



Coal Comminution Characterization for Industrial Scale Circulating Fluidized Bed

a Research Program in the frame of
the International Energy Agency implementing agreement
for co-operation in the field of fluidized bed conversion
of fuels applied to clean energy production

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Acronym used in this report

EDF	= Electricité de France
IRC-CNR	= Istituto di Ricerche sulla Combustione, CNR
TU-VIENNA	= Institute for Chemical Engineering, Fuel and Environmental Technology, University of Technology Vienna
CHALMERS	= Department of Energy Conversion, Chalmers University of Technology
IET	= Institute of Engineering Thermophysics, Chinese Academy of Sciences
TSINGHUA	= Tsinghua University
CUMT	= China University of Mining & Technology

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EXECUTIVE SUMMARY

The management of the bed solids inventory and particle size distribution in fluidized bed combustors has long been recognized as a key factor for the efficient and trouble-free operation of industrial scale fluidized bed combustors. From a coal particle to an ash particle getting out the circulating loop, the particle size reduction factor can be hundred. Detailed pilot scale test observations related to the size of circulating particles and ash particles, show very different behaviours for different coals. In order to describe those behaviours related to a large particle size spectrum, a general frame work is proposed based on several concepts. The key concept is represented by the Primary Ash Particle Size Distribution (PAPSD). According to this concept, Primary Ash Particles are liberated from the carbon matrix under the combined action of Combustion and Primary Attrition of the mother fuel particles. By Primary Attrition it is meant here a complex of particle comminution phenomena closely associated with the progress of reactions (devolatilization/char combustion), along pathways and with mechanisms that are extensively discussed by Chirone et al. (1991). Further (Secondary) attrition of ash particles can occur after char combustion is complete and is not associated with the parallel progress of chemical reactions but to mechanical attrition only.

Those concepts are associated with coal characterization procedures in order to have information on the size reduction of a coal particle and the final size distribution of ash. The measurement obtained on a lab scale is validated by the analysis of some results of industrial-scale tests. In association with procedure definition, a qualitative approach allows prediction and inter-coal comparison. A modelisation work is also presented considering however that quantitative predictions of the particle size distribution inside a CFBC is still a far objective.

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1. Introduction

1.1 Importance of particle size distribution

The management of the bed solids inventory and particle size distribution in fluidized bed combustors has long been recognised as a key factor for the efficient and trouble-free operation of industrial scale fluidized bed combustors. Merrick and Highley (1974) pioneered studies on the subject. They highlighted that attrition phenomena, together with elutriation and fines recycle, play a key role in establishing ash loading and particle size distribution at steady state in the combustor. Their work was based on operating experience with a pilot scale bubbling fluidized bed combustor. Attrition of coals during fluidized bed combustion received since then deeper consideration by several research groups. The state-of-the-art on this subject and the analysis of the combined attrition-combustion pathways determining the fate of carbon until burn-off have been surveyed by Chirone et al. (1991).

Even stronger is the impact of the management of bed solids on the overall performance of circulating fluidized bed combustors. For any given solids hold up in the riser (which largely dictates the overall pressure drop across the riser), the particle size distribution of bed material influences the axial pressure profile, the solids concentration profile along the riser and, in turn, the bed-to-surface heat transfer coefficients along the riser (Wu et al., 1991; Leckner and Andersson, 1992; Wirth, 1995; Basu and Nag, 1996). Furthermore, the extent of axial solids mixing and, in turn, the degree of temperature uniformity along the riser are affected accordingly.

The ash management facilities can also be affected by fuel characteristics. The size distribution of the bed particles can be controlled to some extent by design measures, as have been clearly explained by Herbertz et al. (1989): the small particle side of the size spectrum is determined by the fractional efficiency of the gas-particle separator, whereas the coarse particle side of the spectrum is represented by particles found in the bottom bed, which has to be adjusted by bed drainage. Various types of bed material regeneration devices have been invented to remove the coarse inert particles and send the fine fractions back to the bed at the same time as the loss of unburned particles is minimized. The effective design of the cyclone is critically dependent on the ability to predict the particle size distribution of bed inert material (Muschelknautz and Muschelknautz, 1999; Bursi et al., 1999).

The control of particle size distribution by design measures (gas-particle separator and bed material regenerator) has to be complemented by information relative to the fuel. More knowledge on the performance of the particles after feeding into the combustor is needed : fragmentation and attrition of

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particles, and also the fuel dispersion. Such information should be gained under actual boiler operation conditions to complete experiences made in the laboratory and to form a basis for the validation of prediction models.

1.2 Aim of the research program

The present study deals with the assessment of the inventory and of the particle size distribution of solids establishing at steady state in a circulating fluidized bed combustor under the hypothesis that the bed consists of fuel ash only. The aim of this research program is to define a coal characterization procedure for combustion in industrial scale CFB in order to have information on the size reduction of a coal particle (comminution) and the final size distribution of circulating ash. The procedure is based on existing procedures developed a few years ago by co-workers of this program. The validation is done with the analysis of some results of industrial-scale tests. Associated with the procedure definition, a test storage policy allow qualitative prediction and an inter-coal comparison.

1.3 List of tested coal

Several coals of different rank and from different countries have been tested in this research program :

- Datong = Chinese bituminous coal
- Dayan = Chinese lignite
- Gardanne = French lignite with a high ash content
- Pingzhuang = Chinese low bituminous coal
- Polish = bituminous
- South-African = bituminous
- Wangpingcun mix = mixture of gangue and Chinese anthracite
- Xiazhuang = Chinese anthracite

Their analysis are detailed in the following table. For some coals, two sets of experiments have been done.

Coal name	Proximate Analysis (air-dried basis)					Ultimate Analysis (air-dried basis)				
	Moisture	Ash	Volatile	Fix Carbon	HHV (MJ/kg)	C	H	O	S	N
<i>Datong 1</i>	2,5	12,7	27,4	57,5	28,9	71,5	4,1	7,4	1,1	0,8
<i>Datong 2</i>	4,5	15,1	24,6	55,7	27,1	67,1	3,7	7,7	1,2	0,7
<i>Dayan</i>	20,0	11,8	31,3	36,9	19,6	51,5	3,5	11,8	0,3	1,0
<i>Gardanne</i>	5,8	25,0	41,3	27,9	18,5	49,6	3,4	12,5	4,3	1,4
<i>Pingzhuang</i>	7,4	28,6	21,3	42,7	19,1	52,6	2,5	6,7	1,6	0,6
<i>Polish 1</i>	2,3	17,0	29,2	51,5						
<i>Polish 2</i>	1,7	12,3	27,5	58,5		73,9	4,6	5,3	0,8	1,5
<i>South African 2</i>	7,3	14,6	22,6	55,5						
<i>Wangpingcun Mix</i>	1,3	52,6	5,1	41,0	14,0	43,1	0,7	2,1	0,1	0,1
<i>Xiazhuang</i>	1,4	29,3	9,0	60,4	24,4	61,2	2,7	0,9	3,6	1,0

Those coals have been tested in a lab scale apparatus and in a pilot scale CFBC according to the following list :

Coal name	Lab scale test	Pilot scale test
<i>Datong 1</i>	TSINGHUA, IRC-CNR, TU-VIENNA	IET
<i>Datong 2</i>	TSINGHUA	IET
<i>Dayan</i>	TSINGHUA	IET
<i>Gardanne</i>	TSINGHUA	CERCHAR
<i>Pingzhuang</i>	TSINGHUA	IET
<i>Polish 1</i>	IRC-CNR, TU-VIENNA	CHALMERS
<i>Polish 2</i>	IRC-CNR, TU-VIENNA	CHALMERS
<i>South-African 2</i>	IRC-CNR, TU-VIENNA	CERCHAR
<i>Wangpingcun mix</i>	TSINGHUA	IET
<i>Xiazhuang</i>	TSINGHUA, IRC-CNR, TU-VIENNA	IET

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2. Experimental observations on CFBC pilots

The analysis of coal comminution starts with experimental observations in industrial and pilot CFBC. After a rapid overview of the tests conditions, major test observations are reported.

2.1 Overview of pilot tests

In this research program, three CFBC pilots have been involved in coal combustion tests :

1. IET 1.5 MWth
2. CHALMERS 12 MWth
3. CERCHAR 1 MWth

Experimental observations on particle size inventory require specific information on pilot equipment, pilot measurements and test conditions. For a complete particle size analysis, the following elements should be gathered :

1. Startup material : particle size distribution and mass
2. Coal : particle size distribution and mass flow rate
3. Limestone : particle size distribution and mass flow rate
4. Combustion chamber pressure drop
5. Bottom bed particle : particle size distribution
6. Riser particle analysis : particle size distribution
7. Circulating particle analysis (under cyclone) : particle size distribution
8. Fly ash : particle size distribution and mass flow rate
9. Bottom ash : particle size distribution and mass flow rate

Furnace particle sampling requires a sampling probe adapted to hot conditions. The sampling process needs to be qualified to set the suction velocity, the probe head, etc. The duration of the combustion test should be long enough to reach a solid equilibrium and to get a negligible influence of the start-up material. The furnace hydrodynamic should be as simple as possible. For coals with a low ash content, or with a high sulphur content, the mass of lime compared to the mass of ash present in the bed could be high, which implies to analyse the limestone comminution at the same time as coal comminution. If possible, a combustion test without any limestone would simplify greatly the analysis. Some of those constraints are difficult to obtain especially in large pilots. Ash flow rate is difficult to measure. That's why several of the pilot tests done in the frame of this research program could not lead to a complete particle size analysis. Most of the following observations are done on the IET pilot which

appears to be the most convenient pilot to do such tests.

2.2 Solid concentration

2.2.1 Solid concentration in bottom bed

The solid concentration could be derived from the pressure drop measurement. In Figure 1, the solid concentration is reported for the combustion of five different coals ; the particle density is assumed to be 2500 kg/m³. The smallest average ash concentration in the bottom part of the furnace is only 0.005, which does not mean a dense bed at all, while the largest is over 0.2.

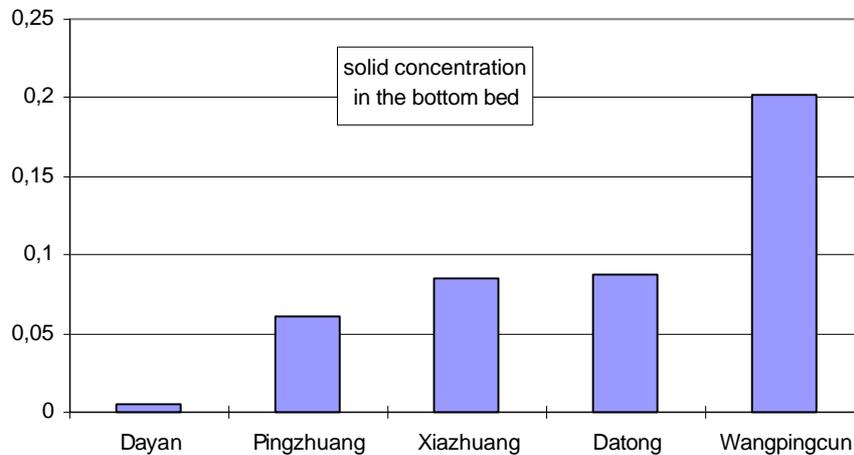


Figure 1 : solid concentration in bottom bed for 5 coals (IET).

2.2.2 Solid concentration in the riser

The differences in the average ash concentrations in the upper part of the furnace among the tested coals are very large. The smallest one is almost 8 times smaller than the largest one. The heat transfer which depends on particle concentration is consequently largely affected. It can be observed that the order of the five tested coals is almost the same as that of ash contents of the coals.

