# **Reduction in coal fines and extended coke production through the addition of carbonisation tar: Environmentally clean process technology**

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# Highlights

- Influence of carbonisation tar addition on proximate analyses is discussed.
- 56.8% reduction in coal fines (powder) was observed
- Carbonisation tar reduced ash content in coal blend.
- Coke qualities reported are comparable to international benchmarks.
- A schematic layout of the carbonisation tar process is proposed.

# Abstract

Production of coal fines in coke making has lately become a problem worth addressing due to its negative impact on ecosystems. The use of coal fines is becoming an issue due to high Quinoline Insoluble formation during the carbonisation process as well as carry over or enhancement of carbon deposition in the upper parts of the coke ovens. In this study, the effect of carbonisation tar addition over a range of 2.0 - 8.0 wt.% was evaluated as a probable partial substitute for expensive coking coals by assessing coal fines reduction, coal blend cost analysis, extended coke production and coke quality. At the optimum condition of adding 6.0 wt.% carbonisation tar, key coke qualities were improved. This occurred even when lower quality coking coal of up to 35% was included in the coal blend. Coke quality results obtained in this study were comparable to international benchmarks. Furthermore, a 56.8 wt.% reduction in coal fines (powder) was observed. By reducing coal fines, there is a considerable reduction in coal dust that personnel are exposed to and a reduction in acid mine drainage as well as a decreased likelihood of spontaneous combustion. Savings of up to USD 1.7 million per year were postulated and supported by mathematical models used to calculate the cost-effectiveness of such a project.

Keywords: Coal; Coal fines; Carbonisation; Tar waste; Process technology

## **1. Introduction**

Coke production plays an essential role in South African national economy and will continue to increase in importance in the near future. In South Africa, coke making plants produce both metallurgical and market coke which is used by the steel plants and the ferroalloy industry respectively. Coke quality has been a subject of intense research for a long time and previous efforts were aimed at developing best techniques for production of good quality coke. Many researches and experiments that are documented and known to improve coke quality include coal pre-treatment (Jackman and Helfinstine, 1970), diesel and tar addition to coal blends (Chatterjee and Prasad, 1982), addition of petroleum coke to coal blends (Forrest and Marsh, 1981), oil addition (Kestner et al., 1981), petroleum pitch addition (Gonzalez -Cimas et al., 1986, Collin and Bujnowaska, 1994), pitch addition (Lin and Hong, 1986), breeze addition (Taylor and Coban, 1987), addition of aluminium metal in scrap form (Alvarez et al., 1989), substitution of coking coals by non-coking coals (Das et al., 2002), formed coke addition (Plancher et al., 2002), chemical additive such as hydrochloric acid (Shevkoplyas, 2002), plastic addition (Nomura and Kato, 2006, Melendi et al., 2011), stamped charging (Saxena et al., 2010), addition of coal-tar pitch to coal blends (Benk, 2010, Benk and Coban, 2011), steam treated coking coals (Shui et al., 2011) and briquetting charging (Díez et al., 2013) have been carried out. Predicted operational challenges associated with most of these techniques include carryover of fines, explosion hazards, increased charging and pushing frequencies, thus limiting the full scale of application of these techniques. Moreover, other shortcomings surrounding these implementations included high cost of reagents, insufficient additives stock and cost of converting current top-charged coke oven batteries to include different charging is so high that is not economically viable to pursue.

In the coke making setting, oven charging process generate significant amounts of waste in the form of coal fines. The use of coal fines is becoming an issue due to high Quinoline Insoluble (QI) formation during the carbonisation process, additional coal loses and to the greater likelihood of spontaneous combustion and acid mine drainage as well as carry over or enhancement of carbon deposition in the upper parts of coke ovens (Nakagawa et al., 1998). This will not only endanger coke making personnel through occupational diseases as well as fatalities but also bring about possible coke making plant closures. According to various authors (Krebs et al., 1996; Nakagawa et al., 1998), carbon deposition brings about high resistance of pushing the coke, narrowing of the cross section of the ascension pipes, giving rise to numerous complications in oven maintenance. Furthermore, depletion of coking coal deposits, the comparatively high price of prime coking coals and their scarcity worldwide generates the need to develop new clean coal technologies to maintain coke production levels for years to come. The earlier statement is supported by another case mentioned by Nomura and Arima (2017) who mentioned that one of the most significant focuses for the coke making industry today is the future depletion of coking coals that forms good quality coke.

Carbonisation tar is a kind of toxic hazardous industrial solid waste produced in the process of coke making. Carbonisation tar is currently being used in chemistry and refractory material industry, production of synthetic carbon materials or electrodes for the metallurgy and in carbon composites technology (Mikociaka et al., 2014). The off-specification carbonisation tar is usually reported pertaining to QI and Matter Insoluble in Tar (MIT) values and has significant implications on its use as a binding material. The off-specification carbonisation tar pose storage requirements, plant availability complications and landfill disposal which requires premium budget. Therefore, partial substitution of more expensive coking coals with carbonisation tar in order to lower coal blend cost and minimise carbonisation tar waste disposal is becoming more critical to the success of coke production in a competitive market.

The addition of carbonisation tar into coal blend was chosen in the current study as it is one of the by-products of the coke making process, which is readily available and is a relatively inexpensive hazardous material. Even though the possibility of using tar as an additive in coal blends has been studied, its influence on coal blending based on proximate analysis, has not

been discussed. Additionally, its environmental pollution impact analysis coupled with economic assessment has not yet been evaluated. The principal aim of the present study was to assess the effect of adding carbonisation tar to an industrial coal blend. Special emphasis was placed on carbonisation tar recycling, coal fines reduction, coal blend cost analysis, extended coke production and improved coke quality.

