Mini-review on hot gas filtration in biomass gasification: focusing on ceramic filter candles

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ABSTRACT

Biomass gasification is increasingly attracting interest in the bio-renewable energy all over the world, as a carbon-neutral supplement to the conventional fossil energy. It can convert biomass to

the burnable producer gas for heat and power production or synthesis of fuels and chemicals. However, its commercial applications are seriously interfered by the minor but unavoidable contaminants or impurities in the raw product gas, which cause severe problems in downstream equipment. This paper reviews the recent progresses on the hot gas filtration technologies for removing particulate matters (PMs) and tars from the biomass-derived product gas, focusing on ceramic filter candles which are widely applied in the biomass gasification systems. Developments of PMs characterization, hot gas catalytic filtration and oxidative filtration, as well as the numerical simulation of computational fluid dynamics (CFD) in hot gas filtration are summarized in details. It also critically discusses the major challenges and future opportunities in hot gas filtration, and concludes that the combined oxidative filtration and catalytic filtration with highly efficient catalyst at moderate temperatures (<600 °C) should be the most economical option for the widespread commercial small-scale biomass gasification systems.

1. INTRODUCTION

Facing the serious challenge of global climate change due to fossil-derived CO_2 emissions, it has to be solved promptly in conjunction with the development and implementation of carbon-neutral routes by using the environmentally sustainable energy.[1] As the most plentiful organic materials produced by photosynthesis in green plants, biomass potentially represents the world's largest and sustainable energy source (ca. 4.5×10^{18} kJ/year) on the earth,[2] which accounts for 13%-14% of the total energy consumption, and more than two-thirds of the renewable energy mix.[3]

The alternative and renewable energy based on biomass, so-called bioenergy, can be converted by thermo-chemical and/or bio-chemical/biological conversion. In general, the thermo-chemical process has the higher efficiency in reaction time and the superior ability to decompose organic

compounds (eg. lignin) compared to the biochemical process.[4] Gasification is one of the promising thermo-chemical conversion technologies,[5] which converts biomass to combustible gases, often termed as product gas or producer gas, sometimes also called as bio-syngas or syngas when its N_2 concentration is very low. When heated to >500 ºC with a gasifying agent, as shown in **Table 1**, biomass can be transformed into the product gas that consists of a mixture of hydrogen (H_2), carbon monoxide (CO), methane (CH_4), carbon dioxide (CO_2), nitrogen (N_2), water vapor (H_2O), and minor impurities or contaminants,[6, 7] depending on the characteristics of the gasification process.

Table 1 Concentration ranges for typical product gas, contaminants and their upper limits [6-9]

Gasifying Agent	Temperature Range, ºC	The composition of the product gas, v/v%						
		H_2	CO	CH_4	CO_2	C_2's	N_2	H_2O
Air	780-830	5-16	10-22	2-6	9-19	0-3	42-62	11-34
Steam	750-780	38-56	17-32	7-12	13-17	2	0	52-60
Steam+O_2	785-830	14-32	43-52	6-8	14-36	3-4	0	38-61

Contaminants/Applications	PMs	Tar	NH_3	H_2S	HCl
The raw product gas	1-50 g/Nm^3	1-150 g/Nm^3	0.1-1.5 v/v%	20-200 ppmv	0.01-0.1 wt%
Internal-combustion engine	<50 mg/Nm^3	<50 mg/Nm^3	n.a.	n.a.	n.a.
Gas turbine	<30 mg/Nm^3	<10 mg/Nm^3	<50 ppmv	<20 ppmv	<1 ppmv
Fischer-Tropsch synthesis	<0.1 mg/Nm^3	<0.1-1 ppmv	0.02 ppmv	0.01 ppmv	0.01 ppmv
Alcohols synthesis	<0.1 mg/Nm^3	<1 mg/Nm^3	0.1 mg/Nm^3	1 mg/Nm^3	0.1 mg/Nm^3

n.a.-not available

In a real gasification system, as illustrated in **Table 1**, a few number of solid and fluid contaminants are unavoidably produced from the biomass gasifier,[10] which can create severe problems in downstream applications.[11] In most cases, the levels of these contaminants have to be reduced to the ppm level or below, prior to the utilization of the producer gas. In recent years, as listed in **Table 2**, significant research attention has been devoted to the cleaning of the biomass-derived producer gas to reduce contaminants below tolerable limits. In principle, raw gas cleanup can be done by several technologies but the one that offers more advantages is the hot gas filtration, which can intensify or simplify the cleaning process, protect the downstream equipment from

erosion and fouling, and prevent blocking by condensation or agglomeration.[12] Owing to the condensation of tars in biomass gasification (<300 ºC), the particulate matters (PMs) have to be preferentially removed at the highest possible temperature to achieve an optimum system efficiency and protect downstream equipment.[13]

Table 2. Summary of the main reviews including hot gas cleanup for biomass gasification.

Authors	Remarks / Main contents	Highlights / Distinctiveness	Ref.
G. Xiao *et.al.*, 2014	Review of granular bed filters for hot gas cleanup, including fixed, fluidized and moving granular beds	Summarize the basic principles, characteristics of various granular bed filters and their performances	14
K. Engvall *et.al.*, 2011	Gas upgrading technologies for pressurized fluidized-bed biomass gasification systems	Focusing on techniques for improved production of biofuels and chemicals via large-scale gasification	15
M.Asadullah *et.al.*, 2014	Advantages and disadvantages of different cleaning methods: physical, thermal, and catalytic gas cleanup	Compare technologies in gas composition, particles content, tar content, gas heating values & efficiency	16
P. Mondal *et.al.*, 2011	Gasifiers and cleaning options are reviewed in view of various feedstocks and downstream applications	Techno-economic analysis of gasifiers and syngas cleaning processes along with the world scenario	17
P. Aravind *et.al.*, 2012	Analysis of HT cleaning systems for the product gas of biomass gasification for fuelling SOFCs	Influence of bio-syngas contaminants on SOFCs, evaluation of cleaning options for CFB gasification	18
P. Woolcock *et.al.*, 2013	Description of contaminants in raw product gas, and overview of the technologies used to remove them	Clean technologies are classified as gas temperatures exiting the cleanup device: HGC, CGC, WGC	19
P. Simell *et.al.*, 2014	R&D works done at VTT in hot gas filtration, gas catalytic reforming, techno-economic evaluation	Highlight some recent research topics in the biomass-to-liquid concept development at VTT	20
Prabhansu *et.al.*, 2015	Removal methods of PMs, tar, alkali and heavy metals, sulphur, nitrogen and chlorine compounds	Summary of cleaning methods for contaminants and impurities from various gasification processes	7
S. Sharma *et.al.*, 2008	Syngas cleaning technologies of particulate removal systems and the practical problems and limitations	Summarize the research of energy technologies at the cLET and CSIRO over many years in Australia	13
S. Sharma *et.al.*, 2010	Status of hot syngas cleaning from coal gasification along with the shortcomings of dry hot gas filtration	Reporting a novel pulse less filtration concept and a system to prevent failure of filter elements	21
S. Anis *et.al.*, 2011	New technologies in tar treatment from biomass gasification with their strengths and weaknesses	Tar components are classified into five classes based on their chemical, solubility and condensability	11
S.Heidenreich *et.al.*, 2013	Review of fundamental aspects of HT filtration, hot gas filter media, systems, and its applications	A detailed survey on hot gas filtration with lots of industrial applications to raise awareness of HGF	12
S. Adhikari *et.al.*, 2015	Review of cold and hot syngas cleanup for major contaminants (tar, NH_3, H_2S, HCl and trace metals)	Extending the review to halides, trace metals and cold gas cleanup, catalysts are illustrated by tables	6
S. Li *et.al.*, 2021	Review of mostly sintered metal fiber filters, and a comparison with ceramic and powder metal filters	Assess the fundamentals, potential applications and design considerations of the metal fiber filters	22
W. Torres *et.al.*, 2018	Applications of basic, acidic, metallic, and redox catalysts for removal of tars, NH_3, H_2S from BGG	Focus on coke accumulation, resistance to sulfur poisoning, and removal reactions below 600 ºC	23

Considering the large number of fine PMs in biomass gasification gas (see Section 2), as shown in **Figure 1**, it could be concluded that the conventional hot gas cleaning technologies such as

cyclones,[24] electrostatic precipitators,[25] and granular bed filters[22] are not appropriate to achieve sufficient particle separation for the higher demands on emission levels, such as PM-10 and PM-2.5.[12] Therefore, the hot gas filtration is one of the important and essential issues for a successful application of biomass gasification. Few comprehensive reviews on hot gas cleanup have been published in the last decade (**Table 2**), focusing on (1) fundamentals and principles of filtration at high temperatures, (2) filter media and devices, (3) high-temperature (HT) removal of tar, inorganics and trace compounds, and (4) examples and advantages of hot gas filtration in various processes. The relevant research and development on biomass gasification were however not summarized in detail, especially for the hot gas catalytic filtration that can simultaneously remove PMs and tars at high temperatures (**Figure 2**). This review updates and complements the earlier ones by extending the review to the recent progresses on characteristics of biomass-derived PMs, HT catalytic filtration, HT oxidative filtration, durability of ceramic filter media, and the numerical studies by computational fluid dynamics (CFD) that have previously only received limited attention. Moreover, some existing barriers, practical issues and important challenges of the hot gas filtration are critically discussed, particularly for the ceramic filter candles, which are widely applied in the bench-scale and the large-scale biomass gasification systems. To narrow the scope, a few articles focusing on the metal filters are not involved, which have been covered by another recent review.[22]

Figure 1. Collection efficiency versus particle size[22, 26] (A) and particle size distributions downstream the cyclone of a CFB biomass gasifier[18] (B). Reproduced from Aravind *et. al.*[18], Baeyens *et. al.*[22] and Hasier *et. al.*[26], Copyright 2021 Elsevier.

Figure 2 Schematic illustrating the different steps and processes included in a gasification system.

2. PARTICULATE MATTERS FROM BIOMASS GASIFIERS

Primary particulate matters (PMs) from biomass gasification originate from three main sources: (1) mineral fly ashes in the solid form from complete combustion or bed materials, (2) tiny char fragments abraded from the pyrolyzed biomass feedstocks in gasifiers, (3) nanometer-sized aerosol particulates formed by condensed mineral vapors and carbonaceous particles of incomplete combustion.[27]

2.1 Mineral fly ashes

It has been reported that mineral fly-ashes in biomass gasification gas had a bimodal particle size distribution with a fine mode (<0.5 μm) and a coarse mode (>0.5 μm).[28] The mineral PMs comprised of ashes and bed materials mainly represent the original solid particles from biomass gasifiers, which are almost invert at the moderate filtration temperature (300-500 ^{o}C). Generally, the fine fly ashes are produced from vaporization of easily volatilized ash components (S, Cl, Na, and K) and some heavy metals (Zn, Hg, if present), and the coarse fly ashes formed from mineral particles ejected mechanically from the fuel bed or intractable ash compounds (Ca, Mg, Si, if present).[29] But it was also mentioned that the mass concentration of mineral PMs could be changed with the feedstocks, bed materials, and gasifiers. If miscanthus was gasified, the fine PMs contained mainly potassium and chlorine, whereas if wood was gasified, magnesium and calcium were the dominant elements.[30] The coarse mineral PMs in the producer gas of a bubbling fluidized bed (BFB) gasifier (20 kW_{th}) mainly comprised chars abraded from the pyrolyzed wood pellets, whereas the fine PMs were dominated by calcium.[27] In contrast, for a steam-blown indirect BFB gasifier (2-4 MW_{th}) using wood pellets, the coarse mineral PMs were found mainly to comprise calcium and silicon, while the fine PMs were dominated by potassium and chlorine.[31] Therefore,

the impact of parameters, such as gasifier type, gasification conditions, biomass feedstocks, sampling position, and measurement system, should be further investigated. In particular, the behavior of the condensed mineral vapors from biomass gasification needs to be addressed for the evaluation of the fine-mode ashes during the hot gas filtration.

