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Development of synthetic light weight soil aggregates utilizing coal fly ash and mine clay as waste materials

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ABSTRACT

This paper presents an investigation of the development, evaluation and micro morphological observations of synthetic light weight soil aggregates utilizing coal fly ash, Okinawa mine clay and used paper as waste materials. In this study series of aggregates have been produced by using several fly ash application percentages from 0 to 100 with other waste materials as an alternative method of waste disposal. Relation-ship between applied coal ash percentage and different aggregate parameters such as bulk density, water holding capacity, permeability, aggregate strength, aggregate stability and mean weight diameter were determined. The results of the study show that fly ash addition percentage had a correlation co-efficient of 0.9107 with the bulk density of the aggregates. Moreover coal ash addition percentage had correlation co-efficients of 0.8625, 0.9519 and 0.9560 with mean weight diameter difference, aggregate strength and loss percentage of finer fraction less than 1mm respectively. Coal fly ash addition percentages below 40% showed low aggregate strength values. Highest aggregate strength was given with 100% of ash application. Bulk densities of all aggregates were in between 0.68 - 1.02 gcm^{-3} which is in the range of light weight aggregates. Permeability of all aggregates showed an average value of 2.7 x 10^{-2} cm/sec. Water holding capacities of all treatments showed high values in between 0.69 - 0.74 l/kg. PH values of aggregates were in the range of 5.85 - 8.25 and nitrogen percentages of all produced aggregates were in between 0.03% - 0.06%. Lowest loss percentage of finer fraction (less than 1 mm) was given at 100% of ash application after subjecting to mechanical shaking. Scanning electron microscopic (SEM) observations indicated that coal fly ash and mine clay particles are well enmeshed in the used paper matrix with starch as the binder to form stable light weight soil aggregates. Low application percentages of coal fly ash showed poor binding in SEM images. The study also revealed that aggregates produced by coal fly ash, mine clay and used paper with starch enhanced the aggregation while improving aggregate performances with increasing percentages of coal fly ash application.

Keywords: Coal fly ash, light weight aggregates, aggregate stability, aggregate strength, mean weight diameter, SEM techniques

INTRODUCTION

From a practical perspective, sustainable development requires the optimization of current natural resources, while optimizing the utilization and minimization of derived waste. Coal has been burnt for well over a century as an important source of energy for most parts of

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the world. As a consequence of its use, it is estimated that approximately 500 million tons of coal ash are produced globally every year (Manz, 1997).

Of this amount, less than one quarter is used commercially with rest being disposed of as land fillings (NRCAN, 1999). Fly ash, a finely divided residue resulting from the combustion of coal of thermal power plant is regarded as an amorphous ferro-alumino-silicate mineral containing naturally occurring essential macro as well as micro-nutrient elements similar to that of soil except humus and nitrogen (Tripathi *et al.*, 1988; Zacharia *et al.*, 1996). Disposal of unused coal fly ash is costly and a burden for the power industry. On the other hand, there is an estimated 275,000 tons of coal ash generated annually in Okinawa, Japan (NIAC, 2003). Furthermore, few uses for the tonnages produced and the disposal of fly ash become a significant problem in Okinawa. Disposal as land fillings is no longer acceptable due to lack of space. Given the large volume of coal fly ash that requires disposal, new application technology are constantly being sought. Therefore several benefits can be obtained by finding an alternative use for coal fly ash. The co-utilization of fly ash and mine clay with used paper to generate synthetic soil aggregates would be a new opportunity for utilization. Utilization of coal fly ash as a resource has been studied for decades, in many research areas such as in-valuable element extraction (Lakshmanan, *et al.*, 1990), in environmental engineering (Polettini *et al.*,2004), in ceramic products (Leroy *et al.*, 2004), in paint and plastic industry, in building products (Sunil *et al.*, 2002). Another use of fly ash is in agriculture, since the hydroxide and carbonate salts give fly ash one of its principal beneficial chemical characteristics, the ability to neutralize acidity in soils (Pathan *et al.*, 2003). Fly ash amendments have been reported to modify soil pH, improve soil texture, water retention capacity and provide essential plant nutrients for increasing crop production (Korcak, 1993: Fail and Wochock, 1977: Page *et al.*, (1979, 1980): Chang *et al.*,1977). Also coal ash has been shown to have a reasonable capacity of absorbing soluble Phosphorous (Donald, 2003). A literature survey revealed that fly ash has been used for removing heavy metals (Weng and Hung, 1994: Bulewicz *et al.*, 1997) and radionuclide (Aptak *et al.*, 1996) from aqueous solutions. The material's chemical composition, as well as its physical properties make fly ash a useful material, mainly in the construction industry. Coal fly ash can be used as a partial replacement for cement in concrete production. After mixing the ash with different materials including other by-products, pellets are formed that can be used in concrete as aggregates (Valenti, 1995).

