Utilization of Grantitoid Rocks in Taif Area as Raw Materials in Ceramic Bodies

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Abstract: Traditionally, granite is used to produce porcelain and stoneware in Saudi Arab. In the present investigation, the granite raw materials were collected from Taif area (Wade Elnoman). The nature granite was characterized with respect to their mineralogical composition by X-ray diffraction, chemical analysis, DTA analysis and microscopic examination. In the second part of the work, the magnetic separation method give a final non-magnetic concentrate (conc.) with relatively high of K2O and Na2O and minimal amount of Fe2O3 and TiO2. All the obtained final non-magnetic concentrate fulfills the chemical and mineralogical constitution required in ceramic industry. In third part of the work, the preparation and characterizing of ceramic bodies with nature granite, concentrate granite and feldspar were studies. Ceramic batch's were made by wet-mixing, drying, pressing (semi-dry press) and fired at temperatures from 1000 to 1350 C. The technological properties determined by physical properties and mechanical strength. Microstructure and phases analysis of the fired bodies were carried out by XRD and SEM. [Journal of American Science 2010;6(10):691-701]. (ISSN: 1545-1003).

Keywords: granite, magnetic separation, ceramic bodies, XRD, XRF, DTA, SEM

1. Introduction:

Granite is a common and widely occurring type of intrusive, felsic, igneous rock. Granites are usually medium to coarse grained, occasionally with some individual crystals larger than the ground mass forming a rock known as porphyry. Granites are pink to dark gray or even black, depending on their chemistry and mineralogy (Xiaa et al., 2008). Saudi Arabia is becoming a relatively large market for local and foreign granite usage. New practices have been introduced in house construction that includes usage of granite tiling of houses. Even when old houses are renovated conventional tiles are replaced with granites (Al-Jarallah et al., 2005).

Granite marbles are the natural stone which are formed under the earth at the compression of heat and pressure. It is an igneous and metamorphic rock which is resistant to extreme temperature, stress and heat. It is easy to wash and clean the dirt, stains and spills using the strong cleaning agent. Granite is the ideal material for countertops, because of its hardest and durable substance. It is a stylish material which combines its natural beauty along with durability. It is subject to heat, water, moisture and salt resistant and also for abrasive material. It can be used for both indoor and outdoor decorations (Xiaa et al., 2008).

Fluxes are raw materials with a high amount of alkaline oxides, mainly K2O and Na2O, which, in reaction with silica and alumina, promote liquid phase formation that facilitates the densification. The liquid phase surrounds the solid particles and by surface tension enables the particles approach, closing the porosity (Vieira el al., 2004; Kingery, 1975; Emiliani et al., 1999; Schmidt-Reinholz and Schmidt, 1995). Granite is considered as flux material due to its large content of alkaline oxides. These oxides derive from feldspars and micaceous minerals that are common constituents of granite rocks (Vieira el al., 2004).

Previous works have utilized granite (Vieira el al., 2004; Mohamed et al. 1992; Robert et al. 1992) in the production of ceramic products. The major observations were that granite shows physical and mineralogical characteristics similar to the raw materials used in the body composition and that the technological properties of some mixture fulfill the required properties. The incorporation of granite in a conventional porcelain stoneware body improved the sintering process and reduced the water absorption (Vieira el al., 2004; Hernández-Crespo and Rincón 2001). However, results obtained by incorporating granite into red ceramic bodies for bricks (Vieira el al., 2004; Menezes et al., 2002), indicated a reduction in mechanical strength and increase in water absorption (Vieira el al., 2004).

