

Article

Syngas Production, Clean-Up and Wastewater Management in a Demo-Scale Fixed-Bed Updraft Biomass Gasification Unit

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Abstract: This paper presents the experimental development at demonstration scale of an integrated gasification system fed with wood chips. The unit is based on a fixed-bed, updraft and air-blown gasifier—with a nominal capacity of 5 MW_{th}—equipped with a wet scrubber for syngas clean-up and an integrated chemical and physical wastewater management system. Gasification performance, syngas composition and temperature profile are presented for the optimal operating conditions and with reference to two kinds of biomass used as primary fuels, i.e., stone pine and eucalyptus from local forests (combined heat and power generation from this kind of fuel represents a good opportunity to exploit distributed generation systems that can be part of a new energy paradigm in the framework of the circular economy). The gasification unit is characterised by a high efficiency (about 79–80%) and an operation stability during each test. Particular attention has been paid to the optimisation of an integrated double stage wastewater management system—which includes an oil skimmer and an activated carbon adsorption filter—designed to minimise both liquid residues and water make-up. The possibility to recycle part of the separated oil and used activated carbon to the gasifier has been also evaluated.

Keywords: biomass gasification; demonstration-scale plant; syngas; circular economy; wastewater management; activated carbon adsorption

1. Introduction

The increasing attention towards climate change and greenhouse gas emissions makes the exploitation of renewable energy sources one of the key pathways for sustainable power generation. It is expected to involve a significant reduction (some 8 Gt/yr by 2050, with a share of 32% among the other low carbon approaches) of CO₂ emissions in the power generation sector, according to the most recent assessment by the International Energy Agency [1]. The same target has been formally assumed by the European Union (EU) with the publication, in December 2018, of the revised “renewable energy directive” (2018/2001/EU), which aims at keeping the EU a global leader in renewable energy and to meet the commitments under the Paris Agreement [2,3]. However, the diffusion of intermittent sources (i.e., wind and solar) makes grid regulation increasingly challenging since ever changing electrical loads must be balanced with ever changing, non-programmable generation [4]. On the contrary, bioenergy can be considered a key option to mitigate greenhouse gas emissions, replace

fossil fuels, and—considering its programmable exploitation—ensure a more secure and sustainable energy system [5]. Additionally, the use of waste biomass (e.g., agro-industrial, municipal and forestry residues) [6] is even more interesting since it allows production of almost CO₂-free energy as an alternative to landfill or inefficient biological processes.

Biomass gasification is an appealing thermochemical conversion process that allows the production of a synthesis gas (syngas) that can be used for power generation in internal combustion engines [7,8] or for other applications, such as liquid fuels production [9].

Gasification technologies for small- and medium-scale combined heat and power (CHP) generation from waste biomass have been significantly developed in the last decades [10]. Most of the attention is focused on fixed-bed downdraft processes, typically available for a capacity between 200 and 700 kW_{th} [11–13], but few studies also consider bubbling [14,15] or circulating fluidised-bed [16] gasification processes, sometimes promoted with specific catalysts [17] or integrated with hydrothermal carbonisation to treat high-moisture biomass [18]. With respect to these technologies, fixed-bed updraft gasification allows a better conversion efficiency (thanks to the countercurrent heat exchanges) [4] and it is typically characterised by simple construction, easy operation, fuel flexibility in terms of type (biomass, coal, wastes, etc.), particle size (5 to 100 mm) and moisture content (up to 60%) [19], but also involves a relatively high production of pyrolysis liquids (i.e., oils and tar) [4]. Such a technology is feasible for applications in the order of a few thermal megawatts.

In fixed-bed updraft gasification reactors, the solid primary fuel is loaded from the top of the reactor and supported by a metallic grate by which the gasification agents (air or oxygen and possibly steam, depending on the specific process) are injected from the bottom, allowing a countercurrent. As fuel flows downwards, it is heated by the hot raw gas that moves upwards, coming from the gasification and combustion zones [20,21], whereas the gasification agents are preheated by cooling the bottom ash [20]. The units developed for small- and medium-scale applications typically operate at atmospheric pressure using air (instead of oxygen, used for the industrial-scale processes) and possibly steam as gasification agents. A number of theoretical studies on these kinds of processes are currently available in the scientific literature, mainly focused on the development of thermodynamic models (in particular by minimising Gibbs free energy) [22,23] or on computational fluid dynamics (CFD) process simulation [24,25], in some cases with the experimental model validation in pilot units [26,27]. Several studies are available on pilot-scale experimental development of the process for waste biomass gasification for power generation and biochar [7,28]. To the best of the authors' knowledge, just a few studies are specifically focused on air-blown fixed-bed updraft technology demonstration at commercial scale. Only Lei et al. [29] recently published an experimental research based on a batch feeding updraft gasifier designed to treat 2 tons per day of rural solid waste in China.

A major issue in biomass gasification plants is the managing of tar, which is the complex mixture of condensable hydrocarbons generated during gasification of such volatile-rich matter as biomasses [30]. Tar is constituted by single-ring to 5-ring aromatic compounds plus other oxygen-containing hydrocarbons and complex polyaromatic hydrocarbons (PAHs), phenolic compounds (cresols, xylenols, etc.), and monocyclic aromatic hydrocarbons such as benzene, toluene and xylene [31,32]. Tar formation is still considered to be the main technological barrier of this kind of technology [22], since it causes several environmental and industrial problems: heavy tars may condense on cooler surfaces downstream leading to blockage of filters and fuel lines [31], it can block valves and clog fuel injectors of engines [33] and, if released in the environment, it can have harmful effects, since its compounds are toxic and potentially carcinogens [34,35]. Tar issues are particularly relevant for updraft gasifiers—with a loading in raw syngas of about 50 g/Nm³ [36]—since the countercurrent heat exchanges involve relatively low temperatures in the upper part of the fuel bed, promoting tar formation and limiting its decomposition into lighter compounds. Therefore, its management is of great relevance to make gasification a feasible option for power generation from biomass. Tar management can be basically achieved through two strategies: reduction of tar formation inside the gasifier, with the so-called primary methods, or separation after the gasification process, with secondary methods [31]. Primary

