



# **Gasification of Solid Fuels (Coal, Biomass and MSW): Overview, Challenges and Mitigation Strategies**

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**Abstract:** Currently, hydrogen energy is the most promising energy vector, while gasification is one of the major routes for its production. However, gasification suffers from various issues, including slower carbon conversion, poor syngas quality, lower heating value and higher emissions. Multiple factors affect gasification performance, such as the selection of gasifiers, feedstock's physicochemical properties and operating conditions. In this review, the status of gasification, key gasifier technologies and the effect of solid-fuel (i.e., coal, biomass and MSW) properties on gasification performance are reviewed critically. Based on the current review, the co-gasification of coal, biomass and solid waste, along with a partial utilisation of  $CO_2$  as a reactant, are suggested. Furthermore, a technological breakthrough in carbon capture and sequestration is needed to make it industrially viable.

Keywords: gasification; solid fuels; coal; biomass; MSW



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# 1. Introduction

In 2020, the total energy consumption in the world was 560 exajoules (EJ) [1], which is forecasted to be 712 EJ by 2040, according to the International Energy Agency's stated policies scenario [2]. Coal currently meets around 27% of the world energy demand [1], which is projected to be about 20% in 2040 [2]. Biomass and municipal solid waste (MSW) are gaining importance rapidly due to their role in global decarbonisation [3,4]. In 2018, global energy consumption from biomass was 55.6 EJ, out of which 85% was supplied by solid biomass, 7% from liquid biofuels, 5% from municipal and industrial solid waste and 3% from biogases [5]. The world MSW generation is about 2.0 billion tonnes (BT) [6], which is forecasted to be 3.4 BT in 2050 [7].

Global fossil-fuel-related CO<sub>2</sub> emissions increased from 9.38 BT in 1960 to 34.8 BT in 2020 [8]. The CO<sub>2</sub> gas emission by sectors includes electricity and heat generation: 37%, transportation: 25%, industry: 23%, building: 9%, agriculture, forestry, and other land use: 6% [2]. In 2020, 37% of the CO<sub>2</sub> emission was caused by coal consumption [8].

Along with other greenhouse gasses, coal-fired power plants typically emit 1.0 kg of CO<sub>2</sub> per kW of electricity production [9]. Hence, advanced technologies are required to reduce emissions and increase thermal efficiency simultaneously. One such technology is gasification, which can efficiently convert solid carbonaceous fuels such as coal, biomass and MSW into syngas [10]. Syngas generated from gasification is subsequently utilised for power production or valuable chemical synthesis.

A detailed review considering the fundamental challenges, including mitigating strategies, for coal, biomass and MSW gasification is limited in the literature. Hence, this article has reviewed various aspects of those issues and proposes possible solutions to overcome those challenges.

## 2. Gasification Technologies

Gasification is the key technology for the cleaner conversion of solid fuels such as coal, biomass and MSW, which have long been used under different names around the globe [11–17]. Gasification technology is primarily divided into three major groups: fixed bed, fluidised bed and entrained-flow gasifier [18]. The fundamental aspect of each category is readily available in a large amount of literature. Therefore, this review summarises a key comparison of those technologies in Table 1. However, interested readers are referred to the following articles for details on those theories and fundamental physics: fixed bed gasifier [19,20], fluidised bed gasifier [20–26] and entrained flow gasifier [18–20,27]. Moreover, due to their importance and widespread industrial application, a comparison of commercial entrained flow gasifiers is reported in Table 2, while schematic views of those gasifiers are in Figure 1.

<b>Operating Conditions</b>	Fixed	l Bed	Fluidis	ed Bed	Entrain	ed Flow
Ash conditions	Dry ash	Slagging	Dry ash	Agglomerating	Slagging	Slagging
Fuel-feeding conditions	Dry feeding	dry feeding	dry feeding	dry feeding	dry feeding	slurry feeding
Reactant type	Air/O <sub>2</sub>	Air/O <sub>2</sub>	Air/O <sub>2</sub>	Air/O <sub>2</sub>	O <sub>2</sub>	O <sub>2</sub>
Reactant requirement	low	low	medium	medium	high	high
Syngas flow direction	up	up	up	up	up or down	up or down
Typical reactor temp (°C)	1000	1500–1800	900–1050	900–1050	1200-1600	1200-1600
Syngas temperature (°C)	425-650	425-650	925–1040	925–1040	1400–1600	1200-1400
Syngas cooling	Water	Water	Coolant	Coolant	Coolant	Water/syngas coolant
Pressure (Mpa)	3.0	2.5	Up to 3.0	1.0-3.0	2.5-3.0	2.5–3.0
Feedstock preference	Low- to high-rank coals and waste	Medium- to high-rank coals, petcoke and waste	Low- to medium rank coals and waste	Low- to medium rank coals, biomass and waste	Low- to high-rank coals, biomass, petcoke and waste	Low- to high-rank coals, biomass, petcoke and waste
Typical particle size (mm)	5-80	5-80	<6	<6	<0.1	<0.1
Residence time (s)	900–3600	900–3600	10–100	10-100	1.5–4	1.5
Moisture (%)	No limit	<28	No limit	No limit	Possible to use coal with high moisture	limited
Ash content limit (%)	<15	<25	<40	<40	2–25	<25
Ash fusion temp limit (°C)	Any	Any	>1100	>1100	Generally <1300	Generally <1300
Commercial gasifier	Lurgi	BGL	IDGCC, HTW and KBR	KRW and U-Gas	Shell, PRENFLO, EAGLE, Siemens, MHI	GE, E-Gas
Conversion	>99	>99	96	95	98–99	100
Typical cold gas efficiency (%)	~88	~88	~85	70–80	~80	74–77
Unit capacity (MW <sub>th</sub> )	10–350	10-350	100–700	20–50	Up to 700	Up to 700
Key technical issues	Agglomeration and use of hydrocarbon liquid	Agglomeration and use of hydrocarbon liquid	Lower carbon conversion and agglomeration	Lower carbon conversion and agglomeration	Syngas cooling and slagging	Syngas cooling and slagging

Table 1. Key characteristics of generic gasifiers [28–32].

