The Influence of Fly Ash After Change to Low-NOx Burners on Concrete Strength – Case Study

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ABSTRACT

Coal-burning electric utilities have increasingly turned to low-NOx burners (LNB) to reduce nitrous oxide emissions. It is well documented that changes in the form and quantity of unburned carbon in low-NOx fly ash, as well as the possible presence of ammonia compounds, have negatively impacted the use of fly ash in concrete. Less well documented are possible changes in low-NOx fly ash that may impact strength activity index (SAI) values, including particle shape and size and glass content. In this study samples of fly ash were obtained before and after the change to LNB at a plant consistently burning a southern lignite from one mine. Samples were tested for Strength Activity Index (SAI) according to ASTM C 311, affinity for air-entraining admixtures, chemical composition, particle size and shape, glass content, and phase assemblage by quantitative x-ray diffraction (Q-XRD). SAI values at 3, 7, 28 and 56 days for low-NOx fly ash were consistently 10% lower than the corresponding values for the fly ash before the burner change. No significant change in bulk chemical composition was found between samples taken before and after LNB installation. Fly ash produced after LNB went online have slightly greater and more active carbon contents, more coarse particles, and slightly less glass which can explain the reduction in SAI observed. Although this study involves samples from only one plant, it does document the possibility of lower SAI values after changing to LNB. Since fly ash is extensively used as a pozzolanic material in concrete, the reduced SAI can affect the strength development of concrete. Chemical admixtures have been used in concrete containing this low-NOx fly ash to restore strengths to the level prior to LNB installation.

INTRODUCTION

Approximately 40% of the over 71 million tons of fly ash collected at American coal-burning power plants is utilized. The primary market for this fly ash is as a pozzolan to replace cement in concrete.[1] Although nitrous oxides (NOx) produced at power plants comprise a relatively small percentage of total NOx emissions to the atmosphere, electric generators have been under increasingly strict limits on NOx emissions since 1990 when the Clean Air Act was approved.

NOx reduction technologies include 1.) Low-NOx burners (LNB), 2.) Overfire air (OFA), and 3.) Post-combustion Selective Catalytic Reduction (SCR) or Selective Non-Catalytic Reduction (SNCR). The key to NOx reduction is a longer, cooler flame and a much more reduced atmosphere compared with traditional burners. Differences in low-NOx fly ash compared to fly ash from conventional boilers have been predicted, however, relatively little is known about how fly ash characteristics are affected by the different types of NOx reduction technology.[2] Key characteristics of fly ash for concrete quality that may change with low-NOx burners include unburned carbon content (LOI), particle size distribution, ash morphology, ash reactivity, composition, and the introduction of ammonium salts.[3,4,5]

This study was undertaken to attempt to determine why concrete mixed with a particular fly ash had approximately 10% lower compressive strength after the power plant made changes to reduce nitrous oxide emissions. Samples of fly ash collected before and after the change to low-NOx burners were examined in detail to attempt to determine what characteristics of the fly ash changed and how these changes may have affected concrete performance.

Effects on Carbon

Reduction of NOx is usually accompanied by an increase in carbon content of the fly ash. [2,3,6-9] This carbon is primarily unburned coal and is greatest in the coarser fractions of fly ash, with higher LOI values in the larger size fractions of the ash.[6,10] Documenting the increase in amount and changes in the form of carbon with conversion to low-NOx burners has been the primary focus of most studies. The nature of fly ash carbon with low-NOx burners is coarser, more coke-like, extremely porous, of high surface area, and very active chemically compared with conventional fly ash carbons. Of great concern for use of fly ash in concrete is this change in carbon with low-NOx conditions to a more highly active form that reduces the effectiveness of admixtures, especially air-entraining admixtures (AEA).[7] Concrete workability is negatively affected by the increased amount of unburned carbon in low-NOx fly ash due to porous carbon particles leading to increased water demand. When LOI is reduced through fly ash beneficiation concrete water requirement has been shown to be reduced.[11]

In the Netherlands fly ashes from low-NOx retrofits have higher unburned carbon levels (LOI of 4-15%) compared with pre-conversion ashes (LOI of 2-3%). However, new low-NOx installations employing modified boiler dimensions have been shown to produce fly ashes with LOI values similar to pre-conversion ashes.[8] Where improved coal grinding and modifications to the air : fuel mixture accompany low-NOx retrofits, carbon contents of coarser fly ash fractions may actually be lower than pre-conversion ashes.[9] Although a change in the distribution of coal components is known to occur with increased coal fineness.[9] Other research has shown no relationship between the particle size of coal powder and fly ash LOI.[8]

