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Influence of coal properties on coal conversion processes-coal carbonization, carbon fiber production, gasification and liquefaction technologies: a review

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Abstract Coal due to its relatively large quantities and wide distribution worldwide has generated renewed interest in research and development with the aim of establishing coal conversion technologies that are technically reliable, environmentally and economically feasible. It has proved to be a prominent energy source in emerging markets with increasing energy demand by accounting for the largest increase in the demand of energy amongst all other energy sources. Furthermore, with its higher mesophase content, coal tar is an appropriate raw material for precursors in the production of carbon fiber. However, whenever a material is put to use, it is important to be able to associate its properties to the behavioral characteristics during a conversion process so as to have a basis for opting for the material in a given process or adjusting the operating conditions in order to optimize the material utilization. Therefore, as with any other material, it is important to be able to relate the properties of coal to its utilization. A review was carried out on the influence of coal properties on four main utilization technologies: gasification, carbonization, liquefaction and carbon fibre production. Among several properties rank, type, mineral matter content, distribution of trace elements, structural composition and pore structure were found to be most influential on the behavior of coal during conversion processes.

Keywords Coal properties · Carbonization · Liquefaction · Gasification · Carbon fibre

1 Introduction

Coal is a combustible sedimentary rock formed from ancient vegetation which has been compressed between other rock strata. The vegetation was accumulated under conditions that either limited or inhibited its decay. Although it is a rock with one evolution process, it is very heterogeneous in nature and the heterogeneity is brought about by the kind of vegetation deposited (coal type), degree of coalification (coal rank) and range of impurities (coal grade) (Edgar 1983; Warwick 2002; Suarez-Ruiz and Crelling 2008). The rank, type and grade are highly influential on the structural composition of coal which can be described as a cross-linked macromolecule comprising of heterocyclic monomers joined by covalent and non-covalent bonds (Marzec 2002). The structure is made up of aromatic layers consisting of aromatic rings ranging from one to three rings and linked by aliphatic chains, as shown in Fig. 1. During coalification, the chemical structure gradually changes to an aromatized structure, acquiring more stability and compactness as the methyl carbons present in the aliphatic chain changes to aromatic carbons thus reducing the spacing between the aromatic units (Blayden et al. 1944; Hirsch 1954; Cartz and Hirsch 1960; Manoj and Kunjomana 2012; Manoj 2014, 2016; Ahamed et al. 2019). As this happens moisture and volatiles are lost while carbon and the energy given out per unit weight of coal increases resulting in different classes of coal, as shown in Table 1.

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Table 1 Coal properties for different coal ranks (Orem and Finkelman 2003; Miller 2011; Speight 2015)

Rank	Proximate properties (%)				Ultimate properties (%)					Atomic ratios		Calorific value (MJ/kg)
	Moisture	Fixed carbon	Volatile matter	Ash	Hydrogen	Carbon	Nitrogen	Oxygen	Sulfur	H/C	O/C	
Anthracitic	3–6	> 98	< 2	4–15	1.5–3.5	75–85	0.5–1	5.5–9	0.5–2.5	0.25	0.05	30.23–34.89
Metaathracite		92–98	2–8							0.25–0.5	< 0.05	
Anthracite		86–92	8–14							0.5	0.05	
Semianthracite												
Bituminous	2–15	78–86	14–22	4–15	4.5–6	65–80	0.5–2.5	4.5–10	0.5–6	0.5–0.75		> 32.56
Low volatile											0.05–0.1	
Medium volatile		69–78	22–31									
		< 69	> 31							0.6–0.75	0.1	
High volatile A												30.23–32.56
High volatile B												24.42–30.23
High volatile C												
Subbituminous	10–25	30–50	28–45	3–10	5.5–6.5	55–70	0.8–1.5	15–30	0.3–1.5	0.75–1	0.15	24.42–26.74
Subbituminous A												22.09–24.42
												19.3–22.09
Subbituminous B												
Subbituminous C												
Lignite	25–45	25–40	24–32	10–50	6–7.5	35–45	0.6–1.0	38–48	0.3–2.5	0.8–1.2	0.2–0.5	9.3–19.3

The chemical constituents present in coal structure impact greatly on its mechanical behavior. The composition of organic matter and its degree of decomposition led to varying coal strength and modulus of elasticity of different coals (Pan et al. 2013; Singh and Mrityunjay 2018b; Ahamed et al. 2019) as the chemical structure was altered. Coals of low rank have a high concentration of aliphatic carbon (C_{al}–C_{al}) bonds while those of high rank are highly concentrated with the aromatic—aliphatic carbon (C_{ar}–C_{al}) and aromatic carbon (C_{ar}–C_{ar}) bonds. These aliphatic and aromatic carbon bonds have different bond energies with the aromatic carbon bonds possessing high bond energies to a maximum of 450 kJ/mol while that of aliphatic carbons is 300 kJ/mol (Li et al. 2015; Ahamed et al. 2019). The increase in bond energies affects the structural skeleton strength and hence coal strength increases as coal matures. Also, the reduction in the aliphatic side compounds and the inter-layer space between the aromatic carbons resulting in reduced space between the aromatic units contribute to the level of condensation and solid form of the chemical structure (Manoj and Kunjomana 2012; Manoj 2016). It is this densely packed coal matrix together with the enhanced bond energies that result in improved coal mechanical properties.

Calcite minerals found in cleats or fracture infillings reduces the inter-granular stresses between particles and this affects the frictional and cohesion strength of coal by increasing coal porosity and permeability (Ward 2002; 2016; Susilawati and Ward 2006; Lebedev et al. 2014). However, under dry conditions calcite minerals improve Young Modulus of Elasticity of coal because of its high resistance. They have resistance higher than that of coal material which tends to improve the overall stiffness of coal material (Gao and Kang 2017).

Clay minerals normally found in cleats, fractures or pore spaces are susceptible to swelling with water adsorption leading to fracturing either on the mineral or maceral phase. The fracturing uses up the elastic energy stored within the coal structure and its reduction causes a decrease in the mechanical strength of coal (Faraj et al. 1996; Ward 2016; Xue et al. 2017).

The complexity of coal and variance in properties has necessitated the need to understand these properties and appreciate their influence on different coal utilization processes (Sprunk 1942; Durie 1982). Not only do they vary across different ranks but also in iso ranks. Properties of coal play a significant role on the behaviour of coal during conversion processes as they define its nature. They are also important in establishing its value for different end-use applications (Sprunk 1942; Mitchell 1997; Taylor et al. 1998; Holuszko and Mastalerz 2015). The article conducted a review of numerous relevant literature material, inclusive of those reported since 1942 to the recent material as an endeavor to capture an update on coal properties in relation to its usage.

2 The role played by coal properties on coal utilization technologies

2.1 Carbonization

Coke is vital during metallurgical processing as it acts as a fuel source, spacer in the blast furnace, reducing agent and carburizes the reduced iron. These roles are quite challenging as coke gets exposed to severe thermal, physical and chemical conditions that no other material is capable of (Devasahayam and Sahajwalla 2013). As a result of this, it is important to control the quality of coke through proper coal selection since carbonization is highly influenced by coal properties. Several coal properties are essential for the accomplishment of the carbonization process.

2.1.1 Organic composition

The organic composition of coals comprises of macerals which are organic substances derived from plant material. The degree at which the macerals have metamorphosed during coal formation era affects the properties of the produced coke (Jasienko et al. 1981; Jee 2012; Devasahayam and Sahajwalla 2013) as it impacts on the ability of coal to melt when subjected to increasing temperatures and forming a coherent residue on cooling. The comparative proportions of both reactive and inert constituents in macerals are influential on the strength of the final carbonized product. Some of the maceral components will melt upon heating making coal to act as a pseudo-liquid material at various temperature ranges thus affecting the strength of coke and coking operations (Patrick 1974). Knowing the coal rank is important because as the quality of coal improves, it molecular structure gets ordered with an increase in aromatic carbon structure while the aliphatic carbon structure decreases (Dı´ez et al. 2002; Jee 2012; Devasahayam and Sahajwalla 2013). The ordered structure means more of the unreactive structure as the aromatics are more stable and will need improved conditions for reactions. Due to this, high-rank coals are inappropriate for the carbonizing process as it requires more reactive material while low-rank coals with more aliphatic structures are preferable.

2.1.2 Chemical composition

The content of volatile matter together with fixed carbon determines the yield of tar, oil, and coke. High volatile content will result in coal yielding large amounts of oil and tar. The increased amount of volatile matter is due to the presence of resins and spores with unusually high volatile compounds. Coals considered good for coking contain volatile matter in the range of 15% to 45% (Sprunk 1942; Jasienko et al. 1981; Devasahayam and Sahajwalla 2013). Increased quantities of fixed carbon in coal will yield large quantities of coke. The production of coke is directly related to the amount of fixed carbon regardless of the type of coal being carbonized.

Coal with high calorific value, carbon and ash content as well as high fusion temperatures results in the production of coke that is slightly harder and less friable (Gupta et al. 2005). While increased oxygen content results in low quantities of tar being distilled as compared to those distilled from coals containing low oxygen with the same amounts of volatile matter mainly because oxygen tends to act as an inert material.

Coals with high moisture, nitrogen, hydrogen, and oxygen content result in the distillation of high volumes of coke gas. It can therefore be concluded that the yield of coking gas increases with an increase in the volatile matter. However, the coke produced from these properties have the tendency to expand and it is this expansion factor that should be given detailed/comprehensive attention during coal selection since high expanding coals can cause damage to coke ovens (Sprunk 1942; Patrick 1974). Low moisture content leads to an increase in bulk density of coal and the degree of rocking attained inside the oven chamber thus contributing to the enhancement of coke quality. Dry coal charging processes capable of reducing moisture in coal by a maximum of 5%–6% has been developed (Nomura et al. 2004). This has resulted in less heat being used during carbonization, increased productivity and improved coke quality being achieved.