Considerable quantity of Paleozoic and Mesozoic limestones in Motobu peninsula of northern part of the Okinawa prefecture, Japan, is being used to make lime materials by crushing in mining industry. These limestones are crushed and washed with water during the commercial process. In this process finer particles of the lime stone are flowing with the water as waste materials. These fine waste materials are being flocculated by adding a flocculent. The flocculated fraction called as mine clay is a waste material which is not utilized for any purpose. Under our research study, the possibility of using this mine clay for the production of synthetic light weight aggregates in combination with coal fly ash was investigated. The purpose of adding used paper was to provide efficient binding surface and to enable porous space within the produced aggregates. Used paper is also an important industrial waste that can be used effectively in agriculture. It is a rich source of carbon and improves soil organic matter content, water holding capacity, soil structure and bulk density (Rasp, 1992).

The formation of aggregate requires both physical rearrangement of particles and the stabilization of the new arrangement. Therefore effective binder should be added in order to obtain stable aggregates. Several binding mechanisms exist between organic polymers and mineral surfaces to provide stable aggregates. Organic polymers have been used quite effectively to stabilize soil structure in recent years. Polysaccharides stabilize soil aggregates because of their contribution as cements and glues. (Taskin *et al.*, 2002). Therefore starch coming out as waste material from Okinawa flour industry was added as an aggregate binder for the production of synthetic aggregates. This paper will envisage the development, evaluation and micro morphological observations of synthetic aggregates which were produced by using different proportions of coal fly ash, Okinawan mine clay, used paper and starch. Moreover waste materials are available free or at a low cost, in Okinawa, Japan. Therefore economics appear to be attractive. Moreover this study also explains an efficient waste management practice.

MATERIALS AND METHODS

Production process of aggregates

Different proportions of raw materials shown in Table 1 were used in the EIRICH mixer (R-02M/C27121) for the production of light weight aggregates. Series of aggregates were produced by varying fly ash addition percentages between 0 % to 100 % (i.e. 0 %, 10 %, 20 %, 30 %, 40 %, 50 %, 60 %, 70 %, 80 %, 90 % and 100 %). Initially respective quantities of mine clay, coal ash and used paper were mixed in the EIRICH mixer. Then starch paste which was made using 100 g of starch and 320 ml of hot water (Table 1) was added to the above mixer. Then whole mixture was mixed until the production of desired aggregates.

Physical and chemical properties of aggregates

Bulk density (Culley, 1993), saturation permeability coefficient (Klute, 1965) and water holding capacity (Culley, 1993) of aggregates were determined. Chemical properties such as pH (aggregate:water:1:2.5), nitrogen (Schuman *et al.*, 1973) and organic carbon (Nelson *et al.*,1996) were also determined.

Structural parameters of produced aggregates

Mean weight diameter

Wet and dry mean weight diameters of different types of produced aggregates were determined by using dry and wet sieving techniques respectively (Yoder, 1936). Three replicates from each type of aggregates were

Table 1. Different proportions of raw material used to produce light weight aggregates

Raw materials	Proportions of raw materials (g)										
Mine clay(g)	1000	900	800	700	600	500	400	300	200	100	0
Coal ash (g)	0	100	200	300	400	500	600	700	800	900	1000
Used paper(g)	50	50	50	50	50	50	50	50	50	50	50
Starch (g)	100	100	100	100	100	100	100	100	100	100	100
Hot water (g)	320	320	320	320	320	320	320	320	320	320	320
Coal ash addition(%)	0	10	20	30	40	50	60	70	80	90	100

used in dry and wet sieving technique. Mean weight diameter differences of produced aggregates were calculated as an indicator for aggregate stability on the basis of dry and wet mean weight diameters.

Different types of aggregates were sieved using 10 mm sieve. Then the dry sieve set was arranged in descending order (i.e.5.60 mm, 3.35 mm, 2 mm, 1 mm and 0.5 mm) and100 g of air dried aggregate sample was placed on the top sieve for the dry sieving technique. Then percentage of dry aggregates retained on each sieve was weighed after the mechanical shaking. Each treatment was triplicated. Then mean weight diameter of dry aggregate was determined.

Aggregate size smaller than 10 mm was used for wet sieving technique. Sieve set was arranged in descending order. Same sample (100 g) which was used in above dry sieving test was saturated for one hour and then placed on the top sieve of the sieve set. Water was poured to the cylinder which contains sieve set (sieve diameters are 5.60 mm, 3.35 mm, 2 mm, 1 mm and 0.5 mm) up to the upper limit of the soil sample in the top sieve. Then all sieve sets were shaken for one hour. After one hour, percentage retained in each sieve was determined. Each treatment was triplicated and then respective wet aggregate mean weight diameter (Van Bavel, 1949) was calculated by using the following equation.

$$MWD = \sum_{i=1}^{n} X_i W_i,$$

Where MWD=Mean weight diameter, X_i is the mean diameter of each size class, and W_i is the fraction of the total sample weight (mass) occurring in the i^{th} size class.

