Catalytic Gasification of Coals and Biochars: A Brief Overview

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Abstract. Coal gasification is a mature technology for conversion of fossil coal to syngas and subsequentially produce chemicals and fuels. In recent years, gasification has been proposed as a suitable technology for conversion of biomass and waste materials, to promote renewable energy sources and circular economy. Gasification of polymeric and miscellaneous wastes often requires a pre-carbonization step to obtain a homogeneous raw material with better fuel properties. However, coal and biochar gasification face specific challenges that require adequate solutions. These materials have high chemical stability and low volatile matter content, characteristics that hinder their full gasification at low temperature. Also, they may yield large fractions of tars and impurities that must be removed from the syngas before it can be used in energy or material applications. The use of catalysts can mitigate these limitations by reducing the activation energy of gasification. Alkali metal and alkaline-earth metal (AAEM), transition metal compounds and natural catalysts are the most widely used catalysts in the coal gasification process, due to their catalytic activity, availability, and low cost. Composite catalysts may allow a higher efficiency, as they have strong synergies, higher stabilities and can improve the gasification conversion rate compared to single catalysts. The scope of this review is the assessment of gasification fundamentals and technology, and the specific conditions for coal and biochar gasification. In addition, the authors comparatively discuss various catalyst typologies in gasification, and sheds light on low-cost and environmentally friendly catalysts as a potential solution for coal and biochar gasification.

Keywords: Coals · Biochars · Catalysts · Gasification

1 Introduction

1.1 Exploring Gasification

The history of gasification is not recent, the first real application was developed in 1798 by William Murdoch that proposed coal gasification to obtain a combustible gas, at the

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time called town gas and that was used for lighting purposes. Various cities in Britain used town gas (also called coal-gas) for street lighting. From 1798 to 1850, the technology was improved, and several reactors were developed, but the invention of the electric bulb (after 1879) led to a significant decline of the town gas industry and gasification became confined to heating and cooking applications [1, 2]. Coal gasification regain interest during World War I and II, due to the scarcity of oil and refined fuels. The Fischer-Tropsch process to produce oil from syngas was proposed and used to obtain alternative liquid fuels [1]. Biomass gasification also became very popular during World War II, when approximately one million downdraft "gas producers" were used to power cars, trucks, boats, trains, and electric generators in Europe and in the United States [3]. After the war, interest in coal and biomass gasification technologies declined again due to the growing availability of cheap crude oil. The first commercial gasification plant (the Wabash River Coal Gasification Project) was implemented in 1999 in the USA [1, 2, 4]. The instability of fossil fuel prices (mainly oil), and concerns of climate change and environmental pollution, have put biomass and waste gasification increasingly under the spotlight, increasing the number of gasification projects around the world[4, 5]. Gasification is a key process for enabling the chemical utilization of carbonaceous resources to produce chemicals and fuels. The main application areas are: (1) chemical recycling of carbonaceous waste to replace fossil feedstock, (2) production of green hydrogen from (biogenic) waste, and (3) CO_2 neutral liquid fuels from biogenic wastes for transportation sectors [6]. The substitution of fossil fuels, such as coal, by biomass/biochar gasification is one of the current decarbonization strategies, allowing the capture of CO_2 or even negative CO_2 emissions [1].

2 Coal and Biochar Gasification: Main Limitations and Concerns

Gasification is the thermochemical conversion of organic solid material (coal, biomass, plastics, and organic waste) into a gaseous mixture of carbon monoxide (CO), hydrogen (H_2), carbon dioxide (CO_2), and methane (CH_4) via partial oxidation. Is typically carried out at temperatures of 700–1500 °C and atmospheric pressure in the presence of pure oxygen, air, steam, and CO_2 [5, 7, 8]. Gasification is very sensitive to the nature of the feedstock, that strongly influences gasification performance and syngas composition [9]. Coal or biochar gasification at moderate temperatures (~700 ºC), may lead to incomplete thermal decomposition given the high chemical stability of the raw materials. In those conditions it is expectable to occur the formation of tars and impurities that preclude direct use of syngas for power generation [10–12]. The efficient operation of engines coupled to gasification plants requires high-quality gas fuel [13], with CO and H_2 concentrations higher than 10% and a high calorific value very low tar content (<100 mg/Nm^3) and total absence of particles, ammonia, and sulphur dioxides (NH_3, SO_2) [14, 15]. Tar compounds may condense in compressors and gas engines, restricting their technical and commercial viability [16]. Other impurities such as ammonia, acid compounds and ashes need to be completely removed, to ensure proper operation of the energy conversion equipment [13, 17]. The high ash content and mineral composition of coals and biochars can also lead to the formation of high levels of solid by-products and fly-ashes [18]. Increasing the gasification temperature can improve the carbon conversion efficiency of coals and biochars, but may also increase CO_2 production, reducing