Small amounts of sulphur and phosphorus in coal is vital for metallurgical processing as both components are highly deleterious affecting the quality of products. Sulphur impacts negatively on the mechanical characteristics of steel by inducing brittleness during the heated state (red-shortness). While phosphorus tends to increase the development of brittleness in a material at reduced temperatures (cold-short) (Devasahayam and Sahajwalla 2013).

2.1.3 Inorganic composition

Great attention needs to be given to the composition of ash, most particularly the presence of Al_2O_3, SiO_3 and alkalis. Silica strongly affects the silicon content of hot metal while alumina affects the fluidity of slag (Ward 2002; 2016; Devasahayam and Sahajwalla 2013). Attempts have been made to decrease the melting point of coal and coke as well as the silicon content of molten metal by simultaneously injecting pulverized coal and dolomite (Yamagata et al. 1990). This was to address the inactiveness of the deadman resulting from the formation of material with high melting temperatures outside the raceway. The presence of alkalis in ash leads to their sublimation in areas of high temperatures in the furnace. They then re-condense in areas of reduced temperatures decreasing the mechanical strength of coke at the bottom part of the furnace, forming scaffolds and attacking furnace lining refractory (Ward 2002; Matjie et al. 2011; Creelman et al. 2013).

2.1.4 *Mineral matter content*

Ash content increases the fuel consumption of the blast furnace by affecting the heat balance in the furnace as the carbon content and calorific value of coal are reduced by the requirement of additional fluxes essential for removing ash as slag (Oshnock 1995; Devasahayam and Sahajwalla 2013). Also experienced is the negative effect on the stability of the blast furnace operation because of the catalytic influence on the reactivity of coke and the alkali attacks (Berkowitz 1979; van Krevelen 1993; Ward 2002; Miller 2017).

2.1.5 *Coal grindability and handleability*

Coal grindability determines the amount of power and throughput required to crush coal to the targeted size by a system used for preparing coal. This property is directly associated with the composition of macerals. It is also influenced by the rank of coal and the amount of mineral matter contained in coal. Low volatile metallurgical coals having a grindability index of 80, get easily milled thus requiring less power for a given coal throughput. Also, they allow for an increase in milling capacity hence an increase in rates of injection without added capital expenses on the added milling capacity (Anon 2 2001).

As the injection rates increase the handleability of coal gets increasingly important because the injection system at high injection rates gets easily affected by coals with poor flow and handleability characteristics. Poor coal handleability in most cases leads to transfer lines getting blocked for the dense phase in the transport system (Bennett and Holcombe 1994) as well as jeopardizing the capability of the system to ignite. In order to assess the handleability, experiments have been conducted (Black et al. 2005) to predict the characteristic behavior of bulk material in hoppers through assessment of characteristics such as friction of walls, compressibility, permeability and cohesion.

2.2 Carbon fibre production

Carbon fibre can be produced through the distillation of carbonaceous natural resources like coal and petroleum to attain pitch as a precursor. Pitch extracted from coal, a by-product of the carbonization process during the production of metallurgical coke, constitutes a valuable aromatic hydrocarbon source essential as a raw material for manufacturing different carbon materials. The carbon materials are derived from the thermal transformation of its highly distributed macromolecules containing high aromatic carbon content and aliphatic constituents substituted by large polycyclic aromatic hydrocarbons (PAH) (Kim et al. 2016a, b; Yang et al. 2016; Apicella et al. 2017; Hiremath et al. 2017). However, scarce information on the chemical structural composition of the precursor pitches which is influential on the yield, quality of the resultant carbon material and that of final products attained from their thermal decomposition/treatment makes it a challenge in manufacturing carbon materials. Several studies have been conducted on carbon fibre production using coal-tar pitch as raw material (Table 2).

Successful production of CFs is dependent on the spinning process which in-turn is influenced by coal parameters such as chemical structural composition, structural molecular weight, and proximate properties.

2.2.1 *Chemical structural composition*

The oxygen bonding structure is important in the production of CFs due to its effect on the composition of pitch used in CF production. Oxygen atoms in coal extract with high oxygen ratios are not directly bonded to the aromatic carbons, which are the major constituents of the pitch. Rather, they are bonded to hydrogen atoms when the energy available for a reaction is relatively low. The oxygen atom positioned at the CH-chain tail such as in the structure formation of low ranked coals makes the coal increasingly reactive and it is this high reactivity that makes it extremely difficult to spin fibres from this type of coal (Fathollahi et al. 2005; Takanohashi et al. 2008; Lee et al. 2019). Also of importance is the double bond of the carbon–carbon atoms. A strong double bond is indicative of a strongly formed aromatic structure that is highly stable. However, this high aromatic structure with solid contents only tends to make it challenging for coal to attain the structural chain required for a pitch with spinnable behavior (Cooke et al. 1986; Odeh 2015; Kim et al. 2014, 2016a, b; Lee et al. 2019).

The aliphatic low molecular components i.e. the methyl groups are essential for pitch spinnability. The carbons connecting the two aromatic rings like a bridge result in the production of CFs with thinner diameter because of the

Table 2 Studies conducted on carbon fibre production using coal-tar pitch as raw material

Reference	Study	Objective	Method	Result
Kim et al. (2016b)	Pitch based carbon fibers from coal tar or petroleum residue under the same processing conditions	Establish the relationship between the raw materials and the properties of the produced carbon fiber	1. Elemental analysis 2. Structural analysis 3. Molecular weight analysis 4. Thermogravimetric analysis 5. Analysis of mesophase development 6. Scanning electron microscope for fiber diameter	1. Petroleum pitch had high carbon, hydrogen, sulfur content, high H/C atomic ratio, low aromaticity with aliphatic hydrocarbon chains of methyl/methylene, low molecular weight substances, 33.5% yield 2. Coal-tar pitch had higher carbon, nitrogen content, low sulfur, hydrogen, H/C atomic ratio; high aromaticity with per condensed aromatic carbons and aromatic carbons bonded to aliphatic chains with small amounts of methyl/methylene carbons; broad molecular distribution,57.3% yield 3. Both produced carbon fibers with uniform morphology and clear surfaces
Apicella et al. (2017)	Analysis of petroleum and coal tar pitches as large PAH	Analyze and compare chemical features of petroleum and tar pitches with different physico-chemical properties	1. Elemental analysis 2. Thermogravimetric analysis 3. Mass distribution analysis 4. Structural analysis	1. Petroleum pitch had two compounds volatilizing at different temperatures; large amounts of methylated PAH and aliphatics 2. Coal-tar pitch had high amounts of unsubstituted PAH from 4 to 7 rings, abundance of lighter PAH, higher aromatic content 3. Both had low N and O content, H/C atomic ratio
Zabihi et al. (2019)	Low-cost carbon fibre derived from sustainable coal tar pitch and polyacrylonitrile: fabrication and characterization	Prepare electro spinnable solutions through CTP to PAN variations for low cost CF production	1. Viscosity analysis 2. Fiber diameter analysis 3. Thermogravimetric analysis 4. Surface area analysis 5. X-ray diffraction measurements 6. Molecular weight distribution	1. Addition of CTP to PAN resulted in significant increase of fiber diameter, precursor viscosity, heat of reaction, specific surface area and pore sizes, carbonization temperature hence increased carbon yield 2. Addition of CTP to PAN decreased electrical conductivity and resistivity 3. No significant change in stabilization temperature and crystallographic planes although evolution of crystallographic structure changed after carbonization at 1200 °C 4. More uniformity in produced fibers was attained for 25% CTP while 50% CTP decreased the uniformity

Table 2 continued

Reference	Study	Objective	Method	Result
Yang et al. (2016)	Preparation of pitch based carbon fibers using hyper-coal as a raw material	Prepare optimal pitch precursor for isotropic CF production by removing volatile matter and increasing softening temperature of spinnable pitch through 1-methylnaphthalene treatment	1. Softening temperature analysis 2. Elemental analysis 3. Structural composition analysis 4. Thermogravimetric analysis 5. Molecular weight distribution analysis 6. Analysis of CF morphology 7. Tensile strength analysis	1. Resultant pitch from bituminous coal had low oxygen content, reduced aromaticity, average molecular weight 2. Resultant pitch from sub-bituminous coal had high oxygen content, lower aromaticity, lower average molecular weight, higher yields 3. Both pitches had decreased softening points, increased contents of aliphatic carbons 4. CFs from the prepared pitches had smooth, homogeneous morphology with diameter of 9 µm 5. CF from bituminous coal tar pitch had higher tensile strength
Lee et al. (2019)	Preparation and characterization of high-spinnability isotropic pitch from 1-methylnaphthalene-extracted low-rank coal by co-carbonization with petroleum residue	Study the effects of co-carbonizing petroleum residue with coal extract from low rank coal on resultant pitch for isotropic carbon fiber production	1. Elemental analysis 2. Proximate analysis 3. Sulfur analyzer 4. Thermogravimetric analysis 5. Molecular weight distribution 6. Chemical structural analysis	1. Spinnability of the resultant pitch (200% PFO-pyrolysed fuel oil) improved significantly—low oxygen and moisture content, reduced C/H mole ration hence lower aromaticity, more developed aliphatic structure 2. Fiber surface cracking was improved hence an improvement of mechanical properties—attained CF diameter 7 µm and tensile strength of 1189 MPa 3. More PFO increment led to decreased softening point and fixed carbon content, high content of lower molecular weight compounds which might lead to fiber breakage

