of wheat straw ash has the highest effect on porosity and water absorption then the sugarcane waste then cullet.

Table 6: Effect of wastes addition on water absorption

Percentage				Firing Temperature, °C	Percent Water Absorption
Main Body	(C)	(W)	(S)		
100	0	0	0	1100	14.425
90	10	0	0	1100	14.914
80	0	20	0	1100	15.612
70	10	20	0	1100	16.101
80	0	0	20	1100	15.232
70	10	0	20	1100	15.720
60	0	20	20	1100	16.419
50	10	20	20	1100	16.907
100	0	0	0	1150	13.941
90	10	0	0	1150	14.413
80	0	20	0	1150	15.089
70	10	20	0	1150	15.561
80	0	0	20	1150	14.720
70	10	0	20	1150	15.193
60	0	20	20	1150	15.868
50	10	20	20	1150	16.340

3.5. Fired Breaking Strength and Modulus of Rupture

The following tables show that the bending strength or MOR decrease by the increase in waste percent. When the porosity increases and then the strength decreases. According to Egyptian standards (ISO13006/2012) [18] the values of breaking strength must exceed the minimum value of 200 N and the values of MOR must exceed the minimum value of 15 MPa for tiles of category with %WA > 10%.

The equations related each waste percent and modulus of rupture (MOR) using Excel program are the following

$$MOR, 1100^{\circ}C = 17.10553 - 0.05232 C - 0.08466 W - 0.05966 S \quad (5)$$

$$MOR, 1150^{\circ}C = 18.29934 - 0.05597 C - 0.09057 W - 0.06382 S \quad (6)$$

Table 7: Effect of wastes addition on MOR

Main Body	Percentage			Firing Temperature, °C	MOR (N/mm ²)
	C	W	S		
100	0	0	0	1100	17.105
90	10	0	0	1100	16.582
80	0	20	0	1100	15.412
70	10	20	0	1100	14.889
80	0	0	20	1100	15.912
70	10	0	20	1100	15.389
60	0	20	20	1100	14.219
50	10	20	20	1100	13.695
100	0	0	0	1150	18.299
90	10	0	0	1150	17.739
80	0	20	0	1150	16.488
70	10	20	0	1150	15.928
80	0	0	20	1150	17.022
70	10	0	20	1150	16.463
60	0	20	20	1150	15.211
50	10	20	20	1150	14.651

The equations related each waste percents and breaking strength (BS) using Excel program are the following

$$BS, 1100^{\circ}C = 410.16023 - 3.87418 C - 6.21997 W - 5.94340 S \quad (7)$$

$$BS, 1150^{\circ}C = 472.10978 - 5.01585 C - 8.27934 W - 6.38507 S \quad (8)$$

From the previous equations it was found that the addition of wheat straw ash has the highest effect on breaking strength and MOR then the sugarcane waste then cullet.

Table 8: Effect of wastes addition on breaking strength

Main Body	Percentage			Firing Temperature, °C	Breaking Strength, (N)
	C	W	S		
100	0	0	0	1100	374.291
90	10	0	0	1100	352.125
80	0	20	0	1100	310.812
70	10	20	0	1100	205.406
80	0	0	20	1100	335.425
70	10	0	20	1100	263.562
60	0	20	20	1100	191.538
50	10	20	20	1100	178.044
100	0	0	0	1150	429.139
90	10	0	0	1150	403.725
80	0	20	0	1150	356.358
70	10	20	0	1150	235.255
80	0	0	20	1150	384.578
70	10	0	20	1150	261.804
60	0	20	20	1150	219.355
50	10	20	20	1150	200.443

4 Conclusion

The main aim of this research is to investigate the possibility of recycling two types of agriculture wastes in addition to broken glass (cullet) as part of raw materials for ceramic wall tiles production. The used agriculture wastes are the agricultural residue obtained from wheat and sugarcane cultivation.

The experimental work starts with performing XRF and sieve analysis for all raw materials. Ceramic wall tiles specimens were made with dimensions $110.4 \times 55.4 \times 8 \text{ mm}^3$ using 27 MPa dry pressing then dried at 120 °C overnight. Tile samples were fired at temperatures in range off 1100 °C - 1150 °C for 15 min soaking time. Water absorption, apparent porosity, and mechanical properties were done to compare them with ISO standards.

According to the previous experimental work, it was found that the samples with the composition (10% Cullet, 15% Wheat ash straw, 20% sugarcane) for samples that fired at 1150 °C or (10% Cullet, 13% Wheat ash straw, 18% sugarcane) for samples that fired at 1100 °C has the optimum properties.

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