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Mineralogy and geochemistry of Greek and Chinese coal fly ash

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Abstract

In this paper the mineralogy and geochemistry of Greek and Chinese coal fly ash are examined. Annual production of fly ash in China is around 160 Mt while in Greece lignite fly ash accounts around 10 Mt. Even though the mineralogical and chemical composition of the fly ashes coming from these two countries differs, there are common questions on the utilization of this material. The variation of the Greek fly ash' chemical composition, from Ca-poor to Ca-rich fly ash, has resulted to applications such as dam construction, use in cement and possibly in concrete and road construction. The Chinese fly ash, which is rich in mullite, is broadly applied for brick making.

Keywords: Fly ash; Mineralogy; Geochemistry

1. Introduction

Lignite plays an important role in Greece's energy sector as it satisfies over 67% of the country's needs in electric power. The remainder is covered by oil (around 19%), hydroelectric (around 10%), renewable (0.1%) and imported electricity (2%) [11]. Greek lignite is of low quality, characterized by low calorific value and high moisture content (Table 1). The extraction of lignite takes place mainly in three regions of Greece, namely Ptolemais–Amynteo, Florina and Megalopolis. The annual production of lignite is around 60 million tons, of which 48 million tons derive from the coal fields of northern Greece (Ptolemais–Amynteo and Florina) (Fig. 1). Almost the entire lignite production is consumed in power generation.

China, the world's most populous country, is the second largest economy and the second larger consumer of primary energy after the United States. According to the data published by State Bureau of Statistics, China's GDP was 15987.8 billion RMB (about 1998.4 billion USD) in 2004. It currently accounts for about 12% of global GDP and primary energy demand. China's total primary energy demand is projected to grow by 2.6% per year from 2002 to 2030 [7].

China is the largest producer of coal in the world, with nearly 12% of total proven reserves (114 billion tons). The majority of these are found in northern China, particularly in the provinces of Hebei, Shaanxi and Inner Mongolia. Hard coal accounts for 84% of total proven reserves. The remainder consists of lower-quality coals, including lignite [7].

Chinese primary coal demand will grow from 1308 Mt in 2002 to 2402 Mt in 2030, at an average rate of 2.2%. Most of new demand will come from the power generation sector (Table 2). The power sector will account for more than 73% of total Chinese coal consumption in 2030 compared with 52% in 2002. Electricity demand grew by 11% in 2002 and more than 15% in 2003, strongly outpacing

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Table 1
Main quality features of the Greek lignites [11]

Deposit	Net calorific value (kJ kg^{-1})[a]	Ash (%)[b]	Moisture (%)
Ptolemais	5452	15.1	52.6
Amynteo	4828	16.4	54.7
Megalopolis	4400	15.5	57.9
Florina	7960	17.0	42.0
Drama	4315	16.0	59.0
Elassona	8590	19.0	41.0

[a] On as received basis.
[b] On dry basis.

Table 2
Electricity-generation mix in China, TW h [7]

	1971	2002	2010	2020	2030
Coal	98	1293	2030	2910	4035
Oil	16	50	59	65	53
Gas	0	17	55	196	315
Nuclear	0	25	82	180	280
Hydro	30	288	383	578	734
Biomass and waste	0	2	31	58	84
Other renewables	0	0	13	31	72
Total	144	1675	2653	4018	5573

economic growth. The trend continued through the first half of 2004 [7].

The utilization of energy resources promotes economic development but causes, on the other hand, serious environmental problems. Particularly, the emitted fly ash cause serious environmental pollution. The aim of this paper is to define the geochemical and mineralogical features of the fly ash, in particular fly ash from China and Greece with dif-