Table 2 continued

Reference	Study	Objective	Method	Result
Huang (2009)	Fabrication and properties of carbon fibers: review	Carry out a review on the research and development activities that has been previously conducted on carbon fibers	1. Carry-out a review on the structure and composition of polyacrilonitrile and mesophase pitches and how they affect the quality of the produced carbon fibers 2. Investigate the process optimization techniques	Structure and composition Crystalline morphologies and defects of fibers are the principal factors affecting the strength of produced carbon fibers 1. Increased crystallinity and well aligned crystals lead to carbon fibers with high modulus of elasticity 2. When Young's modulus is improved in the fiber direction then the absolute coefficient of linear expansion increases 3. When the layers are aligned in parallel along the fiber axis the resultant CF portrays excellent and electrical conductivities 4. Increased content of carbon particle in the pitch results in filament breakage when extruding and thermal treating 5. Characteristics appropriate for pitch fiber spinning are 55% to 65% mesophase content, homogeneous mesophase containing huge coalesced domains having 200 microns in excess and produced from quiescent conditions 6. Pitch with small amounts of solvent in its liquid crystalline structure tends to melt at lower temperatures enabling easier spinning and the resultant CFs have improved melting points 7. Methyl groups in pitch lowers the softening point and improves spinnability and stabilization 8. In-order to avoid fiber breakage when spinning the diameter for the precursor fibers need to be relatively large Optimization 1. Since layer alignment of the precursor fiber is maintained in the produced CF the strength of the CF can be improved through adjustment of the precursor fiber microstructure 2. Subjecting the carbonized fibers to elevated temperatures of up to 3000 °C will assist in attaining an increased carbon content and increased Young's modulus in the direction of the fiber

Table 2 continued

Reference	Study	Objective	Method	Result
Kim et al. (2016a)	Enhancing the tensile strength of isotropic pitch-based carbon fibers by improving the stabilization and carbonization properties of precursor pitch	Produce an isotropic pitch-based CF having excellent tensile strength and elongation properties	1. Thermo-mechanical analyzer 2. Molecular weight distribution analysis 3. Analysis of rheological properties 4. Molecular structure analysis 5. Tensile tester 6. Analysis of surface morphology CF diameter	1. NCO derived pitch and 50:50 NCO:CT both were composed of molecules having three to five aromatic hexagonal moieties 2. Blend of NCO/CT pitch had less aliphatic structures as compared to NCO derived pitch with NCO/CT pitch increased pitch yield 3. NCO/CT pitch had higher molecular weights and its aromatic units had bridges of methylene alkyl chains 4. NCO/CT pitch required the least energy to breakdown the physical structures (i.e. large yield stress values) indicating stronger physical structures due to higher molecular weight and aromatic composition 5. NCO/CT carbonization yield was much more than that of NCO based CFs 6. NCO/CT showed a high degree of shear thinning indicating high degree of molecular orientation during spinning hence improved mechanical properties of the resultant CFs 7. Adding CT to NCO improved the carbonization yield, excellent qualities of NCO/CT based CF were attained from small amounts of oxygen content in the pitch and its homogeneous distribution

improved linearity of the raw material (Kim et al. 2014, 2016a, b). It is important though that they remain in the structure without bridging because they turn into volatile material during spinning since they decompose easily at relatively low temperatures making it difficult to maintain the shape of the fibre (Yoshida et al. 1982; Okuyama et al. 2004; Takanohashi et al. 2008; Lee et al. 2019).

Minimal aliphatic structures present in coal, in particular, high-rank coal like bituminous coal and considered appropriate for carbonization, renders it a challenge spinning CF since high content of aliphatic carbons results in a linear structure which is more spinnable. Additional hindrance to the spinnability process is high content of aromatic carbons (Kim et al. 2016a, b; Lee et al. 2019). However, CFs with uniform and clear surfaces can be successfully manufactured from coal with high aromaticity and low aliphatic carbons by producing pitch with relatively high aliphatic content, high oxygen content and yet relatively low reactivity through chemical refining technique (Wang et al. 1998; Watanabe et al. 1999; Okuyama et al. 2004; Rahman et al. 2013; Yang et al. 2016; Kim et al. 2016a, b; Lee et al. 2019). Blending coal tar pitch with other precursors such as petroleum pitch and subjecting the coal tar pitch to solution or thermal treatment can improve the quality of the resultant precursor pitch (Tables 3, 4).

Table 3 General properties of spinnable pitches (Lee et al. 2019)

Raw material	C	H	N	S	O	O/C	C/H	SP	MW
Coal extract (untreated)	78.6	6.0	0.3	0.2	14.9	0.14	1.09		
Coal extract (treated)	87	6.8	0.0	0.0	6.2		1.06	70	672
Blend of coal extract and petroleum	89.7	7.5	0.3	0.2	2.3		0.99	55	609

Table 4 Aliphatic and aromatic fractions of pitch precursors (Lee et al. 2019)

Precursor pitch	CH_3	CH_2	$C_{\alpha 2}$	$C_{ar\ 1,2}$	$C_{ar\ 1,3}$	F_a
Coal tar (untreated)	1.4	1.4	1.4	21.8	74	0.96
Coal tar (treated)	5.53	8.17	1.2	25.32	41.02	0.85
Coal tar + petroleum	15.74	5.82	2.57	20.21	32.47	0.76

The aliphatic content was improved by blending and treating the coal tar pitch. Initially, the coal tar pitch had low aliphatic content, high aromaticity and oxygen content. Subjecting it to solvent/heat treatment and blending improved the aliphatic structures while the aromaticity and oxygen content was reduced thus rendering the resultant pitch appropriate for carbon fibre spinning. The morphology of the resultant CFs also was improved (Fig. 2).

Carbon fibres produced from different spinnable pitches had a smooth and uniform structure. The fibres showed no sign of fusing, an indication that the stabilization process was a success (Yang et al. 2016). However, the resultant CFs differed in diameters. Coal tar pitch yielded CF with a bigger diameter (15 µm), followed by a pitch from hyper coal at 9 µm and a blend of NCO and coal tar pitch at 7.6 µm. The tensile strength of hyper coal-based CFs was 1150 MPa while that of NCO/CT based CFs was 1800 MPa (Kim et al. 2016a, b; Yang et al. 2016).

Although blending of coal tar pitch with other precursors has been done to achieve the desired results, it can have adversarial effects on the quality of the resultant pitch. Studies conducted in the past (Giray et al. 2013; Hiremath et al. 2017; Zabihi et al. 2019) indicated that adding coal tar pitch to other precursors can drop their charge density consequently leading to a reduction in the repulsion forces hence a decrement in the conductivity of electricity. It also leads to an increase in pitch viscosity and this can be detrimental in achieving dense and smooth fibres. It has been revealed that the addition of more coal tar pitch to polyacrylonitrile (Huson 2017) had a negative impact on the produced carbon fibres (Fig. 3). There was a change in the colour of the fibre mats to brownish and the packing of the carbon fibres got thicker. Also affected was the morphology of the carbon fibers as it got uneven with a surface that has lost its smooth appearance.

The defects on the quality of the resultant CF has also been shown through the blending of coal tar and pyrolysis fuel oil (PFO) (Lee et al. 2019), as shown in Fig. 4. When the amount of pyrolysis fuel oil (PFO) was more in the blend, the thermal stability of the pitch was lowered resulting in fibre breakage during spinning, Fig. 4a. Although the pitch blend of 300% coal tar did not result in the breakage of the fibres, the surface of the CFs was affected, Fig. 4c. The effect on the shape of the fibres could have been due to the uneven distribution of molecular weight (Baumgarten 1971; Deitzel et al. 2001; Okuyama et al. 2004; Takanohashi et al. 2008; Zabihi 2012; Hiremath et al. 2017; Huson 2017; Lee et al. 2019) as the highly aromatic carbons in coal tar pitch mixed with and overpowered the aliphatic carbons in polyacrylonitrile/petroleum pitches hence poor spinnability.

2.2.2 *Molecular weight distribution*

Molecular weight influenced by aromaticity, is important in CF production. High carbon-hydrogen mole ratio indicative of high aromaticity results in large molecular weight due to the polycyclic structure. And because of this high molecular weight distillation is rendered ineffective during the preparation of spinnable pitches. The distillation temperature would, therefore, need to be increased in order to have improved yields of spinnable pitches (Kim et al. 2016a; Lee et al. 2019). On the other hand, the high content of low molecular weight compounds results in easy breakage of the fibers because of the volatile matter given out during the spinning process. Also, an inconstant molecular weight distribution results in the thickness of the fibre being non-uniform due to partial presentation of high molecular weight material (Lee et al. 2019).

2.2.3 *Fluidity*

Coal fluidity is essential during the preparation of the precursor pitch. A detailed attention is vital in controlling the development of mesophase (presence of liquid crystal in an isotropic phase and tends to increase pitch viscosity, Fig. 5). Existence of partial mesophase development can be detrimental to fibre spinning process because of variances in the density and viscosity of the co-existing isotropic and anisotropic phases (Park and Mochida 1989; Kim et al. 2016a, b; Yang et al. 2018). The viscosity of the pitch can adversely affect the initial shape of the droplet, path of the jet and fiber diameter. When the viscosity increases the fibre diameter increases thus negatively affecting the pitch spinnability (Deitzel et al. 2001; Zabihi 2012; Hiremath et al. 2017; Huson 2017).

Coal tar pitch presents a viable precursor alternative in that it facilitates the process of stabilization by increasing the heat that is released during the process thus lowering the energy consumed for stabilization of fibres i.e. minimized stabilization temperatures with high heat of reactions are essential for an efficient thermal stabilization process (Kissinger 1957; Zabihi 2012). In addition, its high aromaticity is indicative of high thermal stability, a quality essential for the steady spinning of the pitch possessing high softening point since melt spinning is normally carried out at a temperature 50 °C higher than the softening point (Kim et al. 2013, 2014; Lee et al. 2019).

