SCIENTIFIC PAPER
Published: October 14, 2023
https://doi.org/10.13167/2023.27.5

Influence of binders, mix proportions, and fabrication method on the characteristics of fly ash aggregate

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Received:
January 5, 2023

Accepted:
July 13, 2023

Published:
October 14, 2023

Citation:
Dash, S.; Gupta, P.; Mustakim, S. M.; and Mohanty, I. (2023). Influence of binders, mix proportions, and fabrication method on the characteristics of fly ash aggregate. *Advances in Civil and Architectural Engineering*. Vol. 14, Issue No. 27. pp. 67-79
https://doi.org/10.13167/2023.27.5

ADVANCES IN CIVIL AND ARCHITECTURAL ENGINEERING (ISSN 2975-3848)

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Abstract:
In this paper, two types of lightweight fly ash (FA) aggregates: cold bonded fly ash (CFA) and sintered fly ash (SFA) aggregates were prepared through the cold bonding and sintering method. During the pelletization process, different ratios of binders to fly ash were used, i.e., 10:90, 15:85, 17:83, and 20:80 with a set amount of water. Cement, metakaolin, sodium silicate, urea-formaldehyde resin, bentonite powder, and phenol-formaldehyde resin were employed as binders. A comparative study on physicochemical, mechanical, phase identification, microstructure, and optical analysis of CFA and SFA was performed. The results showed that CFA (an alkali binder) had a higher water absorption (WA) value of 9,50 % with a crushing strength (CS) value of 6,30 MPa than SFA (sodium silicate binder) with a CS value of 5,80 MPa and a WA value of 10,28 %. Experimental observations also demonstrated that the leaching ability of SFA was considerably lower than that of CFA. Most notably, SFA can be used as a substitute for construction material and structural applications along with solving FA waste disposal and related problems to a considerable extent.

Keywords:
fly ash; binder; cold bonding; sintering; crushing strength; leachability; construction material

1 Introduction

Coal ashes, specifically fly ash (FA) and bottom ash (BA), are waste products of thermal power generation. Thermal power plants produce most of the energy worldwide along with a tremendous amount of coal ash, which poses substantial disposal challenges. The captive power plant in India generates approximately 210 Mt of FA per year [1]. In most countries, a large amount of FA remains unutilized, and only a small amount is employed in building materials and for other purposes. Manufacturing lightweight FA aggregates is a convenient method of increasing their usage. Cold bonding, sintering, and autoclaving are the conventional methods used to convert FA to pellets [2].

Recycling and reducing industrial by-products have become major concerns in the 21st century. In recent years, the evolution of new techniques and ideas for waste management has become a focal issue among researchers. FA obtained from coal combustion can be used effectively in various applications, such as FA bricks, aggregates, pozzolanic cement, lightweight block composite materials, and soil stabilization [3-4]. In addition, FA can be used to prepare geopolymer materials, composite materials, and backfill for mines [5]. Organic and inorganic compounds, harmful pollutants, and colours can be removed from wastewater using FA. FA is used in various applications such as wastewater treatment, soil improvement, zeolite synthesis, and in the concrete and ceramics industries. Concrete can also be fabricated using FA [6-7].

Aggregates are produced via different methods, such as sintering, autoclaving, and cold bonding. Currently, FA, red mud, slag, clay material, and incineration sludge are used to manufacture lightweight aggregates (LWA) [8-10]. In the agglomeration process, various methods (cold bonding, sintering, and autoclaving) are employed to convert FA into aggregates. Cold-bonding and sintering processes have been used to develop a sufficiently strong aggregate. Various factors such as moisture, pelletizer speed, and binder proportion affect the formation of good-quality pellets [11-12]. The production of pellets using disc pelletizers is an easy and effective method. The quality of the pellets depends on the speed of the disc and the pelletizer angle [13-14]. The characteristics of aggregates, to a significant extent, depend upon the types of binders and their dosages used [15]. LWAs are used as filler materials in the manufacture of concrete products. In addition, concrete made with FA as an LWA instead of a natural aggregate provides a plethora of economic and environmental advantages. Reduced weight, low shipping and handling costs, and cost-effective disposal are the most significant advantages of employing LWA [16-17]. The NaOH concentration is directly linked to the strength of the FA aggregate. The FA aggregate can achieve a maximum compressive strength of 64 N/mm^2 at 28 days of curing using mortar containing LWA with 8 M NaOH [18].

This study mainly focuses on the effects of the binder, mix proportions, and manufacturing method (cold bonding and sintering procedures) on aggregate properties. Some essential parameters such as the FA-to-binder ratio, curing time, and sintering temperature on the crushing strength (CS), abrasion test, impact test, and index properties such as water absorption (WA), specific gravity, and leachability were considered. All tests were performed in accordance with Indian standards.

2 Fly ash aggregate specimen preparation

2.1 Materials

The materials used in the preparation of the FA aggregates were FA, a binder, coal dust, and water. Class F Fly ash (FA) (conforming to ASTM C 618) from a captive power plant of the National Aluminium Company (NALCO), Anugul, was used to manufacture CFA and SFA. The FA samples were subjected to X-ray fluorescence analysis at KIIT, University, Bhubaneswar. The typical chemical composition of the FA is listed in Table 1.