In the development and manufacture of ceramics using granite materials, the properties of fired bodies are determined basically by the combination of raw materials and process parameters. When the processing conditions are kept constant, a
number of properties of dried and fired bodies are
determined principally by the combination (or
mixture) of raw materials (Menezes, 2008; Cornell,
2002). That is the basic assumption in the statistical
design of mixture experiments to obtain a response
surface using mathematical and statistical techniques
(Menezes, 2008; Myers and Montgomery, 2002;
Cornell, 2002). To this end, it is necessary first to
select the appropriate mixtures from which the
response surface might be calculated. With the
response surface in hand, a prediction of the property
value can then be obtained for any mixture, based on
changes in the proportions of its components
(Menezes, 2008; Cornell, 2002; Correia et al.,
2004). This methodology has found important applications
in various areas and is becoming popular in the field
of glasses and ceramics (Cornell, 2002; Correia et al.,
2004; Chick and Piepel, 1984; Nardi et al., 2004; Yoon and Lee, 2004; Khalifaoui et al., 2006). In every
reported case, the methodology has led to greater
efficiency and confidence in the results obtained
(Correia et al., 2008) and has simultaneously
optimized the content of raw materials with a
minimum of experiments (Menezes, 2008).

Some researchers (Nardi et al., 2004; Abdakhimova and Abdakhimov, 2006) have
reported using waste materials (Menezes, 2008). One
technological method, which is already being tested
to decrease porosity, is the incorporation of granite
wastes from the sawing process (Monteiro et al.,
2004; Souto et al., 2000; Patri´cio et al., 1998).
Granite is a rock with large amount of quartz,
feldspars and mica. In the initial stages of firing,
these minerals act as non plastic agents which permit
the use of lower amount of water in body forming.
This makes for an easier drying operation. During
firing the quartz generally behaves as an inert
material but may also partially dissolve in liquid
phases, should they occur. Both the feldspars and
mica favor the formation of liquid phases and
contribute to lower the porosity of the final ceramic
product (Monteiro et al., 2004).

In the present paper the effect of granite on
ceramic bodies was investigated to find out the
optimum incorporation. An attempt was made to give
an explanation for the technological results obtained
in terms of flux action on the decreasing of porosity
and their effect on the sintering/densification
behavior and mechanical properties of a ceramic
material.

2.2. Experimental procedure:
2.1. Materials and methods

Samples of materials weighting from 0.5 to
2.0 kg were collected from construction sites and
local suppliers. Six samples of the granite from
different locations in the wade elnoman western taif,
45km from Maka and Gamgogm area northern east
Jeddah parts of Saudi Arabia were also collected. It
Can be used the waste of granite from block granite
decoration. This waste granite is different size
between 170 x220 cm. The average dimension of the
waste granite samples was around 12x12x2 cm3 and
their mass varied from 0.5 to 1.7 kg.

For ceramic bodies, it can be used granite in
wade elnoman in Saudi, feldspar from red sea area
(Safaga in Egypt), Quartz from Alqoser in area and
kaolin from Sinai in Egypt.

2.2. Rock granite description and samples preparation

The raw materials used in this investigation
were collected from location. These raw materials
were dried at 110 °C, manually crushed before
representative samples were separated by quartering.
The second stage grandee's process for samples was
separated by sieves analysis. This particle size which
apparently corresponds to the liberation size, as
confirmed by independent mineralogical analyses,
was found to be optimum after a series of grinding
tests.

2.3. Magnetic separation

The representative ore sample used in
studies was obtained from Wade Elnoman, in Saudi
Arabia. According to the chemical analysis and the
mineralogical studies included microscopic
examination of thin sections; the representative ore
sample contains microcline, albite, quartz and opaque
minerals. To obtain high quality product and
recovery, high intensity of wet magnetic separator
was used for ~200 and 100 µm particle sizes.

Flotation experiments were conducted with
~200 and ~100 µm test samples in a self-aerated
laboratory flotation cell. Conditioning time kept
constant as 5 min for rougher circuit and 3 min for
cleaning circuits. Potassium oleate (1000 g/t) was
added at 4 steps in natural pH of 7.6. Pine oil was
used as a frother in the first stage and in subsequent
stages.

The characterization included chemical
composition (X-ray fluorescence, EDX-700,
Shimadzu), mineralogical composition (X-ray
diffraction, XRD of 0.5 g of randomly oriented
powder was carried out in a Sheifert model URD 65,
diffractometer, equipped with a graphite
monochromator, operating with Cu K_ radiation for a
2θ range from 5° to 40°). The chemical composition
was carried out by fluorescence spectrometry
(Philips, PW 2400) on 7.8 g of pressed powder
pellets. The loss on ignition (L.O.I) was obtained by
determining the weight difference between samples
calcined at 1000 °C and dried at 110 °C, thermal

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behavior (Differential thermal analysis. DTA was run with a coupled (SETARAM TG/DTA 92) DTA-TGA instrument) and The microstructure of the samples was studied by scanning electron microscopy, SEM, using a Zeiss model DSM 962 equipment. The thin sections were investigated by LEITZ SM-LUX POL microscope with the light source from the bottom.