# 2. Materials and methods

## 2.1. Experimental procedure

Carbonisation tests were carried out in one of the coke making battery consisting of 50 slot ovens. The oven dimensions are 450 mm width  $\times$  1385 mm length  $\times$  620 mm height. The coal blend was prepared by mixing four coking coals covering a wide range of rank and coal properties such as volatile matter content, ash content and sulphur content, geographical origin and thermoplastic properties. The carbonisation tar produced from the base blend, was collected from tar decanters of the coke making by-product plant and stored in a tar storage tank at  $55 \pm 2$  °C. Up to 2.0 - 8.0 wt% of carbonisation tar were mixed with prepared coal blend through 8 nozzles of 8 mm diameter fitted in three rows on a single horizontal header pipe. Given that getting the carbonisation tar to get uniformly mixed with the coal blend has been a challenge, this challenge was overcome by spraying carbonisation tar on the specialised conveyor belt feeding the crusher in order to increase contact period as the liquid carbonisation tar infiltrated the porous nature of the coal on its way to the crusher and resulted in a better coal blend carbonisation tar mix. Coal blend was then charged under gravity through charging holes into the coke oven that had reached 1200 °C. A programmable controller was used to keep the oven temperature constant. Temperature at the centre of the coal charge was monitored by means of a thermocouple connected to a computer. The temperature of the wall was kept constant and the coking time was fixed at 19 h.

## 2.2. Required coal and coke analyses

The thermoplastic properties of the coal blend samples were carried out using Gieseler Plastometer (Preiser Scientific, China) following the ISO 10329, 2009 standard procedure. The total dilatation for coal blends was measured using a Rühr dilatometer (Netzsch, Germany) as per ISO 349, 1975 standard procedure. Proximate analyses were performed following the SANS 11722, 2005, ISO 562, 2010, ISO 1171, 2010 standard procedures for moisture content, volatile matter, and ash content respectively. The fixed carbon (FC) content value was determined by subtracting the total of the percentage moisture, volatile matter and ash from a hundred. The measurement of contraction was carried out in a sole heated oven following the ASTM D2014, 2010 standard procedure. Coke samples produced were prepared and tested for Coke Strength after Reaction (CSR) and Coke Reactivity Index (CRI) measurement as per specification in the ASTM D5341 – 99, 2004 standard procedure. The Micum indices measurements and coke cold strength were evaluated as per ISO 556, 1980 standard procedure. Stability factor reflecting the load carrying strength or impact resistance of the coke was determined following the ISO 501, 2012 standard procedure.

## 2.3. GC/MS analysis of carbonisation tar

Carbonisation tar compounds were identified by Gas Chromatograph coupled with Mass Spectroscopy (GC/MS). Carbonisation tar analysis was carried out using an Agilent 6890N GC-MS equipped with a HP – 5 capillary columns and a flame ionization detector (FID).

Table 1			
Coking coals	origin and	chemical	properties.

Coal Origin	Blend %	FC (wt.% db)	VM (wt.% db)	Ash (wt.% db)	Moisture (wt.%)	R <sub>o</sub> Vr (%)	SD (ồi) RoVr (%)	FSI (ISO)	S (Pyritic)	S (SO <sub>4</sub> )	Total S
Coal A RSA Coal B NZ Coal C Austr Coal D USA	35 8 lia 38 19	47.0 57.3 58.9 60.0	37.2 32.0 24.5 26.7	10.0 3.8 9.8 6.9	5.8 6.9 6.8	0.71 1.12 0.90 1.32	0.028 0.034 0.042 0.036	6.0 9.6 8.3 8.5	0,12 0,12 0,10 0,13	0.01 0.01 0.01 0.01	1.06 1.00 0.65 1.08

VM: volatile matter; db: dry basis; FSI: Free Swelling Index; FC: Fixed Carbon; ISO: International Organization for Standardization; S: Sulphur; R<sub>o</sub>Vr: Reflectance; SD: Standard Deviation of Reflectance; NZ = New Zealand; RSA: Republic of South Africa; USA: United States of America.

Helium was used as carrier gas at a flow rate of 1.2 mL/min. The detector and injector temperatures were set at 280 °C and the column initial temperature was kept at 60 °C for 5 min, and then heated to 280 °C at a heating rate of 4 °C/min and held at the temperature for 10 min.

# 3. Results and discussion

## 3.1. Characterisation of blend coking coals

The coal blends used, comprised mainly of New Zealand (NZ), United States of America (USA), South African (RSA) and Australian coals. Table 1 shows the results of the coking coal samples characterisation. The coal blends contained between 0.65 - 1.08 wt% sulphur content, 3.8 - 10 wt% ash, and 24.5 - 37.2 wt% volatile matter. The coal blend compositions were comprised of 35% RSA coal, 19% USA coal, 38% Australian coal and 8% NZ coal. Considering the results shown in Table 1, it can be seen that RSA coking coal (Coal A) is a low rank non-coking Bituminous C coal with a Vitrinite Reflectance (RoVr%) of 0.71. The rank of both the NZ coking coal (Coal B) and the USA coking coal was classified as Bituminous B with the vitrinite reflectance of 1,1 to 1,3 RoVr% respectively. The Australian coking coal C falls within the border lines of Bituminous B and C (RoVr% = 0,9). i.e. marginally out of the prime coking category similar to NZ coking coal B. The vitrinite reflectance (RoVr %) of these coking coals are commonly accepted as a rank parameter indicating the rank or the degree of coalification of coal which controls the coking capacity in the vitrinite macerals in the coal (i.e. the properties of swell, plasticity and fusion).