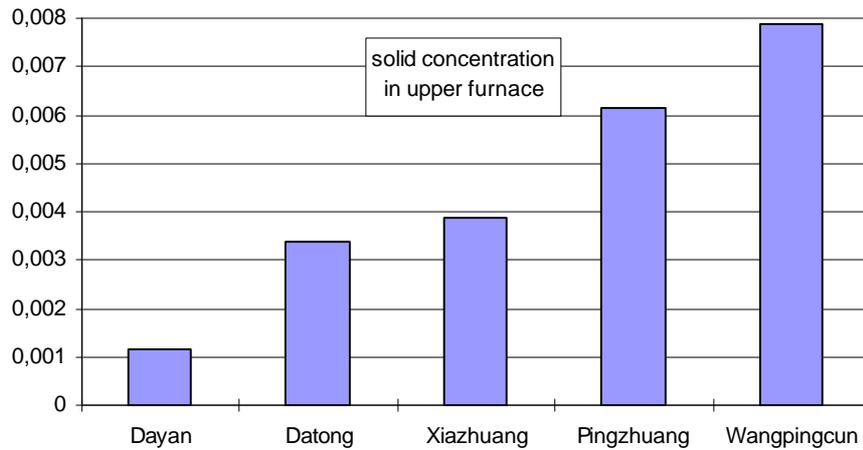


Figure 2 : solid concentration in the upper furnace for 5 coals (IET).

2.2.3 Pressure drop ratio

Combining the two previous observations, it is convenient to define the pressure drop ratio as the pressure drop in the upper furnace divided by the total pressure drop. For the experimental conditions, the maximum of this ratio (homogeneous vertical distribution of solid) is 89 % which is nearly reached by the *Dayan coal*.

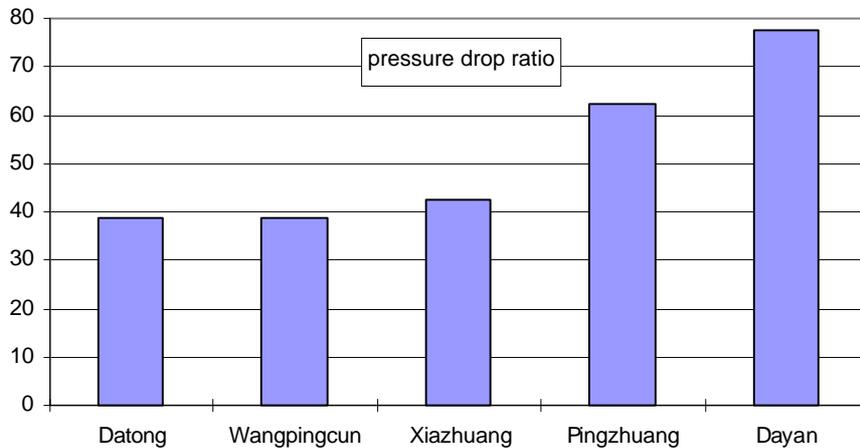


Figure 3 : pressure drop ratio for 5 coals (IET).

2.3 Temperature profile

There are distinct differences among the temperature profiles of the tested coals. Temperature profiles depend on not only combustion but also the ash concentrations and the ash sizes in the furnace. Temperature profiles are more uneven in the lower part of the furnace since it is a part from a dense bed where the secondary air is injected. The uniformity of the temperature profiles is in the order of

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Pingzhuang coal, Dayan coal, Xiazhuang coal, Wangpingcun Mix coal and Datong coal.

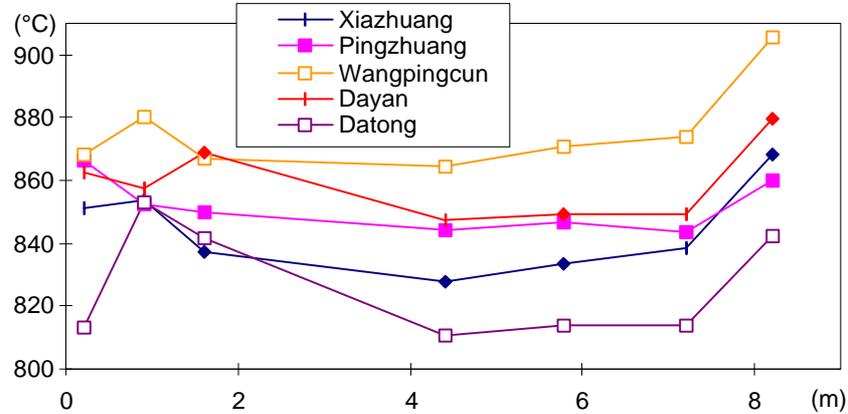


Figure 4 : vertical temperature profile for 5 coals (IET).

The temperature profile of *Datong 900 °C* and *Datong 850 °C* have the same shape which means that the combustion processes at the two temperatures have the same characteristics.

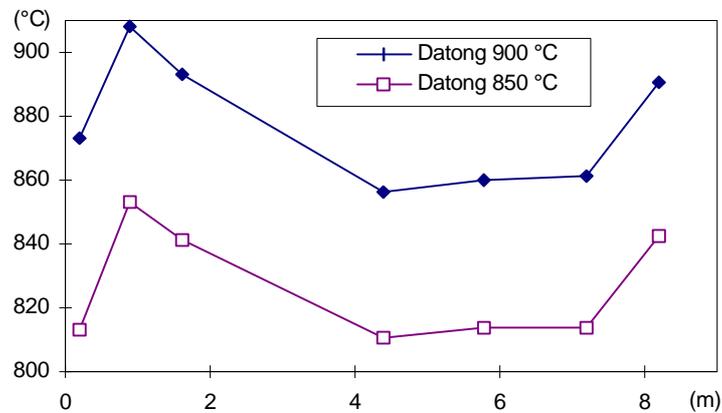


Figure 5 : the shape of the temperature profile is not influenced by the temperature (IET).

2.4 Combustion stability

Due to small thermal capacity of a pilot plant it is easy to see large differences of combustion stability among the tested coals. Combustion stability is influenced by coalification and volatile contents of the coal but also by ash concentration in the bottom of the furnace. The combustion stability order from stable to unstable is basically opposite to the order of the average ash concentrations in the bottom part of the furnace from large to small.

2.5 Fly ash ratio

The fly ash ratio is defined as the flow rate of fly ash divided by the total ashes flow rate. There are large differences of the ratio of fly ash to bottom ash among the tested coals.

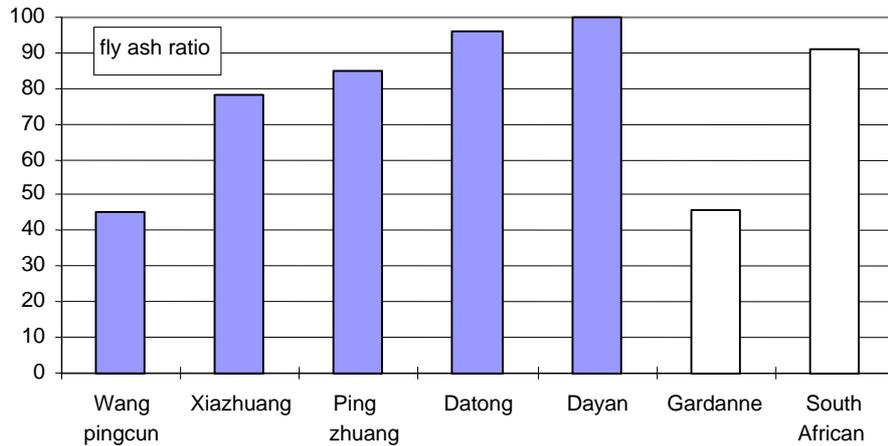


Figure 6 : fly ash ratio for several coals : IET (dark) CERCHAR (white)

2.6 Unburnt carbon

The differences of the carbon contents of fly ash among the tested coals are very large. Except *Datong coal*, carbon content in fly ash is larger as the coalification of the coal is deeper and H, O, volatile and inner moisture contents are smaller. Carbon content in fly ash of *Datong coal* is extremely large, so that the above analysis is not applicable to *Datong coal*. It is necessary to search the relationship between the characteristic of a coal and its combustion efficiency.

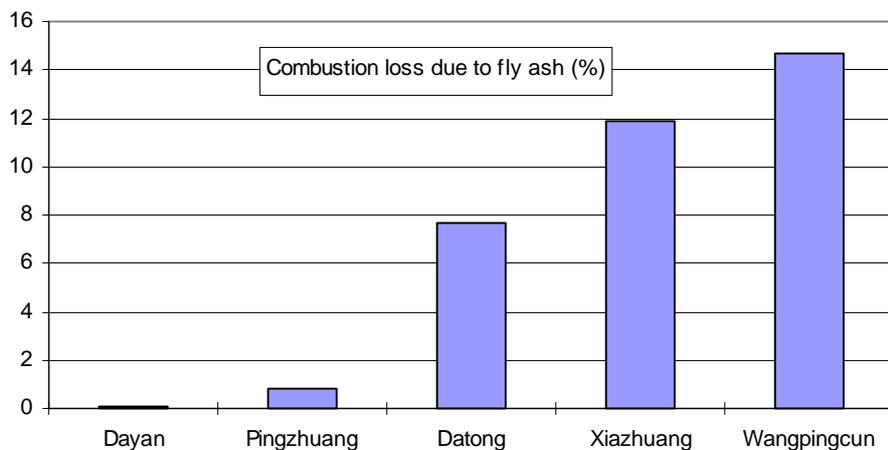


Figure 7 : combustion loss due to fly ash (IET).

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3. Concepts

The initial coal particle size entering a CFBC is approximately 5 mm. The particles coming out the circulating loop could be a hundred times smaller. In the mean time, the particles could circulate in the fluidized loop for a very long time. More than 90 % of those particles are inert. To describe and predict the comminution phenomena, a general framework built from several concepts is proposed hereunder.

1. Primary fragmentation
2. Carbon attrition
3. Primary ash
4. Inert attrition

3.1 Primary Fragmentation

The propensity of coals to break up upon injection into the bed is a consequence of internal stresses due to devolatilization and thermal shock. On the one hand the degree of fragmentation is strongly affected by the coal type and the coal particle size. On the other hand it is not affected by the oxidizing/inert nature of the fluidizing gas. Also the fluidizing gas velocity plays a minor part. Even if the emission of volatile is a necessary condition for the particle to fragment, the porosity of the material, the strength of the coal and its char control the extent of particle fracture. The feature of swelling coals to undergo a plastic stage during heat up is not able to relieve internal thermal stress or internal overpressure as a consequence of possibility of the failure of the external resolidified layer. However the shape of fragments, in particular the formation of spherical shaped fragments, is strongly influenced by coal plasticity.

The average size of particles present in the bed at the devolatilization stage is rather similar for the coals due to the compensating effects of a higher fragmentation probability and a smaller particle multiplication factor.

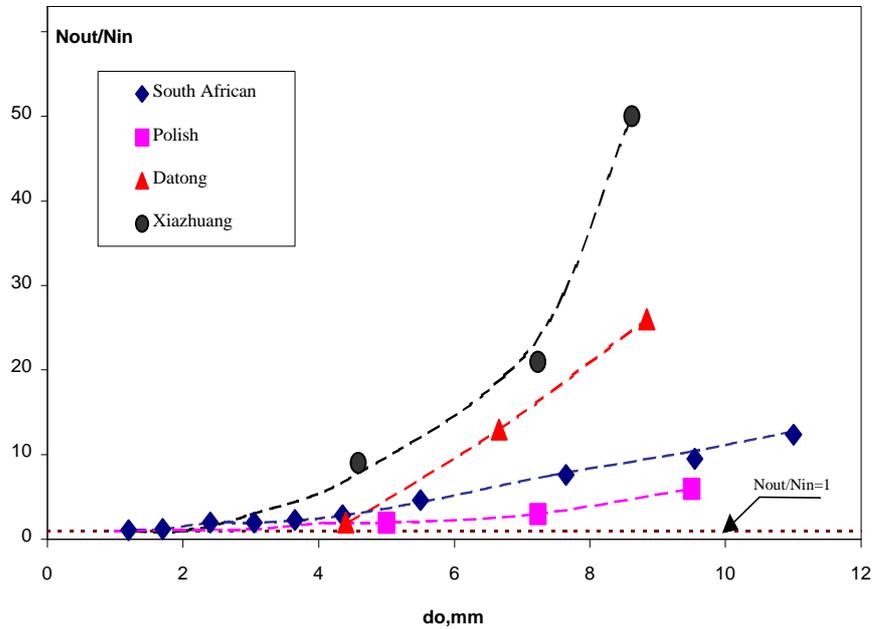


Figure 8 : Primary fragmentation. Particles multiplication factor vs initial particle size (IRC-CNR).

