The development of Indonesian local clay as a lightweight expanded clay aggregate (LECA) for organic growing medium

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Abstract. Lightweight Expanded Clay Aggregate (LECA), a clay-based lightweight aggregate expanding after burning at high temperatures, is among the promising material since it can be used as construction materials for buildings and roads, geotechnical fillings, hydroponic growing medium, humidity control, wall damper, heat insulation, water treatment, and others. However, the potential of clay from various places in Indonesia used for LECA as an organic planting medium is still not explored considerably. In this paper, various local clays in Indonesia were studied to obtain a suitable clay as the single raw material for LECA production. Clay raw materials were originated from Purwakarta, Sukabumi, Klaten and Boyolali. The preliminary selection was evaluated based on XRD and XRF test results. The XRD and XRF analysis showed that Boyolali shale and Boyolali bentonite were the most fitted clay as single raw material for LECA production. Both materials were synthesized into LECA through a firing process at 1100°C and 1200°C. Furthermore, the LECA product was characterized by its bloating coefficient and water absorption. The best results were obtained by Boyolali bentonite with a bloating coefficient of 4.63 or can be referred to as medium type and 14.63% for water absorption.

1. Introduction

There are three types of lightweight aggregate. The first type is produced by mechanical methods, grinding the natural raw material to a certain size. The examples material for the first type were pumice and rock tuff. The second type is produced by the combustion method in a rotary kiln, this type comes from clay, shale, or bentonite that is commonly named LECA. And the last is industrial wastes-by products, fly ash or slag [1].

Lightweight Expanded Clay Aggregate (LECA) is a clay-based lightweight aggregate that expands after being burned at high temperatures (1050°C - 1250°C) [2]. The use of LECA is very diverse including construction materials for buildings and roads [3], geotechnical fillings, hydroponic growing media [4], geology, civil and environmental engineering, agricultural and greenhouse issues [5].

LECA has been used as an organic growing media for gardening and agriculture. When compared with other organic growing media such as coconut fiber, charcoal, tile pieces, and so on, LECA has various advantages. LECA can regulate drainage, maintain moisture, protect roots from freezing and store nutrients and help oxygen flow so that plant maintenance will be easier and cheaper. Moreover,
LECA could be applied in urban areas that do not have much green land. Its lightweight properties make LECA as a very good alternative to be used for vertical farming in a building.

LECA structure is formed through 2 (two) simultaneous events during the firing process, the formation of gas, and the formation of a very viscous liquid phase. After a cooling process, the liquid phase changes to glass and form hollow spaces. Gases trapped in the glass phase are the result of organic and inorganic materials decomposition when the material is subjected to high temperatures. The gas formed during the heating process can be in the form of CO, CO₂, SO₂, H₂O, O₂ and N₂. As temperature and gas concentration increase, the pressure of gases in melted glass increases which results in the expansion of material volume known as bloating [3]. Table 1 shows the classification of clays as raw material for LECA based on the bloating coefficient.

| No | Coefficient of Bloating (Kp) | Degree of Bloating |
|----|-----------------------------|--------------------|
| 1  | > 7 – 8                     | > 4                | Good/ self-bloating |
| 2  | 4 – 5                       | 2.5 - 4            | Medium level        |
| 3  | 2 – 2.5                     | 2–2.5              | Poor               |
| 4  | < 2                         | < 2.5              | Not-self bloating  |

Clay minerals may be grouped into four types, there are kaolinite group, smectite group (montmorillonite group), illite group, and chlorite group [7]. Among all of these groups, montmorillonite is the most swelling clay. Water molecules cause swelling in montmorillonite. The migration of counter-ion, initially bound to montmorillonite surface to the central interlayer plane, leads to swelling in montmorillonite [7]. Montmorillonite is one of mineral contained in clays which could slowly release the crystal of water up to 1200°C provided the gas contributing to the bloating of the clay [8].