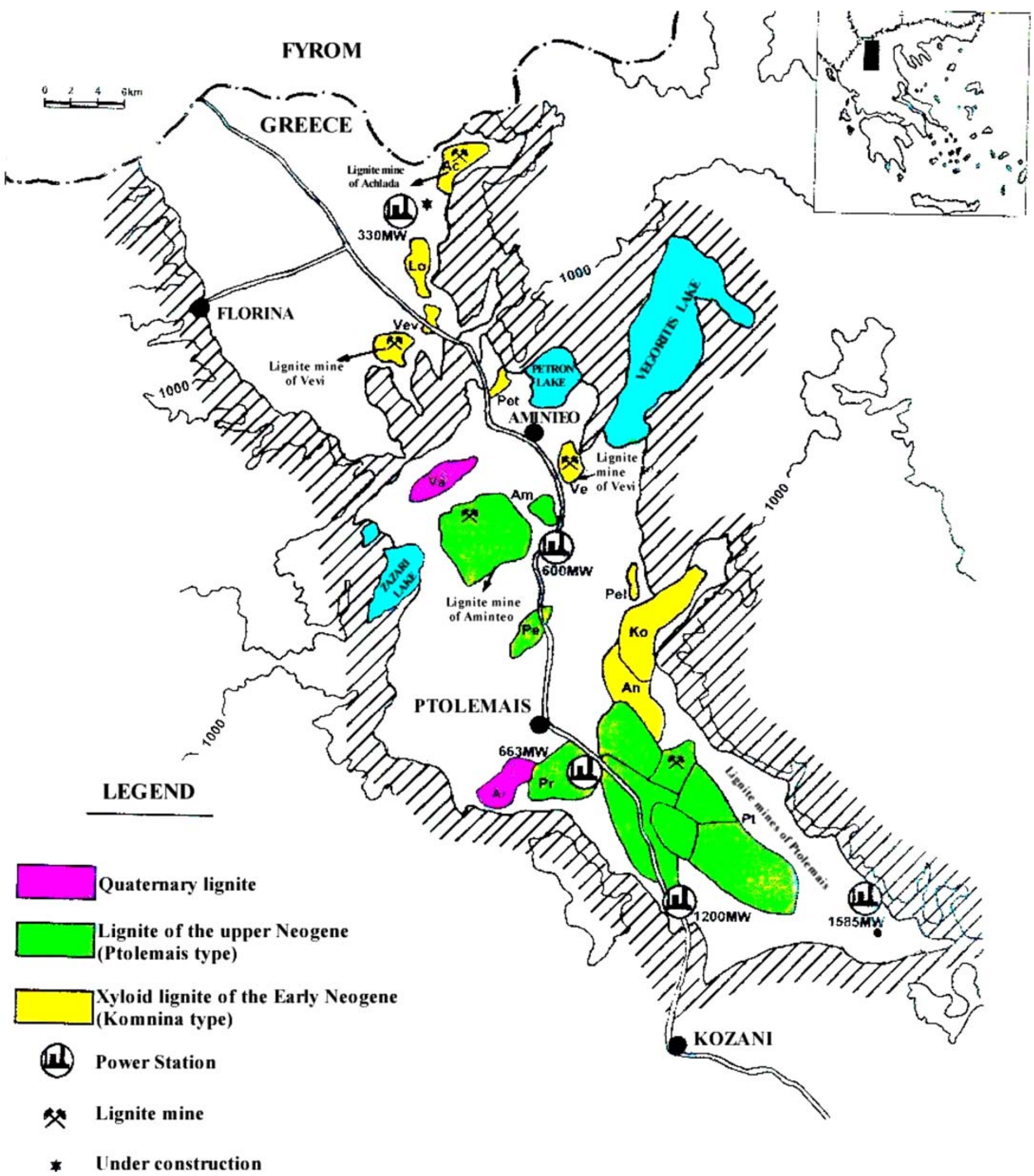

Fig. 1. Location of power stations, deposits and lignite mines of West Macedonia basins, Greece (Pt = Ptolemais, Pr = Proastio, Ar = Ardassa, Ko = Komnina, An = Anatoliko, Pel = Pelargos, Pe = Perdikas, Am = Amynteon, Va = Valtonera, Ve = Vegora, Pet = Petres, Vev = Vevi, Lo = Lofi, Ac = Achlada). Greek fly ash samples were collected from Ptolemais power station (Pt).

ferent characteristics, in order to find out the most suitable use of this lignite by-product.

2. Fly ash in Greece and China

2.1. Greece

The thermal power plants which are located in Ptolemais–Amynteon basin (Fig. 1) are the main sources of fly ash in Greece. Each year approximately 10 millions tons of lignite fly ash is produced in Greece. The fly ash coming out from the Greek Thermal Power plants is classified in type C (according to ASTM C 618), the high CaO, $Ca(OH)_2$, $CaCO_3$ ash. This ash appears to have not only pozzolanic but also hydraulic behavior.

2.2. China

In China the fly ash emitted by the thermal power plants is around 160 million tons [1]. The comprehensive utilization of coal ash amounted to 51.88 million tons in 1995 increased to 70 million tons in 2000, the utilization rated increased from 32% to 44%, it is planned to reach 65% in 2005. Because coal-fired thermal power dominates over power sources, coal ash utilization in China has large potential and brilliant future. With development of electric industry, emitted fly ash will increase year by year, occupy more land and cause serious environmental pollution. Most Chinese fly ash is classified in type F according to ASTM C 618. At the present over 50% of fly ash, produced in China, is used in different purposes such as bricks manufacture which have low economic benefits but also in the construction of dams and cement industry. Large amount of the Chinese fly ash is still emitted into ponds or piled in land.

3. Experimental work

Fly ash was collected by scientists from IGGCAS in China (Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing) and from CERTH/ISFTA in Greece. Chinese fly ash samples come from Douhe power plant, Hebei Province (1550 MW with 2 Mt annual production of fly ash) while Greek fly ash samples come from Ptolemais power station (150 MW with around 1 Mt annual production of fly ash) which is located in Ptolemais–Amynteon basin, northern Greece.