2.2 Tiny char fragments

In a real biomass gasifier, an unavoidable part of biomass or char fragments is entrained by product gas and carried to the high-temperature filter, depending on gas velocity, particle size, particle density, feedstock, and gasifier design.[25] The size distribution of char PMs is usually in the coarse-mode with an aerodynamic diameter (d_{ae}) of 0.5-20 μm, and decreased with increasing particle diameter (d_{ae} > 3μm) due to the inertial losses.[28] These coarse-mode PMs should be very easy to remove by the hot gas filtration if they were just like the normal dusts in combustion gas that are almost kept invariable at the high filtration temperatures. Unfortunately, char PMs from gasification and fast pyrolysis are physically and chemically different from the traditional charcoals prepared by slow pyrolysis, which appears to depend on process temperature and, to a lesser extent, reaction time.[32]

Figure 3. TG curves (A) and FTIR spectrum (B) of the char PMs from a fixed-bed biomass gasifier.[33]

Our group investigated the isothermal pyrolysis characteristics of char PMs at 400 °C,[33, 34] which were collected from a hot gas filter (400-600°C) downstream from an industrial fixed-bed gasifier using wood chips as feedstock. As shown in **Figure 3**, char PMs in the product gas should be incompletely pyrolyzed in the gasifier. Some residual organic functional groups could be found in the FTIR spectrum of char PMs, and its major pyrolysis region is from 350°C to 600°C, which coincides with the temperature ranges of hot gas filtration. As a result, more additional volatiles

(tars) were decomposed and released on the filter candles at the filtration temperatures,[34] which cause the higher press drop (ΔP) during the hot gas filtration in the biomass gasification environment **(Figure 4)**.[35, 36] According to this, a new concept of high-temperature oxidative filtration for biomass air gasification (BAG) had been verified both by laboratory and pilot test, which leads to a simultaneous decrease of ΔP and tar concentration, by continuously introducing a small amount of additional air (~2%O_2) into the raw BAG gas.[37] The XPS and NMR results proved that more surface oxygen-containing organic functional groups could be generated via the partial oxidation of the char PMs in the BAG+2%O_2 atmosphere at 400 °C, possibly inhibiting the aromatization of aromatic clusters and the formation of heavy tar compounds to some extent.[34] Tuomi et. al.[38] also pointed out that thermal reactions of tars could be promoted by the catalytic reactions induced by the unreacted biomass char on the filter surface at high temperatures. In addition, char PMs (d_{ae} <10 μm), even for completely pyrolyzed commercial activated charcoals and pine chars prepared at 900 °C, can help to convert tar (eg. benzene) in the high-temperature filtration at 750-1000°C.[39] Therefore, it is inevitable for the tar-char interactions in a real biomass gasification environment, which has great influences on the hot gas filtration. More attention should be paid to the proper design and functionality of such filters and char PMs in upcoming studies.

Figure 4. The isothermal pyrolysis[33] (A) and the isothermal filtration[36] (B) of char PMs in different atmospheres at 400 °C, BAG: the simulated product gas, BAG+2%O_2: BAG gas containing 2.0vol.% O_2.

2.3 Nanometer-sized aerosol PMs

Both online and offline instruments had been used to characterize the nano-sized aerosol PMs in the hot product gas from biomass gasification,[30, 40, 41] and particle size distributions with two

distinct modes were established.[30] The fine mode (15-140 nm, 640 mg/m^3, 2×10^{10} PMs/cm^3) was supposed to contain mainly heavy tars formed through homogeneous nucleation and condensation on alkali nuclei, another intermediate mode (140-670 nm, 38 mg/m^3, 4.3×10^6 PMs/cm^3) was assumed to be a mixture consisting of original PMs formed by condensed alkali vapors and tars from the gasifier.[30] The fine-mode aerosol PMs, also known as soot, are an absolute majority in the nano-sized PMs from biomass gasification.[42]

For industrial biomass gasifiers, reactions such as pyrolysis and combustion are considered to occur simultaneously with the gasification. So it is inevitable for the generation of soot with tens to hundreds of nanometers, which is formed by incomplete combustion.[42] Very recently, a comprehensive summary of soot formation in biomass gasification was provided by He et. al.,[42] who pointed out that the initial components of the biomass feedstocks play the decisive role in soot formation during biomass thermal conversion. As illustrated in **Figure 5**, the alkali and alkaline earth metals (AAEMs, inherent catalysts) in biomass and the polycyclic aromatic hydrocarbons (PAHs, soot precursors) in heavy tars should be highlighted in the soot formation during biomass gasification.[43, 44] However, the elucidation of soot formation in biomass gasification gas is extremely difficult, for a variety of instantaneous processes are involved during the hot gas filtration, such as nucleation, coagulation, aggregation and surface growth.[45] Until now, the literature concerning soot mechanism from biomass gasification is still very limited.[42]

Figure 5. Effects of inorganic ash[44] (A) and heavy tar [43] (B) on the formation of nano-sized soot. Reproduced from Jarvis *et. al.*[43], Copyright 2021 ACS, and Xiao *et. al.*[44], Copyright 2021 Elsevier.

Gall et. al.[40] developed a volatility tandem differential mobility analyzer (VTDMA) method for the characterization of alkali and heavy tar compounds in the hot product gas formed from a 4-

MWth dual fluidized-bed (DFB) gasifier. As shown in **Figure 6**, the volatile fractions consisting of different tar and alkali compounds evaporate and the particle size decreases with the increasing temperature of the product gas. Most of tars (90 vol.%) evaporate at temperatures below 150 °C, even for the tar compounds with a boiling point around 400 °C (e.g. pyrene, 404 °C) that evaporate around 100 °C to heavier compounds observed by the VTDMA method. The particles continue to evaporate with rising the temperature, and only 3-4 vol.% remains at 300 °C. Additional reductions in size over 500 °C may be associated with the evaporation of alkali compounds.

Figure 6. Remaining volume fraction of size-selected particles sampled from a DFB gasifier (points) as a function of oven temperature. Results from laboratory experiments (lines) are included for comparison.[40] Reproduced from Gall et. al.[40], Copyright 2021 ACS.

It can be clearly seen in **Figure 6** that heavy tars and semi-volatile ashes (e.g. AAEMs) are both present in vapor phase at the high filtration temperatures over 800 °C. However, some major problems are associated with high-temperature (>600 °C) filtration of product gas from biomass gasifiers. First of all, the evaporative alkali and tar species could favor the formation of nano-sized soot PMs in the inner pores of candle filter elements,[42] which often results in the serious blinding of filter elements at temperatures above 600 °C.[20] In addition, alkali and water vapors potentially have deleterious impacts on the life of porous ceramic filters that are currently utilized in advanced biomass gasification.[46, 47] Moreover, alkali vapors could pass through the filter candles at high temperatures, and condense during cooling downstream from the filter,[48] producing new particles or deposits on catalysts or other devices.

In summary, the quantity of carbonaceous PMs is much higher in the product gas from biomass gasification than in combustion gas, owing to the sub-stoichiometric conditions in the gasification

process. The filtration efficiency, the operation and dimensioning of the filter depend upon the particle mass concentration and size distribution, as well as upon the physicochemical properties of the PMs from biomass gasification. For hot gas filtration, therefore, it is crucial to characterize and control the particulate matters present in the raw product gas.

Table 3. Characteristics of the hot gas particulate cleanup technologies [14, 19, 25]

Devices [a]	η, %	T, °C	ΔP, kPa	U_g, cm/s	Advantages	Disadvantages
Cyclones	<80	~1000	<1.0	Very high	Simple, Low-cost, High T	Very low η
Electrostatic Precipitators	<95	<450	<1.0	Very high	Moderate to high	Low to moderate T, Low η
Granular bed filters						
Fixed-bed	>95.0	<650	5-20	<0.8 [b]	High η, Moderate T	High ΔP Intermittent operation
Moving-bed		>800	0.5-5	High	High η & T, Low ΔP Continuous operation	Complex operation, Large footprint, High throughputs
Hot gas filters						
Ceramic candle	>99.0	~1000	2.0-15	1-5	Very high η & T, High permeability, Low weigh	Fragile structure, Corrosion over 800 °C, Length-limited
Ceramic tube			8-12.5	3-5	Catalytically activated Easy to back pulsing	Breakages due to thermal stress, Leakages at joint portions
Honeycomb monoliths			10-20	3-7	Commercial used as diesel soot filter in vehicles	Risk of plugging channels, Poor performance of back pulsing
Metallic candle			5-15	1-5	Thermal and mechanical stability, High porosity	Oxidation and corrosion lead to irreversible plugging of pores

[a] η-Collection efficiency of fine particles, ΔP-Pressure drop, T-Operating temperature, U_g-Face velocity in at T, [b] Means gas velocity in filters, m/s

3. HOT GAS FILTRATION IN BIOMASS GASIFICATION

Hot gas particulate cleanup is one of the most important improvements to commercial applications of biomass and coal gasification in the past 30 years.[49] Many applications benefit thermo-dynamically by cleaning the producer gas at elevated temperatures above 200 °C,[19] such as reduced waste streams, increased efficiencies, avoiding erosion and fouling in downstream units, and removal of alkali, heavy metal and chloride.[50] As shown in **Table 3**, various techniques

have been applied to separate PMs from the gasification gas at high temperatures, most of which are based upon one or more of the following particulate removal technologies: cyclones, electrostatic precipitators (ESP), granular bed filters (GBF) and hot gas filters (HGF). Among them, hot gas filtration, using ceramic or metal candles as the rigid barriers, is one of the most promising method in hot gas cleanup for advanced biomass gasification technologies,[12, 14] which have an extremely high cleaning efficiency, approximately 99.8%, especially when PM-10 and lower levels of PMs need to be removed (**Table 1**).

3.1 Development of hot gas filtration

In general, the gas filtration at temperatures above 260 ºC is called as hot gas filtration based on the VDI guideline 3677 Blatt 3-2012,[51] which can be conducted by removing the PM by rigid barrier filters under hot conditions. Beginning with the 1970 Clean Air Act,[52] hot gas filtration was early used in some incineration plants or nuclear power plants,[12] in order to remove contaminants that would otherwise be emitted to the environment as pollutants. From the end of the 1980s to the end of the 1990s, about 25 large hot gas filter units had been applied in the coal-based IGCC (integrated gasification combined cycles) projects worldwide,[12, 53] to investigate and test different hot gas filter media and filter systems. At the same time, hot-gas-filtration research was initiated at the Technical Research Centre of Finland (VTT), in connection to biomass based IGCC systems via co-gasification of biomass and coal.[20] Unlike the long-term stable operation in coal-gasification tests, ceramic filters were almost completely blocked in several hours during the earlier biomass gasification experiments at high temperatures (690-720℃), which was considered to be caused by the formation of soot and polyaromatic tars on the filter resulting in a sticky filter cake that was difficult to be removed by traditional pulse cleaning.[54] Thanks to the new generation rigid filters and the integrated fail-safe system,[55] the long-term operation of hot gas filtration was partially

achieved in biomass gasification & pyrolysis at the turn of the 21st century.[55, 56] This paper gives a review of hot gas filtration R&D works using rigid filters since that time, particularly in the last twenty years.

Figure 7. Schematic of the hot gas filter elements. Reproduced from Heidenreich *et. al.*[12], Copyright 2013 Elsevier. (请小朱补充该引用授权)

3.2 Ceramic filter candles (CeFC)

As illustrated in **Figure 7**, several types of rigid barrier filter elements exist, including: filter candles, filter tubes, and honeycomb monoliths.[12, 57] It is recognized that ceramic candle filters are the preferred geometry of hot gas filter elements,[55] because of their excellent mechanical, thermal and chemical stability, long-term durability, as well as the high filtration efficiency up to nearly 100%, even for the submicron particles.[12] Sintered metal filter candles are an alternative to the ceramic candles at high temperatures, which are made of different steel grades and various metal alloys.[12] For most biomass gasification environment, however, steam, ammonia, sulfides, and chlorides are present in the producer gas. In this case, only a few special metal alloys can be applied, but oxidation and corrosion lead to irreversible plugging of the pores in the long-running instance.[58] Although the metal filter candle had been practiced in some demonstration trials of coal gasification,[12] its applications in biomass gasification is still rarely reported, especially for hot gas filtration above 800 ºC. This section will only focus on the experimental results obtained for ceramic filter systems (

Table 4), and advances in porous metal filters has been reported in another recent review.[22]

Table 4. Performance comparison of hot gas filtration studies in biomass gasification

Biomass	Bed material	T_g, ºC	ΔP, kPa	U_T cm/s	Pulse time min	Running Time, h	Org. & Ref.
Wood	n.a.	320-400	9.0	n.a.	15	~2000	Bioflow Ltd.[56]
Wood	Sand	580-650	2.5-5.0	5.0	5-10	50-58	TU Delf [59]
	Sand & MgO	800-820	6.0-15	3.0			
	Magnesite [b]	800-820	3.0-3.5	3.0			
Miscanthus	Magnesite		6.5-9.0				
	Olivine	790-820	14-16	2.5-3.0	10-15	50	TU Delf [48]
Straw	Magnesite		12				
Wood	Dolomite	780-810	n.a.	1.5-2.0	n.a.	~30	VTT [38]
Charcoal	Nothing	400-450	1.5-3.5	2.0-4.5	150-300	~50	GIEC [37]
Wood			4.0-5.0	2.0	20-60	~5.0	
Wood	Little oxygen	400-600	1.0-2.0	2.0-3.2	300-500	~200	

[a] ΔP-Pressure drop, T_g-Filter temperature, U_T-Face velocity at T_g, **Pulse-**Jet pulse recleaning, **n.a.**-not available
[b] A new set of DS candles (DS3) was chosen for that tests.