LECA chemical composition consists of SiO₂, Al₂O₃, fraction of Fe₂O₃, CaO, MgO and alkalis Na₂O and K₂O [9]. According to Vaicklionis [3] and Rashad [9], self-bloating properties of clay influenced by its ratio of SiO₂ and Al₂O₃ and sum of oxides contains (Fe₂O₃, CaO, MgO, Na₂O and K₂O) or its chemical composition meet requirements as shown in table 2.

| No | Composition | Vaicklionis [3] (%) | Rashad [9] (%) |
|----|-------------|---------------------|---------------|
| 1  | SiO₂        | 50 - 60             | 53,3 - 70     |
| 2  | Al₂O₃       | 15 - 20             | 15,05 - 27    |
| 3  | F₂O₃        | 7 - 8               | 1 – 14,3      |
| 4  | CaO         | < 8                 | 0,2 – 3,92    |
| 5  | CaO + MgO   | < 5                 |               |
| 6  | R₂O         | 2 - 3               |               |
| 7  | organic     | 0,5 - 3             |               |

Clay bloatability can be enhanced by an addition of organic materials (heavy fuel oil, wood sawdust, fine turf, coal, waste of rubber) or inorganic material (iron compounds, glass granules, cement, fly ash) [3].

Indonesia has abundant clay mineral natural resources but it is currently used for the production of tiles, tableware, potter, and earthenware ceramics [10]. The utilization of clay as raw material of LECA is an alternative to give value added to Indonesia’s local clays. In this study, clay from Purwakarta, Sukabumi, Klaten, and Boyolali were examined. This study aimed to obtain a suitable clay as a single raw material for LECA production.
2. Materials and methods

2.1. Materials
Pasir Pining clay, Sandaan clay, and Cibodas clay were obtained from the Service Technical Implementation Unit of Ceramic Research and Development, Purwakarta. Melikan clay was obtained from Klaten Pottery Conservation Center laboratory. Clay Sukabumi and bentonite Sukabumi were obtained from Department of Industry and Energy Mineral Resources Sukabumi District. Meanwhile, shale and bentonite Boyolali were obtained from Department of Trade and Industry Boyolali District.

2.2. Method and characterization
The mineral contained in each material was investigated using XRD Bruker D8 Advance, then tested for chemical composition using XRF ADVANT XP Thermo ARL9900. After the chemical composition evaluation, the selected clays were dried under the sun for 5 days, grinded, sieved to 0.14 mm – 18 mm size, added with potable water, formed by manual pelletizer then fired in the rotary kiln, length 3 m, 0.3 rpm at temperature 1100°C and 1200°C.

3. Results and discussion

3.1. Mineralogical characterization of clay raw material
The clay raw materials were originated from Purwakarta (Pasir Pining, Sandaan, Cibodas), Sukabumi (Sukabumi bentonite), Klaten (Melikan) and Boyolali (bentonite and shale). The result of clay raw material characterization using XRD was shown in figure 1 to figure 3. Figure 1 were the results of the XRD characterization of Pasir Pining, Sandaan, Cibodas (Purwakarta) samples. From the three diffractogram pattern, all of them indicate the presence of halloysite (Al₃Si₄O₁₀(OH)₈·2H₂O), quartz, muscovite (KAl₂(AlSi₃O₁₀)(F,OH)₂). Pasir Pining clay presence of feldspar (KAlSi₃O₈ – NaAlSi₃O₈ – CaAl₂Si₂O₈) had melting point below 1200°C but the main mineral was halloysite, usually known as aluminosilicate clay mineral that had softening point above 1300°C and the presence muscovite could increase the percentage of alkali R₂O. Based on mineralogy, those clays had low viscosity and non-carbonaceous [3]. Clays needed any additives to decrease the melting point between 1100°C and 1200°C and source of material which produces gases to support the bloatability. Therefore, those clays were not suitable to be applied as single raw material of LECA.

The XRD characterization for Melikan clay from Klaten was shown in figure 2. The diffractogram pattern according to figure 2 indicated that the main minerals were halloysite, α-quartz, muscovite, calcium and illite. Even though halloysite mineral had a high softening temperature which was above 1300°C, but with the presence of muscovite and calcium; the softening temperature of Melikan clay could be lowered into 1100°C. Besides, in this clay presence illite mineral as one of non-expanding clay. Therefore, Melikan clay could not be used as a single raw material for LECA products and required other materials such as potassium feldspar, pure kaolin, or other alkalis that had a high melting point or additives to improve the degree of bloating.