The mineralogical composition of the fly ash was determined by X-ray diffractometry in IGGCAS (China), and in the Technical University of Crete/Greece, Mineral Resources Department. A Siemens D500 powder diffractometer with Cu-Kα radiation, graphite monochromator was used. Quantitative analysis of the Greek fly ash samples was carried out applying the Rietveld method. The Rietveld method enables the fitting of the diffraction pattern calculated from the qualitatively determined phase composition of the sample to the experimentally observed diffraction pattern within a 2θ range. The fitting is achieved, by use of a least-squares technique in which the residual error is minimized by modifying the lattice parameters, the scale factors, atomic parameters and occupancies. When the process is completed, the scale factors provide the quantitative analysis. By addition of a known amount of an internal standard, such as corundum or silicon the amorphous part of the sample can be also determined. The method can be applied only when all the crystal structures of the crystalline phases in the sample are known [12,13].

The content of trace elements was determined by ICP-MS. The major elements were determined by XRF, S2 Ranger (Bruker). The chemical analyses were carried out in ACME analytical laboratories, Vancouver, Canada.

The grain size distribution of fly ashes and the existing state of trace elements were determined by means of scanning electron microscopy (SEM) with energy dispersive X-ray spectroscope (EDX). SEM analyses were carried out at IGGCAS (Beijing) and the University of Athens, Department of Geology.

4. Composition of Greek fly ash

4.1. Mineralogy

The mineralogical composition of the Greek fly ash samples from Ptolemais power station is shown in Table 3. Calcite content varies, among the 12 fly ash samples, within a

Table 3
Mineralogical analysis of the Greek fly ash, from Ptolemais power station

Mineral	Sample											
	FA1	FA2	FA3	FA4	FA5	FA6	FA7	FA8	FA9	FA10	FA11	FA12
Calcite	6.2	6.6	7.9	11.7	15.6	26.5	6.06	12.5	12.5	22.3	39.7	8.7
Quartz	7.6	8.7	7.2	5.1	4.9	7.6	6.2	4.2	4.1	11.7	6.8	9.4
Feldspars	8.2	11.2	9.9	5.8	6.9	5.1	5.9	8.9	8.9	9.4	2.1	10.4
Anhydrite	7.2	7.4	6.3	5.1	4.9	3.3	5.8	4.8	4.8	7.9	14.3	7.8
Lime	3.7	3.9	3.8	4.9	4.5	6.0	7.5	7.2	7.2	3.1	6.9	4.4
Portlandite	1.1	1.3	1.3	2.0	1.2	2.2	4.4	5.8	5.8	0.8	2.1	1.5
Gehlenite	10.2	9.6	9.7	8.7	9.1	9.9	12.2	8.4	10.6	3.5	5.8	9.9
Hematite	2.5	2.5	2.4	2.1	1.9	1.8	2.5	1.8	2.2	1.8	2.4	2.3
Amorphous matter	53.3	48.8	51.4	54.7	50.8	37.4	49.5	44	43.7	39.4	19.9	45.5

wide range (6–40%). This is a result of the variable mineralogical composition of the Ptolemais fly ash as strongly relies to the composition of the lignite which fed Ptolemais power station (Fig. 3). It is well known that the interbeded seams in the lignite deposit and lignite's overburden material consists of calcium-rich sediments such as marl and conglomerates. The appearance of calcite in the fly ash may result either from conglomerate or marl remains. As shown in Table 3 Ptolemais fly ash is calcite-rich with low amorphous content. The amorphous matter is illustrated as glassy cenospheres in the electron microscope (Plate 1).

4.2. Geochemistry

The major and trace elements of 12 samples from the Ptolemais fly ash were determined and the results are given in Table 4.

As indicated in Table 4 the most abundant trace elements in Ptolemais fly ash are Ni, Cr, Zn, Ba, La, Sr and B. It is worthwhile to state that the content of some trace elements, with high pollutant material, such as Cd, Sb, Bi, Cr and Ni, among others is very high compared with typical pulverized coal combustion fly ash. This is due to the geology of the areas surrounding the Ptolemais lignite