2.3 Coal gasification

Coal gasification involves coal reaction with air, oxygen, steam or their mixture to produce a gaseous fuel termed synthesis gas. The principal controlling properties of coal during the gasification process are its reactivity, ash and slag properties, particle size and coal caking and swelling.

2.3.1 *Coal reactivity*

Coal reactivity is one fundamental property essential for the success of the coal gasification process (Ozer et al. 2017). It is influenced by the degree of coalification, organic composition (macerals), mineral contents, coal porosity and surface area.

(1) *Degree of coalification*
Degree of coalification is reliant on the organic composition of coal (macerals), chemical structural composition, mineral matter content and pore structure (Jones et al. 1985; Alonso et al. 2001; Méndez et al. 2003; Choudhury et al. 2008). Coal has been classified using the carbon content of its macerals into a low rank containing carbon content less than 80% and a high rank containing more than 80% carbon. The catalytic effect of coal minerals has been found to be the chief controlling factor of the reactivity for coals in the low rank while the reactivity for high ranked coals is mainly controlled by the available active sites found in the coal matrix (Miura et al. 1989). Low ranked coals contain high volatile material and more hetero-atoms inside its organic structure. Their structure also has more open pores and increased oxygen-functional groups as compared to high ranked coals. Furthermore, low-rank coals comprise of a poorly aligned aromatic structure as opposed to high-rank coals composed of better aligned carbon atoms. The amorphous and more open pore structure of low-rank coals makes it more reactive with oxygen and steam (Ozer et al. 2017).

The highly dispersed catalytic minerals, carboxylic acids as well as phenolic-functional groups also contribute to the high reactivity of low ranked coals (Radovic et al. 1983). Also accountable for this high reactivity is the highly concentrated oxygen-functional groups, increased amount of macropores and the distribution of the catalytic in-organic constituents. The functional groups provide the cations with reaction zones hence improving on the reactivity of coal due to catalysts (Ye et al. 1998). They can also act as active zones as well as exchange zones on which alkali elements can be mounted as exchangeable cations (Takarada et al. 1985). Previous studies (Irfan et al. 2011) revealed that once alkaline elements are removed from low ranked coal, its reactivity would be the same as the reactivity of high ranked coal.

The high moisture content of low ranked coals reduces the temperature generated in the combustion area thus decreasing the thermal efficiency of the gasification process. As a result of this, coal feed rates of low ranked coals need to be more than those of high ranked coals in-order to improve the quality of syngas by increasing the heating value (Durie 1982; Shadle et al. 2002; Ozer et al. 2017; Fletcher 2017).

Low rank coals contain high volatile matter as compared to high-rank coals. This characteristic tends to enhance coal reactivity because the activation energy would be lowered (Hakvoort et al. 1989; Küçükbayrak et al. 2001). It also improves on the carbon burnout, ease of ignition and has been used to predict the number of hydrocarbons that can be given out during the gasification process (Hattingh et al. 2011).

(2) *Organic composition (coal macerals)*

The organic structure of coal is made up of three maceral groups; vitrinite, inertinite, and liptinite/exinite. It is the composition of these constituents that is influential in the transformation of coal structure during the conversion process since maceral groups react differently (Suarez-Ruiz and Crelling 2008; Suarez-Ruiz 2012).

Behavioural characteristics of macerals during the coal gasification process are as followed:

① Vitrinite—the most abundant maceral group derived from woody tissues with qualities and the reactivity varying with the rank of coal. Their high hydrogen content has resulted in their capability to melt, swell and soften after coal has been carbonized. The mobile carbon undergoes nanoscale re-arrangements to align layers that will easily coalesce when subjected to heat treatment leading to the formation of extended layers as well as the loss of edge sites and that of reactivity (Beeley et al. 1996; Zhuo et al. 2000; Zhang et al. 2008). Coals rich in vitrinite produce char with high porous structure and large amounts of mineral matter mainly calcium and potassium improving its catalytic activity. Infact, vitrinite-rich char has high gasification reactivity regardless of the presence of minerals (Megaritis et al. 1999; Sun et al. 2004).

② Liptinite/exinite—this maceral group originates from hydrogen-rich sources like spores, leaf cuticles, plant algae and is very reactive. Although it fails to swell or agglomerate, it has the capacity to melt during heating and hence most of its mass proportion is lost during pyrolysis (Zhuo et al. 2000; Mahagaokar 2004).

③ Inertinite—the most unreactive maceral group rich in carbon and originates from biodegraded wood. Inertinite fails to melt when subjected to heating and would lose only a slight quantity of its mass through pyrolysis and as a result it is a poor gasification agent due to its unreactivity towards CO_2. Coals rich in inertinite possess a structure that is highly oriented with a high degree of crystalline packing (Sharma et al. 2000; Van Niekerk et al. 2008). They are also more aromatic with more polycondensed contents and have been found to be appropriate for combustion and gasification technologies (Sun et al. 2003; van Niekerk et al. 2008).

Inertinite and vitrinite are responsible for the carbon structure conversion of coal during gasification while the change in the mineral matter has been attributed to the presence of inertinite (Gupta et al. 2005; Matjie et al. 2011).

(3) *Porosity of coal and surface area*

Coals of low rank have a highly open porous structure with increased active sites for oxygen-steam reaction during the gasification process. A correlation has been established between the reactivity of coal and its micro porosity basing on the active surface area that is directly proportional to the entire surface area (Irfan et al. 2011). Reactions of gasification mainly take place on the surface of macropores instead of micro pores due to the highly concentrated active zones in the former than in the latter. In fact, macro pores have been found to materialize in crystallite edges or sites that have contact with organic impurities catalytically active whereas the micropores were found to be made up of less reactive basal planes (Hurt et al. 1998).

Also, there is a relation between active sites concentration or surface area reaction and the accessibility of reactants in the gaseous state to the active sites, indicating that the structure of pores can be linked to charcoal reactivity (Slaghuis et al. 1993; Ng et al. 1988; Kajitani et al. 2006; Çakal et al. 2007). When a critical value (70%–80%) for char porosity is attained fragmentation into finely grounded particles occurs improving the reactivity hence char conversion (Jayaraman and Gokalp 2015). Coals with low surface area and porosity have low reactivity (Adschiri et al. 1986; Arenillas et al. 2004; Wu et al. 2009; Ozer et al. 2017).

(4) *Mineral matter in coal*

The most common minerals found in coals used for gasification comprise mainly of kaolinite with small quantities of quartz, illite, dolomite, calcite, pyrite and small traces rutile and phosphate (Ward 2002, 2016; Matjie et al. 2011). Calcite together with dolomite appear within vitrinite macerals while Ca and Ti appears as inorganic elements that associate with the organic matter. Sodium, chlorine and sulfur were also found in some coals.

(5) *Effect of catalytic behaviour of coal minerals*
The interaction of mineral matter with the carbon matrix in coal has a great effect on coal reactivity. Mineral matter can be discrete grains, embedded within or attached to the carbon matrix as is the case with high-rank coals. The embedded mineral matter has a high specific heat capacity and absorb more heat leading to temperature decrease and subsequently low coal reactivity (Wigley et al. 1997; Hattingh et al. 2011). Mineral matter occurring as dissolved salts trapped inside the pores and/or are attached chemically to the carboxylic acids and the phenolic groups in the form of exchangeable ions have strong effect on char reactivity of low ranked coals than that of high ranked coals (Quann and Sarofim 1986; Huggins and Huffman 1995; Ballantyne et al. 2005).

Also of great importance on coal reactivity and hence coal gasification rate are the inorganic metal ions such as K^{2+}, Na^{2+}, Ca^{2+} Mg^{2+}, and Fe^{2+} as they get to trade ions with hydrogen ions (H^{+}) making up the carboxylic acids or phenolic groups. Ca^{2+} influences the syngas composition by promoting the water–gas shift reaction while Ca and CaO organically bound by calcite decomposition impacts on the catalytic effect during coal gasification (Radovic et al. 1983; Wigley et al. 1997; Skodras and Sakellaropoulos 2002; Chodankar et al. 2007; Hattingh et al. 2011; Ozer et al. 2017).

2.3.2 Properties of ash and slag

(1) *Mineral matter conversion into ash*
The fusing, melting and partial crystallization on cooling of mineral matter results in the formation of cristobalite, magnetite, mullite, diopside and anorthite. These new formed phases and their properties (chemical composition, fusion temperatures and slag viscosity) need to be taken into account to reduce risk of clinkering, ash deposits and agglomeration, fouling and corrosion. The occurrence of any would lower the efficiency and capacity of the gasification process resulting in an increase on the power cost (Zevenhoven-Onderwater et al. 2001). Coal components formed at temperatures lower than the ash melting temperature such as magnesia (MgO), ferric oxide (Fe_2O_3), silica (SiO_2), lime (CaO), alumina (Al_2O_3), titanium oxide (TiO_2), alkali compounds (K_2O, Na_2O), some sulphur and chloride compounds may react with each other under high gasification temperatures forming several different compounds with interstitial vesicular glass of calcium to iron-rich calcium (Hu et al. 2006; Kosminski et al. 2006a, b, c; Gupta et al. 2007; Bai et al. 2010).

(2) *Ash fusion temperature*
Ash fusion temperature defines the melting behavior of ash during the gasification process. When gasifying low-rank coals, the operating temperature needs to be less than the temperature at which the ash fuses to avoid molten fused deposits forming whereas for high rank coals the operating temperature needs to be higher than the temperature at which fusion of ash occurs. This would lessen challenges related to slag formation by allowing the melted heated ash (slag) to flow down the reactor walls and drain easily from the reactor (Li 2010). Ash fusion temperature was found to reduce with increasing SiO_2 wt % signifying its fluxing potential on ash and it increased with increasing Al_2O_3 content (Gupta et al. 1998).