Effect on Particle Size

Particle size of fly ash has been found to increase when plants are retrofited with low-NOx burners.[3,4,8,9] Lower combustion temperature and a more reducing environment inherent in low-NOx retrofits lead to a somewhat coarser fly ash with a rougher texture due to increased particle agglomeration.[12] The presence of increased amounts of partially fused coal particles in post-conversion fly ashes has also been attributed to lower combustion temperatures.[9] Fly ashes have been shown to be coarser after low-NOx conversion even when the retrofit also included optimization of pulverizers to reduce the particle size of coal feed.[9] Compared with the pre-LNB fly ash, the retrofit fly ashes also have greater porosity and higher specific surfaces. New low-NOx installations with modified boiler dimensions can produce fly ashes with agglomeration and porosity characteristics comparable to that of conventional boilers before conversion.[8] Reduced pozzolanic reactivity of low-NOx fly ash can be due to an increase in the proportion of coarse particles.[3] Although the lower peak flame temperature in low-NOx burners results in a fly ash with fewer particles under 10 microns, the percentage of particles less than 45 or 32 microns is not necessarily changed. A decrease in particles less than 10 microns reduces the pozzolanic activity of the low-NOx fly ash.[4] The decrease in the fraction finer than 10 microns results in unfavorable workability of concrete containing low-NOx fly ash compared with conventional fly ash. A reduction of minus 10 micron particles leads to poorer particle packing and an increase in water demand in concrete.[4]

Effect on Mineralogy

Mineralogical changes in fly ash after conversion to low-NOx burners are not well documented. The X-ray diffraction patterns of fly ash before and after conversion to low-NOx burners at a Tennessee plant burning Appalachian bituminous coal qualitatively appear similar, however, petrography showed the post-conversion ash had a higher volume percent quartz content which was attributed primarily to the lower combustion temperature of the LNB.[9]

The major and minor element composition of fly ash before and after LNB retrofit has not been widely studied. Given the natural variation in coal geochemistry, little difference in trace element composition of pre- and post low-NOx fly ash was found for a plant burning bituminous coal from the eastern US.[9]

Effect on Glass Phase

Changes in glass content of fly ash with conversion to LNB have been predicted but rarely documented.[2,4,5] The glass content of a bituminous coal fly ash from a

Tennessee plant was found to increase after low-NOx burner retrofit coupled with increased coal fineness and modification of the air:fuel ratio. Petrographic glass determinations on fly ashes sampled before and after conversion to LNB show increases in all size categories and much higher glass contents in coarser fractions.[9] Longer residence times as ash particles pass through the boiler flame and into the dust collection systems in low-NOx systems are thought to permit more glass devitrification than with conventional systems.[4]

Effect on Ammonium

Seconday NOx reduction employing SCR and SNCR can lead to the presence of ammonium salts on fly ash in excess of the threshold of 100-200 mg/kg ammonium.[2,8,12] This effect does not occur from low-NOx burner installations.

TESTING PROGRAM

Test Materials

Fly ash samples from a US power plant burning a southern lignite were obtained. Sufficient ash was obtained to perform comprehensive testing of the fly ash before and after the change to low-NOx burners. Five samples were obtained before the change (fly ashes Pre-1, Pre-2, Pre-3, Pre-4, and Pre-5) and five samples after the change (fly ashes Post-1, Post-2, Post-3, Post-4, and Post-5*)*. These samples spanned a period of about 4 months before the burner change and 9 months after the change. During the period of sampling, the local lignite coal source remained unchanged.

For the strength activity index, an ASTM C 150 Type I Portland cement with 3, 7 and 28-day strengths of 3680, 4460, and 5520 psi, respectively, were use. This low alkali cement had a Blaine fineness of 324.4 m^2/kg, chemical composition as shown in Table 1, and Bogue potential composition of 56% C_3S, 17% C_2S, 7% C_3A, and 9% C_4AF.