methods include optimal design of the gasifier, the optimisation of process parameters, and possibly (depending on the specific technology) the use of suitable additives or catalysts. Secondary methods comprise thermal or catalytic cracking, or mechanical methods such as the use of cyclones and electrostatic filters [37], as well as the use of wet scrubbers. The latter, in particular, is an effective and reliable process for tar removal from syngas. However, the process may generate a high amount of wastewater. The toxicity of many compounds of tar makes the wastewater treatment with biological processes quite difficult. Therefore, chemical and physical treatment must be used, such as advanced oxidation processes, or precipitation with Fe or Al salts [23]. Adsorption on activated carbon is also an effective technology to remove tar from wastewater, although regeneration or disposal of the exhaust sorbent is a drawback of the process [38].

This paper aims to establish the baseline performance of a 5 MW_{th} (1.3 m internal diameter) demonstration-scale updraft gasifier, operating since 2014 at the Sotacarbo Research Centre in Sardinia, Italy [39], and tested for some 1500 h with different kinds of coal and biomass. In particular, the experimental results here reported are focused on the gasification of two kinds of local waste biomass, with the aim to assess operating conditions, syngas composition and properties and the whole plant performance. In addition, the syngas cleaning and wastewater treatment process performance is also evaluated, on the basis of a novel configuration of a tar management system optimised to minimise water consumption and sludge disposal and recirculate part of the separated tar and exhaust activated carbon to the gasification unit (thus improving the efficiency of the whole project).

2. Materials and Method

The analysis presented here summarises the key results of an experimental campaign carried out in order to improve the knowledge of the gasifier's operation in different operating phases (i.e., start-up, steady-state and shut down) and to evaluate the performance of the integration with tar management and wastewater treatment. Moreover, the final scope is to provide useful data so as to improve the system efficiency to make electricity generation suitable by mean of an internal combustion engine fed by clean syngas.

2.1. The Sotacarbo Demonstration-Scale Gasification Unit

The nominal 5 MW_{th} demonstration-scale plant (Figure 1) of the Sotacarbo platform is mainly composed of a gasification section, a wet scrubber for syngas clean-up, a wastewater treatment system and a flare for final syngas combustion, according to the simplified scheme shown in Figure 2.



Figure 1. The Sotacarbo demonstration-scale unit.

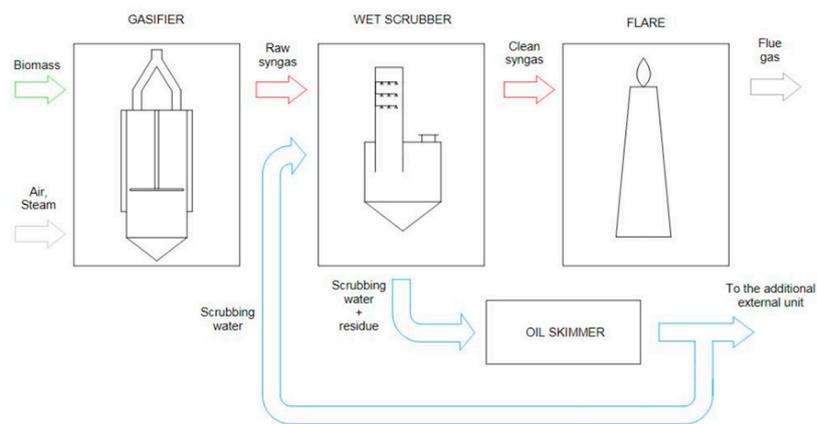


Figure 2. Simplified scheme of the demonstration-scale unit.

2.1.1. Gasification Reactor

The key section of the experimental unit is the gasification reactor. It is based on the historical Wellman-Galusha fixed-bed updraft technology, which has a commercial history in municipal and industrial applications dating back at least 80 years [40]. It was further developed in the United States by Hamilton Maurer International, Inc. (HMI, Houston, TX, USA) for power generation from coal during more than 10,000 h between 1981 and 1985 [41]. It was converted for biomass (wood chips) gasification by Ansaldo Energia (former Ansaldo Ricerche), tested in a 1.3 m internal diameter gasifier, and put in operation between 1999 and 2001 [4].

Biomass is loaded by means of an automatic redler conveyor and it is introduced in the top of the reactor through four different injection points. The fuel bed is supported by an eccentric grate that allows continuous ash discharge. The grate design (with different coplanar plates) has been optimised to allow the reactor to operate in continuous mode, avoiding blockages of the ash extraction system. It is equipped with a robust driving system specifically designed to optimise its operation. The gasification agents (air and/or steam) are injected from the bottom of the reactor through the grate at about atmospheric pressure (with a small overpressure just to win the pressure drops through the whole process). As fuel flows downwards, it is heated by the hot raw gas that moves upwards, coming from the grate [20], so that the following processes take place: fuel drying, devolatilisation, pyrolysis, gasification and partial combustion [38].

The gasifier is also equipped with a cooled stirrer composed of a vertical shaft, with internal water recirculation, which stirs and uniformes the fuel bed in order to maximise the process performance. This device can translate vertically and perform both clockwise and counterclockwise rotation. Moreover, the reactor wall is constantly cooled thanks to a water jacket that allows a slight thermal dissipation. Steam generated in the jacket during plant operations, reaches by natural circulation the upper steam drum, downstream connected to a forced air-cooled condenser.

Due to the experimental nature of the unit, characterised by frequent start-up and shut-down phases, the ignition phase is performed by six infrared ceramic irradiators (instead of the conventional burners) placed above the bottom of the reactor, which is also equipped with several thermo-couples located on different levels, in order to monitor the internal temperature profile.