Parameter	ChevronTexaco	E-Gas	Shell	PRENFLO
Fuel type	bituminous coal	bituminous coal	bituminous coal	petroleum coke and bituminous coal
Gasification process	single-stage entrained flow	two-stage entrained-flow	single-stage updraft entrained flow	single-stage updraft entrained flow
Fuel feeding	slurry feeding	slurry feeding	dry feeding	dry feeding
Reactant	95% pure oxygen	95% pure oxygen	95% pure oxygen	95% pure oxygen
Syngas cooler type	downflow radiant, water tube and fire tube	downflow fire tube	downflow water tube	Downflow or upflow radiant water tube and convective water tube
Controlling particles	water scrubber	metallic candle filter and water scrubber	candle filter	candle filter
Chloride, fluoride and ammonia control	water scrubber	water scrubber	water scrubber	water scrubber
Sulphur recovery (%)	98%	99%	99%	99%
Air separation	cryogenic distillation	cryogenic distillation	cryogenic distillation	Cryogenic distillation
Combustors	multiple cans	multiple cans	twin vertical silos	twin horizontal silos
Firing Temperature, °C	1287	1287	1100	1260
Heat-recovery steam generator	triple-pressure reheat and natural circulation	triple-pressure reheat and natural circulation	triple-pressure reheat and natural circulation	triple-pressure reheat and natural circulation
Slag removal	lock hopper	continuous	lock hoppers	lock hoppers



Figure 1. Major configuration of the commercial entrained-flow gasifiers [36,37].

#### Ash and Slag Characteristics for Entrained-Flow Gasifier

During gasification, inorganic minerals of coal, biomass and MSW generate ash [38]. One of the key features of entrained-flow gasifier is the transformation of ash into molten slag under high-temperature operation. Typically, entrained-flow gasifiers are slagging types. Slagging is one of the greatest challenges for gasification performance as slagging increases the system cost considerably. Furthermore, it is also a barrier to the continuous operation of the gasifier due to the blockage of slag-removal devices, pressure drop and gasifier load reduction [19,39]. Moreover, high-temperature ash slag is accountable for damaging the expensive refractory lining of entrained-flow gasifiers. The steps for the deposition/penetration of slag in the gasifier tube are as follows: coal/biomass > char > ash > molten ash > liquid slag  $\cap$  solid slag [19].

Therefore, it is crucial to understand the characteristics of ash and slag for the fuels to be used in an entrained-flow gasifier. One of the important properties of understanding the characteristics of ash is ash fusion temperature (AFT) [40]. Several studies [39–41] present correlations built between the AFT and different properties such as mineral compositions, thermodynamics and ionic potential to predict the AFT. Recently, Li et al. [40] investigated AFT based on the average ionic potential for ten different types of coal collected from different deposits in China. The predicted correlation was AFT (°C) = 509.12 + 17.98 I<sub>average</sub>, where  $I_{average} = \frac{\sum M_i I_i}{\sum M_i}$ . The term  $M_i$  represents mole fraction of Al<sub>2</sub>O<sub>3</sub>, CaO, FeO and MgO, which sum up to be 100%, whereas  $I_i$  is the ionic potential of Fe<sup>2+</sup> (26.3 nm<sup>-1</sup>), Mg<sup>2+</sup> (30.8 nm<sup>-1</sup>), Ca<sup>2+</sup> (20.2 nm<sup>-1</sup>) and Al<sup>3+</sup> (60.0 nm<sup>-1</sup>) [41].

Song et al. [38] reported that the AFT decreases with increasing CaO up to the concentration of 30%, above which increasing the CaO leads to an increase in the AFT. The result was attributed to the subliquidus phase transformation from anorthite to the gehlenite phase, increasing CaO by over 30%. The flow behaviour of ash and slag primarily depends on the viscosity, which is affected by the thermochemical properties and crystalline structure of ash/slag. Oh et al. [42] demonstrated the effect of crystalline structure on the ash viscosity. Numerous studies investigated the impact of chemical composition on ash viscosity under various operating conditions [43–45]. Table 3 summarises the studies carried out for ash and slag behaviour analysis.

Table 3. Summar	y of the investigations	s regarding ash and	l slag characterisations.
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Ref.	Methods and Materials	<b>Study Parameters</b>	Key Findings
- - [46] -	Experimental viscosity measurement Mineralogical analysis: X-ray diffraction (XRD) Chemical analysis: X-ray fluorescence (XRF) and infrared spectroscopy (IR) Thermal properties: differential thermal analysis (DTA) Expansion process: hot-stage microscopy (HSM)	Thermal-expansion behaviour of fly ash of IGCC power plant	<ul> <li>SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, with a ratio of 1.95:1, were the main components both in ash and slag, accounting for more than 80%, which caused to increase in the viscosity</li> <li>The expansion behaviour of ash is related to the release of gases during gasification</li> <li>A slag with a higher temperature was found to be more porous than a lower-temperature slag</li> </ul>
[38]	Experimental measurement of AFT and temperature of critical viscosity (T <sub>cv</sub> ) Ash and slag characterisation with XRD, XRF and scanning electron microscopy (SEM)	Fusibility and flow properties of laboratory ash	- AFT and $T_{cv}$ of ash are higher than slagging temperature, decreasing with increasing CaO up to 30%. However, the AFT and $T_{cv}$ increase dramatically by increasing CaO concentration by over 35%.

