Evaluation of Alternative Sources of SCMs for Concrete Materials

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ABSTRACT

This study investigated the use of alternative sources of supplementary cementitious materials (SCMs), namely landfilled reclaimed fly ash (RFA) and reclaimed ground bottom ash (GBA), for its beneficial use in concrete. The study characterized RFA and GBA along with conventional Class F fly ash (FA), as control. Furthermore, the influence of the three ashes on the fresh and hardened properties of concrete was investigated (when used to replace 10% of cement by mass). Experimental results revealed that all the ashes were classified as class F fly ash according to ASTM C618. Yet, in contrast to FA, RFA and GBA presented an increased water requirement, which was attributed to the differences in micro-morphology. Furthermore, RFA exhibited a lower strength activity index (SAI), while GBA presented a higher SAI, in contrast to FA. This was attributed to the CaO contents of the ashes, which was lowest for RFA and highest for GBA. RFA admixed concrete exhibited a slight decrease in workability, while GBA admixed concrete exhibited a significant decrease. This was attributed to the irregular shape of GBA particles. The control concrete mixture (with no ashes) exhibited the highest air content among all concrete mixtures, while GBA admixed concrete exhibited the lowest air content. Notably, while RFA exhibited the highest loss on ignition (LOI), RFA admixed concrete presented the highest amount of air from all concrete mixtures implementing ashes, including FA. In terms of hardened properties, all coal ashes had a minimal influence on the compressive strength of concrete, producing marginal decrements in strength. Furthermore, all concrete mixtures with ashes exhibited a slight increase in surface resistivity compared to control.

INTRODUCTION

Concrete is the most used manmade material in the world, with nearly 33 billion metric tons being produced each year (ISO 2016; Lomborg 2003). The wide use of concrete is driven by the readily availability of its raw components and its simplicity of fabrication, cost-effectiveness, and mechanical properties. However, the extensive use of concrete has a significant impact on the environment. Ordinary Portland cement (OPC), which is the most commonly used binder in concrete materials, draws an environmental concern both in terms of the impact caused by the extraction of raw materials and the large amounts of CO_2 emitted during cement manufacture. As such, the use of supplementary cementitious materials (SCMs) to partially replace cement in the manufacture of concrete is an excellent tool to decrease the environmental impact of concrete. Besides the environmental benefits of SCMs, these materials are often implemented to enhance concrete properties and durability. SCMs commonly used are industrial by-products such as fly ash, ground granulated blast-furnace slag (GBBS), and silica fume. Nevertheless, agricultural waste materials such as palm fuel oil ash, rice husk ash, wheat straw ash, and sugarcane bagasse ash have also been used as SCMs due to their pozzolanic characteristics (Chindaprasirt et al. 2007; Ganesan et al. 2007; Nair et al. 2008; Subedi et al. 2019). In the US, the most utilized SCM is fly ash due to its historically vast accessibility and beneficial effects, including: (1) improving concrete's durability; (2) enhancing concrete's fresh and long-term hardened properties; (3) lowering concrete's cost; and (4) reducing concrete's environmental impact (FHWA 1995). As such, fly ash has become an integral part of the concrete industry in the US. However, the pronounced decline in coal-fired power generation in the US in combination with the expected increase in demand of fly ash as SCM raises serious concerns about the long-term accessibility of fly ash (US EIA 2019). Consequently, there is a need to find alternative sources of SCMs that are high quality, cost-effective, and readily available to address the expected shortage of fly ash in the US.

Due to storage constraints, market disparities, or failure to meet the ASTM C618 requirements, large amounts of the fly ash produced are not utilized and disposed of in landfills or surface impoundments (Al-Shmaisani et al. 2019). Landfilled fly ash contains relatively low amounts of water (i.e., 10-20% moisture content) and can be reclaimed through minor processing (i.e., drying) for its use in concrete (Al-Shmaisani et al. 2019). Some recent studies have shown that even after disposal, reclaimed fly ash (RFA) remains pozzolanic and can exhibit similar properties as those of conventional Class F fly ash (Boral Resources 2018; Diaz-Loya et al. 2019). Along with fly ash, bottom ash is also produced in coal-fired electrical power stations. While fly ash is the fine particle ash that rises up with the flue gas, bottom ash is the heavier ash that does not rise and is collected from the bottom of the furnace (FHWA 2016). The coal bottom ash is a granular, coarse, and incombustible ash that is typically used as fine aggregates in concrete and asphalt mixtures, or disposed of in landfills (Churchill and Amirkhanian 1999; Ghafoori and Bucholc 1996). Nevertheless, this coal ash can exhibit some pozzolanic activity due to its content of silica, alumina, and iron oxide (Argiz et al. 2017). Yet, bottom ash has a large particle size and a highly porous microstructure, resulting in high water requirement and low compressive strength. Nevertheless, processed bottom ash (by grinding) with particle size comparable to that of fly ash decreases its porosity and enhances its pozzolanic activity (Argiz et al. 2017). Such types of bottom ash can be used as SCMs in concrete. Some recent studies have developed bottom ash with sufficient quality to be used as SCMs (Argiz et al. 2017; Mangi et al. 2019a; Mangi et al. 2019b)