(3) *Slag viscosity*
Coals with similar ash fusion temperature can have differing slag viscosities and portray different slagging behavioural traits because the ash fusion temperature is dependent on ash mineral composition. Therefore, ash having varying compositions may exhibit similar ash fusion temperatures but dissimilar slag viscosities (Gupta et al. 1998; Lolja et al. 2002; Trapp et al. 2004; Liu et al. 2013). Slag viscosity is highly influenced by the temperature and chemical composition of ash. When temperatures are low the slag gets more viscous with the poor erratic flow while high temperatures lead to fast refractory wear (Groen et al. 1998). Slag viscosity increases with increasing alumina and silica but reduces in the presence CaO, FeO, MnO_2 and MgO (Wang and Massoudi 2013).

(4) *Ash fouling, deposition and agglomeration*
Some mineral constituents can get entrained within the stream of the synthesis gas being deposited on the cool reactor surfaces or wall tubes causing corrosion, slagging and fouling. They can even negatively impact on the devices for emission control and other equipment (Vamvuka et al. 2008). The deposition of inorganics affects the transfer of heat within the reactor thus altering the operation and capacity of the reactor hence the system efficiency. Fouling and deposition challenges are promoted by the existence of the alkali metals (alkaline earth metals, chloride, silicon, Na) and sulphur in ash (Ward 2002, 2016; Creelman et al. 2013; Ozer et al. 2017). Furthermore, vapours due to alkali metals lead to the final end-use equipment

being corroded. The dominant alkali species are sodium and potassium hydroxides as well as chlorides. Small amounts of chlorine contained in some coals during the gasification process produce gaseous HCl that interact with metal compounds accumulating in syngas coolers leading to deposits of chlorides like $FeCl_2$, NaCl, and CaCl (Kosminski et al. 2006a). Ash agglomeration results in channel burning. It also leads to challenges related to pressure drop in the reactor and the reactor performance being unstable hence cut-backs of the gasification loading (van Dyk and Waanders 2007).

2.3.3 *Coal particle size*

Large sized particles have a tendency to reduce the reactivity of coal and hence poor syngas quality. Also experienced in relation to large sized particles are challenges with transporting the large particles in the reactor and start-up difficulties of the equipment. Gasifying low rank coals using large particle sizes can lead to the formation of clinker and channeling (Speight 2014). Fine coal particles, on the other hand, reduce carbon conversion thereby decreasing syngas production (Dittus and Johnson 2001).

2.3.4 *Coal caking and swelling*

Coal caking and swelling apply in particular to bituminous coal and it leads to particles adhering to one another and forming clusters to create a phase known as metaplast. The forming agglomerate phase can interrupt flow patterns of gas and thus reduce the thermal efficiency. However, the tendency of coal to cake reduces as the volatile matter content falls below 20 wt % (Shadle et al. 2002). The swelling of coal affects the reactivity as the operation of the reactor gets unstable due to fluctuations in pressure drop and channeling. Also, the size of coal particles and density of the produced char are affected (Jayaraman and Gokalp 2015). In addition, caking coals tend to form tars with high molecular weight which if not controlled can lead to blocking and fouling of lines, heat exchangers and filters thus tampering with the system sufficiency.

2.4 Liquefaction

Coal liquefaction involves the conversion of coal into liquids. There are two methods for liquefying coal; direct liquefaction, also known as hydrogenation involves the production of liquid fuels directly from coal using a hydrogen donor solvent and indirect liquefaction. Indirect liquefaction process coal is liquefied through the Fischer–Tropsch (FTS) process. The FTS process uses synthesis gas from the gasification process and converts it into liquidous hydrocarbons and associating oxygenated compounds using different catalysts from Group VIII. The product mixture depends on the carbon monoxide to hydrogen ratio (Berkowitz 1979; Durie 1982; Höök and Aleklett 2009; Khan 2011; Vasireddy et al. 2011; Heydari et al. 2016; Miller 2017).

Coal consists mainly of solid organic matter with an atomic hydrogen to carbon ratio ranging from 0.1 to 4. Therefore to produce liquids from coal requires breaking down the chemical structure into compounds with a low molecular weight that is in a fluid state under ambient conditions, eliminating heteroatoms in particular O, N and S and increasing the hydrogen to carbon ratio. These generate saturated aromatic compounds and stabilizes the resultant liquids (Durie 1982; Gibson 1983).

Different coal properties have varying significant impact on the coal liquefaction process. Comprehensive studies have been carried out on several chemical and physical characteristics of coal and their importance during the conversion of coal into liquid products (Table 5).

2.4.1 *Degree of coalification*

The degree of coalification is the most crucial coal parameter in liquefaction. Coal rank through vitrinite reflectance can be used to determine the potential of a coal seam for producing hydrocarbons. A model by Tissot and Welte (1984) was used to predict the primary hydrocarbon potential through the evolution stages of coal (Fig. 6). Coals which have passed the evolution phase of diagenesis (a phase when coal significantly loses oxygen as carbon dioxide and water) and reached to the catagenesis stage (when hydrogen and carbon are lost due to oil and gas generation when the organic matter under-goes thermal decomposition as temperature increases with burial in the sedimentary basins) have great potential for generation of hydrocarbons (Tissot and Welte 1984; Suarez-Ruiz and Crelling 2008; Suarez-Ruiz 2012; Singh and Mrityunjay 2018a, b).

A high liquefaction yield is obtained near the transition from brown coal to bituminous coal i.e. coals with carbon content in the range of 80% to 85% (Randolph 1944; Vasireddy et al. 2011). When the carbon content is between 87% and 89% (medium volatile bituminous coal), syncrude yield declines sharply. Coals of high rank with a carbon content greater than 90% (low volatile bituminous coal and anthracite coal) are mostly unreactive (Vasireddy et al. 2011). The yield of oil decreases and that of moisture and carbon dioxide increases with a decrease in the carbon content of the bright coal hydrogenated (Randolph 1944). Hydrocarbon yield is limited by high oxygen content approaching 30 wt% dmf. The oxygen reacts with carbon

Table 5 Coal parameters with significant influence on liquefaction process

Parameter	Techniques applied	Observation	Reference
Coal rank	1. Petrographic analysis 2. Proximate and ultimate analysis	1. More tar and oil can be distilled from coals with high volatile matter 2. Less tar and oil are distilled from high oxygen coals than from low oxygen coals of equal volatile matter content 3. Coals with high hydrogen content, high content of aliphatic structures, simple aromatic structures, low oxygen and nitrogen contents are preferable for the conversion of coal into liquid fuels 4. High oxygen content limits the production of liquid fuels because it reacts to form water and carbon oxides while hydrogen content of coal contributes the maximum to the generation of liquid hydrocarbons 5. Products of coal liquefaction decrease in quantity with an increase in the rank of coal. Coals with high carbon content are difficult to liquefy while those with low carbon convert more easily into liquid products 6. Yields of products with high viscosity is obtained from coals of low rank resulting in pumping difficulties 7. Although low rank coals give low yields, the fraction of low boiling distillates increase with decreasing coal rank. Their products, in particular from sub-bituminous coals are highly volatile with more saturated hydrocarbons and are less aromatic with higher hydrogen content and fewer heteroatoms	Sprunk (1942), Davis et al. (1976), Durie (1982), Bertrand (1989), Newman et al. (1997), Peters et al.(2000), Sykes (2001), Vasireddy et al. (2011), Singh et al. (2013), Speight (2014), Singh and Mrityunjay (2018a, b)
Organic composition	1. Microscopic investigation 2. Megascopic analysis 3. Calorific analysis 4. Rock–eval pyrolysis analysis	1. Macerals with high constituents of spores and resins hence high volatile contents yield large quantities of oil and tar 2. Bright coals are best for liquefaction than splint coals 3. Fusain macerals yielded 15%–27% 0f liquefaction products 3. Banded coals had high calorific value hence high liquefaction yields due to more presence of high reactive macerals 4. Coals with high content of reactive macerals (vitrinite and liptinite) with high H/C atomic ratios in the range of 0.8–0.9 are suitable for liquefaction process 5. Hydrogen and carbon make up the complex organic compounds of coal while nitrogen occurs mainly in organic matter 6. Macerals of Type III kerogen, that is macerals from plants associated with terrestrial inputs into lacustrine or marine environments, have the capability to produce oil as well as gas but most of them are better suited for production of gaseous hydrocarbons	Sprunk (1942), Jones et al. (1985), Durie (1982), Falcon and Snyman (1986), Newman et al. (1997), Suarez-Ruiz and Crelling (2008), Singh et al. (2013), Speight (2014), Mishra et al. (2018), Singh and Mrityunjay (2018a)

Table 5 continued

Parameter	Techniques applied	Observation	Reference
Mineral matter content		1. Pyrite in coal tends to subject catalytic effect on coal hydrogenation 2. Alumino-silicate minerals behave as cracking catalysts at elevated temperatures during hydrogenation of coal 3. Calcium in coal leads to formation of deposits hence blockage on the used equipment as calcium carbonate forms	Durie (1982), Vasireddy et al. (2011), Singh et al. (2013)
Particle size		Decreasing the particle size increased the total conversion of coal into liquid products. With reduced particle size the solvent molecule had less distance to penetrate into the coal particle for extraction of products	Neavel et al. (1981), Giri and Sharma (2000), McMillen and Malhotra (2006), Li et al. (2008a, b), Heydari et al. (2016)

and hydrogen to form water and carbon dioxide during oxygen elimination processes. In addition, moisture content needs to below as it has to be removed during the hydrogenation (Durie 1982; Vasireddy et al. 2011).

