

# EFFECT OF BLENDING PET-COKE ON COMBUSTION CHARACTERISTICS OF PCI COALS<sup>1</sup>

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## Abstract

Low and high volatile bituminous coals are widely used in PCI blends, but petroleum coke has also been used, often as a cheaper substitute for low volatile coal. The paper aims to study the effect of blending petroleum-cokes with bituminous coals on the combustion characteristics under laboratory conditions. Both physical and chemical properties of coals and pet-cokes were characterized using a range of analytical techniques. The carbon structure of coal and char samples was characterized using X-ray diffraction. Coal and pet-coke blends were injected in an experimental drop-tube furnace (DTF) to characterize char characteristics and burnout under controlled combustion conditions. Char reaction rates were measured in an experimental fixed-bed reactor. The burnout of high volatile coal was higher compared to low volatile coal as expected, however the burnout and reaction rates could not be related to total volatile matter of coal blends. In general, burnout of pet-coke blends was lower than the burnout of coals. The carbon structure of pet-cokes is shown to have a strong influence on the burnout such that up to 10% blending of pet-coke did not show any appreciable effect on the burnout of coals.

**Key words:** PCI; Pet-coke; XRD; Blast furnace.

## Resumo

Carvões minerais dos tipos baixo e alto voláteis são largamente utilizados em misturas para PCI, mas coques de petróleo tem também sido utilizados, geralmente como substitutos para os carvões baixo voláteis. O objetivo deste trabalho é o de estudar a nível laboratorial o impacto causado pelo coque de petróleo nas características de combustão da mistura de PCI quando o mesmo é utilizado conjuntamente com o carvão mineral. As propriedades físicas e químicas dos carvões e coques de petróleo foram caracterizadas através do emprego de uma gama de técnicas analíticas. A estrutura do carbono das amostras de carvão e do "char" foi determinada por meio de difração de raios-X. As misturas de carvão e coque de petróleo foram injetadas em um forno experimental do tipo tubo de queda (Drop-tube furnace - DFT) a fim de determinar as características do "char" e da queima sob condições controladas de combustão. As taxas de reação do "char" foram medidas em um reator experimental do tipo plataforma fixa (fixed-bed reactor). Como esperado, a queima do carvão alto volátil foi maior do que a do baixo volátil. Entretanto não foi possível estabelecer uma relação entre a queima e taxas de reação a matéria volátil total das misturas de carvão. Em geral a queima do coque de petróleo foi inferior a queima dos carvões. A estrutura do carbono do coque de petróleo provou ter forte influencia na queima de tal forma que ate 10% de participação do coque de petróleo na mistura não mostraram nenhum efeito significativo na queima dos carvões.

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## 1 INTRODUCTION

Pulverized coal injection (PCI) is now a proven and well established technology and is becoming increasingly utilized in blast furnace operations due its potential to produce greater flexibility, lower cost and increased productivity. Most importantly, it reduces the dependence on ageing coke plants throughout the world. However, anticipated economic and environmental advantages of PCI technology are constrained by several factors including coal characteristics, coke properties and iron-ore feed properties. Such constraints become increasingly stringent as coal injection rate exceeds 150 kg/t-HM.

For a successful high-rate injection operation that is assured of achieving desirable effective economic gains, it is imperative that high coke substitution rates are achieved, which in turn is strongly influenced by the fact that the injected coal must burn as fast as possible in the raceway where residence time is very short, i.e. ~20 ms.<sup>[1]</sup> At high injection levels ~ 200 kg/t-HM, about 50% of coal is burnt in front of the tuyeres.<sup>[2]</sup> Under such conditions, unburnt char particles will be carried out of the raceway and accumulate in lower parts of the blast furnace causing disturbance to several operational problems including reduced permeability, undesirable gas and temperature distribution and hence decreased productivity. Ensuring that the amount of unburnt char is maintained at a reasonable level is one of the key requirements for the efficient PCI operation.

To some extent, the degree of combustion can be controlled by the operating conditions such as blast temperature and oxygen enrichment.<sup>[3]</sup> However, the injected fuel properties are known to have the most significant effect on their combustion behavior. Low Volatile Matter (LVM) coals, i.e. those with <20% daf (dry ash free) content and in some cases other carbon rich fuels such as petroleum cokes (PC), are believed to provide a higher coke replacement ratio due to their high carbon content. However, they are often difficult to burn when compared to Higher Volatile Matter (HVM) coals, those with >30% daf content. Therefore, coals are often blended in order to optimize the replacement ratio and combustion performance.