For the same type of SCM, properties can vary significantly depending on the supplier and source (Al-Shmaisani et al. 2019). This highlights the importance of characterizing potential SCM materials on a supplier and source basis. The objective of this study is to investigate the feasibility of alternative sources of SCMs, namely landfilled reclaimed fly ash (RFA) and reclaimed ground bottom ash (GBA) for its beneficial use in concrete. The sources evaluated in the present study have not been investigated in previous research and could be used to produce concrete materials in Region 6. The utilization of unconventional sources of SCMs can expand the portfolio of SCMs available in the region and enhance the cost-effectiveness and greenness of concrete materials by valorizing waste products and reducing cement consumption.

METHODOLOGY

Characterization of Coal Ashes

Three different types of coal ashes, i.e., ASTM C618 Class F fly ash (FA), reclaimed fly ash (RFA), and reclaimed ground bottom ash (GBA), were utilized in this study. The FA, GBA, and RFA were received from Illinois, Texas, and Georgia, respectively. All the ashes were used as received from the supplier, with no further processing. The three ashes were characterized for their chemical composition, micro-morphology, and physical properties. X-ray fluorescence (XRF) and X-ray diffraction (XRD) were performed to identify mineral compositions. XRF scan was done with Rigaku Supermini200 (Rigaku Corporation, Japan) and then calibrated with three standard fly ash samples from the National Institute of Standards and Technology (NIST). XRD was done with a Bruker-AXS D8 Advanced (Bruker Corporation, MA, USA) with Cu source (Cu Kα radiation, $\lambda = 1.54178$ Å) and a Lynxeye PSD detector. The XRD spectra were analyzed with the Profex software (Doebelin and Kleeberg 2015). SEM was performed with the JEOL JSM-7500F (JEOL USA Inc., MA, USA) to understand the micro- morphology. The SEM samples were prepared by depositing a thin layer of ash onto carbon tape on top of an aluminum stub. The samples were then sputter-coated with 5nm of palladium-platinum alloy to avoid charging in SEM. Several physical properties such as moisture content (MC), loss on ignition (LOI), water requirement, and strength activity index (SAI) were evaluated following ASTM C311/ C311M standard (ASTM 2018).

Table 1. Concrete Mixture Proportions

ID	Ash Type	Cement (kg/m^3)	Ash (kg/m^3)	Ash (%)[a]	Coarse Aggregate (kg/m^3)	Fine Aggregate (kg/m^3)	Water (kg/m^3)	HRWR (L/m^3)	AEA (L/m^3)
CO	-	344.1	0	0	1058.2	743.7	260	0.42	0.36
FA-10	FA	309.7	34.4	10	1058.2	734.0	260	0.42	0.36
RFA-10	RFA	309.7	34.4	10	1058.2	738.5	260	0.42	0.36
GBA-10	GBA	309.7	34.4	10	1058.2	732.4	260	0.42	0.36

[a] % replacement of cement by mass

Concrete Mixture Proportions and Testing

***Materials*:** All concrete mixtures were produced using Type I ordinary Portland cement (OPC), limestone as a coarse aggregate, and concrete sand as fine aggregate. The coarse aggregate presented a maximum nominal particle size of 19 mm, a specific gravity of 2.68, and absorption of 0.8%. On the other hand, the concrete sand presented a maximum nominal particle size of 4.75 mm, a specific gravity of 2.65, and absorption of 0.4%. Aggregate size was determined from sieve analysis, while the specific gravity and absorption of the coarse and fine aggregate were determined according to ASTM C127 and ASTM C128 standards, respectively (ASTM 2015a; ASTM 2015b).

***Concrete Mixture Proportions*:** Concrete mixtures were prepared to evaluate the effect of the different ashes on the fresh and hardened properties of concrete at a cement replacement level of 10% by mass, as shown in Table 1.