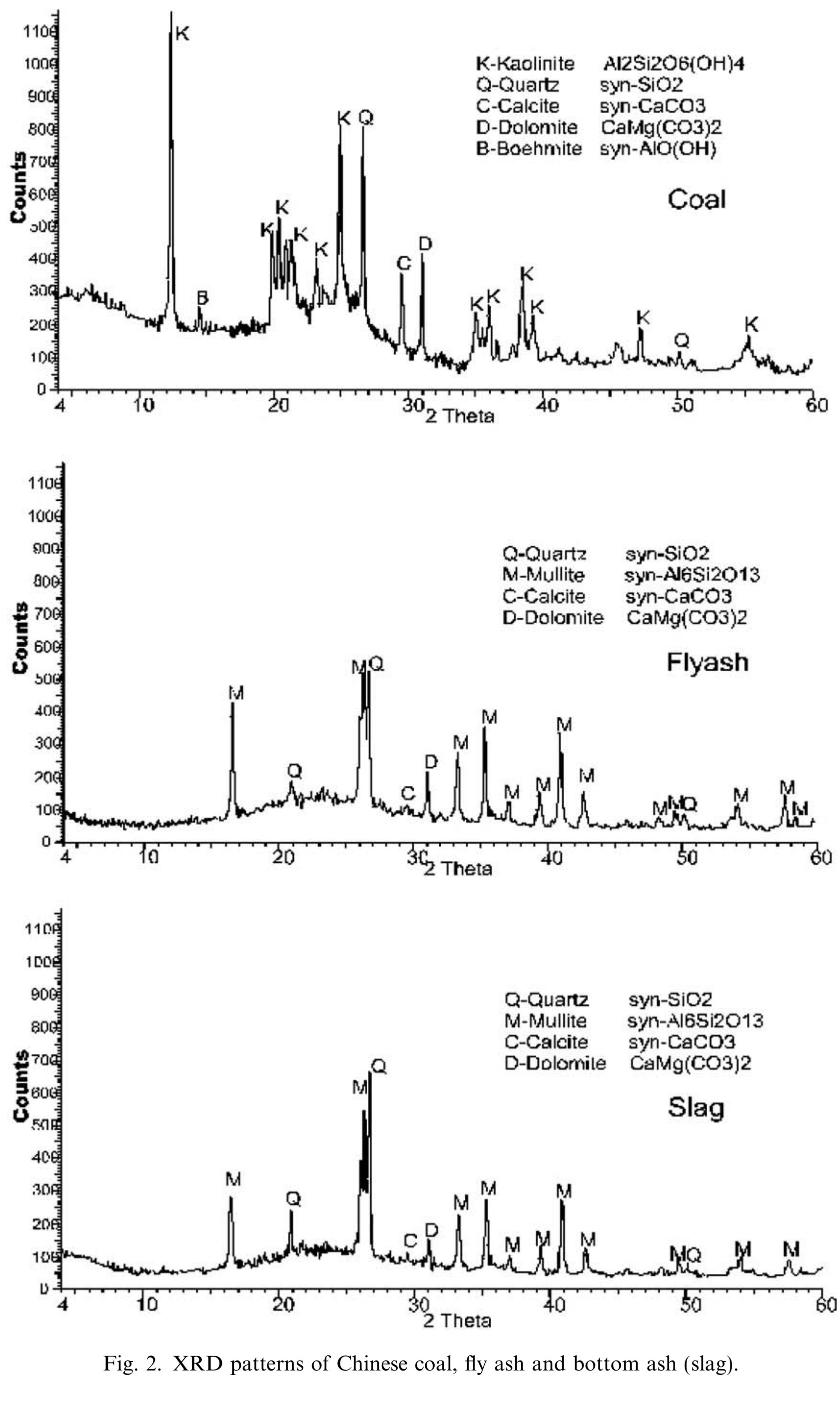

Fig. 2. XRD patterns of Chinese coal, fly ash and bottom ash (slag).