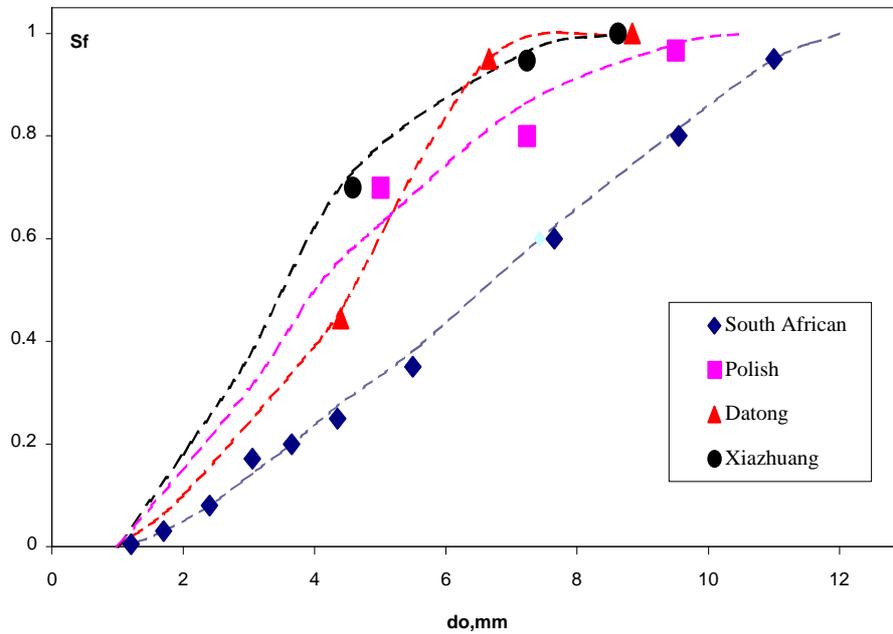


Figure 9 : Primary fragmentation. Fragmentation probability vs initial particle size (IRC-CNR).

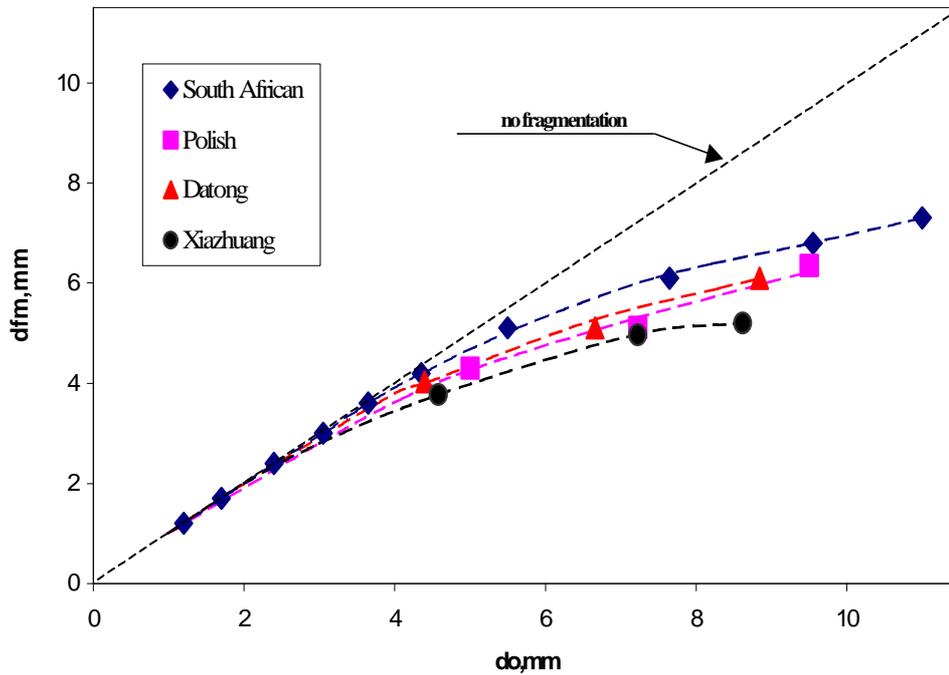


Figure 10 - Primary fragmentation. Average size of particles present in the bed after devolatilization stage vs initial particle size (IRC-CNR).

3.2 Carbon Attrition

When the carbon attrition rate is expressed per unit char particles external surface, a monotonically increasing trend is observed : a very rapid increase to the specific attrition rate is observed at the beginning ; a plateau is reached at intermediate carbon conversion degrees ; a final tail characterised by further increase of specific attrition rate is observed at late burn-off. This phenomenology led to the formulation of the Combustion-Assisted concept. According to this concept, attrition rate is dramatically enhanced by the progress of combustion. Internal burning enlarges pores, regenerates surface irregularities and, in turn, decreases the mechanical strength of the outer region of the particle, making it increasingly prone to failure upon surface wear. One consequence of the Combustion-Assisted Attrition mechanism is that the attrition constant k is a function of the oxidising conditions.

The validity of the equation $E_C = k(U - U_{mf}) \frac{W_c}{d}$ relating the carbon elutriation rate to bed carbon loading, has been proven with reference to a number of coals under bubbling fluidized bed combustion conditions. The dependency of k on the size of bed inert material has been assessed. The robustness of the experimental procedure for evaluation of the fuel-specific parameter k with respect

to the scale of the experimental apparatus has been positively checked. The same equation has been proven to apply to circulating fluidized bed combustion of a subbituminous coal char. The attrition rate constant k turned out to be dependent on the solids mass flux along the riser G_s .

The properties described here are illustrated by the following two figures where carbon attrition curves are reported for the *Polish* and the *South African coal* under different oxidising conditions. The curves have the same general trend. However some differences are : i) combustion efficiency is more sensitive to oxygen concentration in the case of the *South African coal* ; ii) less regular curves have been obtained in the case of *Polish coal*.

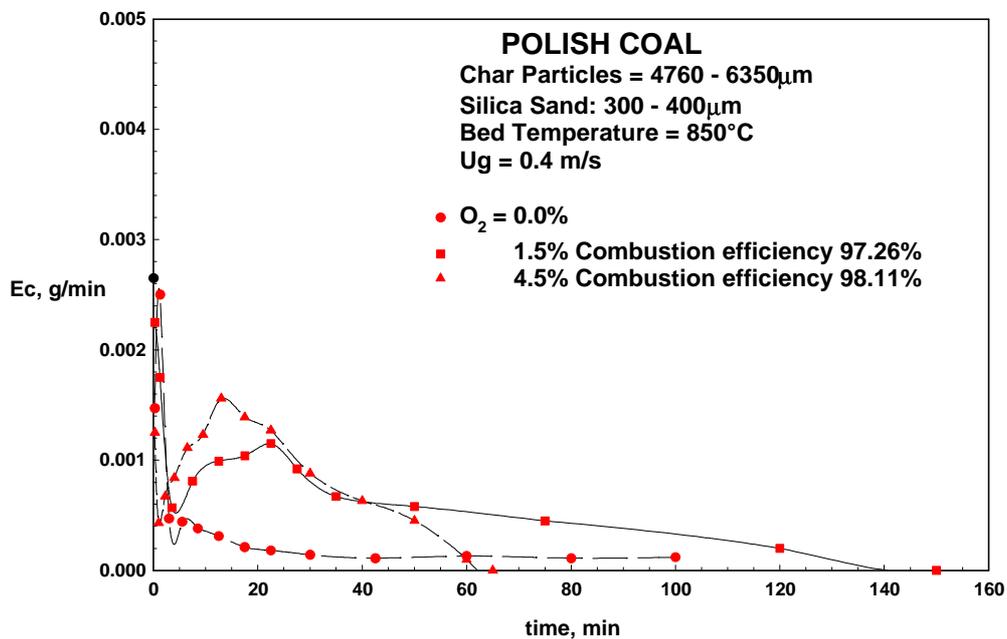


Figure 11 : characteristic attrition curves for the Polish coal (IRC-CNR)

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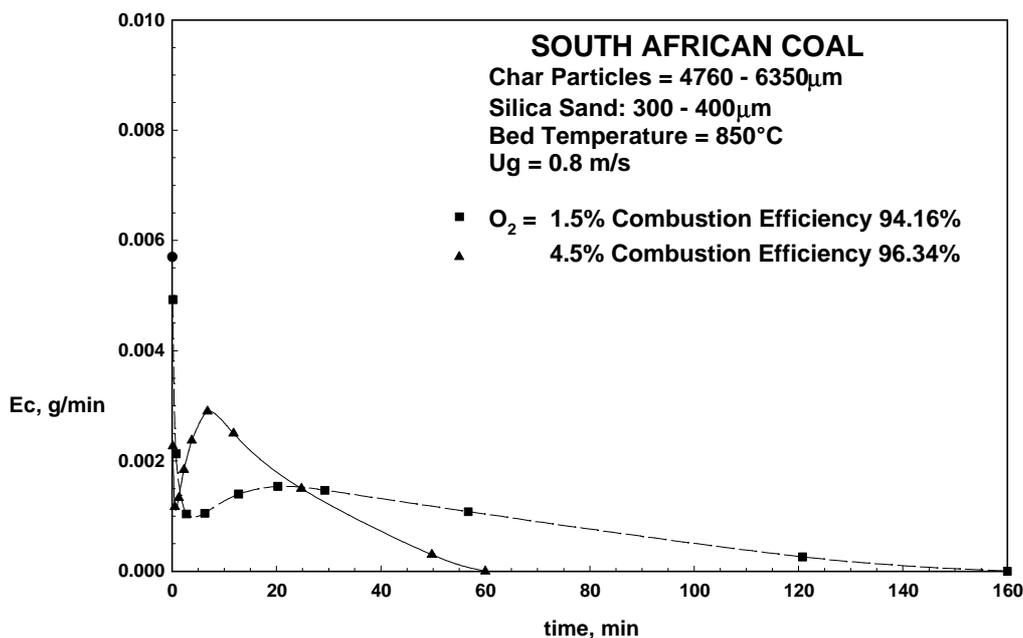


Figure 12 : characteristic attrition curves for the South African coal (IRC-CNR).

3.3 Ash formation

The comparison between the inert content of a coal particle and the ash obtained after combustion of that coal in a CFBC pilot brings some understanding on the ash formation mechanisms. The study presented here includes the qualitative and quantitative analysis of main mineral matters under both the reflected light and the transmitted light, using polarising light microscope and the X-ray diffraction analysis. The following description is based on the analysis of five Chinese coals of different rank before and after combustion.

In analysed coal particles, the minerals observed are mainly clay minerals, pyrite, quartz, carbonate minerals, rutile. The occurrence of clay minerals with various size fractions are quite different. Those less than 60 μ m in size are predominately disseminated in organic matrix, the minerals with the size range 60-100 μ m normally filled the cell cavity or occurred as small nodules, minerals ranging from 100-300 μ m occurred as large nodules or lenticules and the minerals more than 300 μ m occurred as large nodules long-bar shape or large irregular lumps. Pyrites are only observed in the small range (typically 0-250 μ m). The pyrites less than 60 μ m in size are predominately distributed in clay minerals while in the size range 50-100 μ m and 90-250 μ m, they occur as small and irregular lumps. Quartz occurs in the form of excluded particles, normally less than 250 μ m in size.

