Granulation of coal fly ash by using different types of granule agents

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Abstract. The use of coal produces about 5% solid pollutant in the form of ash (fly ash and bottom ash). Of the total ash produced, about 10-20% is bottom ash and 80-90% is fly ash. This study was aimed at obtaining a type of adhesive which could be used as a fly granulation material for soil conditioner. The study was conducted at the pilot plant of Surfactant and Bioenergy Research Center (SBRC) LPPM IPB from April to August 2016. The fly ash used in this study was obtained from Kalimantan. A pan granulator was used in fly ash granule making process. Granule agent materials were diluted in the concentration of 5, 10, and 15%. Different types of granule agents, namely SBRC-M, SBRC-T, and SBRC-SC were used. The formed fly ash granules were then analyzed for their physical properties including particle density, fly ash granule pH, fly ash granule durability, and fly ash granule water holding capacity. Results showed that fly ash granules made from 15% of SBRC-M had the highest particle density (0.75 g/cm³). Fly ash granules made with SBRC-M had higher pH (10) than those made by using SBRC-SC adhesive (9.3) and SBRC-T (9). SBRC-T was found as the granule agent material which produced fly ash granules with the highest durability levels on average. In this study, the use of SBRC-M granule agent resulted in higher water holding capacity (WHC) (40.62%) than did SBRC-SC (38.79%) and SBRC-T (36.85%). As a granule agent, compared to SBRC-SC and SBRC-T, SBRC-M could produce fly ash granules with highest particle density, highest pH, good durability, and best water holding capacity.

1. Introduction
The use of coal produces about 5% solid pollutant in the form of ash (fly ash and bottom ash). Of the total ash produced, about 10-20% is bottom ash and 80-90% is fly ash. Physically, fly ash is a material containing granules which are fine in size and gray in color. Fly ash is a heterogeneous mixture of amorphous and crystalline phases and generally considered as ferroaluminosilicate. It comprised of about 69 percent silt and clay size fractions [1]. Chemically, fly ash contains silica (SiO₂), alumina (Al₂O₃), ferric oxide (Fe₂O₃), and calcium oxide (CaO). It also contains some additional compounds and element including magnesium oxide (MgO), titanium oxide (TiO₂), alkaline (Na₂O and K₂O), sulfur trioxide (SO₃), phosphorus oxide (P₂O₅), and carbon. Fly ash was also found to have Ca and Mg contents, high pH value (11), and high cation exchange capacity [2].
Fly ash has been much used as a soil conditioner material to solve agricultural problems in Greece [3], Thailand [4], India [5, 1, 6], USA [7], Japan [8,9], and other countries. Fly ash was proven to be able to repair the structure, bulk density, water holding capacity, conductivity, and particle size of soil [5]. Fly ash, which was alkaline, could increase soil pH. It was also found to increase plant production as it contained high K\textsuperscript{+}, Zn\textsuperscript{2+}, Ca\textsuperscript{2+}, Mg\textsuperscript{2+}, and Fe\textsuperscript{3+} ions which were important for plant growth.

These physical and chemical properties have made the potential of fly ash utilization as ameliorant/soil conditioner very high. In addition to improving soil pH, fly ash also provide nutrients required by plants. However, when it is used as a soil conditioner, there is a concern that fly ash may pollute the air and affect health when it is inhaled as its particles are very small in size and volatile. Therefore, fly ash needs to be granulated in order to make it uneasy to get inhaled by humans and easy to be transported. This study was aimed at obtaining an adhesive which could be used as a fly granulation material for soil conditioner.

2. Methodology
The study was done at the pilot plant of Surfactant and Bioenergy Research Center (SBRC) LPPM IPB from April to August 2016. Fly ash was obtained from Kalimantan. Granules were made by using a pan granulator. Granule agent material was diluted in a concentration of 5, 10, and 15%. During the granule production process, the pan granulator was kept rotating and granule agent spraying was done simultaneously. At least two personnel were needed, one as a stirrer and the other took care of the spraying. The granulation speed was set at 28 RPM. The granulation process was done for 15 minutes to produce granules with a diameter of 2-10 mm. The formed granule pellets were then taken out from pan granulator. The process proceeded to drying stage done by putting the granule pellets under direct sunlight and in an oven. The drying was done until the water content of the granules reached 9-15%. Good granule agents are easy to get, affordable, able to absorb water, and have good adhesive property and dispersion/release time which is correspondent to the rhythm of nutrient absorption by plants. In this study, granule agents were consisting of sugar by-products (SBRC-M), starch (SBRC-T), and soil cement (SBRC-ST).