The control concrete mixture (i.e., CO) was designed to resemble the characteristics of a type A1 structural class concrete per the Louisiana Department of Transportation and Development specifications (Louisiana Department of Transportation and Development 2016). The water-to-binder ratio (w/b) and binder content were kept constant for all concrete mixtures at 0.45 and 344.1 kg/m^3, respectively. Furthermore, a polycarboxylate-based high-range water reducer (HRWR) was used for all concrete mixtures at a constant dosage of 1.2 mL per kg of binder to enhance concrete workability. Carbon content in SCMs such as coal ashes can disrupt air-entrainment in concrete materials, which in turn can negatively affect the performance of concrete subjected to freeze/thaw conditions (American Coal Ash Association 2013; Folliard 2009). To evaluate the air-entrainment disruption potential of the different ashes evaluated in this study, all concrete mixtures implemented an air-entrainment admixture (AEA) at a constant dosage of 1.1 ml per kg of a binder. All concrete mixtures were prepared using a drum mixer with 1 ft^3 capacity. Initially, the coarse aggregate, 2/3 of the mixing water, and AEA were added and mixed for two minutes. Subsequently, all the components were added and mixed for another three minutes. Next, the mixture was allowed to rest for three minutes, and finally mixed for three additional minutes.

Concrete Testing: Upon completion of the concrete mixing process, the slump and air content were evaluated according to ASTM C143/ C143M and ASTM C231/C231M (ASTM 2020a; ASTM 2017), respectively. Subsequently, three cylinders with diameter of 101.2 mm and height of 202.4 mm (4 in x 8 in) were cast to evaluate the compressive strength and surface resistivity (SR) of all the concrete mixtures. After casting, all the cylindrical specimens were covered with a plastic cap to prevent moisture loss and allow to harden in the laboratory. After 24 hours, specimens were demolded, placed in a lime saturated water tank, and allowed to cure for 28 days, according to ASTM C511 (ASTM 2019b). Upon completion of curing, AASHTO T358 SR test was conducted using a Wenner four-pin array with a 38 mm (1.5 in.) spacing (AASHTO 2017). This test was used to gain insight into the concrete materials' durability as the SR test provides a rapid indication of concrete’s resistance to chloride ion penetration. A curing correction factor of 1.1 (for lime saturated water tank curing) was applied on the average SR values for all concrete mixtures (AASHTO 2017). Right after the SR test was completed, the 28-day compressive strength was evaluated according to ASTM C39/C39M (ASTM 2020b). The specimens were tested at a loading rate of 15 MPa/min utilizing a concrete hydraulic compression testing machine (Forney LT-8031-FTS).

RESULTS AND DISCUSSION

Chemical and Physical Properties of Coal Ashes

***XRF, Moisture Content, LOI, XRD, and Microstructure*:** The results of XRF are presented in Table 1. As shown, both FA and GBA have a significant amount of calcium oxide (CaO) compared to RFA. However, none of these ashes reached the necessary threshold to be classified as Class C fly ash (i.e., >18% CaO) per ASTM C618. Furthermore, FA contained more iron oxide (Fe_2O_3) than the other two ashes. It is worth mentioning that the sum of the total pozzolanic component (i.e., the sum of SiO_2, Al_2O_3, Fe_2O_3) in all coal ashes was significantly higher than 50%; thus, meeting ASTM C618 requirement to be classified as Class F fly ash (ASTM 2019a). Moreover, none of the ashes evaluated surpassed the sulfur trioxide (SO_3) limit of 5%.

Table 2. Chemical composition results from XRF.

%	CaO	SiO_2	Al_2O_3	Fe_2O_3	SO_3	MgO	K_2O	Na_2O	SiO_2+ Al_2O_3+Fe_2O_3
FA	8.4	57.2	20.2	10.2	1.2	1.6	2.7	1.1	87.6
RFA	1.8	53.4	28.0	7.7	0.1	0.99	2.2	0.3	89.1
GBA	11.0	62.0	20.8	6.9	0.5	2.8	0.9	0.3	89.7

The moisture content and LOI for all coal ashes are presented in Table 3. It is important to notice that the moisture content for all coal ashes is lower than that of OPC. The results show that RFA has the lowest moisture content but highest LOI among the three ashes. LOI is a measurement of unburned carbon remaining in the material. As such, high LOI indicates that RFA has higher carbon content than FA and GBA. Nevertheless, all ashes met the requirement for moisture content (i.e., maximum limit of 3%) and LOI (i.e., maximum limit of 10%) to be classified as a Class F fly ash per ASTM C618 (ASTM 2019a).