Ref.	Methods and Materials	Study Parameters	Key Findings
[47]	AFT analysis Size, composition and abundance of mineral grains of ash and slag study by SEM	To characterise the slag and mineral transformation	<ul> <li>The main minerals contained in the coal ash are kaolinite, quartz, dolomite, calcite, muscovite, pyrite and Microline</li> <li>The key element that causes the bonding of the slag is Ca</li> <li>The modification of mineral phases to decrease viscosity and AFT is crucial</li> <li>Addition of additives helps to modify the ash and slag behaviour by interrupting the formation of liquid-phase bonding</li> </ul>
- [39] <sup>-</sup>	Experimental study Thermodynamic equilibrium modelling using FactSage software	Slag viscosity prediction of the ash	<ul> <li>It is possible to predict actual viscosity at different portions of the slag rather than just prediction of average viscosity using the slag-viscosity model (i.e., urban model)</li> <li>The concentration of CaO and Na<sub>2</sub>O change with increasing temperature</li> <li>Na<sub>2</sub>O influences the formation of slag at a lower temperature</li> <li>Increasing CaO decreases the viscosity</li> </ul>
[48]	An experimental study was carried out using image profile of ash pellets	Ash fusion characterisation along with dilatometry and sintering strength tests using the image-based technique	- An image profile can predict the slag and fouling behaviour of the boiler and can provide more information in comparison to a conventional ash fusion test
[49] _	Experimental measurement of AFT while doping the ash with several materials TGA, SEM, XRD and energy-dispersive spectrometry (EDS) were conducted	The effect of the addition of inorganic materials in coal or ash	<ul> <li>The addition of GeS, PbCO<sub>3</sub>, NaCl and SrCO<sub>3</sub> reduces the ash fusion temperature, whereas the addition of CrO<sub>3</sub> and GeO<sub>2</sub> increases the AFT</li> <li>Cr<sub>2</sub>(SO4)<sub>3</sub> and nickel compounds did not have a significant effect on AFT</li> <li>The diverse characteristics of different additives are due to the difference in ionic potential among the additives</li> </ul>

## Table 3. Cont.

An experimental study using ten

different types of coal ash

[40]

# 3. Current Status of Syngas Production from Gasification

Development of a correlation

to predict AFT based on the

ionic potential of major

minerals in the ash

The primary gasification product is syngas, used for various applications directly or after downstream treatment. Syngas obtained from gasification is cleaned before use in subsequent applications, such as hydrogen production, methanol synthesis, liquid-fuel synthesis, power generation, etc. [33,34]. Some available syngas treatment and purification techniques include water–gas shift reaction, methanation, membrane adjustment, acid gas removal, pressure swing adjustment and cryogenic separation [50]. On the other hand, various types of feedstock are used for gasification, such as coal, biomass, waste, petcoke and natural gas.

A correlation to predict AFT was

 $\times$  *I*<sub>average</sub> with an error ±20 °C.

developed as AFT = 509.12 + 17.98

This section outlines the current status of gasification technologies based on the application, gasifier type, feedstock and status of plants worldwide. Table 4 summarises the world gasification plants and their production capacity. Globally, 44% of the syngas plants are under operation; 27% are under development; and the rest, 29%, are planned [51].

Status	Projects	Gasifiers	Syngas Capacity (GW <sub>th</sub> )
Operating	379	938	173
Development	131	348	108
Planned	146	734	116
Total	656	2020	397

Table 4. The status of gasification plants and syngas capacity [51].

According to Table 5, syngas is predominantly used to produce chemicals with 45%; followed by 30% for gaseous fuels; 4% for industrial gases; 15% for liquid fuels; and the rest 6% for power generation [51].

Table 5. Worldwide applications of syngas [51].

Scheme 2017	Chemicals (MW <sub>th</sub> )	Gaseous Fuels (MW <sub>th</sub> )	Industrial Gases (MW <sub>th</sub> )	Liquid Fuels (MW <sub>th</sub> )	Power (MW <sub>th</sub> )	Total (MW <sub>th</sub> )
Operating (2017)	95,000	18,000	9000	42,000	11,000	175,000
Development (2020)	50,000	27,000	6000	12,000	9000	104,000
Planned	33,000	74,000	2000	6000	3000	118,000
Total	178,000	119,000	17,000	60,000	23,000	397,000

Various fuels are used to produce syngas, of which coal plays the dominant role, accounting for 84%, while the shares of natural gas, petcoke and petroleum are about 5% each [51]. The production of syngas from renewable biomass is negligible, with a share of less than 1.0%. However, biomass is a promising feedstock that may play a significant role in the foreseeable future due to the restriction on the use of coal in some countries. Table 6 illustrates the worldwide application of syngas.

Table 6. Worldwide production of syngas from different feedstocks [50,51].

Status	Coal (MW <sub>th</sub> )	Natural Gas (MW <sub>th</sub> )	Petcoke (MW <sub>th</sub> )	Petroleum (MW <sub>th</sub> )	Biomass (MW <sub>th</sub> )	Waste (MW <sub>th</sub> )	Total (MW <sub>th</sub> )
Operating (2017)	140,000	17,000	4000	13,000	1000.0	1000	175,000
Development (2020)	84,000	0	11,000	2000	0	0	97,000
Planned (2021)	110,000	2000	5000	6000	2000	2000	125,000
Total	334,000	19,000	20,000	21,000	3000	3000	397,000

# 4. Properties of Coal, Biomass and MSW

The following sections outline different properties of coal and biomass affecting gasification performance.

#### 4.1. Chemical Properties of Coal, Biomass and MSW

# 4.1.1. Chemical Properties of Coal

The chemical composition of coal is complex, consisting of organic and inorganic compounds. Up to 76 of the 90 naturally found elements listed in the periodic table can be found in coal, although most are in trace amounts [52]. The main elements present in the complex organic compounds of coal are C, H, N, S and O. On the other hand, more than 120 inorganic minerals can be found in coal, out of which 33 are common in most coal samples, and only eight are high enough to be considered major [52,53]. Table 7 shows the proximate and ultimate data for coal from different origins, whereas Table 8 shows the mineral content of some bituminous coal, which is studied for pyrolysis and gasification studies.