Table 1. Chemical composition of fly ash (FA)

Constituents	SiO_2	Al_2O_3	Fe_2O_3	Fe	CaO	MgO	Cr	Zn	Pb	LOI
Weight (%)	57,100	24,730	10,50	7,509	2,500	1,400	0,048	0,039	0,024	4,620

The binder plays a vital role in pelletization. Technical-grade chemicals were used during the preparation. This aided in the binding of the FA particles to create a pellet. The binders used were sodium hydroxide (NaOH), urea-formaldehyde (UF) resin, bentonite powder (BP), phenol-formaldehyde (PF) resin, cement (C), metakaolin (M), and sodium silicate (SS). Finar Chemicals Supplier Ltd. (India) supplied the chemicals required for the procedure.
The importance of coal in the manufacturing of SFA cannot be overstated. In downdraft sintering, burning coal produces the required heat. Consequently, a predetermined amount of coal dust was added to the sinter mixture. The coal dust burns in the sinter mix and supplies the heat required for the sinter bed during sintering.
Water is essential in the aggregate mix design process. The amount of water used influences the aggregate mix proportions. Water was added regularly in a controlled manner to affect the workability and setting time of the mixtures.

2.2 Sample preparation

Essential raw components such as FA, binder, and water were mixed in appropriate proportions to prepare FA aggregates. Seven unique aggregate types were created using various binders to FA ratios (Six types of FA aggregates were prepared using the cold bonding method (CBM), and one using the sintering method (SM). Homogeneous mixtures of the binder and FA were prepared in a disc pelletiser with binder-to-FA ratios of 10:90, 15:85, 17:83, and 20:80. The optimal amount of water (0,30 mL) was sprayed during the procedure. Nucleation caused a few seeds to form within 5-10 min, and the seeds steadily expanded over time. Finally, pellets were collected from the trays. Table 2 displays the mix ratios used to prepare the CFA and SFA aggregates.

Table 2. Mix proportion of cold bonded fly ash (CFA) aggregate and sintered fly ash (SFA) aggregate (kg/m^3)

Aggregate type	Duration (min)	Methods adopted	FA	NaOH	UF	BP	PF	C	M	SS	Water
FA-Na	15	CBM	0,85	0,15	0	0	0	0	0	0	0,30
FA-UF			0,80	0	0,20	0	0	0	0	0	
FA-B			0,90	0	0	0,10	0	0	0	0	
FA-PF			0,80	0	0	0	0,20	0	0	0	
FA-C			0,85	0	0	0	0	0,15	0	0	
FA-M			0,85	0	0	0	0	0	0,15	0	
FA-SS		SM	0,83	0	0	0	0	0	0	0,17	

2.3 Production of FA aggregate via the cold bonding method

In the first stage of the experiment, a disc pelletizer with a base depth of 250 mm and disc diameter of 500 mm was employed, with the disc angle oscillating between 45 ° and 50 ° and a speed of 55 rpm. The components, that is, FA with a binder, were dry-mixed for 2 min in a pan mixer before being placed in a pelletizer to complete the FA aggregate generation. Pellet formation occurred within 10-15 min of experimental manufacturing. Pelletization is the process through which FA aggregates are formed. The FA aggregates were then allowed to air-dry for one day before being water-cured for 1, 3, or 7 days.

2.4 Production of FA aggregate by down-draft sintering method

The FA aggregates were collected in a tray and sintered at 1100-1150 °C for 30 min using a downdraft sintering process aggregates. Figure 1 shows the aggregate production process flowsheet (cold-bonding and sintering techniques). The SFA aggregates were prepared using

a batch-type suction grate sintering machine with a cross-sectional area of 300×300 mm and a hearth height of 500 mm. An experiment was performed by keeping a 400 mm BH (bed height) on granulated particle with a 50-mm thick hearth layer and a suction pressure of 400 mm Hg down the grate to achieve 1150 °C temperature for preheating and finally, the aggregate was allowed to cool for 25-30 min.