Several studies have been conducted in past investigating effect of coal properties on combustion behavior in a blast furnace.<sup>[1,3-15]</sup> Fuel ratio (fixed carbon/volatile matter) is commonly used as an indicator of the expected combustion performance of coals.<sup>[16-17]</sup> Unexpectedly, on some occasions, low volatile coals were seen to display a better combustion performance.<sup>[18-19]</sup> However, the understanding of the effect of coal blending on the combustion behavior is limited and often contradictory. Unlike, calorific value, volatile matter and ash content of coals, the combustion efficiency of blends does not necessarily always show additive behavior, particularly when related to the volatile matter of blends<sup>[19-20]</sup> or when coals of widely different ranks are blended.<sup>[21]</sup> The non-additive effect of coal blends on combustion has been proposed to be attributed to heterogeneous nature and hence complex interactions of blend components at higher temperatures including mineral transformations.<sup>[9]</sup> In case of coal blend, as the high temperature behavior of particles of one coal can also be influenced as a consequence of interaction with components of other coal particles. Moreover, the intensity of interaction of the blend components will be further complicated by the local environment around coal particles such as the proximity of particles, stoichiometric conditions depending on the injection rate and practice.

Due to differences in PCI operating practice as well as the heterogeneous nature of coals, no consensus exists regarding any specific properties of coal that can be reliably used to assess the PCI performance in a blast furnace particularly those of blends.<sup>[22]</sup> Based on current understanding, coal volatile matter is the most reasonable and widely accepted parameter to estimate the combustion performance of PCI. However, the uncertainties regarding the reliability of effect of volatile matter alone increases significantly as coals of different geological regions, ranks particularly low volatile coals or pet-cokes are used as the supplementary fuels in the PCI blend.

Pet-coke is a commonly selected choice for blending with coal for power generation and metallurgical applications due to lower cost, high carbon content, low ash content and high calorific value, despite the fact that, pet cokes are generally believed to display low reactivity compared to coal under similar combustion conditions.<sup>[16, 23-24]</sup> The physical structure of pet-coke char has been shown to be similar to that of low volatile bituminous vitrinite char particle.<sup>[23]</sup> However, unlike bituminous coals, pet coke does not necessarily always provide high burnout, especially within the short residence times experienced in the blast furnace raceway.<sup>[23-24]</sup> There is, therefore, a limited understanding of the effects of pet-coke blending on the combustion mechanisms of PCI coals.

In this paper, an experimental study is presented to further clarify the effect of pet-coke on the combustion characteristics of coals for PCI application in a blast furnace. Burnout measurements of coal and pet-coke blends in a range of controlled conditions in a drop-tube furnace (DTF) are provided. Physical and chemical properties of coals and pet-cokes are characterized using a range of techniques including X-ray diffraction. The burnout of blend samples and the reaction rate are related to the coal, pet-cokes and the char properties.

## 2 MATERIALS AND METHODS

Two bituminous coals were selected on the basis of differences in their volatile matter being of the order of 20% and 30%. Two low and high S pet-cokes were obtained from Brazil and USA. Proximate and ash analysis of all the four samples is given in Table 1.

**Table 1.** Proximate and ultimate analysis of coals (samples A & B) and pet-cokes (samples C & D).

	A	B	C	D
<b>Proximate analysis (adb)</b>				
Moisture % (adb)	1.2	2.3	0.9	1.6
Ash %, (adb)	8.4	9.7	0.4	0.7
Volatile % (adb)	20.1	30.1	13.6	13.3
Fixed carbon %	70.4	57.9	85.1	84.4
<b>Ultimate analysis (daf)</b>				
Carbon %	89.9	86.4	90.8	87.0
Hydrogen %	4.7	5.3	4.2	4.0
Nitrogen %	1.8	1.9	2.4	2.2
Sulfur %	0.5	0.6	0.8	4.6
Oxygen %	3.2	5.9	1.8	2.2

**Table 2 .** Ash composition of coal and pet-coke samples expressed as oxides (wt.%).

Elements	A	B	C	D
SiO <sub>2</sub>	49.1	60.0	18.7	52.0
Al <sub>2</sub> O <sub>3</sub>	36.3	25.3	8.0	10.1
Fe <sub>2</sub> O <sub>3</sub>	4.5	5.8	11.9	10.9
CaO	3.9	1.0	2.9	4.7
MgO	0.8	1.0	0.7	1.8
K <sub>2</sub> O	0.9	3.5	0.6	0.4
Na <sub>2</sub> O	0.9	1.2	1.8	1.8
TiO <sub>2</sub>	1.6	1.2	1.1	0.5
SO <sub>3</sub>	0.7	0.4	0.0	0.4
P <sub>2</sub> O <sub>5</sub>	1.1	0.4	0.2	0.5
Mn <sub>3</sub> O <sub>4</sub>	0.0	0.1	0.2	0.2
V <sub>2</sub> O <sub>5</sub>	0.1	0.1	44.5	10.7
NiO	0.0	0.0	9.2	5.7
Others	0.6	0.6	1.2	2.3