Table 3. Moisture content and LOI of OPC and coal ashes

Material	Moisture Content	LOI
OPC	0.58%	-
FA	0.40%	1.42%
RFA	0.06%	2.95%
GBA	0.27%	0.92%

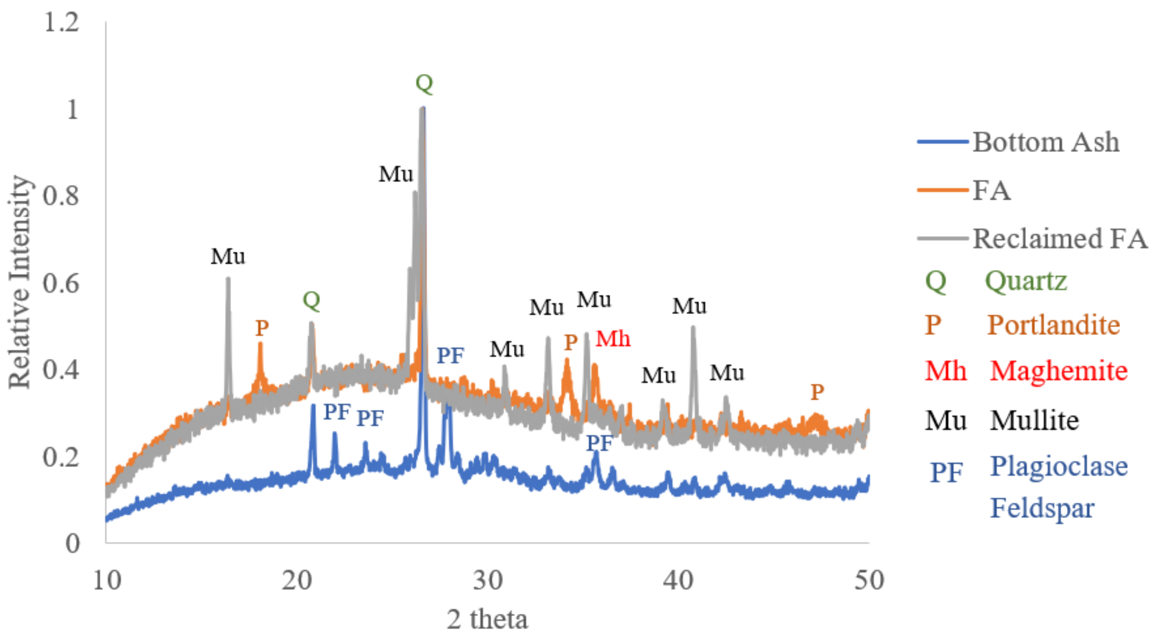
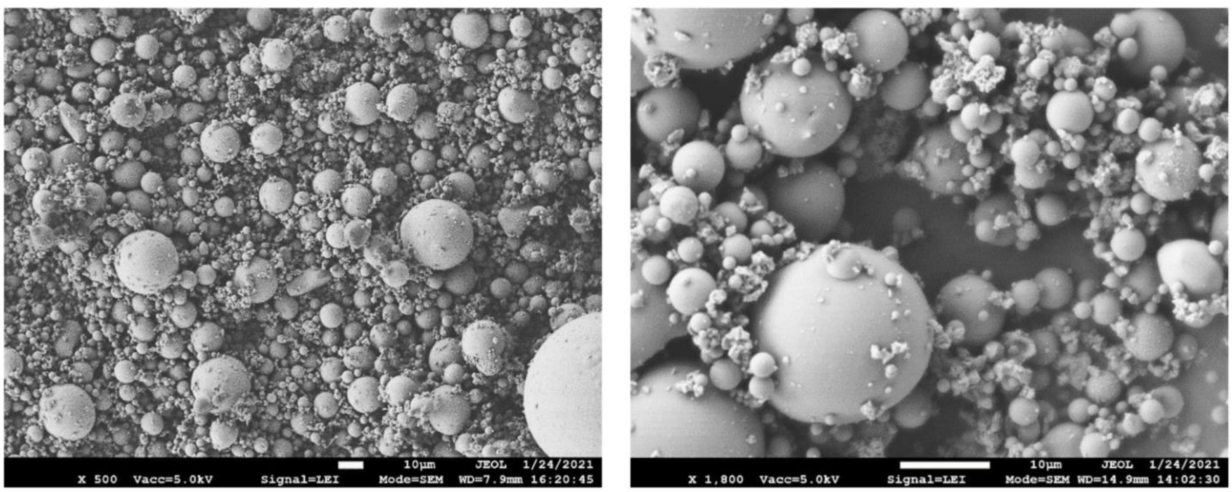


Figure 1. XRD spectra of coal ashes with identified mineral phases.