Ceramic candle filters had performed satisfactorily over 2000h in the biomass IGCC demonstration plant in Värnamo, Sweden,[56] at the temperature range of 320-400 ºC. Reaching filtration temperatures above 500-550 ºC has, however, shown to be challenging,[38] due to the serious blinding of filter elements at temperatures above 600 ºC. Thus, recent researches in Europe have been focused on increasing the filtration temperature closer to the temperature in the fluidized-bed gasifier outlet, in the range of 800-850 ºC; which would increase the efficiency of biomass gasification by keeping off the extra cooling and heating steps upstream and downstream the filter.[20] As a part of the CHRISGAS project (Clean Hydrogen-rich Synthesis Gas), a detailed study on the filtration performance at temperatures about 800 ºC was performed at Delft University of Technology (UT Delft).[48, 59] During the lab-scale test runs with Dia-Schumalith ceramic filter candles, it is proved that the selection of bed material and biomass feedstock combinations plays

an important role on the stable filter operation and gas quality, especially for the pressure drop and the heavier tar compounds.[48] But tar behavior in filtration conditions at high temperatures is not yet well-known. Soon after that, a deeper understanding of the behavior of tars in hot gas filtration over 600 ºC was proposed by the VTT,[20, 38] based on the air/steam gasification of woody biomass. Thermal tar reactions were found on the high-temperature filter surface induced by unreacted biomass char and carry-over bed material, regardless of the biomass feedstock, bed material or gasifying agent. As a result, a dense and sticky cake layer is formed on the filter candle, and the thermally generated soot may penetrate the filter pores which ultimately leads to filter blinding at elevated temperatures.[20] As shown in **Figure 8**, the higher filtration pressure was found to intensify the filter-blinding effect, though the presence of steam in the gas could inhibit the blinding effect to a certain extent, owing to the steam gasification of carbon that may take place on the filter surface.[20]

Figure 8 Test runs performed in nitrogen/steam atmosphere at 800 °C.[20] Reproduced from Pekka *et al.*[20], Copyright 2021 Springer Nature.

A similar filter-blinding phenomenon was discovered by our group at the intermediate temperature (300-600 ºC) as well,[37] due to the polycondensation of gasified chars and tars (Section 2).[34] As shown in

Table 4, the pressure drop (ΔP) during the wood gasification was over twice than that of the charcoal gasification with low tar content, which gave rise to unstable operation of wood

gasification-filtration. It's confirmed that tars should be the controlling resistance in hot gas filtration of biomass gasification. Thus, an innovative concept of oxidative hot gas filtration for biomass gasification had recently been developed by the Guangzhou Institute of Energy Conversion (GIEC) in China,[35, 37] which leads to a simultaneous decrease of ΔP and tars, by continuously introducing a small amount of additional oxygen into the producer gas; Meanwhile, it also allows efficient particulate removal (>99.0%), and a steady oxidative filtration with low pressure drop (<2.0 kPa) was operated for over 200 h during the pilot-scale woody biomass gasification in 400-600 ºC.[37]

3.3 Catalytic filter candles (CaFC)

Since 2000, a concept of catalytically activated ceramic filter candles was introduced to perform the simultaneous removal of tars and particulates from hot biomass gasification gas.[60] It has been carried out in the bench-scale fluidized bed biomass gasifiers in recent years,[61] and the best performance of the catalytic filter candle is proven, concerning pressure drop, water conversion, tar content and ammonia decomposition. **Table 5** summarizes the ongoing development of CaFC studies in biomass gasification. The nickel-based catalysts using MgO/CaO as a promoter have been the most effective at temperatures ranging from 800 to 900 ºC, even in the presence of sulfur compounds.

3.3.1 Proof of the CaFC concept

The catalytic feasibility of the CaFC concept was firstly demonstrated on lab-scale by G.V. Baron's group, the high removal efficiencies of benzene and naphthalene (tars) were observed using α-Al_2O_3 filter loading 1 wt% nickel at 900 ºC while applying a sulphur-free and dust-free simulated producer gas.[60] Considering the trace H_2S in real biomass gasification gas (50-200 ppm),

a series of promoters such as CaO, MgO, ZrO_2 and CeO_2 were then added to the Al_2O_3-based filter to enhance the resistivity to sulphur poisoning of nickel (Ni) catalysts, which were prepared by various methods like incipient wetness,[60, 62] urea precipitation,[63] and co-precipitation with urea.[64] The NiO-MgO-Al_2O_3 component was subsequently exposed as the high efficiency Ni catalyst with a much better sulfur tolerance.[64] Increasing the surface area and the Ni loading could additionally provide an effective improvement to the catalytic filter disk, allowing a nearly 100% naphthalene conversion even in the presence of a H_2S concentration of 200 ppmv.[65] In case of the similar catalyst coating on the pore walls of commercial filter candles, however, the lower naphthalene conversion of 58% and 97% was achieved in the presence and absence of H_2S, respectively.[66] Recently, another kind of the calcium silicate (CaSiO3) based filter disk was catalytically activated by coating a Ni/γ-Al_2O_3 layer. The higher methane and benzene conversion (77%-88%) were achieved in the absence of H_2S, while H_2S-containing gas caused significant activity loss despite adding the MgO promoter to suppress sulfur-catalyst interaction, especially for the CH_4 conversion.[67] Although the estimated naphthalene conversions in a disk model system can be used as bases for the prediction of real tar conversions in a bench-scale gasifier,[68] to investigate the catalytic activity of the CaFC candle under real biomass gasification environment is often more important for the simultaneous removal of particulate and tar.

Figure 9 Scheme of configurations of the catalytic filter candles

3.3.2 CaFC studies on bench-scale biomass gasification

There are at least five different catalytic filter designs have been investigated in the biomass gasification process. All of them are illustrated in **Figure 9** with the following specifications:

CeFC: the ceramic candle without catalysts

CaFC-A: the ceramic candle coated catalytic layers in the pores

CaFC-B1: the ceramic candle partially filled with catalyst grains

CaFC-B2: the ceramic candle totally filled with catalyst grains

CaFC-C: the ceramic candle filled with a catalytic inner tube (porous ceramic foam)

CaFC-X: the arbitrary combination of double or multiple CaFCs described above

Figure 10 Schematic illustrating the new concepts of the UNIQUE project.[69] Reproduced from Rapagna *et al.*[69]. Copyright 2021 ACS.

Table 5. Summary of some typical CaFCs in the hot gas filtration for biomass gasification.

Catalysts	Filters	Reaction conditions (running time, h)	T_g °C	U_T cm/s	ΔP kPa	X_{tar} %	Ref.
1.0% Ni	α-Al_2O_3 disk	Naphthalene=5g/Nm^3, MHSV=1.39s^{-1}	900	2.5-5.0	n.a.	100	60
1%Ni0.5%CaO	α-Al_2O_3 disk	Benzene=15g/Nm^3, H_2S=100ppm	900	2.5-4.0	n.a.	>78	70
1%Ni0.5%MgO 2.5%Al_2O_3	α-Al_2O_3 disk	Naphthalene=5g/Nm^3, H_2S=100ppm Benzene=15g/Nm^3, H_2S=100ppm	900	2.5	3.0-5.0	99.3 95.4	62 64
1.0%/0.5 % Ni/CaO	α-Al_2O_3 disk	Naphthalene=5g/Nm^3, H_2S=100ppm Benzene=15g/Nm^3, H_2S=200ppm	900	2.5	n.a.	98.0 57.0	63
Ni@γ-Al_2O_3	$CaSiO_3$ disk	Benzene=15g/Nm^3, GHSV=20000h^{-1}	750	5.5	~2.5	~85	67
6%Ni-MgO	CaFC-B1	Naphthalene=5g/Nm^3, H_2S=100ppm	800	2.5	~5.5	~100	65
NiO@MgO-Al_2O_3	CaFC-A	Naphthalene=5g/Nm^3, no H_2S Naphthalene=5g/Nm^3, H_2S=100ppm	800	2.5	~5.5	97.0 58.0	66
NiO@MgO-Al_2O_3	CaFC-A	*in situ* FBG & olivine, F_B=8-10g/min using both CaFC and CeFC filters	675-840	2.5	n.a.	58.0	69
NiO@MgO-Al_2O_3-X	CaFC-B1 CaFC-A	Naphthalene=5-10g/Nm^3, H_2S=100ppm 50 h long-term tests	800	2.0	n.a.	100 74.0	71
6%Ni-MgO	CaFC-B1	*in situ* FBG & olivine, F_B=4-8g/min, 22 h	800	2.0	n.a.	79.0	72
NiO@MgO-Al_2O_3	CaFC-A CaFC-C	Naphthalene=5g/Nm^3, H_2S=100ppm water content of up to 30vol.%	850	2.5	n.a.	87.0 98.0	73
NiO@MgO-Al_2O_3	CaFC-A (I) CaFC-B1(II)	FBG & olivine, F_B=4-6g/min test-I: 6h, test-II: 20h,	810	1.95 2.35	1.7-3.0 2.5-3.0	59.8 93.5	74
NiO-MgO	CaFC-C	Model/real gas, F_B=10g/min, H_2S=40ppm	850	2.0	n.a.	97	68
NiO@MgO-	CaFC-A	*in situ* FBG & olivine, F_B=8-10g/min	800-	2.5-	4.0-	87.5	61

Al_2O_3	CaFC-C	three types of filters were tested	815	2.8	4.5 5.5-6.5	98.0	
6%Ni-MgO	CaFC-B1	FBG & Fe/olivine, 25h, GHSV~3000h^{-1}	~800	1.5-2.5	n.a.	75.0	[75]
NiO-MgO	Four types	FBG & Fe/olivine, 1-4h, GHSV~2000h^{-1}	800	1.5-2.5	n.a.	80.0	[76]
NiO-MgO	CaFC-C	*in situ* FBG & olivine/dolomite, F_B=800g/h	~800	2.5-3.2	5.6-6.8	99.3	[77]
15wt% Nickel*	CaFC-B1 CaFC-B2	*in situ* FBG & olivine, F_B=800g/h, GHSV~5500h^{-1}, catalyst=550-750g	~800	2.8	3.5 5.5	92.4 88.1	[78]

CaFC: Catalytic Filter Candle, **FBG:** Fluidized-bed gasifier, ***F_B*:** the feeding speed of biomass feedstock

* it is a commercial conventional steam reforming catalyst supplied by Johnson Matthey

According to the European UNIQUE project (**Figure 10**), an innovative concept of integrating a catalytic filter candle in the freeboard of a fluidized-bed gasifier was put into practice.[69] Compared with a non-catalytic filter, a 58% conversion of tar and a 28% conversion of methane was obtained, and led to a simultaneous increase of the gas yield and hydrogen content. Meanwhile, two types of full-size catalytic filter candles (CaFC-A&B1) were also investigated for their catalytic activity via hot gas cleaning of biomass-derived syngas.[71] The CaFC-B1 filter had proven higher catalytic performance than the CaFC-A filters with a series of five developed catalytic layers, and showed a complete naphthalene conversion at 800 °C in the 50h long-term test in the presence of 100 ppmv H_2S at a typical face velocity of 2.0 cm/s. Subsequently, this CaFC-B1 candle was applied into the bed freeboard of a lab-scale biomass gasifier in 800-820 ^oC.[72] After a long-term gasification test (22 h), the catalytic filtration gave rise to notable improvements in gas and hydrogen yields, with corresponding decreases in methane and tar content of 20% and 79%, respectively. Several parameters affecting the performance of the CaCF-B1 filter were systematically analyzed in a dual fluidized bed gasifier, which shows that the amount of tar did not affect the tar conversion (~75% at 800 ^oC), but the tar conversion decreased when the face velocity increased from 1.1 to 2.5 cm/s.[75] Recently, the CaCF-B1 and CaCF-B2 filters

filled with commercial Ni-catalyst pellets were investigated at the freeboard of a bench-scale fluidized-bed gasifier, in order to verify the effectiveness of diverse layouts for the inner bed of catalyst pellets.[78] At the same gas velocity (2.8 cm/s), it was found that the ΔP value of CaCF-B1 is approximate to that observed for the empty candle (~3.5 kPa), while for the CaCF-B2 candle it was significantly higher (~5.5 kPa). As shown in **Figure 9**, it is explained that the gas flow path is different in each configuration: gas flows radially and uniformly through the CaFC-B1, on the other hand, a deviation of the gas flow path occurs in the bottom region of the CaFC-B2 due to its comparatively higher resistance, mainly towards its upper part. This is contributed to the more effective promotion of tar and methane steam reforming reaction by the CaFC-B1 candle. So that the pilot testing with the CaFC-B1 configuration is highly recommended, which could leave a cylindrical, internal hollow space for the hot gas to flow towards the candle head.