2.4. Preparation of ceramic bodies

Selected five ceramic mixtures containing 25% feldspar, 25-30% granite and 25-30% concentrated granite after processing were prepared and homogenized for 4 h in a planetary mill with alumina grinding balls (see in Table1).

Table 1: Batches composition (mass %) of the ceramic bodies.

<table>
<thead>
<tr>
<th>Batches</th>
<th>Kaolin %</th>
<th>Quartz %</th>
<th>Feldspar %</th>
<th>Granite %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Batch 1</td>
<td>60</td>
<td>15</td>
<td>25</td>
<td>---------</td>
</tr>
<tr>
<td>Batch 2</td>
<td>60</td>
<td>15</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td>Batch 3</td>
<td>60</td>
<td>10</td>
<td>30</td>
<td>-----------</td>
</tr>
<tr>
<td>Batch 4</td>
<td>60</td>
<td>15</td>
<td>25 (conc.)</td>
<td>30</td>
</tr>
<tr>
<td>Batch 5</td>
<td>60</td>
<td>10</td>
<td>30 (conc.)</td>
<td>-----------</td>
</tr>
</tbody>
</table>

The powders were uniaxially pressed into test disc (25 mm diameter x 5mm thickness) and bars (50x10x10 mm³) under a load of 20 MPa. All specimens were dried at room temperature for 48 h and then at 110 °C in a laboratory oven until constant weight were achieved. Powder compacts and drying were sintered at temperatures between 1000 and 1350 °C for 2 h, with a heating rate of 300 °C/h. The heating rate and the sintering time used in this work were chosen to simulate the actual sintering process used in the ceramic industry.

Five batches for each composition were then tested to obtain the firing technological properties related to the bulk density, water absorption and three points flexural rupture strength. The bulk density was measured dividing the dry mass by the external volume. Water absorption was determined by using the Archimedes water displacement method, as specified by the ISO 10545-3 standard. The flexural rupture strength was determined in an Instron 5500 Universal Testing Machine, according to standard procedure (ASTM, 1977).

3. Results and Discussion:
3.1. Characteristics of the rock granite

Table 2 shows the chemical composition of the raw materials. The chemical composition of the granite is typical of feldspar-based material with high amount of silica oxide, low amounts of alkaline oxides and relatively low amount of Al₂O₃. The chemical composition of the granite, in addition to SiO₂ and Al₂O₃, shows a relatively low amount of alkaline and alkaline earth oxides. The granite is containing 3.77% Fe₂O₃, 0.45% TiO₂, 3.37% Na₂O and 6.25% K₂O. This confirms the flux potential of the granite. The significant amount of Fe₂O₃ in the raw materials is responsible for the reddish color of the specimen after firing.

Fig. 1 shows the XRD patterns of the granite raw materials. These diffractograms indicate that the granite presence of a micaceous mineral, quartz, albite and microcline can be observed. Albite and microcline are sources of alkaline flux materials, such as K₂O and Na₂O, which favor the formation of a liquid phase above 700 °C (Reed, 1976). The fluxing capacity of the granite, which is associated with lower porosity after firing, is also confirm by the presence of K₂O and Na₂O containing minerals. The granite is the sources of K₂O and Na₂O, which act as fluxes to improve the sintering process.

Fig. 2 shows the thermal behavior of the granite. The DTA curve shows two endothermic peaks at 40.70 and 736.46 °C that are associated with the weight loss. Fig. 2 shows also an exothermic peak at 1112 °C, that can be attributed to the formation of mullite (3Al₂O₃·2SiO₂).