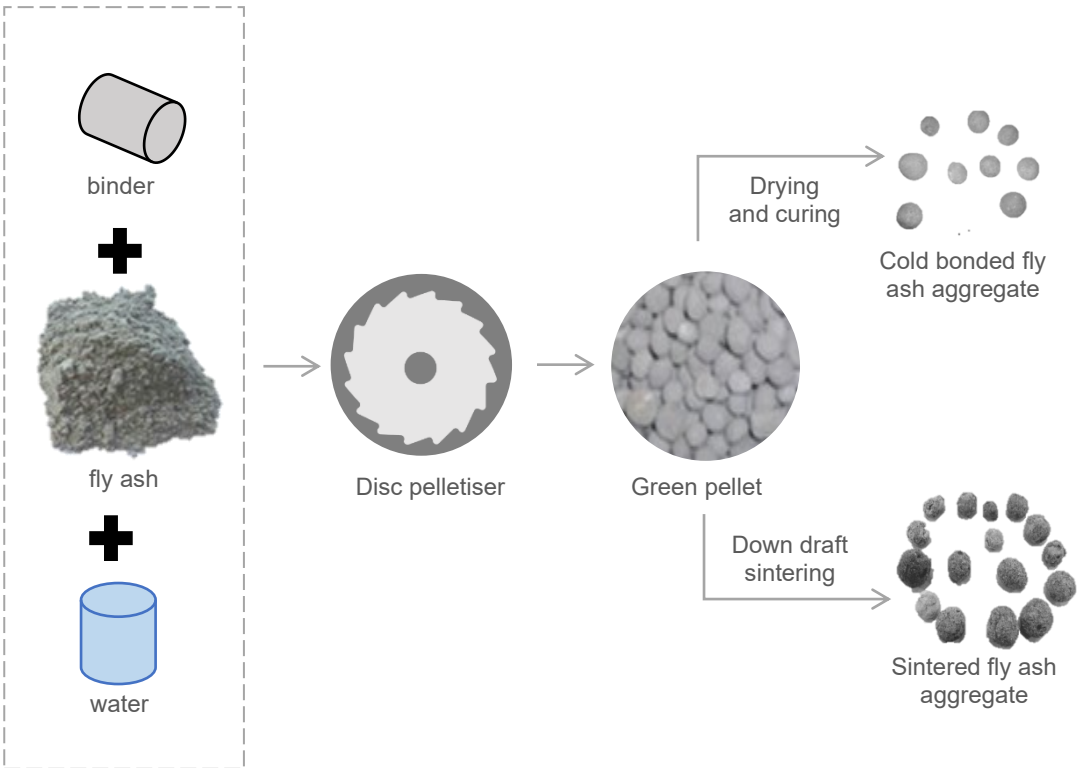


Figure 1. Process flow of manufacturing the cold bonded and sintered fly ash (SFA) aggregate

3 Experimental methods for fly ash aggregate characterization

The experimental methods included the investigation of FA aggregates based on their morphological, chemical, physical, and mechanical properties, as described below.

3.1 XRD, SEM, and optical microscope study of FA and FA aggregates

X-ray diffraction (XRD) studies of FA and FA aggregates provided insights into the possible phases formed. A Cambridge Stereo Scan 200 SEM unit was used to study the microstructural changes and related morphologies of the aggregates. Close views of the aggregates fabricated using both methods were imaged using an optical microscope. All the aforementioned tests were performed at the Council of the Scientific Industrial Research Institute of Minerals and Materials Technology (CSIR-IMMT), Bhubaneswar.

3.2 Mechanical properties

The mechanical characteristics of the aggregates were tested in accordance with Indian Standards. The CS of the aggregates was determined in accordance with IS:2386 (Part-IV)-1963. The aggregates were subjected to impact and abrasion tests in accordance with IS: 2386 (Part IV)-1963 and IS2386 (Part IV), respectively. In addition, specific gravity and WA tests of the FA aggregate were conducted in accordance with IS2386 (part-III)-1 [19].

3.3 Water quality test

Generally, FA contains various hazardous materials, thus necessitating a leachability test to determine the possibility of heavy metal leaching. The disposal of FA, a by-product of thermal power plants, causes serious environmental problems. When coal FA is disposed, it may leach, posing the risk of contamination, particularly in aquatic environments, when ash comes into contact with water. The presence of heavy metals/metalloid(s) such as Pb, Cd, Cr, Fe, and As in groundwater is the main factor contributing to FA pollution. Therefore, developing a processing technology that can effectively remove toxic elements from FA before it is disposed of in the environment is crucial. This is because the amount of FA released into the environment increases, leading to environmental problems [20]. To perform the water quality test (in terms of the leaching test), the CFA and SFA were initially placed in a 500 ml beaker filled with distilled water. The cured water samples collected at different time intervals (1, 3, 7, 12, 24, 48, and 72 h) were tested for pH, TDS, and turbidity.

4 Results and discussion

4.1 Physical appearance and texture

To determine the physical appearance of the aggregate samples, optical images were captured from the front perspective of the samples. To maintain the same magnification for all images, the distance between the camera and aggregate was kept constant during imaging.

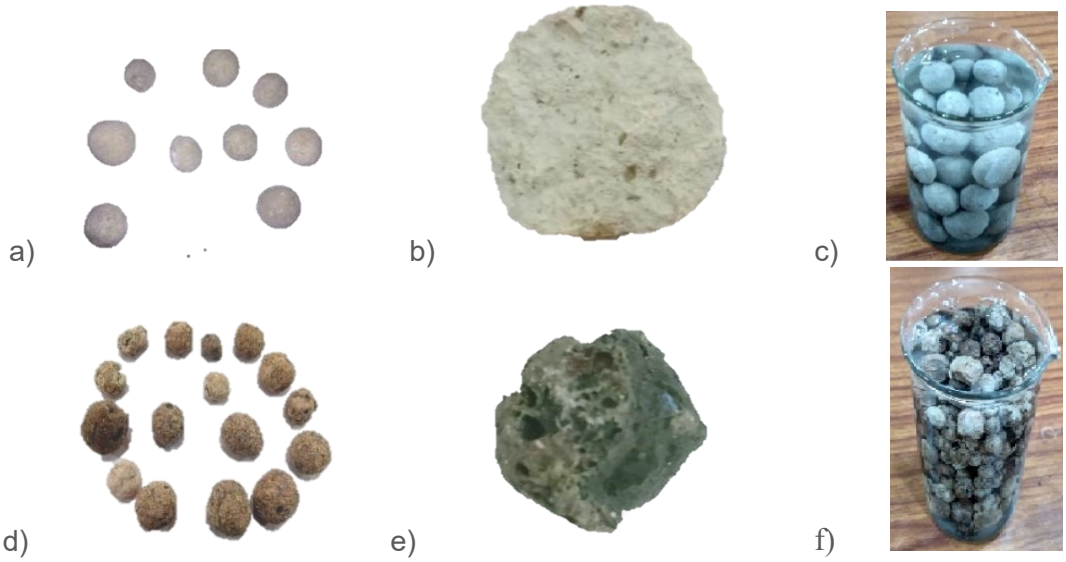


Figure 2. Physical appearance of FA: a) front view of cold bonded fly ash (CFA) aggregate; b) internal core of CFA; c) CFA aggregates in water; d) front view of sintered fly ash (SFA); e) internal core of SFA; c) SFA aggregates in water