Table 1. Cement Chemical Composition

Component	Amount, %
CaO	62.54
SiO_2	20.61
Al_2O_3	4.45
Fe_2O_3	3.05
SO_3	2.78
MgO	4.05
Na_2O	0.23
K_2O	0.42
Total Na_2O eq.	0.50
Insoluble Residue	0.29
LOI	1.35

General Characterization

Several fly ash characterization methods were used to identify changes resulting from the burner modifications. Obvious visual differences between samples were evaluated for color using the Munsel color chart. Oven drying to 105 °C was used to determine moisture. Overall changes in rapidly soluble components, such as free line and alkalis, were evaluated by measuring the pH of a solution (containing 1.5 grams of ash in 50 ml of water) one minute after mixing using pH paper test strips.[13] All fly ash samples were evaluated by a calorimetric method for bulk CaO content,[14] here the temperature rise (ΔT in °C) from mixing 20 grams of ash with 75 ml of 15% HCl is correlated with the CaO content. The laboratory is CaO weight percent = $3.35 + 0.2986\Delta T$. Positive evolution of H_2S in these ash-acid mixtures was noted.

Particle Size: The amount of material retained on a 45µm sieve was determined according to standard procedures.[15] The material retained was examined microscopically.

Chemical Analysis: Each fly ash was chemically analyzed by X-ray fluorescence. Unburned carbon contents were determined by loss on ignition.[15]

Characterization of Glass in the Fly Ash: The glass phase present in the fly ash samples was characterized by X-ray diffraction using a PANalytical X'Pert PRO Instrument. A 4% Si internal standard was used to quantitatively determine the crystalline phases.[17] The weight percent of glass was determined by subtraction of crystalline phases.[17,18] The position of the glass diffraction maxima in the X-ray patterns was also determined as an index for glass compositional changes. In general, shifts to smaller d-spacing have been correlated to increased content of CaO in the glass phase and a more reactive glass.

Properties Affecting Performance in Concrete

Two main properties of all fly ash samples were investigated that are significant for utilization in concrete, the affinity for air entraining admixtures (foam index) and the ability to react as a pozzolan (strength activity index).

Affinity for Air-Entraining Admixtures: A modified foam index procedure was done on the samples to determine changes to the affinity of the carbon for air-entraining admixtures with conversion to low-NOx.[7] In this test a gram of ash and a gram of cement are mixed with 10 ml of water and a 0.1% sodium lauryl sulfate (SLS) solution is added incrementally until a stable foam is formed; at each increment the vial is systematically shaken to generate foam. The end point of the test is the weight of SLS solution required for 1 ml of foam to remain stable for one minute multiplied by 1.75.

Strength Activity Index: Mortar cubes were made with the fly ashes according to standard procedures.[15] In this procedure, the flow of mortar mixtures containing the a 20% fly ash replacement level is maintained within 5 units of the reference mixture (containing only cement) by adjusting the amount of water in the mixture. Three 2in x 2in (5cm x 5cm) cubes were broken at 3, 7, 28, and 56 days.[16] Mixing was carried in two days using the same cement; a reference mixture was run on each day.

TEST RESULTS

General Characterization

General characterization results for all the fly ash samples are shown in Table 2. In addition to these results, all samples contain less than 0.1 % moisture. With the exception of pH and minor differences in calculated CaO, all the categories in the Table 2 showed differences. As the result of the burner change, the fly ash became darker, there was an increase in LOI, Foam Index, and particle size. The presence of sulfide was identified in all the post-ash samples and none of the pre-ash samples.

As is typically experienced, the Low-NOx fly ashes in this study have more carbon and it is more active, compared with the pre low-NOx fly ashes. The low-NOx fly ash samples are darker in color and have an average LOI of 0.52%, compared with an average LOI of 0.43 for the pre low-NOx fly ashes.