the calorific value of the syngas [11, 12]. The use of catalysts seems to be an option to mitigate these limitations by reducing the activation energy and improving the reaction efficiency of gasification and facilitating tar cracking at lower temperatures [17, 19]. Catalysts ease the thermal and mass transfer resistance through the particles while providing an alternative lower-energy pathway for the reaction to proceed [4].

3 Investigating Common Catalyst in Coal and Biochar Gasification

Alkali and alkaline-earth metals (AAEM) such as potassium (K), sodium (Na), calcium (Ca), and magnesium (Mg), and transition metal like iron (Fe), nickel (Ni), cobalt (Co) compounds, are the most frequent components of catalysts used for coal gasification [20, 21]. Sodium salts have been recognized to be excellent catalysts, which can accelerate gasification reaction rate and lower reaction temperature by 200–300 °C [22]. Most of these metals (Na, K and Ca) are already abundant in biochars therefore may act as natural catalysts, decreasing the cost of an added industrial catalyst [18]. The biochar reactivity in the gasification process is associated with structural features of char such as mineral content, porosity and surface area and active sites [23]. The AAEM'S that are inherently present in the biochar could also serve as catalytic active sites to induce tar reforming reactions [8]. However, the activity of aaem catalysts increases with increasing loading, so the number of catalytically active species contained in the biochar may not be sufficient. Therefore, increasing the number of active species by adding catalyst to the feed or to the gasification bed may be a more efficient solution [24].

3.1 Catalyst Classification: Single Component, Composite, and Disposable

Catalysts can be classified into single or composite materials, and reusable or disposable catalysts. In the case of single-component catalysts it has been shown that the catalytic activities of alkali metal are higher than those of alkaline earth metal and transition metal for three main gasification reactions [25]. For alkali catalyst the catalytic activity increases with the increase of relative atomic mass of alkali metal. For example, the order for catalytic activity of the alkali metal carbonates is $Cs_2CO_3 > K_2CO_3 > Na_2CO_3 > Li_2CO_3$. Due to the Cs high price, It is not suitable for industrial application, which makes k is the first choice, presenting a good catalytic activity and less tendency of coking. Catalysts containing K used in gasification are mostly k_2co_3 and koh which are compounds naturally found in biochar [26]. Apparent realizations of promoters can be obtained by combining catalysts. The combination of catalysts, results in Catalysts that may include binary composites or ternary composites. The K catalytic effect can be influenced by the presence of Ca, so positive synergic effects can be obtained when combining K and Ca catalysts. Several roles of ca had been verified during the gasification, such as promoting the gasification rate, protecting potassium avoiding deactivation, sulfur fixation, and *in-situ* carbon dioxide capture [20, 25, 26]. The steam gasification of corn cob biochar was evaluated using four alkali salts as catalysts (KOH, K_2CO_3, NAOH and NA_2CO_3) and the highest catalytic activity was obtained for alkali hydroxides. The maximum H_2 yield of 197.8 g/kg coal was obtained with 6%wt KOH [12]. Karimi *et al.,* (2011) reported that K_2CO_3 and Na_2CO_3 were the most effective catalysts for bitumen

