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Characterization of Marble and Granite Processing Waste and its Utilization in Brick Production: Case of Saba and Semayata Dimensional Stone Companies, Tigray, Ethiopia

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ABSTRACT
In Ethiopia, the marble and granite dimension stone industries have been growing significantly in the last decade and have become a part of the flourishing construction industry. Accordingly, the amount of quarrying and processing has increased significantly. This has contributed to increased waste of time. Semayata and Saba Dimensional Stone plc are the leading dimension stone-based companies located in the Wukro and Adwa areas, respectively in the Tigray region are involved in producing tiles and slabs of marble and granite. Due to its processing techniques and its highly alkaline nature, marble, and granite wastes in the Adwa and Wukro areas have become causes for environmental problems. Consequently, the demand for the recycling of waste has come to the forefront due to both environmental and financial considerations. Keeping this in view, a study was conducted on the waste generated by the Saba and Semayata processing units. The study aims to characterize wastes; and discuss the possibilities of using marble and granite wastes as an alternative raw material in the production of bricks. Based on the chemical, physical, and mechanical properties, the waste can be used to produce bricks without modifying and adding other contents.

Keywords: Dimension stone, Marble and granite, Waste, Bricks, Tigray, Ethiopia.

1. INTRODUCTION

Dimension stone played a great impetus in human endeavors worldwide (Ciccu et al., 2007; Walle and Zewude, 2000). A sustainable growth in the consumption of rocks is observed all over the world in the current years (Ciccu et al., 2007 and Vijayalakshmi et al., 2010). Various types of rocks used for construction purposes included, granite, marble, limestone, slate etc. With time, this has become a major industry as dimension stone industry producing tiles and slabs. These industries produce different size/type tiles also contribute a significant amount of waste, which often becomes an environmental issue. As a result, an effort has begun to focus on finding new ways of using the waste materials in order to lessen environmental issues and generate revenue from waste. Many countries like India, America, China, Italy, Egypt etc working on how to reuse the waste material. Ethiopian marble and granite industries are also looking for an alternative use of waste such as brick production (Sentayehu, 2011).

Waste thus may be used to supply new products so that natural resources are used effectively, and the environment is protected. During the cutting and polishing processes

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marble and granite produce about 30% and 10% dust respectively (Ciccu et al., 2007). It is mainly composed of oxides such as SiO_2, Al_2O_3, Fe_2O_3 and CaO and small amounts of MgO, TiO_2, MnO, and K_2O. These constituents have enough potential to damage the environment by contaminating soil and water if it is not efficiently treated before disposal (Ciccu et al., 2007 and Vijayalakshmi et al., 2010). Dimension stone is also one of the upcoming industries in Ethiopia. Several dimension stone processing units are working all over the country including in Tigray region, northern Ethiopia mainly processing marble and granite. Marble and granite deposits and the quarry sites distributed in Tigray region are provided in table 1.

Table 1. The location of both granite and marble quarry sites, with their estimated resources (according to the report of Tigray Region of Water, Mineral and Energy Bureau, 2006).

S. No	***Type of Deposit***	***Location***	***Estimated resource (m^3)***
1	Naeder marble	Naeder- Adet wereda, 55km from Axum	$8.23x10^8$
2	Dichinamo marble	Western Tigray, 30 km from Shiraro	$242x10^6$
3	Newimarble	Kola Tembien wereda, 42km NW of Abi-Adi	$14.72x10^6$
4	Adiwoyane marble	Kola Tembien wereda, 13km SW of Abi-Adi	$6.87x10^5$
5	Taget marble	Kola Tembien wereda, 8km SW of Abi-Adi	$5.7x10^5$
6	Emnizong marble	Medebay Zana wereda, 60km South of Axum	$2.9x10^4$
7	Akmara marble	Kola Tembien wereda, 44km NW of Abi-Adi	$534x10^6$
8	Tekeze marble	Western Tigray, 52km SW of Shiraro town	$193.76x10^6$
9	Negash granite	Eastern Tigray, 55km North of Mekelle town	$5.8x10^6$
10	Adi Elena granite	Western Tigray, about 55km from Axum	3.l5 x l0
11	Adikelete granite	Western Tigray, 68km NW of Shire town	$5.7x10^8$
12	Borpuah granite	Western Tigray, about 16km far from Rama	$4.6 x10^7$
13	Kisadgaba granite	Western Tigray, 37km west of Shire town	$5.2x10^6$
14	Embamadre granite	Western Tigray, 38km west of Shire town	$13.2x10^2$
15	Adiet granite	Central Tigray, 14 NW of Aksum town	$9.27x10^7$
16	Sebeya granite	Eastern Tigray, 35km east of Adigrat town	l.359xl0^7

