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Comparison of fly ash properties from Afsin–Elbistan coal basin, Turkey

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Abstract

Afsin–Elbistan (AE) coal fly ashes obtained by burning coal samples from top, middle and bottom sections of the AE coal seam were characterized and their properties were compared. Chemical analysis of the AE coal fly ashes showed that they are mainly composed of CaO, SiO₂, Fe₂O₃ and Al₂O₃. Quantitative X-ray diffraction (XRD) analyses were carried out using an interactive data processing system (SIROQUANTTM) based on Rietveld interpretation methods. Lime is found in all the samples, ranging from around 7% to just over 38%. Amorphous contents of fly ashes are ranged between 19% and 25%. Different types of AE fly ashes revealed that bottom section coal fly ash is very similar to Class F, while medium and top section coal fly ashes are close to Class C and they might be used as mineral admixture in concrete. But also they do not comply with any of the standard. The results presented here show new possibilities for AE coal fly ashes in a wide range of fields, resulting in great advantages in waste minimization, as well as, resources conservation. © 2004 Elsevier B.V. All rights reserved.

Keywords: Afsin-Elbistan A power plant; Coal fly ash; Mineral composition; Chemical properties; Quantitative XRD analyses; Waste minimization

1. Introduction

The Afsin–Elbistan (AE) basin is a multi-layer coal deposit with associated clays and calcareous gyttja partings. The basal part of the coal seam consists of clay, marl and sandy clay. These formations are overlain by gyttja enveloping coal seam. The term gyttja refers to a formation that is of sapropelic, black or brown mud with organic matter and have many gastropod shells. The AE coal basin is divided as upper seam, where the gyttja formation occurs as a sterile intercalation and lower seam, where clay occurs instead of the gyttja [1]. These intercalations strictly affect mineral matter contents of the AE coals. Generally, the most of mineral matter in coal ends up as a fly ash. Physical properties and chemical composition of a fly ash depends on the composition of mineral matter of the coal from which the ash is derived. Coal production is currently exploited in Kislakoy

opencast mine and supply feed coal to AE A power plant, which has four boiler units each with 340 MW capacity. The AE A power plant consumes 18.0×10^6 metric tonnes of coal per year and generates about 3.24×10^6 metric tonnes of fly ashes. Then, these ashes return to the dumping area of the mine as combustion waste.

The management of fly ash produced by coal fired power plants remains a major problem in many parts of the world. Although, significant quantities are being used in a range of applications and particularly as a substitute for cement in concrete, large amounts are not used and this requires disposal. This is not an ideal situation as there are concerns that this may cause long-term adverse environmental effects. Fly ash consists of fine particles that contain leachable heavy metals and is therefore, classified as a toxic waste [2,3].

Research is needed to develop new alternative applications that can further exploit coal fly ash, which needs to be increasingly regarded as a raw material, with potential for processing into new products rather than a waste. Potential applications for fly ash are grouped into following

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main categories: construction materials (cement production, concrete, ceramics, glass and glass–ceramics), geotechnical applications (road pavement and embankments), agriculture and miscellaneous (sorbent and sludge conditioning) [3]. A large amount of information is available on the application of coal fly ash [4–10]. Bayat [11,12] studied the AE fly ashes, for their ability to remove nickel [Ni(II)], copper [Cu(II)], zinc [Zn(II)], Cr(VI) and Cd(II) from an aqueous solution. They were found to be a metal adsorbent, as effective as, activated carbon. Yasar et al. [13] studied the strength properties of lightweight concrete made with the basaltic pumice and the AE fly ash.

The aim of this work is to investigate the physical, chemical and mineralogical properties of the AE coal fly ashes obtained by burning coal samples from various sections in the coal seam and to compare their properties.

2. Experimental

Chemical compositions of the AE fly ashes change at the vertical direction of the coal seam [14]. For that reason, the coals were produced from the top, middle and bottom sections of the coal seam and they were burnt up to 1100 °C at the boiler of the AE A power plant, separately. The representative samples were taken from the top section, middle section and bottom sections of coal fly ashes. These samples were then named as L1, L4, M11, M21, H32 and H36. L1 and L4 for the bottom section coal fly ash and H32 and H36 for the top section coal.