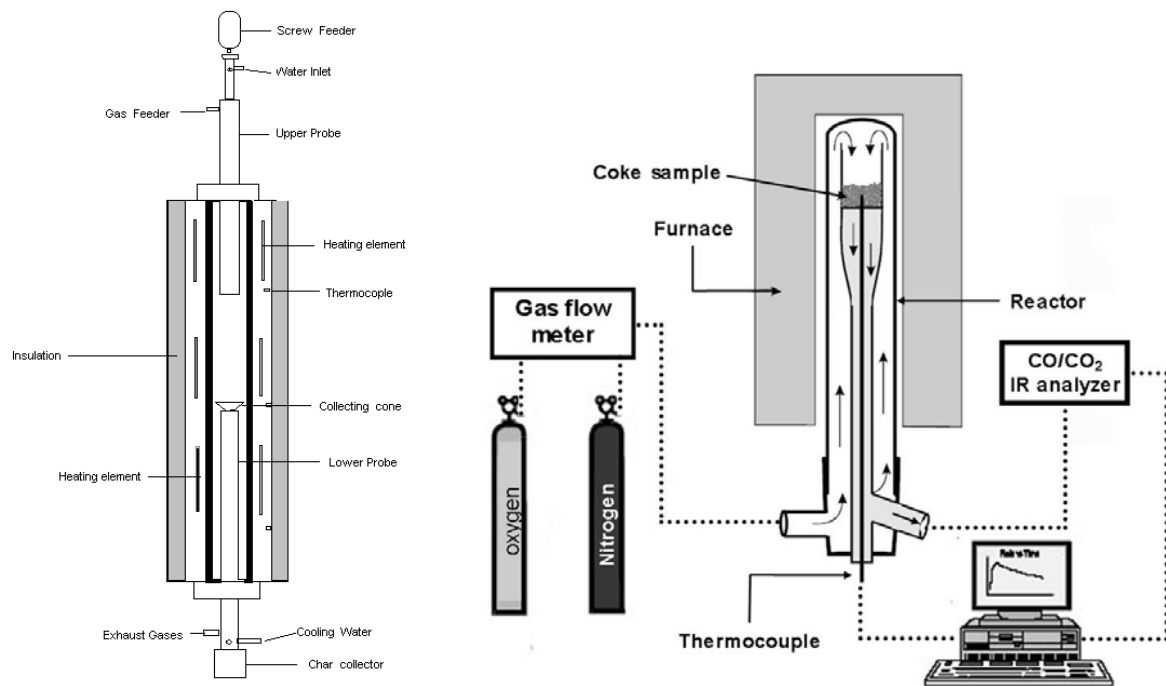
## 2.1 Drop Tube Furnace

Bulk coal samples were initially pulverized using a standard preparation approach and then stored in a sealed container inside a cold store. After pulverizing, the bulk samples were dry sieved to obtain sub- samples in the range of 38 to 75 microns to avoid feeding problems in drop tube furnace (DTF) during injection as well as to minimize effect of variation due to size distribution. The drop tube furnace is equipped with a particle feeding system, sample collector and gas distribution system (Figure 1). The DTF was electrically heated up to the test temperature. Coal particles were injected from the top through a water-cooled feeder into the hot-zone of DTF at the controlled rate of 0.15 g/min with a steady gas flow rate of 1.22 l/min. In this system, particles are heated at the rate of the order of 10<sup>4</sup> degree/second. Char samples were collected at the bottom of the DTF after a residence time of particles about 2 seconds. The feeder was water-cooled to prevent either pre- or post-experiment reactions. The gas composition was maintained at 76% N<sub>2</sub> and 24% O<sub>2</sub> using Brooke’s mass flow controller. Pyrolysis char samples were prepared from coals and their blends at 1473 K in the presence of 99% N<sub>2</sub> and 1% O<sub>2</sub>. A small amount of (1 %) oxygen was used to avoid soot and tar condensation on char particles. Residual volatile matter in pyrolysed char samples was less than 5%. Burnout percentage was calculated by using the ash tracer method by assuming that ash loss during combustion was insignificant using equation 1.<sup>[13]</sup>

$$\text{Coal Burnout} = (1 - A_o/A_i)/(1 - A_o/100) \dots\dots\dots\text{Equation 1}$$

where A<sub>o</sub> is the ash content (dry basis) of the feed coal and A<sub>i</sub> is the ash content (dry basis) of the combusted char sample.