The SFA was brown. Visually, the ash particles on the surface are strongly connected, which might be related to the formation of the liquid phase at the sintering temperature. Certain SFA were partially erupted. Because of the high LOI concentration, this behaviour might be attributed to the creation of massive gas bubbles during sintering. The black component of the aggregate displayed in the image is thought to be the molten ash of the aggregate (liquid phase). The oxidation states of iron and carbon are represented by the interior black core of the SFA (Figure 2).

4.2 Particle size distribution

The particle-size distribution of the aggregates was highly graded (Figure 3). The results of the tests showed that the aggregate properties were affected by the pelletization time and the cement employed in the hardening process. During the pelletization process, spherical aggregates with sizes of 4,75; 9,00; 12,50; 16,00 and 20,00 mm were generated. Gradation examination of aggregates revealed a maximum size range of 20,00 mm to 12,50 mm and a minimum size range of 12,50 mm to 4,75 mm.

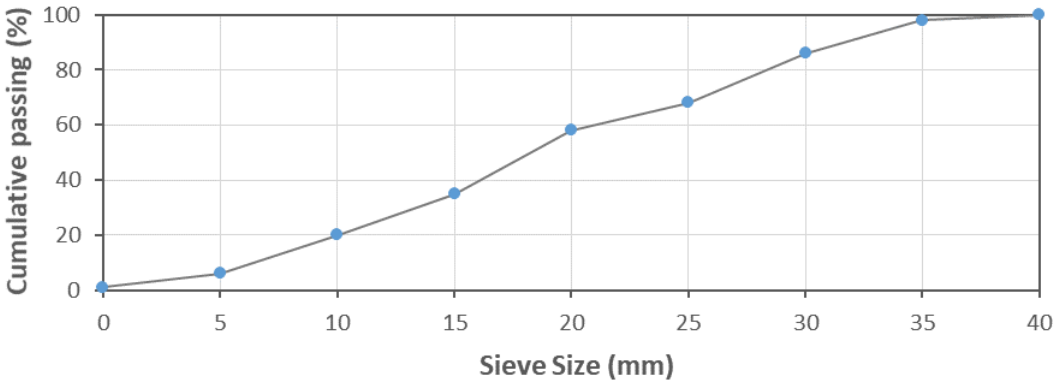
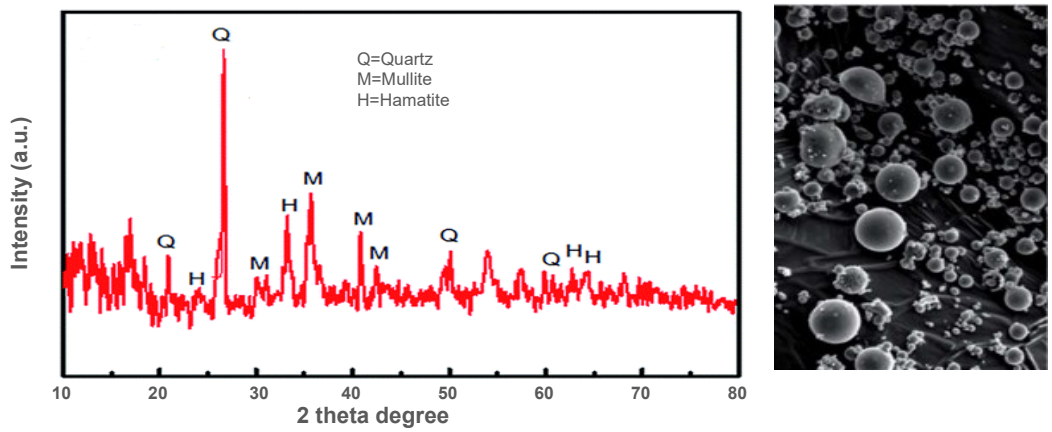


Figure 3. Grain size distribution of the FA aggregate

4.3 XRD, SEM and optical analysis of FA aggregate

The XRD patterns of the FA confirmed the presence of quartz, mullite, and haematite phases. The surface topography of FA appeared to be spherical. The XRD and SEM images of FA are shown in Figure 4.