In order to improve the tar reforming performance of a CaFC filter in biomass-derived syngas, as new approach, ceramic foams were often used as the support for catalyst integration in recent years. M. Nacken et. Al. reported that the overall activity of a CaFC-C candle with an integrated hollow-cylindrical catalytic ceramic foam (HCCF) was precalculated to 98% naphthalene conversion that was by 18% higher than the achieved conversion of a CaFC-A candle without integrated HCCF.[73] Based on this result, the catalytic activity of the CaFC-C candle with a HCCF catalyst was further tested in the freeboard of a biomass gasifier, and compared with other type filters.[61, 74] It is confirmed that the catalytic performance of the CaFC-C candle with HCCF is much more promising and stable over the 20h steam biomass gasification, which shows an increase in tar conversion from 59.8% to 93.5% compared with the experimental data of the CaFC-A candle.[74] Concerning all the gasification tests, the rate of ΔP increase of noncatalytic filter candles (CeFC) always showed higher values with respect to the catalytic filters (CaFC), and the best performance

of the CaFC-C candle with integrated HCCF is proven, also for what concerning water conversion, hydrogen content and ammonia decomposition.[61] Furthermore, the tar removal efficiency of four different CaFC filters was evaluated with real biomass tar produced in a dual fluidized-bed gasifier. Despite the tar conversion was higher for all the CaFC candles, as illustrated in **Table 5**, the most promising CaFC design is the catalytic layer integrated with a catalytic alumina foam tube (CaFC-A + CaFC-C), in which the tar removal efficiencies up to 95% at 850 °C with corresponding tar contents down to 200 mg/Nm^3.[76] As far as tar is concerned, its concentration in the producer gas is also determined by the amount and the kind of the bed materials used in the fluidized-bed gasifier; because bed materials like olivine, dolomite, and magnesite have been widely used as primary catalysts to reduce the tar content of the product gas. With that in mind, the synergic effects of the bed material and the CaFC-C candle were recently investigated in one reactor vessel, which confirmed the tar content could be brought down to 57 mg/Nm^3 from 8600 mg/Nm^3 of dry gas, by gasifying biomass with steam at 820 ºC in a bed of olivine and dolomite with a CaFC-C candle inserted in the freeboard of the fluidized-bed gasifier.[77]

3.4 Recleaning and durability of filter elements

Although extensive efforts have been paid to develop new kinds of CeFC and CaFC filter elements for biomass gasification gas cleanup at higher than 800 ºC, it still remains two main technical issues to be solved, which are the durability of the ceramic filter elements and the high recleaning efficiency of the jet-pulse cleaning system.

3.4.1 The durability of ceramic filter elements

Hot gas filters need to operate reliably for more than 10,000 h, maintaining particulate removal efficiencies and high flow capacity.[79] According to many test results, however, the filters often

break or become fragile after less than 1000 operation hours.[80] In these applications, damages may occur because of the two key reasons caused by the severe thermal or mechanical stresses, and the aggressive gasification environments at high temperatures.

Generally, a too strong cleaning pulse or degradation of the filter due to a repeated cold cleaning pulse are considered to be the most common causes of the thermal or mechanical stresses in ceramic filters.[80-83] She et al.[81] evaluated the thermal shock behavior of silicon carbide (SiC) ceramic filters by the water-quenching technique, which showed that the residual strength of the quenched SiC ceramics decreases gradually with increases in the quenching temperature and specimen thickness, but the fracture strength of the quenched specimens was not affected by the increase of quenching cycles. Kitaoka et al.[84] tested thermal shock fatigue resistance of cordierite and SiC filters under simulated pulse cleaning conditions. It was found that failure of both filters was initiated at the inner sub-surfaces due to the effects of thermal shock, and concluded that thermal shock behavior may vary over different temperature ranges since the loss in fracture strength with increasing temperature. Li et al.[82] used the Reynolds stress model to simulate the flow field during the filtration and back pulse process in a ceramic filter vessel, which showed that velocity distribution was non uniform outside the filters, and the open end of the filter candle was subject to thermal stress during the back pulse process. Shin et al.[83] investigated the effect of operating conditions, thermal shock and pressure shock on the durability of cordierite filters. It was reported that the thermal shock was the most important factor decreasing the durability, and caused the formation of cracks at the filter surface; while the pressure shock didn't deteriorate the durability, and the pulse cleaning per specific pressure drop could improve the durability of ceramic filters, compared with the periodical pulse cleaning. In recent studies, therefore, the pulse

cleaning system is designed in such a way that the heating nitrogen vessel is always used to avoid the risk of thermal shock when the candles are regenerated.[48, 59]

Unlike combustion systems, the gaseous sulfur (H_2S), chlorine (HCl), alkaline (earth) metal species and water vapor, released from the biomass gasifier, can potentially have toxic impacts on the lifetimes of porous ceramic filters. The corrosion behavior of ceramic filter elements in contact with biomass chars or ashes under simulated gasification conditions has been well explored.[47, 79, 85, 86] Müller et al.[85] investigated the corrosion of three ceramic materials (Al_2O_3, mullite and SiC) in contact with straw, wood and miscanthus ashes at 850 ºC for 250h. They concluded that all filter materials could react with potassium and calcium at high temperatures, while Al_2O_3 showed the best corrosion resistance, mullite and SiC showed severe reaction of grains with potassium, which cause in case of mullite formation of cracks in grains. The similar corrosive effects on the binder materials in corundum (Al_2O_3)- and mullite-based filter candle materials were also studied by Schaafhausen et al.,[86] which indicated that the reliable use of ceramic filter candles in biomass gasification was possible, and the best solution with regards to filter material composition was a corundum based filter candle with a silica-free binder phase. They also reported that the employment of SiC filter candles in water vapor containing, alkali-rich gasification environment at high temperature is problematic, owing to the formation of an alkali silicate melt.[79] Recently, the corrosion behavior of SiC filter materials with alumina additives was investigated in presence of steam and coal ashes at 1000 ºC for 96-240h. It turns out that SiC filters prepared with alumina additives showed better corrosion resistance, and water vapor was the perpetrator for strength degradation due to formation of cracks at the bonding necks between SiC particles.[47]

In summary, there has been significant improvement in the performance of ceramic candle filters but with significant scope remaining for further improvement to achieve a better stability

and durability at the ultrahigh temperature over 800 ºC. On the other hand, there are certain fundamental limitations to improvement owing to the intrinsic material properties of ceramic filters and biomass ashes; for instance, microstructural changes and degradation of ceramic candles due to exposure to HT water vapor environments,[87] the infiltration and slag formation actuated by the melting point decrease due to alkali-rich ashes,[85, 86] and the noticeable increase in adhesive force of calcium-rich ashes over 800 ºC, especially under CO_2 containing condition.[88] As a result, it should be also clearly aware that alternate routes involving medium-temperature operating conditions (<500 ºC) and process related changes should be explored to develop more reliable and efficient hot gas cleaning technology.

3.4.2 The regeneration of ceramic filter elements

The recleaning of ceramic filter candles from sticky biomass chars and ashes is still challenging. The conventional jet pulse cleaning systems are always compromised when the filter is dealing with high contents of very fine particles or sticky particles.[55] Moreover, one potential risk of ceramic filter elements is the candle fractures subjected to the high thermal and mechanical stresses during the jet-pulse process. An innovative coupled pressure pulse (CPP) cleaning method was developed to integrate a fail-safe system and to improve the cleaning efficiency.[89] As shown in **Figure 11**, the CPP technology enables higher recleaning intensities at lower recleaning gas pressures compared to the conventional jet-pulse technology. Meanwhile, a CPP filter model was also developed helping to understand filter performance, recleaning efficiencies, and support calculations for scale-up considerations of the hot gas filter.[90] Another promising example is the pulse-less filter technology proposed by Sibanda et al.[91] and Sharma et al.[21], as shown in **Figure 12**, which is also known as the cross-flow filtration.[12] Main advantages of such systems have been proved on the limited growth of cake thickness and the less frequent pulse cleaning of filter

elements via the high annular (or shear) velocity on the filter surfaces generated by an in-line jet ejector. These concept studies had been performed on lab-scale experiments, which show that a thin continuous candle layer during pulse-less filtration can minimize breakthrough of fine PMs and protect the filter surface from erosion and corrosion.[21] However, the scale-up pilot test of these interesting concepts has not been reported so far. Very recently, a novel oxidative filtration concept was practiced by our group on both bench scale[35] and pilot scale[37], in order to decrease the pulse-cleaning frequency and pressure drop by the partial oxidation of carbonaceous PMs in the biomass-derived product gas. By introducing a low concentration of oxygen into the product gas in 300-500 ℃, the carbonaceous PMs could be oxidized to a maximum while maintaining a minimum degradation of product gas at the optimum reaction conditions,[35] and the frequent jet-pulse cleaning is not necessary for the oxidative filtration with a long-term pulse interval of 5-8h.[37]

Figure 11. Schematic comparison of the jet pulse and the coupled pressure pulse.[89] Reproduced from Walch *et. al.*[89]. Copyright 2021 John Wiley and Sons.

Figure 12. Conceptual design of a large-scale pulse less filter unit.[21, 91] Reproduced from Sharma et al.[21] Sibanda *et. al.*[91]. Copyright 2021 Elsevier.

4. CFD NUMERICAL STUDIES IN HOT GAS FILTRATION (请余老师补充 Ref.)

Computational fluid dynamics (CFD) is a powerful tool which uses numerical analysis and data structures to model and analyze hot gas filtration that involve complex multiphase fluid flows with mass, momentum and heat transfers. CFD has been widely used in biomass thermochemical conversion [92]. As an imperative computer aided tool at the particle and reactor scales, CFD is as important as other simulation methods (e.g. molecular modeling life cycle assessment and artificial neural network modelling) at different scales [93]. CFD modeling can be applied to different type of biomass gasifier, for example, circulating fluidized bed gasifier [94] and entrained flow cyclone

gasifier [95]. To simulate unit operation of hot gas filtration, CFD is the most powerful numerical tool available, empowering researchers to achieve economic and environmental targets as they optimize reactor and product's performances. Computer aided design of hot gas filtration with CFD mainly solves the problems faced by experimental methods: (a) high cost of the experiment (b) the flow and concentration field are difficult to perceive and measure, (c) the complexity of the internal structure leads to the optimization problems. CFD effectively broadens the scope of experimental research. For hot gas filtration system, the complex interplay of multi-phase flow, residence time, mixing, and heat transfer with chemical kinetics poses important challenge for practitioners to achieve optimal design and operation conditions. CFD modeler have made intensive efforts to unlock the complex interplay of those factors on hot gas filtration system. **Table 6** summarizes of the main CFD numerical models for different filter vessels.