The coal macerals found in the coking coal samples is comprised of vitrinite, liptinite, semifusinite, inertinite and pseudovitrinite as shown in Table 2. The significance of macerals in the context of the current study is that vitrinite and liptinite groups of coking coals behave as reactive macerals during carbonisation process (Varma, 2002). The vitrinite maceral group is grey in colour, with a reflectance that is between that of liptinite and inertinite. Generally, the coal rank is directly proportional to the volatile matter and carbon content in European and USA coals but due to the heterogeneous nature of RSA coals, rank is more reliably determined by vitrinite reflectance (Falcon and Ham, 1988). The liptinite macerals occur as fine particles embedded in a matrix of vitrinite components (Fig. 1). The coking coal C is rich in vitrinite followed by Coal D. Coal A has the lowest vitrinite maceral followed by Coal B. The coal A liptinite content of 4.6 vol % is higher than the rest of the coking coals. According to Teichmüller (1989), liptinites were derived from hydrogen-rich plant organs and also from decomposition products. A high inertinite content was found in Coal B. Coal D has a high semifusinite content of 15.1 vol %.

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Maceral	Coal A	Coal B	Coal C	Coal D			
Vitrinite [vol%]	78.4	70.2	88.0	84.0			
Liptinite [vol%]	4.6	1.2	2.4	0.0			
Semifusinite [vol%]	3.5	5.1	5.0	15.1			
Pseudovitrinite [vol%]	8.0	0.8	1.4	0.0			
Inertinite [vol%]	5,5	13.0	3.0	0.0			

Table 2 Maceral analyses of individual coking coals.



**Fig. 1.** Typical macerals found in coking coal include A: Vitrinite, B: Liptinite, C: Semifusinite, D: Pseudovitrinite, E: Inertinite.

## 3.2. Characterisation of carbonisation tar

Table 3 shows the properties of coke oven tar used in this investigation. From the analyses, it was determined that the moisture content was 3.1 wt% and 3.4 wt%, against a target of  $\leq 5$  wt%. Low moisture content in coke oven tar is considered acceptable as high moisture bring about bulk density effect on coal blend. A low ash content of 0.04 wt% was observed against a target of 0.16 wt%. Low ash content in coke oven tar is necessary as it may cause reduced ash content when added to high ash content coal blend. Specific gravity (SG) of 1.12 g cm<sup>-3</sup> and 1.15 g cm<sup>-3</sup> against a target of 1.20 g cm<sup>-3</sup> was reported. Furthermore, flue temperature of 1230 °C and 1233 °C were observed against a target of 1233 °C.

QI is used to determine the quantity of solid and high molecular weight material in carbonisation tar. According to Patrick et al. (1983), the presence of inert QI particles influences the anisotropy present in the carbonised tar, and the current work reveals that the rank of the coal from which the carbonisation tar was obtained also has a marked influence. High QI values of 7.4 wt.% and 7.8 wt.% against a target of 6.0 wt.% were reported. These values are in agreement with classification made by Panaitescu and Predeanu (2007), who reported QI values in the precursor carbonisation tar in the range of 5 - 8 wt%, also in accordance with the study of Díez et al. (1994), who characterised coal tar as follows: Ash (0.04 wt%), SG

Table 3							
Properties	of cart	onisation	tar	with	various	moisture	content.

	Moisture (wt.%)	MIT (wt.%)	Ash (wt% db)	QI (wt%)	SG (gcm <sup>-3</sup> )	Flue Temperature (°C)
Target	5.0	6.0	0.16	6.0	1,20	1233
Sample 1	3.4	7.3	0.04	7.8	1.12	1230
Sample 2	3.1	7.2	0.04	7.4	1.15	1233

MIT - Matter Insoluble in Tar; db - dry basis; QI - Quinoline-Insoluble; SG - Specific Gravity.

(1.17 gcm<sup>-3</sup>), mean flue temperature (1230 °C) and QI (2.9 wt%). MIT of 7.2 wt% and 7.3 wt% against a target of 6.0 wt% was also recorded.

The carbonisation tar characterisation required the use of specialised analytical tool that is able to work over a range of molecular masses such as GC-MS. Fig. 2 depicts typical hydrocarbons found in carbonisation tar as indicated by GC-MS. Table 4 shows that about sixteen (16) predominant hydrocarbons were detected by GC-MS.



Fig. 2. Typical hydrocarbons found in Carbonisation tar.

No	RT(min)	Compound	MF	MW (g/mol)
1	18.01	Benzene	C <sub>6</sub> H <sub>6</sub>	78
2	19.50	Thiophene	C <sub>4</sub> H <sub>4</sub> S	84
3	21.45	Phenol	C <sub>6</sub> H <sub>6</sub> O	94
4	22.00	o-cresol	C <sub>2</sub> H <sub>8</sub> O	108
5	23.09	Xylenol	C8H10O	120
6	25.38	Naphthalene	C10H8	128
7	25.51	Quinoline	C <sub>9</sub> H <sub>7</sub> N	129
8	26.26	Methylquinoline	C <sub>10</sub> H <sub>9</sub> N	143
9	29.13	biphenyl	C12H10	154
10	30.22	Phenylpyridine	$C_{11}H_9N$	155
11	32.02	Carbazole	C <sub>12</sub> H <sub>9</sub> N	167
12	33.24	Dibenzofuran	C12H8O	168
13	34.10	Anthracene	C14H10	178
14	36.00	Benzoquinoline	C <sub>13</sub> H <sub>9</sub> N	179
15	36.44	Fluorenol	C13H10O	181
16	38.07	Pyrene	C16H10	202

Table 4 Qualitative data from GC-MS analysis of carbonisation tar samples.