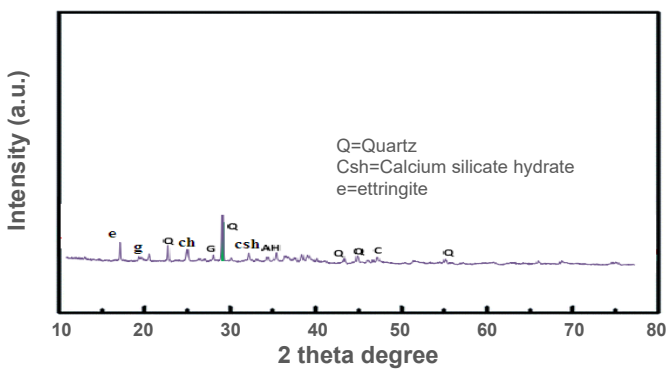


(a) XRD of FA (b) SEM of FA

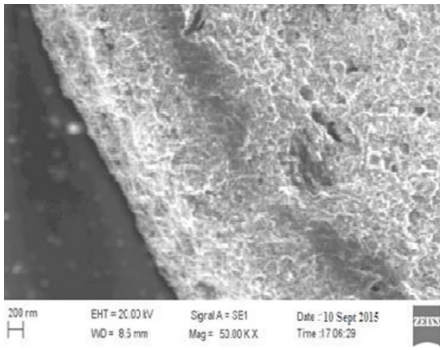
Figure 4. X-ray diffraction (XRD) and scanning electron microscopy (SEM) of FA

The XRD study of CFA showed that the phases present in FA, such as anhydrite and dicalcium silicate, were transformed into hydration products such as gypsum, ettringite, and calcium silicate hydrate when the water curing time increased from one to seven days. Microscopic structural analysis showed that the CFA aggregates had a relatively dense outer shell with relatively low porosity. The XRD analysis of SFA aggregates showed that various phases like quartz, mullite, anhydrite, gehlenite, and haematite appeared when the sintering temperature increased to 1200 °C. The microscopic structure of the SFA appears to have a lower outer shell density with increasing porosity. The optical image of the SFA clearly shows small pores

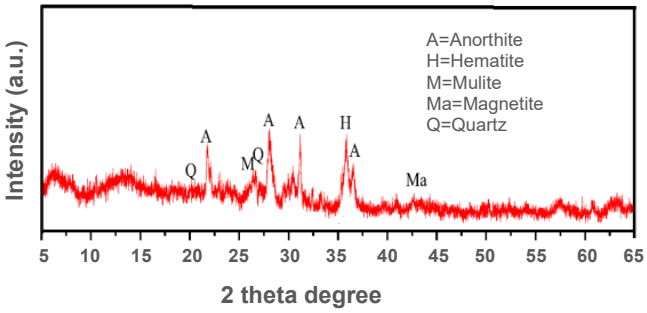
in the outer shell of the body system. The optical image of the SFA showed small pores on the surface. The XRD and SEM analyses of the CFA and SFA are shown in Figure 5.



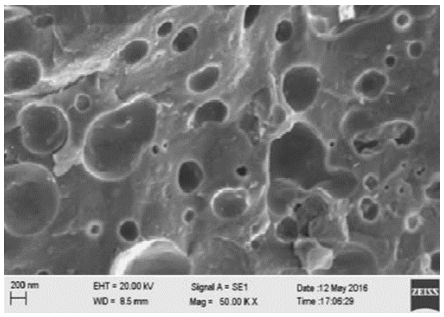
(a) XRD of CFA



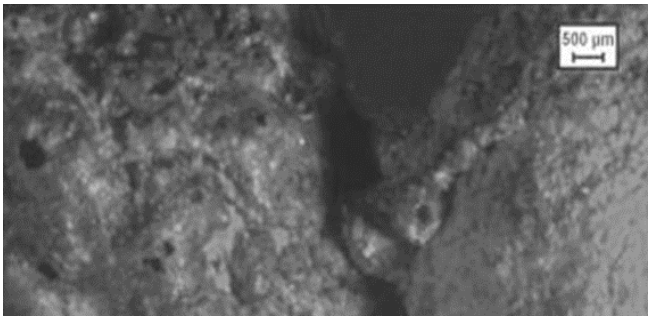
(b) SEM of CFA



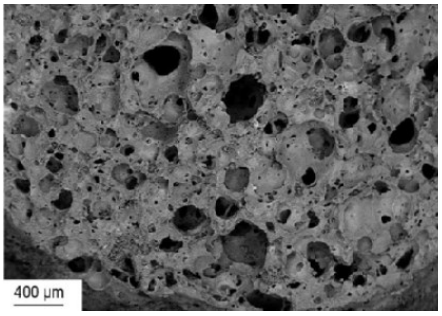
(c) XRD of SFA



(d) SEM of SFA



(e) Optical study of SFA



(f) Inner view of SFA

Figure 5. XRD and SEM analyses of CFA and SFA

4.4 Mechanical characteristics of FA aggregate

The mechanical/engineering properties of the different types of aggregates were analyzed according to the standard tests mentioned in Section 3.2. Figures 6-10 depict the different engineering characteristics of CFA and SFA.

FA with NaOH binder-based aggregate in the CBM showed a maximum CS of 6.30 MPa, whereas the second largest CS of 5,80 MPa was obtained for FA with sodium silicate binder-based aggregate from SM (Figure 6). A value of <10 signifies an exceptionally strong aggregate, whereas values of >35 typically signify weak aggregates; accordingly, all aggregate types are classified as strong.

Impact value of aggregate should not exceed 30 % for aggregate used for surface wearing course and should not exceed 45 % for surface other than wearing course; accordingly, all aggregate types are suitable for use according to impact test results (Figure 7.)