Table 6 Summary of the main CFD numerical models for filter vessel. **(请余老师精简表达)**

Authors	Device/Software/Method	Predicted results/conclusions	Ref.
Ahmadi and Smith, 2002	Device: Pinon Pine filter vessel, 3.05 m in diameter and 13.4 m long. Software: Fluent, Method: Euler (gas)-Lagrange (solid)	Predicted results: pressure contours, velocity magnitude contours,v vector fields, turbulence kinetic energy contours, particle trajectories. Conclusions: particle size determines the particle deposition rate, and leads to nonuniform cake compositions and thicknesses	96
Casari *et al.* 2020	Device: piping system for 125kg/h (biomass) downdraft gasifier Software: CAD and CFD Method: Euler (gas)-Lagrange (tar)	Predicted results: temperature pattern, deposit height. Conclusions: CFD helps in the prediction of the rate and area of deposit growth. The tar deposition law implemented relies on a compound-dependent condensation temperature.	97
Li *et. al.* 2007	Device: ceramic filter vessel containing three candle filters, filter length of 1.5 m Software: Fluent Method: Euler (gas) with Reynolds stress transport model	Predicted results: velocity vector, radial velocity, gas temperature. Conclusions: Sharp temperature change takes place due to thermal stress. Temperature increase during the pulse cleaning process was observed owing to gas compression.	98
Savuto *et. al.* 2019	Device: ceramic filter diameter of 40/60 mm, and filtration length of 440mm Software: Fluent Method: Euler (gas) with tar	Predicted results: tar conversion and concentration, temperature profile, reaction progress. Conclusions: The temperature drop causes very low conversion of tar. Small injections of O_2 can rise the temperature and increase tar conversion	99

	removal kinetics		
Lee *et. al.* 2015	Device: HTHP dust filter (ceramic candle filter), 60/40mm in diameter, 1500mm in length Software: Fluent, Method: Euler (gas)-Lagrange (solid)	Predicted results: pressure contours, velocity distribution contours. Conclusions: The pressure drop and filtration velocity has a linear relationship. The inertial inlet method has simplified the HTHP process and enhance the efficiency	[100]
Liu *et. al.* 2020	Device: pilot-scale ceramic filter, 40/60mm in diameter, 1520mm in length Software: Fluent, Method: double precision solver, steady and transient simulation	Predicted results: mass decomposition contours, particle velocity magnitude contours, velocity stream contour, pressure drop. Conclusions: High tangential velocity decreases the radial velocity. The results of downdraft flow filtration and updraft flow filtration are compared.	[101]

CFD numerical platform primarily consists of hydrodynamics equations, heat transfer model, turbulent model, tar removal model, porous medium model, reaction kinetics, species transport model and particle tracking model. The development of CFD model for hot gas filtration mainly includes: (1) Tar removal model: Tar (heavy hydrocarbon or polycyclic aromatic hydrocarbon) is mainly decomposed into light hydrocarbons such as CO and hydrogen, and those reactions takes place in the catalyst bed inside the candle. Savuto et. al. used the chemical reactions of tar (benzene, toluene, naphthalene) and methane steam reforming and water gas shift to predict the tar removal process.[99] (2) The porous media model: the porosity of the ceramic filter media is considered by correcting the equations of momentum, energy, and species transport. The momentum equation is also calibrated with an extra source term composed of a viscous loss and an inertial loss. Furthermore, the thermal property such as effective thermal conductivity is calibrated to model the heat transfer through porous medium. (3) Particle removal model: a Lagrangian approach of solid dispersed phase is Newton's second law of motion for particle in a gas phase. The particle equation of motion includes nonlinear drag force and gravity to track particle trajectories in the filter vessel. Ahmadi and Smith used particle tracking model to evaluate the particle deposition patterns and the effect of particle size on particle deposition rate for the candle filters.[96] **Figure 13** illustrates the fundamental structure of the CFD framework, parameter

inputs, typical numerical results and mechanism of optimization. The developed CFD model could evaluate the influence of operation conditions (filtration temperature, composition of hot gas, gas flow rate, oxygen content, etc.) and geometric configuration (design, and catalyst and bed materials) on purification efficiency, and obtain the optimal scenario. The three-dimensional numerical results (velocity field, temperature field, concentration field, pressure field, reaction progress, particle tracking trajectory) will be helpful to analyze the deficiencies of the existing filter units (flow dead zone, low heat and mass transfer zone).

Figure 13. Schematic drawing of CFD framework toward evaluation and optimization of hot gas filtration

5. CHALLENGES AND PERSPECTIVES

The key challenge to commercializing biomass gasification is to generate a clean fuel gas meeting the global emission standards.[7] Hot gas filtration is a reliable and well proven technology already applied in hundreds of installations all around the world,[12] which can remove PMs efficiently down to the submicron range (η>99.5%) and decrease clean gas concentrations down to <1.0 mg/m^3. Although some small hot gas filters containing 1-100 candles had been operated in lab or demonstration plants for gasification and pyrolysis of biomass since 1990s,[12] there is not much large-scale experience of hot gas filtration for biomass gasification.[12, 22] Therefore, to obtain breakthroughs and cost reductions of the high-temperature filtration technology for the biomass-derived product gas, much more attention should be devoted to the following issues.

(1) Characterization and reduction of soot PMs from biomass gasification

Blinding of ceramic filter candles has been reported at temperatures above 600 ^{o}C,[20] which is consider as the formation of soot and high-molecular-weight PAHs on the filter resulting in a dense

and sticky cake layer that could not be effectively removed by conventional pulse cleaning.[38] According to the recent studies on soot formation and reduction techniques,[42] despite catalytic gasification could significantly reduce the soot formation in biomass gasifiers,[102, 103] the regenerative treatment of catalysts is still a great challenge, which is needed to be in-depth studied in future.

In a real biomass gasification system, furthermore, the levels of PMs impurities can vary in a wide range of concentrations, owing to the inherent variability of biomass and the wide variability in biomass implies. It is consequently necessary to develop stable and accurate monitoring devices and methods for PMs measurements in the hot product gas, especially for the nano-sized soot PMs formed during the hot gas filtration (see Section 2). Although a lot of optical diagnostic techniques has been applied in the PMs measurement for flame, few of them are applicable to biomass gasifiers because char and soot PMs significantly attenuate the laser signal.[42] Combination of these approaches is necessary to get a complete view of the nano-sized PMs formation characteristics in the biomass derived gas.

(2) Catalytically activated hot gas filter elements and process optimization

The most important challenges of hot gas filtration are related to the behavior of tars in the biomass-derived producer gas.[37, 38] For process simplification and intensification, therefore, the utilization of catalytic ceramic filters for hot gas filtration is one of the most popular technologies for HT gas cleanup, which provides simultaneous abatement of PMs, tars and some poisonous gases (eg. NH_3, VOCs).[77]

Figure 14. Flow chart of hot gas filtration for the gasification processes operating at different temperatures.[38] Reproduced from Toumi *et. al.*[38]. Copyright 2021 Elsevier.

As illustrated in **Figure 14** A&B, recent researches in this field were mostly focused on increasing the filtration temperature closer to the outlet temperature of the biomass gasifier (800-850 ºC), which could increase the system efficiency and reduce the production costs by avoiding the extra cooling and heating steps upstream and downstream the hot gas filtration.[20, 38] Although there has been significant improvement in the performance of catalytic ceramic filter candles (see section 3.3), a long-term and large-scale test has not been achieved at such high temperatures until now. It should be aware that there are certain fundamental limitations to improvement due to the inherent material properties of ceramic candles over 800 ºC, such as corrosion of ceramic materials, adhesive property of PMs, thermal and mechanical shock of the pulse cleaning, AAEMs volatilization, and so on. Consequently, alternate routes involving filtration temperature and process optimization should be explored to develop more reliable and efficient hot gas catalytic filtration. For example, a hot gas filter with inner cyclone was developed and applied successfully for a 2000 t/d coal gasifier (**Figure 15**), which could remarkably reduce the pressure drop, lengthen the life of ceramic candles, and bring the direct economic benefit about 17.09 million CNY per year.[104] The oxidative filtration of the wood-derived product gas was proved by our group that could simultaneous removal of tars and PMs in the range of 350-550 ºC, but additional long-term tests are needed to optimize operational conditions in the presence of oxygen. Thus, much more research efforts should be focused on the hot gas catalytic filtration to emulate large-scale economics by utilizing innovative process technology and process intensification.

Figure 15. Scheme of the original filter system (A) and the modified system with inner pre-separators (B)[104]. Reproduced from Tao *et. al.*[104], Copyright 1994-2021 China Academic Journal Electronic Publishing House.

(3) Integrated design and development of the small-scale biomass gasification systems

So far there are not much large-scale experiences of biomass gasification systems, because of the economic problems and physical difficulties of long-haul transport transportation.[105] Over the last decade, a large number of commercial fixed-bed biomass gasifiers (70 kWe-3.0 MWe) have been more successful applied in the small-scale combined heat and power facilities in Europe[106] and in China[107], which provide a high thermal efficiency and a competitively priced product. Nowadays, it has been recognized that if biomass gasification could be an economically viable technology, some form of process intensification/integration is inevitable, especially for the heat management of the multi-step gas upgrading processes (**Figure 2**).[20] In order to recover the high-grade waste heat during hot gas filtration, an integrated equipment was developed for the ceramic candle filter by coupling heat exchangers. As shown in **Figure 16**, the integrated equipment could be applied for the combustible gas over 1000 ºC, with the dust removal efficiency over 99.98% and the heat exchange efficiency over 70%;[108] which provided a new way for efficient energy utilization and hot gas filtration. Furthermore, as illustrated in **Figure 14**, the hot gas catalytic filtration over 800 ºC, designed for the large-scale fluidized-bed gasifiers, would be not suitable for the small-scale biomass gasification systems, considering that the product gas temperatures in the outlets of fixed-bed gasifiers are usually below 600 ºC.[106] It should be a subject of great efforts in future to develop the low-cost and stable filter candles loading the highly active catalysts that could achieve satisfactory tar conversion below 600 ºC,[6, 23] which could realize the hot gas catalytic filtration at moderate temperatures (500-600 ºC).

Figure 16. Scheme of integrated equipment for hot gas filtration and waste heat recovery[108]. Reproduced from Xiong *et. al.*[108], Copyright 1994-2021 China Academic Journal Electronic Publishing House.

(4) CFD numerical platforms for the hot gas filtration

As CFD models for the problem of multiphase phase in hot gas filtration are relatively robust. The promising future direction of CFD modeling on hot gas filtration is to improve modelling accuracy of multiphase phenomenon, particularly for the tar removal model and particle removal model. Tar formation in biomass gasification is complex, therefore, the treatment of tar component is important to reveal the detailed chemistry taking places in the catalyst bed inside the candle. Mellin et al.[109] presented a comprehensive chemistry scheme (134 species and 4169 reactions) to describe tar formation using CHEMKIN. The Lagrangian model is a suitable model for particle removal in hot gas filtration, which is to investigate particle motion, where the observer follows an individual particle parcel as it moves through space and time. Xiong et. al.[110] pointed out the improvement of particle flow modelling for biomass thermal conversion. The Lagrangian method requires engineering accuracy but computationally economic sub-models to include the effects of intraparticle transport phenomena. Furthermore, reducing computational cost is another challenge for CFD modelling of hot gas filtration processes as CFD modelling of large-scale industrial systems is computationally expensive, particularly if the Lagrangian method is employed. Nowadays, supercomputing and parallel computing are becoming more accessible for the field of research and development, therefore, computer-aided design of large-scale unit operation using CFD platforms has great potential in the future.

6. CONCLUSIONS

Biomass conversion into the product gas by gasification provides an alternative renewable source of chemicals and fuels to replace the fossil-based fuels, and much effort had been expended to develop commercial biomass gasification processes with support from governments since 1980s.[106] However, the commercial developments of biomass gasification were elusive, owing to the presence of aerosol contaminants or impurities at the concentrations detrimental to most

downstream applications.[6] Nevertheless gas cleanup technologies developed so far have brought a lot of remarkable achievement, the high-quality gas production via gasification is nowadays a major challenge in this field. Since the beginning of the 21st century, hot gas filtration, a reliable and well proven technology applied in many industrial processes,[12] has received more and more attention in the gasification and pyrolysis of biomass, especially for the removal of particulate matters (PMs) and tars.

This paper summarized the main published reviews about hot gas cleanup in the last decade, and reviewed the recent developments of ceramic filter candles applied in biomass gasification, especially for the hot gas catalytic filtration that can simultaneously remove PMs and tars at high temperatures. The different HT gas cleanup systems were briefly introduced and compared in Section 1.

Section 2 divided the PMs from biomass gasifiers into three groups: I-mineral fly ashes, II-tiny char fragments, and III-nanometer-sized aerosol PMs. It is concluded that the quantity of carbonaceous PMs is much higher in the biomass gasification gas due to the sub-stoichiometric conditions. For hot gas filtration, it is firstly important to characterize and control the PMs present in the biomass-derived product gas.