A study was conducted to characterize the waste generated by the Semayata and Saba Dimension Stone processing units and its possible application in brick making industry. It involved conducting various tests chemical and physical so as to understand the utility value of the waste.

Semayata and Saba Dimension Stone companies are the leading ones with well-developed processing units in Tigray region (Ethiopia) (Fig 1). These are producing granite tiles and slabs, with an annual capacity of 240,000m² of tiles and cut-to-size slabs, which are located at Wukro and Adwa respectively. Semayata Dimension Stone Factory was built on 3.7 hectares of land, and it has a three-storey building and warehouses. About ETB 400 million

(€17.06 million/US$18.65 million) was spent by Ayiga Trade & Industry, the owner of Semayata, to build all the facilities, quarrying, and granite processing technology and it started production of granite tiles and slabs on February 18, 2016. Saba and Semayata processing units have a capacity of processing 300,000 and 240,000 m^2 of tiles and cut-to-size marble and granite slabs respectively annually. But due to the conflict in Tigray, both factories are not functioning now.

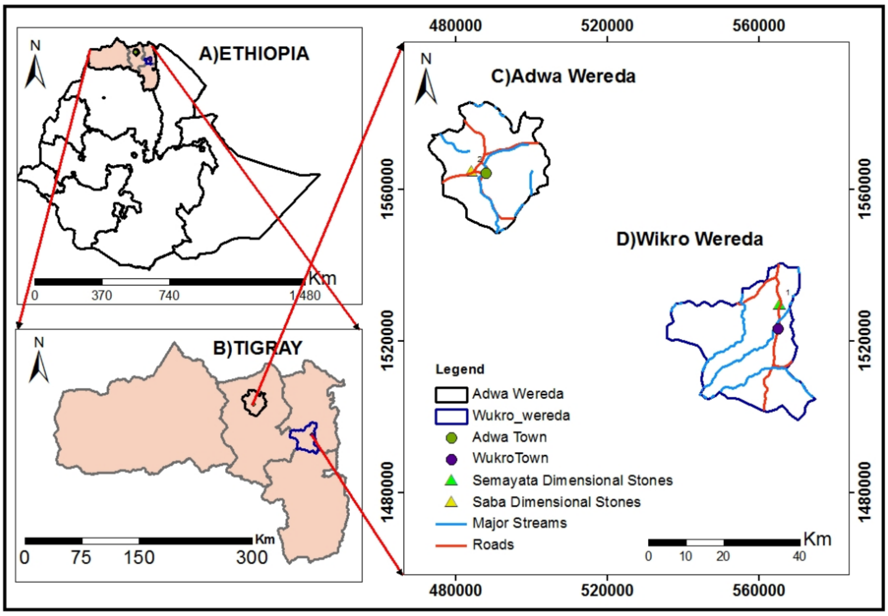

Figure 1. Location map of Semayata and Saba dimension stone processing units.