Mechanical shaker analysis

50 g of aggregates grater than 1mm from each treatment were put into 200 ml bottle. Then 150 ml of water was added to the bottle and kept for 12 hours of saturation. Subsequently saturated samples were shaken in mechanical shaker at 100 rpm for 5 hr, 10 hr, 24 hr and 48 hrs respectively. Each treatment was triplicated. Finally average loss percentage of finer fraction smaller than 1 mm was calculated.

Aggregate strength

Aggregate strength was determined by using hardness testing machine (Kiya Digital Hardness (Rigidity) tester, KHT-20, Japan). Randomly selected 5 samples of 5 mm diameter aggregates were used to determine the average aggregate strength.

Statistical analysis

Data were subjected to analysis of variance (ANOVA) for determining treatment effects and, Duncan multiple comparison test procedure was used for mean comparisons by using SAS package (SAS Institute, 1990).

Scanning electron microscopic study

Coal fly ash, mine clay, used paper, starch samples as raw materials and randomly selected aggregate samples were dried at 105°C for 24 hours and stored in desiccators. Prepared starch paste was dried at 105 °C for 24 hours. Then it was critical point dried in a critical point drier JCPD-3 (JEOL, Japan). Then samples were mounted on brass stubs with double adhesive carbon tape and gold coated (20 nm thick)in a fine Coat-Ion Sputter JFC1100 (JEOL, Japan) for 3 minutes. Micromorphology of the samples was observed under high vacuum conditions (HV) at 10-12 kV using a JSM-5600 LV scanning electron microscope (JEOL, Japan) equipped with a black scattered electron detector as described by Davey 1978.

Results and Discussion

Bulk densities of synthetic aggregates decreased from 1.02 gcm^{-3} to 0.68 gcm^{-3} with increasing percentage of coal fly ash from 0% to 100 % (Table 2), which showed a correlation coefficient of 0.9107 (Table 3). Average bulk density of the aggregates was 0.85 gcm^{-3} which can be graded as light weight aggregates (ACI, 2000: Jansen, 2000). It is also evident that there were significant differences between bulk densities of aggregates produced with different fly ash levels. Page *et al.*, (1979, 1980) reported that fly ash amendment to a variety of soils tend to decrease the bulk density. Therefore these low bulk density values in turn improve the porosity and water retention capacity of the aggregates. All aggregates show high value of water holding capacities in between 0.69-0.74 l/kg. Moreover it revealed that there is a 0.05 l/kg increment of water holding capacities of aggregates with the increasing percentages of coal fly ash addition from 0 % to 100 %. But there are no significant differences in water holding capacities of different aggregates at 0.05 level (Table 2). The average water holding capacity for produced aggregates was 0.72 l/kg. Chang *et al.*, (1977) reported that an addition of fly ash increased water holding capacities of the soil. Aggregate's permeability is a measure of the ability of air and water to move through it. Permeability is influenced by the size, shape and continuity of the pore spaces which in turn depend on the bulk density, structure and the texture. Here permeability values of produced aggregates increased slightly within the range of 1.6×10^{-2} - 3.7×10^{-2} cm/s (i.e. normal silt type soil range) with the increasing percentage of coal fly ash addition, but did not show any significant difference between treatments.Low bulk densities with high water holding capacities of aggregates using fly ash and mine clay enabled them to be used as a medium for crop growth. Original pH values of coal fly ash and mine clay were 5.95 and 8.45 respectively. All aggregates produced by using coal fly ash showed pH values (Table 4) between 5.85 - 8.25 which fluctuated between slightly acidic to alkaline range. The pH of coal fly ash varies from 4.5 to 12.0 depending largely on the sulphur content of the parent coal (Plank and Martens, 1974) and it can modify the pH (Korcak, 1993). So these aggregates can be used to buffer the pH values of problematic soils.

But nitrogen content of all aggregates showed low limits which were in the range of

Table 2. Physical properties of different types of aggregates

Fly ash Application (%)	Bulk Density (gcm^{-3})	Water holding capacity (l/kg)	Saturation Permeability co efficient (cm/sec)
0	1.02^{a}	0.69^{a}	1.60×10^{-2a}
10	0.87^{bc}	0.70^{a}	1.86×10^{-2a}
20	0.87^{bc}	0.71^{a}	1.86×10^{-2a}
30	0.86^{cd}	0.72^{a}	2.24×10^{-2a}
40	0.85^{d}	0.72^{a}	2.24×10^{-2a}
50	0.81^{e}	0.73^{a}	2.24×10^{-2a}
60	0.80^{e}	0.73^{a}	2.80×10^{-2a}
70	0.74^{f}	0.74^{a}	2.80×10^{-2a}
80	0.72^{g}	0.74^{a}	2.80×10^{-2a}
90	0.69^{h}	0.74^{a}	3.70×10^{-2a}
100	0.68^{h}	0.74^{a}	3.70×10^{-2a}

(Means followed by the different letter are significantly different at 5% level in the same column)

Table 3. Relationship between the different aggregate parameters with the coal fly ash addition percentage