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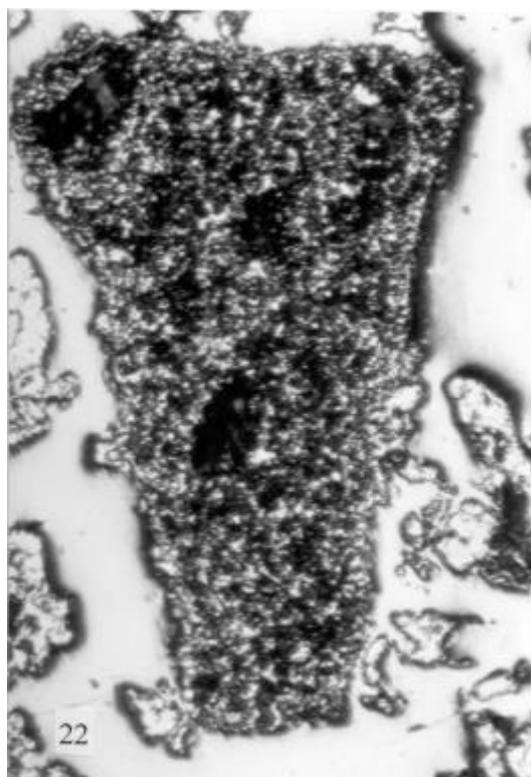


Figure 13 : coal minerals : clay, reflected light, single polar (x160) (CUMT)

Obvious changes can be observed between the occurrence of the minerals in coal and the mineral matter in ash after coal combustion, they can be described with the following aspects : after combustion, the original minerals in coal did not occurred as single particles, but coalesced together. In addition, rapid cooling normally caused the formation of glassy materials, and most particles are composed of less amount of crystals and large amount of glassy materials, so the analysis of the mineral of the ash samples was carried out by identification of the different particles. In this experiment, under the transmitted light of polarising microscope, five kinds of particles were recognised, namely clayey particles, quartz, mineral aggregate, ferruginous particles and unburnt carbon. Mineral aggregate, glassy material and ferruginous particle are typically new materials.

After combustion, the original clay disseminated in coal matrix or filled in cell cavity has changed, most of clayey particles occurred as irregular lumps, only a little of them was encapsulated into the unburnt carbon. The percentage of mineral particles is not evenly distributed in each size fraction. The number of the clayey particles and mineral aggregates in coarser fractions is more than that in finer size fraction. The mineral aggregates are new material obtained after combustion. They are composed by a great number of small particles such as quartz or clayey materials, maybe originated from the coalescence of the disseminated minerals in coal during combustion. Quartz occurred as irregular

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single particles. Due to clay minerals decomposition, the percentage of quartz particles in ash is more than that of quartz particles in coal at the same size fraction. Ferruginous particles are an iron oxide formed with pyrite lost sulfur after coal combustion. They enriched in finer fractions, normally less than 120 μm occurred as small excluded round particles or distributed in clayey particles.

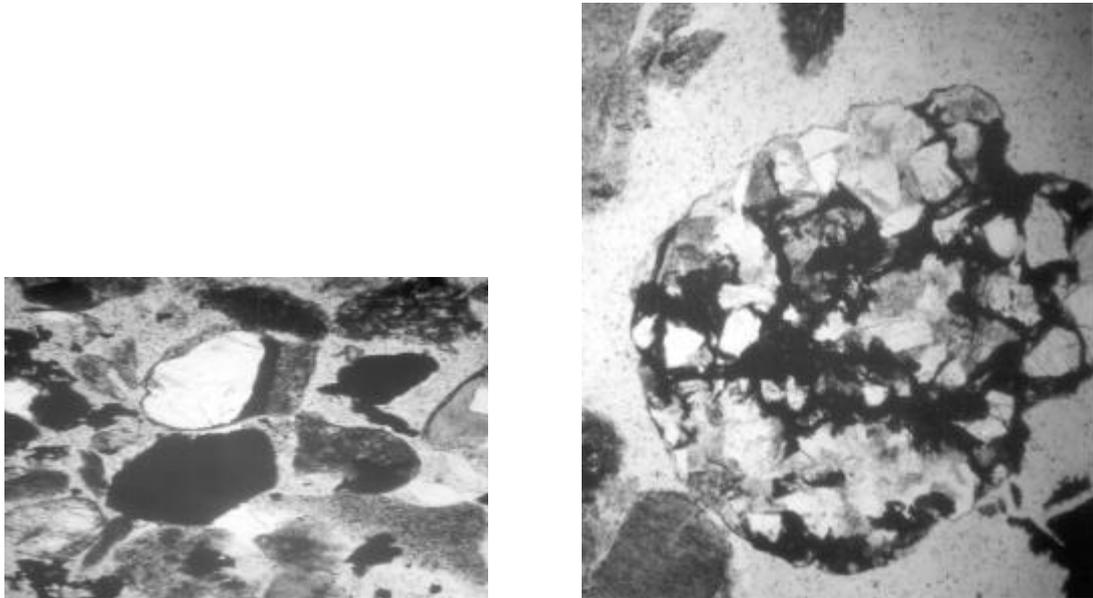


Figure 14 : Ash particles. left : unburnt carbon and quartz, right : mineral aggregate, transmitted plainlight (x100) (CUMT)

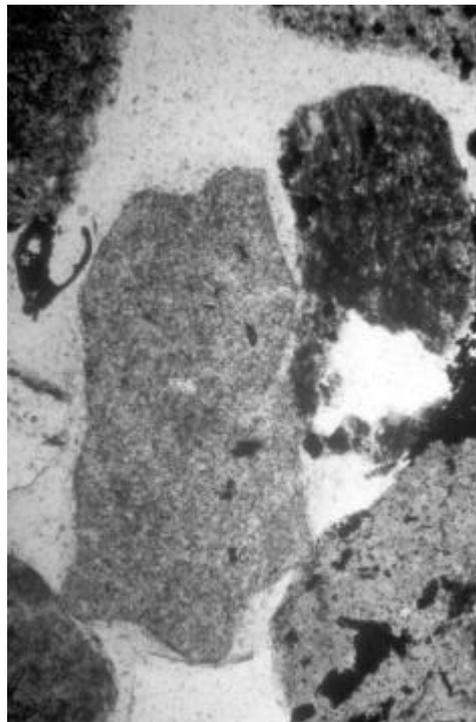
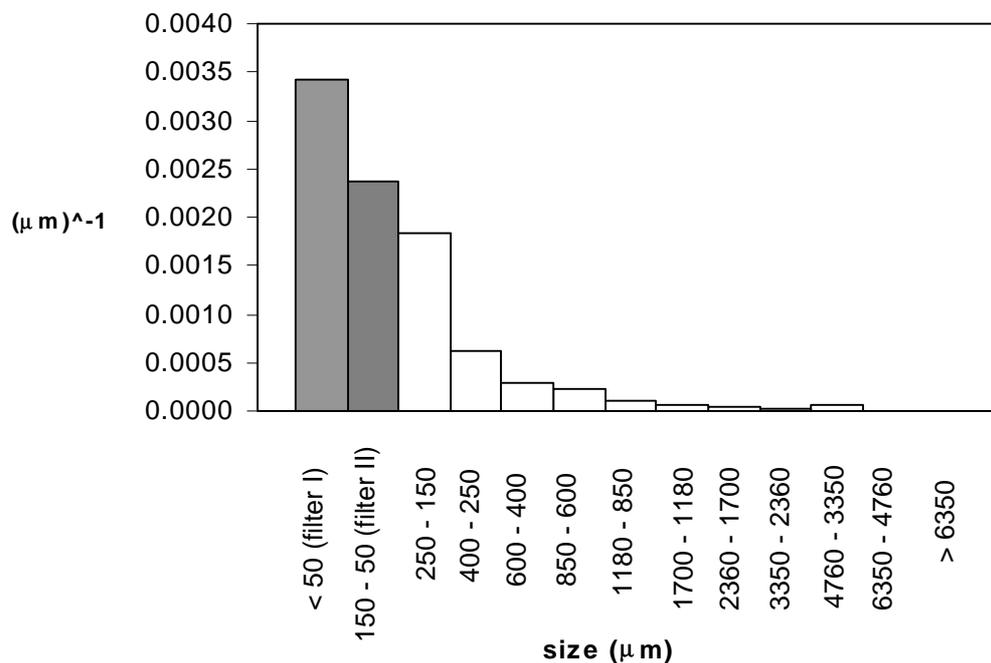


Figure 15 : clayey particle transmitted plainlight (x100) (CUMT)

3.4 Primary Ash

The key concept in the proposed approach is represented by the *Primary Ash Particle Size Distribution (PAPSD)*. According to this concept, Primary Ash Particles are liberated from the carbon matrix under the combined action of *Combustion* and of *Primary Attrition* of the mother fuel particles. *Primary Attrition* means here a complex of particle comminution phenomena closely associated with the progress of reactions (devolatilization/char combustion), along pathways and with mechanisms that are extensively discussed by Chirone et al. (1991). The concept of Primary Ash can be defined as ash obtained after complete combustion of coal particles and after a sufficient mechanical stress to get ashes as individual particles. Primary ash is therefore a characteristic image of the inert content of coal.

PAPSD of SOUTH AFRICAN coal



PAPSD of POLISH coal

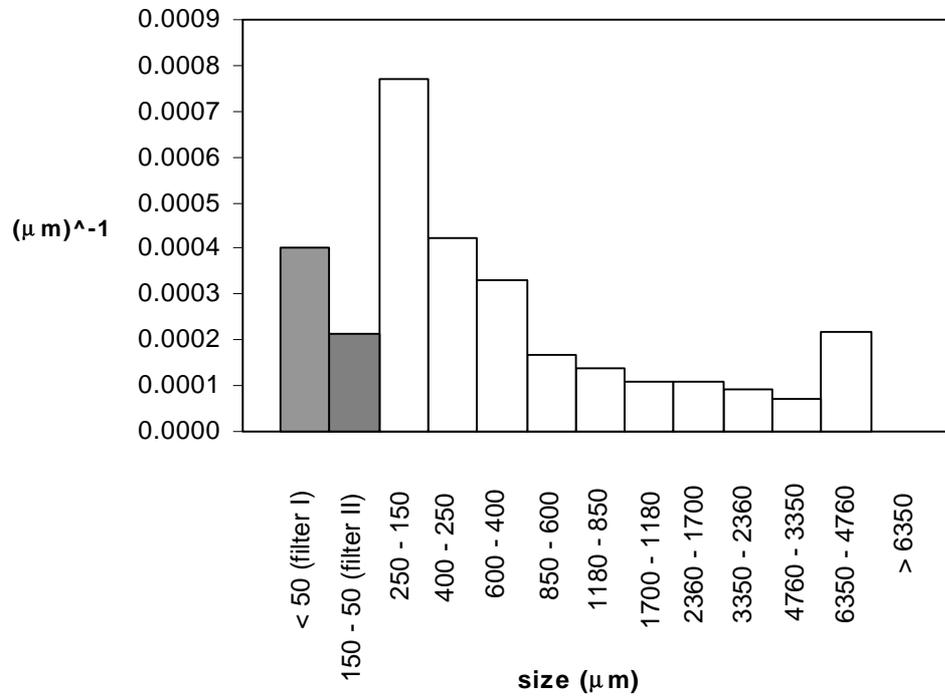


Figure 16: PAPSD of the Polish and South African (IRC-CNR)

3.5 Secondary Attrition

Secondary attrition of ash particles can occur after char combustion is completed and is not associated to the parallel progress of chemical reactions but to mechanical attrition only.