2.1.2. Wet Scrubber

Gas cleaning is an essential component of any biomass gasification plant to meet the specifications of the syngas end user. Internal combustion engines for power generation require limits of about 30 mg/Nm³ for particulate and 100 mg/Nm³ for tar [36,42]. Conventionally, gas cleaning is performed by means of different systems, such as wet or wet-dry scrubbing (the former being the most common in this kind of applications) or hot gas conditioning.

In this specific case, raw syngas from the gasification unit is sent to a wet scrubbing system (Figure 3) by means of an insulated pipe (to reduce tar condensation), installed with a slight slope to allow condensates to move to the scrubber by gravity. The scrubber operates in co-current mode, in order to remove tar and dust and to prevent backfire. The scrubbing tower is provided with a dehydration section for the separation of the micro drops of water dragged in the purified syngas. Heavy non soluble tar (C20–C40 and more), together with inert matter and un-reacted dust separated from syngas, is collected in the conic bottom of the scrubber and removed through a screw pump [43]. On the other hand, water with light tar (C10–C20) moves to the oil skimmer section.

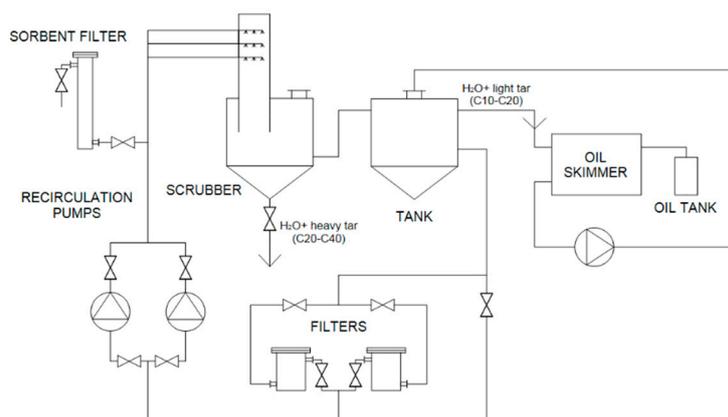


Figure 3. Water recirculation system.

2.1.3. Wastewater Treatment

The wastewater leaving the scrubber (and temporarily stored in a 6 m³ tank that allows a thermal flywheel effect) is loaded with tar, dust and various contaminants and must be treated before being recirculated. As mentioned, the integrated wastewater treatment system (schematically represented in Figure 3) has been specifically designed to minimise freshwater consumption and sludge disposal and to recirculate part of the separated tar and exhaust activated carbon to the gasification unit (thus recovering its energy).

Firstly, wastewater from the scrubber is pre-treated by means of an oil skimmer (specifically designed and assembled using only commercial components to contain capital cost, in view of commercial applications), with the aim to continuously separate insoluble oils and tar from the surface of the liquid phase (thanks to the low specific weight of the compounds and their affinity with the materials) and potentially to recirculate a part of the pre-treated water directly to the scrubbing system. About 8 m³/h of wastewater is collected from the tank, filtered and recirculated to spray nozzles of the wet scrubber through proper pumps. On the other hand, about 0.4 m³/h of light tar mixed with water (about 5% of the scrubber wastewater recirculation flow) is collected from the free surface of the tank and delivered to the oil skimmer. An additional external unit provides further treatment in a proper chemical–physical unit, equipped with different sections for chemical reagent dosing, destabilisation, flocculation, sedimentation and sludge purging. Treated water is then collected in an external tank before disposal.

Among the available technologies (i.e., wet oxidation, adsorption on active carbon and/or carbon-rich ashes from gasification, as well as chemical, physical and biological treatment [43], the latter not feasible due to the toxicity of the components to be treated [23]), a chemical–physical treatment unit has been selected.

Physical treatment as ultraviolet light-induced wet oxidation or adsorption on various coke sorbents was suggested to treat this type of wastewater. Moreover, chemical precipitation using various salts of Fe and Al can promote the formation of flocs, then reducing the concentration of colloidal and particulate matter in the wastewater [23]. Finally, adsorption on activated carbon is demonstrated

to be an effective technology to remove organic pollutants from wastewaters. The main benefits of using activated carbon are the large surface area, the economic viability and the easy operational procedures [44], whereas the disadvantages are a difficult and expensive regeneration process that can limit its application, considering also that the spent carbon generally involves a serious disposal problem [38].

In this specific case, the different commercial diluted solutions dosed in this unit are based on the following chemical reagents: hydrochloric acid for pH neutralisation, ferric chloride for destabilisation and flocculation [45], calcium hydroxide as a coagulant, polyelectrolytes as a thickener. The reactions take place in different phases, starting with destabilisation and coagulation and ending with flocculation, and are carried out in two separate tanks, equipped with mechanical stirrers set at different speeds (the first fast and the second slow). The separation between purified water and sludge takes place in a tank equipped with lamellar septa and a single-screw mud pump for sludge extraction.

2.1.4. Auxiliaries

The unit is also equipped with different auxiliary systems to allow the experimental runs: fuel temporary storage and charging system, dust extraction and filtration system, process air and steam production and adduction systems, and LPG storage and adduction system (to support syngas combustion by the flare). For safety reasons, such pipeline inertisation or emergency shutdown of the plant, require a nitrogen storage and adduction system; it comprises a vertical cryogenic vessel for liquid nitrogen and a system of vaporisers.

The plant includes industrial data acquisition and control equipment in order to continuously monitor the main process parameters, with particular reference to temperatures, pressures, and flow rates of the gasification agents. Particular attention is dedicated to monitor the internal temperature profiles of the gasifier, crucial for correct operations and for reliable data processing.