(1) *Chemical structure*
The differences in the yield of syncrude from different coal ranks can be attributed to the chemical configuration of coal with the potential to influence the nature of its chemical reactivity (Zhou et al. 2016). Low ranked coals have an increased content of aliphatic carbon (C_{al}–C_{al}) bonds whereas high ranked coals have an increased concentration of aromatic-aliphatic carbon (C_{ar}–C_{al}) bonds and aromatic carbon (C_{ar}–C_{ar}) bonds. An enhancement in the carbon content results in an increased number of aromatic carbon bonds (Shi et al. 2013; Zhou et al. 2016).
The bond energies of the aliphatic carbons and the aromatic carbons are different with the aromatic carbons having high bond energy (Fig. 7) (Shi et al. 2013; Li et al. 2015; Ahamed et al. 2019). The increased bond energy reduces the reactivity of coal as more energy will be required to breakdown the aromatic structure hence the low syncrude yield by coals of high rank. Coal liquefaction is favoured by the simple skeletal structure that comprises mainly of simple aromatic rings which are less likely to be removed from the lignin basing on the building block for phenyl propane (Durie 1982).

(2) *Porosity*
Coal porosity is most relevant in relation to direct liquefaction. Determining the surface area and pore volume is essential for establishing the correlation between the surface area and chemical reactivity with regard to the access of gases and liquids to the interior of coal particles (Vasireddy et al. 2011). Coals from different ranks have a structure made up of varying pores being the macropores having diameters greater than 50 nm, mesopores with pores ranging from 2 nm to 50 nm and the micropores with pores less than 2 nm. The macropores reduce with an increase of coal rank while the mesopores and micropores increase with an increase in the rank of coal (Zhu et al. 2016).

2.4.2 *Organic composition*

The organic composition of coal is directly related to the oil chemistry and potential of coal to generate hydrocarbon compounds hence its importance in the liquefaction process. Organic constituents in coal differ greatly in ease of liquefaction and yield of the various products. The type of maceral usually with the highest degree of capacity for liquefaction is fusain. Fusain is a mixture of opaque fibers termed fusinite and translucent or semi-opaque material ranging from 10% to 30%. The yield of liquefaction is dependent primarily on the amount and nature of the translucent material. The translucent material consisting of constituents such as leaves, spores, resins, cuticles are usually considered good for hydrogenation (Sprunk 1942; Randolph 1944).

The estimated oil yields based on weight that can be obtained from clean macerals are 78%–80% for liptinite, 6% to 10% for vitrinite while the inertinite yields 0%–2% of liquefaction products. The vitrinite macerals readily dissolve and as such assist in conversion of coal into liquid products through their natural thermoplastic characteristics. The high yields by liptinites is due to the presence of aliphatic structures while the inertinites simply remain inert during a conversion process (Saxby and Shibaoka 1986; Bertrand 1989; Hunt 1991; Newman et al. 1997; Peters et al. 2000; Sykes 2001; Suarez-Ruiz and Crelling 2008; Singh and Mrityunjay 2018a, b). Generally, for coal to be considered suited for liquefaction, its organic matter should be 10% to 20% Type I kerogen or 20% to 30% with Type II kerogen (Powell and Boreham 1994). Type I kerogen is from a different precursor with high aliphatic content from various sedimentary environments whereas Type II is related to the planktonic organic matter found in open marine as well as freshwater from lacustrine environments (Suarez-Ruiz 2012).

The liquid yield and the nature of solid residue from coals of constant coal rank condition is highly dependent on the maceral composition (Kalkreuth et al. 1986). Coals rich in vitrinite and exinite have high hydrogen content and increased hydrogen has been found to increase the products of liquefaction (Singh et al. 2013; Mishra et al. 2018).

2.4.3 *Size of coal particle*

Particle size is deemed a significant parameter during coal liquefaction conversion. Small-sized particles have reduced distances of diffusion by the solvent molecule in order to access the core of the coal matrix for the extraction of liquid products. Large-sized particles result in a diffusing component being consumed quicker than its diffusion into the particle due to the long distance it has to travel to reach the core of the particle. This leads to a reaction in zones near the center of particle which in turn will lead to an increased level of mixing of free radicals. The free radicals result in products with stable high molecular weight being formed which is undesirable during liquefaction (Schlosberg 1985; McMillen and Malhotra 2006; Li et al. 2008a, b). Extraction yields have been found to be more for small sized particles as they provide increased coal surface area and enhance the ability of the solvent to easily penetrate into the coal structure hence an increment of yields (Giri and Sharma 2000; Heydari et al. 2016).

2.4.4 *Inorganic matter*

The heterogeneity of coal has resulted in mineral matter distribution of various forms, compositions and associations. These mineral matter constituents are not really inert during the conversion of coal into liquids and can affect the distribution and yields of liquid products. It has been reported to act beneficially as a catalyst for liquefaction. Pyrite and alumino-silicates exert a catalytic effect during coal hydrogenation. Furthermore, the mineral matter can be detrimental on the equipment used for liquefaction by formation of scale and deposits on the reactor walls and can lead to blockage problems due to formation of compounds like calcium carbonate adversely affecting the reactor capacity (Durie 1982; Heydari et al. 2016).

The ash content of coal should not exceed 10 wt % as constituents likely to form ash exist as cations associating with carboxylic acid compounds in coal. Their nature and quantity can have a significant effect on the behavior of coal as it undergoes thermal decomposition and aid expedite oxygen elimination. In addition, high ash content adversely affects the liquefaction process by negatively impacting the reactor capacity. Also, a ballasting impact relating to the recoverable yield of distillable oil can be experienced as the added catalyst gets deactivated by the presence of mineral matter (Durie 1982).

Coals with vitrinite reflectance ranging from 0.57% to 0.62%, hydrogen to carbon atomic ratio falling in the range of 0.73 to 0.87 with reactive maceral content of 72% to 96.8% and volatile matter content in the range of 29.4% to 57.1% are considered suitable for conversion into liquid products (Davis et al. 1976).

3 Discussion

Physical and chemical characteristics of coal are essential in determining coal usage. The level of coalification reached by coal can be deemed the primary factor as it influences all other coal parameters. Different ranks of coal vary in the composition of macerals, plasticity, porosity, reactivity, chemical structural composition, and the inorganic compounds, all these being the parameters essential in selecting the coal technology to implement for a particular coal. Also of importance is the organic/maceral composition of coal as it is responsible for most of the coal benefits such as its energy output during combustion, its role in metallurgical processing, its capacity for in situ methane absorption and its potential as an alternative hydrocarbon source (Sprunk 1942; Falcon and Snyman 1986; van Krevelen 1993; Nas 1994; Mitchell 1997; Holuszko and Mastalerz 2015). An understanding of the effect of maceral composition during coal conversion process is essential as the variation in its physical and chemical properties influences coal utilization technologies. High yields of volatile matter can be achieved from liptinite upon heating making it ideal for bitumen and tar production (Falcon and Ham 1988). Vitrinite found in bituminous coal of low rank is suitable for liquefaction process due to its high hydrogen content, more aliphatic structures and low aromaticity (Falcon and Ham 1988; Sun et al. 2003). Inertinite molecular structure characterized by low hydrogen and volatile matter content, higher thermal stability, high aromaticity and condensation in comparison to other maceral groups renders them the least reactive yielding low gas and tar during pyrolysis (Zhao et al. 2011).

The composition of macerals influences the composition of elements in coal which in turn impacts on the coal reactivity. Vitrinite contains less hydrogen than liptinite but more oxygen while inertinite has high carbon content and lower hydrogen than vitrinite but similar oxygen (Durie 1982; Suarez-Ruiz and Crelling 2008; Suarez-Ruiz 2012). It is the arrangement of these elements in a coal structure that determines the reactivity of different coals. These elements can exist either as aromatic or aliphatic structures. Coals consisting of more aromatic structures as compared to the aliphatic structures have low reactivity as more energy will be needed to breakdown the covalent bonds existing in the aromatic structures. Hence for easier coal conversion more aliphatic structures are essential.

However, because of the intimate association of macerals within coal, determining the effect of one maceral group can be a challenge. As a result, separation techniques such as chemical fractionation, density separation and centrifuging, have been used to better appreciate the structural properties and the reactivity of individual maceral groups (Maledi et al. 2017). These techniques may have an adversarial effect on the maceral composition hence misleading results.

The degree of coalification together with maceral composition impact on the mechanical behaviour of coal by altering its structure. Coal structure is made up of aromatic and aliphatic carbon structures. As coal matures the aliphatic carbon structures turn into aromatic carbon structures. The high bond energies for the aromatic structures are responsible for the low reactivity of high rank coals while low bond energies for the aliphatic carbon structures renders low rank coals high reactivity (Pan et al. 2013; Li et al. 2015). The functional groups making up the structure of coal are inclusive of methylene, methyl, phenolic hydroxyl and carboxyl groups. The methyl groups exist as substituent in the aromatic nuclei and are responsible for almost 20% of the hydrogen found in coal structure. While 60%–80% of organic oxygen is due to phenolic hydroxyl groups. An appreciable oxygen content found in low rank coals is due to the carboxyl groups (Durie 1982). These functional groups tend to impact negatively on coal strength. Although they hardly react with water, others act as hydrophilic sites attracting water molecules that fill up the capillary pores. The water adsorption leads to more pressure being applied on particles causing the relaxation of the intergranular stresses hence reduced frictional and cohesion strength (Kaji et al. 1986; Yu et al. 2013). An average molecular model have been proposed using vitrinite macerals to gain information on the structure of coal. However, this model was found to be misleading as it was suitable for the particular vitrinite used. It also included a mixture of low molecular weight entities (Durie 1982). As a result, understanding coal structure still remains a challenge because of its complex heterogeneous cross linked polymeric structural networks.