Section 3 summarized the developments of hot gas filtration in biomass gasification by using the ceramic filter candles and catalytically activated ceramic filter candles. The long-term stable hot gas filtration in biomass gasification was shown to be challenging, due to the serious blinding of ceramic filter elements at high temperatures above 600 ºC. In recent years, the best performance of hot gas catalytic filtration has been proven over 800 ºC, concerning pressure drop, water conversion, tar content and ammonia decomposition. But it still remains two main technical issues:

the durability of the ceramic candles and the high frequency jet-pulse cleaning. Better yet, an innovative concept of oxidative pulse-less filtration developed by GIEC, provides a new solution for the simultaneous removal of PMs and tars in 350-500 ºC, which should be more suitable for small-scale biomass gasification system.

Section 4 illustrated the latest research progress of the CFD numerical studies on hot gas filtration. The development of CFD model for hot gas filtration mainly includes tar removal model, porous medium model and particle removal model. The parameter inputs, typical numerical results and the mechanism of optimization are discussed.

Section 5 finally assessed the main challenges to commercializing hot gas filtration in biomass gasification, and the hot gas catalytic filtration and oxidative filtration is regarded as one of the most promising technologies for the economically viable biomass gasification, particular in CFD numerical simulation and integrated design and of the commercial small-scale biomass gasification systems.

FIGURES

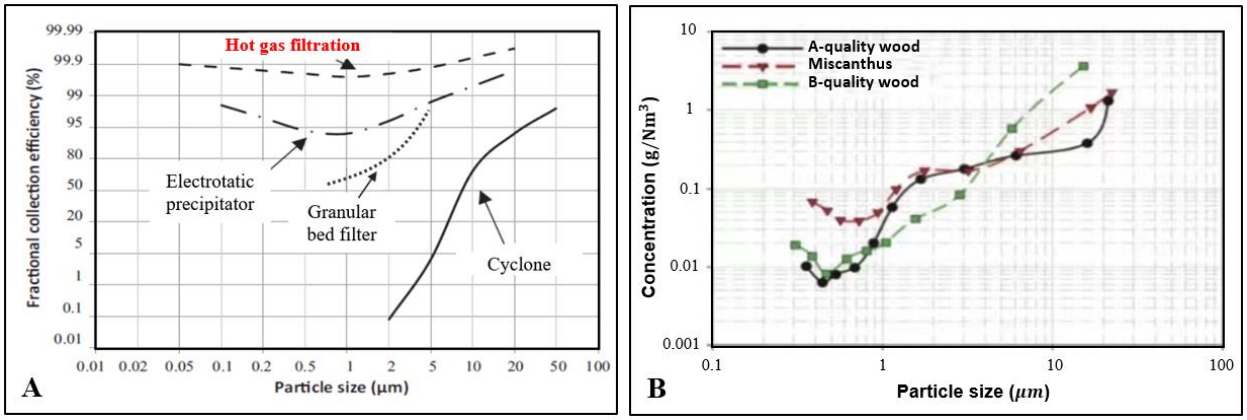


Figure 1.

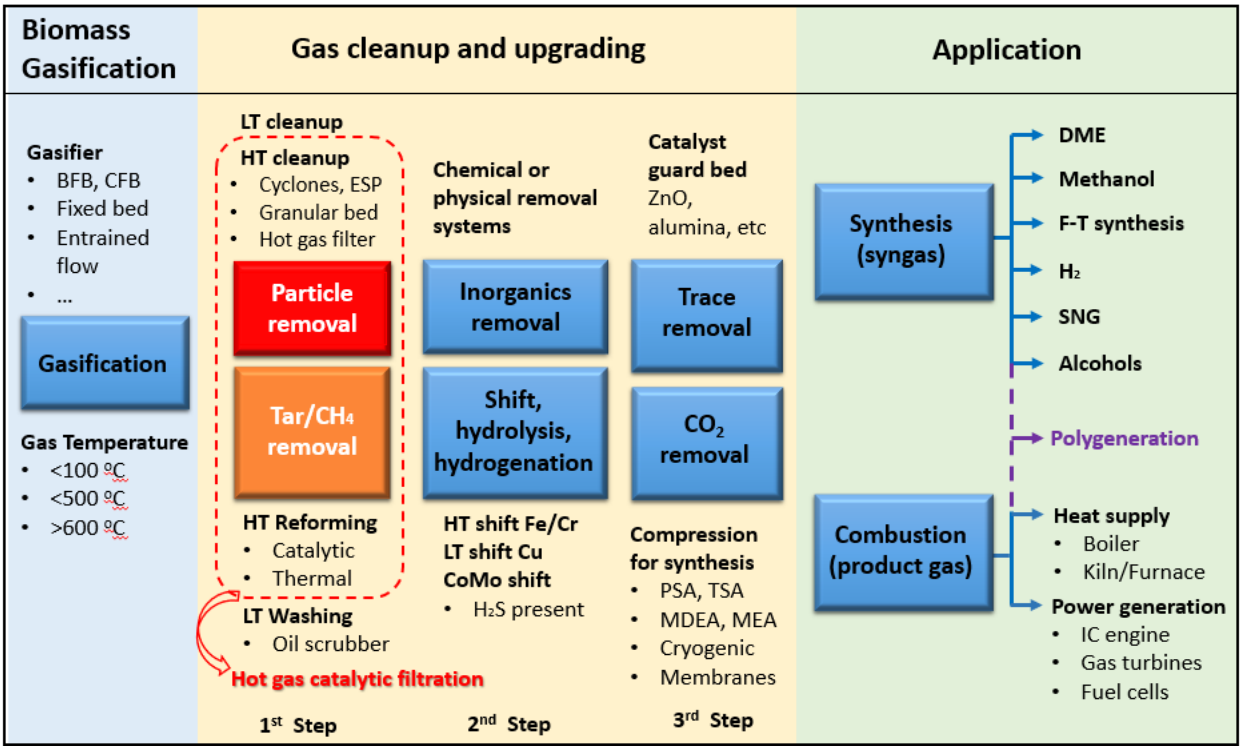
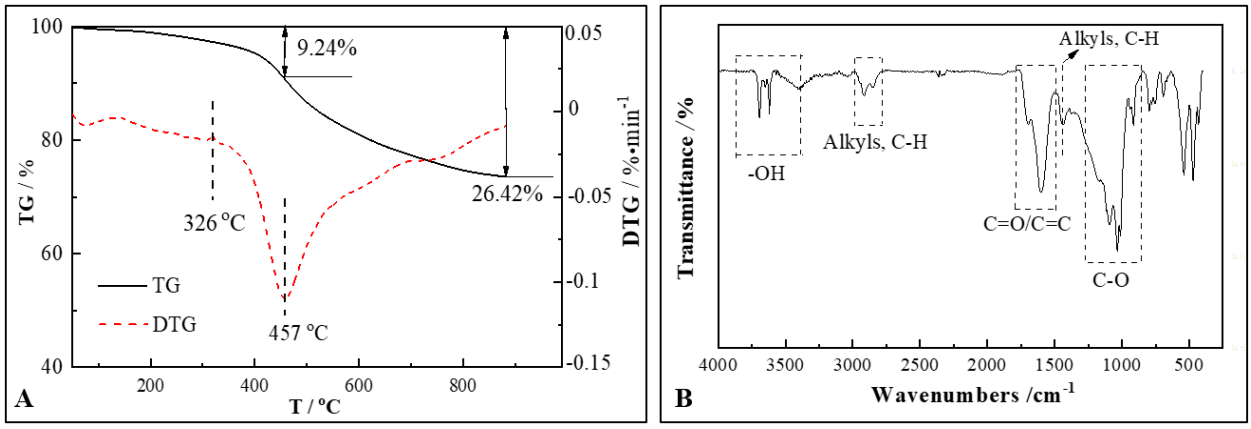


Figure 2.



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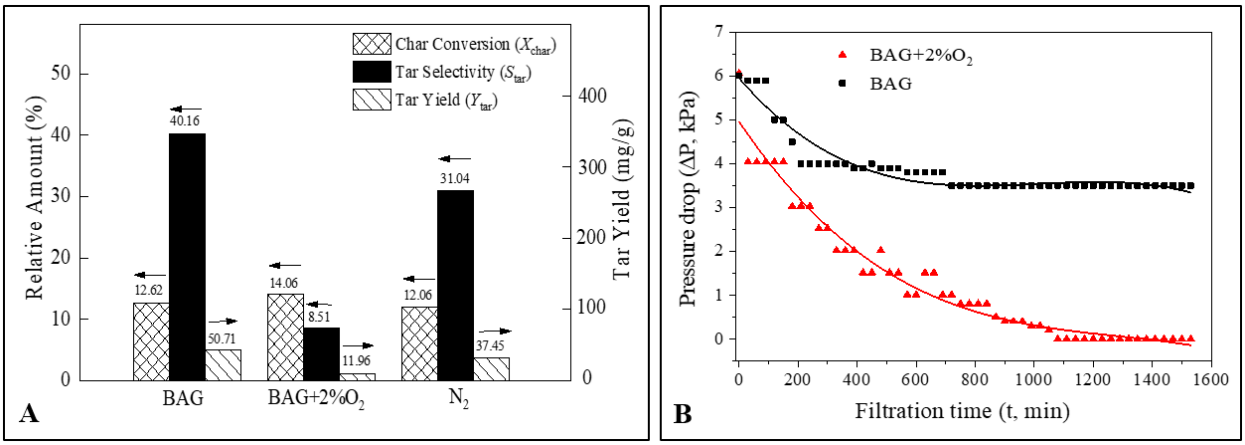

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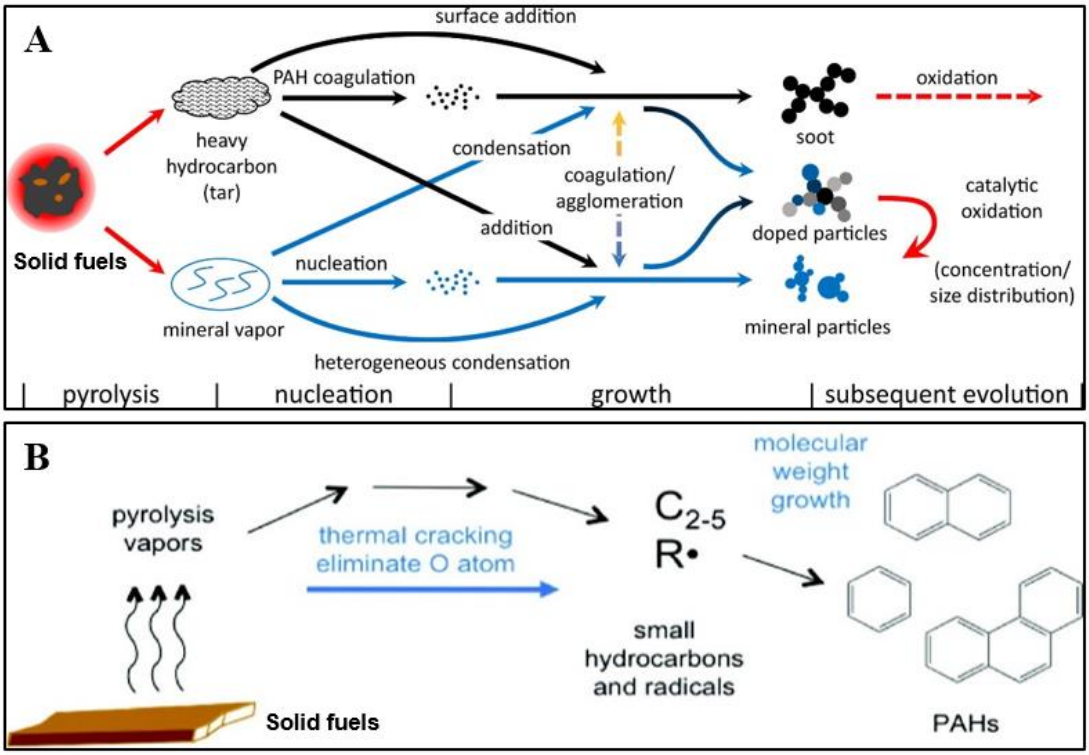