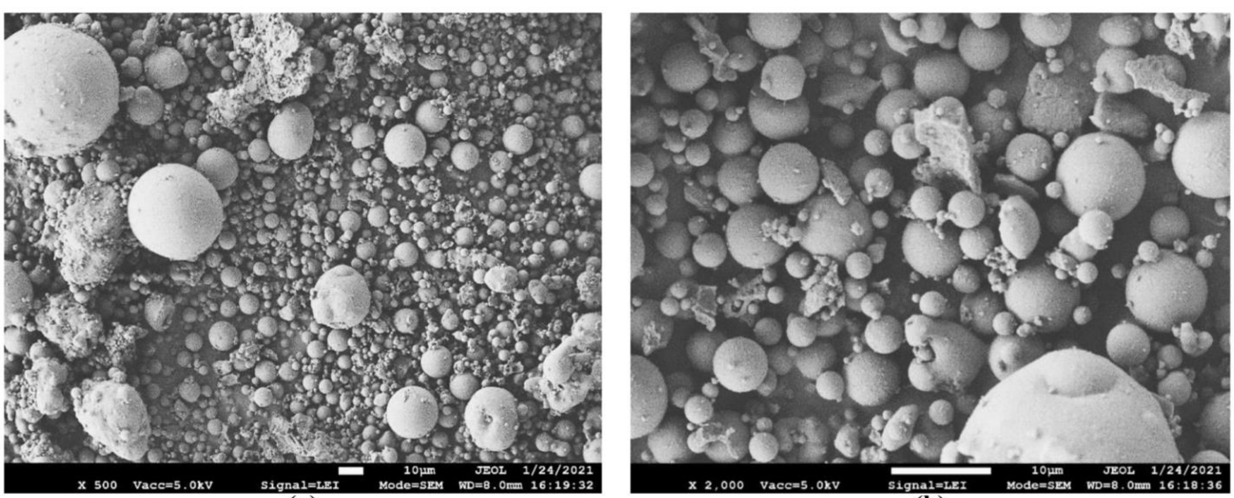
Figure 1 shows the XRD spectra for all three ashes. While FA and RFA have a significant portion of amorphous phases (indicated by the large hump between 15 to 30° 2θ), a significant amorphous hump cannot be observed in the plot for GBA. It is worth mentioning that XRF was conducted and analyzed prior to conducting XRD so that it could aid in identifying the phases from XRD. As for phase identification, all three ashes contain large quartz peaks, which is common among coal ashes. For the rest of the phases, using the information obtained from XRF, FA has peaks that correspond to portlandite ($Ca(OH)_2$) and maghemite (γ-Fe_2O_3), RFA contains mullite ($2SiO_2 \cdot 3Al_2O_3$), while GBA contains plagioclase feldspar (likely anorthite due to the high Ca and low Na content from XRF).

The morphology of SCMs has a significant influence on the fresh and hardened properties of concrete mixtures. As such, SEM imaging of all three coal ashes was performed in this study. Figures 2, 3, and 4 show the micromorphology of FA, RFA, and GBA, respectively, in low and medium magnification. Both FA and RFA have similar morphology with the spherical particles. However, the impurities in FA are mostly small in size, while those in RFA were similar to or larger than the impurities observed in FA. On the other hand, GBA has an entirely different morphology. GBA contains irregular prismatic particles, which are much larger than those of FA and RFA.



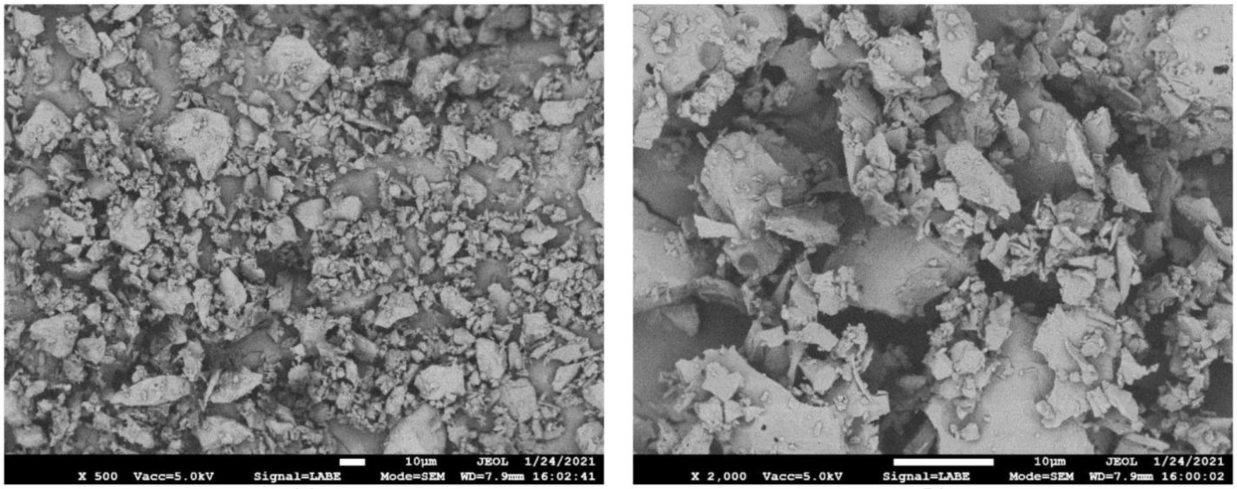
(a) (b)