Composition of raw syngas from the gasifier, clean syngas downstream of the scrubber, exhausts from flare and vents are monitored by four different gas sampling lines. A real time multi module industrial analysis system and a portable micro gas chromatograph (GC) are dedicated to continuously monitor the syngas composition. The first provides a quick online measure of H₂, CO, CO₂, CH₄, H₂S and O₂. It consists of several gas analysers, which provide: H₂ concentration measured by thermal conductivity by means of a CALDOS 25 module (within a range between 0% and 100% by volume); CO, CO₂ and CH₄ concentrations measured by an infrared URAS26 module (within the following ranges: 0–30% for CO, 0–25% for CO₂, and 0–5% for CH₄, all by volume); H₂S concentration measured by an ultraviolet Limas 11 module (between 0% and 2% by volume); and O₂ concentration measured by a paramagnetic Magnos 206 module (between 0% and 25% by volume). Moreover, a micro GC (Agilent 2000, Santa Clara, CA, USA) analyses H₂, CO, CO₂, CH₄, H₂S, and O₂ in the syngas, as well as N₂, COS, C₂H₆ and C₃H₈. The plant is also equipped with a sampling line that allows measuring the amount of tar at the exit of the gasifier. To this aim, syngas is collected downwards of the reactor and sent—through a hot line (200 °C)—to three ice cooled traps to allows tar condensation and sampling. Samples are then analysed in the laboratory, typically in terms of weight, calorific value and composition.

2.2. Primary Fuels

The experimental campaign presented here has been carried out with stone pine (*Pinus pinea*) and eucalyptus (*Eucalyptus camaldulensis*) wood chips from local wood management. For each biomass, two different samples have been delivered and analysed, with slight differences mainly in terms of moisture content.

Table 1 shows, for each sample, proximate and ultimate analysis, as well as lower heating value (LHV) and bulk density. The analyses have been carried out in the Sotacarbo laboratories according to the international standards. In particular, proximate analysis has been performed by a LECO TGA-701 thermogravimetric analyser; ultimate analysis has been carried out on a LECO Truspec CHN/S analyser;

finally, the energy content of the sample (higher heating value) has been measured using an adiabatic oxygen bomb calorimeter by a LECO AC-500 calorimeter, according to the ISO 1928:1995 standard, and then converted into LHV considering the moisture content.

Table 1. Characterisation of fuel samples, as received.

Parameter	Stone Pine (1)	Stone Pine (2)	Eucalyptus (1)	Eucalyptus (2)	Standard
Proximate analysis (% by weight)					
Fixed carbon	22.33	21.09	15.37	15.67	By difference
Volatiles	61.87	66.91	56.09	57.41	ASTM D 5142-04
Moisture	11.93	9.25	26.86	25.76	ASTM D 5142-04
Ash	2.87	2.75	1.68	1.16	ASTM D 5142-04
Ultimate analysis (% by weight)					
Total carbon	50.72	50.88	49.62	49.75	ASTM D 5373-02
Hydrogen	6.32	6.71	6.17	6.54	ASTM D 5373-02
Nitrogen	0.63	0.50	0.48	0.35	ASTM D 5373-02
Oxygen	27.53	29.91	15.19	16.54	By difference
Moisture	11.93	9.25	26.86	25.76	ASTM D 5142-04
Ash	2.87	2.75	1.68	1.16	ASTM D 5142-04
Other properties					
LHV (MJ/kg)	16.07	16.08	13.12	13.34	
Bulk density (kg/dm ³)	0.20	0.22	0.25	0.26	

2.3. Experimental Procedures

Basically, each run is organised in four different phases: plant preparation, start-up (which typically lasts for about four hours), steady-state operation and plant shut-down (that requires about five hours). Before each run, all the auxiliary systems are started up and their functionality is verified, in order to assure the right operation of the whole plant. In parallel, some instruments are calibrated to avoid significant errors in the measurement of the key parameters. As soon as the auxiliary operation is verified, the start-up process can begin: primary fuel is heated by means of the six ceramic infrared irradiators, until fuel locally reaches the temperature of 750–800 °C (typically this phase takes some 45 min, with 15 extra min to complete fuel heating); then air is injected through the ceramic irradiators to start the fuel ignition; in this phase, gas is vented in the atmosphere. When the ignition of the fuel bed is confirmed (it needs just few additional minutes), the ceramic irradiators are turned off, the air flow through the irradiators is stopped, a sub-stoichiometric air flow is injected through the grate and gas is sent to the flare and burnt; this can be consider the beginning of the operation phase [39]. In general, during the operation phase, a steady-state regime is kept for several hours (through a continuous feeding of the primary fuel selected for the specific run) or pre-determined operating procedures are followed, on the basis of the specific aims of each run. The shut-down phase of the plant begins at the end of the experimental test, interrupting the fuel loading and decreasing air flow to cool the reactor down.

The runs are operated with a primary fuel (as-received) characterised by a particle size between 10 and 50 mm (with an amount of fines within 5% by weight), maintaining the main operating parameters constant (in particular air flow) and regulating fuel loading to achieve an almost constant fuel bed level (at about 1800 mm), with a maximum fluctuation of ± 30 mm.

During the experimental tests, wastewater samples are collected from different sections of the circuit with a pre-determined frequency: from the bottom of the scrubber tank (every 3 h), from a floating skimmer (1 h), in the decantation tank (1 h), from the rotating oil skimmer (3 h), and downstream of the activated carbon filter (5 to 10 min). Samples are then analysed in the Sotacarbo labs by measuring

pH (by means of a portable pH-meter Medidor PH BASIC 20 and a Yokogawa online pH-meter) and suspended and dissolved solids.

3. Results and Discussion

Gasification results that were collected, for each fuel sample, during about 80 h of continuous steady-state operation have been processed in order to determine plant operating conditions and performance. In parallel, wastewater has been collected and analysed. With respect to the previously published data [39], referred to a preliminary biomass gasification experimental campaign, the results here reported come from a specific optimisation of the operating procedures for this kind of fuel.