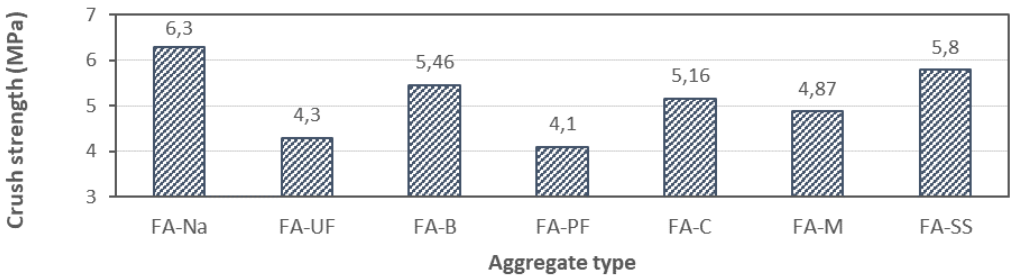


Figure 6. Crushing strength of different aggregate types

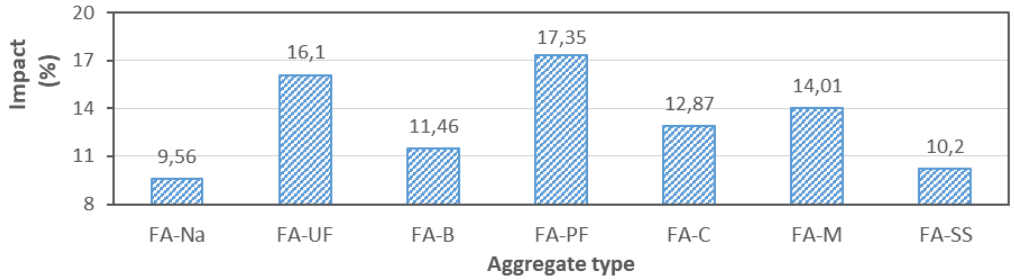


Figure 7. Impact test results of different aggregate types

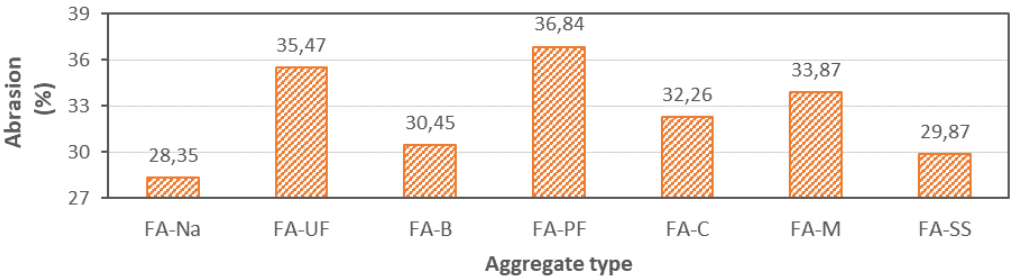


Figure 8. Abrasion test results of different aggregate types

The abrasion value of the aggregate should not exceed 30 % for aggregates used for the surface wearing course; only FA-Na and Fa-SS are suitable aggregate types (Figure 8). According to the values presented in Figure 9, all aggregates were classified as LWAs.

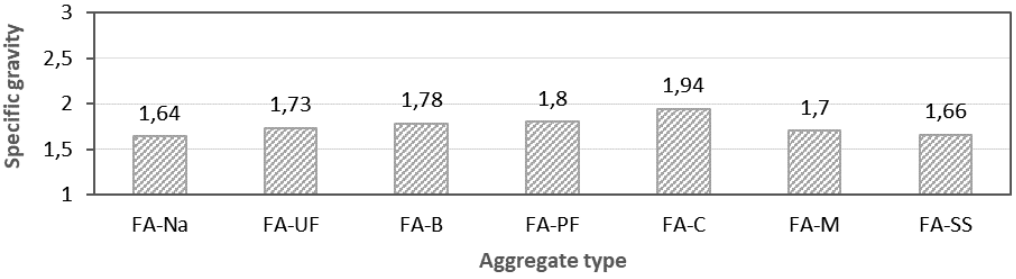


Figure 9. Specific gravity results of different aggregate types

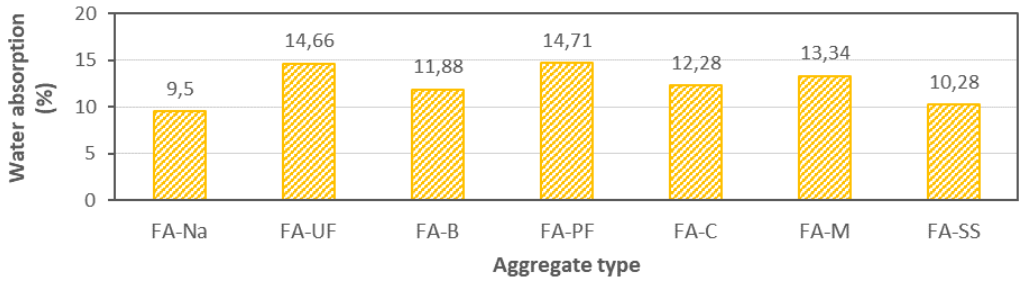


Figure 10. WA results of different aggregate types

The WA value decreased as the curing period increased from one to seven days. For cement-based aggregates, the specific gravity during curing increased, reaching the highest range (1,94). The WA test findings of the aggregates were used to estimate the aggregate porosity. The porous nature of the aggregate, as shown in Figure 10, accounted for its high absorption. This high absorption rate is not conducive to the creation of excellent concrete. Sealing all the holes is not a good way of limiting the WA because it increases the density of the aggregates. In addition, commercially available FA aggregates have a WA capability of 10-25 %. Surface treatment with certain polymers also reduces the absorption ability of the aggregates. The WA of the aggregates decreased as the sintering temperature increased. At high temperature levels, the surface of the aggregates develops a glassy texture, which may impede interpore connections. The use of a binder in the aggregate production process also resulted in significant reduction in the aggregate WA capacity.