Origin/Reference	М	VM	FC	Ash	С	Н	Ν	S	0	LHV (MJ/kg)
Chilean Sub-bituminous [54]	14.3	34.6	35.8	15.3	52.4	3.6	0.8	0.2	13.4	18.9
Spanish Alcorisa Lignite [54]	11.1	38.6	39.0	11.3	54.7	4.1	0.4	8.1	10.3	20.8
Kentucky, USA [13]	-	39.1	50.7	8.3	81.4	5.6	1.7	3.3	7.94	-
Illinois No. 6 bituminons, [55]	8.5	36.0	44.8	10.7	69.3	5.0	1.1	3.5	9.3	26.3
India (Tirap) [56]	4.30	32.2	55.7	7.7	-	-	-	1.3	-	27.6
Chines bituminons coal [57]	1.66	34.3	48.4	15.5	55.3	2.1	0.8	0.4	5.3	-
Chines Datong coal [58]	3.2	25.7	57.3	13.7	82.7	5.0	0.8	2.4	8.9	26.5
South African bituminous coal [59]	3.5	25.5	55.3	15.7	66.3	3.6	1.8	0.5	8.6	24.9
Taiheiyo bituminons coal, Japan [60]	5.3	46.7	35.8	12.1	77.6	6.5	1.1	0.2	13.9	27.4
Shenhua bituminous coal, China [61]	5.51	32.2	54.48	7.81	70.5	4.8	1.0	0.7	9.58	26.3
Victorian brown coal (Loy Yang) [62]	11.1	48.2	-	8.0	50.1	4.3	0.4	0.2	-	-
Victorian brown coal (Morwell) [62]	14.9	49.3	-	3.6	60.7	5.3	0.5	0.04	24.0	-
Datong coal, Korea [63]	10.5	29.2	51.6	8.6	80.3	6.4	11.4	0.9	1.0	27.5
Shenmu bituminous coal [64]	5.2	31.9	58.2	4.8	75.4	4.6	12.1	1.1	0.5	25.9
Kentucky, USA [65]	4.2	36.3	51.6	7.9	74.8	5.1	7.2	1.6	3.0	-
	5.2	27.1	45.3	22.4	57.8	4.0	9.3	1.0	0.3	-
Shenhua, China [66]	1.9	20.7	70.6	8.7	80.8	3.9	4.9	1.2	0.5	-
Australian bituminous coal, Brisbane [67]	14.1	42.1	34.4	9.4	79.6	5.5	-	-	4.0	-
Polish bituminous coal [68]	5.2	31.9	58.2	4.8	80.3	6.4	11.4	0.9	1.0	27.5

Table 7. The proximate (wt.%) and ultimate (wt.%) analyses of coals from different origins.

Table 8. Major mineral contents (wt.%) of bituminous coals.

Coal	SiO <sub>2</sub>	$Al_2O_3$	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	SO <sub>3</sub>	$N_2O$	TiO <sub>2</sub>
Chinese sub-bituminous Datong coal [69]	41.4	17.8	28.2	6.4	2.9	1.6	0.2	-
Chinese shenmu bituminous coal [64]	27.7	12.2	12.2	33.9	1.9	7.4	0.8	0.5
Australian bituminous coal, Brisbane [67]	41.3	28.2	6.1	13.3	0.0	6.7	3.8	0.1
Polish bituminous coal [68]	48.5	26.1	10.1	2.9	1.6	-	2.5	-

4.1.2. Chemical Properties of Biomass

Table 9 includes the typical elemental and proximate analysis values for certain regularly used biomasses. Carbon (C), hydrogen (H), oxygen (O), nitrogen (N) and sulphur (S) values are presented on a percentage of weight basis in elemental analysis, and moisture (M), volatiles (VM), fixed carbon (FC) and ash values are shown on a percentage-of-weight basis in proximate analysis. Table 9 also includes lower heating values (LHV) for several common biomasses.

Origin/Reference	Μ	VM	FC	Ash	С	н	Ν	S	0	LHV (MJ/kg)
Wood [70]	20	82	17	1	51.6	6.3	-	0.1	41.5	18.6
Wheat straw [70]	16	59	21	4	48.5	5.5	0.3	0.1	38.9	17.3
Barley straw [70]	30	46	18	6	45.7	6.1	0.4	0.1	38.3	16.1
Sawdust [71]	3.19	78.57	17.09	1.15	45.66	5.81	0.11	-	45.32	16.08
Waste wood [71]	6.27	78.11	15.04	0.58	43.46	6.20	0.64	0.15	43.49	17.48
Palm kernel shell [71]	5.92	71.31	17.81	4.99	44.60	6.50	2.92	0.1	40.20	18.74
Empty fruit bunch [71]	9.63	64.95	19.48	5.94	43.84	6.01	0.88	-	39.17	16.38
Pinewood [72]	6.8	71.7	19.2	2.3	48.9	6.2	0.1	0.1	42.5	18.1
Timothy grass [72]	5.6	78.2	12.6	3.6	43.4	6.1	0.4	0.1	45.4	15.9
Wheat straw [72]	5.2	70.1	20.3	4.4	44.1	6.0	1.3	0.1	45.0	15.6

Table 9. The elemental (wt.%) and proximate (wt.%) analyses for some typical biomass.

4.1.3. Chemical Properties of MSW

The typical values of elemental analysis and proximate analysis for some material of interest in MSW are also shown in Table 10. In the case of elemental analysis, values for C, H, O, N and S; and in the case of proximate analysis, values for M, VM, FC and ash, are shown on a percentage-of-weight basis. Moreover, LHV for some materials of interest in MSW is also shown in Table 10.