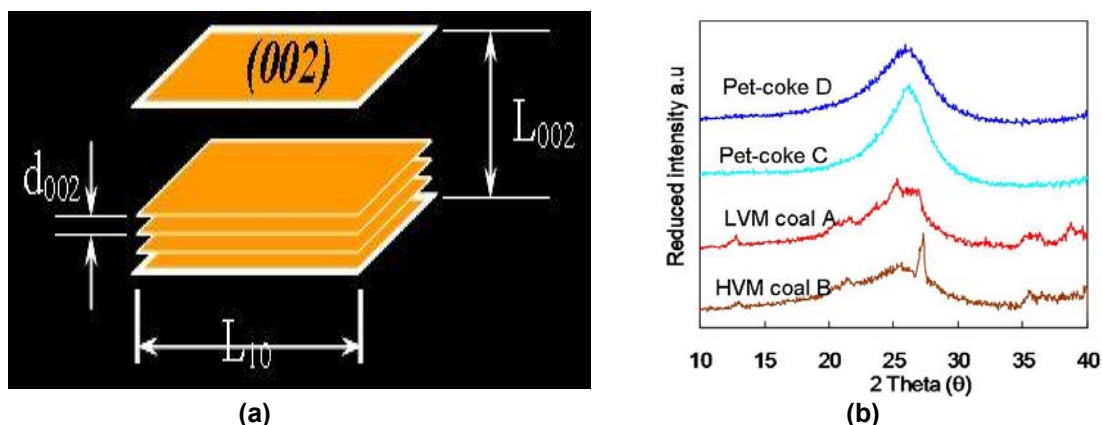
Apparent reaction rate of coal blend char samples is measured using a fixed bed reactor at about 773 K by oxidizing in the presence of 40% O<sub>2</sub> and 60% N<sub>2</sub>. Further details are described in our previous studies.<sup>[12]</sup>



**Figure 1.** (a) Schematic of the Drop Tube Furnace (DTF) and (b) fixed bed reactor (FBR).

## 2.2 Carbon Structure Measurements- X-ray Diffraction

On the basis of carbon structure, coal carbon matter can be considered as composed of crystalline and amorphous carbon matter. With increasing coal rank, more carbon layers are arranged in vertical stack, and is often represented by a carbon structure parameter, also known as stack height, often denoted by  $L_c$  or  $L_{002}$ . The  $L_{002}$  value represent total thickness of planes of carbon crystallites while  $L_{01}$  represent lateral dimension of carbon crystallite as illustrated in Figure 2a and detailed elsewhere.<sup>[25, 26]</sup>



**Figure 2.** (a) Illustration of structural parameters of carbon crystallite and (b) comparison of reduced intensity of XRD spectra of original coal and pet-coke samples.

Crystalline proportion of carbon in all samples was measured by analyzing the shape of 002 carbon peak of the XRD spectrum. XRD data of char and coal samples was obtained by using less than 0.2 gm of sample using Philips 112 X-ray Diffractometer using Copper  $K\alpha$  radiation (30 kV, 30 mA) as the X-ray source. Samples were packed into a glass holder and scanned over an angular range ( $2\theta$ ) of 5-115° by using a step size of 0.05° and collecting the scattering intensity for 5 seconds at each step. The average stack height ( $L_{002}$ ) of carbon crystallite was calculated using Equation 2 after processing of raw data as detailed elsewhere. <sup>[25]</sup>

$$L_{002} = 0.89 \lambda / B_{002} \cos \phi \dots\dots\dots\text{Equation 2}$$

where  $\lambda$  is the wavelength of the X-ray radiation,  $B_{002}$  is the full width at half maximum intensity (FWHM) of 002 carbon peak and  $\phi$  is the two theta position of the same peak. A narrow peak width means that a greater proportion of carbons are organized in crystalline planes or graphitized, and is characterized by high  $L_{002}$  values.

Figure 2b compares the XRD spectra of four samples and the shape of 002 carbon peaks. Each sample has different peak shape and hence different proportion of crystalline and amorphous carbon such that with increasing rank, peak becomes narrow and more carbons are arranged in vertical stack. Accordingly, low volatile coal A indicates a narrower 002 peak compared to high volatile coal B. The  $L_{002}$  values of pet cokes C and D is much higher than two coals A & B indicating carbon structure of pet-cokes is more ordered (Table 3). The interlayer spacing,  $d$ , of both pet-cokes C & D is less than 0.340 nm being close to typical interlayer spacing of graphite (0.335 nm) which is known to be less reactive compared to coals.

