

Assessment of Sintering of Fly Ash in a Circulating Fluidised Bed Combustor

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Abstract

This paper describes a comparative study of ash deposition in two 125 MWe circulating fluidised bed combustor power generation plants (CFBC 1 and CFBC 2) undertaken to provide a comparative data for the identification of the possible causes of ash deposition in CFBC 2. Samples of the feed coal, limestone, deposits and ashes were collected from various sampling points within the furnace and from areas of ash deposition. The samples were analysed using a combination of ash chemistry and quantitative X-ray diffraction analysis, optical petrography and QEMSCAN analysis. Although the results indicated that the nature of the superheater deposits in both power plants is similar in terms of mineralogy and texture (dominated by iron oxide and anhydrite occurring as layers), the ash chemistry and mineralogy are both significantly different. The study confirmed that CFBC 2 ashes are unusual in the occurrence of complete sulphation of the decarbonated limestone with no evidence of either the occurrence of intermediate phases such as CaO or the presence of sulphate reaction rims on decarbonated limestone as observed for CFBC 1 ashes and commonly described in the literature. A possible reason for this unusual behaviour is the high sulphur content of feed coal in CFBC 2 leading to complete sulphation of limestone due to an insufficient feed rate. Another possible factor is the greater proportion of fine particles in the milled CFBC 2 limestone which would react completely. This observation is supported by the occurrence of fine anhydrite particles in the CFBC 2 back pass sample and a subsequent increase in grain size in the back end of the boiler, suggesting that winnowing of the fine particles has occurred in the hotter sections of the back pass. This is in contrast to the relatively limited size variation exhibited by anhydrite in the ash samples of CFBC 1.

Introduction

Combustion is the largest and fastest growing application of circulating fluidized-bed (CFB) technology and a very promising coal combustion technology for cleaning flue gases. The CFBC technology is more efficient and advanced than pulverized coal combustion (PCC) due to large reductions of SO₂ and NO_x emissions in flue gases [1]. The CFB technology is used around the world to generate electric power by utilizing various low grade fuels, including coal with reduced pollutants in an environmentally friendly manner. Both the capacity and number of CFB boilers in use have been increasing over the last 10-15 years or so with the largest CFB boiler in operation being a 460 MWe supercritical once through utility (OUT) CFB Boiler in Poland. In a CFBC boiler, powdered coal is burnt in the presence of ground limestone which is used as a sorbent of SO₂ at 850–900 °C, significantly below the typical operating temperature (1200–1400 °C) of a pf plant. In order to increase the efficiency of SO₂ removal, typical Ca/S molar ratios are often increased to 2–2.5 [2-3].

The solid inventory usually consists of crushed refractory or natural sand, and fuel-originated ash as single particles or attached on the sand particles, and sorbents such as limestone. Usually the operating temperature (bed temperature) of a FBC reactor is 800-880 °C. Bed particles absorb the heat generated during combustion and distribute it throughout the furnace by convection and radiation. The principal reason for the low temperature is the optimum sulphur capture: To reduce SO₂ emissions, the combustor is typically fed with limestone or dolomite particles which calcine and then fix SO₂ in the temperature range of 800-900 °C, which reacts to form anhydrite (CaSO₄) [3-5]. This low-temperature range avoids ash softening; a further advantage that follows out is that virtually no thermal NO_x is produced. However, a very important mechanism associated with ash-formation is sintering which could eventually lead to operational difficulties. Sintering is largely governed by the coal quality, limestone characteristics as well as the combustion conditions (based on air flow distribution) in the boiler. In an attempt to study the sintering propensity of fly ash, a case study has been undertaken to investigate the causes of ash deposition in the superheater sections of CFBC boilers. The paper also addresses the most probable mechanisms and explanations for the various phenomena seen with sulphur capture in both CFBC plants.

Sampling and analytical procedures

One sample each of the feed coal, limestone, several ash and deposit samples from different sections of both identical plants (CFBC 1 and CFBC 2) were considered for analysis (see Table 1 for details). However, Sub-samples were taken of the coal sample for chemical analysis, to prepare a radiofrequency plasma mineral matter sample for X-ray diffraction analysis and to prepare the QEMSCAN™ (an automated electron beam image analysis instrumentation package based upon a SEM) analysis mount. The limestone and ash samples were sub-sampled to provide material for chemical analysis and X-ray diffraction. The deposit sample was subdivided into sixteen sub samples based upon textural characteristics. Further samples were taken of the ash and deposit for QEMSCAN analysis and of the deposit sub-samples for preparation of polished blocks and thin sections for petrographic analysis.