coke steam gasification at 600–800 °C. When K_2CO_3, KCl, Na_2CO_3, $CaCO_3$, CaO and MgO are selected as catalysts [27]. Indeed, alkaline carbonates and hydroxides relatively have greatest catalytic activity compared with metal sulfates, nitrates, oxides, and acetates [20, 25, 26]. Other studies proved that composite catalysts (NA_2CO_3- $FECO_3$) have better catalytic activity than individual catalysts during coal gasification with CO_2. They have strong synergy, higher stability and can increase the gasification conversion rate by 100–500% compared to the effect of individual Na_2CO_3 and Fe_2CO_3-based catalysts [20]. However, composite catalysts, especially ternary ones, are very difficult to recover. on the other hand, disposable catalysts are cheap catalysts, usually corresponding to residues from agriculture, forestry, and industry, existing in abundance and widely available. They are economically and environmentally friendly catalysts. nevertheless, their catalytic activities are often low [25]. The comparisons of different types of catalysts are shown below in Table 1.

Table 1. Comparison of different types of catalysts. Adapted from [25].

Parameter	Alkali metal	Alkaline earth metal	Transition metal	Composite catalysts	Disposable catalysts
Representative	K	Ca	Fe	K-Na-Li	Biomass ash
Catalytic loading amount	Large	Medium	Small	Large	Small
Recovery	Difficult	-	-	Very difficult	No
Price	High	Low	Low	Medium	Very low
Activity	High	Medium	Medium	Very high	Low

Numerous studies on the catalytic gasification of coals and biochars with single, composite, and disposable catalytic species have been performed and are discussed in the next sections.

4 Investigating Catalysts for Tar Removal and Syngas Improvement in Coal and Biochar Gasification

Most of the work for tar minimization has been done in catalytic cracking of the tar because of the multiple advantages of catalytic degradation compared to the alternatives like mechanical method such as dry gas cleaning and wet gas cleaning, thermal cracking, and plasma method. Catalysts can degrade comparatively stable compounds such as aromatics and polycyclic aromatic hydrocarbon (PAHs). They can be employed *in-situ* during the gasification reactions or *ex-situ* after gasification reactions for tar removal and syngas quality improvement [4]. *In situ* gas cleaning (primary measures) involves incorporating or mixing catalyst with the feed or by the utilization of catalytically active bed material to achieve catalytic gasification [28]. *In situ* measures are attractive since they reduce the need for downstream cleaning equipment, and the energy content of the

undesirable species is retained in the product gas. Generally, naturally occurring minerals such as dolomite, limestone, bauxite, and olivine are utilized as the bed materials owing to their higher catalytic activities [29]. Some these catalytically active bed materials such as, olivine and bauxite are suitable options for adjusting the H_2/CO inside the gasifier, promote water-gas-shift reactions (WGS) and gas quality [29, 30]. The *ex-situ* gas cleaning strategies (secondary measures) use an additional reactor down-stream the gasifier to remove or convert the tar. It includes catalytic cracking with active bed material as used in the *in-situ* gas cleaning strategies. Catalytic tar removal takes place at a comparatively lower range of temperature 700–900 °C when compared to regular thermal processes (1100–1300 °C) [29]. *In-situ* catalysts are the most suitable and effective strategy for tar catalytic removal and H_2-rich gas production by reducing tar formation and enhancing the tar reforming into useful product gas [28]. For tar cracking, the most applied catalysts are nickel-based. Ni is characterized by high catalytic activity for the reformation reactions but its resistance to sulphur poisoning, sintering and carbon deposition strongly depends on the support material, promoters and other additives that are utilized in its manufacture. Also, they require cheap carbon sorbents to have a profound effect [4]. To date, potassium carbonate (K_2CO_3) has been the most suitable for catalytic gasification of industrialized coal. The K-Ca composite catalyst is a promising catalyst due to high gasification rate, CO_2 and H_2S capture removal, but is difficult to recover [25]. The main catalysts used in removing tar and promoting H_2 production from the syngas of catalytic gasification of coal and biochar are presented below in Table 2.

Table 2. Catalysts used in tar removing and H_2 production during coal and biochar gasification.