2. WASTE GENERATION, QUALITY, QUANTITY AND ASSOCIATED PROBLEMS

2.1. Marble and Granite Waste Generation

Two types of waste is generated while making granite and marble tiles/slabs. Firstly, during mining (cutting/sawing), and secondly during processing/polishing. At some point of the processing of stone, the raw stone block is cut as per demand either into tiles or slabs of different thickness (commonly 2 or 4 cm), using diamond blades. Water is showered on blades to cool the blades while cutting the blocks into sheets of various thickness and the waste is produced throughout the cutting operation (Ciccu et al., 2007, Niyaziogurkockal, 2014 and Vijayalakshmi et al., 2010). Water is generally not re-cycled due to the alkalinity problems which can make the slabs dull. In the factory, where the blocks are cut into slabs and tiles, the cooling water is stored in sedimentation tanks until the suspended particles settle, then the slurry is collected in trucks and disposed of on the ground to dry. After it becomes dry, the particles become airborne (Rania et al., 2011) and often cause air pollution problems in the surrounding area. Some other solid waste generated by using the marble and granite gadgets is the cutting waste, which ends from reducing slabs into the desired dimensions. According to Rania et al. (2011), after the stone has been cut to the specific dimensions, the slabs are finished by either polishing or texturing, as requested. The polishing operation is completely automatic with the use of powdered abrasives that maintain on scrubbing the floor of the marble until it will become smooth and shiny (Rania et al., 2011).

2.2. Marble and Granite Waste Quantification

Since it is not monitored by the government or any other interested body, it is difficult to know the actual data on the amount of waste generated in the processing units. However, as the product thickness increases, some of the waste decreases (Celik et al., 2008), which is almost coherent with the study area (Table 2). Whereas other references like Rania et al (2011); Haggar (2007) and Pareek (2007), estimate that 20 to 25% of a marble-granite product produces suspended powder, as with any 20 mm thick marble and granite slab produced; The 5 mm powder is crushed during the cutting process.

Table 2. Physical and chemical properties of marble and granite waste powder (for both Saba (marble) and Semayata (granite) dimensional stone wastes).

Physical properties of marble and granite waste			***Waste chemical composition***		
Properties	***Marble***	***Granite***	***Oxides***	***Marble wt.%***	***Granite wt.%***
Bulk Density(kg/m^3)	1118	1210-1465	Fe_2O_3	1.09	18
Specific Gravity	2.69	2-2.84	Na_2O	0.63	6
Moisture content(dry) %	1.59	0.065	MgO	18.94	0.05
Moisture content(wet) %	23.35	26-28	Al_2O_3	1.09	19
Fineness modulus	2.04	0.9-2	CaO	32.23	1-3
Effective size(mm)	0.17	0.32	SiO_2	4.99	63
Coefficient of uniformity	1.58	1.8	K_20	0.52	3
Coefficient of gradation	1.37	1.26	LOI	40.63	5

2.3. Marble and Granite Waste Composition and Problems

Chemical composition of the granite and marble vary significantly, and marble absorb more water compared to granite (Table 2). The loss on ignition (LOI) values thus will be high for marble compared to granite. This also has bearing on their physical and mechanical properties.

The dust can affect their surrounding environment in different ways, such as: (1) choking of drains in rainy season, (2) dust nuisance, (3) fine particles of slurry (with size < 363 micron) become air borne and cause air pollution, (4) slurry affects productivity of land due to decreased porosity, water absorption and percolation, (5) slurry dumped areas cannot support vegetation and remain degraded and (6) due to long-term deposition of slurry on land, the finer particles block the flow regime of aquifers (Celik et al., 2008; Rania et al., 2011). Marble being carbonate in composition, dissolves easily in water can affect the plant life (Ciccu et al., 2007; Celik et al., 2008 and Rania et al., 2011). In addition, wear and tear of the steel grit and blades used in processing transfer some quantities of toxic metals like chromium (Celik et al., 2008; Rania et al., 2011). Due to high cation exchange capacity, the waste can increase soil alkalinity, decrease soil fertility and plant productivity (Haggar 2007; Rania et al., 2011).