Table 10. The elemental (wt.%) and proximate (wt.%) analyses for some materials of interest in MSW.

Origin/Reference	Μ	VM	FC	Ash	С	н	Ν	S	0	LHV (MJ/kg)
Mixed food waste [73]	3.02	69.35	19.31	8.32	42.25	6.47	5.25	0.45	34.24	21.38
Mixed paper waste [4,74]	10.2	75.9	8.4	5.4	43.3	5.8	0.3	0.2	44.3	14.1
Mixed plastics waste [75]	0.38	94.71	4.37	0.54	82.41	13.42	0.18	-	2.8	43.7
Yard wastes [4,76]	60.0	30.0	9.5	0.5	46.0	6.0	3.4	0.3	38.0	15.60
Solid recovered fuel (fluff) [77]	18.67	70.88	2.94	7.51	51.81	7.68	0.07	0.07	30.83	16.02
Solid recovered fuel (treated) [78]	5.04	78.09	7.52	0.43	43.24	6.03	0.44	-	41.89	21.54

#### 4.2. Physical Properties of Coal, Biomass and MSW

# 4.2.1. Particle Size

Particle size significantly impacts the reactivity of gasification, and thus, carbon conversion. Generally, decreasing particle size increases carbon conversion [79]. A higher carbon conversion from particles with smaller sizes results from a higher specific surface area. Moreover, smaller particle size results in higher residence time in the gasifier, which again helps to accelerate the carbon conversion [80].

Kirtania and Bhattacharya (2016) [81] studied the effect of particle size of spruce and coconut-shell biomass char in an entrained flow gasifier. The study chose three gasification temperatures of 800, 900 and 1000 °C, and two particle sizes of 150–250 and 500–600  $\mu$ m were selected for the study. Based on the particle size, the residence time for spruce chars was 9.0 and 4.5 s, while the residence time for coconut-shell char was 7.0 and 2.5 s. The results

showed that smaller particle sizes showed 47 and 58% higher carbon conversion under similar operating conditions than larger particle sizes using spruce and coconut-shell chars.

Apart from residence time, particle size determines the diffusion behaviour of the char particle. A larger particle size causes diffusion resistance to be higher and thus inhibits reactant flow into the micropores, especially at higher temperatures [67]. Hence, bulk diffusion becomes a rate-limiting step due to the limited active surface area for the gasification reactions. Therefore, it is essential to maintain the particle size to achieve higher carbon conversion while maintaining entrainment conditions and residence time [79].

## 4.2.2. Porosity

Porosity is an important physical property of coal/biomass, which is the ratio of void space over the bulk volume of the particle. Particles with higher porosity lead to an increase in the devolatilisation rate and subsequent gasification reactivity of char. Particles can be classified into three types based on the porosity: Group I is highly porous particles having porosity >70% and a very low wall thickness of  $<5 \mu$ m; Group II is the particles with medium porosity between 40–70% and a wall thickness of  $>5 \mu m$ ; and Group III is the particles with porosity as low as <40% and a wall thickness of >5  $\mu$ m [82]. Typically, particles with higher porosity show higher reactivity. However, this might not always be the case since other interdependent factors affect reactivity [28]. The operating conditions of gasification significantly affect the porous structure of the char. However, there is no general trend for porosity concerning pressure and temperature. Terry Wall et al. [82] studied four different types of bituminous coal char under different pressures of 5.0, 10.0 and 15.0 atm using different particle groups. The result shows that increasing operating pressure increases the porosity of Group I particles while decreasing Group II and Group III char particles. Regarding the effect of temperature, the porous structure might be altered with extremely high temperature if the mineral matter is melted and sintered into the pore of the char particles [83,84].

#### 4.2.3. Specific Surface Area

The internal surface area of coal/biomass/MSW is another crucial parameter determining gasification reactivity. Higher internal surface area results in more active sites for the gasification reactions to take place. This physical property depends on the coal, biomass and MSW type and process condition. Generally, the surface area is inversely proportional to the operating pressure and is considered the function of fluid behaviour during pyrolysis [82,85]. Vyas et al. [86] studied the effect of temperature on the surface area of different coal and biomass blend using the pyrolysis temperature between 300 and 900 °C. The BET surface area increases with increasing temperature of up to 600 °C for pure coal before declining with processing temperature. The effect of the biomass blend with coal shows that the peak surface area of the blend differs concerning temperature. Higher initial surface area leads to an increase in the temperature to reach the peak surface area. In addition, similar results have been reported under CO<sub>2</sub> [87,88] and steam [89] gasification conditions.

#### 5. Pyrolysis and Gasification Mechanisms

#### 5.1. Pyrolysis Mechanism of Solid Fuels

Pyrolysis is the primary step in the solid-fuel-conversion process, which plays a significant role during gasification. Depending on the organic properties and operating conditions, coal loses up to 70% of its mass under pyrolysis [90]. During pyrolysis, coal/biomass/MSW particles are fragmented due to breaking the weak bond between aromatic clusters. Under elevated temperature, the molecular mass of the fragments becomes considerably low, which then vapourises and escapes from the coal/biomass/MSW. The devolatilised part is known as light gas and tar [91]. The product of the devolatilisation of coal and biomass is shown below. where *x* represents the mole fraction of the yield gases.