**Table 3.** Carbon structure parameters of coal and pet-coke samples.

	A	B	C	D
Lc (nm)	1.05	0.73	1.26	1.49
Xa (nm)	0.38	0.40	0.23	0.20
Ar	0.80	0.75	0.74	0.73
d (nm)	0.350	0.345	0.340	0.339

\*A & B are coal samples; C and D are pet-coke samples

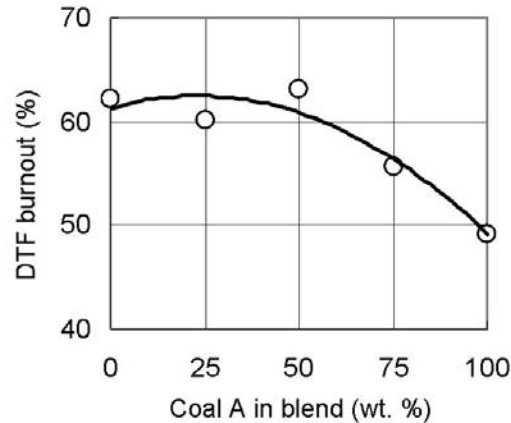
### 3 RESULTS AND DISCUSSION

#### 3.1 Effect of Blending Coals

Figure 3 shows the burnout variation of blends of two coals A and B after combustion at 1473 K in the DTF. The burnout of high volatile matter Coal B and low volatile matter coal A is about 60% and 50% respectively. Therefore, under similar DTF combustion conditions, coal A provides about 17% less burnout compared to coal B. This observed difference is consistent with general expectation in which low burnout is often associated with low volatile matter, high rank and high degree of carbon structure ordering.

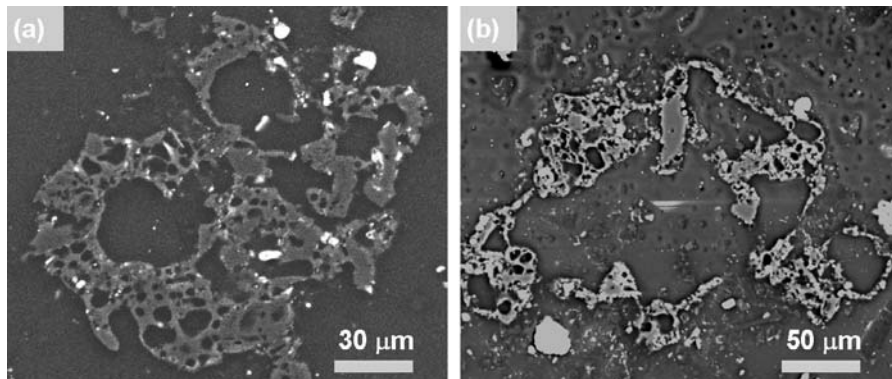
Figure 4 and Figure 5 show the physical characteristics of dominant char particles of coal A and coal B after devolatilization and partial combustion. Coal A char sample contained high proportion of typical thick carbon wall particles as shown in Figure 4. The

average thickness of typical char particles of coal A is observed to be greater than the average thickness of char particles of coal B after devolatilization (Figures 4a & 5a) as well as after partial combustion. Therefore, higher burnout of coal B can be contributed by high porosity and thin carbon walls of its char particles.

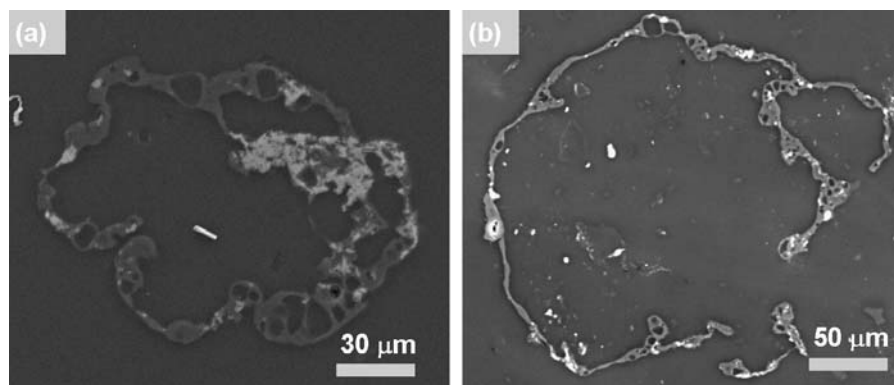


**Figure 3** Variation of burnout with coal A content in the blend samples of LVM coal A and HVM coal B after combustion in DTF at 1473 K in the presence of 24%O<sub>2</sub>.