The mineralogical analyses reveal that granite ore is composed of quartz, microcline, orthoclase, plagioclase and albite. The microscopic examination is presented in Figs. 3 and 4 which indicate that the rock granite characterizes by hypidomorphic texture at particles size between medium and big size. It is containing two type of feldspar as plagioclase and microcline.
Table 2: Chemical analysis of granite raw materials from Wade Elnoman area.

<table>
<thead>
<tr>
<th>sample No.</th>
<th>SiO₂</th>
<th>Al₂O₃</th>
<th>Fe₂O₃</th>
<th>TiO₂</th>
<th>CaO</th>
<th>MgO</th>
<th>Na₂O</th>
<th>K₂O</th>
<th>MnO</th>
<th>SO₃</th>
<th>P₂O₅</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>68.10</td>
<td>14.00</td>
<td>5.50</td>
<td>0.65</td>
<td>1.09</td>
<td>0.13</td>
<td>3.55</td>
<td>6.00</td>
<td>0.12</td>
<td>≤0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>2</td>
<td>70.35</td>
<td>14.1</td>
<td>3.70</td>
<td>0.45</td>
<td>1.13</td>
<td>0.06</td>
<td>3.25</td>
<td>6.50</td>
<td>0.10</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>3</td>
<td>70.56</td>
<td>13.33</td>
<td>4.10</td>
<td>0.50</td>
<td>0.99</td>
<td>0.07</td>
<td>3.45</td>
<td>6.40</td>
<td>0.11</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>4</td>
<td>71.60</td>
<td>13.96</td>
<td>3.30</td>
<td>0.38</td>
<td>0.74</td>
<td>0.05</td>
<td>3.30</td>
<td>6.22</td>
<td>0.09</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>5</td>
<td>71.63</td>
<td>14.00</td>
<td>3.20</td>
<td>0.37</td>
<td>0.84</td>
<td>0.05</td>
<td>3.35</td>
<td>6.24</td>
<td>0.09</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>6</td>
<td>71.95</td>
<td>14.05</td>
<td>2.84</td>
<td>0.35</td>
<td>0.85</td>
<td>0.07</td>
<td>3.20</td>
<td>6.15</td>
<td>0.07</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Average</td>
<td>70.70</td>
<td>13.91</td>
<td>3.77</td>
<td>0.45</td>
<td>0.94</td>
<td>0.08</td>
<td>3.37</td>
<td>6.25</td>
<td>0.1</td>
<td>0.05</td>
<td>0.05</td>
</tr>
</tbody>
</table>

Fig. 1: XRD pattern of raw granite. Q: quartz, M: microcline, A: albite, F: iron oxide.

Fig. 2: DTA of the behavior of raw granite material.

Fig. 3: Equigranular texture of quartz (Q), orthoclase perthite (OPr), microcline perthite. Microcline crystal shows cross-hatched twinning (MPr), plagioclase (P), biotite (B) and iron oxides (Fe).

Fig. 4: Perthitic texture is also occurring due to the exsolution of albite within K-feldspar (Pr). Plagioclase (P) shows lamellar twinning with somewhat altered to sericite.
The main constitutes of rock granite consist of feldspar and quartz minerals and secondary minerals as biotite, fluorite and iron oxide. The quartz in granite is characteristic by crystal face incomplete which represented crystallization in equilibrium crystallization environment and containing some small crystal inter the monoclinic feldspar. Feldspar consists of orthoclase crystal incomplete or microcline crystal, both crystals reflection as perthite consists of orthoclase crystal incomplete or microcline feldspar. Feldspar which represented crystallization in equilibrium granite is characteristic by crystal face incomplete as biotite, fluorite and iron oxide. The quartz in feldspar and quartz minerals and secondary minerals granite concentrate containing 0.27% Fe$_2$O$_3$ and 0.12% TiO$_2$ was produced by the weight of 84.5%.

3.2. Magnetic separation

3.2.1. Dry magnetic separation

The sample between -200 and +38 µm was fed to high intensity dry magnetic separator. The granite concentrate containing 0.27% Fe$_2$O$_3$ and 0.12% TiO$_2$ was produced by the weight of 84.5%.