Figure 3 illustrated the XRD diffractogram for Gunung Guruh clay from Sukabumi and indicated halloysite, muscovite, montmorillonite, calcite, and small fraction of α-quartz minerals. The presence of calcite shown that clay was carbonaceous clay which could play a role in the bloating process [11]. Montmorillonite as acid clay would provide releasing gases to support the bloatability. The presence of muscovite provided high alkali R₂O. As a result, by the presence of (1) alkalis as flux and bloating actor; and (2) gases and pore materials, Sukabumi clay had a potential to be developed into LECA product.

The XRD pattern in Boyolali bentonite clay was shown in figure 4. The diffractogram indicated presence of montmorillonite, halloysite, quartz, cristoballite and feldspar minerals. Montmorillonite was contained the acid clay. It released the crystal water slowly up to 1200°C provided the gas contributing to the bloating of the clay [8]. Therefore, Boyolali bentonite clay can be developed into a single raw material of LECA product.
Figure 1. XRD patterns of pasir pining, sandaan and cibodas.

Figure 2. XRD pattern of melikan clay.
Based on XRD pattern from all clay sample; Boyolali bentonite, Boyolali shale and Sukabumi clay were selected to be used in preliminary and main research. These raw materials were the most fitted to develop into LECA product since they only required fewer additives or even used as single raw material. Moreover, the XRD diffactogram analysis must be supported by other characterization in order to some materials consider suitable as LECA raw material especially if it is used as single raw material.

3.2. Chemical characterization of clay raw material
The clay ability to expand was closely related to the oxide composition. The X-RF results were presented in table 3. Sukabumi bentonite clay had high SiO$_2$ that was above 60%, 15% Al$_2$O$_3$, 2.8%
Fe₂O₃, 1.74% R₂O (less than 2%), and (CaO + MgO) more than 5% that is 12,49%. (CaO + MgO) more than 5% also appeared in Sukabumi clay that is 13,36 %. Moreover, it could be seen that Sukabumi bentonite clay and Sukabumi clay had a total flux of 16,90% dan 17,37%, respectively. Sukabumi bentonite clay and Sukabumi clay had a lower SiO₂/fluxing ratio of 3.79 and 3.12, respectively. The lower ratio indicated both clays were non-bloating raw material. That meant both clays had lower viscosity that material could not trap a sufficient amount of gas and prevented from expanding during firing [11]. Therefore, clays did not suitable to be applied as raw material of LECA. However, Sukabumi bentonite clay and Sukabumi clay still could be develop as raw material of LECA by using additives to decrease the total flux and to increased SiO₂/fluxing ratio.

Theoretically, Boyolali bentonite and shale had met the standard requirements for LECA product. These raw materials had (CaO and MgO) less than 2% and total flux less than 10% as could be seen at table 3. In this study, Boyolali bentonite and shale were qualified to be used as single raw material for LECA production.

### Table 3. Chemical composition of bentonite, shale and clay.

| No | Oxide  | Boyolali Bentonite (%) | Boyolali Shale (%) | Sukabumi Bentonite (%) | Sukabumi Clay (%) |
|----|--------|------------------------|--------------------|------------------------|-------------------|
| 1  | SiO₂   | 51.17                  | 61.20              | 64.00                  | 54.28             |
| 2  | Al₂O₃  | 17.62                  | 17.63              | 15.18                  | 9.65              |
| 3  | Fe₂O₃  | 7.32                   | 8.14               | 2.67                   | 1.17              |
| 4  | MgO    | 1.46                   | 1.90               | -                      | 4.42              |
| 5  | CaO    | 1.52                   | 1.44               | 12.49                  | 8.94              |
| 6  | K₂O    | 1.89                   | 2.09               | 1.74                   | 1.10              |
| 7  | Na₂O   | 0.91                   | 1.54               | -                      | 1.74              |
| 8  | TiO₂   | 0.72                   | 0.77               | 0.32                   | 0.05              |
| 9  | SO₃    | 0.35                   | 0.35               | 0.63                   | -                 |
| 10 | MnO    | 0.14                   | 0.07               | 0.14                   | -                 |
| 11 | P₂O₅   | 0.08                   | 0.12               | 2.83                   | -                 |
| 12 | LoI    | 14.37                  | 5.62               | -                      | 17.97             |
| 13 | Total flux | 13.10             | 15.11              | 16.90                  | 17.37             |
| 14 | Si/Flux| 3.91                   | 4.05               | 3.79                   | 3.12              |