RT = Retention Time; MF = Molecular Formula; MW = Molecular Weight.

Many investigators have studied the characteristics of coal tar. The results as shown in Table 4 are also in good agreement with those reported by other (Benk and Coban, 2011, Wang et al., 2013; and Si et al., 2017). The results obtained for these hydrocarbons are significant, confirming carbonisation tar characteristics regarding its influence on coal blend binder quality.

#### 3.3. Effect of carbonisation tar addition on coal proximate analyses

The effect of carbonisation tar addition on the proximate analyses of coal blends was evaluated and the results are shown in Table 5. Ash content, sulphur content and volatile matter content of individual coking coal values are additive properties which means the value of the coal blend will be the average value of the individual coking coals within the blend. The rule of mixture is the simplest way of predicting a coal property from its coal blend using Eq. (1):(1)

$$Z_i = \sum_{i=1}^n x_i Z_a \tag{1}$$

where:

 $Z_i$  = the property such as ash content, sulphur content and volatile matter content of the coal blend,

 $Z_a$  and  $x_i$  = the property and the percentage mass fraction of coal *i* respectively

n = the number of coals blended.

#### Table 5

Coal blend proximate analysis.

Coal Blend	Ash (wt% db) Measured	Ash (wt% db) Eq. 1	Sulphur (wt.% db) Measured	Sulphur (wt.% db) Eq. 1	VM (wt% db) Measured	VM (wt% db) Eq. 1
0 wt% Tar	9.20	8,84	0.87	0.90	31.1	30.0
2 wt% Tar	8.55	8.66	0.99	n.a	30.0	n.a
4 wt% Tar	8.63	8.66	0.98	n.a	30.1	n.a
6 wt% Tar	8.62	8.66	0.97	n.a	30.2	n,a
8 wt% Tar	8.66	8.66	0.99	n,a	30.4	n.a

n.a: not available; VM: volatile matter; db: dry basis.

#### Table 6

Coal blend plasticity, fluidity, G and FSI values.

Plasticity	0 wt% Tar	2 wt% Tar	4 wt% Tar	6 wt% Tar	8 wt% Tar
Max Fluidity (ddpm)	466	476	488	489	491
Max Fluidity Temp. °C	357	360	367	388	397
Plastic Range °C	78	81	84	86	90
Max C%	27.5	29.8	29.5	29.6	29.8
Max D%	36.0	35,3	38.9	47.3	55.1
G value	1.01	1.01	1.01	1.03	1.04
FSI	8.1	8.1	8.2	8.2	8.3

Max C = Maximum Contraction; Max D = Maximum Dilatation; FSI = Free Swelling Index; ddpm = Dial Division per Minute.

Eq. (1) makes the implicit but reasonable assumption that volume fractions and mass fractions of the blend components are the same, i.e. that all coals have the same density.

Prior to carbonisation tar addition, the measured coal blend sulphur content of 0.87 wt% was reported as opposed to 0.90 wt% calculated using Eq. (1). Therefore, sulphur content of individual coking coals confirmed to be additive with low standard deviation of 0.03 as per the standard statistical procedure. The sulphur content increased from 0.87 to 0.99 wt.% at 2.0 wt % and remained constant at 0.99 wt.% with further carbonisation tar addition. Although the coal blend sulphur content increased, it is still within the acceptable range of <1 wt.%. The sudden increase in sulphur content with carbonisation tar addition might be due to hydrogen sulphide (H<sub>2</sub>S) and sulphur in the carbonisation tar. According to a study by Hou et al. (2018) who investigated transformation of sulphur and nitrogen during pyrolysis of Shenmu coal found that 43.7 wt.% of coal blend sulphur was transferred into coal tar with main forms of benzothiophene (52 wt.%), dibenzothiophene (41%), thiophene (0.15 wt.%) and the other (6.85 wt.%). About 56.2 wt.% of coal blend sulphur was transferred into retorting gas with main forms of H<sub>2</sub>S, carbonyl sulphur and a small amount of SO<sub>2</sub>, Hou et al. (2018). The remaining 0.1% sulphur was possibly lost during the carbonisation process. Sulphur is the single most influential chemical component in coal that affect coke CSR therefore it is undesirable that carbonisation tar increases coal blend sulphur although the increase was within the acceptable margin of <1 wt%.

Coking coal is commonly distinguished from thermal coal in having lower contaminants of ash content. Typical values of low ash content ranges were published by Varma (2002). Prior to carbonisation tar addition, the measured coal blend ash content of 9.20 wt.% was reported as opposed to 8.84 wt.% calculated using Eq. (1). A small standard deviation of 0.36 can be observed for the ash content in the coal blend. After the addition of 2.0 wt% carbonisation tar, the ash content was significantly reduced to 8.55 wt%. The ash content remained constant with further increase in carbonisation tar addition up to 8.0 wt.%. The reduction in ash content and constant values of ash content with further carbonisation tar addition can be attributed to the low ash content available in the carbonisation tar and low percentage of carbonisation tar added to the coal blend respectively.