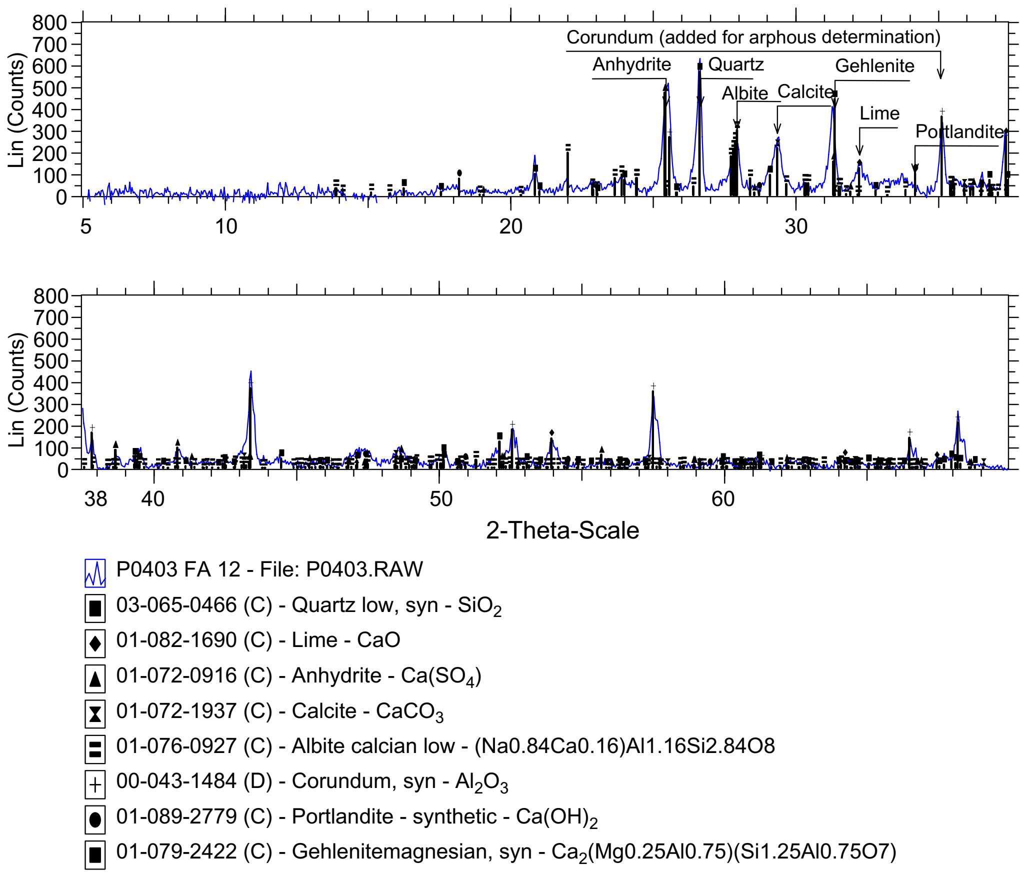

Fig. 3. XRD patterns of Greek fly ash.

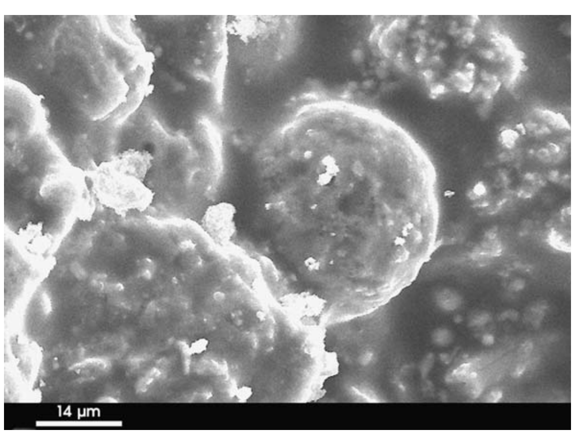

Plate 1. Typical shape of Ptolemais' fly ash cenospheres.

basin. Basic and ultra basic rocks, which are rich in Cr and Ni elements, surrounded Ptolemais basin. As indicated above the fuel of Ptolemais power plant is lignite which is extracted from the open-cast mine in the affinity of the power plant. This fuel contains, apart of the lignite seams, material from the overburden and interbeded seams which mainly consists from material that was deposited in the lake and its composition is very much related to the mountains surrounded the lake.

The linear correlation coefficient for the trace elements, with $r > 0.7$, is given in Table 5. It is clear that there is correlation between the calcium mineral (calcite) and elements typically associated, such as Sr and Ba. There is a negative correlation among Cu and these elements (Sr and Ba). On the contrary it exists high correlation of Cu with Th.

The concentration of the trace elements identified in Ptolemais' fly ash is similar to this reported from other researchers from Greece [2–6,8,9].

The correlation among the fly ash's trace elements, as shown in Table 5, is due to the high geochemical affinity among the corresponding elements and to the operational conditions of Ptolemais power plant. The elements Cr, Ni and Co occur as oxides in the aluminum-silicate glass matrix of the fly ash.

5. Composition of Chinese fly ash

5.1. Mineralogy

The coal used in the power plant comes from Carboniferous and Permian deposits, from Hebei and Shanxi Provinces. Their mineralogical composition consists of kaolinite, quartz, calcite, dolomite and boehmite. Kaolinite is the major mineral phase. Its crystal structure was changed after combustion at high temperature in the boiler, and mullite is produced. Mullite is indicated in the fly ash as material resulted from the thermal decomposition of kaolinite. Results for coal, fly ash and slag analyzed