Elemental analyses of these samples were carried out by X-ray fluorescence (XRF) spectrometry technique. Knowledge of mineralogy is a key component of any investigation into why a fly ash behaves in a certain way in its utilization and disposal. Quantitative X-ray diffraction (XRD) analyses were carried out using an interactive data processing system (SIROQUANTTM)¹ based on Rietveld interpretation methods [15]. Diffractograms of the samples were obtained using a Philips diffractometer system with Cu Ka radiation. Samples were run from 5° to 65° 2θ with a step of increment of 0.02° and counting time of 2 s/step. Study of the particle size, specific surface area, bulk density, specific gravity and pH development of ash is interesting from the point of view of characterization of the fly ash itself. Fineness of fly ash affects the performance of concrete and grouts, when it is used as a replacement material. It was noted by Sear [16] that an increase in the percentage retained on the 45 µm sieve results in a decrease in the strength of the concrete. Particle size measurement of fly ash sample was carried out by sieving technique. A pycnometer was used to determine the specific gravities of fly ash samples. The specific surface area of the ashes was found by the laser beam method. Development of pH in suspensions of each fly ash in water was monitored by a universal glass pH probe and pH meter. Fly ash samples of weigh 2 g were suspended in 100 ml of deionized water by continuous stirring. The immediate pH of the solution and pH values each hour, thereafter for a period of 24 h, was recorded. The water-soluble contents of the AE fly ashes are examined according to ASTM C 593 [17].

3. Results and discussion

Chemical compositions of the fly ash samples are presented in Table 1. They are mainly composed of CaO, SiO_2 , Fe_2O_3 and Al_2O_3 , accounting for 74–85% of the material on a dry weight basis.

The results of the quantitative XRD analysis (Table 2) represent the final output from each task, when the best possible fit had been achieved between the observed and calculated XRD patterns (Fig. 1). Table 2 also shows the error associated with each individual component from the estimated standard deviation (e.s.d.). The error is expressed in each case as an absolute percentage with respect to the last digit; thus, a determination of 12.5 (4)% refers to 12.5% with an error of $\pm 0.4\%$. An estimate of the overall goodness of fit of each analysis is also provided, expressed as the relevant global χ^2 value derived as indicated by Taylor [18]. This value should approach 1.0 for a perfect fit between the measured and interpreted patterns. Hematite, quartz and anhydrite are the major crystalline phases in the bottom section coal fly ash sample. Lime and anhydrite are the major crystalline phases in the middle section and the top section coal fly ash samples. Gehlenite and melilite are present in significant proportions in the bottom section and the middle section coal fly ash samples. C3A, C4AF and BC2S cement minerals are present in small proportion in all the samples.

Fig. 2 presents the range of the particle size distribution of the AE fly ash samples. It indicates that the fly ash contains important proportion of (20-72 dry wt.%) clay and silt size ($<75 \,\mu$ m). Generally, there is a relationship between the percentage retained on the 45 μ m sieve and the coal type or ash source. Fig. 2 shows the large variation between the ashes from the small amount of coarse particles found in ash H36, to the ash L1 where coarse particles comprise about 85% percent of the ash. The specific surface areas of the fly ash samples are given in Table 3. As also seen from Table 3, the bulk density of the fly ash samples ranged from 0.63 to 1.36 g/cm^3 , and specific gravity ranged from 2.80 to 3.27 g/cm³. In addition, it is evident that both bulk density and specific gravity increased as the particle size of the fly ash decreased. The low bulk density makes these fly ashes a good material for lightweight building material. When used as a partial replacement for cement, the effects of coal fly ash on the pH of the mixing water, may be masked by the rapid rise in pH resulting from the reaction of the cement grains. Data on the development of pH with time of solutions of ashes in water are presented in Fig. 3. Ash samples except L4 are highly alkaline at around 12.5. In the cases of fly ashes M21, H36 and H32, the pH of

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Table 1	
Chemical composition of the AE coal fly ash s	amples