Parameter	r^2	Equation
Bulk density	0.9107	Y= - 0.0029X + 0.9566
Mean weight diameter difference	0.8625	Y= - 0.0303X + 2.9686
Aggregate strength	0.9519	Y= 0.0257X + 0.8573
Loss percentage of finer fraction(less than 1 mm)	0.9560	Y= - 0.1897X + 53.502

Table 4. Chemical properties of different types of aggregates

Fly ash Application (%)	pH (Aggregate: water:1:2.5)	Carbon (%)	Nitrogen (%)
0	8.25	5.48	0.03
10	8.05	5.47	0.03
20	7.90	5.47	0.04
30	7.85	5.46	0.04
40	7.83	5.45	0.04
50	7.79	5.44	0.04
60	7.77	5.44	0.05
70	7.75	5.43	0.05
80	7.72	5.42	0.05
90	7.50	5.41	0.06
100	5.85	5.40	0.06

0.03 % - 0.06 %. Therefore nitrogen source should be supplied when it is used as an agricultural growth medium. The average organic carbon content was 5.44 %.

It is revealed that mean weight diameter difference of produced aggregates decreased with increasing amount of coal fly ash application percentage, which was explained by a linear function (Table 3) with a correlation coefficient of 0.8625. Mean weight diameter difference values among the treatments were not significantly differed at 0.05 level (Table 5).

Aggregate strength increased with the increasing amount of coal fly ash application percentage. There was a statistically significant relation-ship between percentages of coal fly ash application and the aggregate strength of the produced aggregates, which was explained by a linear function (Table 3) with a correlation coefficient of 0.9519. Highest aggregate strength (3.16 kgcm^{-2}) was given at 100 % of fly ash application while the lowest (0.93 kgcm^{-2}) was given at 0 % fly ash application. It reveals that fly ash addition above 40 % gave the more strengthen aggregates than treatments below 40 % of fly ash addition (Table 5). Therefore aggregates above 40 % of ash application have the ability to resist water and wind erosion more effectively.

It is evident that loss percentages of finer fraction (< 1 mm) of produced aggregates decreased with the increasing amount of coal fly ash application percentage, which was

Table 5. Effect of fly ash application percentage on mean weight diameter difference and aggregate strength of different types of aggregates

Fly ash Application (%)	Mean weight diameter difference (mm)	Aggeregate strength (kgcm^{-2})
0	3.18^{a}	0.93^{d}
10	3.03^{a}	1.08^{c}
20	2.47^{a}	1.22^{c}
30	2.02^{a}	1.58^{c}
40	1.87^{a}	1.59^{c}
50	0.64^{a}	2.39^{b}
60	0.62^{a}	2.68^{ab}
70	0.58^{a}	2.85^{ab}
80	0.56^{a}	2.92^{ab}
90	0.52^{a}	3.10^{a}
100	0.50^{a}	3.16^{a}

(Means followed by the different letter are significantly different at 5 % level in the same column)

Table 6. Effect of coal fly ash application percentage on the loss percentage of finer fraction (< 1 mm) in mechanical shaker (100rpm)

Fly ash Application (%)	Shaking time (hours) 5	10	24	48
0	50.27^{a}	54.23^{a}	68.36^{a}	86.75^{a}
10	49.91^{a}	53.35^{a}	63.62^{b}	84.00^{b}
20	46.40^{b}	50.32^{b}	57.92^{c}	82.96^{b}
30	41.47^{c}	46.37^{c}	55.01^{d}	73.68^{c}
40	32.79^{d}	44.01^{d}	54.28^{de}	68.21^{d}
50	29.66^{e}	43.76^{d}	52.32^{ef}	63.91^{e}
60	28.77^{e}	42.31^{e}	51.42^{fg}	61.47^{f}
70	24.75^{f}	38.66^{e}	50.13^{fgh}	59.90^{fg}
80	20.63^{g}	37.49^{fg}	48.97^{ghi}	58.17^{g}
90	19.69^{g}	36.86^{g}	47.75^{hi}	55.75^{h}
100	19.47^{g}	36.80^{g}	47.43^{hi}	54.23^{h}

Means followed by the different letter are significantly different at 5% level in the same column)