Table 2. Fly Ash General Characterization Results

Ash	Color	pH	LOI, weight %	Cal. CaO, weight %	H_2S evolution in HCl	Particle size >45µm, wt. %
Pre-1	Light Gray	11.0	0.35	12.0	Negative	20.8
Pre-2	Light Gray	11.5	0.36	11.4	Negative	26.4
Pre-3	Light Gray	11.0	0.43	13.5	Negative	25.2
Pre-4	Light Gray	11.0	0.50	9.6	Negative	28.7
Pre-5	Light Gray	11.5	0.90	13.5	Negative	22.0
Average pre- ash		11.2	0.43	12.0		24.6
Post-1	gray	11.0	0.55	14.0	Positive	23.4
Post-2	gray	11.0	0.50	12.6	Positive	30.3
Post-3	gray	11.0	0.57	12.9	Positive	30.2
Post-4	gray	11.0	0.69	14.5	Positive	26.6
Post-5	gray	10.5	0.28	12.5	Positive	30.6
Average post- ash		10.9	0.52	13.3		28.2

The content of coarse particles in the fly ash samples increased after the change to low-NOx burners as shown in the last column of Table 2. The 45µm retain averages approximately 4% greater in the low-NOx ashes. Microscopic examination of the material retained in the sieve shows an increase in char and coke particles, angular quartz grains, and agglomerated particles in the low-NOx fly ash samples compared with the samples before the burner change.

Chemical Analysis

No significant differences in fly ash chemical composition between the pre-ash and post-ash samples were found as shown in Table 3 and Figure 1. In this figure, the bars indicate average values of the set and error bars the range in composition. A small increase in sulfate content in the post-ash samples is shown in Figure 1, but part of these sulfur ions are present as sulfide given the evolution of H_2S identified in this ash group (Table 2). Given that the lignite source used was the same before and after the burner change, these results indicate that the burner modification had no substantial effect on the overall chemistry of the ash. A similar result was reported for an Eastern Bituminous coal fly ash studied before and after the change to low-NOx burners.[9] As expected from the similarity in major element chemistry, little change is found in pH and CaO calorimetry after burner change.

Table 3. Chemical Composition of Fly Ashes (weight percent).

Ash	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O
Pre-1	49.93	24.57	3.85	13.76	2.38	0.57	0.22	0.78
Pre-2	50.37	24.47	4.54	13.58	2.26	0.59	0.20	0.85
Pre-3	48.42	24.13	4.09	15.14	2.37	0.57	0.18	0.73
Pre-4	52.76	24.70	4.59	11.10	1.99	0.48	0.14	0.91
Pre-5	47.62	24.21	4.19	15.40	2.51	0.63	0.21	0.70
Average pre- ash	49.82	24.42	4.25	13.80	2.30	0.57	0.19	0.79
Post-1	51.24	24.09	3.45	13.39	2.23	0.64	0.18	0.83
Post-2	49.42	23.58	4.72	14.30	2.25	0.70	0.21	0.78
Post-3	49.49	23.97	4.51	14.13	2.32	0.63	0.19	0.67
Post-4	50.18	24.30	4.14	13.91	2.33	0.62	0.19	0.71
Post-5	50.13	22.98	5.15	14.18	2.32	0.66	0.24	0.75
Average post- ash	50.09	23.78	4.39	13.98	2.29	0.65	0.20	0.75

Mineralogy

Mineralogy of the samples determined by XRD is shown in Table 5. The major changes from the pre-ash to the post-ash observed in this table are a reduction in glass and magnetite, and an increase in both quartz and anorthite. Of these, the increase in anorthite is the only change that shows no overlap between the groups. The position of the glass diffraction maxima was found to be similar for