Figure 2. SEM images of FA (a) Low magnification and (b) Medium magnification



(a) (b)

Figure 3. SEM image of RFA: (a) Low magnification and (b) Medium magnification



(a) (b)

Figure 4. SEM image of GBA (a) Low magnification and (b) Medium magnification

***Water Requirement and SAI*:** The water requirement, 7-day cube compressive strength, and SAI for all mortar mixtures containing coal ashes are presented in Table 4. The results show that FA can reduce the water requirement by 10%, while neither the RFA nor the GBA reduces the water requirement significantly. However, both RFA and GBA required less water than OPC to achieve similar flowability as the control mixture. The higher water requirement for RFA compared to FA is most likely due to the prominent presence of large mullite particles (Figure 3). In the case of GBA, the higher water requirement in contrast to FA is likely due to the irregular and larger particles (Figure 4).

In terms of the compressive strength at seven days of curing, the replacement of OPC with GBA showed no significant difference, while FA shows a minor decrease, and RFA shows a significant decrease. The corresponding SAI for FA, RFA, and GBA was 94.7%, 79.4%, and 98.1%, respectively. As such, all ashes exhibited higher SAI than 75% at seven days of curing, thus meeting ASTM C618 requirement to be classified as a Class F fly ash. It is expected that both FA and RFA would show a more significant increase for the 28-day compressive strength from the dissolution of the amorphous aluminosilicate phases. Therefore, the decrease in early strength could be compensated by the long-term strength gain.

Table 4. Physical properties of the ashes.

Mixture	Water Requirement	W/C Ratio	7-Day Compressive Strength (MPa)	SAI (%)
OPC	100.0%	0.484	26.7 ± 1.1	-
OPC + 20% FA	89.5%	0.433	25.3 ± 0.6	94.7
OPC + 20% RFA	95.0%	0.460	21.2 ± 1.0	79.4
OPC + 20% GBA	97.8%	0.473	26.2 ± 1.3	98.1

Properties of Concrete Admixed with Coal Ashes

***Slump and Air Content*:** The slump and air content for all concrete mixtures evaluated in the study are presented in Figure 5a and 5b, respectively. It is observed that the replacement of cement with FA did not influence the slump (i.e., the slump was 3.5 inches for both control and FA-10). On the other hand, RFA admixed concrete exhibited a slight decrease in slump (i.e., to

3.25 inches), while the GBA admixed concrete exhibited a dramatic reduction in slump (i.e., to 1.75 inches). From the SEM images, it was observed that GBA consists of irregularly shaped prismatic particles that are highly angular. This in turn, is likely the reason for the negative influence of GBA on the workability of concrete mixtures. It is also important to mention that even though both RFA and GBA mortars presented slightly lower water requirements than the pure OPC mortar (as shown in Table 4), a decrease in a slump was observed for both RFA and GBA admixed concrete in contrast to the control concrete mixture. Nevertheless, the tendency in water requirement was in agreement with the observed slump tendency (i.e., water requirement for GBA>RFA>FA, while slump for GBA-10<RFA-10<FA-10).