The produced fly ash granules were analyzed for their physical properties, namely particle density, pH, dispersion time, durability, water holding capacity.

- **Particle density**: Particle density of fly ash granule was measured by putting 100 mL sample in a 100 mL beaker glass. The granules were weighed to get the particle density in 100 cm\textsuperscript{3}.
- **pH**: Five grams of fly ash granules were mixed with water in a ratio of 1:5. pH of the precipitate of the mixture was measured by using a pH meter.
- **Durability**: Five grams of fly ash granules were put in a beaker glass and stirred. Stirring was done by using a shaker set at the speed of 200 rpm for 10 minutes. Five replicates were allocated. When the shaking was finished, intact granules were taken out and weighed. The percentage of unbroken granules was determined.
- **Water holding capacity**: Water holding capacity was measured by referring to the method of [10]. The test was done by soaking the granules in water for an hour. Wet granules were then weighed for their wet weight. The granules were then ovened at 60°C for 24 hours before they were weight for their dry weight. The test was done to determine the ability of soil conditioner granule to absorb water. Water holding capacity was calculated by using the following equation:

\[
\text{Water holding capacity} = \frac{\text{Wet weight} - \text{dry weight}}{\text{Wet weight}} \times 100\%
\]
3. Results and discussion

3.1 Particle density

Particle density is the weight of fly ash granules per a volume unit. Mineral soil has a particle density of 2.6 g/cm³ [11,12]. Particle density is stated in gram per volume unit (cm³). Low value of its particle density established its potential for dust formation. The bulk density of fly ash was 1.01 mg/m³. Organic carbon content in the ash was 0.36 percent [1].

Results showed that the particle density of fly ash granules made from SBRC-M, DBRC-SC, and SBRC-T was 0.74, 0.68, and 0.67 g/cm³, respectively. Organic soils in general have a particle density of 0.5-0.8 g/cm³. The particle density of fly ash granules used in this study is listed in Table 1.

| Granule agent (concentration) | Particle density | ST. Dev |
|-------------------------------|-----------------|--------|
| SBRC-M 5% (M5)                | 0.73c           | 0.01   |
| SBRC-M 10% (M10)              | 0.74b           | 0.01   |
| SBRC-M 15% (M15)              | 0.75a           | 0.00   |
| **Average**                   | **74.08**       |        |
| SBRC-SC 5% (SC5)              | 0.65f           | 0.01   |
| SBRC-SC 10% (SC10)            | 0.70d           | 0.01   |
| SBRC-SC 15% (SC15)            | 0.69d           | 0.01   |
| **Average**                   | **67.91**       |        |
| SBRC-T 5% (T5)                | 0.65f           | 0.01   |
| SBRC-T 10% (T10)              | 0.68e           | 0.01   |
| SBRC-T 15% (T15)              | 0.68e           | 0.01   |
| **Average**                   | **66.80**       |        |

It was found that fly ash granules made from 15% of SBRC-M had the highest particle density (0.75 g/cm³) while that made from SBRC-T 5% and SBRC-SC 5% shared the lowest particle density (0.65 g/cm³). In general, higher adhesive concentration results in higher particle density. This can be used as a consideration in selecting adhesive material as particle density gives significant effect on granule packaging and transportation. The size of granule pellets affected the results of this study. Bigger granule size made more space in the beaker glass which then affected the total weight of the granules in the beaker glass.

Similar to soil, there were several factors including texture, organic material, structure, and bulk density affecting the particle density of fly ash granule [13]. Texture is a visual appearance of soil or granule resulted from qualitative composition of granule pellet size in a given granule mass. The texture of fly ash granule represents the composition of particles constituting the granule. High organic material content gives significant effect on particle density as found in coal fly ash granule used in this study. Fly ash obtained from coal-fired power plant was a very fine particle or a dust material. The size of fly ash of coal-fired power plant origin varies from the finest of 11-25 μm to the coarsest of 40-150 μm.
3.2. pH

Plants require complete nutrients including macro, micro, and beneficial elements in proportional amount. Fertilizer formula containing these elements is beneficial in improving plant productivity and maintaining nutrient balance in the soil. Acidity level (pH) is an important factor affecting the process of nutrient absorption by plant roots. Soil with neutral acidity level is the most ideal condition as most nutrients are easily dissolved in water.