The volatile matter content of coal blend affects both coke quantity and coke quality. In this study, as the carbonisation tar increased to 2.0 wt.%, the coal blend volatile matter decreased from 31.1 to 30.0 wt.%. On the other hand, as the carbonisation tar increased from 4.0 wt.% to 8.0 wt.%, the volatile matter content remained constant. The volatile matter content obtained for the carbonisation tar addition were in the same range of 30.0 wt.% - 30.4 wt.%. The volatile matter of the coal blends was expected to decrease with carbonisation tar addition because coking coals with high volatile matter content are being replaced by carbonisation tar, which has low volatile matter content. Conclusively, ash content, sulphur content and volatile matter content for coal blends under consideration were found to be additive.

#### 3.4. Effect of carbonisation tar addition on maximum contraction and dilatation

The dilatation percent of the coking coal specifies its coking properties. Table 6 shows the effect of carbonisation tar on the maximum contraction and maximum dilatation of coal blend. It can be observed that as the carbonisation tar increased from 2.0 wt.% to 8.0 wt.%, maximum dilatation increased from 36.0 to 55.1 correspondingly. The dilatation values for the coal blend studied confirm the concept of the requirements that a minimum total

dilatation of 29.6 is necessary to obtain a completely fused coke structure of minimum M10 and therefore a high value of tensile strength (North et al., 2018). In the event of maximum contraction, at 2.0 - 8.0 wt.% carbonisation tar addition, maximum contraction increased from 27.5 at 0 wt.% to 29.8 at 2.0 wt.% addition and remained constant with further addition of carbonisation tar up to 8.0 wt.%. The degree of maximum contraction of the coal blend charge seems to be one of the vital factors for coking pressure since this directs the final volume of the carbonised mass relative to the initial coal blend charge.

Free Swelling Index (FSI) test was used to determine the agglomeration or swelling properties of the coal and its blend. FSI indicates the caking ability through swelling behaviour. However, FSI is not an additive property for coal blend. The effect of carbonisation tar addition on FSI is shown in Table 6. As shown on Table 6, there was no significant change in FSI of 8.1 with carbonisation tar addition. According to Collin and Bujnowaska (1994), a FSI greater than 4 means well coking coals and a FSI greater than 7 indicates high quality coking coal.

The G factor is one of the predicting tools used to determine the coke quality. The effect of carbonisation tar addition on G values is shown in Table 6. There was insignificant change in G values of 1.01 with carbonisation tar addition. According to Collin and Bujnowaska (1994), prime coking coals have G values between 1.02 and 1.1, and the results are in good agreement with findings by Collin and Bujnowaska (1994).

## 3.5. Effect of carbonisation tar addition on plasticity and fluidity

The fluidity of coal is considered one of the important parameter in the coke formation and in determining the quality of the coke produced. In order to ensure optimum coal particle interaction, it is important that the temperature intervals of the plastic state of each coal constituting a blend should overlap. The effect of carbonisation tar addition on plasticity and fluidity was investigated and the results are reported in Table 6. Coal blend fluidity effects due to carbonisation tar addition showed a linear increase from 466 ddpm at 0 wt % to 491 ddpm at 8.0 wt.%. It is well known, that coal blend fluidity is a non-additive property and is considered as the superposition of the fluidities of the individual components at a specific temperature (Fernández et al., 2012). The phenomena occurring in the plastic stage are influenced by carbonisation tar and are indicated by an increase in plastic temperature range of 357 °C (0 wt %) to 397 °C (8 wt.%). The results are in good agreement with the findings by Lin and Hong (1986) who noted the significance of pitches to improve fluidity of coal blends. The results are further supported by an additional case, reported by Chatterjee and Prasad (1982), whose results reported an increase in maximum fluidity due to tar addition. The escalation in the maximum fluidity of most coal blends due to tar addition, plainly points out the improvement of the intrinsic coking characteristics of such blends due to carbonisation tar additions. These results are consistent with the view that during heating through the thermoplastic range, the development of Gieseler fluidity is strongly dependent on a source of transferable hydrogens to cap radical species and thereby generate the low molecular weight 'solvating' species favourable to fluidity development.

## 3.6. Effect of carbonisation tar addition on coal fines

In order to quantify the effect of carbonisation tar addition on particle size distribution, crushed coal blend samples were collected and analysed. Table 7 shows the influence of carbonisation tar addition on coal fines (-0.106 mm). The coal blend with the carbonisation

tar addition of 2.0 wt.% reduced coal fines from 19 wt.% to 16 wt.%. As the carbonisation tar increased to 4.0 wt.%, the coal fines were further reduced to 12 wt.%. With further carbonisation tar addition to 6.0 wt.%, coal fines substantially reduced to 8.0 wt.%. Lastly, with carbonisation tar addition of 8.0 wt.%, the coal fines reduced to 6.0 wt.%. The overall percentage reduction in coal fines (powder) due to carbonisation tar addition over a range of 2.0 wt.%, 4.0 wt.%, 6.0 wt.% and 8.0 wt.% was found to be 16%, 37%, 58%, 68% respectively. The decrease in coal fines by means of carbonisation tar addition may be attributed to the carbonisation tar serving as a binder, wetting coal fine particles (which may cause suppressing of coal fines), which subsequently bind to larger particles. These results are important because more coal fines are less desirable owing to high dust, high QI formation, carbon deposits and bulk density increase. The latter is supported by Yu et al. (1995) who mentioned that the bulk density is strongly affected by the particle size distribution.