Table 1 Description of samples considered

CFBC 1	CFBC 2
Feed coal, limestone	Feed coal, limestone
Bed Ash	Bed Ash
Seal Pot Deposits Left side Back Pass Inlet left	Cyclone ash
Reheater 2 Deposits in B/W banks	Air preheater ash
Superheater deposits	Superheater deposits
Economiser Right side banks top, Fly ash	ESP ash, Fly ash

Results and Discussion

Feed coals

Although the alumina contents of both coal feed ashes are comparable, silica is lower and iron higher in the CFBC 2 lignite ash. Calcium and sulphur concentrations are greater in the CFBC 1 lignite ash and sodium significantly lower. Based on ash chemistry, several indices have been developed as indicators of slagging and fouling propensity and values for some of these are given in Table 2, having been calculated using several expressions detailed elsewhere [6]. Slagging indices indicate that both coals would have a similar tendency to form ash deposits. Although the sulphur content of the CFBC 1 ash is much higher than in the CFBC 2 ash, total sulphur in the CFBC 1 lignite is 2.26 wt% S on a dry basis compared to 7.35 wt% S in the CFBC 2 lignite. The mineralogy of both lignites reflects the differences in chemistry. Silica and kaolinite contents are higher in the CFBC 1 lignite, although interlayered illite/smectite is present in the CFBC 2 lignite. Calcium sulphate contents are higher in the CFBC 1 lignite, although all three calcium sulphate polymorphs are present in the CFBC 2 lignite, bassanite being the most abundant. Natrojarosite is present in the CFBC 2 lignite and hexahydrite in the CFBC 1 lignite; the pyrite content of the CFBC 2 lignite is double that of the CFBC 1 lignite.

Table 2 Slagging indices of lignite fuels

Ash deposition index	CFBC 1	CFBC 2
Si ratio	48.32	60.92
Base/Acid	0.76	0.43
iron Index	20.24	7.01
Fe/Ca	5.95	2.26
Fe+Ca	31.13	23.42

Limestones

The CFBC 2 limestone is essentially pure calcium carbonate containing only minor amounts of silica. In contrast, the CFBC 1 limestone has higher silica, alumina and iron contents. The purity of the CFBC 2 limestone is also shown by the X-ray diffraction analysis, the calcite content being 96.9 wt% occurring in association with minor dolomite, quartz and illite. Calcite content in the CFBC 1 limestone is 82.0 wt%; kaolinite and goethite contents are approximately 8 wt% and quartz content 1 wt%. A Hardgrove test was performed on both limestones to gauge their relative resistance to abrasion. The results are practically identical, the Hardgrove index being 89 for the CFBC 2 limestone and 88 for the CFBC 1 limestone, indicating that the two limestones have very similar abrasion characteristics. Particle size analysis of the milled limestones (see Figure 1) indicated that CFBC 2 limestone has a coarser maximum particle size of 320 μ m as compared to 140 μ m for the CFBC 1 milled limestone. The median particle size for CFBC 2 is also coarser being 20 μ m as opposed to 14 μ m for CFBC 1. However, finer particles are more abundant in the CFBC 2 milled product which has a modal value of only 12 μ m compared to the value of 38 μ m in the CFBC 1 milled product.

Ashes

Although the ash samples are not strictly comparable as the samples have been taken from different locations some generalised observations can be made (see Tables 3 and 4). Silica, alumina and sodium contents are higher in the CFBC 1 ashes. Iron and calcium contents are comparable but both elements show greater variability in the CFBC 2 ashes. Sulphur contents are consistently greater in the CFBC 2 ashes. The higher silica and alumina contents of the CFBC 1 ashes are reflected in the higher quartz and mullite contents (Tables 5 and 6). Anorthite contents are comparable although higher values tend to be present in the CFBC 2 ashes. Nepheline is a common phase in the CFBC 2 ashes but is absent in the CFBC 1 ashes. A characteristic feature of the CFBC 1 ashes is the occurrence of calcite, lime and portlandite which show a systematic variation with respect to position within the plant; only minor calcite is present in the CFBC 2 ashes. Iron oxide contents are higher in the CFBC 2 ashes, although a greater variability in the nature of the iron oxides is present in the CFBC 1 ashes with the occurrence of magnetite, maghemite and hematite as compared to the

dominance of hematite in the CFBC 2 ashes. Anhydrite contents are much higher in the CFBC 2 ashes. Amorphous contents are significantly higher in the CFBC 1 ashes.