The mineral matter during a conversion process goes through thermal decomposition, disintegration, fusion and agglomeration (Chakravarty et al. 2015). These transformations are essential in providing information with regard to coal suitability for a particular technology (Matjie et al. 2011; Zhang et al. 2011). As the minerals react and undergo transformations there is formation of molten deposits (fouling and slagging) on the walls of conversion units resulting in great operational challenges during the process (Fig. 8).

The deposits decreases the absorption of heat in the unit thus decreasing the thermal performance as well as efficiency. Previous studies have been conducted where soot blowers using high pressurized air, steam or water jets were used to remove the deposits. However, the method did not prove effective enough to get rid of strongly adhered deposits since excessive blowing led to erosion damage of the equipment (Wee et al. 2005).

A model was devised to determine the usage of coal (Falcon and Snyman 1986). This model is based on the rank and maceral composition of coal. It uses the vitrinite reflectance and the percentage of the reactive macerals to determine the end-use application of coal (Fig. 9).

When applied to Indian coals from Argada and West Bokaro coals (Singh et al. 2015; Singh and Mrityunjay 2018b) the model indicated that the coals were best used as blending coking coals because of the high amount of reactive macerals found in them.

Also developed to assist in determining the use of coal was a Seyler diagram (Fig. 10) basing on the content of hydrogen and carbon (Singh et al. 2015).

The use of coal in relation to hydrogen and carbon contents can be deduced from the diagram as shown in Table 6.

Although the models have proved applicable in determining the coal utilization technology to implement for those particular coals, they were specifically developed for coals rich in minerals hence difficult to beneficiate for coals

Table 6 Coal utilization basing on hydrogen and carbon contents as per Seyler diagram

Utilization	Carbon content (%)	Hydrogen content (%)
Gasification	68–80	2.5–5.5
Metallurgy	82–93	4.9–5.5
Carbon electrodes	Greater than 93	Greater than 4.5
Liquefaction	77–87	Greater than 5.0

Table 7 Coal properties critical during carbonization, gasification, liquefaction and carbon fiber production processes

Property	Technology			
	Carbonization	Gasification	Liquefaction	Carbon fiber production
Organic matter (macerals) composition	Vitrinite, liptinite	Vitrinite, liptinite	Liptinite (78%–80% yields), vitrinite (6%–10% yields)	Vitrinite, liptinite
Structural composition (aromatic and aliphatic structures)	Minimal aliphatic structures	High aliphatic structures, increased hetero-atoms	Simple aromatic rings, increased aliphatic carbon	Low aromatic carbon, high aliphatic carbon
Elemental composition	Low O, C, S, P content	High H,O content, low S content	High C (80%–85%), H content Low O content (below 30 wt % dmf)	High O content
Calorific value	Low	High	NS	NS
Coal reactivity		High		Low
Proximate characteristics	High volatile matter content-coking gas, low volatile matter content-coke low moisture and ash content	Low moisture and ash content, high volatile matter and fixed carbon content	Low moisture and ash content, 29.4%–57.1% volatile matter content	NS
Inorganic matter composition	Al_2O_3, SiO_3 and alkalis lead to poor coke quality	MgO, Fe_2O_3, SiO_2, CaO, Al_2O_3,TiO_2 and alkali compounds may react with each other	Pyrite and amino-silicates tends to act as catalysts	NS
Mineral matter content	Low	Low	Low	Low
Ash characteristics	Low ash fusion temperatures	Ash fusion temperatures less than 1600 °C	NS	NS
Caking tendencies	Low	Low	NS	NS
Grindability	Grindability index of 80	NS	NS	NS
Coal fluidity	NS	NS	NS	High
Molecular weight distribution	NS	NS	Formation of products with stable high molecular weight is undesirable during liquefaction	Low content of low molecular weight compounds, uniform molecular weight distribution
Particle size	NS	Small sized	Small sized	NS

Note: NS means there was no available specification from the reviewed literature

varying greatly in maturity and composition of organic matter. Therefore a tool suitable for coals portraying dissimilar characteristics is needed. Furthermore, the properties important in coal utilization are inversely related (Durie 1982; Mishra et al. 2005; Yang et al. 2016) therefore a compromise is necessary for satisfactory performance of the utilization technology deemed applicable.

4 Summary and Conclusion

The degree of coal coalification is the most fundamental parameter in determining the end-use application of coal as it influences all the other coal properties and as such is applicable in all coal utilization technologies. Final influence of coal properties found to be critical on carbonization, carbon fiber production, gasification and liquefaction was summarized in Table 7.

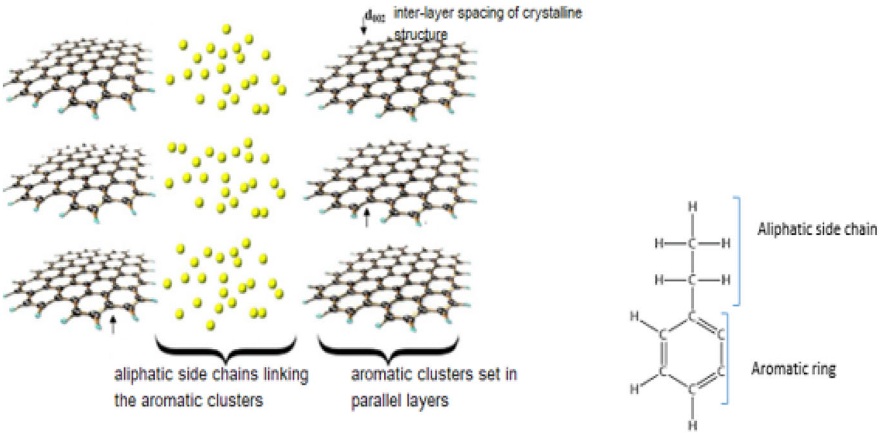

Fig. 1 Clusters of aromatic layers linked to the aliphatic structures (Ahamed et al. 2019)

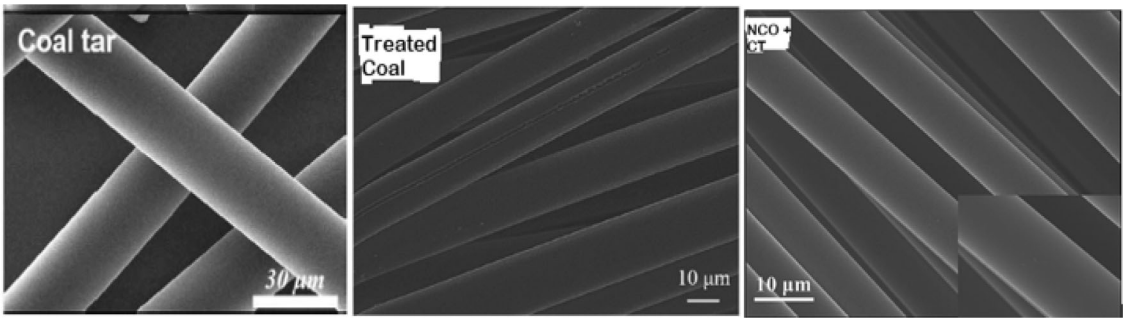

Fig. 2 Carbon fibers from different precursor materials (Kim et al. 2016b; Yang et al. 2016)

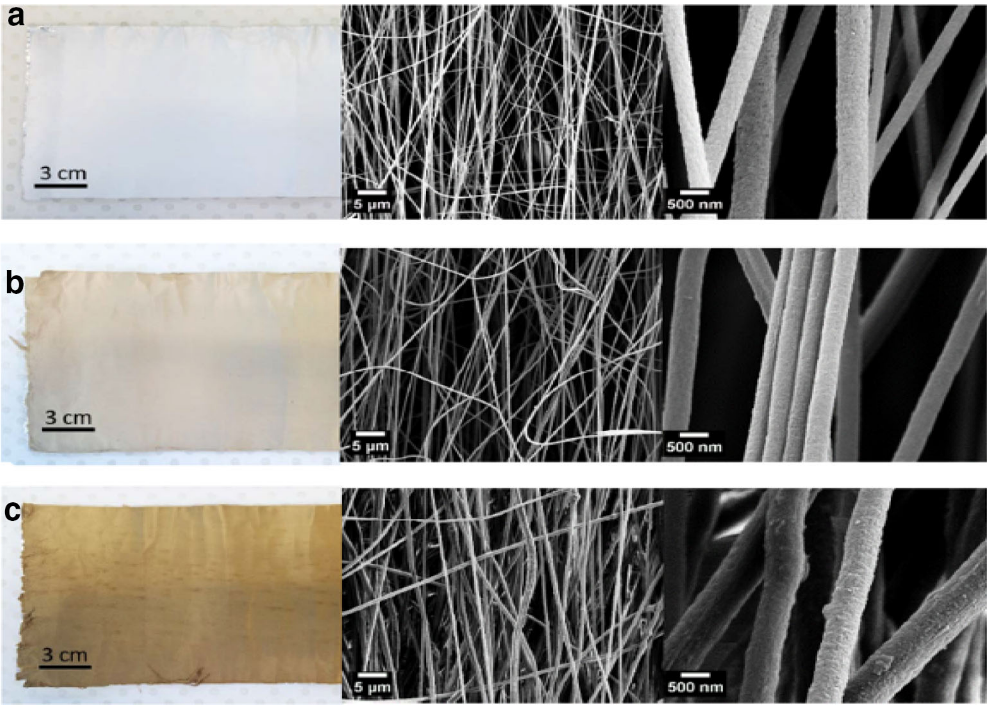

Fig. 3 Carbon fibers from Polyacrylonitrile precursor with 0% coal tar **a**, 25% coal tar **b**, 50% coal tar **c** (Modified after Zabihi et al. 2019)