2.4. Marble and Granite Waste Uses

Based on the physical and chemical properties the waste can be used in different industries. For example, as a coloring agent, aggregate, brick making, as binder, paints, plaster, grout, cosmetics, paper etc (Sentayehu, 2011; Niyaziogurkockal, 2014) (Table 3). Additionally, because of its low hardness and solubility, it is also used as an animal feed, calcium additive in conjunction with a small calcium carbonate pill, to protect against acid reflux. In the chemical industry the marble waste is used for acid neutralization in acidic reactions. Since it has high amount of Ca, it is used for dairy cows and chickens that lay eggs, and because of its low hardness abrasive, it is used to scrub kitchen and bathroom fixtures. (Sentayehu, 2011; Niyaziogurkockal, 2014).

Table 3. Marble and granite waste utilization in percentage (Niyaziogurkockal, 2014; Sentayehu, 2011).

Use	***Concrete, roofing, and aggregate***	***Cement and Ceramic***	***Bricks and tiles***	***Paint, plaster, and binder***	***Land disposal***	***Structural fill and soil stabilization***	***Particle board & wood substitute***
%	10	18	10	7	17	11	10

3. METHODOLOGY

10 waste samples, 5 each from granite and marble were collected from Semayata and Saba dimensional stone processing units. They were subjected to various tests at Axum University which included atterberg limit, grain size, specific gravity, density, water absorption, mix design (liquid limit, plastic limit, and plastic index). Physical and chemical analyses were also carried out at Aksum University (Chemistry and Mechanical Engineering Labs).

3.1. Atterberg Limits and Grain Size

The plasticity of marble and granite processing waste particles was determined in accordance with ASTM D 4318-00 - Liquid limit and Plastic limit. The grain size of the marble and granite residue was determined by sieve analysis according to ASTM C136-01. The particle size of the marble and granite wastes was determined by wet sieving in accordance with ASTM D 422-63.

3.2. Specific Gravity, Density and Water Absorption

Density, specific gravity and water absorption of marble and granite waste was determined based on ASTM C127-07. The measured water absorption, specific gravity and density of the granite and marble waste were correlated with the international standards and calcite minerals. In addition, both granite and marble waste were analyzed based on their specific gravity, density, and water absorption.

3.3. Chemical and Mechanical Analysis

The samples were first dried in a dry oven, crushed in crusher, homogenized, and pulverized. 100gram sample was collected after coning and quartering. Major oxide data was obtained (SiO_2, MgO, Fe_2O_3, CaO, MnO, Na_2O, K_2O, Cl, SO_3 and P_2O_3) using X-ray fluorescence at Mesebo Cement Factory Central Laboratory using internal standards. For testing mechanical properties, as suggested by Rania et al. (2011), the mixed brick residue was mixed with 0, 10, 20, 30, 40 and 50 wt% raw clay and briquette samples (5.0 x 2.5 x 2.5 cm) were made. The mixing was carried out in a planetary mill and at least 100 briquettes of achievable durability were formed, and the samples were dried in an oven at 110 °C for 24 hours. Briquette samples were treated for two hours at 600 to 900 °C under atmospheric oxidation conditions at a heating rate of 10°C/minute. After sintering at selected temperatures, the samples were subjected to several tests to verify their geotechnical properties (compressive strength, crack resistance, water absorption, porosity, and bulk density). The compressive strength was determined by dividing the maximum load by the applied load area of the brick samples (Rania et al., 2011). The flexural strength was measured with a universal testing machine (Bend test machines) in a flexible three-point test at a constant loading speed of 0.5 mm/min. The water absorption, porosity and density of most relevant samples were determined using the Archimedes water displacement method.

4. RESULTS AND DISCUSSION

4.1. Chemistry of Marble and Granite Waste

Granite waste shows higher concentration for SiO_2 (62.84 to 70.68%), with lower LOI (maximum 2%), followed by Al_2O_3 (10.44 to 13.41%), Fe_2O_3 (5.56 to 8.65%), CaO (2.89 to 5.25%) and low concentration of K_2O and Na_2O (2.51 to 3.56) as shown in figure 1. On the contrary, marble shows CaO as the major component (>56%), with much higher level of LOI (maximum 20%), with SiO_2 values between 0.56 and 1.2 %, Al_2O_3 values of 0.16 to 0.56%, MgO values of 0.15 to 0.32%, and traces of Na_2O and K_2O (0 to 0.15 %). The higher values of LOI for marble is ascribed to carbonates. These results compare well with the published data for the waste by Vijayalakshmi et al. (2010); Rania et al. (2008, 2011); Torres et al. (2009).