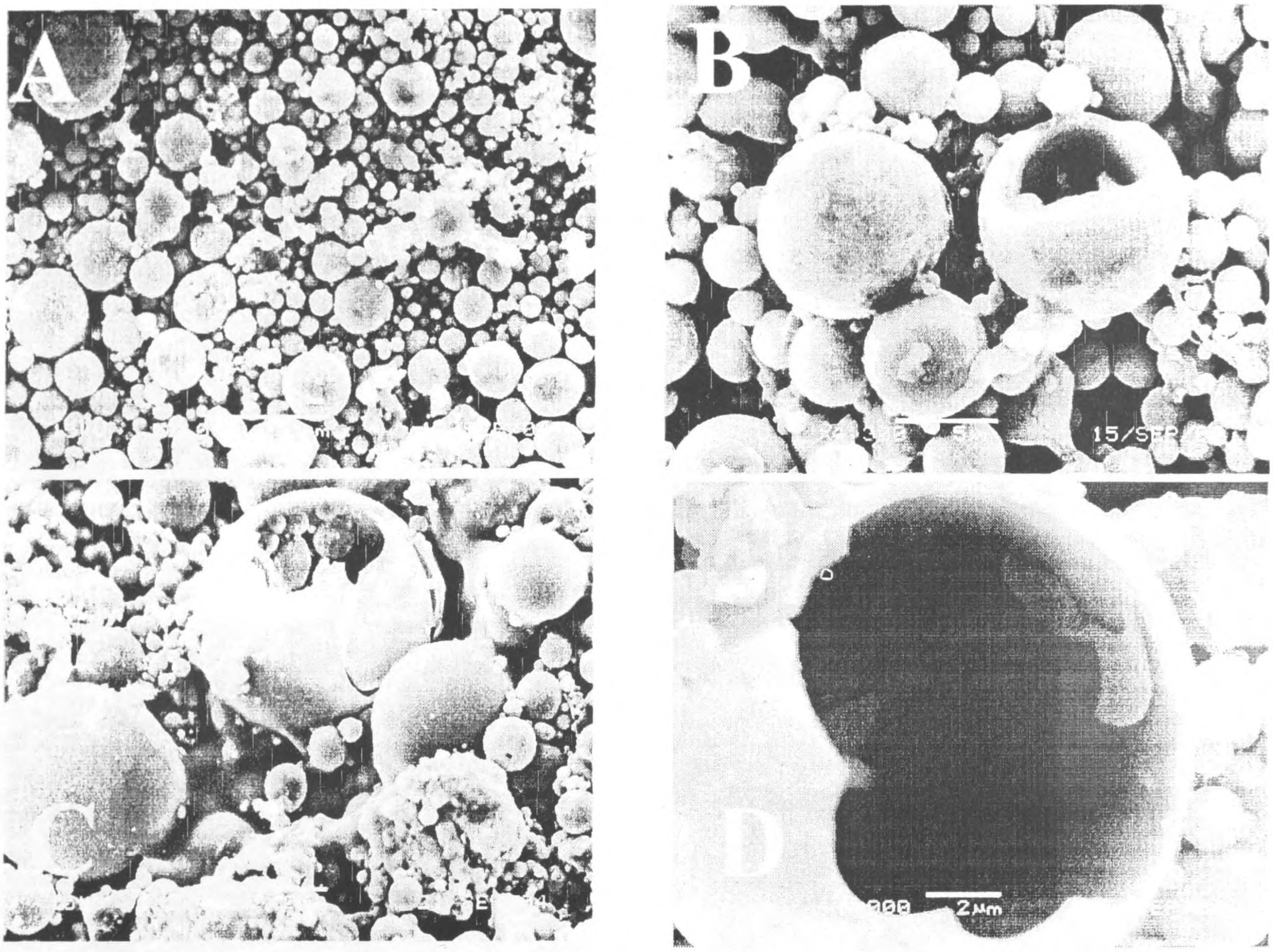

Figure 1. Scanning electron micrographs of coal fly ash particles, showing the micro morphology and various sizes of coal ash particles. (**A and B**) : Occurrence of round shape coal fly ash grains with various sizes. (**C and D**): Fly ash shows cenospheres (Hollw spheres)

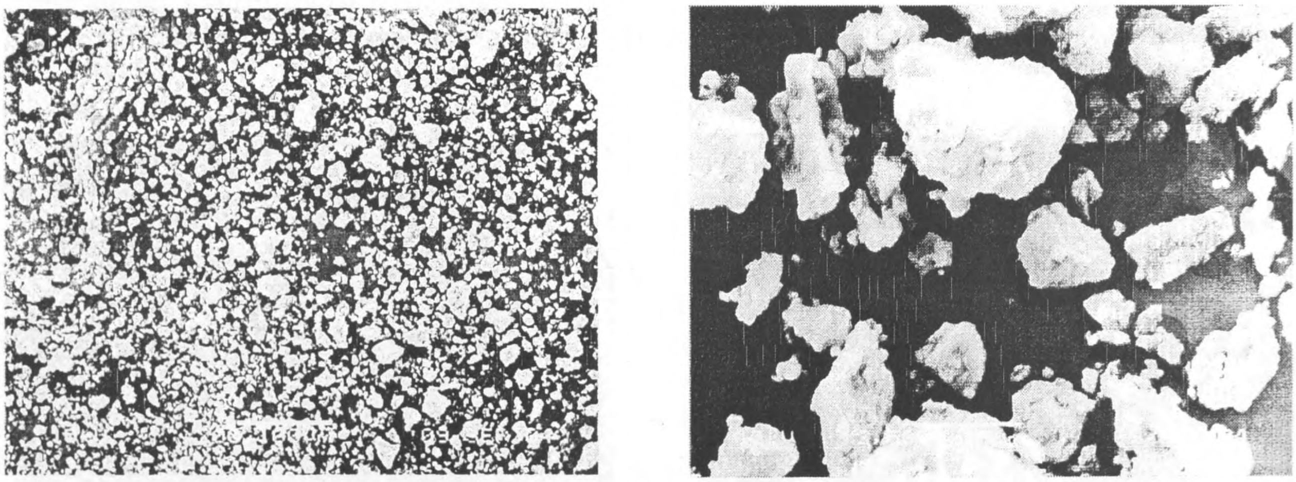

Figure 2. Scanning electron micrographs of Okinawa mine clay particles, showing the micro morphology and various sizes of particles.