The pyrolysis process of coal/biomass generally starts at a temperature around 350 °C, releasing CO, H<sub>2</sub> CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub> and other trace elements. The syngas yield during pyrolysis is due to the increased decomposition of the functional groups, structural components of the char and partial gasification. In addition to syngas, under low temperatures (<1100 K), pyrolysis yields tar [92]. Generally, increasing pyrolysis temperature increases the yield of CO and H<sub>2</sub> while decreasing CO<sub>2</sub>, CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub>. The cracking of heterocyclic oxygen groups produces CO. At elevated temperatures, the pyrolysis products CO<sub>2</sub> and steam react with carbon present in coal/biomass char and alter the yield of CO and H<sub>2</sub>. The formation of CH<sub>4</sub> is due to the decomposition of methyl and methylene bridge groups at lower and higher temperatures, respectively [83]. The drop of CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> with increasing temperature is potentially due to the gas-phase reaction and condensation or crosslinking within the char structure [83,93]. At a temperature around 1200 °C, available CO<sub>2</sub> is consumed, and the yield of CO becomes stable [83].

## 5.2. Gasification Mechanism of Solid Fuels

The gasification of coal/biomass/MSW involves both homogeneous and heterogeneous reactions. Key steps for the gasification of coal, biomass and MSW are illustrated in Figure 2. As mentioned earlier, pyrolysis is the first step in the gasification process, where volatile matter, tar, oil, naphtha and some oxygenated compounds are released, leaving char behind [91].



Figure 2. Key gasification steps for the solid carbonaceous solid fuels (redrawn based on [94]).

During gasification, multiple reactions occur within the volatile compounds through cracking, reforming and CO-shift conversion, whereas some parts of the volatile species undergo reaction with char and yield syngas. The key reactions during gasification are the Boudouard reaction, reverse water–gas shift reaction, partial oxidation, CO oxidation, CO shift reaction and steam–methane reforming, as illustrated in Table 11.

Immediately after devolatilisation, the released combustible gases and char are partially oxidised with inherent oxygen in the char matrix and the feed oxygen (if any). These partial oxidations are exothermic, and supplying heat energy requires endothermic char gasification reactions. The exothermic reactions are very first and thereby reach equilibrium quickly. Due to insufficient oxygen, char particles remain unburnt and participate in gasification reactions [95].

In the second stage of gasification, pyrolysed gases, reactants and char particles are involved in the heterogeneous gas–solid reaction according to Boudouard, steam gasification and hydrogasification reactions. It is to be noted that the first two reactions are highly endothermic, and thus the reaction rate is not fast enough to reach equilibrium. Furthermore, gas-phase homogeneous reactions occur according to the water–gas shift reaction, steam-methane-reforming and dry methane-reforming reactions and provide final syngas compositions.

Reaction Type and Name	Reaction	Enthalpy, $\Delta H^{\circ}$ ( $\frac{kJ}{mol}$ )		
Partial oxidation				
CO oxidation	$CO + {}^{1\!/_2}O_2 \to CO_2$	-283.0		
Hydrogen combustion	$\mathrm{H}_2 + ^{1\!\!/_2}\mathrm{O}_2 \to \mathrm{H}_2\mathrm{O}$	-242.0		
Partial oxidation of carbon	$C + \frac{1}{2} O_2 \rightarrow CO$	-111.0		
Heterogeneous gasification reactions				
Boudouard reaction	$C + CO_2 \rightleftharpoons 2CO$	+172.0		
Steam gasification	$C + H_2 O \rightleftharpoons CO + H_2$	+131.0		
Hydrogasification	$C+2H_2\rightleftharpoons CH_4$	-75.0		
Homogeneous gasification reactions				
Reverse water-gas shift reaction	$CO + H_2O \rightleftharpoons CO_2 + H_2$	-41.1		
Steam–methane reforming	$CH_4 + H_2O \rightleftharpoons CO + 3H_2$	+206.3		
Dry methane reforming	$CH_4 + CO_2 \rightleftharpoons 2CO + 2H_2$	-247.0		

Table 11. Key gasification reactions and corresponding enthalpy [96,97].

It is worth noting that these reverse reactions play a pivotal role in determining the final syngas yield, depending on the temperature and pressure of the gasifier. Some factors governing all reactions involved in gasification are temperature, pressure, gasification reagent concentration and gasifier type. For example, increasing the operating pressure leads to an increase in the CH<sub>4</sub> and CO<sub>2</sub> concentrations in the product gas. On the other hand, increasing the gasification temperature decreases H<sub>2</sub> and CH<sub>4</sub> but increases CO [96].

#### 5.2.1. Reactivity of Coal, Biomass and MSW

The gasification performance vastly depends on the reactivity of coal/biomass/MSW char. For example, the reactivity of coal/biomass/MSW determines the degree of recycling and size and configuration of the gasifier, which subsequently dominates the oxidant requirement and overall system performance [98,99]. However, the reactivity of char is not constant for coal with a specific rank or biomass but varies with their physical and chemical properties [100]. Furthermore, operating conditions such as temperature, pressure and heating rates and gasifying agents also play a significant role in coal char reactivity [98].

Common factors that determine the reactivity include the formation of active sites, the microporous structure of char, diffusivity of reactants and inorganic mineral matter in coal [3,89,94,101]. The alkali and alkaline earth materials (AAEM) present in the ash helps to catalyse the char reactivity [79,83,84]. However, the inorganic ash minerals may have an adverse effect if they melt and sinter in the pores of char particles [99]. Micropores blocked by ash minerals prevent the gasification reagents from accessing the active sites of the char, resulting in a lower reactivity [83].

Whilst considering the effect of fuel properties, there is no general trend on the gasification reactivity based on the coal rank [102]. Generally, lower-rank coal possesses higher reactivity than higher-rank coal due to the higher proportion of oxygen-containing functional groups, micropores, and greater dispersion of catalytic inorganic compounds [99,103].