Figure 3 further shows that as the proportion of Coal A in the blend is increased up to 50%, the burnout of blend is unexpectedly not changed significantly even though volatile matter and other properties change depending on the ratio of two coals in the blend. However, as the coal A proportion exceeds 50%, the burnout of blend begins decreasing as normally expected based on coal parameters particularly volatile matter. Unlike current perception, these results clearly demonstrate that burnout of coal blends is not an additive property, and hence cannot be reliably estimated based on simply combining individual coal properties.



**Figure 4.** (a) Typical morphology of char particle of LVM coal A after devolatilization at 1473 K (1% O<sub>2</sub>) and (b) after partial combustion in DTF at 1473 K (24% O<sub>2</sub>).

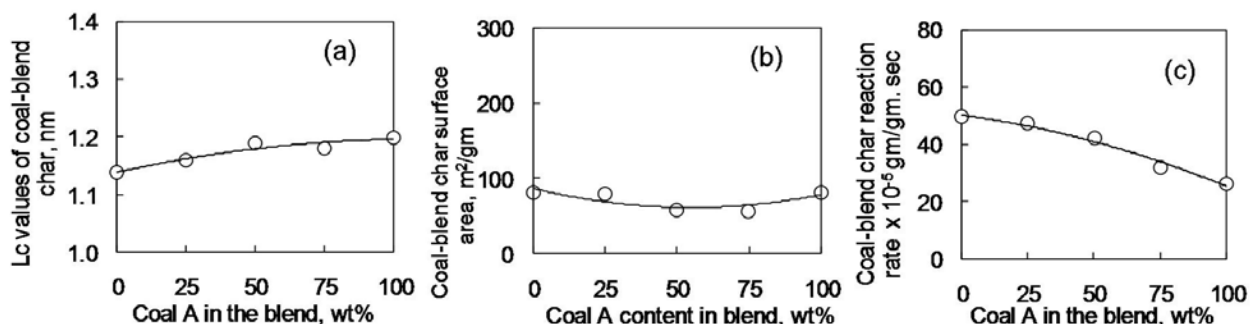


**Figure 5.** (a) Typical morphology of char particle of HVM coal B after devolatilization at 1473 K (1% O<sub>2</sub>) and (b) after partial combustion in DTF at 1473 K (24% O<sub>2</sub>).

Burnout of blends is influenced by several factors including the combustion environments as well as physical and chemical properties of char. However, this non-linear burnout of the blends cannot be explained on the basis of coal properties including volatile matter and carbon structure. A similar trend of effect of blending coal A and B was also observed at a higher combustion temperature of 1673 K.

In order further clarify the interactions of two coals during devolatilization, the char samples were prepared at 1673 K by using blend samples of the same coals. Char properties of individual coal-blends appear to be modified as a consequence of interaction of volatiles released during pyrolysis (Figure 6). The L<sub>002</sub> values of coal-blend char samples increases with increasing proportion of coal A in the blend as expected (Figure 6a). Figure 6b shows that surface area of devolatilized coal-char samples did appear to change significantly (Figure 6b). However surface area of the char samples of the same set of coal blends decreased rapidly as the coal A content in the blend exceeds more than 50% (not shown here). This evolution of surface area of coal-blend char samples during combustion was found to follow the trend of the burnout variation seen in Figure 3.

The effect of char properties modification is also reflected in the reaction rate changes of coal-blend char samples. Figure 6c illustrates that reaction rate is not significantly modified when coal A is less than 50% in the blend. On the other hand, the reaction rate starts declining as coal A content exceeds 50% in the blend. This suggests that the effect of blending on char reactivity is modified as a consequence of interaction of volatiles released from each of blend components during devolatilization.