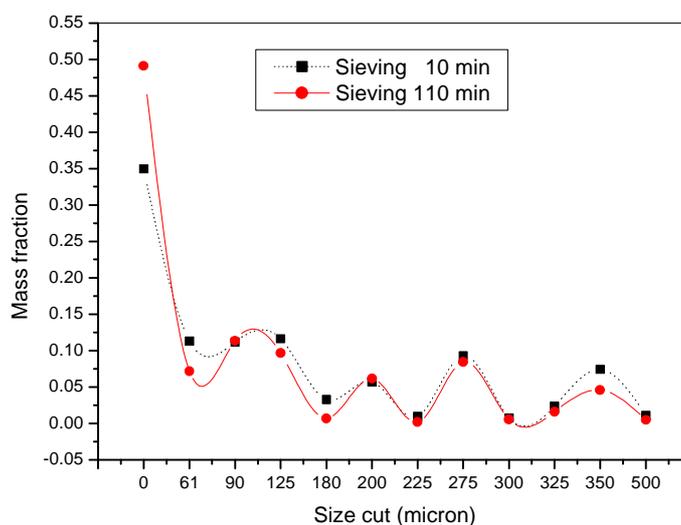


Figure 17: Xiazhuang ash particle size distribution at different times (size cut: 0-450micron, sieving amplitude: 3.0mm) - (TSINGHUA).

An obvious mass shift, which is from large size component to small one, occurs with the time pass-by. The previous text described the attrition phenomenon, showing that attrition resulted in a relatively large mother particle and a group of fine particles. And for Xiazhuang coal sample, the change of the mass fraction of mid size cuts (180-300 microns) could almost be ignored.

Moreover an important conclusion can be deduced from these figures. It can be observed that the mass amount of mid size cut did not increase. Contrarily the middle size cut also contributes to the generating of fines. This strongly supports the abrasive mechanism hypothesis of the attrition process in a fluidized bed.

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4. Methodological results : experimental procedure

4.1 Primary Fragmentation Characterization

The characterization of raw fuels fragmentation upon fast heating up and devolatilization that follow particle injection into the hot fluidized bed are based on devolatilization experiments in a bench-scale basket-equipped fluidized bed reactor (40 mm I.D.) (Chirone and Massimilla, 1989). The reactor are operated batchwise with respect to the fuel charge using nitrogen as fluidizing gas.

The primary fragmentation of each coal is studied by iterating several times (30 or more in order to get statistically significant experimental results) the following sequence:

1. The wire mesh basket is immersed into the bed, resting on the gas distributor, before fuel feeding;
2. One coal particle is fed to the combustor;
3. About three minutes after coal particle injection, the basket is raised about 40 cm above the bed surface (that is in the relatively cold freeboard section). The devolatilized particles (mother particle + fragments) are cooled down by injecting nitrogen to prevent combustion as particles are exposed to ambient air.
4. Particles are extracted from the reactor by means of the basket. The number of residual particles and their size are quantitatively characterized. The shape of fragments is qualitatively classified.

Experiments are carried out under inert conditions, i.e. by fluidizing the bed with nitrogen, at a gas velocity of 0.8 m/s. Coal particles belonging to the size ranges 9.5-6.35, 6.35-4.76 and 4.76-3.35 mm have been tested. Silica sand in the size range 0.3-0.425mm (true density: 2560 kg/m³; minimum fluidization velocity: 0.045 m/s @850°C) was used as inert bed material.

Experimental data have been worked out to calculate the primary fragmentation parameters : the particle multiplication factor ; the probability that a particle would undergo fragmentation ; the mean size of particles (unfragmented particles plus fragments) found in the bed after devolatilization.

4.2 Char Attrition characterization

Char attrition is characterized by feeding batches of pre-devolatilized char particles into a 40 mm ID fluidized bed equipped with a special two-exit head to collect particulate in the flue-gas (Chirone et al. 1985). Attrition is characterized in batchwise experiments by measuring the time-resolved carbon

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elutriation rates. These are monitored by operation of the two-exit head, i.e. by connecting alternatively the two exits. Using several filters in sequence, carbon emission within the combustion time after char is charged into the bed, is obtained. To keep oxidizing conditions substantially uniform during the combustion time batches of about 2g of fixed carbon are used in each run. Operative conditions at which experiments are carried out are listed in the following table :

Operative variables of the combustor	
Bed Temperature, °C	850
Fluidizing Velocity, cm/s	40, 80, 120
% O ₂ in Inlet Gas	0, 1.5, 4.5
Size distribution of the batches of char tested	
Particle size, mm	6.35 - 4.76

Table 1 : experimental conditions for attrition characterisation.

4.3 Primary Ash characterization

4.3.1 Primary Ash characterisation in oven

An electrically heated oven and a cold sieving shaker are used to carry out the experiment. The procedure uses 50 g of coal particles with a size range 2-2.5 mm. The coal is burnt at 850°C in an oven in the absence of any external mechanical forces which might affect the formation of ash. The shape of the ash particles obtained is similar to the original coal particles if they are kept untouched. However, if any mechanical forces are active, these loose ash agglomerates were destroyed. Parts of fine ash particles are found on the surfaces of larger ash particles.

The ashes produced in oven are put in the sieving machine for attrition tests. The sieving done at certain sieving conditions only destroyed the loose ash agglomerates and separated the ash particles from each other. A complete separation by sieving is not possible. Fine ash particles usually stick on larger ones. However, the amount of mass might be low and the PSD obtained is still representative for the coal. The sieving time is 30 minutes.

This procedure has been developed and applied by TSINGHUA and TU-VIENNA and is referred hereafter respectively as PAPSD 1 and PAPSD 3.

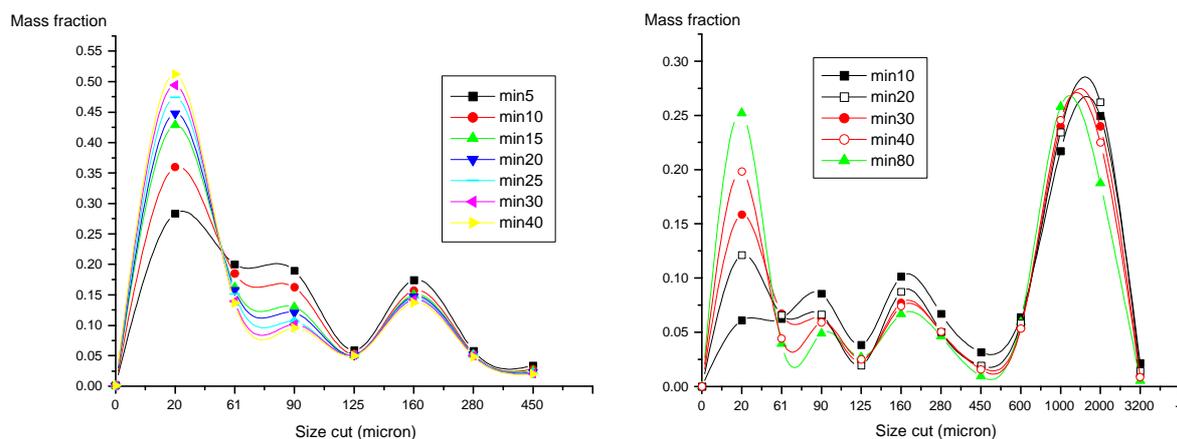


Figure 18 : Gardanne coal, Size cut:0-450 microns. Sieving amplitude:1.5mm / Gardanne, Size cut:2000-3200 microns. Sieving amplitude:1.5mm (TSINGHUA)

4.3.2 Primary Ash characterisation in a bubbling bed

A procedure for the experimental determination of the PAPS in a bubbling bed is described hereunder :

Apparatus: The apparatus consists of a 40 mm ID stainless steel combustor, 1 m high, electrically heated. The lower section of the combustor is filled with ceramic rings and acts as gas preheater. The gas distributor consists of a perforated plate with 55 holes of 0.5 mm diameter in a triangular pitch. The combustor is equipped with sintered brass filters at the exhaust for collection of elutriated particles. Monitoring of gas composition at the exhaust is accomplished by on line CO, CO₂ and O₂ analyzers. Prior to the test, a cylindrical wire mesh basket (40 mm outer diameter, 400 µm mesh size) is located just above the gas distributor. Then the combustor is charged with about 150 g of silica sand in the size range 125-150 µm.

Procedure: The procedure consists of the following sequence of steps:

1. Devolatilization (in nitrogen) followed by combustion (in air) of a coal sample (about 3g) until complete burnout. Gas superficial velocity is kept at the value U_1 slightly larger than the incipient fluidization velocity of the bed. Combustion is carried out at 850°C. Collection at the exhaust and further characterization of fly-ashes is performed after this stage.
2. Elutriation of ashes from the bed at gas superficial velocity U_2 larger than U_1 but smaller than the terminal velocity of bed inert material. Collection and further characterization of elutriated material is performed after this stage.

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3. Extraction of coarser ($d > 400 \mu\text{m}$) bed ash by means of the wire mesh basket followed by discharge of the remaining bed material (sand+ash). Separation of ash material from the sand by sieving and its further characterization is performed after this stage.

In order to collect significative quantities of ash samples, steps 1-3 are iterated several times (typically six), and both elutriated fines and material withdrawn from the bed cumulatively subjected to further characterization.

The operating procedure is optimized considering the following requirements:

- a) Limited surface wear is beneficial to overcome weak particle-to-particle forces (typically van der Waals forces) that keep together fragments otherwise disconnected.
- b) Surface wear must not be exceedingly large to give rise to extensive secondary attrition of ash particles.
- c) Combustion time must be small in order to minimize the occurrence of secondary attrition of ash particles during burn off.
- d) The coal/char particle temperature must not be high to the point of giving rise to ash sintering.

A trade off between conflicting needs a) and b) has been established by operating at moderate gas superficial velocity during Step 1 ($U_1 = 0.06 \text{ m/s}$) using relatively fine bed material (silica sand with particle diameter in the size range $125\text{-}150 \mu\text{m}$). At the same time U_1 was selected in orders to elutriate carbon fines smaller than the typical cyclone cut-off size in CFBC, thus removing fine particles that would be rapidly lost in a real combustor. During Step 2 the combustor acts like an elutriator, with the aim of collecting at the exhaust ash particles (not elutriated during Step 1) whose sizes fall within the size interval of the bed inert material. At the same time, inert bed material must not be elutriated to a significant extent (accordingly U_2 was set at the value 0.2 m/s).

Constraints c) (which would call for severely oxidizing conditions) and d) (that would rather call for mild oxidation) have been reconciled by operating in air at 850°C under conditions that apparently did not bring about ash sintering.

This procedure has been developed and applied by IRC-CNR and is referred hereafter as PAPS5.

4.4 Ash attrition characterization

The procedure for the hot attrition test consists of oven combustion and CFBC tests.

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1. Coal particles are put into an oven to produce the ashes under the following operating conditions : 100 g of coal is used for particles in the size range 2-2.5 mm, the oven temperature is 850°C, the duration of the test is 5 hours in steady-state operation.
2. When the ash is cooled down, it is put on the sieving machine for 30 minutes to obtain primary ash particle size distribution (PAPSD).
3. After the sieving the ash is fed into the CFBC under the following operating conditions : the bed temperature is 850°C, isothermal along the riser ; the fluidizing gas is air with a superficial velocity of 3 m/s ; the bed material is 200 g of silicon carbide particles in the size range 250-280µm ; the operation time is 60 min.
4. After 60 min operation, the gas supply is switched off and simultaneously, the heating shells are switched off. When the temperature of the CFBC falls down to the room temperature, all bed material : the ash and the silicon carbide are collected from the CFBC to measure final ash particle size distribution. Because the silicon carbide is a very hard and stable material, no fragments are produced from the silicon carbide after high temperature CFBC operation. After sieving, all silicon carbide is collected between 280µm and 250µm. All information about ash attrition is obtained, except the size range (280-250µm) which is mixed with silicon carbide. This data is estimated by averaging the data of the size range below (250-224µm) and above (315-280µm). In CFBC operation the finest ash could not be collected since it escapes as fly ash, so this ash mass loss is considered to belong to the size range which is smaller than 40µm (the smallest size of the used sieve is 40µm).