3.1. Gasification Performance

The experimental tests have been arranged regulating the process operating parameters (mainly fuel and air injection) to optimise syngas composition and maximise its lower heating value as well as process stability. Table 2 reports a summary of the key operating parameters (including the equivalence ratio, ER, defined as the ratio between the oxygen actually injected as gasification agent and the oxygen theoretically required for the stoichiometric fuel combustion) and the subsequent syngas composition, intended as the average value during the steady-state operation. In particular, the key index assumed as a measure of the global performance of the gasification process is the so-called cold gas efficiency (η_{CG}), conventionally defined on the basis of the first law of thermodynamics as the ratio between the chemical energy of raw syngas (calculated as the product of syngas mass flow and its lower heating value) and the chemical energy of primary fuel:

$$\eta_{CG} = \frac{m_{syn} \cdot LHV_{syn}}{m_{fuel} \cdot LHV_{fuel}} \quad (1)$$

where m is the mass flow rate (in kg/s) and LHV is the lower heating value (in MJ/kg) of syngas and fuel, indicated by the subscripts syn and $fuel$, respectively.

First of all, it can be noticed that raw syngas from eucalyptus chips gasification is characterised by a slightly higher CO_2 concentration and a lower CO concentration than those measured during the stone pine gasification tests. This is one of the effects of the higher moisture content of eucalyptus biomass, which promotes the water–gas shift reaction. In parallel, it can be noticed that eucalyptus runs have been performed with a lower specific air flow rate (about 1.9 kg of air per kilogram of biomass, compared with 2.2 kg/kg for stone pine, both previously determined as the optimum values for the considered fuels), which leads to a slightly higher heating value. Cold gas efficiency is in the order of 79–80% for all the runs. Moreover, with respect to eucalyptus gasification, the higher equivalence ratio used for stone pine gasification led to higher temperatures in the combustion zone and in the freeboard with, as a consequence, a significantly lower tar production.

Figure 4 shows the trend with time of H_2 , CO_2 , CO and CH_4 concentrations in the syngas from one of the runs with stone pine chips (similar results have been obtained during the other three runs). Syngas composition is quite constant during the whole steady-state operation. It can be also noticed that the gasification conditions have been reached after about 3 h from the beginning of the start-up procedure, with an increasing of CO and H_2 concentration, whereas the steady state is reached after about 10 h with the stabilisation of syngas composition.

One of the key aspects for the evaluation of a fixed-bed updraft gasifier operation is represented by the temperature profile into the reactor, shown in Figure 5—with reference to wall temperatures—for the same pine-fueled run. Excluding the start-up phase, when the whole reactor is at almost ambient temperature, the profile shows good stability during the whole run. The trend reveals the characteristic tendency of fixed bed updraft gasifiers with the maximum temperature in the combustion zone [39]. And, thanks to the presence of the water jacket, the inner temperature in the freeboard (above about 1000 mm from the bottom of the reactor) is typically lower than 200 °C and around 100 °C at the outlet

of the gasifier. This involves the condensation of the heaviest tar compounds, which remain trapped in the reactor in a sort of natural recirculation.

Table 2. Operating parameters and syngas properties.

Parameter	Stone Pine (1)	Stone Pine (2)	Eucalyptus (1)	Eucalyptus (2)
Operating parameters				
Fuel loading (kg/h)	280	275	345	340
Air flow (kg/h)	630	620	655	650
Equivalence ratio (%)	33.02	32.85	26.48	26.36
Fuel bed level (mm)	1800	1800	1800	1800
Maximum temperature (°C)	870	880	830	824
Freeboard temperature (°C)	380	376	300	310
Syngas composition (molar fraction, dry basis)				
CO	0.29	0.29	0.27	0.28
CO ₂	0.05	0.05	0.06	0.06
H ₂	0.12	0.12	0.12	0.12
CH ₄	0.02	0.02	0.02	0.02
H ₂ S	0.00	0.00	0.00	0.00
N ₂	0.52	0.52	0.53	0.52
Syngas properties and process performance				
Syngas mass flow (kg/h)	860	845	918	910
Syngas mass flow (kg/h, dry)	783	772	839	832
Lower heating val. (MJ/kg, dry)	4.58	4.58	4.29	4.29
Specific heat (kJ/(kg·K))	1.11	1.11	1.12	1.12
Cold gas efficiency	0.797	0.800	0.795	0.787
Byproducts				
Tar production (kg/h)	50	45	80	92
Ash production (kg/h)	5.5	7.0	5.0	5.4

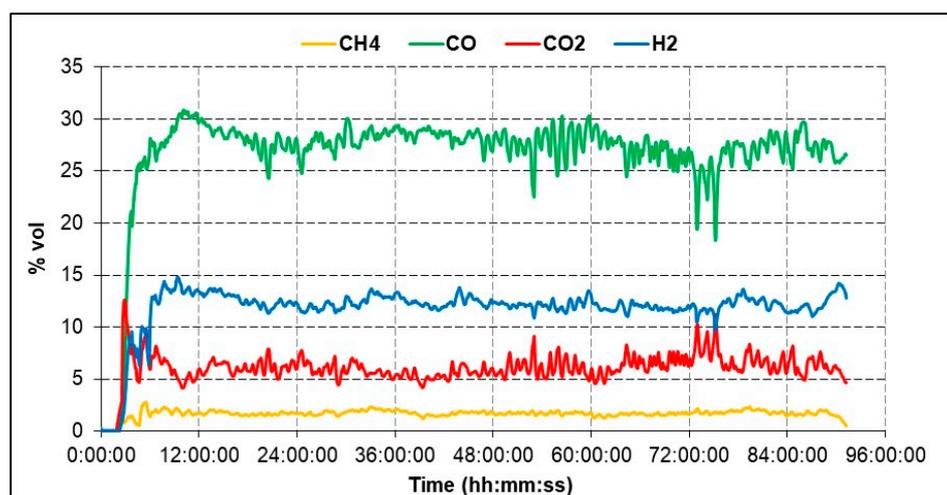


Figure 4. Raw syngas composition trend for stone pine gasification.