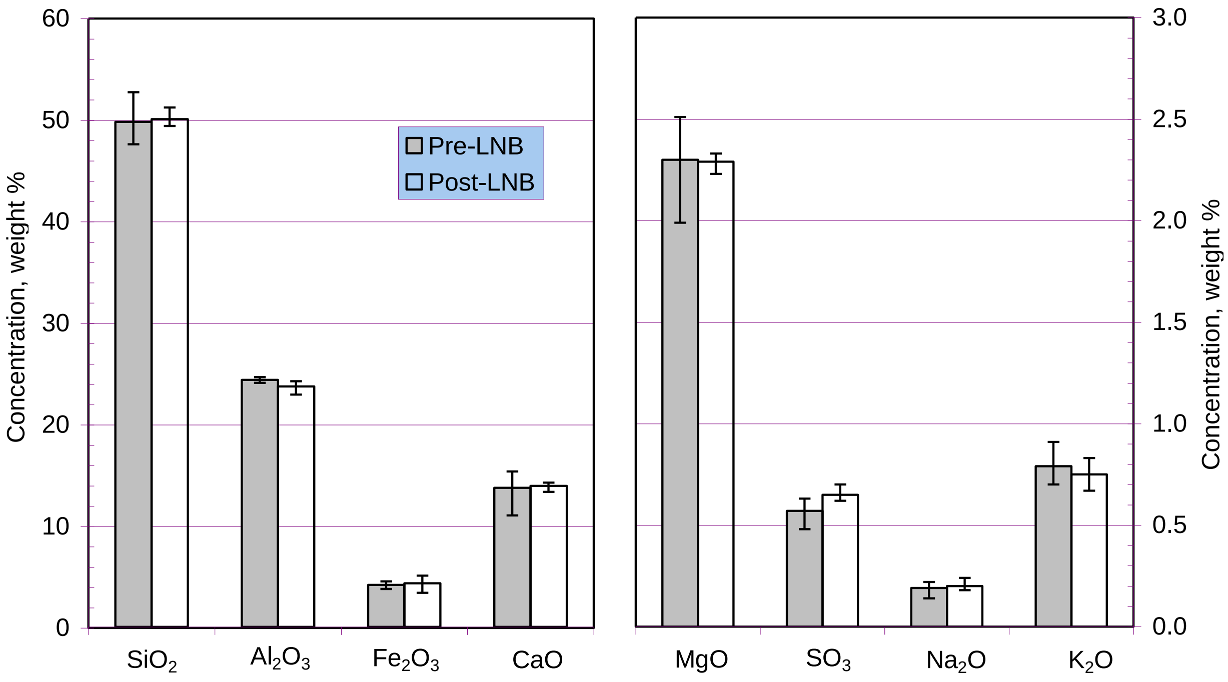

Figure 1. Elemental composition expressed as oxides of pre- ash and post- ash.

Table 4. Fly Ash Mineralogy Based on XRD, weight %

Ash	Glass	Mullite	Quartz	Calcite	Anhydrite.	Magnetite	Anorthite	Lime	Hematite	Portlandite
Pre-1	81.4	8.6	6.0	0.0	0.7	1.5	0.6	0.4	0.3	0.5
Pre-2	80.6	8.8	7.5	0.1	0.1	1.0	0.7	0.3	0.6	0.3
Pre-3	84.7	7.1	5.3	0.0	0.1	0.8	0.6	0.3	0.4	0.7
Pre-4	75.8	13.2	8.6	0.0	0.1	0.7	0.4	0.3	0.7	0.2
Pre-5	80.8	9.5	5.9	0.0	0.2	0.8	0.8	0.4	0.8	0.8
Average pre- ash	80.7	9.4	6.7	0.0	0.2	1.0	0.6	0.3	0.6	0.5
Post-1	79.5	10.2	7.9	0.1	0.2	0.3	0.5	0.3	0.7	0.4
Post-2	76.8	10.1	8.9	0.1	0.2	0.6	1.7	0.4	1.1	0.1
Post-3	77.1	10.1	8.2	0.1	0.2	0.9	1.7	0.3	1.0	0.4
Post-4	80.8	8.4	7.3	0.0	0.2	0.6	1.4	0.2	0.6	0.5
Post-5	77.8	8.8	8.8	0.0	0.2	0.8	2.3	0.3	0.7	0.3
Average post- ash	78.4	9.5	8.2	0.1	0.2	0.6	1.5	0.3	0.8	0.3

the pre-ash group and the post-ash group, varying from 26.1-26.5 °2θ with average of 26.3°2θ and 26.3-26.7°2θ with average of 26.4°2θ, respectively

Affinity to Air-Entraining Admixtures

Table 5 indicates that the foam index is significantly greater for the low-NOx fly ash. Although the increase in carbon content in fly ashes after the change to low-NOx burners is not dramatic (Table 2), the foam index is significantly greater for the low-NOx fly ash, indicating a more active form of carbon compared with fly ash from conventional burners. The carbon in the low-NOx fly ashes is seen microscopically to be open, porous, large particles with a surface resembling coal char and coke.

Table 5. Foam Index Results.

Ash	Foam index, ml
Pre-1	0.12
Pre-2	0.19
Pre-3	0.13
Pre-4	0.12
Pre-5	0.10
Average pre- ash	0.13
Post-1	0.15
Post-2	0.15
Post-3	0.25
Post-4	0.22
Post-5	0.21
Average post- ash	0.20

Strength Activity Index

The compressive strength data and calculated SAI values are shown in Table 3. Mortar cubes made with varying w/cm to maintain consistent flow display a reduction in compressive strength. Differences in amount and type of carbon and possibly in particle size and morphology led to an average w/cm of 0.46 for the pre-low-NOx cube mixes and 0.48 for the post low-NOx cube mixes.