3.2.2. Flotation after magnetic separation

In this study, the granite ore was first subjected to magnetic separation then the concentrate was cleaned of colored impurities using flotation with the optimum conditions. At the end of the high intensity dry magnetic separation followed by flotation tests performed with -200 and + 38µm, a concentrate containing 0.18% Fe$_2$O$_3$ and 0.05% TiO$_2$ was obtained by the weight of 76.6%.

The magnetic separation method gives a final non-magnetic concentrate (conc.) containing 0.18% Fe$_2$O$_3$ and 0.05% TiO$_2$, with relatively high of K$_2$O and Na$_2$O show in Table 3. The anther part is tailing contain high amount of Fe$_2$O$_3$ and TiO$_2$. All the obtained final non-magnetic concentrate fulfills the chemical and mineralogical constitution required in ceramic industry. The curves in Fig 5, respectively, illustrate the XRD patterns of granite and granite concentrate; the latter indicates that the intensity of quartz and albite peaks decreases compared to the granite raw. But, the intensity of orthoclase peaks increases; these results clearly indicate the success of selective separation.

3.3. Behaviors of granite (conc.) in the ceramic bodies.

3.3.1. Characteristics of the raw materials in ceramic bodies.

Table 4 gives the chemical compositions of the raw materials used in this work. The clay material presents a typical composition and is constituted mainly by silica and alumina and minor contents of Mg, Ti, Ca, Na and K oxides. The significant amount of iron oxide (2.73 wt. %) is responsible for a white beige coloring of the sintered samples. The loss on ignition (13.22%) is within usual range for white-clay material and is associated with volatile components and organic matter. The raw of granite is formed basically by SiO$_2$, Al$_2$O$_3$, K$_2$O and Na$_2$O with small amounts of MgO, Fe$_2$O$_3$ and CaO. The high alkaline earth oxide content (particularly K$_2$O and Na$_2$O); present in the granite material will act as a fluxing agent during the sintering process. Fig. 1 shows the X-ray diffraction patterns of granite material. It can be noted that the granite contains quartz, microcline and albite. The crystalline phases identified are in agreement with the results observed by XRF (Table 2). Table (5) presents the calculated chemical composition of the investigated mixtures. One should notice that with increasing concentrated granite addition there occurs an increase in the percentage of alkaline fluxes and alumina content as compared to the simple ceramic body in batch 1 (see in Table 1).

3.3.2. Physical properties

Figures 6-7 show the graphs corresponding to the results of the technological properties after firing. The maturing temperature of ceramics batches were studies. The batches 1 and 4 are containing feldspar and concentrated granite at 25% have maturing temperature at 1300 ºC while the batches 2 and 3 are containing raw granite at 25-30% have maturing temperature at 1200ºC. But, the batch 5 contains concentrated granite at 30% has maturing temperature at 1350 ºC. These results indicate that the concentrated granite addition increases the bulk density (see in Fig. 7), while the water absorption in Fig. 6 is decreased. The increase bulk densities as well as the decrease in water absorption with concentrated granite and feldspar addition are consequences of the higher fluxes content in Table 5, as compared to that of the raw granite as batches 2 and 3. But, the result indicate that the concentrated granite batches 4 and 5 increase the bulk density and decreased the water absorption are better than feldspar addition in batch 1 (see in Fig. 6 and 7). At higher temperatures the fluxes agent cause an increase in the liquid phase promoting densification (Vieira et al., 2004, Hernández-Crespo and Rincón 2001).

This indicates that the observed differences on the technological properties of the batches are not influenced by the bulk density or compactness of the ceramic. It is also important to mention the significant reduction observed on the water absorption with concentrated granite addition. For example, the body with 25 and 30 wt. % of concentrated granite as batches 4 and 5, shows water absorption 0.47% , 0.67% lower than the body contain feldspar as batch1, the water absorption 0.87%, and with raw granite as batches 2 and 3 the water absorption 0.66% and 1.40%. This is an indication that the formation of a larger amount of liquid phase contributes to reduce porosity.
The result indicates the behavior of raw granite less than concentrated granite and feldspar from maturing temperature at 1200 °C due to the impurities of Fe₂O₃ which act as reflux agent in ceramic bodies.