If the cold gas efficiency is the key performance indicator for the gasification process, a general performance on the whole plant (excluding the power generation system, not considered in this study) can be assessed on the basis of a carbon balance, based on the simplified scheme reported in Figure 6.

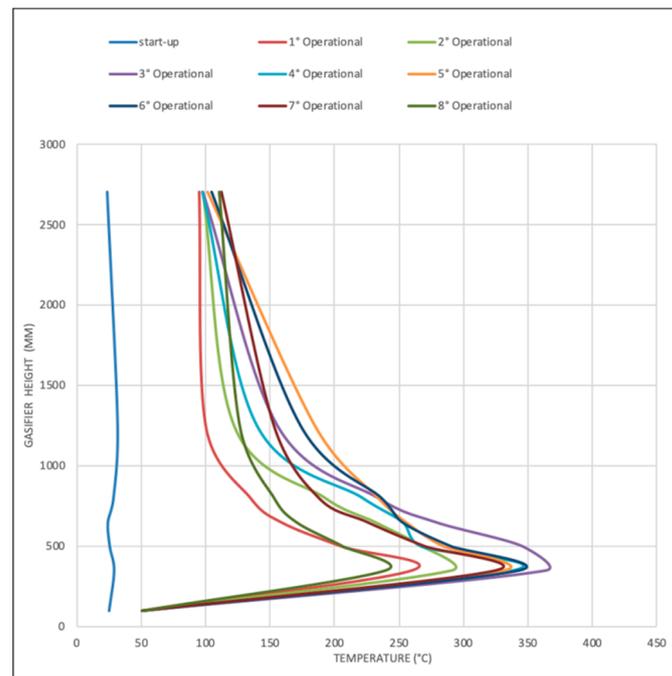


Figure 5. Wall temperature profiles for stone pine gasification.

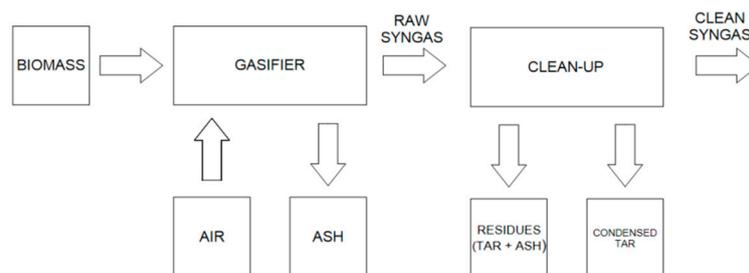


Figure 6. Process scheme for carbon balance assessment.

Globally, carbon balance can be easily represented by the following equation:

$$C_{in} = C_{out} + C_{acc} \quad (2)$$

where C_{in} is carbon that enters the plant through the primary fuel, C_{out} is carbon that leaves the plant and C_{acc} represents carbon accumulation within the process. In particular, C_{out} includes carbon contained in bottom ash (which is negligible, indicating a very efficient conversion during the gasification process), in the scrubber residues (some 50 kg/h of heavy tars and dust discharged from the bottom of the scrubber), in light condensed tar (about 10 kg/h separated from the free surface of the scrubber and discharged by the oil skimmer), and in clean syngas as CO, CO₂ and CH₄. The residues analyses are summarised in Table 3. On the other hand, C_{acc} does not include any contribution from the gasification unit (the process works at steady state and the fuel level into the gasifier is kept almost constant during the whole run), whereas some accumulation occurs in the wet scrubber (carbon residues in the washing water) and in the filters of the water recirculation system (about 7 kg/h). Due to these accumulations, and in order not to saturate the washing water and keep the water levels constant in the tanks, a make-up of fresh water of about 60 kg/h is required.

Table 3. Residues characterisation (% by weight).

Parameter	Bottom Ash	Liquid Residues	Condensed Tar
Fixed carbon	0.14	n.a.	n.a.
Volatiles	18.66	0.00	0.00
Moisture	0.13	30.00	20.00
Tars (C10–C40)		68.00	80.00
Minerals	81.08	2.00	0.00
Lower heating value (MJ/kg, dry basis)	n.a.	26.00	22.00

3.2. Wastewater Treatment Optimisation and Performance

The efficiency of the implementation of an oil skimmer in the scrubber loop system is evaluated in terms of pH, and removal of total suspended solids (TSS). Wastewater from the scrubber tank is characterised by pH values from 3.5 to 4.5, with TSS of about 200 mg/dm³ (measured by filtration and drying of the collected samples).

During the first gasification campaigns (before the installation of the oil skimmer), the content of tar and inorganic compounds in clean-up water tended to increase, while pH decreased down to a pseudo steady-state value of about 3.5. Lab scale titration experiments with 1.2 M HCl showed a high buffer capacity of the clean-up water around this value. This behaviour has been confirmed also after CO₂ removal from the samples by insufflation of air: the separation between liquid and solid phases (the latter constituted by the heavy tar deposited on the bottom of the scrubber) was observed. Light tar is partially separated by the bubbles rising in the liquid phase in the scrubber, and forms a layer above the free water surface. The dissolved fraction of tar is almost completely recirculated in the liquid stream and tends to accumulate, increasing the organic load with time. The high content of pollutants in the clean-up water reduces the effectiveness of scrubbing and may lead to fouling of surfaces and blocking of the recirculation pumps. Different strategies have been implemented or planned to remove organics from the clean-up water with in-line processes, to increase the effectiveness of the scrubbing and reduce water make-up. Furthermore, this complete in-line treatment system makes it possible not to use the chemical separation process, with a decrease in the investment and operation costs. In particular, the residues, consisting mainly of tar with a high specific weight C20–C40, are expelled from the bottom of the scrubber tank where washing takes place. The tar with low specific weight (C10–C20), floating on the top layer of the tank, is taken from the oil skimmer, concentrated and separated from the water by means of a mechanical rotary filter and subsequently expelled from the plant.