The concentrations of SO_3, P_2O_5, Cl and MnO in the waste in both granite and marble are insignificant. Chemical analysis results suggest that SiO_2 dominates the granite waste while CaO the marble waste. The oxides in granite waste follow the trend from higher to lower as $SiO_2 > Al_2O_3 > Fe_2O_3 > Na_2O > K_2O >$ CaO > MgO. In the case of marble, CaO > MgO > $SiO_{2,} > Fe_2O_3 > Al_2O_3 > Na_2O > K_2O$ (Table 4 and Fig 2). Higher amounts of Fe_2O_3 in granite waste compared to marble suggest the contribution from iron grit used in the cutting as abrasive material (Rizzo et al., 2008; Menezes et al., 2005; Lin et al., 2006 and Corinaldesi et al., 2010). CaO and K_2O in both granite and marble waste will act as a fluxing agent during the sintering process (Saboya et al., 2007).

Table 4. Chemical analysis results in waste of marble (M1 - M5) and granite (G1 - G5) (wt.%).

S. No	***SiO₂***	***Al₂O₃***	***Fe₂O₃***	***CaO***	***MgO***	***Na₂O***	***K₂O***	***SO₃***	***P₂O₅***	***Cl***	***MnO***	***LOI***
M1	0.57	0.56	0.21	55.26	0.25	0.12	0.11	0.08	0.06	0.02	0.05	20
M2	0.78	0.25	0.32	54.35	0.32	0.06	0.03	0.12	0.07	0.01	0.06	23
M3	1.2	0.18	0.4	56.32	0.3	0.11	0.09	0.07	0.12	0.03	0.08	2.5
M4	0.56	0.16	0.11	45.6	0.2	0.15	0.08	0.06	0.04	0.02	0.04	15
M5	1.12	0.45	0.19	56	0.15	0.07	0.07	0.13	0.05	0.04	0.07	3.6
G1	62.84	12.22	8.65	4.15	0.17	3.32	3. 56	0.06	0.08	0.05	0.12	1.7
G2	65	13.41	7.74	3. 71	0.08	2.51	2.99	0.04	0.05	0.02	0.08	2
G3	68.5	11.51	7.25	3.52	0.11	2.89	3.23	0.05	0.04	0.01	0.06	1.8
G4	70.68	10.44	6. 37	5.25	0.15	2.95	2.89	0.11	0.03	0.04	0.09	1.6
G5	63.45	13.11	5. 56	2.89	0.09	3.11	3.75	0.07	0.06	0.02	0.11	1.3

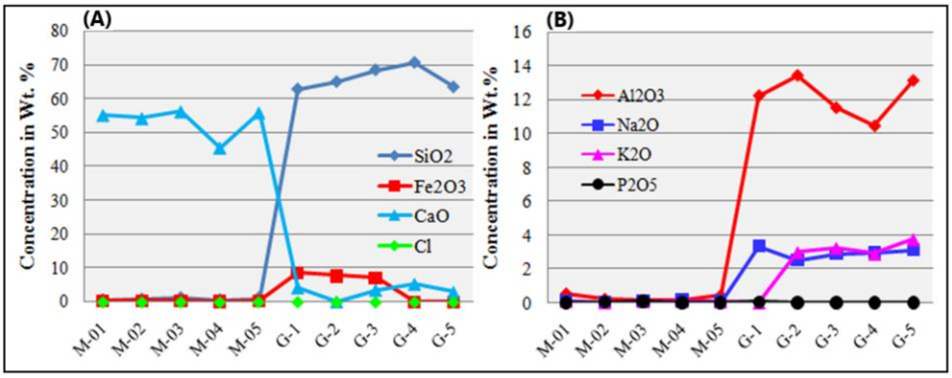

Figure 2. The chemical composition of marble and granite waste, A) the trend of CaO, SiO_2, Fe_2O_3 and Cl; B) Al_2O_3, Na_2O, K_2O and P_2O_5.