explained by a linear function (Table 3) with a correlation coefficient of 0.9560 after subjecting to a mechanical stress. It reveals that fly ash addition above 80 % gave the lowest loss percentage of finer particles less than 1mm than other treatments (Table 6) in all

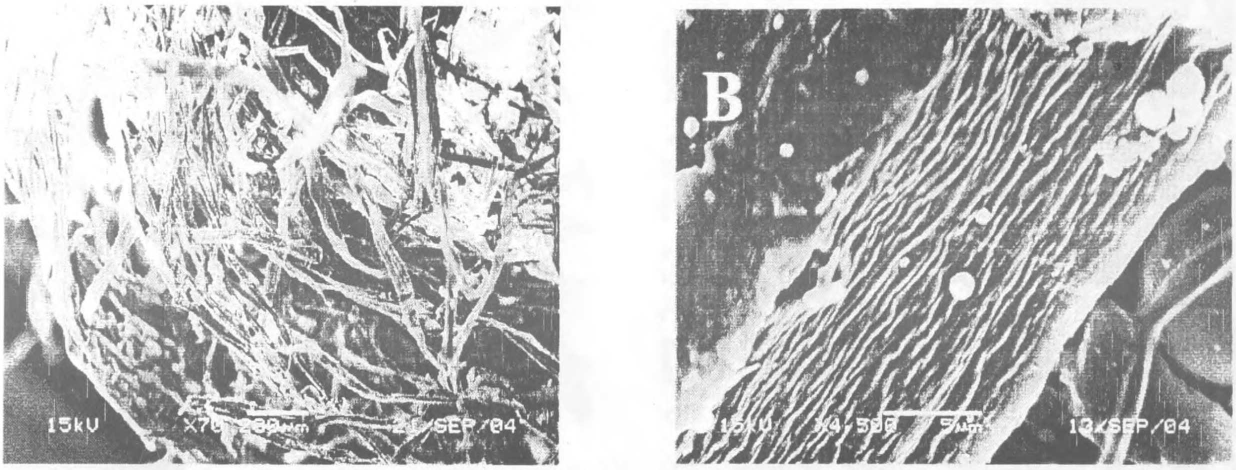

Figure 3. Scanning electron micrographs of used paper fibers, showing the micro morphology (A) Shows the porous structured arrangement of fibers (B) Shows micro morphology of binding surface of an individual Fiber.

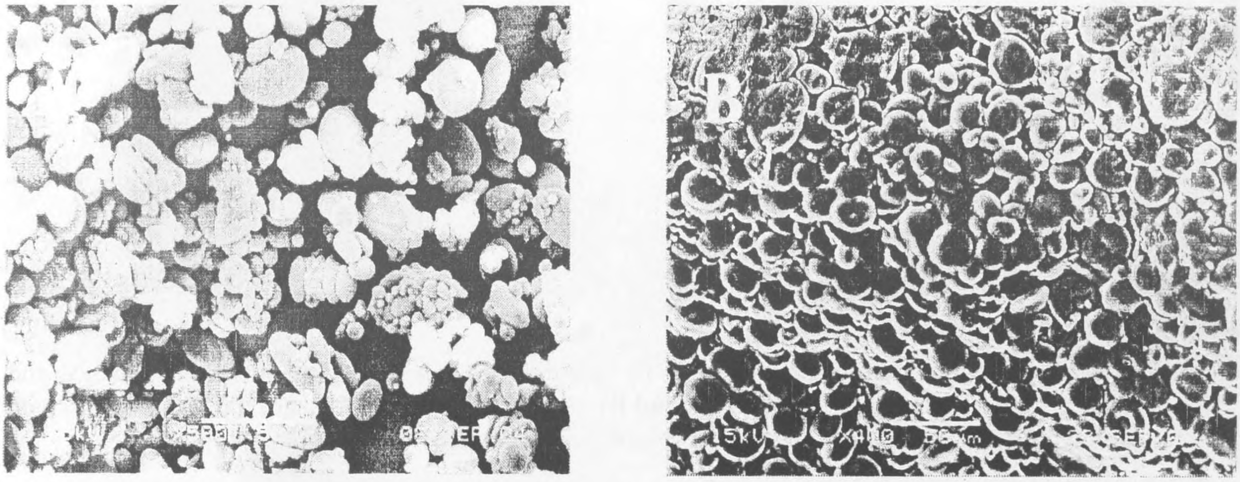

Figure 4. Scanning electron micrographs of starch, showing the micro morphology (A) Starch powder (B) Starch paste.

