CHARACTERIZATION OF THERMAL TREATED CLAY OBTAINED FROM LEBANESE LOCAL RESOURCES ASSUPPLEMENTARY CEMENTITIOUS MATERIALS

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BAALBAKI, OUSSAMA Professor, Department of Civil & Environmental Engineering, Faculty of Engineering; ABDALLAH, BACHAR; Elkordi, Adel; and Khatib, Jamal (2018) "CHARACTERIZATION OF THERMAL TREATED CLAY OBTAINED FROM LEBANESE LOCAL RESOURCES ASSUPPLEMENTARY CEMENTITIOUS MATERIALS," BAU Journal - Health and Wellbeing. Vol. 1 : Iss. 3 , Article 37. Available at: https://digitalcommons.bau.edu.lb/hwbjournal/vol1/iss3/37

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Abstract
The purpose of this research was to evaluate a kaolin clay obtained from local Lebanese source after being subjected to thermal treatment. A sample of kaolin clay was collected from local resources and subjected to a treatment process for enhancing its pozzolanic properties. The treated sample was characterized through testing procedure as per ASTM C311. It was found that the physical and chemical properties were in compliance with the relevant ASTM C618 standard. This indicates that the proposed treatment succeeded in developing and exhibiting the pozzolanic activity of the end product. The activity index test of the treated samples was investigated to verify the quality and performance of the end product which was found to be satisfactory. Therefore, the treated product is accepted to be classified as natural Pozzolan as per ASTM C618. The potential of producing Metakolin in Lebanon is of high importance due to its availability in large quantities. Moreover, it might be used to replace part of the Portland cement to reduce the CO2 emission and energy consumption resulting from the cement production.

Keywords
Calcined Clay, Activity Index, Natural Pozzolan, Metakaolin)
CHARACTERIZATION OF THERMAL TREATED CLAY OBTAINED FROM LEBANESE LOCAL RESOURCES AS SUPPLEMENTARY CEMENTITIOUS MATERIALS

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ABSTRACT: The purpose of this research was to evaluate a kaolin clay obtained from local Lebanese source after being subjected to thermal treatment. A sample of kaolin clay was collected from local resources and subjected to a treatment process for enhancing its pozzolanic properties. The treated sample was characterized through testing procedure as per ASTM C311. It was found that the physical and chemical properties were in compliance with the relevant ASTM C618 standard. This indicates that the proposed treatment succeeded in developing and exhibiting the pozzolanic activity of the end product. The activity index test of the treated samples was investigated to verify the quality and performance of the end product which was found to be satisfactory. Therefore, the treated product is accepted to be classified as natural Pozzolan as per ASTM C618. The potential of producing Metakolin in Lebanon is of high importance due to its availability in large quantities. Moreover, it might be used to replace part of the Portland cement to reduce the CO₂ emission and energy consumption resulting from the cement production.

KEYWORDS: Calcined Clay, Activity Index, Natural Pozzolan, Metakaolin

1. INTRODUCTION

Use of Supplementary Cementitious materials is nowadays a new challenge kurdì et al. (1992 & 1997). Development of construction materials from available resources for use in concrete offers technical and environmental benefits (Baalbaki, 1994; Khatib and Mangat, 2003; Mangat et al 2006; Khatib, 2014). Active Metakaolin (MK) is one of these cementitious materials (Khatib, 2009; Khatib et al 2009). MK is composed of kaolin clay, consisting predominantly of silica and alumina, while the remaining portion contains small amount of iron, calcium and alkalies. MK is a natural Pozzolan obtained from the process of calcination of kaolin Clay at a temperature range from 600°C to 800°C resulting in the formation of amorphous alumina-silicate. This reaction occurs under specific conditions of heating and cooling. The hydrated products, resulting from the cement hydration process; calcium-silicate hydrate (CSH) and the lime reacts with MK to form additional CSH that densify the cement paste leading to a stronger concrete. The MK is incorporated into concrete by partial substitution of cement to improve the fresh and hardened concrete properties. The engineering benefits are manifested by the direct effect on the different properties of concrete such as compressive strength, permeability, workability; PCA book) 2009, another advantage of using MK is that it is first naturally occurring and do not need blasting of quarries and second is the cost of MK production which is lower than that of cement due to incineration at lower temperature. This would be beneficial in reducing the rate of CO₂ which is 0.9 tones emissions per ton of cement. The MK is generally used with typical addition about 10% to 20% by mass of cement when very low permeability is needed or a high compressive strength.