4.2. Particle Size Analysis and Atterberg Limits

The grain size of marble and granite waste are <75 microns, with 90% of the samples <25µm in marble and <35 µm in granite. In terms of diameter, 50% is <5µm from marble, and <8 µm from granite wastes. Thus, the size range from clay to silt grain size. So, the finer the material, the greater the water holding capacity and the harder the brick produced. Both marble and granite wastes show shrinkage limits, though they are non-plastic materials. According to ASTM D427-98-Shrinkage factors of wastes by the mercury method, the shrinkage limits as a percentage of dry mass (SL) of marble and granite is 23.25, and 27.25 respectively and shrinkage ratio (R) of 1.51 and 1.47 respectively. These results are similar with those reported by Rania et al. (2011).

In addition to grain size, atterberg data is important check the quality of the final product as air drying shrinkage can distort the high-water content body. As the Atterberg limits are closely related to the optimum moisture content in the soil mass, the plasticity index, liquid, and plasticity limit decrease significantly as the level of rejection increases (Table 5). These results are consistent with the results reported by Menezes et al. (2005), which is suitable to produce bricks. Similarly, the durability of a brick is affected by water absorption and porosity i.e., the less water enters the brick, the longer the brick will last (Vijayalakshmi et al., 2010, Rizzo et al., 2008 and Torres et al., 2009).

Table 5. Average liquid limit, plasticity limit and plasticity index values (N=10).

Marble and granite wastes (%wt.)	***0***	***10***	***20***	***30***	***40***	***50***
Liquid limit (%)	42.2	39.6	37.5	35.4	32.8	30.7
Plasticity limit (%)	24.3	22.6	20.9	19.2	17.0	15.2
Plasticity index (%)	17.9	17.0	16.6	16.2	15.8	15.5

4.3. Mechanical Behavior

For the evaluation of the mechanical properties, the samples were prepared by mixing clay at levels of 0%, 10%, 20%, 30%, 40% and 50% marble and granite waste powder. After firing at selected temperatures, the samples were tested for compressive strength, bending fracture force, water absorption, porosity, and bulk density. Initially, the compressive and flexible breaking forces of standardized industrial bricks were measured. The strength achieved for 10% residue at 600°C is approximately equal to the forces measured for standardized industrial bricks. With residues up to 20%, 30%, 40% and 50% added to the clay mixture, and the strengths measured at 900°C, shows higher values for higher % residue mixing compared to lower. The strengths, water absorption, porosity and bulk density values obtained in this study for mixing clay with marble and granite are much better and meets the specifications of the bricks manufacturing industry under the same conditions (Saboya et al., 2007; Delgado et al., 2006; Almeida et al., 2007; Dhanapandian et al., 2008). Thus, waste dust of both marble and granite can act as a potential raw material to produce bricks. The higher brick density is directly proportional to the amount of waste added to the mixture and temperature (Vijayalakshmi et al., 2010; Rizzo et al., 2008).

Table 6. Compressive strength (MPa), water absorption (%) and powder (%) as well as standard grades of the bricks (as per ASTEM-55 concrete brick and as per IS 1077 burnt clay bricks).