Soil pH level also shows the presence of elements which are toxic to plants. Aluminum (Al) is found a lot in acidic soil. Al is not only toxic to plants but it also binds phosphorus making the phosphorus not available for adsorption by plants. In addition, acidic soil contains excessive micro elements which could intoxicate the plants. In contrast, sodium (Na) and molybdenum (Mo) are found a lot in alkaline soil. Soil pH also affects microorganism development in soil. Organic material decomposer fungi and bacteria grow well in soil with pH 5.5-7. In such soil, microorganisms which are beneficial for plant roots also grow well.

Liming is commonly used to overcome acidity and high level of Al saturation in soil. Liming is done to increase soil pH from acidic to rather neutral or neutral and lower Al content. Lime has a pH of 8.5-10. Fly ash has a pH level of 11.7 and high content of K⁺, Zn²⁺, Ca²⁺, Mg²⁺, and Fe³⁺ ions which are important for plant growth. Dolomite commonly used by farmers only contains Ca²⁺ and Mg²⁺. It has even been proven that fly ash was able to improve the structure, bulk density, water holding capacity, conductivity, and particle size of soil [1]. Therefore, fly ash is potential to be used as soil conditioner or ameliorant. The addition of fly ash increased the soil reaction (pH), the concentration of available P, exchangeable K, Na, calcium and magnesium [14].

In this study, the resulted fly ash granules had a pH of 9-10. It was found that types of granule agent significantly affected fly ash granule pH. Fly ash granules made with SBRC-M had higher pH (10) than those made by using SBRC-SC adhesive (9.3) and SBRC-T (9). This indicated that fly ash granules produced in this study could be used as materials to improve soil pH. pH levels of fly ash granules made by using different granule agents are depicted in Figure 2.
3.3. Durability

This test was aimed at assessing the durability of fly ash granules under a condition where there were certain friction and stress. Durability is a physical property indicating the granule’s resistance to damages during transportation. The percentage of intact fly ash granules after steering process is listed in Table 2.

![Graph of the effects of granule agent types on fly ash granule pH.](image)

**Figure 2.** Graph of the effects of granule agent types on fly ash granule pH.

**Table 2.** Percentage of durable fly ash granules after stirring process

| Material        | Durability (%) | St. Dev |
|-----------------|----------------|---------|
| SBRC-M 5% (M5)  | 76.39          | ±8.29   |
| SBRC-M 10% (M10)| 93.91          | ±1.73   |
| SBRC-M 15% (M15)| 92.53          | ±3.53   |
| **Average**     | **87.61**      |         |
| SBRC-SC 5% (SC5)| 86.74          | ±2.88   |
| SBRC-SC 10% (SC10)| 92.09        | ±2.74   |
| SBRC-SC 15% (SC15)| 90.69        | ±1.71   |
| **Average**     | **89.84**      |         |
| SBRC-T 5% (T5)  | 87.50          | ±6.21   |
| SBRC-T 10% (T10)| 93.99          | ±3.33   |
| SBRC-T 15% (T15)| 95.69          | ±1.48   |
| **Average**     | **92.39**      |         |

It was shown in Table 2 that fly ash granules made by using SBRC-M had lower average durability (87.61%) than those made by using SBRC-SC (89.84%) and SBRC-T (92.39%). SBRC-T was found as the granule agent material which produced fly ash granules with the highest durability levels on average. The addition of fly ash can increases strength of soil [15].

The results of this study were found to be in line with the work of [16] who found that adhesive property of starch was better than that of sugar by product. This was caused by different chemical compositions of...
the adhesives. Starch contains sticky complex carbohydrate (amilopectin) while sugar by-product do not. The main component found in SBRC-M is sucrose which is a simple carbohydrate. While in SBRC-T, as it is sticky, amilopectin improves inter-material binding in fly ash granules which in turn improves the durability of fly ash granules to pressure and friction they receive during transportation. Sago and starch meal containing complex polysaccharides are commonly used as adhesive material.