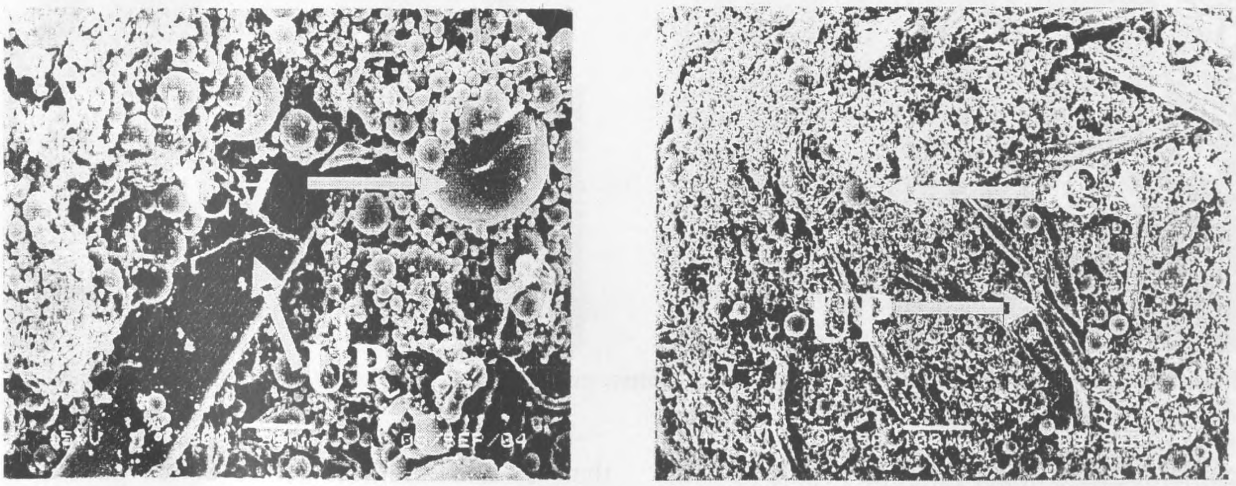

Figure 5. Part of macro-aggregate is from treatment containing 100% of fly ash application (CA= Coal fly ash, UP =Used paper).

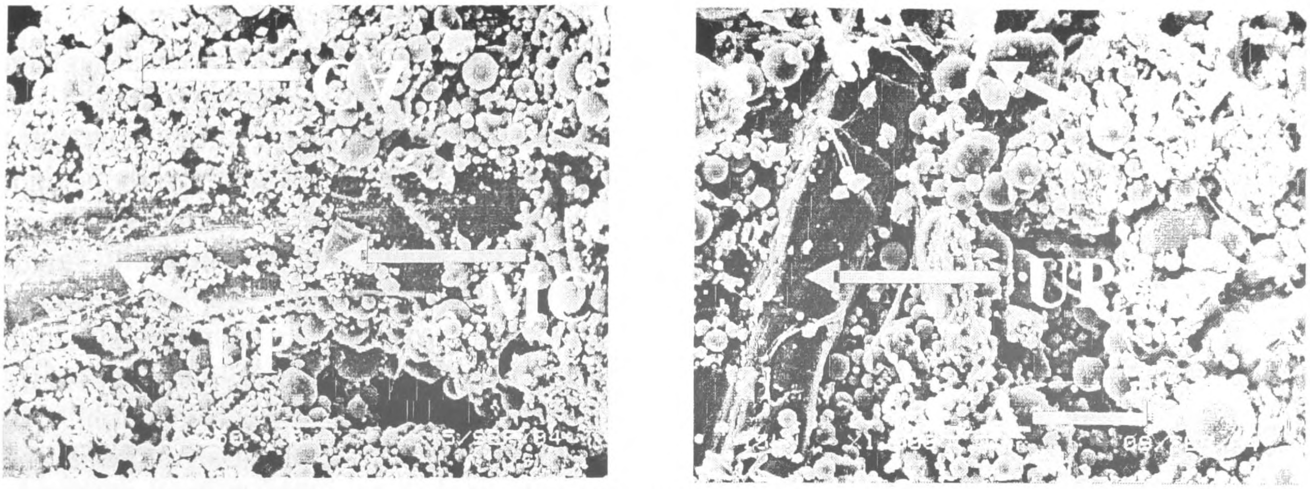

Figure 6. Part of macro-aggregate is from treatment containing 80% of fly ash application (CA – Coal fly ash, MC =mine clay, UP=Used paper)