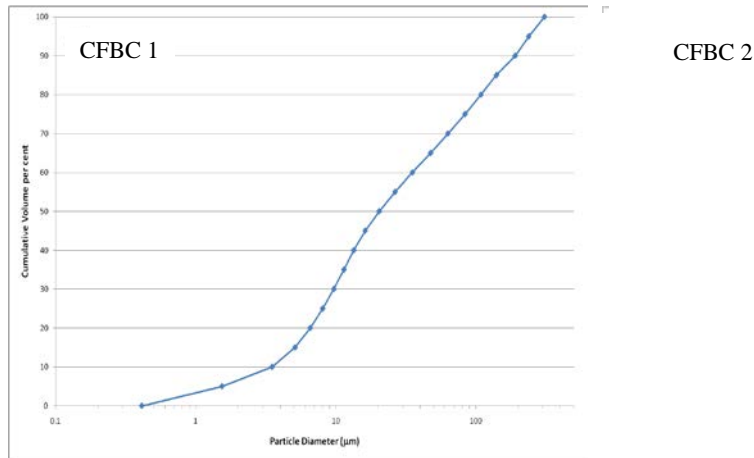


Figure 1 Cumulative volume per cent grain size plots of milled limestones

Table 3 Chemical analyses of the coal, limestone and ashes of CFBC 1

Oxide	Limestone	Lignite	Bed Ash	Cyclone	APH	ESP	Fly Ash	Superheater Sample 1	Superheater Sample 2
SiO ₂	8.81	40.53	41.10	38.39	35.51	24.36	27.79	4.66	7.37
TiO ₂	0.57	3.02	2.94	2.81	2.49	1.66	1.77	0.56	0.65
Al ₂ O ₃	4.48	19.45	16.73	19.49	17.74	11.49	11.33	3.38	4.36
Fe ₂ O ₃	7.36	16.24	27.84	13.70	14.59	11.26	11.08	9.77	8.79
MnO	0.13	0.15	0.84	0.16	0.19	0.16	0.16	0.15	0.13
MgO	0.54	2.58	1.53	1.66	1.66	1.95	2.33	40.38	38.78
CaO	40.47	7.18	5.76	15.11	16.51	31.95	26.35	2.73	2.66
Na ₂ O	<0.01	1.00	0.53	0.73	0.94	0.42	0.81	0.08	0.09
K ₂ O	0.05	0.20	0.19	0.25	0.23	0.10	0.22	<0.01	<0.01
P ₂ O ₅	0.05	0.05	0.17	0.07	0.07	0.06	0.09	0.04	0.03
SO ₃	0.09	8.47	2.00	7.93	7.93	9.81	7.34	39.11	36.35
L.O.I.	36.88	1.00	0.35	None	2.09	7.24	9.26	0.32	0.20
TOTAL	99.43	99.87	99.98	100.30	99.95	100.46	98.53	101.18	99.41