Element/element oxide (dry wt.%)	Bottom section coal fly ash	Middle section coal fly ash	Top section coal fly ash	Number of samples
Al ₂ O ₃	12.370-23.100	6.230-11.400	1.800-9.100	24
CaO	6.200-20.600	22.510-50.850	48.100-74.700	24
Cl	0.006-0.008	0.003-0.007	0.006-0.013	6
Cr ₂ O ₃	0.053-0.070	0.092-0.111	0.061-0.074	6
Fe ₂ O ₃	9.550-25.790	3.500-6.850	1.750-4.960	24
K ₂ O	0.240-0.820	0.200-0.490	0.110-0.555	24
MgO	2.190-4.200	1.570-4.000	1.270-3.100	24
MnO ₂	0.060-1.100	0.120-0.240	0.020-0.090	24
MoO ₃	0.013-0.027	_	_	6
Na ₂ O	0.200-0.830	0.104-0.490	0.100-0.250	24
NiO	0.035-0.042	_	_	6
P ₂ O ₅	0.250-0.413	0.460-0.494	0.531-0.620	6
SO ₃	5.750-17.210	12.320-24.200	6.600-19.000	24
SiO ₂	27.700-46.600	14.000-30.000	6.918-20.400	24
SrO	0.052-0.071	0.056-0.064	0.037-0.059	6
TiO ₂	0.260-2.332	0.200-0.515	0.230-0.550	24
V ₂ O ₅	0.000-0.095	0.109-0.117	0.066-0.087	6
ZrO ₂	0.131-0.186	_	_	6
CO ₂	0.300-0.350	0.600-0.750	1.200-1.900	6
LOI	1.290–1.340	0.640-3.560	2.460-4.940	6

Table 2

Mineralogy of the coal fly ash from the AE power plant (wt.%)

Mineral	Bottom section coal fly ash	Middle section coal fly ash	Top section coal fly ash
Anhydrite (CaSO ₄)	12.3 (3)	18.6 (3)	23.5 (4)
Lime (CaO)	7.2 (3)	24.3 (3)	38.2 (3)
Quartz (SiO ₂)	13.2 (3)	3.8 (3)	1.9 (4)
C3A (orthogonal) (3CaO Al ₂ O ₃ + Na ₂ O)	0.3 (3)	0.5 (2)	2.7 (4)
C3A (cubic) (Ca ₃ Al ₂ O ₆)	1.4 (4)	2.7 (3)	3.8 (4)
Calcite (CaCO ₃)	1.8 (4)	1.3 (2)	2.5 (3)
Portlandite (Ca (OH) ₂)	2.9 (4)	1.4 (2)	2.1 (3)
Spinel (MgAl ₂ O ₄)	0.5 (3)	-	-
Hematite (Fe ₂ O ₃)	16.6 (4)	1.1 (3)	-
C4AF (Ca ₂ Fe 1.28Al 1.72O ₅)	1.0 (4)	2.9 (4)	4.2 (4)
Gehlenite (Ca ₂ Al (Si, Al) ₂ O ₇)	8.1 (3)	11.1 (4)	-
Melilite [(Ca, Na) ₂ (Mg, Al)(Si, Al) ₃ O ₇]	6.8 (4)	6.2 (4)	-
$\beta C2S (Ca_2 SiO_4)$	2.7 (3)	3.6 (4)	1.6 (4)
Amorphous content	25.1 (3)	22.4 (4)	19.3 (4)
Global χ^2	1.7	1.6	1.6

the solution reached a peak value of 12.5, 12.8 and 13.0 at 6 h, respectively, and then remained at the same level for the period of 24 h. In the case of fly ash L4, the pH value reached a peak value of 11.2 at 0.5 h and decreased 9.6 at 4 h and then remained almost constant at approximately 9.5.