Table 6. Compressive Strength and Strength Activity Index (SAI) of Cubes

Ash	w/cm	Flow %	3 day Strength Mpa	3 day SAI, %	7-day Strength Mpa	7-day SAI, %	28-day Strength Mpa	28-day SAI, %	56-day Strength Mpa	56-day SAI, %
Pre-1*	.46	92	17.7	77.3	22.3	82.0	32.5	96.7	38.5	110.9
Pre-2#	.47	99	15.4	69.6	19.4	70.4	28.3	83.7	33.5	96.6
Pre-3#	.47	101	15.7	70.5	19.9	72.2	31.7	93.7	34.4	99.2
Pre-4#	.46	97	18.8	84.8	23.1	84.0	31.8	93.9	37.9	109.1
Pre-5*	.46	100	18.7	81.9	23.0	84.3	31.0	92.4	37.4	108.0
Average pre- ash	.46	98	17.3	76.8	22.0	78.6	31.1	92.1	36.3	104.8
Post-1*	.48	94	16.4	71.9	20.8	76.2	29.9	89.1	35.4	102.0
Post-2#	.48	99	13.9	62.4	17.7	64.2	26.4	78.0	29.0	83.5
Post-3#	.48	93	15.1	68.0	17.8	64.7	28.8	84.9	31.9	92.0
Post-4#	.48	101	15.1	68.0	19.7	72.2	28.1	83.4	32.0	92.2
Post-5*	.48	93	15.5	77.0	20.9	76.7	27.9	83.0	35.4	102.2
Average post- ash	.48	96	15.2	69.5	19.35	70.8	28.2	83.7	32.7	94.4
Reference mixtures										
*	.48		22.8		27.2		33.6		34.7	
#	.48		22.2		27.5		33.9		34.7	

DISCUSSION OF RESULTS

Burner Effects on Fly Ash

As stated previously, the low-NOx fly ash samples have slightly more quartz and plagioclase feldspar (anorthite) and less glass compared with the pre low-NOx fly ashes. The CaO content of these lignite ashes is intermediate between that typically seen in a Powder River Basin Class C fly ash and a Class F Bituminous coal ash from the Illinois Basin and the Eastern coal basins. Intermediate fly ashes thermally relax relatively quickly at temperatures as low as 1000°C causing the more lime-rich glass particles to crystallize anorthite and clinopyroxene. In this particular case, it is likely that slightly more quartz remained unmelted in the somewhat lower peak flame temperature of low-NOx burner. Also, the longer residence time of fly ash particles in the low-NOx burner resulted in a small but significant anorthite increases at the expense of glass. But the overall properties of the glass phase as indicated by the position of the glass peak did not appear to be significantly affected by the minor increase in anorthite and quartz.

In addition, the low-NOx fly ashes produced a H_2S odor when acidized in the CaO calorimetry testing that was not apparent with the pre low-NOx samples, indicating the presence of a small amount of sulfide, perhaps oldhamite. The amount of sulfide is too small to resolve by XRD.

Potential Burner Effects on Concrete Performance

Darkening of the ash, slightly higher LOI, and significantly higher Foam Index in the ash after the low-NOx burner installation strongly suggest that the carbon is significantly more active in relation to air-entraining admixtures. Differences in carbon may have contributed to the higher w/cm in the low-NOx mixtures.

Figure 2 shows that a clear reduction in activity index of approximately 10% is observed at all ages tested for the ash obtained after the low-NOx burners were installed. Differences in amount and type of carbon, particle size and morphology led to an average w/cm of 0.46 for the pre-low-NOx mixtures and 0.48 for the post low-NOx mixtures. The post-ash did not afford any water reduction to the mortar mixtures, but the w/c was the same as the as the reference mixture without ash. A loss of approximately 10% compressive strength was reported for concrete batched with this lignite fly ash after the