#### 3.3. Bloating coefficient analysis

According to Vaickelionis [3] the suitable clay for LECA should have bloating coefficient (Kp) at least 2 and initial melting temperature less than 1300 °C [3]. Bloating coefficient analysis was performed on Boyolali bentonite dan Boyolali Shale. Table 4 shows bloating coefficient calculation results. Both materials were synthesized into LECA through firing process at 1100°C and 1200°C. Boyolali bentonite clay firing at 1200°C gave the best result with Kp > 5 implying medium level of bloating coefficient.

Boyolali bentonite gave a better bloating coefficient result than Boyolali shale with firing process at 1200°C. The firing process held important part that can affect bloating phenomenon. Bloating phenomenon occurred at LECA product as the forming of black core due to the reduction of Fe₂O₃ to FeO at high temperature [8]. The reduction also affects the increase of internal gas pressure as shown in following chemical reaction:

\[
6\text{Fe}_2\text{O}_3 (s) \rightarrow 4\text{Fe}_3\text{O}_4 (s) + \text{O}_2 (g)
\]

\[
2\text{Fe}_3\text{O}_4 (s) \rightarrow 6\text{FeO} (s) + \text{O}_2 (g)
\]
Another reduction process occurred in LECA due to organic and carbon oxidation in natural materials as shown in following chemical reaction [12]:

\[
3\text{Fe}_2\text{O}_3 (s) + \text{C} \rightarrow 2\text{Fe}_3\text{O}_4 (s) + \text{CO} (g)
\]

\[
\text{Fe}_3\text{O}_4 (s) + \text{C} \rightarrow 3\text{FeO} (s) + \text{CO} (g)
\]

Those processes produce CO gas that can increase internal pressure and FeO. FeO as flux supposed to decrease the melting point and influence viscosity of material. Kang Hoon Lee, et al, divided the bloating mechanism into three stages: (1) the formation of gas inside material body, (2) the formation of pores by internal pressure and (3) the formation of pores by internal pressure between small and large pores. LECA formation occurred with the forming of gas and pore due to internal pressure. Based on chemical composition results, Boyolali bentonite and shale have LoI of 14.37% and 5.62%, respectively. Boyolali bentonite with higher LoI produced more gases than Boyolali shale. Therefore, this significant point caused the bloating coefficient of Boyolali bentonite was higher than Boyolali shale.

### 3.4. Water absorption analysis

As shown in table 5, the water absorption of LECA which was made of Boyolali bentonite increased as the firing temperature increase. At a temperature of 1100°C, glass phase formation occurred on the surface of the bentonite material before bloating. Through bloating the volume of material expands, porosity got larger and gases release as the temperature rises. Then LECA from bentonite with a temperature of 1200°C had the biggest water absorption, 14.62%. A different case occurred in Boyolali shale, at a temperature of 1100°C the material was still large porous but with increasing temperature the phase glass of the flux will begin to close the pores. Therefore, increasing the temperature would decrease in water absorption properties from 8.46% to 7.49%.

Based on XRD diffractogram pattern and chemical composition analysis, Boyolali bentonite and shale could be used as single raw material for LECA. Both materials were synthesized into LECA.
through a firing process at 1100°C and 1200°C. Furthermore, the LECA product was characterized by its bloating coefficient and water absorption. The best results were obtained by Boyolali bentonite with a bloating coefficient of 4.63 implying medium type and water absorption 14.63%.

4. Conclusion
In this study, the lightweight expanded clay aggregate had been produced using Boyolali bentonite as single raw material without any additives for expanding the bloating coefficient. The water absorption increased with increase firing temperature. These result showed that Indonesia’s clay which currently used as a raw material for ceramic industry could also be used as a growing medium LECA.

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