Table 3: Chemical analysis of concentrate granite after magnetic separation

<table>
<thead>
<tr>
<th>Oxide</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>average</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>74.05</td>
<td>73.85</td>
<td>73.80</td>
<td>75.75</td>
<td>74.35</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>13.00</td>
<td>13.66</td>
<td>13.50</td>
<td>12.50</td>
<td>13.17</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>0.30</td>
<td>0.06</td>
<td>0.30</td>
<td>0.07</td>
<td>0.18</td>
</tr>
<tr>
<td>TiO₂</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>CaO</td>
<td>0.11</td>
<td>0.42</td>
<td>0.30</td>
<td>0.16</td>
<td>0.25</td>
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<tr>
<td>MgO</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Na₂O</td>
<td>3.88</td>
<td>4.20</td>
<td>3.90</td>
<td>3.92</td>
<td>4.00</td>
</tr>
<tr>
<td>K₂O</td>
<td>8.30</td>
<td>7.44</td>
<td>8.00</td>
<td>7.15</td>
<td>7.80</td>
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<tr>
<td>MnO</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>SO₃</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
<td>0.05</td>
</tr>
</tbody>
</table>

Table 4: Chemical composition (wt. %) of the granite conc. and raw materials used for bodies' composition design.

<table>
<thead>
<tr>
<th>Oxide</th>
<th>SiO₂</th>
<th>Al₂O₃</th>
<th>Fe₂O₃</th>
<th>TiO₂</th>
<th>CaO</th>
<th>MgO</th>
<th>Na₂O</th>
<th>K₂O</th>
<th>MnO</th>
<th>P₂O₅</th>
<th>LoI</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unite</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
</tr>
<tr>
<td>Raw granite</td>
<td>70.70</td>
<td>13.91</td>
<td>3.77</td>
<td>0.45</td>
<td>0.94</td>
<td>0.08</td>
<td>3.37</td>
<td>6.25</td>
<td>0.05</td>
<td>0.05</td>
<td>0.4</td>
</tr>
<tr>
<td>Granite conc.</td>
<td>74.00</td>
<td>13.17</td>
<td>0.18</td>
<td>0.05</td>
<td>0.25</td>
<td>0.05</td>
<td>4.00</td>
<td>7.80</td>
<td>0.05</td>
<td>0.05</td>
<td>0.40</td>
</tr>
<tr>
<td>Feldspar</td>
<td>67.33</td>
<td>16.45</td>
<td>0.41</td>
<td>0.04</td>
<td>1.48</td>
<td>0.17</td>
<td>3.00</td>
<td>10.78</td>
<td>---</td>
<td>0.01</td>
<td>0.33</td>
</tr>
<tr>
<td>Quartz</td>
<td>99.25</td>
<td>0.55</td>
<td>0.15</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>Kaolin</td>
<td>52.10</td>
<td>27.45</td>
<td>2.73</td>
<td>0.72</td>
<td>0.43</td>
<td>0.82</td>
<td>0.58</td>
<td>1.82</td>
<td>0.05</td>
<td>0.08</td>
<td>13.22</td>
</tr>
</tbody>
</table>

Table 5: Chemical analysis of the mixtures (wt. %) for bodies' composition design.