The ternary composition plot for the AE coal fly ashes based on the content of the SiO_2 , CaO and Al_2O_3 is depicted

Table 3 Specific surface areas and density distribution of the samples

1	2		1
Sample	Specific surface area (cm ² /g)	Bulk density (g/cm ³)	Specific gravity (g/cm ³)
L1	1560	1.34	3.16
L4	2550	1.36	3.27
M11	2040	1.10	2.97
M21	2160	1.07	3.03
H32	2820	0.81	2.84
H36	3020	0.63	2.80

in Fig. 4. It shows the typical composition range for ordinary portland cement (OPC) and coal fly ash based on their typical Si, Al and Ca oxides content [19]. It is apparent that the bottom section coal fly ash sample positions in the ternary diagram have pozzolanic and also cementations properties. However, for the pozzolanic properties to be exhibited, silica must be present as amorphous glass SiO₂, which accounts for its chemical reactivity. The absence of significant amorphous material was revealed for the AE fly ashes by inspection of XRD patterns. Chemical composition of OPC [20] and the AE fly ash samples compared in Table 4. Chemical composition of the middle section and the top section coal fly ash samples are close to that typical of OPC. But mineralogical compositions of the AE fly ashes are not similar to OPC [21] (Table 5).

Fly ashes classified as high calcium and low calcium fly ashes according to its CaO content. If the CaO content is less than 10% then the ash classified as low calcium or Class F. If



Fig. 1. Comparison of observed XRD trace (top), calculated trace (middle) and difference between them (bottom) for bottom section coal fly ash.



Fig. 2. Particle size distribution of the AE coal fly ash samples.

 Table 4

 Chemical composition of OPC and the AE fly ashes

Element oxide (dry wt.%)	OPC	L1	L4	M11	M21	H32	H36
Al ₂ O ₃	5.60	19.80	12.37	9.55	6.23	7.27	4.31
CaO	61.87	10.10	15.95	35.40	45.673	55.60	64.15
Fe ₂ O ₃	4.13	9.55	25.79	4.10	3.70	3.47	3.92
K ₂ O	0.83	0.45	0.43	0.20	0.47	0.35	0.56
MgO	2.60	2.80	3.32	3.65	1.57	1.80	1.27
Na ₂ O	0.14	0.83	0.63	0.20	0.10	0.25	0.17
SO ₃	2.79	7.55	11.22	19.30	22.72	10.65	17.51
SiO ₂	20.65	45.80	27.67	26.35	18.25	17.24	6.92
LOI	0.50	1.29	1.34	3.56	0.64	4.94	2.46



Fig. 3. pH development of the AE coal fly ash samples.

the CaO content is more than 10%, then the ash is classified as high calcium or Class C. ASTM C 618-97 defines the chemical and physical requirements of coal fly ash and raw or calcined natural pozzolan for use as a mineral admixture in concrete [22]. Different types of the AE fly ashes revealed that bottom section coal AE fly ash is very similar to Class F, while middle section and top section coal fly ashes are close to Class C, as defined in the ASTM C 618-97 [22] standard (Table 6). But also that each type fly ash failed to meet one or more criteria of the standard. All the types of AE fly ash have high SO₃ content and coarse particle size, while middle section and top section coal fly ashes are below the Class C requirement for (Si + Al + Fe) oxides. Concrete structures can suffer from alkali-aggregate reaction. It is also seen from Table 6 that the total alkali content of Na2Oeq of all the types AE ash is made up of both Na₂O and K₂O and comply with ASTM C 618-97 [22] standard. It was shown that a nonstandard fly ash can also be used in concrete, for example Atis et al. [23] studied the effect on compressive strength of replacing cement with the AE coal fly ash. It is reported that, the AE coal fly ash might be used in concrete between 15 and 30% as cement replacement.

The most effective way to improve the properties of the vitrified products without major alterations to the process itself, is the induction of a controlled crystallization, i.e. by forming a glass–ceramic [24]. Erol et al. [25] studied the characterization of glass–ceramics developed from Seyitomer/Turkey thermal power plant fly ash and reported that the glass–ceramic samples produced from Seyitomer/Turkey fly ash have several desirable properties that would make them attractive to industrial use in construction, tiling and cladding



Fig. 4. Ternary composition grid for the AE coal fly ash samples as compared with OPC.