Wang et al. [66] conducted  $CO_2$  and steam gasification using bituminous coal under different operating conditions. The carbon conversion was reported to be 15–45% at temperatures between 800 and 900 °C using a heating rate of 20 °C/min. In contrast, the conversion was 50–60% with higher temperatures between 950 and 1000 °C. The highest conversion (65%) was achieved using a mixture of 60% H<sub>2</sub>O and 40% CO<sub>2</sub> with a heating rate of 1000 °C/min. In addition, the kinetic study of that study reported that the activation energy of coal was 192.56 kJ/mol using temperatures between 800–950 °C. Considering reactant type, steam is more reactive than  $CO_{2-}$ ; thus, gasification using steam leads to a higher carbon conversion under comparable operating conditions [104–106]. One way of measuring reactivity is by calculating the reactivity index. The reactivity index of coal/biomass gasification can be determined by the following equation [84].

$$R_s = \frac{0.5}{t_{0.5}}$$
(2)

where  $t_{0.5}$  refers to the time required to reach the 0.5 fractions of the fixed carbon conversion. Studies conducted by different authors [66,67] reported that the reactivity of lower-rank coal is much higher than that of higher-rank bituminous coal, which increases with increasing temperature, as shown in Figure 3.



**Figure 3.** Reactivity indices for different coal and char samples: (**a**) effect of  $H_2O/CO_2$  ratio at a temperature of 1000 °C and a heating rate of 20 °C/min [66]; (**b**) effect of temperature using a heating rate of 50 °C/min [67].

## 5.2.2. Modelling of Gasification Kinetics

The following generic formula can be used to express the immediate carbon-conversion rate during gasification [99,107]:

$$\frac{dX}{dt} = k(C_g, T)f(x) \tag{3}$$

where  $\frac{dx}{dt}$  denotes the rate of carbon conversion over time *t*; the reaction rate *k* is a function of the concentration of the reactant  $C_g$  and the temperature *T*; and the degree of change in physicochemical characteristics of char and the corresponding char structure is represented by f(x). The nth-order rate equation is commonly used to simulate the effect of temperature on carbon conversion. The rate constant and kinetic parameters were computed using the experimental carbon-conversion data.

Kinetic parameters determined for different types of coal under  $CO_2$  and steam atmospheres are illustrated in Table 12. It is to be noted that these kinetic parameters are highly sensitive to the sample preparation, experimental method and operating conditions and models used for the calculations. Overall, increasing coal quality/rank leads to increased activation energy. Furthermore,  $CO_2$  gasification requires 1.1 to 1.4 times the activation energy of steam gasification, while the reaction order is mostly between 0.4 to 0.6 under both gasification conditions.

Char Samples	E <sub>a</sub> (kJ/mol)		A <sub>0</sub> (s <sup>-1</sup> )		n ()		Ref.
Char Samples –	CO <sub>2</sub>	Steam	CO <sub>2</sub>	Steam	CO <sub>2</sub>	Steam	
Barapukurian bituminous coal	173.4	143.4	$1.21  imes 10^4$	$1.47  imes 10^3$	0.67	0.95	[99]
Morwell Brown coal	169.0	152.18	$1.69  imes 10^5$	$8.25  imes 10^4$	0.39	0.55	[105]
Yallourn Brown coal	168.8	152.18	$2.07  imes 10^5$	$7.14 \times 10^3$	0.48	0.44	[105]
Loy Yan Brown coal	164.9	119.24	$3.58  imes 10^4$	$4.59  imes 10^2$	0.44	0.53	[105]
Indonesian sub-bituminous coal	144.0	-	$1.74  imes 10^2$	-	0.4	-	[108]
High-ash Indian sub-bituminous coal	216.0	204	$3.05  imes 10^6$	$2.88  imes 10^6$	0.6	0.11	[109]
Australian (NL) bituminous coal	257.0	-	$2.54  imes 10^7$	-	0.56		[110]
Australian (BA) bituminous coal	283.0	-	$1.09  imes 10^9$	-	0.54	-	[111]
Chinese (S) bituminous	261.0	214	$1.23  imes 10^9$	$2.45  imes 10^4$	0.49	0.86	[111]
Semi-anthracite coal	282	-	$7.2  imes 10^8$	-	0.35	-	[112]
Jingcheng Anthracite coal	151.5	239.8	$1.53  imes 10^2$	$5.15  imes 10^6$	-	0.46	[113]

**Table 12.** Kinetic parameters for different types of coal using CO<sub>2</sub> and steam.

Several scholars have theorised models to match experimental data to find char gasification kinetics. The intrinsic kinetic model, interparticle heat and mass-transport models and the pore-structure model are the three major types of models [103]. The gasification rate is mostly controlled by a chemical reaction in the intrinsic kinetic model. Homogeneous and shrinking-core models are two simple intrinsic kinetic models [65]. The homogeneous model assumes that a uniform reaction occurs throughout the particle during the reaction time. Furthermore, it is considered that particle density varies relative to particle size during gasification, but particle size does not. The reaction rate can be equated as follows using the homogeneous model, often known as the volumetric model (VM) [114]:

$$\frac{dX}{dt} = k_{VM}(1-X) \tag{4}$$

where  $k_{VM}$  is the volumetric model's reaction rate constant.

The reactant diffuses via the gas layer around the particle, according to the grain model (GM), often known as a shrinking-core model. The reactant diffuses the unreacted core via the ash layers. The unreacted cores continue to shrink as the reaction advances, forming new unreacted cores [103]. The grain model can be stated in a general manner as [115]:

$$\frac{dX}{dt} = k_{GM} (1 - X)^{2/3}$$
(5)

where,  $k_{GM}$  is the grain model's reaction rate constant.