#### Table 7

Effect of carbonisation tar on coal fines.

Coal Screens	0 wt.% Tar	2 wt% Tar	4wt.% Tar	6 wt% Tar	8 wt.% Tar
- 0.106 mm (%)	19.0	16.4	12.3	8.2	6.0
	19.2	16.1	12.0	8.0	6.2

By reducing coal and coke fines, there is considerable reduction in coal dust that personnel are exposed to, and a reduction in acid mine drainage when storing the discarded coal fines as well as a decreased likelihood of spontaneous combustion. In addition, coal dust being particulate in nature is a significant hazard factor that can cause explosions, pneumoconiosis and other health related conditions. Therefore, coal fines reduction is a major plus for the industries concerned. Alternatively, using water as an additive to lower the coal fines dust is generally unfavourable for the coke quality. Nomura and Arima (2008) reported that a large amount of vaporized coal moisture (moisture content 20 wt.%) could break through the plastic layer and cause at the thin and uneven plastic layer in the case of wet coal charging, which can seriously affect the coking property. Secondly, as climate change takes effect, many countries are facing serious water scarcity and many others will face severe water shortages within a short period of time.

#### 3.7. Coke hot and cold strength properties: comparative study

In assessing coke quality, many parameters are used such as coke size, ASTM stability, CSR, CRI, grindability, hardness and various drum indices. Table 8 shows for blast furnaces, the physical and chemical properties of coke produced at optimum conditions of 6 wt % carbonisation tar addition compared together with those reported by other operation for European, Australian BHP Port Kembla, American and Japanese blast furnaces as reported by Díez et al. (2002). It should be noted that the abrasion resistance, M10 of 7.0 is greater than Australian Port Kembla reported value of 6.5 and falls within European range. The lowest coke fragmentation, M40 of 67 is reported compared to other furnaces operations reported. I20 of 80 is reported which is in good agreement with European Range of >77.5.

Table 8					
Physical properties requirements	of blast	furnace	coke	in current	operation.

Properties	European Range <sup>a</sup>	Australian Port Kembla <sup>a</sup>	American Range <sup>a</sup>	Japan Range <sup>a</sup>	South African [Current Study]
Mean size (mm)	47-70	50	50	45 - 60	58
M40	>78->88	85	n.a	n.a	67
M10	<5-<8	6.5	n.a	n.a	7.0
140	53 - 55	n.a	n.a	n,a	51
120	>77.5	n.a	n.a	n.a	80
ASTM Stability	n,a	63.6	60	n.a	56
CRI	>60	74.1	61	50 - 65	65
CSR	20 - 30	17.7	23	n,a	20

n.a: not available.

<sup>a</sup> Díez et al. (2002).

Although lower I40 of 51 is reported in the current study as compared with European range of 53 - 55, this value is still within the usual range for coke quality used in coke making.

Coke size depends on fissured occurring in the coke. The mean coke size of 58 mm is however within the range of both the Japan and European Range. The Australian Port Kembla and American Range reported lower mean coke particle size values of than in the the current study. It is well known that coke oven flue temperature is one of the factors controlling coke size (Nomura and Arima, 2013) which was found to be fairly stable in the current study.

As shown in Table 8, a stability of 56 reflecting the load carrying strength or impact resistance of the coke for the current study, was shown. Although the current study reported less coke stability among other blast furnaces operations, the reported value has improved from the base coal blend and meets the requirement for the current blast furnace operation in the country (Mangena and du Cann, 2007). Furthermore, the current study results are in good agreement with findings presented by DuBroff et al. (1985), who mentioned that the stability index of about 50 - 60 is preferred for an acceptable strength metallurgical coke.

The CSR and CRI are valuable parameters used to evaluate the behaviour of coke in blast furnace. Therefore, CSR and CRI were also determined for all the samples. Fig. 3 shows the relationship between CRI and CSR of cokes produced as a function of the amount of carbonisation tar added to the blend. CRI of 65 against a target of 60 was achieved at the optimum 6 wt% carbonisation tar addition. Therefore, the coke from carbonisation tar addition follows the general trend observed for the CRI and CSR of the blast-furnace cokes: i.e. the lower the CRI, the higher the CSR index. Fig. 3 shows a high degree of correlation (i.e.  $R^2 = 0.986$ ) between CSR and CRI indices achieved with carbonisation tar addition. The results are in good agreement with the previous findings by MacPhee et al. (2009) who stated that the CSR drop is accompanied, as expected, by a concomitant rise in CRI. Even though CSR values for the current study indicated lower values as opposed to other regions as reported by Díez et al. (2002), this value is within the usual range of coke of acceptable quality.



Fig. 3. The relationship between CSR and CRI indices due to carbonisation tar addition.

# 4. Economic evaluation of carbonisation tar addition

In order to assess the economics of carbonisation tar addition, a schematic layout of the process was designed, based on the required steps to facilitate metallurgical coke production from the carbonisation tar addition technology, as shown in Fig. 4 depicts schematic layout of the process based on the additional equipments required to facilitate metallurgical coke production. The manufacturing process involves equipment such as carbonisation tar storage tank (1), carbonisation tar filters (10), and canopy with sparing nozzles (22) and coal conveyers system (23) as well as associated connecting pipes.



Fig. 4. Layout for carbonisation tar addition plant.