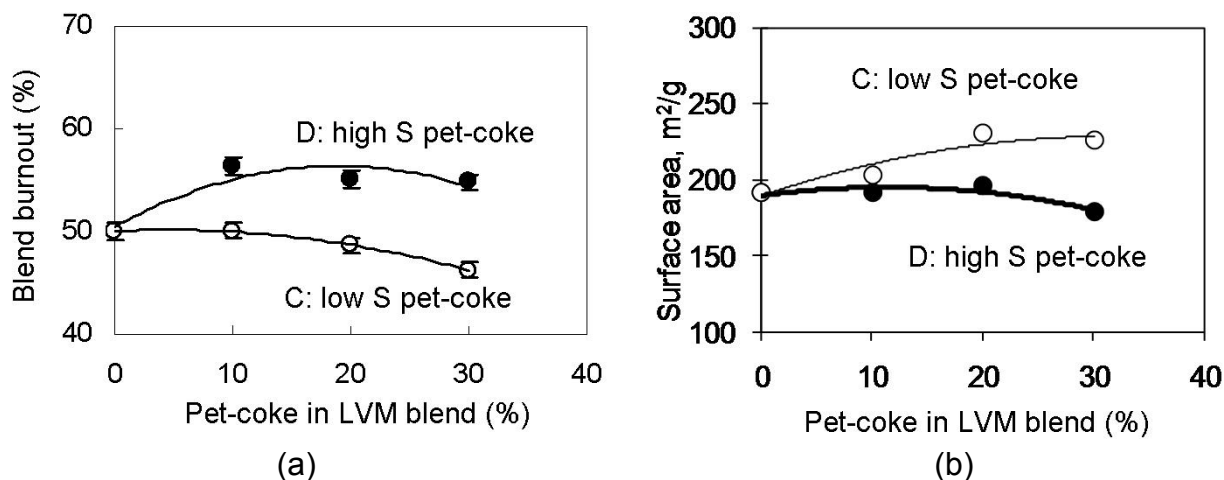
**Figure 6** Variation of properties of coal-blend chars made at 1673 K (1%O<sub>2</sub>) with coal A content; (a) L<sub>c</sub> values; (b) surface area and (c) FBR char reaction rates at 15% conversion at 773 K (40%O<sub>2</sub>).



### 3.2 Effect of Blending Pet-cokes

Figure 7a shows that the effect of blending two pet-cokes with coal A. Figure 7a shows that pet-coke C decreases the burnout of coal A. The burnout of the blend sample containing high-S pet-coke D is marginally greater than that of coal A, but still low being of the order of 55%. There is no obvious explanation of this unexpectedly higher burnout of pet-coke D and coal A blend. High amounts of nickel and vanadium may have some catalytic effect but is less likely as similar higher burnout was not seen in any other cases or reported before in previous studies.<sup>[23-24]</sup> The same figure further shows that the burnout of pet-coke D blend samples is greater than the burnout of pet-coke C blends. The burnout differences can be related to the differences in the carbon structure. Low burnout of both pet-coke blends can be attributed to high ordering of carbon structure as seen in the XRD patterns of pet-cokes and coal A (Figure 2a). Carbon structure of pet-cokes is generally more ordered when compared to that of coals including Coal A. The  $L_{002}$  values of pet-cokes C and D is 1.25 nm and 1.45 nm respectively which is 25% to 50% more than  $L_c$  value of coal A (Table 3).

Figure 7b shows that both pet-cokes did not indicate any significant increase in surface area as was seen in case of only coal blends. This implies that interaction of coal and pet-coke volatiles is not significant and growth of surface area of char samples is not having significant influence on the modification of coal burnout.

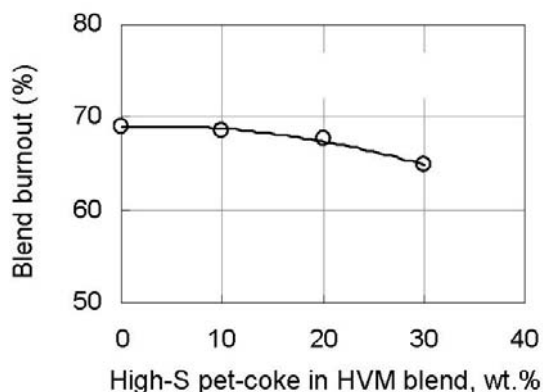


**Figure 7** (a) Burnout of coal A blend with varying pet-cokes (C & D) percentage after combustion in DTF at 1473 K (24%O<sub>2</sub>) and (b) surface area changes of char samples after devolatilization.