Several studies have been made on the treatment process of MK and its effect on the chemical composition of the treated material and on the properties of concrete. Concerning the strength activity index, it shows that kaolin increase the concrete compressive strength when treated appropriately by a thermal method and used within the correct dosage Biljana et al (2010). Aboubakar et al (2013) studied the strength activity index of Libyan kaolin treated by a thermal method, where natural materials from Libya was heat treated, and then the
resulting materials where introduced in cement to find its effects on strength activity index BS EN 196-5 (2011). MK does not only improve the concrete strength but also enhances other properties such us workability, durability, permeability, setting time. Nova John (2013) has studied the strength properties of MK admixed concrete. Concerning the thermal treatment technique of kaolin clay and its effect on the pozzolanic activity, Tironi et al (2012) studied the influence of different thermal treatments on the pozzolanic activity of raw kaolin with 98% kaolinite. Poon et al. (2011) conducted studies on rate of pozzolanic reaction of MK in cement paste. The objective of this research is to evaluate the physical and chemical properties of the calcined kaolin clay obtained from local resources after treatment in the aim of using it in concrete to improve its performance in terms of strength and durability. The activity index of Pozzolanic MK was the main test used to validate the use of MK as SCM to partially substitute cement in concrete

2. CLAY SOURCES IN LEBANON

Clay is distributed overall the Lebanese territory. The type of clay in Lebanon varies among the different regions of Lebanon. A study showed (CNRS 1994; National Council of Science Research, Information and Documentation Center) that the soils having high kaolin content are localized in the central Bekaa valley, south east of Tripoli, and the southern part of Lebanon. Central Bekaa, south Bekaa and north of Tripoli also showed week kaolin contents. It was found that the southern region of Lebanon near to the Litani River showed a unique clay composition comprising high kaolinite content (40-50%), and all of the fluvial systems between the rivers Litani and Awali manifesting similar signatures.

3. TREATMENT PROCESS

The clay sample used for the preparation of MK was collected from Debbieh, Lebanon, located at the South- East of Beirut and North-East of Saida. The geographical location of the sample was 33°40'26" N and 35°27'53" E at an altitude of 372 meters above the sea level. The sample of 5 Kg weight was dried in the oven for 24 hours at 105°C. The sample was ground using balls mill grinder. The raw materials resulting from the grinding process were fine materials of brownish color. Following grinding process the resultant materials of size mostly finer than 75 micron were collected for the final phase of the heating process. The sample was heated in the furnace at 800 °C. After cooling, it was removed and stored for testing and evaluation

![Figure 1. Transformation of Clay from dark brown (left) to reddish (right) after heating](image)

4. CHARACTERIZATION

The ASTM C618 (2015) covers the specifications of Fly Ash, and other raw or calcined natural pozzolan for use in concrete. MK comply with the requirements of class N (Natural pozzolan) whereas fly ash meet the requirements of class F or C based on the bituminous coal type and burning process. Sampling and testing procedure for fly ash and raw or calcined pozzolans for use in Portland-cement concrete are covered in The ASTM Standard C311 (2017), Table 1.

<table>
<thead>
<tr>
<th>Fineness (max % retained on 45)</th>
<th>Min Strength Activity</th>
<th>Minimum SO$_3$+Al$_2$O$_3$+Fe$_2$O</th>
<th>Max SO$_3$</th>
<th>Max LOI</th>
</tr>
</thead>
<tbody>
<tr>
<td>34%</td>
<td>75%</td>
<td>70%</td>
<td>4%</td>
<td>10%</td>
</tr>
</tbody>
</table>

4.1. Chemical Composition: X-Ray Fluorescence Analysis

The principle of the test is that an X-Ray collides with the atom and disturb its equilibrium and making it unstable. The energy released to restore stability is emitted as a secondary X-Ray which is detected by the detector. The preparation of the material for X-Ray analysis starts by drying the sample
at 105˚C till obtaining constant mass. The moisture content was found to be equal to 1.89 %. A portion of the dried sample was fused at high temperature. Another portion of the dried sample was taken to test the loss on ignition, noted as “LOI”. LOI is determined by igniting the sample at 950˚C until constant mass. Two samples were tested conducted at in two different laboratories for confirmation of the obtained results (Table 2).

The summation of oxides (Si₂O₃ + Al₂O₃ + Fe₂O₃) in both samples was found to be greater than 70%: 82.1 % and 87.45% which comply with the relevant ASTM C618 standard related to natural pozzolan. The requirement related to sulfur content SO₃ was 0.09% which is very low in comparison to the target specification (< 4%). The obtained Loss of Ignition which is 5.35% is also satisfactory with respect to the limit specified by ASTM C618 (LOI < 10%). Thus, the chemical composition of the treated material meets the required chemical requirements of the natural pozzolan as per ASTM C618. The XR diffractometer indicated that the selected sample is classified as weak kaolin of low kaolin content.

### Table 2. Chemical Oxides Composition

<table>
<thead>
<tr>
<th></th>
<th>Sample</th>
<th>Sample B</th>
<th>Requirem</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>56.96</td>
<td>56.07</td>
<td>&gt; 70%</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>15.73</td>
<td>20.72</td>
<td></td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>9.42</td>
<td>10.65</td>
<td></td>
</tr>
<tr>
<td>CaO</td>
<td>5.46</td>
<td>5.51</td>
<td></td>
</tr>
<tr>
<td>MgO</td>
<td>2.1</td>
<td>2.6</td>
<td></td>
</tr>
<tr>
<td>SO₃</td>
<td>0.09</td>
<td>0.09</td>
<td>&lt; 4%</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.4</td>
<td>0.13</td>
<td></td>
</tr>
<tr>
<td>K₂O</td>
<td>1.13</td>
<td>1.17</td>
<td></td>
</tr>
<tr>
<td>P₂O₅</td>
<td>0.2</td>
<td>0.08</td>
<td></td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>0.036</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>LOI %</td>
<td>5.35</td>
<td>-</td>
<td>&lt; 10%</td>
</tr>
</tbody>
</table>