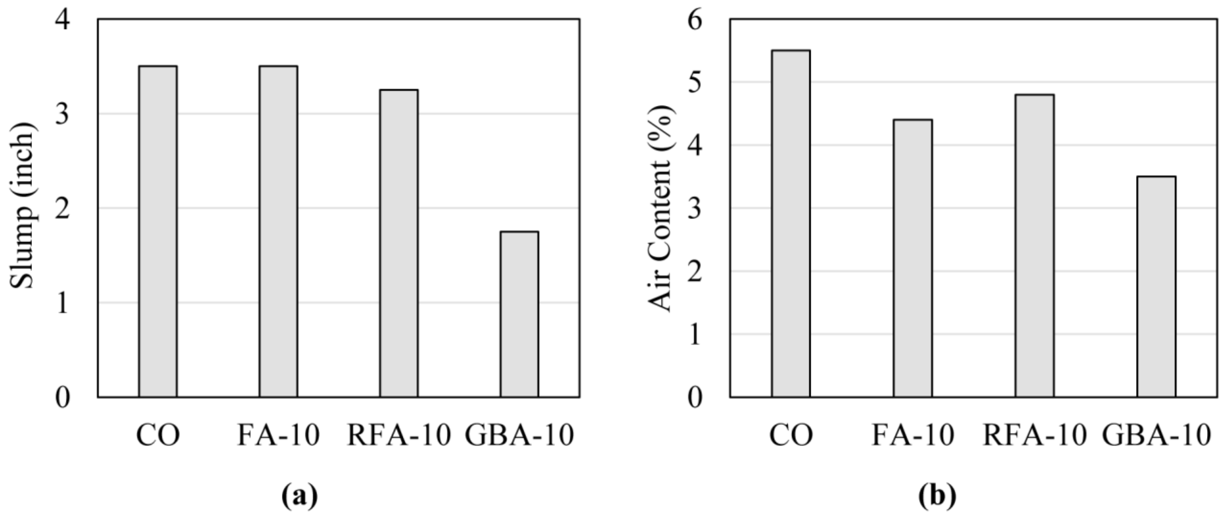
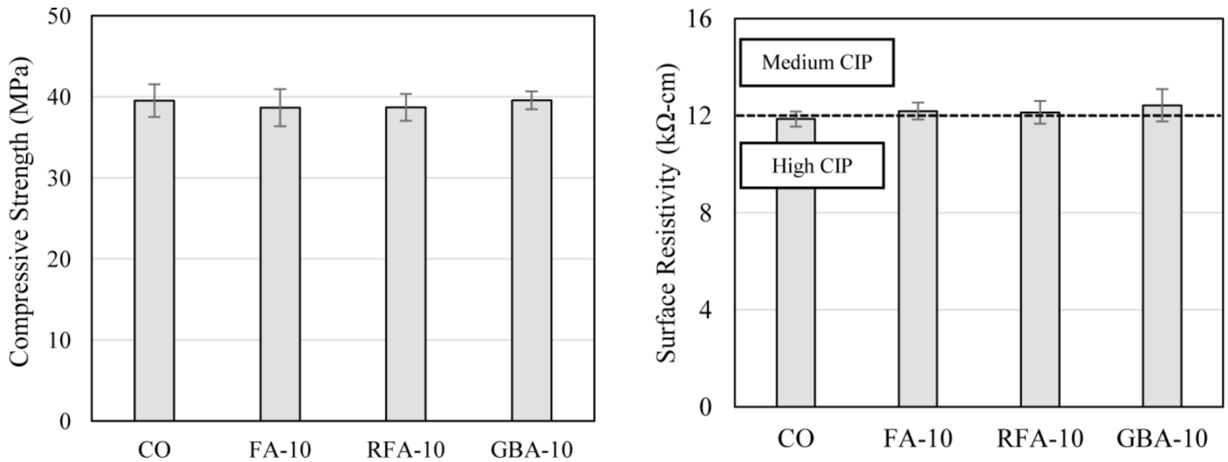


Figure 5. Concrete Fresh Properties: (a) Slump and (b) Air Content

In terms of air content, it is observed that the control mixture exhibited the highest air content (i.e., 5.5%) among all concrete mixtures, while GBA admixed concrete exhibited the lowest air content (i.e., 3.5%). From previous studies, it is known that the amount of entrained air decreases with the increase in surface area (fineness) of cement and/or SCMs and increase in organic coal residuals (unburned carbon, char particles) (Folliard 2009). This is primarily due to the AEA adsorption effect. During the mixing procedure, AEA (surfactant) is adsorbed from solution onto the organic coal residuals surface; thus, making AEA less effective in stabilizing air bubbles in fresh concrete mixtures (Folliard 2009). Even though RFA exhibited the highest LOI among the three coal ashes, interestingly, RFA-10 did not exhibit the lowest air content among all concrete mixtures. The RFA-10 concrete exhibited a higher air content than FA-10. Based on various studies, it is also known that apart from organic coal residuals mass, coal residuals form, surface area, surface polarity, and pore size also play roles in AEA adsorption (Freeman et al. 1997; Hill et al. 1997; Hurt and Suuberg 2004). As such, there may not be a general correlation between LOI and air content (Folliard 2009).