The char structure alters as the conversion progresses during gasification. To model char-gasification reactivity, the Random Pore Model (RPM) was created based on the char structure. The pores of the char become larger as the reaction progresses, according to this model, as carbon is consumed. However, after a given conversion, the pores merge, causing the reactivity to decline. The maximum reactivity is defined as the moment where the reactivity curve reaches a peak before dropping [116]. The separable form of the RPM is as follows:

$$\frac{dX}{dt} = k_{RPM} \sqrt{1 - \psi \ln(1 - X)} (1 - X)$$
(6)

where the reaction rate constant is  $k_{RPM}$ , and the term  $\psi$  is the no-dimensional structural attribute, which can be found using regression of experimental data fitting. In all of these models, the rate constant k is temperature-dependent and may be calculated using the Arrhenius equation as Equation (7). In contrast, the reaction-rate constant can be calculated

from the linear form of the models with the help of experimental carbon conversion, as shown in Table 13.

$$k = A_0 e^{E_a/RT} \tag{7}$$

where the pre-exponential/frequency factor is  $A_0$ , the apparent activation energy  $E_a$ , the molar gas is R, and the temperature is T.

Table 13. Summary of the kinetic models [105].

Models	Separable Form	Linear Form			
Volumetric model (VM)	$\frac{dX}{dt} = k_{VM}(1 - X)$	$k_{VM}t = -\ln(1-X)$			
Grain model (GM)	$\frac{dX}{dt} = k_{GM}(1-X)^{2/3}$	$k_{GM} = 3[1 - (1 - X)^{1/3}]$			
Random Pore Model (RPM)	$\frac{\frac{dX}{dt}}{k_{RPM}} = k_{RPM} \sqrt{1 - \psi \ln(1 - X)} (1 - X)$	$\frac{k_{RPM} t}{\frac{2}{\psi} \left[ \sqrt{1 - \psi \ln(1 - X)} - 1 \right]}$			

## 6. Challenges of Coal, Biomass and MSW Gasification

Gasification is a mature technology with an age of over two centuries. However, numerous challenges still need to be addressed to make the technology widely accepted [117]. The challenges include lower carbon conversion, low-quality syngas, the formation of tar and the issues related to the slagging of ash [33,118]. Several studies are available in the literature considering catalytic gasification to enhance carbon conversion [119–122]. Those studies showed a significant improvement in product gas quality, tar formation and pollutant emission, besides increasing carbon conversion.

Nonetheless, catalytic gasification has not been commercialised due to the high cost associated with the industrial catalyst [123]. Furthermore, other problems related to catalysts include degradation of their properties and evaporation at high temperatures [124]. Another critical issue with coal/biomass gasification is its slagging behaviour, which is more dominant using high-rank coals. The temperature required to form slag is significantly high, thus increasing the gasifier's operating temperature, which is energy-intensive [118].

Another key challenge faced during coal gasification is greenhouse-gas-emission reduction. To reduce the greenhouse-gas emission during coal gasification, clean coal technologies (CCT) can be applied at different stages of coal processing, such as preparation, gasification, removal of pollutants and capturing-storing CO<sub>2</sub> [125]. Advanced gasification technologies such as the HYCOL (hydrogen-from-coal) process, the EAGLE (coal Energy Application for Gas, Liquid, and Electricity) process, the Nakoso air/oxygen-blown IGCC, the CO<sub>2</sub>-recovery-type IGCC system and advanced IGCC/IGFC with exergy recovery technology (IGFC stands for integrated coal-gasification fuel-cell combined-cycle power-generating technology) are some of the examples of CCT for coal gasification [126]. Co-gasification of coal, biomass and MSW are also gaining importance due to their renewability and CO<sub>2</sub>-reduction potential [3,33,123].

Furthermore,  $CO_2$  recovery and utilisation technologies, such as in situ  $CO_2$  capture and utilisation technologies and  $CO_2$  recovery from the exhaust gas, can be applied to reduce greenhouse gas emissions [3,79,127]. Three basic  $CO_2$ -capture systems can be utilised during coal gasification.

- (i) Pre-gasification capture is utilised in IGCC power plants, where gaseous components (syngas) are produced from solid fuel by applying heat under pressure in the presence of oxygen and steam; CO<sub>2</sub> is captured from the syngas before the combustion process is completed [128].
- (ii) For in situ CO<sub>2</sub> capture during coal gasification, there are two that are sufficiently efficient: (1) calcium looping, which results in a gas stream with a low CO<sub>2</sub> content [129] and (2) chemical absorption using monoethanolamine, which is a commercially available technology that is routinely employed on a large scale [130]. Of the two, calcium looping is the most promising owing to its low cost and high reactivity [131].

(iii) Post-gasification CO<sub>2</sub> capture technology includes a water–gas-shift (WGS) reaction system, pressure-swing adsorption (PSA), and chemical-looping combustion (CLC). The WGS reactors use steam to convert vast quantities of CO generated during the gasification process into a CO<sub>2</sub> and H<sub>2</sub> blend. The PSA system provides a hydrogenrich fuel that is used to generate electricity in a combined cycle. The CLC system oxidises the residual CO and methane in the flue gas stream to produce a CO<sub>2</sub> stream that can be sequestered and a gas stream that can be delivered to the combined cycle to generate electricity [132].

# 7. Conclusions and Recommendations

The use of coal in conventional combustion technology has been shutting down worldwide. Hence, gasification technology might be crucial in utilising this cheap, reliable energy resource in the foreseeable future. Despite gasification being a mature and well-established technology, innovative ideas and drastic technological breakthroughs must address numerous technical issues. The biggest challenge in coal gasification is reducing greenhouse-gas emissions. Potential ideas currently becoming important include co-gasification of coal, biomass and waste (i.e., MSW), utilisation of CO<sub>2</sub> in the same cycle partially or fully and carbon-capture utilisation and sequestration (CCUS). Despite research and development in the last few decades, carbon-capture and sequestration technology has not been widely commercialised due to the high cost and the requirement of suitable geological locations. Recently, co-gasification technology has been gaining more priority, potentially replacing a proportion of coal with renewable biomass and solid water. Co-gasification generally increases carbon conversion, improves syngas qualities and lowers the slagging temperature. Hence, the combination of co-gasification and carbon capture and utilisation might be considered for pilot and commercial-scale demonstrations.

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