Feed	Atmosphere	Catalyst	Tar content (g/Nm^3)	H_2 content (vol.%)	Reference
Bitumen coke	Steam	K_2CO_3	-	68.3 (mol. %)	[27]
		Na_2CO_3	-	71.5 (mol. %)	
Yuyang coal	Steam	Alkali-feldspar and Quartz sand (bed material)	10.2	57.1	[29]
		Quartz sand (bed material)	55.5	47.9	
Bituminous coal	Steam	Calcined olivine (bed material)	16.8	50.9	[31]
Bituminous coal	Steam	Na^+ (1 wt%)	-	62.0	[24]
50% Brown coal + 50% pine sawdust	Steam	Calcined dolomite (bed material)	7.2	50.6	[32]

(*continued*)

Table 2. (*continued*)

Feed	Atmosphere	Catalyst	Tar content (g/Nm^3)	H_2 content (vol.%)	Reference
		Olivine (bed material)	9.1	47.2	
		Sand (bed material)	16.4	38.1	
Sawdust biochar	Steam	KCe (5%K + 5%Ce)	-	67.6	[33]
		KCo (5%K + 5%Co)	-	65.4	
Sub-bituniboous coal	Steam	Chicken calcined eggshell (20wt%)	-	64.0	[34]
Anthracite coal	Steam	Spirit-based distillers' grains (1:1)	-	68.0	[35]
80%coal + 20%polyethylene	Steam/O_2	Ni-dolomite	13.0	31.0	[36]
Bituminous coal + wood residue	O_2	K_2CO_3 (mass ratio 1.0)	9.62	52.05	[37]
60% sub-bituminous coal + 40% sawdust	Air	Silica sand (bed material)	5.61 (g/kg)	11.0	[38]
Lignite	Air	Quartz sand (bed material)	0.365	8.3	[39]
Turkish coal (25.62 wt% ash)	Air	Calcined dolomite (10wt%)	-	17.70	[40]
Bituminous coal	Air	Calcined dolomite (5wt%)	25.2 (g/kg)	-	[41]
70%Coconut shell + 30%Biochar	Air	Biochar (30wt%)	-	8.84	[42]

Although many types of catalysts can catalyze tar upgrading and improve syngas quality during gasification, some disadvantages may limit their industrial application. The high cost of the catalyst materials themselves, the decline in their performance and activity over time, and the difficulty in recovering and recycling some catalysts, triggers the search for efficient, cost-free, and highly stocked waste materials as gasification catalysts [43]. The search for environmentally friendly catalysts in coal and biochar gasification is a current goal to make these processes cost competitive in the markets.

Studies have been done in this direction. For example, Fan *et al.,* (2017), used calcined chicken eggshell as a catalyst in the steam gasification of sub-bituminous coal in a tubular fixed bed. The results showed that compared to the raw coal non-catalytic gasification, the use of calcined eggshell could improve the carbon conversion, reaction rate and yield of H_2 (80% higher) in the syngas [34]. Lv *et al.,* (2019), in steam gasification of blends of anthracite coal using spirit-based distillers' grains as a catalyst with mass ratios of 1:1, also recorded an increase in H_2 concentration compared to not using a catalyst [35]. Development on known catalysts and work on new and more innovative and suitable catalysts are promising options to minimize tar and improve syngas quality.

5 Conclusion

Gasification is a key process for enabling the chemical utilization of carbonaceous resources to produce chemicals and fuels. Coal or biochar gasification has some specific challenges such as the high ash content and mineral composition of these materials as well as the significant amounts of produced tars that must be removed from the syngas. The use of catalysts seems to be an option to mitigate these limitations. Most studies of catalytic gasification of coal and biochar use steam as the reaction medium, which ultimately favors hydrogen production. The use of air despite decreasing the costs of the process seems to have received little attention. Natural catalysts such as dolomite, olivine and quartz sand and composite catalysts (essentially carbonate catalysts) are the most used in catalytic gasification of coal and biochar, showing good catalytic activity in reducing tar and promoting H_2 production. More disposable waste catalysts with acceptable catalytic activities need to be developed to make these processes cost competitive. Catalytic gasification of coal and biochar can potentially become a widely used process on an industrial scale if the catalysts selected are cheap and efficient.

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