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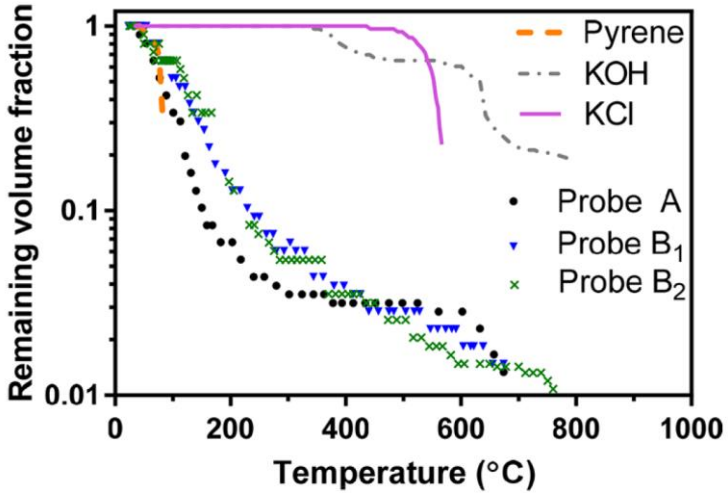

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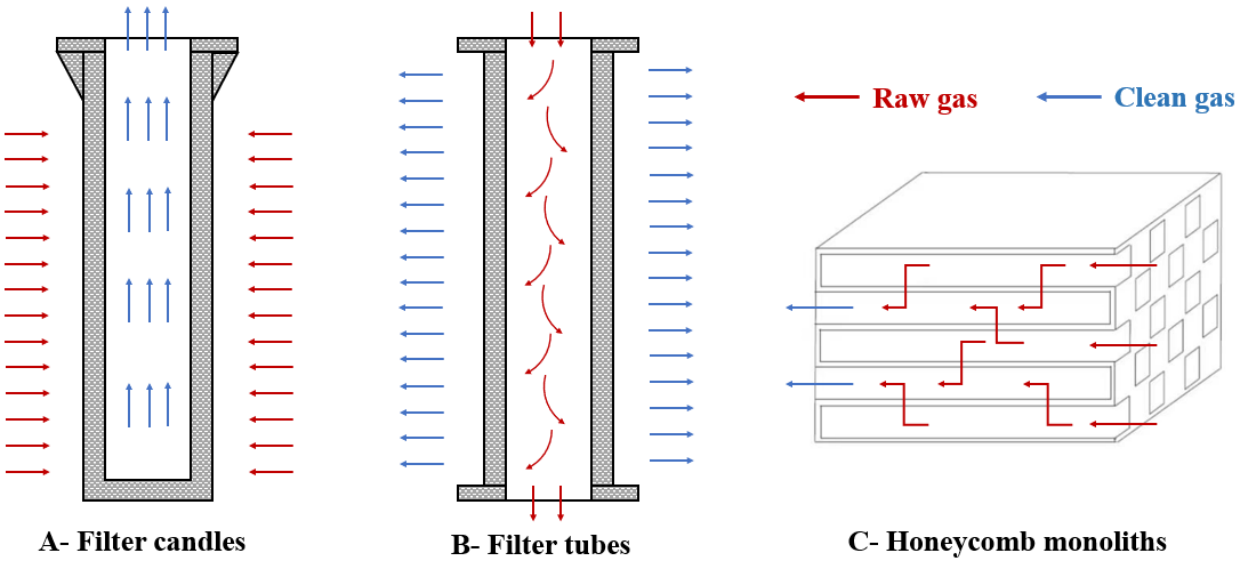

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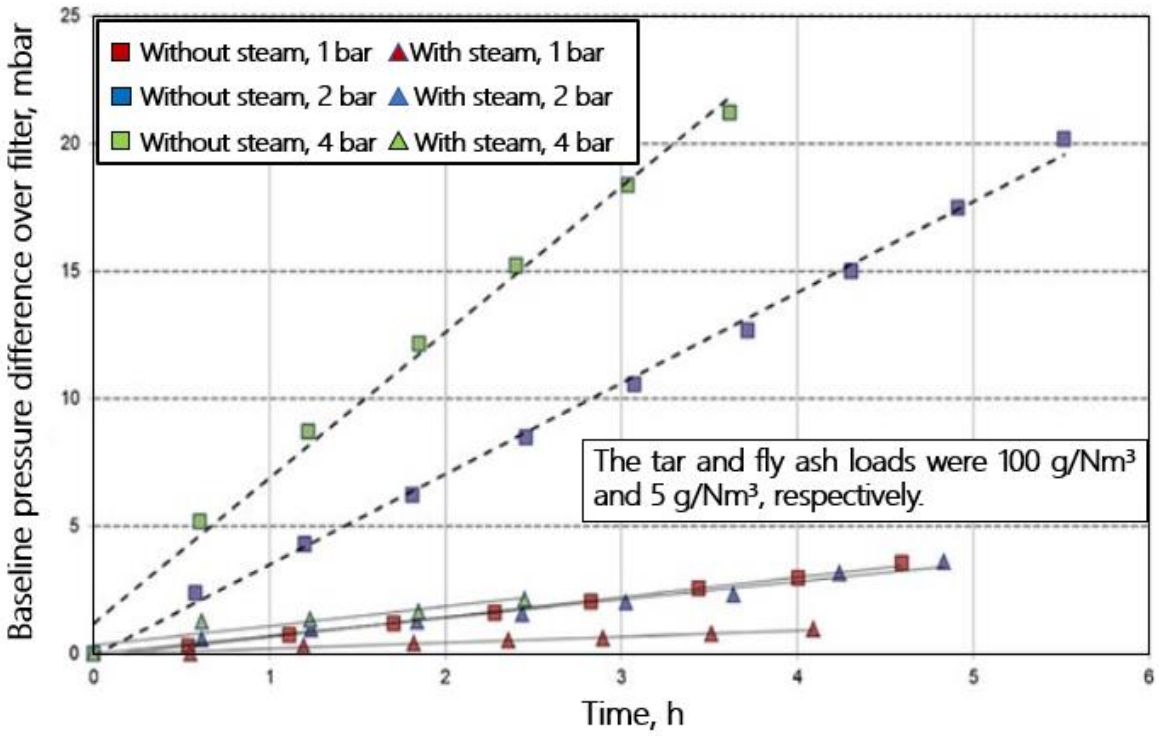

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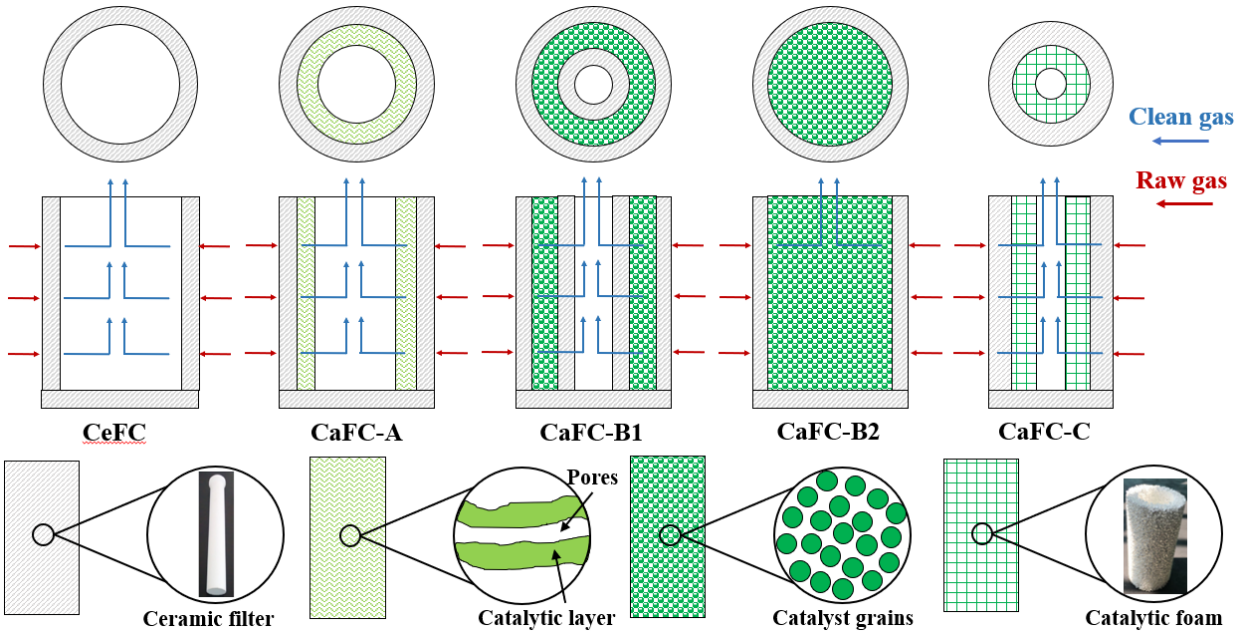

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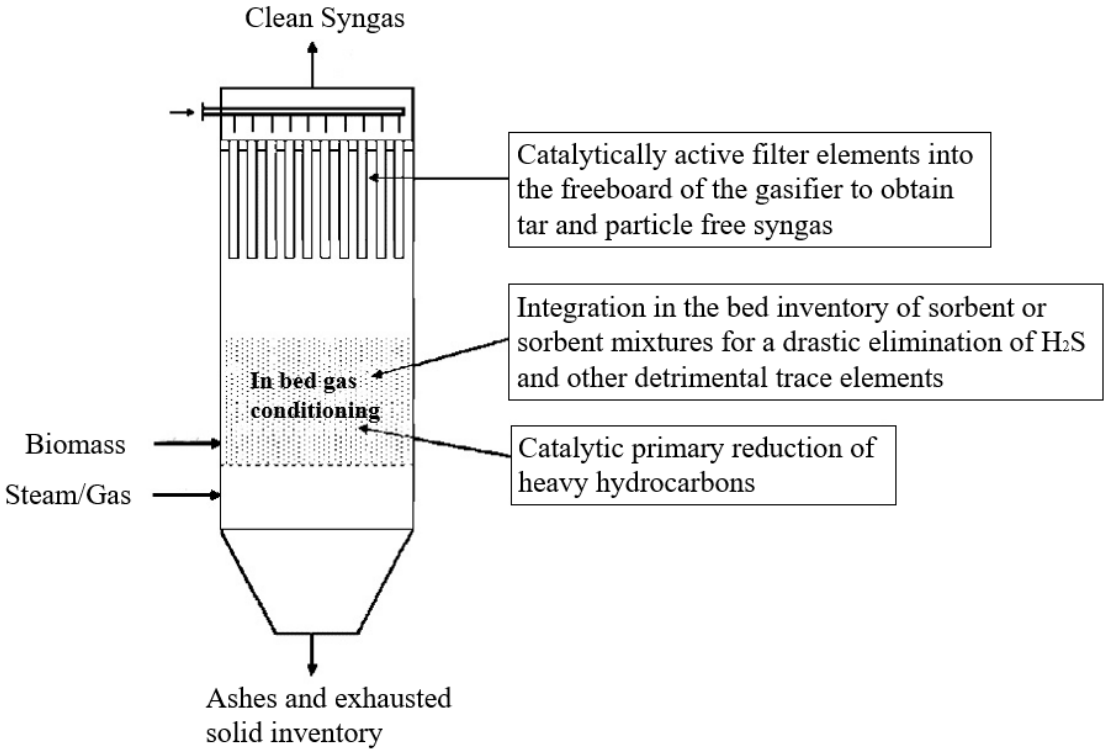

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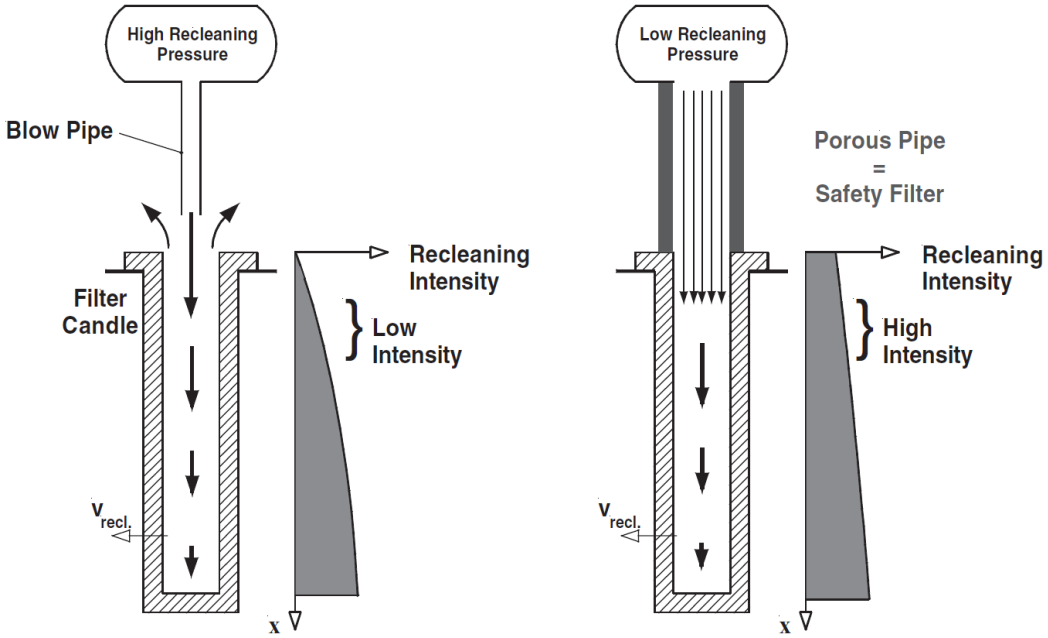