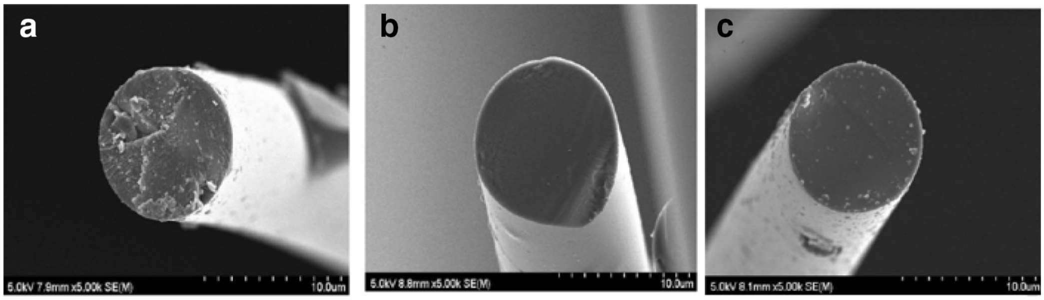

Fig. 4 Carbon fibers from pyrolysis fuel oil with 100% coal tar **a**, 200% coal tar **b**, 300% coal tar **c** (Modified after Lee et al. 2019)

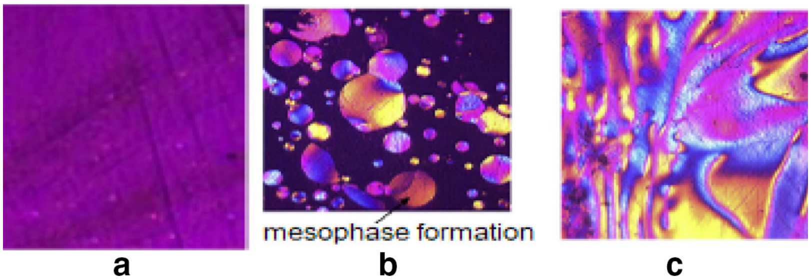

Fig. 5 Isotropic phase **a**, formation of mesophase in an isotropic phase **b**, anisotropic phase **c** (Modified after Huson 2017)

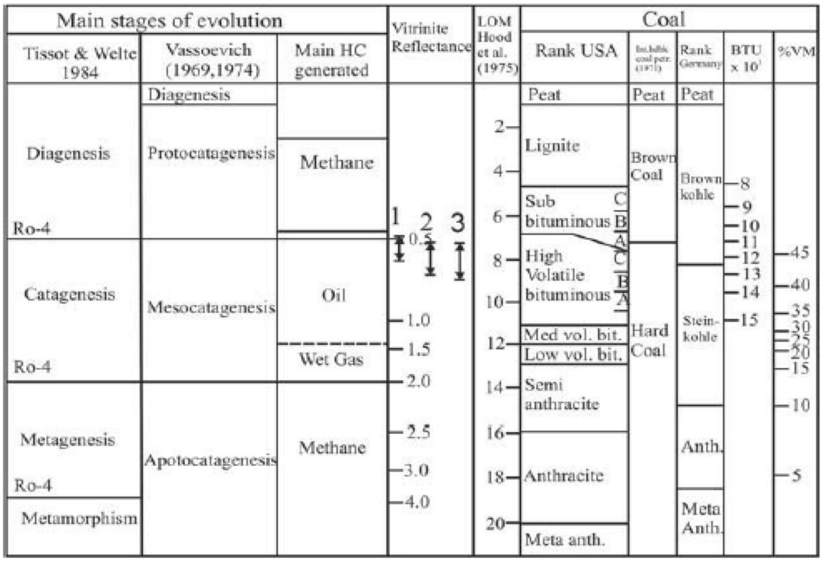

Fig. 6 Main stages of maturity relating coal organic composition and rank (Singh and Mrityunjay 2018a)

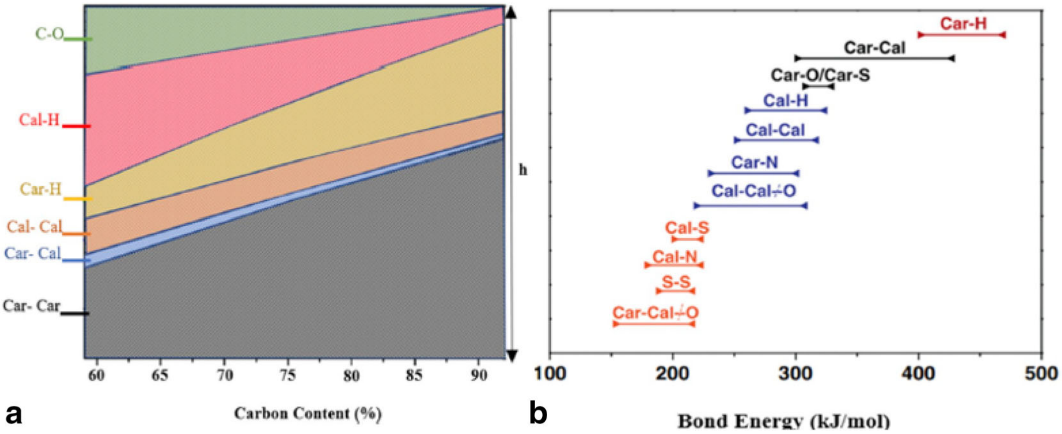

Fig. 7 Aliphatic and aromatic carbon bonds in relation to carbon content **a**, bonding energies for major covalent bonds in coal **b** (Ahamed et al. 2019)

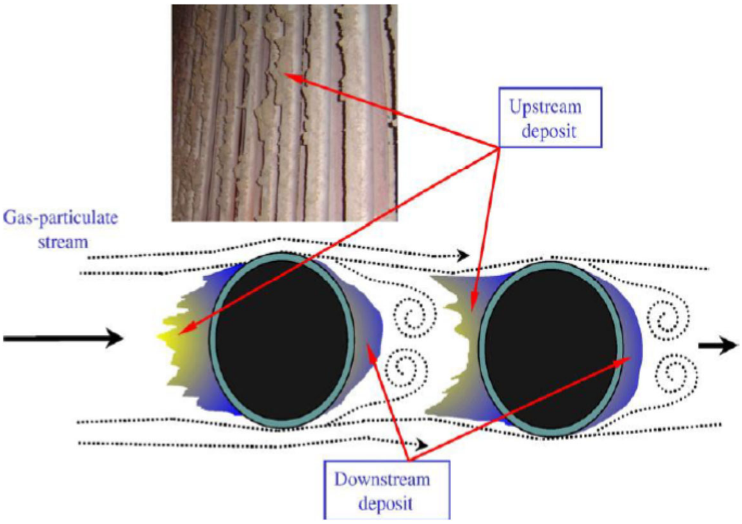

Fig. 8 Fouling deposits on tubes in convective pass (Ma et al. 2007)

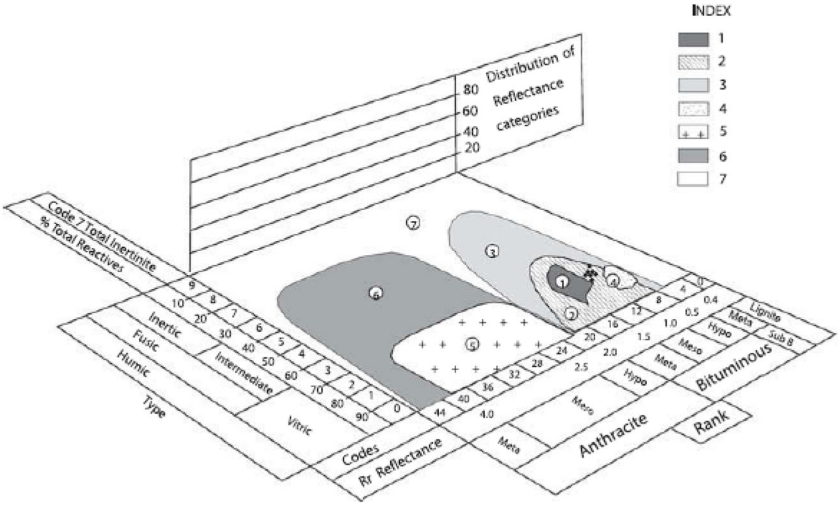

Fig. 9 Model used to determine coal utilization basing on vitrinite reflectance and percentage of reactive macerals (Singh et al. 2015; Singh and Mrityunjay 2018b)

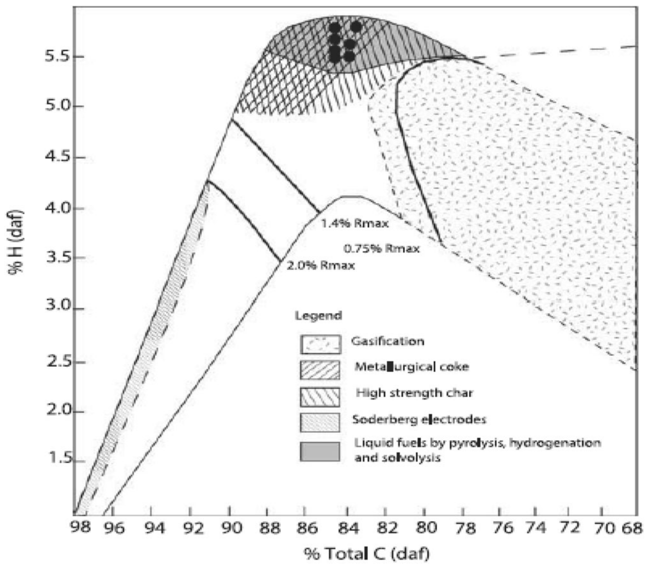

Fig. 10 Seyler diagram used for determining coal utilization basing on hydrogen and total carbon contents (Singh et al. 2015)

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