<table>
<thead>
<tr>
<th>Oxide Batches</th>
<th>SiO₂</th>
<th>Al₂O₃</th>
<th>Fe₂O₃</th>
<th>TiO₂</th>
<th>CaO</th>
<th>MgO</th>
<th>Na₂O</th>
<th>K₂O</th>
<th>MnO</th>
<th>P₂O₅</th>
<th>LoI</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unite</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
<td>%</td>
</tr>
<tr>
<td>Batch1</td>
<td>62.98</td>
<td>20.66</td>
<td>1.76</td>
<td>0.44</td>
<td>0.63</td>
<td>0.53</td>
<td>1.10</td>
<td>3.80</td>
<td>0.02</td>
<td>0.02</td>
<td>8.06</td>
</tr>
<tr>
<td>Batch2</td>
<td>63.83</td>
<td>20.03</td>
<td>2.60</td>
<td>0.54</td>
<td>0.50</td>
<td>0.51</td>
<td>1.19</td>
<td>2.66</td>
<td>0.02</td>
<td>0.02</td>
<td>8.10</td>
</tr>
<tr>
<td>Batch3</td>
<td>62.40</td>
<td>20.70</td>
<td>2.79</td>
<td>0.57</td>
<td>0.53</td>
<td>0.51</td>
<td>1.36</td>
<td>2.98</td>
<td>0.02</td>
<td>0.02</td>
<td>8.12</td>
</tr>
<tr>
<td>Batch4</td>
<td>64.65</td>
<td>19.83</td>
<td>1.71</td>
<td>0.44</td>
<td>0.31</td>
<td>0.50</td>
<td>1.35</td>
<td>3.05</td>
<td>0.01</td>
<td>0.01</td>
<td>8.14</td>
</tr>
<tr>
<td>Batch5</td>
<td>63.39</td>
<td>20.48</td>
<td>1.71</td>
<td>0.45</td>
<td>0.33</td>
<td>0.51</td>
<td>1.55</td>
<td>3.46</td>
<td>0.02</td>
<td>0.02</td>
<td>8.10</td>
</tr>
</tbody>
</table>
Fig. 5: XRD pattern of granite (G1) and granite concentrate (G2). Q: quartz, M: microcline, A: albite, F: iron oxide.

Fig. 6: Water absorption of fired ceramic bodies with temperatures

Fig. 7: Bulk density of fired ceramic bodies with temperatures

3.3.3. Mineralogical composition of the ceramics bodies

The sintering behavior is closely related to the mineral phases formed during the firing process and the materials obtained by firing show the smaller percentage of water and maximum bulk density. The chemical composition of the tested bodies is shown in Table 2. It has been detected by XRD that the crystalline phases of the fired samples at the corresponding temperatures (Fig. 8). Therefore, it has been demonstrated that the firing products of the batches 1, 2, 3, 4, and 5, at temperatures of 1200, 1300 and 1350 °C respectively, are constituted by quartz and mullite minerals coming from the kaolin, granite and concentrated granite.

Mullite is formed by the reaction of the metakaolinite that is provided by the dehydroxilation process of the kaolinite materials. This mullite phase is hardly detected by XRD, which indicates that it can be considered as a primary mullite and secondary is extremely big. The mullite peaks could be clearly detected at 1200 °C especially for the batches 2 and 3. The intensity of the quartz peaks tend to decrease with increasing sintering temperatures due to their gradual dissolution into glassy phase. This was more evident for batches 2 and 3. The intensity of fluxing minerals, Fe₂O₃ included, tends to decrease for temperatures, as expected. Batches 1, 4 and 5 have been high intensity of mullite peaks tend to
increasing sintering temperature due to their growth.

In general, firing resulted in the formation of mullite and glassy phase, with some non-dissolved quartz still remaining in the sintered bodies. As the concentrated granite addition increases, the peak intensities of mullite also increase. The common crystalline phases between batches 1,2,3,4 and 5 are quartz and mullite. While quartz is a residual phase, mullite results from spinel phase, with an approximate composition of 2Al₂O₃, 3SiO₂, formed at temperatures around 925 ºC (De La Fuente et al., 1990; Johns, 1965).

3.3.4. Mechanical strength

The flexural strength is a property strongly dependent on the porosity and microstructural defects of the specimen. In principle, due to the densification, one would expect an increase on the flexural strength with the concentrated granite addition (see in Table 6). This result can be explained by the higher amount of quartz content, Table 5, introduced with the granite and concentrated granite addition. The quartz can promote the appearance of micro-cracks due to their volumetric variation at 573 ºC. These micro-cracks are known to act as stress raisers, contributing to reduce the mechanical strength (Abajo, 2000; Kilikoglou et al., 1995).

All ceramic bodies increase their flexural strength with temperature in approximately the same way. There is a tendency toward higher strength values for the experimental ceramic body batch 5, which has the greater amount 30% in Table 1 of concentrated granite. For this class of materials, the mechanical strength is determined by the stress concentration in structural defects such as pores, voids and microcracks (Schneider, 2000). The bending strength for fired bodies at maturing temperature is increased from 31.1 to 45.4 MPa with increase the bulk density from 2.3062 -2.5119 g/cm³ (see in Table 6). The ceramic bodies which containing concentrated granite higher in bulk density and bending strength than the raw granite and pure feldspar in Table 6. The ceramic batch 5 has high bulk density at 2.5119 g/cm³ and bending strength at 45.4 MPa, because high amount of concentrated granite.