Note: LOI Loss on ignition at 1000°C

Table 4 Chemical analyses of the coal, limestone and ashes of CFBC 2

Oxide	Limestone	Lignite	Bed Ash	Seal Pot	Back pass	Reheater	Superheater Deposit	Economiser	Fly Ash
SiO ₂	2.41	31.22	21.08	18.32	21.62	8.99	4.71	12.23	14.66
TiO ₂	0.03	1.89	1.14	1.02	1.32	0.52	0.35	0.69	0.95
Al ₂ O ₃	0.60	17.00	11.18	9.83	12.16	4.81	2.79	6.94	8.28
Fe ₂ O ₃	0.31	26.65	11.61	12.30	34.79	15.39	10.65	16.47	18.93
Mn ₃ O ₄	0.01	0.13	0.17	0.07	0.11	0.09	0.09	0.14	0.13
CaO	59.45	4.48	25.38	18.83	6.97	26.87	39.99	22.93	28.54
MgO	0.47	2.27	1.33	1.42	2.18	1.11	1.19	1.06	2.01
Na ₂ O	0.03	4.34	3.36	2.89	3.61	1.66	1.16	2.33	2.31
K ₂ O	0.03	0.32	0.20	0.14	0.24	0.11	0.07	0.14	0.16
P ₂ O ₅	0.23	0.05	0.17	0.12	0.07	0.15	0.21	0.13	0.18
SO ₃	0.43	5.49	23.34	24.50	7.51	31.19	36.18	28.72	19.18
SrO	0.03	0.04	0.05	0.04	0.05	0.04	0.04	0.04	0.05
BaO	0.00	0.01	0.01	0.00	0.01	0.00	0.00	0.00	0.00
LOI	35.97	6.11	0.99	10.53	9.38	9.08	2.56	8.18	4.61
Total	100.01	100.00	100.0	100.0	100.0	100.00	100.00	100.00	100.0

Note: LOI Loss on ignition at 1000°C

Table 5 Quantitative X-ray diffraction mineralogy of the coal mineral matter, limestone and ash samples (in wt%) of CFBC 1

Oxide	Limestone	Lignite	Bed Ash	Cyclone Ash	APH	ESP Ash	Fly Ash
Mineral matter wt%		28.9					
Quartz	1.1	10.6	15.8	2.8	3.0	5.2	5.9
Kaolinite	8.2	48.4					
Mullite			10.8	8.4	4.9		
Sillimanite				1.6	3.5		
Anorthite			2.3	1.8	3.1	4.2	2.9
Rutile			0.6	0.6	0.4		
Goethite	8.7						
Magnetite		1.0	9.5	4.5	5.6	1.9	3.5
Maghemite			5.5	2.9	3.2	2.0	0.1
Hematite			6.9	5.1	3.8	6.6	4.6
Ilmenite			2.1	1.4	2.2		
Hercynite				3.3	2.2		
Spinel			4.1	0.3			
Lime			1.8	3.1	1.9	8.3	1.4
Portlandite					5.0	2.8	4.2
Calcite	82.0				0.9	12.2	14.0
Anhydrite		6.7	4.4	9.8	14.5	17.9	14.8
Bassanite		17.5					
Hexahydrate		4.9					
Aphthitalite							0.5
Srebrodolskite					1.5	3.7	3.6
Mackinawite		0.9					
Pyrite		10.0					
Amorphous			36.3	54.4	44.4	35.2	44.6
Total	100.0	100.0	100.1	100.0	100.1	100.0	100.1

Table 6 Quantitative XRD mineralogy of the superheater deposit sub-samples (wt%) of CFBC 1

Description	A (-1.7 mm)	D (-5mm + 1.70mm)	E	F	G	H	I	J	K	L	N	O
Quartz	4.3	0.4	0.3	0.4	1.2	0.4	0.1	0.3	0.4	0.1	3.3	0.6
Cristobalite									1.3			
Mullite												
Anorthite	4.6	3.4	2.7	2.9	3.0	3.9	1.7	2.3	2.9	1.1	2.7	6.3
Tephroite		1.8	1.9	3.7				2.4				
Ca ₂ SiO ₄			2.4	3.4								2.2
Gehlenite	2.0	9.2	7.8	4.1		6.5	7.3	11.4	9.7	4.0		4.0
Wollastonite												13.8
Augite	5.8	2.5	0.9	0.9								
Diopside	5.8	3.8	3.3	2.3	2.5	6.0	4.0	4.4	5.8	7.8	1.9	6.5
Corundum												
Maghemite	0.7	10.7	11.9	11.6	3.9	10.7	12.9	15.0	11.2	6.2		
Hematite	12.5	6.9	7.9	2.8	10.4	3.1	1.2	0.4	2.6	3.4	15.7	15.2
Ca ₃ Al ₂ O ₆		3.1	1.9			0.8	2.0	1.9	1.3			
Ca ₂ Fe ₂ O ₅						5.5	2.1	3.9	2.2			
Lime	0.2											0.3
Calcite	0.8	0.6	0.6	0.5		0.5	0.4	0.5	0.4			1.6
Anhydrite	42.3	57.3	57.7	59.2	78.4	62.1	68.3	57.4	62.2	77.6	71.9	35.7
Hexahydrite	0.9	0.1	0.4		0.5						4.5	0.5
Melanterite	0.5	0.3	0.4	0.8	0.2	0.4		0.1				0.5
Srebrodolskite				7.5								
Amorphous	19.6											12.8
Total	100.0	100.1	100.1	100.1	100.1	99.9	100.0	100.0	100.0	100.2	100.0	100.0