This procedure has been developed and applied by TU-VIENNA and is referred hereafter as PAPSD 4.

5. Qualitative results : a Coal Comminution Classification System

For a qualitative purpose, for example inter coal comparison relative to their comminution properties, the Primary Ash concept could give enough information. Although an additional work for the definition of a standard procedure is clearly needed, this concept is proved to be sufficiently robust to serve as a basis for a Coal Comminution Classification System.

5.1 PAPSD procedure comparison

For the same coal, the PAPSD measured by the different procedures are significantly different. This is observed in figure 19 and 20 where the PAPSD of the *Datong coal* and *Xiaozhuang coal* are reported. The first important reason why the differences are so large is the quality of the coal particles sampling. Due to the development stage of those procedures, no regular sampling methods have been applied before measurement. In current stage the procedure uses few number of particles which induces a larger influence on the sampling. The second reason is the difficulty to measure the particle size distribution of fine particles (below 50 μm) with sieves. The use of two different apparatus for small and large particles would also lead to some difficulties. Finally, the different operating conditions of the proposed PASPD measurement procedures are also responsible for discrepancies. There is a need for the definition of one standard procedure.

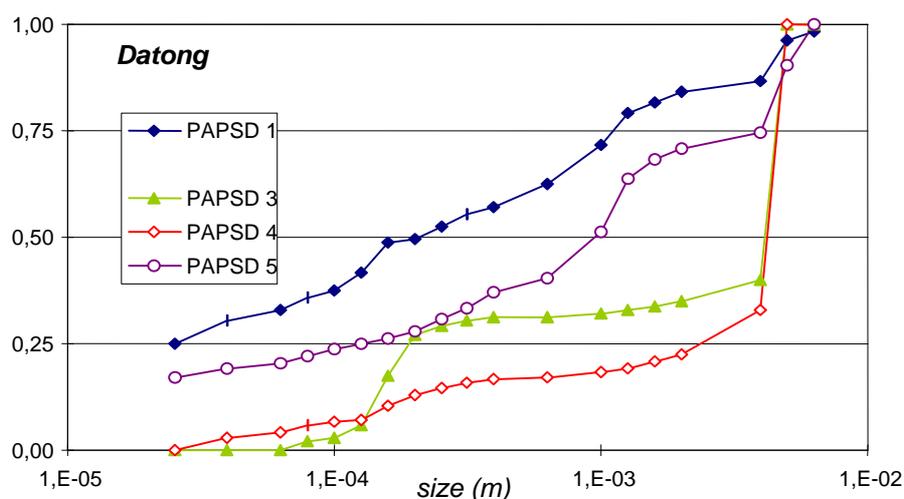


Figure 19 : PAPSD procedure comparison for Datong coal.
 PAPSD 1 & 3 : procedure using an oven (TSINGHUA & TU-VIENNA)
 PAPSD 4 : procedure using a hot CFBC (TU-VIENNA)
 PAPSD 5 : procedure using a bubbling bed (IRC-CNR)

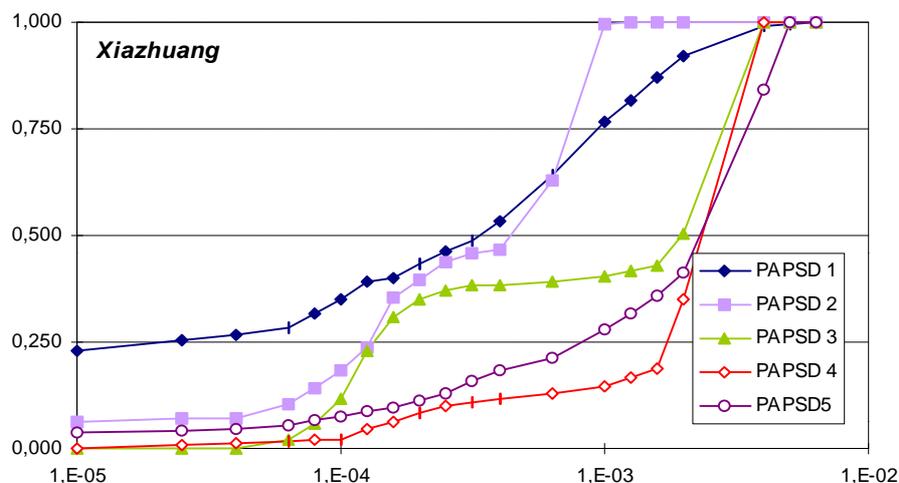


Figure 20 : PAPS D procedure comparison for Xiazhuang coal.
 PAPS D 1 & 3 : procedure using an oven (TSINGHUA & TU-VIENNA)
 PAPS D 2 : procedure using an oven and a cold CFBC (TSINGHUA)
 PAPS D 4 : procedure using a hot CFBC (TU-VIENNA)
 PAPS D 5 : procedure using a bubbling bed (IRC-CNR)

5.2 PAPS D coal comparison

For different coals, the measured PAPS D are significantly distinct which means that the inert content of different coals have different structure. However the curve shapes are quite similar for all the coals. In the medium size range, 0.1-1 mm, the cumulative Primary Ash size distribution is approximately linear with Log(size) with a coefficient similar for 4 coals. The *Pingzhuang coal* has a special distribution with few fines but many medium size range inert particles. The *Dayan coal* has a particularly high content of inert fines : 50 % of its inert content has a size below 100 μm .

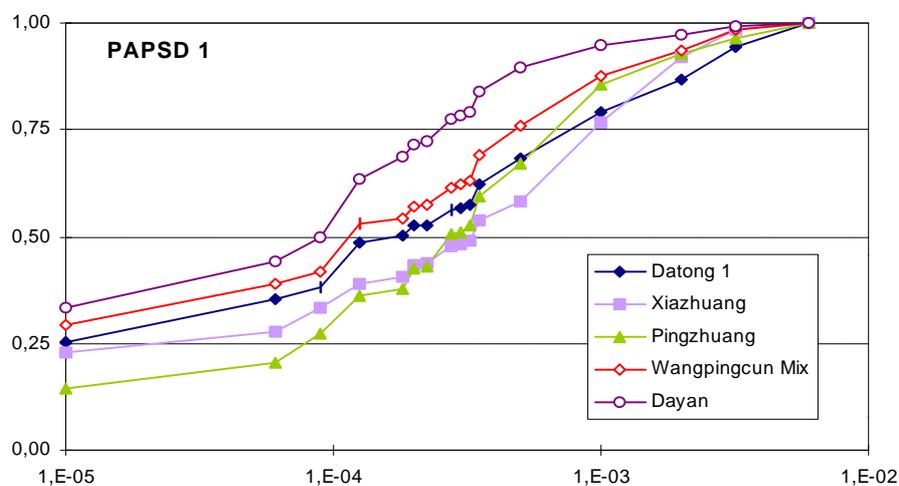


Figure 21 : PAPS D coal comparison for the oven procedure (TSINGHUA).

In spite of the discrepancies observed between the measurement of PAPS_D with different procedures, the measured PAPS_D are coherent. To assess this point, a comparison is made between the PAPS_D measured by TSINGHUA (figure 21) and the one by IRC-CNR (figure 22). In those two sets of measurements, the *Datong coal* and the *Xiazhuang coal* are the same. The relative position of the two curves is similar for the two graphs.

The previous remarks concerning the shape of the cumulative PAPS_D are also valid for those results. The *South-African coal* has a special inert content with many fines.

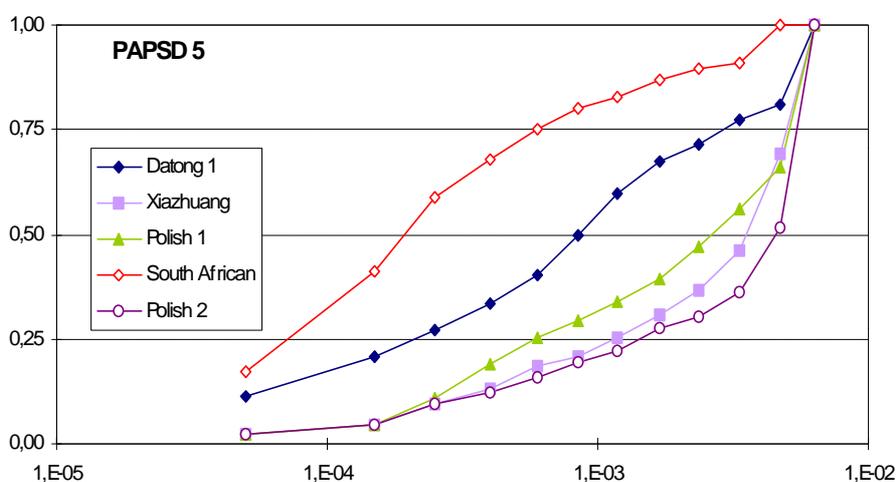


Figure 22 : PAPS_D coal comparison for the bubbling bed procedure (IRC-CNR).

5.3 PAPS_D and secondary attrition

After Primary Ash formation, the secondary attrition leads to the production of a large amount of fines. The classification between coals is not modified. The ability of PAPS_D to serve as a coal classification scale is thus confirmed.

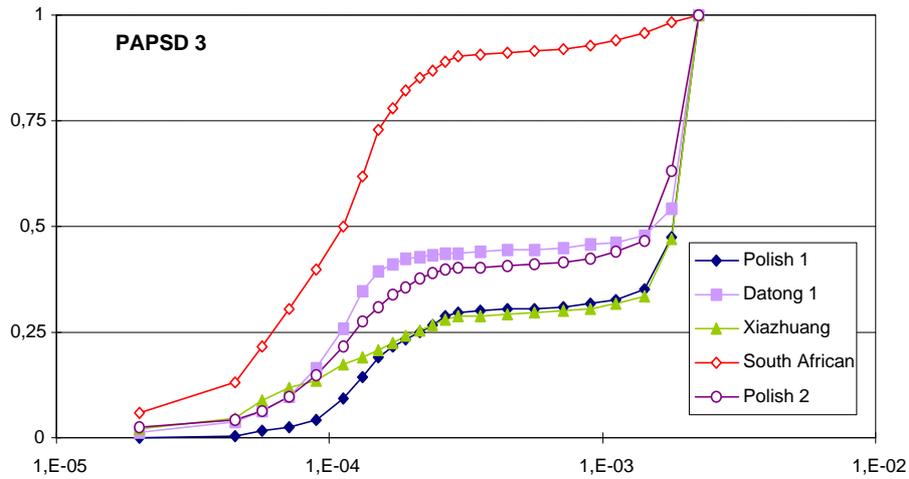


Figure 23 : PAPS D coal comparison for the oven procedure (TU-VIENNA).

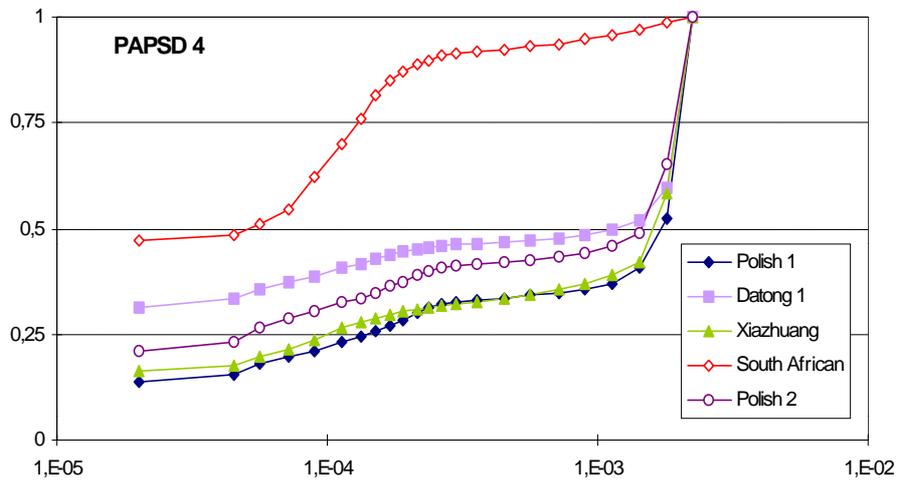


Figure 24 : PAPS D coal comparison for the hot CFBC procedure (TU-VIENNA).