Table 9 shows the initial investment costs for the carbonisation tar addition plant. The costs of the equipment used were estimated based on South African market prices. The equipment was not scaled up or down since it was not different with the required size of the process. The total capital investment of almost USD 370, 000.00 includes all costs incurred to purchase the equipment needed for the control system (purchased equipment costs), the costs of labour and materials for installing that equipment (direct installation costs) and certain other costs (indirect installation costs). Operational costs were broken into maintenance costs – which include cost of keeping equipment in good working condition by undertaking regular repairs as needed, training and miscellaneous. The major costs of coke production through carbonisation tar addition related to site preparation, carbonisation tar storage and transportation as well as capital costs of such an equipment.

#### Table 9 Initial investment costs for carbonisation tar addition plant set-up.

	No	Item Description	2018 Cost (USD)
I. Equipment	1,	Carbonisation tar storage tank	31,250.00
	2.	Bund Wall	31,875.00
	3.	Motor-pump unit	65,625.00
	4	Hand Shut-off valve	812,50
	5.	Drain valve	812,50
	6.	Non-return valve	812,50
	7.	Pressure gauge	437.50
	8.	By-pass valve	812,50
	9.	Hand valve	812,50
	10.	Filters	20,687.50
	11.	Hand valve	5,625.00
	12.	32 mm pipe	15,625.00
	13.	Auto-shut off valve	1,275.00
	14.	By-pass valve	812,50
	15.	Flow control valve	1,275.00
	16	Manual control valve	812,50
	17.	Pressure gauge	437.50
	18.	Flow meter	1,875.00
	19.	Programmable Logistic Controller	9,375.00
	20.	Manual shut off valve	812,50
	21.	Weighing scale	1,875.00
	22.	Canopy with nozzles	3,125.00
	23.	Tar resistant Conveyor Belt	25,000.00
	24.	From Carbonisation tar Storage	1,500.00
	25.	Hand valve	812,50
	26.	Level Indicator	1,875.00
		Total I	226,050.00
II. Project Costs		Delivery of tank to site	4,150.00
		Cranage for offloading	1,512,50
		Labour for Bund area	3,125.00
		Labour for installation	
		Safety Requirements	625.00
		Paint and consumables	637,50
		Total II	65,050.00
III. Operations Costs		Maintenance costs	62,500.00
		Training	9,375.00
		Miscellaneous	6,250.00
		Total III	78,125.00
Grand Total USD		Γ/	369,225.00

The financial viability inputs were summarised in Table 10. The international bulk coking coal prices are usually quoted by independent media organisations in USD/T, representing spot prices for loading or delivery within 90 days, with standardise specifications relating both to cargo size and location (Australia, China, India, Japan, Colombia, Europe, and the USA) and also for coal qualities Matyjaszeka et al. (2018). The costs of imported coals were weighed against coke yield. Carbonisation tar yield is another important factor in assessing the feasibility of adopting this method as it would have a marked effect on the overall economics. It is shown in this study that for the optimum condition achieved, coke yield decreased by 2 wt%. Since raw materials prices vary greatly with the international market fluctuations, the production costs were determined in a range according to the maximum and minimum values of the annual average raw material over the last ten years (from 2008 to 2018). Prices quoted for coking coals are as delivered prices, whereas the price quoted for coke excludes transportation. Scholtz et al. (2006) mentioned that the coke manufacturing process is a continuous production operation and as such costs related to carbonisation tar addition were calculated using this basis. The capacity of the plant was estimated to be 950 tons/day (Scholtz et al., 2006), when operating 85% of the time which then corresponds to 310 days per year. Each coke oven was found to produce approximately 18.62 tons of coke (Scholtz et al., 2006) and the production was planned for 22 days per month excluding planned maintenance. It was determined that the plant would produce 245,784.00 ton/year, resulting in a considerable savings per month estimated as indicated in Table 10.

#### 4.1. Net present value

The sum of discounted net cash flows derives the Net Present Value (NPV). The NPV allows for simulations in which multiple uncertainties and risk factors are taken into account. In other words, it calculates the NPV of all cash flows in a carbonisation tar addition project and focuses directly on the profitability of a project instead of only its costs. The NPV is one criterion by which to simultaneously examine costs (cash outflows) and revenue (cash inflows). Estimated daily profit of USD 7,000.00 is made due to the difference in blend saving (USD 11, 194.16) and decrease in coke throughput (USD 4,512.50) as a result of carbonisation tar addition. Carbonisation tar addition results in savings of USD 150,000.00 per month, amounting to USD 1,7 million per year. The reported price was the minimum at which the coke production investment could make optimal profits. Although coke yield decrease of about 2% was reported, major savings have risen as a result of carbonisation tar addition to coal blend. Given that initial investment of USD 370,000.00 was used and an additional saving of USD 150,000.00 per month was realised.

#### 4.2. Break-even

Break-even (BE) analysis is used to determine the minimum level of coke sales that ensure the project is not experiencing any loss. According to Tsorakidis et al. (2000), break-even is the analysis of the level of sales and can be calculated using Eq. (2):

$$BE = \frac{F_C}{(S_P - V_P)} \tag{2}$$

where BE = Break-even;  $F_C = fixed cost$ ,  $S_P = Selling price$ ,  $V_P = variable price$ .