Table 5

Comparisons of mineral content of OPC with the AE fly ash (wt.%)

	C3S	C2S	C3A	C4AF	CaSO ₄	Free CaO
OPC [20]	49 (42–67)	25 (8-31)	12 (5–14)	8 (6–12)	2.9 (2.6–3.4)	0.8 (0-1.5)
Bottom section coal fly ash	_	2.7	1.7	1.0	14.3	16.2
Middle section coal fly ash	_	3.6	3.2	2.9	18.6	24.3
Top section coal fly ash	-	1.6	6.5	4.2	23.5	38.2

Table 6

	ASTM [21] Class F	ASTM [21] Class C	L1	L4	M11	M21	H32	H36
$SiO_2 + Al_2O_3 + Fe_2O_3 (min, \%)$	70.0	50.0	75.15	65.83	30.45	28.18	27.98	15.14
SO ₃ (max, %)	5.0	5.0	7.55	11.22	19.30	22.72	10.65	17.51
Moisture content (max, %)	3.0	3.0	0.3	0.3	0.3	0.4	0.3	0.4
Available alkalies, as equivalent, as Na2O (max, %)	1.5	1.5	1.13	0.91	0.33	0.41	0.48	0.53
Loss on ignition (max, %)	12.0	6.0	0.50	1.29	1.34	3.56	0.64	4.94
Amount retained when wet-sieved on 45 µm sieve (max. %)	34	34	84	66	78	50	75	33

Comparison of chemical composition and some physical properties of the AE fly ashes with to limits of ASTM C 618-97 standards of fly ash

applications. Especially, chemical and mineralogical composition of the AE bottom section coal fly ash is very similar to Seyitomer fly ash.

The alteration of soil properties to meet specific engineering requirements is known as soil stabilization [26]. Principal applications of chemical soil stabilization are in highway and airfield engineering. Some additives such as Portland cement, lime and fly ash can be used for improving the engineering properties of soil. Stabilization of coarse-grained soils having little or no tines can often be accomplished by the use of lime-fly ash or lime-fly ash-cement combinations [27]. Fly ash contains silicon and aluminum compounds that, when mixed with lime and water, forms a hardened cementitious mass capable of obtaining high compressive strengths. Lime and fly ash in combination, can often be used successfully in stabilizing granular materials since the fly ash provides an agent, with which the lime can react. Thus, lime-fly ash or lime-fly ash-cement stabilization is often appropriate for base and sub-base coarse materials [27]. Class C fly ash is self-reactive or cementitious in the presence of water, in addition to being pozzolanic because it contains a high percentage of lime. Class F fly ash requires the addition of lime to form a pozzolanic reactions. The AE fly ashes might be used in soil stabilization, as a substitute of lime or cement due to their pozzolanic characteristics. Quality requirements of fly ash used for stabilization are indicated in ASTM C 593 [17]. Different types of the AE fly ashes revealed that water-soluble fraction and fineness properties of the top section coal fly ash meets the requirements indicated in ASTM C 593 (Table 7). However, lime-pozzolan strength should be investigated according to ASTM C 593. The protection of open pit slopes against water and earth pressures, frequently calls for grouting. Grouts are important to improve strength and stability, therefore, they must penetrate deeply and seal effectively against the ingress of bulk water. Ingles et al. [26] reported that cement and lime are the most common grouts.

Ural and Yuksel [28] indicated that slope instability is a continuing problem in the open pit mines located at the AE coal basin. The AE fly ashes might be used as a substitute of cement or lime replacement for grouting. Bulk density of the AE fly ashes is less than other fill materials used in construction of embankments. Due to bulk density values of the AE fly ash are between 0.63 and 1.34, there is an advantage in replacing conventional fill materials with fly ash, since smaller loads would be imposed on soils thus resulting in less significant ground settlement.