QEMSCAN particle maps of the bed ash, cyclone air preheater, electrostatic precipitator and fly ash samples of CFBC 1 revealed sharp reduction in grain size when passing from the air preheater to the electrostatic precipitator ash. Abundant aluminosilicate particles are present in the bed ash frequently in intimate association with iron silicate. Iron oxide grains are also present, some of which have a complex zonal structure, the iron oxide occurring in association with a calcium-iron silicate. A narrow, incomplete rim of Ca-Al silicate is present on several grains. Anhydrite grains are common and frequently have a core of calcium mineral which is likely to be calcium oxide. The results of the QEMSCAN examination of selected sub samples of the superheater deposit are shown in Figure 2. The most characteristic feature is the dominance of anhydrite and iron oxide, both of which define a layering based upon their relative proportions. As shown in the optical examination, the layering varies from a simple planar geometry to a complex curvilinear pattern. Aluminosilicate particles occur as inclusions in some samples and also as layers enclosed by anhydrite/iron oxide layers. Associated with the aluminosilicate are grains of calcium iron silicates which occasionally occur as rims on the aluminosilicates.

The results of the QEMSCAN examination of the thin sections of deposits from CFBC 2 indicated that the seal pot deposit is comprised of loose aggregates of particles with aluminosilicate particles (derived from the breakdown of clays) predominating. Discrete anhydrite particles are also common; particles consisting of a fine intergrowth of anhydrite and iron oxide are also present and the association also occurs as rims on other particles. In both the superheater deposits anhydrite is the dominant phase as shown in Figure 2. The superheater deposit can be seen to consist almost entirely of massive anhydrite, often containing disseminated iron oxide+anhydrite intergrowths occurring in discrete layers. Rare aluminosilicate particles are also present and coarse lenticular silica rich areas are also common. The QEMSCAN data confirms that the development of deposit strength is related to the amount of anhydrite present and its mode of occurrence. Of the three deposit samples, the

superheater deposit is the strongest in which the anhydrite content is highest and its occurrence is as massive aggregates.

Texturally, as shown by the QEMSCAN analysis, the CFBC 2 ashes are unusual in the absence of any calcium phases other than anhydrite. In contrast many of the anhydrite grains in the CFBC 1 ashes have a core of a phase identified as “Ca mineral” in the QEMSCAN analysis which most likely corresponds to the lime and portlandite identified in the X-ray diffraction analysis. Iron oxide grains are also less abundant in the CFBC 1 ashes.

Table 7 Quantitative X-ray diffraction mineralogy of the coal mineral matter, limestone and ash samples (in wt%) of CFBC 2

Oxide	Limestone	Lignite mineral matter (%)	Combustor Bed Ash	Seal pot deposits	Back Pass Inlet	Reheater Deposits	Superheater Deposits	Economiser	Fly Ash
Quartz	0.6	4.7	1.8	3.6	2.3	3.3	0.2	3.2	1.5
Cristobalite				3.1	2.6	2.0			
Mullite				3.2	2.8	1.6			
Diopside			0.9				2.4	0.4	
Illite	0.4								
Illite/smectite		15.5							
Kaolin	0.6	33.8							
Montmorillonite	0.4								
Orthoclase	0.6								
Albite	1.1								
Anorthite			3.1	5.8	7.7	0.6		1.9	8.5
Nepheline			9.1	4.0	2.7	1.1	3.0	6.0	6.8
Hauyne			1.6						
Calcite	95.8					0.8	0.5	0.2	0.8
Dolomite ferroan	0.6								
Hematite			4.5	17.9	44.2	23.2	10.9	18.1	20.7
Maghemite			6.9				6.9	5.0	9.8
Portlandite					0.1				
Anhydrite		5.2	47.0	44.5	14.8	61.1	75.5	51.4	50.9
Bassanite		9.0							
Gypsum		3.8							
Hexahydrate				2.2		1.2		0.7	0.5
Calcium magnesium sulfate					x	x			
Natrojarosite		5.9							
Loweite				x	x	x	x	x	
Butlerite			0.3		0.2	0.3	0.6	0.5	0.6
Pyrite		22.1							
Amorphous			24.6	15.6	22.4	4.6		12.6	
Total	100.1	100.0	99.8	99.9	99.8	99.8	100.0	100.0	100.1