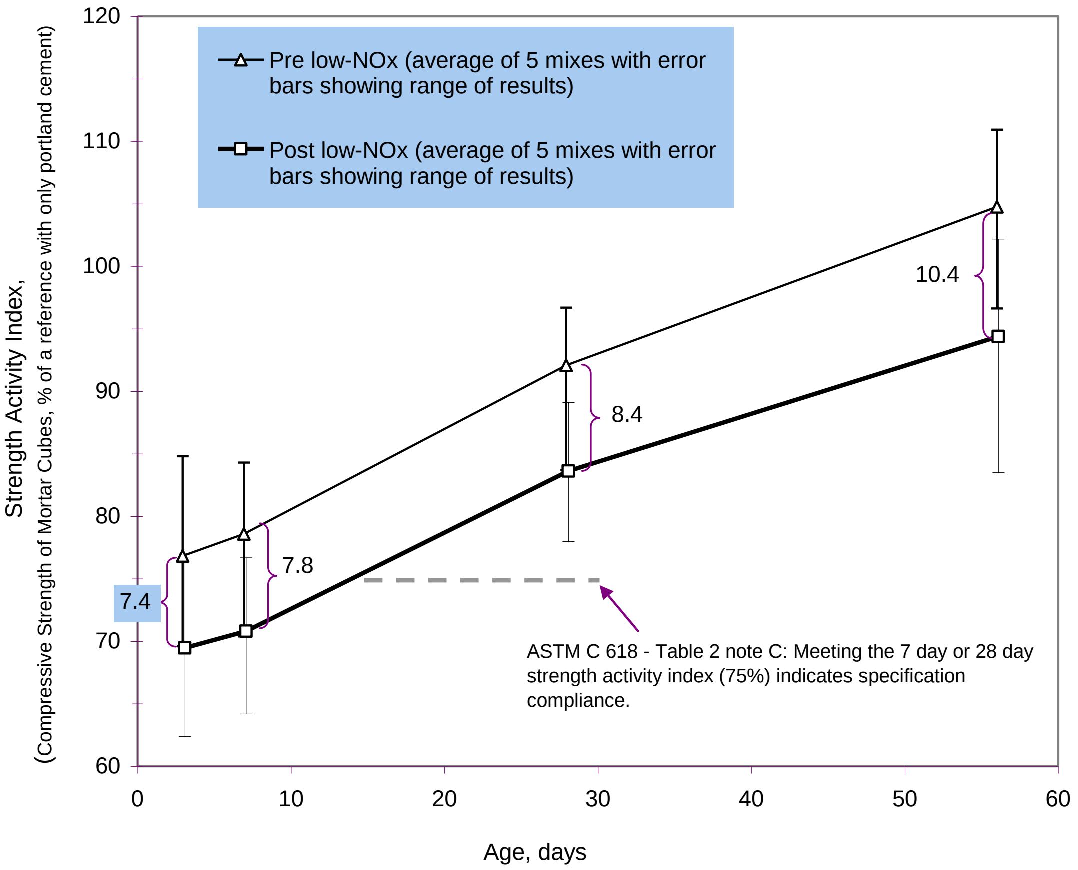

Figure 2. Average Strength Activity Index (SAI) values over time.

change to low-NOx burners. The results of this investigation indicate that this strength loss is primarily due to an increase in water demand in concrete produced with the fly ash after the change to low-NOx burners. This is due to a change in amount and type of carbon present in the fly ash.

The lower strength obtained in the field were overcome by increasing the amount of Water-reducing admixtures to reduce the w/cm and return concrete strengths to pre low-NOx levels.

CONCLUSIONS

The following conclusions can be made based on the study of fly ash obtained from one coal-burning plant undergoing installation of low-NOx burners (LNB).

- SAI values for low-NOx fly ash were consistently 10% lower than the corresponding values for the fly ash before the burner change.
- Fly ash produced after LNB went online have slightly greater and more active carbon contents, more coarse particles, and slightly less glass which can explain the reduction in SAI observed.
- Chemical admixtures have been used in concrete containing this low-NOx fly ash to restore strengths to the level prior to LNB installation.
- No significant change in bulk chemical composition was found between samples taken before and after LNB installation.
- Fly ash produced after LNB has significantly higher affinity for air-entraining admixtures from the increased activity of carbon.
- Slight decreased in glass content in the LNB ash, results from less quartz melting (lower flame temperature) and devitrification of the glass phase to form feldspar (longer residence time).

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