5.4 A PAPSD based Coal Comminution Classification System

The PAPSD contains many information, some of them could be extracted to serve as a comminution classification system.

For a one-number classification system, an example with PAPSD-200, the proportion of Primary Ash whose size is below 200 μm , is detailed. The following table includes some information obtained on both lab and pilot scale for different coals. The coals are ordered from the coal which gives the coarsest Primary Ash (*Polish 1*) to the coal which gives the finest one (*South-African 2*). It can be observed that :

1. The order given by PAPSD-200 is roughly similar to the one given by the fly ash ratio.
2. The order given by PAPSD-200 is roughly similar to the order given by the pressure drop ratio.
3. It is not possible to make a link between PAPSD-200 and the carbon in fly ash. Although the loss of ignition from fly ash should depend on both the coal rank and the size of carbon fines, the first parameter appears to be dominant.
4. Detailed information on the relative position of fly ash ratio and pressure drop ratio can be obtained from PAPSD : the high pressure drop ratio for the *Pinzhuang coal* could be explained by the particular curve of its PAPSD (cf. §5.2). A one number classification system could not characterize this.

Coal name	Lab Scale			Unit	Pilot & Industrial Scale		
	PAPSD1-200	PAPSD3-200	PAPSD5-200		Fly ash ratio (%)	Pressure drop ratio (%)	C in the fly ash (%)
Polish 1		25	5	Chalmers			
Gardanne	40			Provence 4	58	29	< 5
Gardanne	40			Cerchar	46		
Xiazhuang	43	25	5	IET	78	42	25
Polish 2		38	5	Chalmers			
Pingzhuang	42			IET	85	62	2
Datong	52	43	20	IET	96	39	23
Wangpingcun Mix	57			IET	45	39	26
Dayan	71			IET	100	78	1
South-African 2		85	40	Cerchar	91		

Figure 25 : Example of a one-number comminution classification system with both lab-scale and pilot-scale results.

It is also possible to use a three-numbers classification system to get a more complete description of the comminution behavior of a coal. Three different size classes (0-100 μm , 100-800 μm ; 800-6000 μm) are defined according to their potential fluidization characteristics : fly ash, circulating ash, dense bed particles respectively. Each coal revealed a certain amount of ash in those size classes.

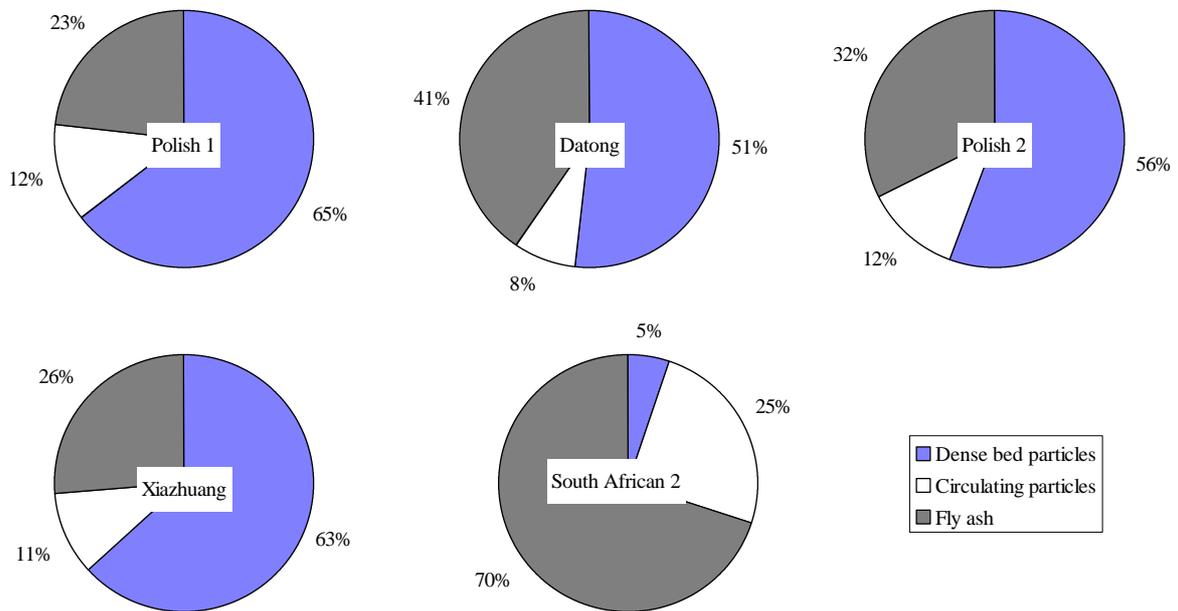


Figure 26 : Example of a three-number comminution classification system. Results based on PAFSD 4 measured in a lab-scale hot CFBC at TU-VIENNA.

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6. Towards quantitative predictions : modelisation procedure

The main goal of this modelisation work is to study the relative importance of the concepts previously introduced. Quantitative predictions of the particle size distribution inside a CFBC is still considered as a far objective.

6.1 Modelisation of the circulating loop

The circulating loop is considered here as one unique system to avoid to modelise the transfers between zones controlled by complex two-phase flow hydrodynamics and to focus on particle balance. Therefore the modelised system has one inlet which is the coal feeder and two outlets which are the cyclone exit and the bottom bed drain. The outlets are controlled by design and operation parameters (cyclone and bed drain efficiency) and by the internal circulation flow rate.

This simple approach has several drawbacks including to neglect the vertical segregation of particles regarding to their size and to suppose a uniform system for the modelisation of combustion. In spite of the evidence of extensive axial solids segregation, no established equation is available in the literature except correlation for the transport regime (Geldart, 1981).

In CFBC, the modelisation of the internal circulation rate is a difficult task since it needs detailed information on turbulent two-phase hydrodynamics with a large range of particle sizes. Here, a basic model is used for the circulation flow rate of particles of size d which is supposed to be equal to the weight $Wf(d)$ of particles of size d divided by a characteristic time of circulation $t_c(d)$. Introducing the cyclone efficiency $h_c(d)$, the cyclone outlet can be modeled by :

$$E_c(d) = Wh_c(d)f(d) / t_c(d) \quad (1)$$

$f(d)$ is the particle size distribution, W is the total weight of the carbon particles. The modelisation of the circulation time is only needed for particle which could effectively get out the circulating loop by the cyclone.

For bed drainage, the same basic model is used, where the emphasis is this time on large particles which are preferably collected. For those particles, the characteristic circulating time should be interpreted as the circulation time in the dense bed.

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$$E_d(d) = Wh_d(d)f(d) / t_d(d) \quad (2)$$

In this model, to study the influence of coal comminution characteristics on the particle population balance, the bed drainage efficiency is considered as a constant and the problem is solved to get the population balance inside the circulating loop. In real systems, the bed drain is an operation control parameter used to adjust the internal mass of circulating particles.

6.2 Carbon comminution

Considering the previous remarks and the previous work on that subject (see Chirone 1991 for a review), the carbon comminution equation for CFBC could be written as follows :

$$\begin{aligned}
W \frac{d(f(d)R(d))}{dd} = & E_0 f_0(d) - Wf(d) \frac{h_c(d)}{t_c(d)} - Wf(d) \frac{h_d(d)}{t_d(d)} \\
& - \frac{3W}{d} \left[\int_d^{d_{\max}} f(s)R_a(s)y(d,s)ds - f(d)R(d) \right] \\
& - W \left[\int_d^{d_{\max}} f(s)R(s)F(s)j(d,s)ds - f(d)R(d)F(d) \right]
\end{aligned} \quad (3)$$

With the boundary condition

$$f(d)R(d) = 0 \text{ for } d = d_{\max} \quad (4)$$

$R(d)$ is defined as the comminution rate. Following the assumption of Arena et al. (1983), $R(d)$ is made of additive contribution of the rate of shrinkage due to attrition and combustion.

$$R(d) = \frac{dd}{dt} = R_a(d) + R_c(d) \quad (5)$$

The right hand terms of equation (5) are respectively feed, cyclone and bed drain outlet, comminution gain and loss due to combustion and attrition, comminution gain and loss due to fragmentation. $y(d,s)$ and $j(d,s)$ are the distribution functions of fines and fragments produced by attrition and fragmentation.

The attrition comminution rate is modelled by :

$$R_a = -\frac{k_a}{3}(U - U_{mf}) \quad (6)$$

In CFBC, the fines produced by attrition may stay in the circulating loop regarding their size. Without precise knowledge on this size, all the fines are put in the finer class modelised $y(d_{\min},d)=1$.

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The combustion model uses the combustion rate per surface q given by the competition between kinetics and diffusional mechanisms (Basu and Fraser, 1991)

$$q = k_c [O_2]_g \left(1 - \frac{q}{k_d [O_2]_g}\right)^n \quad (7)$$

The combustion constants (k_c, n) are taken from the same reference. The combustion rate is modelled by :

$$R_c = -2 \frac{q}{r_c} \quad (8)$$

The fragmentation model uses the similarity hypothesis and the constants reported in Salatino (1987).

6.3 Inert comminution

The modelisation of inert behaviour during the whole combustion phase is an other difficult task since the particles are very different from the beginning of combustion to the end. A first distinction can be made between external or structural inert material and internal inert material. The characteristic size of the first type is of the same order as the initial coal particle ; therefore those ashes should go out the coal particle structure in the first stage of combustion. The second type of coal inert material could be seen as mineral inclusions inside the coal structure. Those ashes are liberated at the same time as the combustion occurs. But this behaviour has not been taken into account here.

The size of the liberate ashes is obviously a major parameter of coal comminution in CFBC. To answer this question the concept of Primary Ash Particle Size Distribution (PAPSD) introduced previously is used. The concept of Primary Ash can be defined as ash obtained after the complete combustion of coal particles and after a sufficient mechanical stress to get ashes as individual particles. Primary Ash is therefore a characteristic image of inert content of coal.

Considering those elements, an equation similar to equation (4) is written to describe the comminution of inert (see also Li et al. 1997) :

$$\begin{aligned} W' \frac{d(f'(d)R'(d))}{dd} &= E'_0 f'_{\text{PAPSD}}(d) \\ &- W' f'(d) \frac{h_c(d)}{t_c(d)} - W' f'(d) \frac{h_d(d)}{t_d(d)} \\ &- \frac{3W'}{d} \left[\int_d^{d_{\max}} f'(\mathbf{s}) R'(\mathbf{s}) \mathbf{y}'(d, \mathbf{s}) d\mathbf{s} \right. \\ &\quad \left. - f'(d) R'(d) \right] \end{aligned} \quad (9)$$

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where W' is the total mass of inert and $f'(d)$ the size distribution function. $R'(d)$ is the comminution rate of inert particles. From Primary Ash, inert attrition is supposed to be the only effective mechanism. The inert attrition model will be discussed later.

6.4 Simulation results

6.4.1 Base case definition

To set up the parameters of the model, a pilot test of the 1.5 MWth CFBC of the Institute of Engineering Thermophysics in Beijing is used (Qian et al., 2000). The set of data needed to run and to validate the model is listed bellow :

Carbon parameters :

- Fixed carbon,
- Initial coal size distribution (after devolatilisation and primary fragmentation),
- Comminution constants in particular combustion and attrition constants,
- Carbon in fly and bed ash.