Table 4
Chemical composition of Ptolemais' fly ash samples

	Sample											
	FA1	FA2	FA3	FA4	FA5	FA6	FA7	FA8	FA9	FA10	FA11	FA12
Major oxides (%)												
SiO_2	70.10	67.39	67.19	65.69	65.23	63.87	62. 89	63.04	64.12	69.40	68.92	60.44
Al_2O_3	4.95	4.91	4.83	6.01	6.07	4.85	4.97	4.22	4.20	5.16	5.22	4.17
TiO_2	0.17	0.18	0.19	0.13	0.13	0.12	0.18	0.14	0.15	0.13	0.12	0.19
Fe_2O_3	4.18	4.19	4.19	4.58	4.61	3.43	4.19	3.61	3.09	3.41	3.19	3.57
MgO	1.65	1.77	1.80	2.24	2.27	2.17	2.29	2.16	2.23	1.38	1.41	1.40
MnO	0.45	0.48	0.50	0.42	0.35	0.46	0.40	0.32	0.34	0.34	0.28	0.30
CaO	16.35	18.50	18.68	18.74	19.14	23.10	22.74	23.94	23.91	17.53	18.12	26.82
Na_2O	0.12	0.14	0.14	0.06	0.06	0.07	0.12	0.10	0.14	0.05	0.04	0.08
K_2O	0.49	0.51	0.52	0.48	0.49	0.45	0.48	0.38	0.41	0.49	0.50	0.30
SO_3	1.45	1.81	1.85	1.54	1.54	1.37	1.61	1.96	1.28	2.02	2.09	2.62
P_2O_5	0.09	0.12	0.11	0.11	0.11	0.11	0.13	0.13	0.13	0.09	0.11	0.11
Total	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Trace elements (ppm)												
Mo	7	8	7	4	4	3	6	7	6	5	5	6
Pb	16	17	15	17	18	19	27	18	19	19	20	20
Zn	51	52	49	70	71	62	65	62	45	59	59	37
Ni	139	130	133	671	671	387	340	333	228	138	145	163
Co	20	22	21	35	37	23	24	22	16	16	16	13
Th	10	11	10	8	8	8	10	8	9	11	11	8
Sr	205	225	233	248	257	289	333	348	400	195	196	262
Cd	0.42	0.39	0.34	0.58	0.59	0.80	1.23	0.81	0.50	1.04	1.10	1.20
Sb	0.21	0.23	0.21	0.36	0.34	0.37	0.42	0.49	0.38	0.33	0.33	0.58
Bi	0.34	0.35	0.31	0.27	0.28	0.23	0.32	0.26	0.25	0.36	0.37	0.36
V	142	132	128	147	150	111	129	121	111	115	118	138
La	26	28	27	25	25	24	30	26	29	31	32	26
Cr	158	152	155	477	475	335	265	244	170	167	171	174
Ba	104	106	124	158	160	170	163	135	184	139	119	132
B	24	26	24	73	66	107	146	163	189	33	38	53
Ga	13	13	13	15	15	13	14	12	11	13	14	12
W	0.5	0.6	0.5	0.6	0.6	0.7	1.5	1.4	1.1	0.2	0.3	0.9
Sc	11	11	11	16	11	11	10	8	9	9	8	11
Tl	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Hg	61	59	50	50	43	46	58	59	47	42	33	45
Se	2	2	2	2	2	2	4	3	2	2	2	4
Te	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1

Table 5
Geochemical correlation between trace elements with $r > 0.70$

Element	Element	r (linear correlation coefficient)
U	Se	0.72
Cd	Se	0.75
U	B	0.73
Cu	B	−0.87
As	B	0.72
Ni	Cr	0.99
Zn	Cr	0.81
Co	Cr	0.9
Th	La	0.82
U	Sb	0.91
Pb	Cd	0.84
Ni	As	0.76
Zn	Co	0.83
Ni	Zn	0.8
Sr	Cu	−0.82
Th	Cu	0.75
Ba	Cu	−0.77
Sr	U	0.75

by XRD are given in Fig. 2. The main minerals found in the ash of Douhe Power plant are mullite, calcite, dolomite and quartz.

The quantitative composition of the fly ash, as determined by the Rietveld method, is: mullite, calcite and dolomite 71%, quartz 5%, amorphous 24%.

5.2. Morphological research of bottom ash and fly ash

The ash from coal combustion melts and forms glass (glass drop) in high temperature. The air in the microstructure of coal expanded during the burning, and formed microsphere and spherical grain. There is some unburned coal particle remained in the ash (Plate 2).

The grain size of fly ash varies from 0.1 mm to 1 mm, while the majority of fly ash sizes are about 0.1 mm, the shapes of grains are mainly spherical grain, irregular melted grain and porous grain (Plates 3 and 4). The major mineral compounds of the spherical grains are quartz, alu-

Plate 2. Coal particle in the Chinese fly ash.

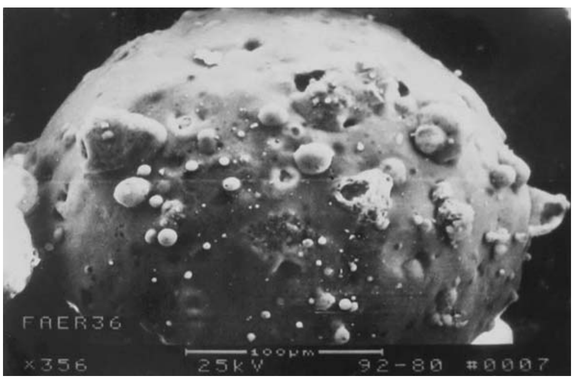

Plate 3. Spherical grain of the Chinese fly ash.