***Compressive Strength*:** The 28-day compressive strength of all concrete mixtures is presented in Figure 6a. It can be observed that the different ashes had a marginal influence on the compressive strength. It is relevant to mention that the differences in the average compressive strength between the concrete mixtures were not statistically significant per ANOVA (p-value=0.87). While the hydration reaction occurs as soon as the water is added to the cement, the pozzolanic reaction occurs at a later age. Since the compressive test was conducted at 28 days of curing, the pozzolanic reaction did not produce a significant effect in strength. Nevertheless, it is expected that the concrete mixtures implementing the different ashes will exhibit a higher long-term compressive strength in contrast to control. It is also important to mention that the investigated cement replacement level with the different ashes was only 10%. Therefore, significant effects on strength are not expected. As such, future research should be directed towards evaluating concrete mixtures using the different ashes at higher replacement levels of cement and different curing ages.



(a) **(b)**

Figure 6. Concrete Hardened Properties: (a) Compressive Strength and (b) Surface Resistivity

Surface Resistivity**:** Figure 6b presents the surface resistivity test results of all concrete mixtures. It can be observed that all concrete mixtures implementing ashes exhibited a slightly higher SR compared to control. For instance, the SR of the control mixture was 11.9 kΩ-cm, which increased to 12.2 kΩ-cm (i.e., an increase of 2.5%), 12.1 kΩ-cm (i.e., an increase of 1.6%), and 12.4 kΩ-cm (i.e., an increase of 4.2%) for FA-10, RFA-10, and GBA-10, respectively. It is worth mentioning that all concrete mixtures, except for control, presented SR values higher than 12 kΩ-cm, thus falling into the category of medium chloride ion penetrability as per AASHTO T 358 (AASHTO 2017). The control mixture fell in the category of high chloride ion penetrability, which might be attributed to the high air content presented in Figure 5b. As such, the slight increase in the SR of concrete mixtures implementing the different ashes is likely due to the lower air content compared to the control mixture. It is also possible that the ashes produced a filler effect, thus decreasing concrete permeability.

CONCLUSION

This study investigated the feasibility of using reclaimed fly ash (RFA) and reclaimed ground bottom ash (GBA) as SCMs for concrete mixtures. The study thoroughly characterized RFA and GBA along with conventional Class F fly ash (FA), as control, to determine whether these materials met the requirement to be classified as a pozzolan according to ASTM C618. Furthermore, the influence of the different coal ashes (FA, RFA, and GBA) on the fresh and hardened properties of concrete was investigated when used to replace 10% of cement by mass. Based on the experimental results, the following conclusions can be drawn:

- All the ashes evaluated met the requirements for pozzolanic component, CaO, SO_3, moisture content, LOI, water requirement, and SAI to be classified as Class F fly ash according to ASTM C618. Compared to FA, RFA presented a lower SAI, an increased water requirement, and a lower CaO content. On the other hand, GBA exhibited a higher SAI, an increased water requirement, and a higher CaO content, in contrast to FA. The differences in SAI reported were mainly attributed to the CaO content of the ashes, while the increments in water requirement for RFA and GBA were associated with their micro-morphology. Both FA and RFA presented similar morphology with spherical particles, however, RFA presented more and coarser impurities. On the other hand, GBA consisted of irregular angular prismatic particles. The three ashes contained large quartz peaks in the XRD spectra. Furthermore, FA and RFA consisted of a significant portion of amorphous phases, while bottom ash did not exhibit a significant amorphous phase hump in the XRD spectra.

- FA admixed concrete produced the same slump as that of the control concrete mixture (i.e., 3.5 inches). In the case of RFA admixed concrete, a slight decrease in the slump was observed (i.e., 3.25 inches). Interestingly, GBA admixed concrete exhibited a significant decrease in workability (i.e., 1.75 inches). This was attributed to the micro-morphology of GBA. In terms of air content, the control mixture exhibited the highest amount of air, while the GBA admixed concrete exhibited the lowest air content. Notably, even though RFA exhibited the highest LOI among the three coal ashes investigated, the concrete mixture containing RFA exhibited the highest amount of air from all concrete mixtures implementing ashes, including FA. All ashes had a minimal influence on the 28-day compressive strength of concrete, producing marginal decrements in strength (i.e., up to 2.2%). Furthermore, all concrete mixtures implementing coal ashes exhibited a slight increase in surface resistivity compared to control (i.e., up to 4.2%).

Overall, from the present experimental results of an ongoing Tran-SET funded research project, it can be concluded that both RFA and GBA could be potentially used as SCMs for the production of concrete materials. Both coal ashes met ASTM C618 requirements to be classified as Class F fly ash and produced concrete with similar strength and surface resistivity as that of concrete using conventional Class F fly ash (i.e., FA-10). Nevertheless, further research is being conducted to: (1) determine the influence of higher levels of cement replacement with RFA and GBA on the mechanical properties of concrete; and (2) determine the influence of RFA and GBA on the long-term properties of concrete.

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