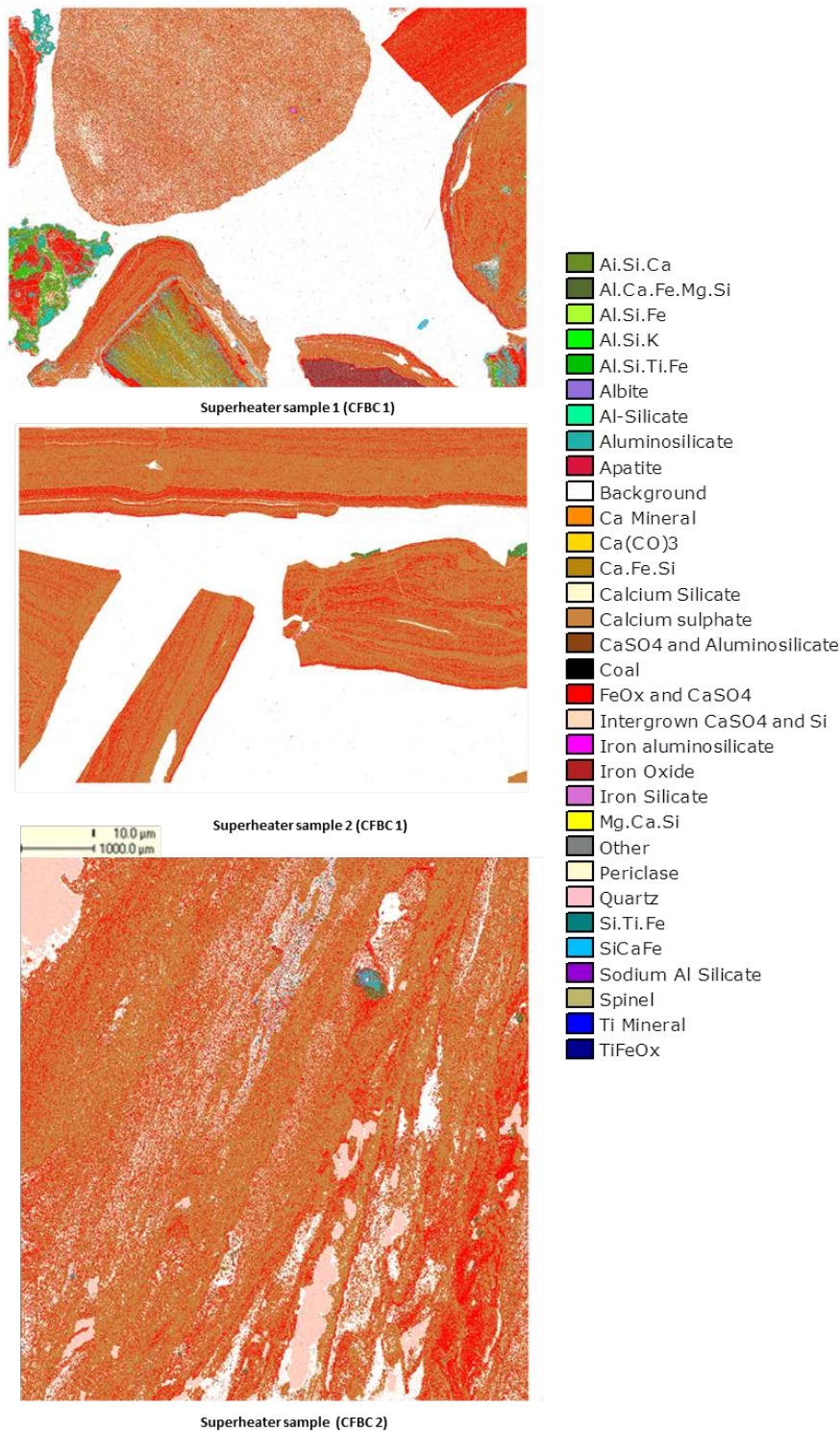


Figure 2 QEMSCAN images superheater deposits of CFBC 1 and CFBC 2

Based on the analytical results it appears that the CFBC 2 ashes were unusual in the dominance of anhydrite to the almost complete exclusion of other calcium phases such as calcite (CaCO_3), lime (CaO) and portlandite (Ca(OH)_2), a characteristic which has not been previously reported in the literature (3, 5). The CFBC 1 plant was selected for a comparative study as it is of a similar design, although the coal and limestone feeds have different characteristics. The results obtained from this study of the CFBC 1 plant confirm that CFBC 2 is unusual in the chemistry and mineralogy of the ashes, although the nature of the superheater deposits from both power plants is similar with respect to