4.5 Effect of binder on FA aggregate production

The ease with which the pellet is formed based on the quantity of raw FA modified to aggregate during pelletization. The mechanical strength of the pellets depends on the binder to water ratio. Optimum pelletization of aggregate was achieved by the addition of 17 % of sodium silicate binder. The maximum time limits for stable pellet formation and compaction were 10 min and 5 min, respectively. Thus, FA aggregate formation varies according to the type and quantity of the binder. The relationship between FA aggregate production and the binder is shown in Figure 11.

4.6 Effect of binder on CS

CFA (using a sodium hydroxide binder) yielded the highest CS value (6,30 MPa) followed by SFA (using a sodium silicate binder) (5,80 MPa). The strength property variation of the aggregate produced in both CFA and SFA techniques depends on its chemical composition, solid-to-liquid mass ratio, and the percentage of binder added to the FA. The CS of the aggregates plays an important role in determining the compressive strength of concrete materials.

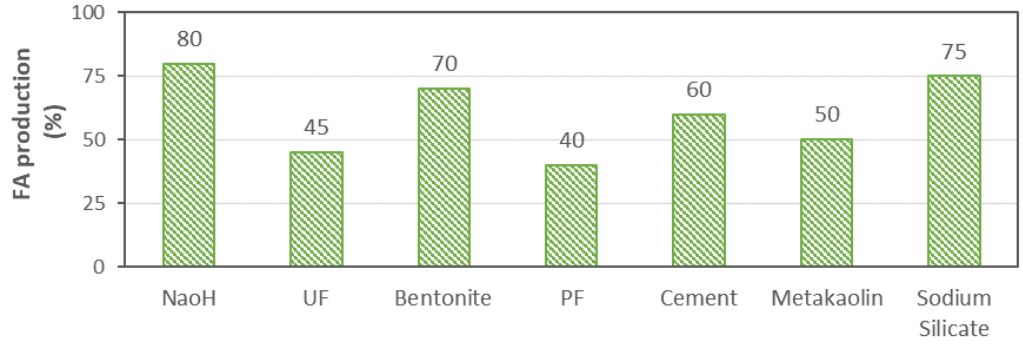


Figure 11. Relationship between FA aggregate production (%) and binder

The compressive strength of concrete depends on the CS of the aggregates. SFA aggregates showed optimum CS and low WA with no change in water quality parameters compared to CFA aggregates. Therefore, the sintered-aggregate-based concrete material exhibited more optimized performance.
Another advantage of the SFA aggregate is that as there is no change in the water quality parameter characteristics, this aggregate can be used for roadway applications.

4.7 Curing days and WA

WA values of the aggregates generated by both procedures decreased with increase in the typical water-curing period due to the contraction of the aggregate porosity.
The anhydride and calcium silicate phases in raw FA gradually transform into hydrated products such as ettringite, gypsum, and calcium silicate hydrate when the water curing period is increased from 1 to 7 d [15]. However, in the case of SFA aggregates, there was a linear decrease in the total porosity with increasing sintering temperature, which led to the low WA capacity of the sintered aggregate.

4.8 Water quality test

The following procedure can be used to prevent heavy metal leaching from FA. Using X-ray diffraction analysis, bioelectrochemical systems (BES) and electrolysis reactors (ER) were investigated for heavy metal removal from FA leachate, which had high detectable amounts of Zn, Pb, and Cu. The removal of heavy metals from FA leachate using a combination of BESs and ER was demonstrated. Cu (II), Zn (II), and Pb (II) metal removal efficiencies were 98,5 %, 95,4 %, and 98,1 %, respectively [21].
A plant extract called saponin and extracted from the fruits of Sapindus laurifolia (S. laurifolia) using both chemical and aqueous processes was used to remove harmful components from coal-based FA used in thermal power plants [22]. Thermal and chlorination methods can be used to remove heavy metals from MSS FA efficiently. The findings show that during the thermal treatment, approximately 80 to 89% of Pb, 48 to 56 % of Cd, 27 to 36 % of Zn, and 6 to 24 % of Cu were volatilised in the pure MSS FA.
In particular, for the mid-volatile elements, Cu and Zn, the chlorinating agents appeared to promote the removal of heavy metals. More than 96 % of Pb, 78 % of Cd, 76 % of Zn (excluding NaCl), and 86 % of Cu (excluding NaCl) may be eliminated from MSS ash with the use of 10 wt% chlorinating chemicals. With an increase in the Cl proportion, the evaporation rate of heavy metals increased; however, the effect clearly varied [23].
Leaching tests were conducted using CFA and SFA aggregates, and the results are presented in Figures 12-14 as measures of the water quality. This demonstrates that there was a gradual rise in water parameters such as pH, total dissolved solids (TDS), and turbidity with an increase in the curing time of CFA aggregates. The maximum pH value obtained after 3 days (72 h) of curing was 12,26 and the TDS value was 6.96 with a turbidity of 88,24 NTU. In SFA, negligible changes were observed in the water parameter values as pH was 7,2; TDS was 1,73; and turbidity 3,88 NTU with 3 days of curing.
Figure 9 depicts the results of the CFA and SFA. Although the binder is important for the pellet strength in the case of CFA aggregates, heavy metal leaching from these pellets cannot be controlled. In the case of SFA aggregates, increasing the sintering temperature aids in boosting the aggregate strength while maintaining stable water quality parameters. According to previous studies, the SFA aggregate is superior to the CFA aggregate.