Based on a lab scale study, a pilot scale system for treatment of the clean-up water has been designed and implemented, with the aim to study a possible plant-scale process for the removal of dissolved tar, thus allowing wastewater recirculation in a closed loop with minimum make-up. Adsorption with carbon-based sorbents has been tested, with commercial activated carbon (AC) but also with two coal samples from South Africa and Venezuela (both characterised by similar properties: a lower heating value of 25 MJ/kg and a content of carbon, moisture and ash of 54%, 8% and 15% by weight, respectively), to assess the possibility to recirculate the exhaust sorbent in the gasification unit. The detailed results of this analysis at lab- and pilot-scale can be found elsewhere [43]. Lab scale experiments show that samples of coal have very low sorption capacity, one order of magnitude lower than the corresponding values of AC, and low retention times in the breakthrough curves.

Based on these results, the scaling up of the process to plant size has been assessed. A further experimental campaign has been carried out, where the pilot-scale column has been inserted in the secondary line and fed for several days, evaluating the turnover time of the sorbent with different flow rates. As a result, a turnover of about 50 h is required to ensure almost complete removal of dissolved tar with columns of 2.5–3.0 m height. The exhaust sorbent has been then characterised, in order to evaluate whether it can be mixed with the biomass in the feed to gasification. Table 4 shows the comparison between the characteristics of raw sorbent, exhausted sorbent, and a biomass already

used in the feed of the gasification unit. The assessment shows that the exhaust AC can be added to the feed: heat power is higher than the corresponding amount of biomass, the content of ashes is similar. The moisture content is considerably higher, which can be a problem in a field application. However, AC may constitute a considerable fraction of the feed: 20% of AC results in a 2% increase of moisture in the feed, which can be acceptable for the plant. Moreover, a possible pre-treatment of drying may be implemented.

Table 4. Characterisation of raw and exhaust activated carbon (% by weight).

Parameter	Raw AC		Exhaust AC	
	As Received	Dry Basis	As Received	Dry Basis
Fixed carbon	79.07	82.03	56.08	79.80
Volatiles	2.89	3.00	6.60	9.40
Moisture	3.62	0.00	29.75	0.00
Ash	14.43	14.97	7.58	10.80
Lower heating value (MJ/kg, dry basis)	26.39	n.a.	18.47	n.a.

This study, in addition to all the previous experimental activities on the Sotacarbo demonstration-scale unit [39], has allowed confirmation of the efficiency of the fixed-bed updraft gasification process with a further improvement of the technology, with particular reference to the applications for power generation from waste biomass, such as wood chips from wood management but also agricultural residues. Most of the gasification results have been used to optimise a commercial-scale unit under design in Alaska [4].

Particular attention has been paid to develop and optimise the wastewater management; the introduction of the oil skimmer in the pilot unit reduced the organic load from 10% to 30%, depending of the runs. Moreover, several experiments have been carried out on the use of active carbon to treat the pyrolysis liquids (oils and tars), with the potential reuse (and energy recovery) of the spent material as primary fuel of the gasification unit. New tests are currently underway to assess the content of other contaminants that can be found in raw syngas generated by different feedstock and in different operating conditions. The stated goal is to make the necessary changes to connect the system to the electricity grid.

As an alternative solution, recovery of pyrolysis oils separated from raw syngas as a fuel additive in diesel engines will be exploited [4].

4. Conclusions

This paper reports the main results of the experimental activity—carried out on a demonstration plant at the Sotacarbo Research Centre in Southwest Sardinia (Italy)—to optimise and develop a fixed-bed updraft gasification process for power generation from biomass. In particular, the experimental campaign with stone pine and eucalyptus wood chips as primary fuels has shown a very high gasification performance, around 79–80% in terms of cold gas efficiency. Syngas has a very stable composition during the whole run (also thanks to the robust grate driving system, specifically designed and modified by ENEA and Sotacarbo to optimise the process operation), with a mean lower heating value in the order of 4.3–4.6 MJ/kg (syngas being composed of 52–53% by nitrogen from the gasification air).

Even if the maximum fuel bed temperature ranges between 820 and 880 °C, the highest zone of the fuel bed is relatively cold, with a freeboard mean temperature of about 300–380 °C. It involves a relatively high tar formation that required a careful design of the syngas cleaning section and particular attention to optimise the wastewater management to reduce make-ups and wastewater disposal and possibly recover the high energy content associated with tar and spent sorbents. A first characterisation of the process has been carried out and the modalities and key parameters to reach the standard operating conditions have been identified. Moreover, the optimal process parameters for the operation of the syngas cleaning section have been identified and the configuration has been

modified (in particular, with the introduction of the oil skimmer integrated with the wet scrubbing system), with the result of a 60% reduction of wastewater disposal.