Use of the AE fly ashes as a soil amendment has the potential as an increasing outlet for the AE fly ashes, if the public can be shown through appropriate research that the AE fly ashes can improve certain types of soils without significant damage to the environment. Several studies have shown that attributes of some types of fly ash can improve plant growth and crop yield. However, high concentrations of Al, B, Mn or soluble salts, as well as deficient levels of P and K, may cause problem [29,30]. The sulfur (SO₃) content is found to be high for all the AE fly ash samples. Bradsley and Lancaster [31] stated that virtually all of the sulfur in fly ash is in soluble form, as revealed by NH₄OAc analysis. The importance of this finding in terms of soil and vegetation enrichment of fly ash-derived sulfur is very important for the AE fly ashes. Since most of Turkish soils are mainly calcareous and alkaline, iron chlorosis is a very common and serious problem. In practice, gypsum and elemental sulfur are used for fertilization to prevent iron chlorosis. Use of fly ash could be more economical. Phung et al. [32] indicated that addition of fly ash to soil tends to increase soil pH, soil salinity, soluble calcium, magnesium, sodium, sulfur and boron, and decrease the concentration of water-soluble phosphorus. Since most of Turkish soils are calcareous, their high buffering capacity will resist changes in pH induced by fly ash additions. The increase in pH of acid soil suggests that, fly ash could be used as a limiting agent to raise the low pH of acid soils in humid

Table 7

Comparison of some physical properties of the AE fly ashes with to limits of ASTM C 593-89

	ASTM C 593-89 [16]	L1	L4	M11	M21	H32	H36
Water-soluble fraction (max, %)	10.0	4.1	4.6	6.2	4.8	1.1	1.9
Fineness, amount retained, when wet-side	eved:						
No. 30 (600 µm) sieve (max, %)	2.0	10.0	5.0	3.0	3.0	1.4	1.2
No. 200 (75 µm) sieve (max, %)	30.0	72.0	44.0	69.0	32.0	66.0	20.0

regions such as the Black Sea coast of Turkey. On the other hand, excessive use of the AE high-calcium fly ash in soil could possibly impede root development by creating hard areas near the soil-fly ash interface in a closed ash disposal area.

Leaching of fly ash to recover minerals to produce other useful products has been investigated and reported in detail in the literature [33–37]. Physical properties and chemical compositions of the AE fly ashes show that the AE bottom section fly ash may have a potential for iron gaining. However, the absolute values from economic analyses must be considered only in connection with the basis of the analyses (value and marketability of the product, costs of the facility, etc.).

4. Conclusions

Physical properties and chemical compositions of the AE coal fly ash depend on the location in the coal seam from which it is produced. CaO content of fly ashes vary from 6.20 to 74.7%. Chemical composition of bottom section coal fly ash is very similar to Class F, while middle section and top section coal fly ashes are close to Class C. Hematite, quartz and anhydrite are the major crystalline phases in the bottom section coal fly ash sample whereas lime and anhydrite are the major crystalline phases in the middle section and the top section coal fly ash samples. C3A, C4AF and BC2S cement minerals are present in small proportion in all the samples. Ash samples are highly alkaline except the bottom section fly ash sample L4. The bottom section coal fly ash sample positions in the ternary diagram have pozzolanic and also cementations properties. The absence of significant amorphous material was revealed for the AE fly ashes. The chemical composition of the middle section and the top section coal fly ash samples are close to that typical of OPC, and they might be used as mineral admixture in concrete. The bottom section fly ash could be used in tiling and cladding applications due to its chemical and mineralogical composition. Water-soluble fraction and fineness properties of the top section coal fly ash meets the requirements and it might be used in soil stabilization and for grouting the open pit slopes, as a substitute of lime or cement. The AE fly ashes could act as a source of some essential elements to plants, such as sulfur, calcium, magnesium and manganese. By increasing soil pH, they have the capacity of being used as an amendment for acid soil capable of improving soil conditions for proper plant growth. Moreover, they could be also used as a limiting material to raise the low pH of acid soils. Bulk density of fly ash samples ranged from 0.63 to 1.36 g/cm³, and specific gravity ranged from 2.80 to 3.27 g/cm^3 . It is evident that both bulk density and specific gravity values increase as the particle size and CaO content of the ash samples decrease. In addition, due to the wide range of the density of ash samples, particular attention has to be paid for the design and construction of ash handling system.

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