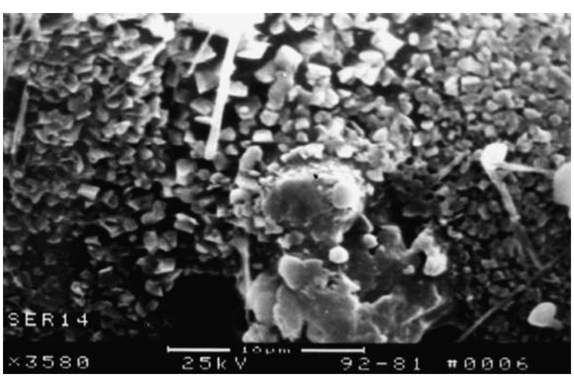

Plate 4. Irregular melted grain of the Chinese fly ash.

Plate 5. Radial shape of mullite.

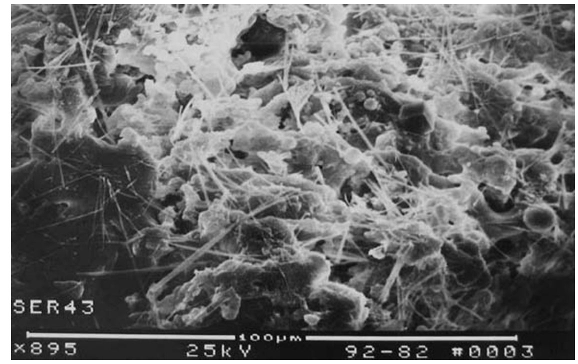

Plate 6. Needle shape of mullite.

mina and magnetite; the major mineral compounds of irregular grains are quartz, calcium oxide and magnetite. These inorganic minerals in coal are mostly silicates, clays, carbonates, sulfides and quartz. The clay lost water and formed glass when coal burning temperature is over 1000 °C and the vitreous body often co-exist with some

Table 6
Chemical composition of fly ashes in Douhe Power Plant

	CFA1	CFA2	CFA3	CFA4	CFA5
Major oxides (%)					
SiO_2	98.38	98.51	98.44	98.48	98.38
Al_2O_3	0.06	0.05	0.06	0.05	0.06
TiO_2	0.04	0.04	0.04	0.04	0.04
Fe_2O_3	0.55	0.51	0.53	0.52	0.55
MgO	0.06	0.06	0.06	0.06	0.06
MnO	0.06	0.06	0.06	0.06	0.06
CaO	0.79	0.71	0.75	0.73	0.79
Na_2O	0.01	0.01	0.01	0.01	0.01
K_2O	0.02	0.02	0.02	0.02	0.02
SO_3	0.01	0.01	0.01	0.01	0.01
P_2O_5	0.02	0.02	0.02	0.02	0.02
Total	100.00	100.00	100.00	100.00	100.00
Trace elements (ppm)					
Mo	3	2	3	2	2
Pb	3	3	2	3	3
Zn	5	5	4	4	5
Ni	3	4	4	3	3
Co	1	1	1	1	1
Th	5	4	4	4	5
Sr	121	117	118	120	117
Cd	1	1	1	1	1
Sb	1	1	1	1	1
Bi	1	1	1	1	1
V	10	10	10	10	10
La	15	14	15	14	15
Cr	2	2	2	2	2
Ba	250	237	240	245	250
B	21	20	20	21	21
Ga	3	3	3	3	3
W	1	1	1	1	1
Sc	2	2	2	2	2
Tl	1	1	1	1	1
Hg	16	21	20	18	21
Se	1	1	2	2	2
Te	1	1	1	1	1

minerals, such as vitreous phase and mullite (Plates 5 and 6), and carbonates changed into calcium oxide after releasing CO_2, sulfide changed into Fe_3O_4 and Fe_2O_3.

5.3. Geochemistry

The major and trace elements of the selected five Douhe fly ashes are quoted in Table 6. The extremely high content of silica is demonstrated in all fly ash samples from Hebei Province. The chemical composition is related to the mineralogy of the fly ash, as minerals rich in silica and alumina (quartz and mullite) were detected by XRD. Although the calcium content in the fly ash is very low compared to Ptolemais fly ash, minerals such as calcite and dolomite are clearly indicated in the mineralogical analysis.

The main trace elements of Douhe fly ash are Sr, V, La, and Ba which are closely related with the major elements.

The elements Ni, Cr, Zn, Ba, La, Sr and B have lower concentrations in the fly ashes compared to the Greek fly ashes. This is due to the composition of the rocks which surrounded the coal basins; granitic in the case of the Douhe ashes, basic and ultra basic for the Greek fly ashes.

6. Applications

The utilization and disposal of the fly ash residues has been the subject of development activity for many years. Emphasis has been put on the increased use of fly ash materials in high volumes, as constituents of cements and mortars, or for the manufacture of concrete and aggregate materials. The utilization of power station fly ashes depends on their physical and chemical properties.