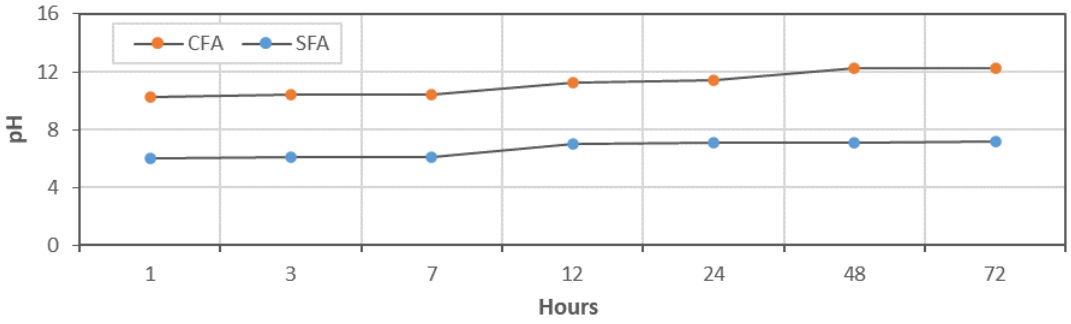


Figure 12. Change of pH during curing for CFA and SFA aggregates

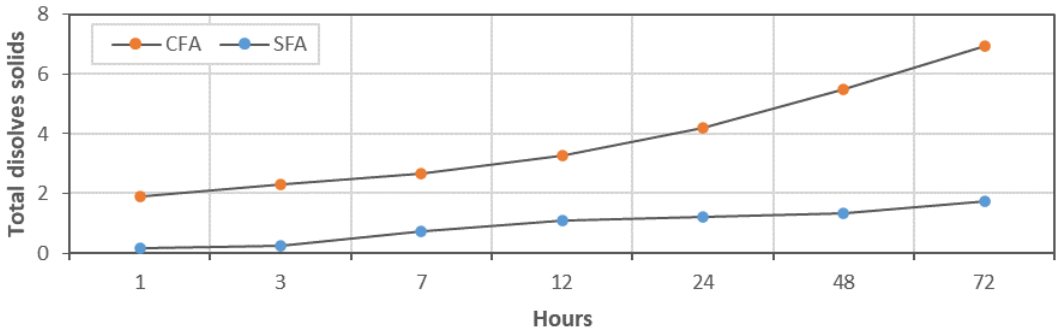


Figure 13. Change in TDS during curing for CFA and SFA aggregates

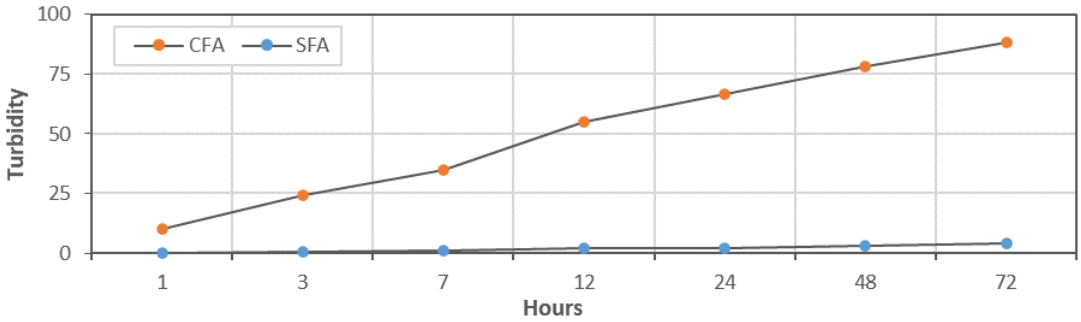


Figure 14. Change in turbidity during curing for CFA and SFA aggregates

5 Conclusions

In this study, FA was successively assembled into lightweight aggregates using cold bonding and sintering. The outcomes of this study, based on the experimental observations and results, are as follows:

- In both cases, as the duration of pelletization increased, the pellets increasingly became spherical with remarkable strength, indicating the high efficiency of the pellets.
- Production of FA aggregates (80 %) can be achieved with the addition of FA to binder (sodium hydroxide) at a ratio of 15:85. Similarly, it can be obtained (75 %) with the inclusion of FA a binder (sodium silicate) at a ratio of 17:83.
- In the case of CFA aggregates, the strength depends on the duration of water curing, whereas for SFA aggregates, the strength depends on the increase in temperature.
- CFA (using a sodium hydroxide binder) showed a maximum CS value of 6,30 MPa compared to 5,80 MPa for SFA (using a sodium silicate binder).

- The WA value of CFA (using sodium hydroxide binder) was found to be 9,50 %, whereas that of SFA (using sodium silicate binder) was 10,28 %.
- SFA exhibited zero leachability but CFA may present the possibility of leaching.
- The above results revealed that SFA aggregates exhibited optimum CS, zero leachability, and standard WA values. Thus, SFA aggregates have improved as building materials and have more optimized potential than CFA aggregates.

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