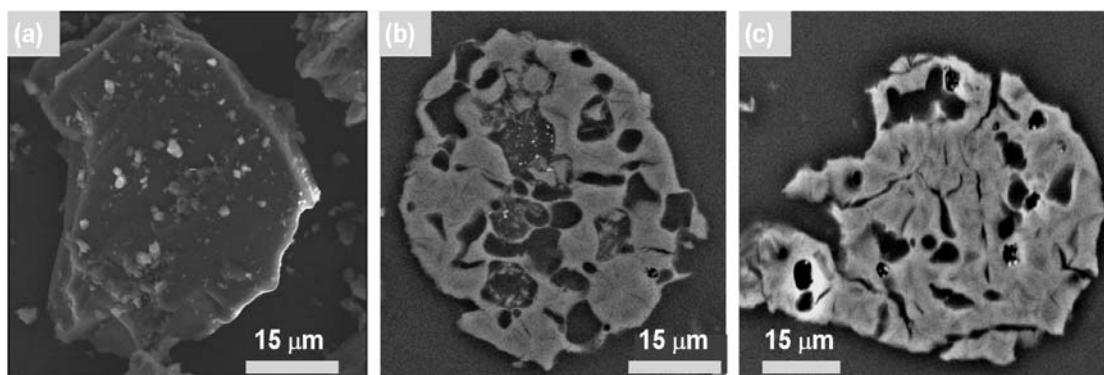
Normally, the pet-coke is blended to replace carbon rich low volatile coals. Therefore, only one of the very high volatile coal was tested in this study. In this case also, the burnout of high volatile coal is also seen to decrease with increasing amount of pet-coke D in the blend (Figure 8). This implies that that even in the high volatiles released from HVM coal did not have any positive influence on the pet-coke char reactivity as well as the burnout.

Figure 9 shows typical pet-coke A particle and changes in morphology after devolatilization and partial combustion at 1473 K. Clearly, pores present in original pet-coke are opened after devolatilization but there is small changes in the physical structure after combustion. Similarly, pet-coke D char also do not show a significant change in pore structure after combustion. The SEM analysis of pet-coke char samples

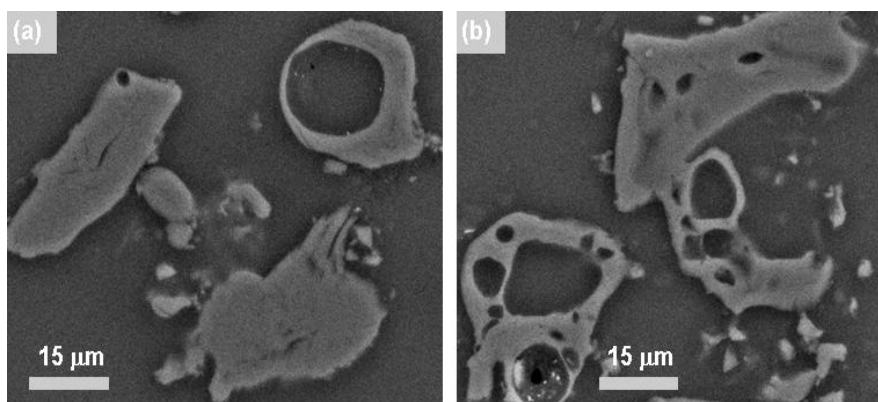
further shows large thickness of char particles which will consequently make it more difficult to burn. This observation of thick wall char structure of pet-coke char particles is consistent with previous studies.<sup>[23]</sup> Therefore, both carbon structure and SEM analysis support our burnout observations of pet-coke blends in the DTF such that generally pet-cokes would burn with greater difficulty when compared to coals. The study also implies that carbon structure of pet-cokes has a strong effect on the burnout of coal blends. Under the tested conditions up to 10% blending of either of the two pet-cokes did not show any appreciable effect on burnout of the tested coals



**Figure 8** Burnout variation with different percentage of pet-cokes D in blend samples of a HVM coal after combustion in DTF at 1473 K in the presence of 24%O<sub>2</sub>.



**Figure 9** (a) Typical morphology of particles of pet-coke sample C; (b) pet-coke char sample after devolatilization at 1473 K (1% O<sub>2</sub>) and (c) after partial combustion in DTF at 1473 K (24% O<sub>2</sub>).



**Figure 10** (a) Typical morphology of particles of pet-coke char sample D after devolatilization at 1473 K (1% O<sub>2</sub>) and (b) after partial combustion in DTF at 1473 K (24% O<sub>2</sub>).

## 4 CONCLUSIONS

Two petroleum cokes were blended with low and high volatile Australian coals to study the effect on burnout of blends for PCI applications. The study demonstrated that coal blending influences the char properties of individual coals. The burnout of tested blends is found to be non-additive such that burnout of the blends cannot be explained on the basis of individual coal properties particularly volatile matter. Unlike coal blends, pet-cokes did not display significant effect on the modification of char characteristics of the blend which is most likely attributed to limited interaction of volatiles of coals and pet-cokes. The pet-cokes were generally found to burn with a greater difficulty. Carbon structure of pet-cokes was found to have the most significant effect on the burnout differences such that pet-coke with more ordered carbons indicated lower burnout when blended with the same coal. The study implies that under similar combustion conditions, up to 10% blending of either of the two pet-cokes may not show any appreciable effect on burnout of the tested coals. As the pet coke of the blend exceeds more than 10%, the effects become increasing counterproductive.

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