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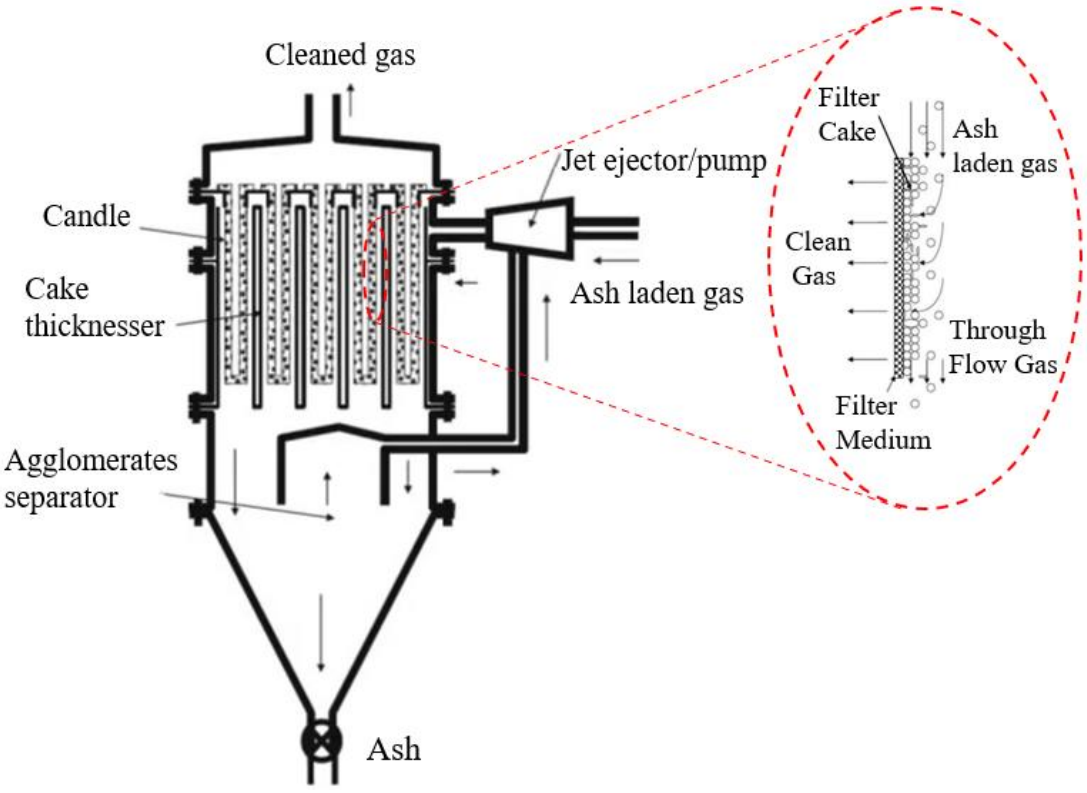
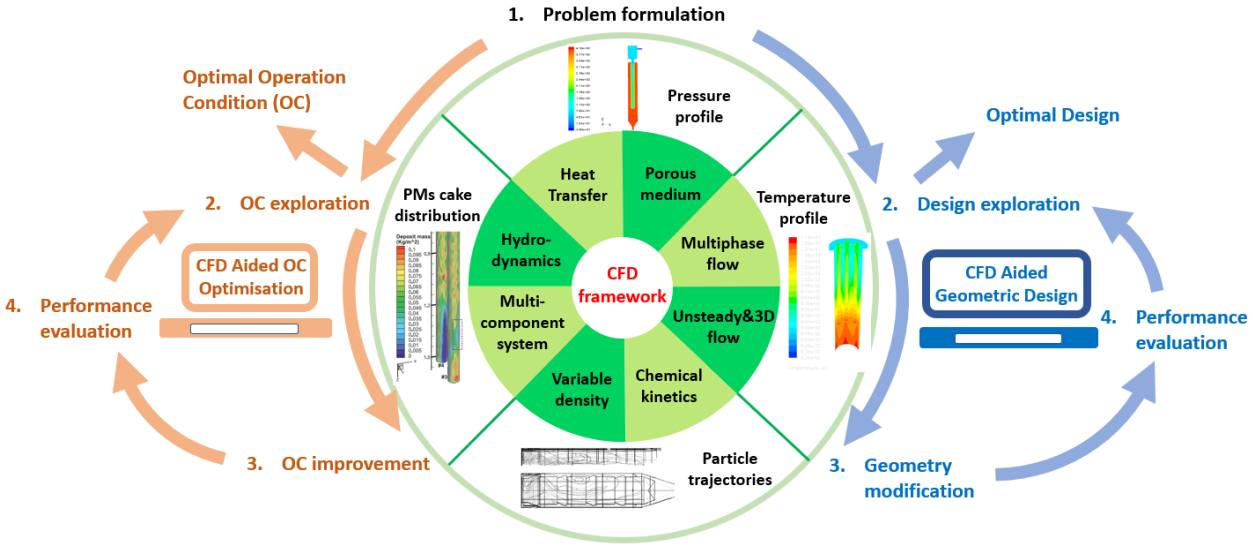


Figure 12.



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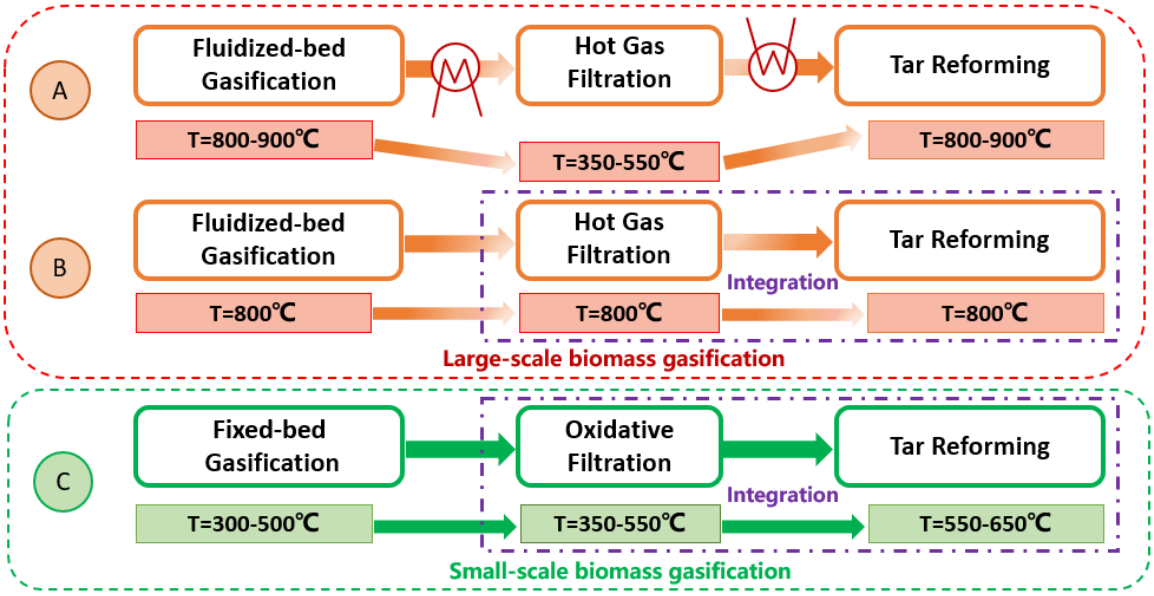

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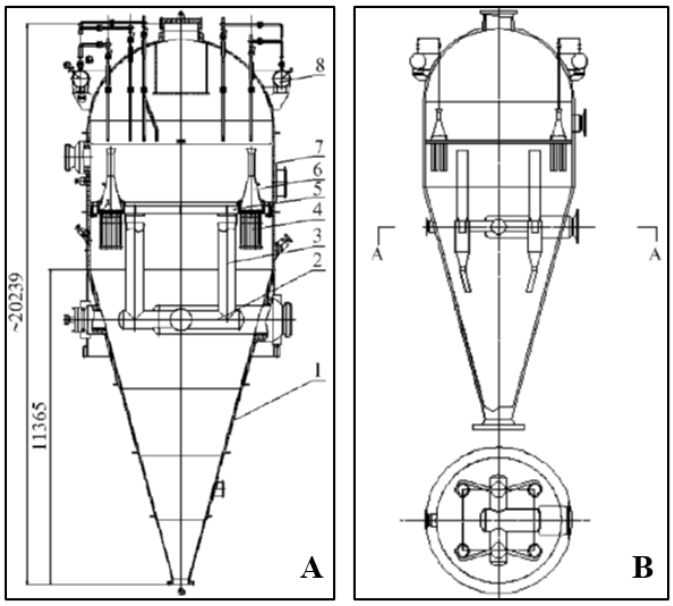

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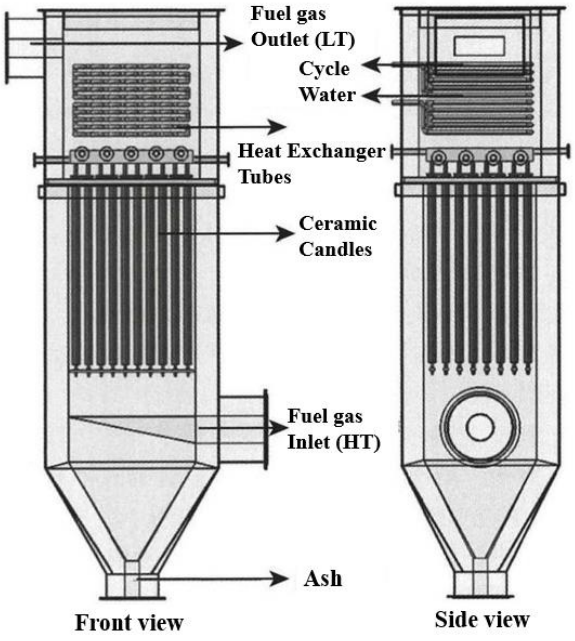

Figure 16.

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Notes

The authors declare no competing financial interest

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ABBREVIATIONS

AAEMs	Alkali and alkaline earth metals
BAG	Biomass air gasification
BFB	Bubbling fluidized bed
BTL	Biomass to liquid
CAD	
CFB	Circulating fluidized bed
CFC	Ceramic filter candle
CFD	Computational fluid dynamics
CHRISGAS	Clean Hydrogen-rich Synthesis Gas
CGC	Cold gas clean-up
CNY	Chinese yuan (Renminbi, RMB)
cLET	Centre for Low Emission Technologies

CSIRO	Commonwealth Scientific and Industrial Research Organization
CPP	Coupled pressure pulse
DFB	Dual fluidized bed
d_{ae}	Aerodynamic diameter
ESP	Electrostatic precipitators
FTIR	Fourier-transform infrared spectroscopy
GBF	Granular bed filters
GIEC	Guangzhou Institute of Energy Conversion
HCCF	Hollow-cylindrical catalytic ceramic foam
HGC	Hot gas cleanup
HGF	Hot gas filtration/filter
HT	High temperature
HTHP	
IGCC	Integrated gasification combined cycle
LT	Low temperature
OC	Operation condition
Org.	Organization
PAHs	Polycyclic aromatic hydrocarbons
PM-2.5	PMs with aerodynamic diameter <2.5 μm,
PM-10	PMs with aerodynamic diameter <10 μm
PMs	Particulate matters;
Ref.	References
SOFCs	Solid oxide fuel cells
UT Delft	Delft University of Technology
VTDMA	Volatility tandem differential mobility analyzer
VTT	Technical Research Centre of Finland

WGC	Warm gas cleanup
ΔP	Pressure drop

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SYNOPSIS (TOC graphic)