mineralogy and texture. The possible factors which may contribute to these differences in ash characteristics and subsequent behaviour include variation in operating conditions, differences in coal properties and differences in limestone characteristics. From information supplied by the power station operators, it would seem that operating conditions of both power plants are similar so variation in operating parameters is unlikely to be a significant factor. The sulphur content of the CFBC 1 lignite is much higher than that of the CFBC 2 lignite, being almost 3.5 times greater. The implication of this difference is that a higher rate of limestone consumption would be required for effective desulphurisation to occur. Unfortunately, although figures for limestone feed rates were supplied for the CFBC 1 plant, they are not available for CFBC 2 so a comparison cannot be made. Variation in limestone characteristics may also be an important factor. As noted above, the modal grain size of the CFBC 2 milled limestone is only 12 μ m, considerably smaller than the modal value of 38 μ m obtained for the CFBC 1 milled limestone. Although not strictly comparable with respect to sample location, it is interesting to note that the particle size of anhydrite as determined by QEMSCAN for the CFBC 1 cyclone sample is 32 μ m and for the CFBC 2 back pass sample 10 μ m, both of which are similar to the modal size values of the respective limestone feeds. In contrast, the average anhydrite particle size in the air preheater, electrostatic precipitator zone and of the fly ash in the CFBC 1 samples is approximately 20 μ m, finer than the 40 μ m found in the economiser and fly ash samples of the CFBC 2 plant. This apparent coarsening in the back sections of the CFBC 2 plant suggest that winnowing of the finer anhydrite particles may have occurred by deposition of these fine particles on the boiler tubes.

Conclusions

This study of the CFBC 1 plant was undertaken in order to provide comparative data for the identification of the possible causes of ash deposition the CFBC 2 plant. Although the nature of the superheater deposits in both power plants is similar with respect to mineralogy and texture, both being dominated by iron oxide and anhydrite occurring as layers defined by grain size and relative proportions of the two minerals, the ash chemistry and mineralogy are both significantly different. The most notable difference is the occurrence of calcite, lime and portlandite in the CFBC 1 ashes and the relationship of these minerals to anhydrite. The CFBC 1 feed coal has a significantly lower total sulphur content. The CFBC 1 limestone has a greater level of impurities such as quartz and kaolinite but the Hardgrove index values are practically identical. However, there is a greater proportion of fine (10 μ m) particles in the CFBC 1 milled limestone. Slagging indices based upon ash chemistry and mineralogy of both coals indicated that they would have a similar propensity to form boiler deposits. Optical examination and QEMSCAN analysis both indicated that the strength of the deposits is related to the abundance and mode of occurrence of the anhydrite, abundant fine, interlocking grains imparting the greatest degree of strength to the deposit.

This study confirms that the CFBC 2 ashes are unusual in the occurrence of complete sulphation of the decarbonated limestone with no evidence of either the occurrence of intermediate phases such as calcium oxide or the presence sulphate reaction rims on decarbonated limestone as observed for the CFBC 1 ashes and commonly described in the literature.

A possible reason for this unusual behaviour is the high sulphur content of the CFBC 2 lignite which may result in complete sulphation of the limestone due to an insufficient feed rate. The significance of this factor cannot be evaluated as limestone feed rate data was not available for the CFBC 2 plant. Another possible factor is the greater proportion of fine particles in the milled CFBC 2 limestone which would react completely. This observation is supported by the occurrence of fine anhydrite particles in the CFBC 2 back pass sample and a subsequent increase in grain size in the back end of the boiler, suggesting that winnowing of the fine particles has occurred in the hotter sections of the back pass. This is in contrast to the relatively limited size variation exhibited by anhydrite in the CFBC 1 ash samples.

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