Ash balance analysis :

- Coal ash content,
- PAPS_D,
- PSD of circulating particle [result],
- PSD of fly ash [result],
- PSD of bottom ash [result],
- fly ash over total ash ratio [result].

Operating and design parameters :

- Coal feed rate
- Oxygen concentration after volatile combustion,
- Characteristic circulation times,
- Efficiency of cyclone and bottom bed extraction.

The following figure shows, in cumulated form, the size distribution of circulating particle, the PAPS_D and the size distribution of fly and bottom ash. The sample circulating particles were extracted from the cyclone return leg. The PAPS_D was measured by IRC-CNR.

From experimental values, it is possible to solve equation (9) for the cyclone and the bed extraction efficiency. For example, if we suppose that $R'(d)=0$, the resulting particle size distribution of fly and bottom ash is equal, in their size range, to f_{PAPSD} . From figure 27, we can deduce that this hypothesis gives good indication on the particle size distribution of fly and bottom ash. It should be noted that those distributions are independent of $t_c(d)$ and $t_d(d)$. The cyclone and bed drain efficiencies here obtained are used in the rest of the study.

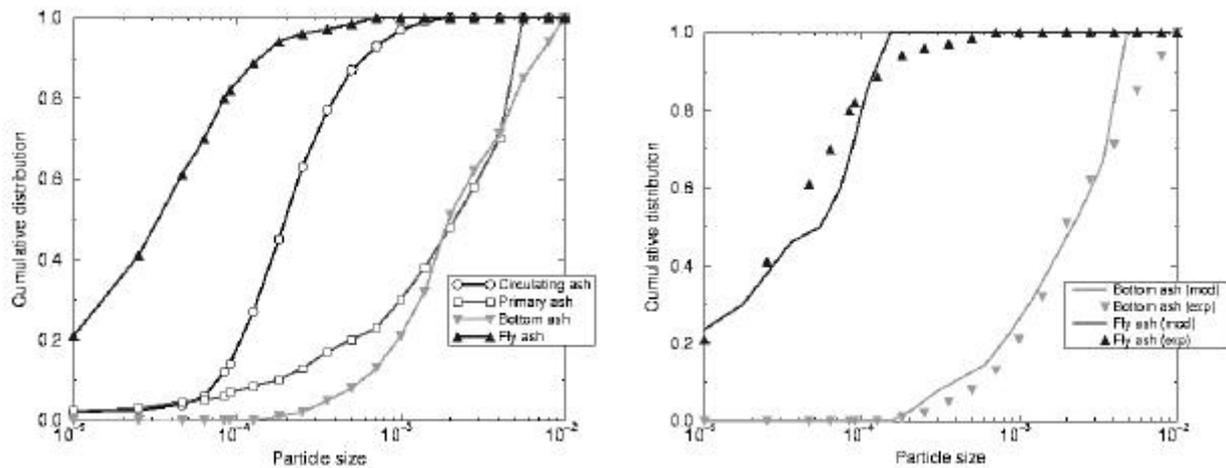


Figure 27: (left) experimental particle size distributions. (right) comparison between model and experimental fly and bottom ash with the assumption that the inert attrition rate is null.

6.4.2 Main simulation results

The main simulation results are listed below :

1. mass of circulating particles (furnace pressure drop)
2. particles size distribution in circulation loop
3. fly ash over total ash ratio
4. carbon concentration per class
5. carbon in fly ash
6. carbon in bed ash

Main Model Results	
Furnace pressure drop	4.7 kPa
Mean ash diameter	290 μ m
Carbon fraction	0.5 %
Inert fly ash ratio	10 %
Carbon fraction in fly ash	0.01 %
Carbon fraction in bottom ash	30 %

Table 2 : results of the reference case

The resulting particles size distribution of carbon and inert content is shown in figure 28.

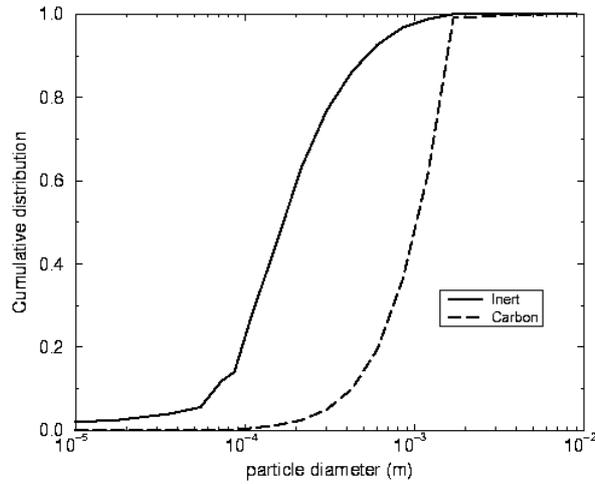


Figure 28 : equivalent size distribution of carbon and inert content of circulating particles.

6.4.3 Fly ash to total ash ratio

The fly ash to total ash ratio measured on the pilot (85 %) is different from the predicted one (10 %). This discrepancy could have several origins. On the one hand, on the pilot, the bed extraction operation is discrete while this operation is modelled as continuous. On the other hand, the resulting fly ash over total ash ratio may be interpreted as the relevance of an inert attrition mechanism.

Following Pis et al. (1991), we can modelled the inert attrition rate by :

$$R'(d) = -\frac{k'_a}{3}(U - U_{mf})d \quad (10)$$

with $k'_a = 1.6 \cdot 10^{-7} \text{ s}^{-1}$. The attrited fines are distributed through the y' function to the size ranges below $50 \mu\text{m}$. The relative importance of inert attrition versus cyclone or bed drain extraction could be estimated by computing a ratio of the characteristic times :

$$r(d) = \frac{W'}{E'_0} k'_a (U - U_{mf}) \frac{f'(d)}{f'_{\text{PAPSD}}(d)} \quad (11)$$

Since $r(d) < 1$ we can assume that this modelled attrition mechanism has little significance in our context. This analysis is confirmed by the simulation.

To study the influence of attrition, we use the model described by equation (6). With this model, attrition is very effective for small particles and the inert balance is significantly modified. For

example, the evolution of the equivalent furnace pressure drop and the mean circulating particles size distribution with the inert attrition constant have been reported in figure 29. When the attrition constant is increased, we observe an important decrease of the furnace pressure drop, since a large amount of mass of particle leaves the circulating loop by the cyclone. Therefore the fly ash over total ash ratio increases.

Nevertheless it has not been possible to find a set of model parameters coherent with experimental values. Our knowledge on inert attrition is insufficient to reach the experimental observations.

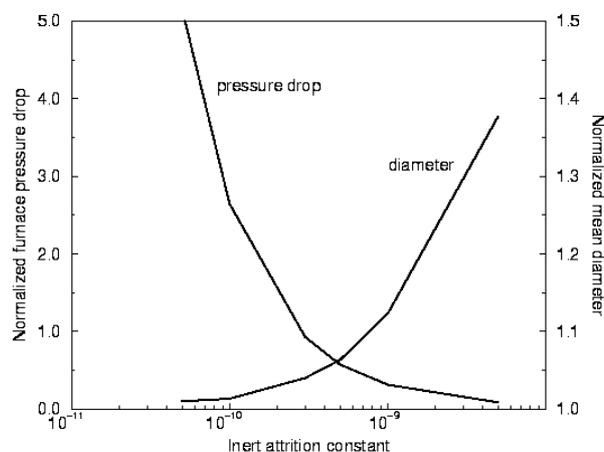


Figure 29 : the influence of attrition constant on furnace pressure drop and mean circulating particle diameter (cf. equation 7).

6.4.4 Carbon in ash

The carbon load in the circulating particles is low (0.5 % of the total mass) which is representative of experimental observations. Nevertheless, the predictions of carbon in ash are far from experimental values.

The carbon load in bottom-ash is over-estimated. In the present model, the kinetic of carbon combustion is used while the kinetic of Primary Ash formation is ignored : those two mechanisms should be coupled. As a consequence, the inert bed extraction is calculated on an equilibrium inert distribution while the carbon bed extraction is calculated on a distribution controlled by comminution. Too many big "carbon" particles are extracted.

The carbon load in bottom-ash is under-estimated. This discrepancy could have several origins. The carbon attrition could be responsible for a high level of carbon in fly ash. However, increasing the attrition constant has no influence on that value since fines would burn very quickly. This analysis is

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confirmed by simulation cases where the oxygen concentration is decreased : the carbon in fly ash increases sharply. The need for a model on char reactivity is therefore enlighten.

6.5 Modelisation conclusion

From this analysis, we can conclude that the present model could serve as a basis for a complete comminution model which would take into account the carbon and the inert content of coal. The use of the Primary Ash concept allows to predict favourably the size distribution of fly and bottom ash. However several disagreements between experimental and model data are observed. While the discrepancy concerning carbon-in-ash could be explained, the difference concerning the fly over total ash ratio is not fully understood.

7. Conclusion

The research program « Coal Comminution Characterization for Industrial Scale Circulating Fluidized Bed » aimed at defining a coal characterization procedure for combustion in industrial scale CFB, in order to get information on the size reduction of a coal particle and the final size distribution of ash. This goal has been approached with the help of the concept of Primary Ash and with the associated definition of a coal characterization procedure on lab scale apparatus. Also effectively usable, the definition of the experimental procedure still needs to be improved to get results more independent of unspecified operating conditions. For a qualitative analysis, the obtained results have proven to be enough accurate to make an inter-coal classification. Furthermore this classification can be favorably compared to pilot scale measurements such as fly ash or pressure drop ratios. For a quantitative analysis, the prediction of the particle size in the circulating loop is still considered as a far objective.

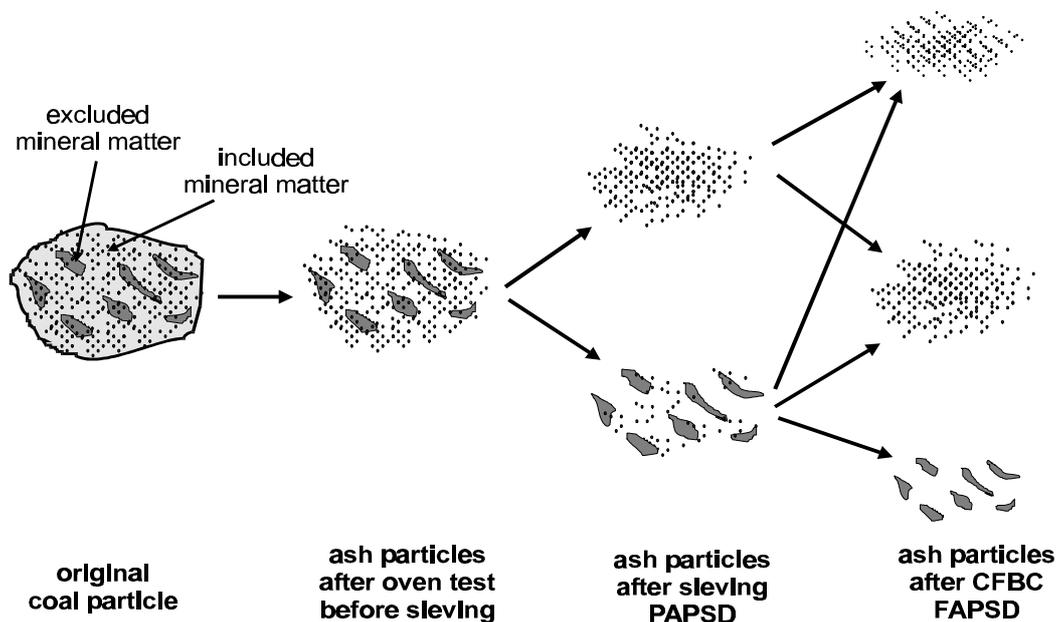


Figure 30: The ash formation mechanism and the concept of primary ash particle size distribution (PAPSD) and final ash particle size distribution (TU-VIENNA).

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