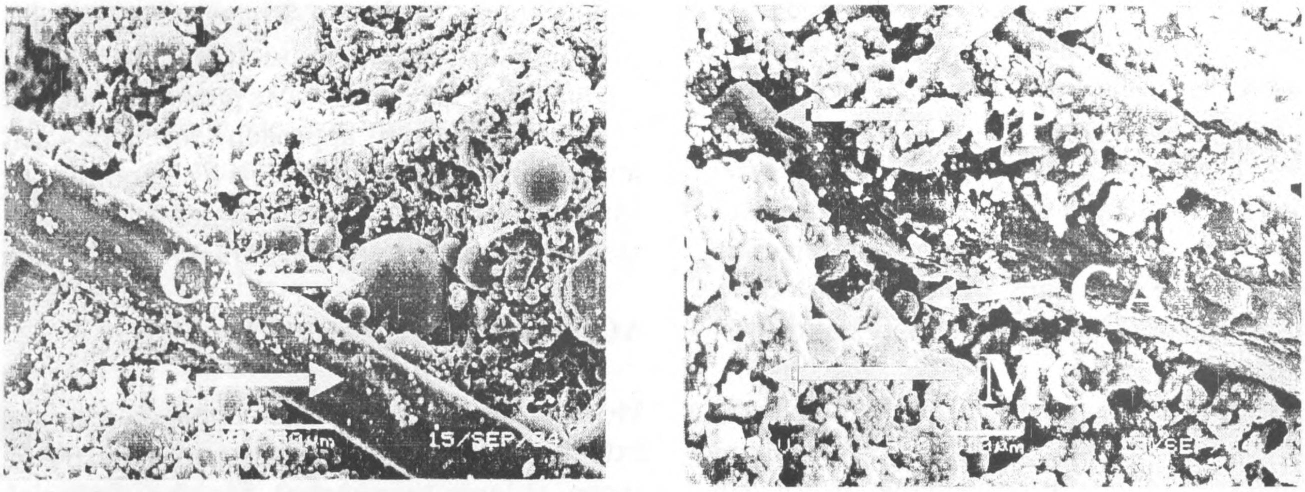

Figure 7. Part of macro-aggregate is from treatment containing 10% of fly ash application percentage (CA=Coal fly ash, MC =Mine Clay, UP=Used paper)

time intervals. Moreover, aggregates produced without coal ash application (0 %) gave the highest mean loss percentage at all time intervals.

Scanning electron microscopic study

The scanning electron micrographs of figure 1 show the detailed micro morphology of coal fly ash particles. Shapes of coal fly ash particles were more or less round and were varying in size. (Figure 1- (A and B)). SEM observations show (Figure 1- (C and D) that coal fly ash contains thin walled hollow spheres called cenospheres. Therefore, bulk densities of aggregates decreased with increasing amount of coal fly ash. Figure 2 shows the micro morphology of Okinawa mine clay, which was angular in shape. SEM observations of used paper fibers indicated (Figure 3) the ability to provide efficient and higher binding surface area and also structure for the aggregation. Coal fly ash particles and mine clay particles can be easily bound to fibers with strarch paste because of high binding area of used paper. Morphological observations of starch powder and starch paste are given in Figure 4.

A part of macro-aggregate having 100 % of coal fly ash application is shown in Figure 5 which showed efficient binding and wide spread arrangement of coal fly ash with used paper fibers. Moreover, SEM observations indicated that fly ash particles are well enmeshed into the used paper matrix with starch binder. In this treatment, coal ash particles are very high. Therefore, this treatment contains high percentage of cenospheres, which gives the lowest bulk density. Consequently 100 % coal fly ash application treatment showed high pore space, permeability and aggregate strength. Overall micro morphological observations of 80 % coal fly ash application showed (Figure 6) well bound enmeshments of coal fly ash and mine clay with used paper fiber. Moreover, these light weight aggregates could be considered as dual composite material consisting of rigid fly ash and mine clay particles in a soft used paper fiber matrix. Synthetic light weight aggregates produced with the high fly ash application with low percentages of mine clay show higher strength than high applications of mine clay.

Figure 7 showed a part of macro-aggregate, which is from a treatment containing a lower percentage of fly ash application (10 %). It showed weak arrangement of materials due to high quantity of mine clay. Scanning electron microscopy indicates that high concentration of mine clay did not well enmesh into used paper matrix. Therefore, it gives lower aggregate stability and aggregate strength, which has high dispersion under a mechanical stress.

Overall morphological observations of synthetic light weight aggregates explain their high aggregation ability and aggregation strength, which also has a positive correlation with the increasing percentages of coal fly ash applications.

CONCLUSION

The study concludes that there is scope for utilization of coal fly ash, in combination with Okinawa mine clay and used paper fiber for the production of synthetic light weight aggregates with starch as an organic binder. Synthetic light weight aggregates showed low levels of nitrogen percentages, high permeability values and high water holding capacity values. Aggregates showed high stability and high strength with the increasing amount of coal fly ash application. Aggregates produced in > 40 % of coal fly ash application showed the highest stability. Scanning electron micrographs indicates the fly ash and mine clay enmeshments in the used paper fiber matrix. Therefore, aggregate production from coal fly ash and mine clay with waste paper can be used as an alternative approach for coal ash utilization. Further research is required to study the suitability of aggregates as a growth medium for different types of plant species.

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