	M1	***M2***	***M3***	***M4***	***M5***	***G1***	***G2***	***G3***	***G4***	***G5***
Compressive strength	13.22	14.24	13.84	14.12	13.93	14.33	13.99	14. 48	14.55	13.89
Water absorption	17.5	19.7	20	18.25	19.99	24.25	19.74	23.5	22.9	23.6
Powder	70	85	90	87	75	87	95	91	78	85
Testing of Bricks (standards)										
As per ASTEM C-55 concrete brick				***As per IS 1077 burnt clay bricks***						
Grade	***Compressive strength (MPa), avg. of 3 units***	***Compressive strength (MPa), individual units***		***Class designation***		***Average compressive strength not less than (MPa),***				
				5		5				
N	24.1	20.7		7.5		7.5				
S	17.3	13.8		10		10				

4.4. Compressive Strength and Water Absorption

Compressive strength test was performed according to IS 3495 (1992) and ASTEM C-140. According to IS 1077 (1992) the average compressive strength of 10 blocks should not be <7.5 MPa for 2nd class bricks. The average compressive strength of the samples in the study is >7.5 MPa thus making them suitable for making brick products. Similarly, according to ASTEM C-55 compressive strength for S class bricks (good quality bricks, which is good in quality compared to the other bricks) for an individual unit must be about 13.8 MPa. The highest compressive strength achieved for granite waste is 14.48 MPa also indicate that the granite waste meets the required standards for S class bricks.

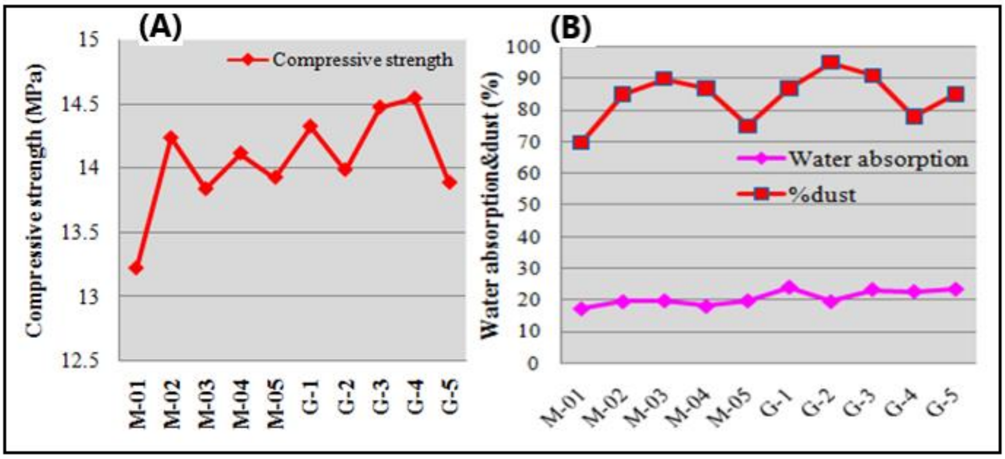

Figure 3. A) Compressive strength, and B) water absorption with respect to the amount of dust.

Regarding water absorption, the test was conducted according to IS 3495 (1992) and ASTEM C-140. According to the standards, the water absorption should not be >20% by weight. All the bricks pass the water absorption test in the present case. When the percentage of dust content from both marble and granite increases uniformly, the compressive strength does not follow the same trend (Table 6 and Fig 3). Hence, the compressive strength of the bricks depends not only on the granite and marble waste content, but also on the additives. The aggregate absorption of granite slurry powder is 23%, due to the high surface area of the particles which requires high water content for saturation, whereas that of marble varies between from 18 and 19% in coarse and fine stone aggregates.

From figure 3, one should notice that the compressive strength and water absorption values for the bricks decreases with increased percentage of dust, thereby increasing its weathering resistance. This result indicates that marble and granite wastes are sufficiently fused at higher temperatures and acted as flux agent. The bricks made with clay normally have a bulk density of 1.8-2.0g/cm^3 (Lin et al., 2006). Thus, the internal structure of the brick must be intensive enough to avoid the intrusion of water.

5. CONCLUSION

The collected waste samples indicate that they can be used for the production of brick without modifying their mechanical properties. The presence of marble and granite waste makes it possible to obtain a clay brick with better properties as conventional bricks at low temperatures, as is commonly used for brick products in the brick industry, resulting in energy saving and waste reduction and finally they ensure an environmentally friendly recycling product that is safe for health and environment.

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