3.3.4. Microstructure

The morphologies of the crystalline materials were different from those observed in ceramics bodies. This difference in the crystal morphology may be due to the difference in the characteristic of raw materials used in conventional ceramic viz. clay, quartz and feldspar. Fig. 8 shows SEM observations on the fracture of the ceramic body fired at 1200-1350 ºC. It is observed the typical grain and bond microstructure of porcelain consisting of quartz grains held together by a finer matrix or bond that is almost fully dense (Lee and Rainforth, 1995). The matrix is composed by primary mullite, originated from pure clay agglomerate relics and consisting in aggregates of small scaly crystals and secondary mullite composed of elongated needle shape crystals derived from feldspar–clay relics (Iqbal and Lee, 1999; Ludin, 1964). The SEM of the ceramic bodies 1, 4 is containing quartz crystal and mullit. This mullite showed a needle-like microstructure (mullite whiskers), which is characteristic of mullite obtained by the decomposition of feldspar or concentrated granite (see in Fig.9 and 11). The sintering bodies 1, 4 and 5 have high mechanical strength because microstructure formation of mullite whiskers. Figures 10 is shows small crystalline quartz dissolving in a glassy matrix and mullite crystals. This mullite was a needle crystal form and little mullite whiskers in sintering bodies 2 and 3, this crystals form is effect on mechanical strength. The sintering bodies 2 and 3 are less mechanical strength than sintering bodies 1, 4 and 5 because the crystal form of mullite is strong influence on the microstructure formation.

Table 6: Properties of the firing ceramic bodies.

<table>
<thead>
<tr>
<th>Batches</th>
<th>Maturing temperature ºC</th>
<th>Water absorption %</th>
<th>Bulk density (g/cm³)</th>
<th>Bending strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Batch 1</td>
<td>1300</td>
<td>0.87</td>
<td>2.33</td>
<td>40.5</td>
</tr>
<tr>
<td>Batch 2</td>
<td>1200</td>
<td>0.66</td>
<td>2.31</td>
<td>32.5</td>
</tr>
<tr>
<td>Batch 3</td>
<td>1200</td>
<td>1.40</td>
<td>2.31</td>
<td>31.1</td>
</tr>
<tr>
<td>Batch 4</td>
<td>1300</td>
<td>0.47</td>
<td>2.37</td>
<td>41.7</td>
</tr>
<tr>
<td>Batch 5</td>
<td>1350</td>
<td>0.67</td>
<td>2.51</td>
<td>45.4</td>
</tr>
</tbody>
</table>
4. Conclusions
1- The chemical composition of the granite is typical of feldspar-based material with high amount of silica oxide, low amounts of alkaline oxides.
2. According to the mineralogical studies, the representative ore granite contains microcline, albite and quartz.
3. The intensity of the magnetic separation and flotation methods were applied separately and combined on the raw granite to produce an alternative raw material to feldspar using ceramic industries.
4- The incorporation of granite raw and concentrated granite up to 30 wt. % in ceramic bodies has led to the following conclusions:
   • The concentrated granite incorporation decreases the open porosity of the fired bodies. Consequently, the water absorption decreased with the concentrated granite content and increases the bulk density. The flexural strength is a property strongly dependent on the porosity, bulk density and microstructure.
   • The SEM microstructure analysis showed that the composition with 30 wt. % of concentrated granite has a finer fracture surface with fewer defects as compared to the granite raw incorporation.
   • XRD analysis indicated that no significant effect on the crystalline phases was observed with concentrated granite incorporation.
   • The use of concentrated granite in ceramic bodies can be a technological solution to the problems caused by the indiscriminate disposal into the environment.

Acknowledgment
The financial support provided by Taif University is gratefully acknowledged. The authors are also thankful to National research center in Egypt.
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5. References:


8/2/2010