#### Table 10 Coke production 2018 investment annual operating costs.

Description	Unit Cost	0 wt% Tar		6 wt.% Tar	
	USD/Ton	Composition	Weight (Ton)	Composition	Weight (Ton)
Coal B	231,25	8%	92	7.3%	83.95
Coal C	225.00	38%	437	34.5%	396.75
Coal D	218.75	19%	218.5	17.2%	197.80
Coal A	118.75	35%	402.5	35%	402.50
Carbonisation Tar	120.00				
Carbonisation tar Added	61.62	0%	0	6%	69.00
Coal Blend per Oven			23.00		23.00
Coal Blend per 50 Ovens			1150		1150
Coal Blend Cost USD		215,193.75		203,999.60	
Blend Saving USD		_		11,194,16	
Coke per Oven			19.00		18.62
Coke Yield		69%		67%	
Coke Production/Day			950		931
Coke USD	237.50	225,625.00		221,112.50	
Production Lost USD		_		4,512,50	
Savings/Day USD		_		6,681.66	
Saving/Month USD		_		146,996.47	

The results are presented in Table 11. Table 11 shows that as long as coke production and sales are above 6.3 Months, coke making industry will experience profit and the returns on investment will be within the same period.

Range	1 Month	6.2 Month	6.3 Month
Coke sold (237.50 USD)	619 (Ton)	3837 (Ton)	3 899.70 (Ton)
Total coke Sales (USD)	146, 996, 52	911, 477.50	926, 178.75
V <sub>P</sub> (USD)	88, 197.91	546, 886.50	555, 707.25
Contribution Margins	58, 798.61	364, 591.00	370, 471.00
FC (USD)	369, 225.00	369, 225.00	369, 225.00
Profit/Loss (USD)	(-310, 426.39)	(-4, 634)	(1,246)

Table 11 Profit/Loss margin for different coke volume sold.

V<sub>P</sub> = variable cost; USD = United States Dollar; F<sub>C</sub> = Fixed carbon.

#### 4.3. Discounted payback period

Discounted payback period is the number of years that will pass before the investment cost is recovered and it is calculated while accounting for the time-value of money. The discounted payback period works as a quick assessment of the time period during which an investor's capital is at risk (Short et al., 1995). The discounted payback period is determined by Eq. (3):

$$NPV = \sum_{j=0}^{N} \frac{S_j}{(1+c)^j}$$
(3)

where j = analysis year.

where j = analysis year.

NPV = net present value of the capital investment,

 $S_j = \text{cash flows received at time } j$ ,

c = rate that equates the present value of positive and negative cash flows, when used as a discount rate.

Approximately, 6.3 months payback period is required to recover the initial investment cost used for carbonisation tar addition project. In addition, it is expected that maximum of up to 50% of carbonisation tar added be recovered after carbonisation (Chatterjee and Prasad, 1982).

#### 4.4. Sensitivity analysis

Sensitivity analysis was performed to understand the influence of the uncertain variables on the NPV value of the three carbonisation production processes. In this study, variations in the purchase price of carbonisation tar and the selling price of coke were investigated based on a 20 year plant life. The prices of all these parameters were varied between -50% and 50% of the original values and the results are as shown in Table 12.

 Table 12

 Sensitivity analysis values of varied price parameters for carbonisation process.

Range	0%	50%	-50%
Saving/Month USD	146, 996.47	220, 494.69	73, 498.23

# **5.** Conclusions

Carbonisation tar addition over 2.0 - 8.0 wt.% range was studied as a possible route for recycling coke oven tar waste into a coal blend. The use of carbonisation tar for coal blends is selected as an alternative to its disposal. The effects of carbonisation tar addition on coal blend proximate analyses were evaluated. The ash content, sulphur content and volatile matter content of individual coking coals making a blend were found to be additive. Carbonisation tar addition reduced ash content in the coal blend from 9.20 to 8.63 wt.%. A 56.8% reduction in coal fines (powder) was observed which consequently resulted in less coal dust pollution to the encironment. This occurred even when lower quality coking coal of up to 35% was included in the blend. Coke quality results are comparable to international benchmarks and mathematical models used to calculate the cost-effectiveness of such a project. Savings of up to USD 1.7 million per year were postulated and supported by the results. Recycling carbonisation tar has a great realistic significance in saving coal resources and protecting both the environment. Therefore future work should focus on quantifying the impact of coke oven gas emissions as a result of carbonisation tar addition.

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# Nomenclature

ASTM	American Society for Testing and Materials
с	rate that equates the present value of positive and
	negative cash flows
BE	Break-even
CRI	Coke Reactivity Index
CSR	Coke Strength after Reaction
db	Dry Basis
ddpm	Dial Division per Minute
FC	fixed cost
FC	Fixed Carbon
FID	flame ionization detector
FSI	Free Swelling Index
GC-MS	Gas Chromatograph coupled with Mass Detector
h	hour
i.d	internal diameter
ISO	International Organization for Standardization
i	analysis year
Max C	Maximum Contraction
Max D	Maximum Dilatation
MF	Molecular Formula
MIT	Matter Insoluble in Tar
MW	Molecular Weight
n	is the number of coals blended
n.a	not available
NZ	New Zealand
NPV	net present value of the capital investment
QI	Quinoline Insoluble
RSA	Republic of South Africa
RT	Retention Time
RoVr	Reflectance
S	Sulphur content
SANS	South African National Standard
SD	Standard Deviation of Reflectance
SG	Specific Gravity
Si	cash flows received at time j
Sp	Selling price
USA	United States of America
USD	United States Dollar
UV	Ultraviolet
VM	volatile matter
vol%	Volume percentage
VP	variable price
wt.%	Weight percentage
Zi	the property such as ash content, sulphur content and
-	volatile matter content of the coal blend
$Z_a$ and $x_i$	the property and the percentage mass fraction of coal <i>i</i>
	respectively
	- <b>-</b>

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