6.1. Greece

In Greece, ashes are mainly used in cement industry replacing cement clinker and aiming to the production of special types of Portland cements. Furthermore, they are successfully tested in road construction, several mortars, waste treatment, and embankments and cement grouting. CERTH/ISFTA has participated in pilot project concerning the utilization of fly ash in the production of concrete, using fly ash, with encouraging results. As indicated in the report resulted from the project, the use of fly ash from Ptolemais power station in concrete increase the strength of the final product and the porous structure of fly ash enhances the quality of concrete [10].

The only massive use of Ptolemais fly ash took place in the construction of Planatovryssi dam. In this dam the roller compacted concrete (RCC) technique was applied, utilizing fly ash as basic cementitious material. The participation of fly ash in the concrete was 82%. RCC is defined broadly as no-slump concrete compacted by roller, usually a vibratory roller. The dam is 95 m high with a crest length of 270 m and a volume of 450,000 m^3. The Planatovryssi dam is about 12 km downstream the 170 m high Thissavros rock fill dam and acts as the lower reservoir of Thissavros pumped storage plant of 300 MW (Fig. 4). Since 1992, the year of Platanovryssi's dam construction, no problems have been reported.

Fig. 4. Platanovryssi dam.

Fig. 5. Three Gorges dam.

6.2. China

In China coal deposits range from Tertiary to Carboniferous in age, and the coal ranks from lignite to anthracite. The comprehensive utilization of fly ash in China is developed. Fly ashes have been blended in concrete and sand slurry in construction industry since 1950s. In addition to this, fly ash is used to produce brick and as base material on road construction. At the moment fly ash is used mainly in the following applications: sintered brick, steam-curing brick, silicate block, aerated-concrete, instead of clay as raw material in cement, as blending material to manufacture cement, and in the road construction.

There are considerable social, environmental and economic benefits from the utilization of fly ash in China. In the Jinan-Qingdao freeway, fly ash was used in the construction of the road, in 1992. The part of the freeway using fly ash was of 4 km, with an average fill height of 2.7 m. Around 0.4 million tons of fly ash were used, saving 0.325 M yuan and 22.5 ha of ash-ponding area. In 1990 the road connection project in the north of the Qianjiang second bridge, Hangzhou, Zhejiang Province, 0.21 Mt of fly ash was used to build road with a length of 1.7 km, average height of 4.2 m, and width of 26 m, and 4 ha for ash-ponding area and 3 M yuan were saved.

The construction of a dam within the framework of the "Three Gorges Project" was commenced at the end of 1994. Around 15 million tons of fly ash was used in the construction of the dam (Fig. 5). The dam is of 185 m in altitude, and its length is over 2300 m. In 2006 the running water level will be 165 m, in 2009, 175 m. The capacity of the overall reservoir is 39.3 billion m^3, and prevent flood capacity of the reservoir is 22.15 billion m^3. The total pump capacity plant of the dam is 1768 MW and the annual electricity generation is 84 billion KW h. In 2006, the first batch set will generate electricity.

The large utilization of the Chinese fly ash in various applications, such as road construction, dam construction and brick manufacturing, provide useful information to initiate similar projects in Greece.

7. Conclusions

China and Greece are two countries heavily relying on coal to meet their electricity generation needs. China, which is the largest producer of coal in the world, generated around 1300 TW h in 2002 from coal-fired power plants and produced around 160 million tons of fly ash. Greek lignite satisfies over 67% of the country's needs in electric power, producing around 10 million tons of fly ash yearly.

Selected samples from China (Hebei Province) and Greece (Ptolemais) were analyzed in order to determine their mineralogical and chemical composition. Chinese ash contains mullite, calcite, dolomite and quartz (76%) and amorphous material (24%) while the main mineral phases included in coal are quartz, kaolinite, calcite, dolomite and bohemite. The mineralogical composition of the Greek fly ash includes calcite, quartz plagioclase, anhydrite, lime, portlandite, gelehnite, hematite and amorphous material.

The Chinese fly ash is classified in type F, according to ASTM C 618, while the Greek fly ash in type C. The examined Chinese fly ash is rich in Al_2O_3 (35%) while the Greek fly ash is CaO-rich (28%). As far as the trace elements is concerned, the Chinese fly ash contains lower concentrations of Ni, Cr, Zn, Ba, La, Sr and B compared to the Greek fly ash. This is due to the surrounded the coal basin rocks' composition.

Fly ash is used in China in the construction industry since 1950s. At present it is used as blending material to produce cement, in road construction, and in brickworks. The "Three Gorges Project", a 185 m height dam, is also constructed using 15 million tons of fly ash. On the contrary the utilization of fly ash is only extended in the construction of Platanovryssi dam (82% participation of fly ash in the concrete) and in cement production.

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