3.4. Water holding capacity

In this study, the use of SBRC-M granule agent resulted in higher water holding capacity (WHC) (40.62%) than did SBRC-SC (38.79%) and SBRC-T (36.85%). The highest WHC (44.22%) was obtained fly ash granules made by using 10% SBRC-M granule agent. While the water holding capacity of fly ash without granulation was 56.9 percent [1].

Fly ash granules made by using SBRC-M were easy to adsorb and mix with water as the granules were immediately damaged when they were in touch with water. This phenomenon was more obvious with 10 and 15% SBRC-M which were easy to be washed out during the separation process of water and granules before the granules were weighed for their wet weight. This became an obstacle in the weighing process as water was still attached to the granules, making the wet weight of the granules was not easy to determine. Forced weighing would result in more granules being washed out in the water. In contrast, 5% SBRC-M were not immediately damaged when they were soaked in water. Therefore, no such obstacle as the one in 10 and 15% SBRC-M was found in the weighing process. Granules made by using SBRC-SC adhesive in various concentration did not absorb water well. However, no different water holding capacity was found on granules made by using all adhesive materials in all concentrations except in granules made by using 10% SBRC-M. SBRC-T in all concentration, except SBRC-T 5%, did not absorb water as high as SBRC-M 10%. Meanwhile, SBRC-T 5% and SBRC-T 15% could absorb water by 39.45% and 36.92%, respectively, but the two figures were not significantly different.

Water holding capacity is affected by the structure of the particles contained in granules. High water holding capacity of ash was due to its dominant silt and clay size fractions. Fly ash contained about 93 percent of silica and sesquioxides (Al₂O₃ and Fe₂O₃), and in the remaining portion, Ca²⁺ was the dominant cation followed by Mg²⁺, Na⁺ and K⁺[1]. And in the remaining portion, Ca²⁺ was the dominant cation. The addition of fly ash granules to the soil will improve soil water holding capacity as fly ash granules have a capability to increase the number of soil micropores and macropores [17,18]. Macropores have pore diameter ≥ 0.1 mm while that of micropores is < 0.1 mm [19].

![Figure 3. Water holding process of fly ash granules made by using various types and concentration of adhesive materials, before drying (at) and after drying (b).](image-url)
Figure 4. Water holding capacity (WHC) of fly ash granules in various types and concentration of granule agents

Table 3. Water holding capacity (WHC) of fly ash granules made by using various types and concentration of adhesive materials

| Adhesive material | WHC (%) | St. dev |
|-------------------|---------|---------|
| SBRC-M 5% (M5)    | 40.68ab | ±2.91   |
| SBRC-M 10% (M10)  | 44.22a  | ±2.93   |
| SBRC-M 15% (M15)  | 36.95b  | ±4.62   |
| Average           | 40.62   |         |
| SBRC-SC 5% (SC5)  | 38.56b  | ±1.10   |
| SBRC-SC 10% (SC10)| 38.46b  | ±1.59   |
| SBRC-SC 15% (SC15)| 39.35b  | ±1.49   |
| Average           | 38.79   |         |
| SBRC-T 5% (T5)    | 40.75ab | ±2.69   |
| SBRC-T 10% (T10)  | 39.45b  | ±3.25   |
| SBRC-T 15% (T15)  | 36.92b  | ±1.96   |
| Average           | 36.95   |         |

4. Conclusions
It was found that fly ash granules made from 15% of SBRC-M had the highest particle density (0.75 g/cm³). Fly ash granules made with SBRC-M had higher pH (10) than those made by using SBRC-SC adhesive (9.3) and SBRC-T (9). SBRC-T was found as the granule agent material which produced fly ash granules with the highest durability levels on average. In this study, the use of SBRC-M granule agent resulted in higher water holding capacity (WHC) (40.62%) than did SBRC-SC (38.79%) and SBRC-T (36.85%). As a granule agent, compared to SBRC-SC and SBRC-T, SBRC-M could produce fly ash granules with highest particle density, highest pH, good durability, and best water holding capacity.

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