### 4.2. Physical Characteristics

#### 4.2.1. Wet Sieving

The wet sieving test was performed according to ASTM C430, Standard Test Method for Fineness of Hydraulic Cement by the 45-μm (No. 325) Sieve. The sample of 2 g weight was wetted carefully with a gentle stream of water. Materials passed and retained on the sieve were dried in and the percentage of materials retained on the sieve was calculated. Two samples were tested; one with grinding followed by heating treatment and the other one with only grinding. The results obtained showed that the percentage retained on sieve No. 325 (45μm) was 20% for both samples which are less than the ASTM C618 maximum specified limit (34%). So, the natural raw materials meets the standard requirement to be considered as natural pozzolan in terms of fineness properties. It was concluded that the heating process does not affect the degree of fineness.

#### 4.2.2. Specific Surface Area by Physical Adsorption (B.E.T)

The BET (Brunauer, Emmett and Teller) theory is commonly used to evaluate the gas adsorption data and generate a specific surface area result in m²/g. Prior to analysis, the sample was preconditioned to remove physically bonded impurities from the surface of the powder in a process called degassing or outgassing. This is typically accomplished by applying elevated temperature to the sample in conjunction with vacuum or continuously flowing inert gas. The specific surface is then determined by the physical adsorption of a gas (typically nitrogen) onto the surface. The test result indicated that the specific surface area of the treated material tested was 31.7319 m²/g. The specific surface of the treated material is considered very high in comparison to the natural pozzolan and meets the required condition to be considered as natural pozzolan as per ASTM C618. This high value which is greater or equal to that generally obtained on micro- silica is might be due to the high energy of grinding resulting in very fine particles.
4.3. Strength Activity Index

The activity index test was performed according to ASTM 311 Standard Test Method using 50 mm cubical Specimens. Four sets were prepared for comparison purpose. The first set consists of the control mortar specimens. The second set consisted of partial substitution of cement by 10% of micro-silica. The remaining two sets were respectively substituted by 10 and 20% of treated materials. The testing program is indicated in Table 3. The mortar cubes were prepared and tested at 7 and 28 days according to ASTM C109. The w/c was 0.48 and the cement to standard sand proportion was 1:2.75. The activity index is the ratio of compressive strength at 7 and 28 days for the mortars with partial replacement of cement over that of control specimens at the same age.

As shown in Table 4 and Figures 2 and 3, the activity index of the sample with 10% substitution by silica fume has the largest strength activity index which is obvious. Both treated samples with 10% and 20% partial substitution of cement gave good results; greater than 75% which meet the ASTM C618 related to natural Pozzolan. It is to be noted that the 20% is considered the optimal one in terms of economical and strength performance (105.8%). However, the production cost of MK is lower than that of cement since the heating temperature is lower; 800°C instead of 1500°C.

Table 3. Testing Program

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>CON</td>
<td>Control Mix</td>
</tr>
<tr>
<td>S.F</td>
<td>10% Silica fume</td>
</tr>
<tr>
<td>A10</td>
<td>10% Treated Clay</td>
</tr>
<tr>
<td>B20</td>
<td>20% Treated Clay</td>
</tr>
</tbody>
</table>

Table 4. Compressive strength and Activity Index Test Results

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Compressive Strength</th>
<th>Activity Index</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>7 days</td>
<td>28 days</td>
</tr>
<tr>
<td>CON</td>
<td>32.11</td>
<td>49.63</td>
</tr>
<tr>
<td>S.F</td>
<td>40.92</td>
<td>59.91</td>
</tr>
<tr>
<td>A10%</td>
<td>33.40</td>
<td>49.23</td>
</tr>
<tr>
<td>A20%</td>
<td>33.98</td>
<td>51.88</td>
</tr>
</tbody>
</table>
5. CONCLUSIONS

Based on the evaluation process and the test results, it was found that the resulting product from the proposed methodology of treatment, is qualified to be classified as per ASTM C618, a natural pozzolan that meets the physical and chemical requirements. Considering the high specific surface (30 m²/g) and high activity index (105%) achieved by this new proposed material, it can be classified as metakaolin that can be used in concrete to improve its hardened properties and durability. The high fineness of the material was due to the high efficiency of grinding. The time of grinding can be controlled and reduce to minimize the cost of production. The replacement of cement by metakaolin is generally beneficial since it enhances both fresh and hardened concrete properties. Further research is required to be conducted on different local sources of clay with higher kaolin content for evaluation purposes. The proposed product can be locally produced and is classified as natural materials and not waste product which may contain hazardous materials. However, fresh and hardened properties need to be determined on concrete containing locally produced metakaolin for validation of the product’s performance.

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