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References

1. International Energy Agency. *World Energy Outlook 2019*; International Energy Agency: Paris, France, 2019.
2. European Union. Directive (EU) 2018/2001 of the European Parliament and of the Council of 11 December 2018 on the promotion of the use of energy from renewable sources. *Official J. Eur. Union* **2018**, *61*, 82–209.
3. Krzyzaniak, M.; Stolarski, M.J.; Graban, L.; Lajszner, W.; Kuriata, T. Camelina and crambe oil crops for bioeconomy—Straw utilisation for energy. *Energies* **2020**, *13*, 1503. [[CrossRef](#)]
4. Ward, C.; Goldstein, H.; Maurer, R.; Thimsen, D.; Sheets, B.J.; Hobbs, R.; Isgrigg, F.; Steiger, R.; Revay Madden, D.; Porcu, A.; et al. Making coal relevant for small scale applications: Modular gasification for syngas/engine CHP applications in challenging environments. *Fuel* **2020**, *267*, 117303. [[CrossRef](#)]
5. Ferreira, S.; Minteiro, E.; Calado, L.; Silva, V.; Brito, P.; Vilarinho, C. Experimental and modeling analysis of brewers' spent grains gasification in a downdraft reactor. *Energies* **2019**, *12*, 4413. [[CrossRef](#)]
6. Marcantonio, V.; Bocci, E.; Monarca, D. Development of a chemical quasi-equilibrium model of biomass waste gasification in a fluidized-bed reactor by using Aspen Plus. *Energies* **2020**, *13*, 53. [[CrossRef](#)]
7. Gallucci, F.; Liberatore, R.; Sapegno, L.; Volponi, E.; Venturini, P.; Rispoli, F.; Paris, E.; Carnevale, M.; Colantoni, A. Influence of oxidant agent on syngas composition: Gasification of hazelnut shells through an updraft reactor. *Energies* **2020**, *13*, 102. [[CrossRef](#)]
8. Chambon, C.L.; Karia, T.; Sandwell, P.; Hallett, J.P. Techno-economic assessment of biomass gasification-based mini-grids for productive energy applications: The case of rural India. *Renew. Energy* **2020**, *154*, 432–444. [[CrossRef](#)]
9. Ren, J.; Liu, J.L.; Zhao, X.Y.; Cao, J.P. Methanation of syngas from biomass gasification: An overview. *Int. J. Hydrogen Energy* **2020**, *45*, 4223–4243. [[CrossRef](#)]
10. Szul, M.; Iluk, T.; Sobolewski, A. High-temperature, dry scrubbing of syngas with use of mineral sorbents and ceramic rigid filters. *Energies* **2020**, *13*, 1528. [[CrossRef](#)]
11. Situmorang, Y.A.; Zhao, Z.; Yoshida, A.; Abudula, A.; Guan, G. Small-scale biomass gasification systems for power generation (<200 kW class): A review. *Renew. Sustain. Energy Rev.* **2020**, *117*, 109486.
12. Siddiqui, H.; Thengane, S.K.; Sharma, S.; Mahajani, S.M. Revamping downdraft gasifier to minimize clinker formation for high-ash garden waste as feedstock. *Bioresour. Technol.* **2018**, *266*, 220–231. [[CrossRef](#)] [[PubMed](#)]
13. Sikarwar, V.S.; Zhao, M.; Clough, P.; Yao, J.; Zhong, X.; Memon, M.Z.; Shah, N.; Anthony, E.J.; Fennell, P.S. An overview of advances in biomass gasification. *Energy Environ. Sci.* **2016**, *9*, 2939. [[CrossRef](#)]
14. Porcu, A.; Sollai, S.; Marotto, D.; Mureddu, M.; Ferrara, F.; Pettinau, A. Techno-economic analysis of a small-scale biomass-to-energy BFB gasification-based system. *Energies* **2019**, *12*, 494. [[CrossRef](#)]
15. Kittivech, T.; Fukuda, S. Investigating agglomeration tendency of co-gasification between high alkali biomass and woody biomass in a bubbling fluidized bed system. *Energies* **2020**, *13*, 56. [[CrossRef](#)]

16. Huang, Y.; Zhao, Y.J.; Hao, Y.H.; Wei, G.Q.; Feng, J.; Li, W.Y.; Yi, Q.; Mohamed, U. A feasibility analysis of distributed power plants from agricultural residues resources gasification in rural China. *Biomass Bioenergy* **2019**, *121*, 1–12. [[CrossRef](#)]
17. Ren, J.; Cao, J.P.; Zhao, X.Y.; Yang, F.L.; Wei, X.Y. Recent advances in syngas production from biomass catalytic gasification: A critical review on reactors, catalysts, catalytic mechanisms and mathematical models. *Renew. Sustain. Energy Rev.* **2019**, *116*, 109426. [[CrossRef](#)]
18. Lee, S.Y.; Park, S.W.; Alam, M.T.; Jeong, J.O.; Seo, Y.C.; Choi, H.S. Studies on the gasification performance of sludge cake pre-treated by hydrothermal carbonization. *Energies* **2020**, *13*, 1442. [[CrossRef](#)]
19. Singh Siwal, S.; Zhang, Q.; Sun, C.; Thakur, S.; Gupta, V.K.; Thakur, V.K. Energy production from steam gasification processes and parameters that contemplate in biomass gasifier—A review. *Bioresour. Technol.* **2020**, *297*, 122481. [[CrossRef](#)]
20. Hobbs, M.L.; Radulovic, P.T.; Smoot, L.D. Modeling fixed-bed coal gasifier. *AIChE J.* **1992**, *38*, 681–702. [[CrossRef](#)]
21. Hahn, O.H.; Wesley, P.D.; Swisshelm, B.A.; Maples, S.; Withrow, J. A mass and energy balance of a Wellman–Galusha gasifier. *Fuel Process. Technol.* **1979**, *2*, 332–334. [[CrossRef](#)]
22. Yu, J.; Smith, J.D. Validation and application of a kinetic model for biomass gasification simulation and optimization in updraft gasifiers. *Chem. Eng. Process. Process Intensif.* **2018**, *125*, 214–226. [[CrossRef](#)]
23. Mehta, V.; Chavan, A. Physico-chemical treatment of tar-containing wastewater generated from biomass gasification plants. *Int. J. Chem. Mol. Eng.* **2009**, *3*, 458–465.
24. Manek, B.; Javia, M.S.; Harichandan, A.; Ramani, H. A CFD based comprehensive study of coal-fired updraft gasifier in ceramic industry. *Therm. Sci. Eng. Prog.* **2019**, *9*, 11–20. [[CrossRef](#)]
25. Murgia, S.; Vascellari, M.; Cau, G. Comprehensive CFD model of an air-blown coal-fired updraft gasifier. *Fuel* **2012**, *101*, 129–138. [[CrossRef](#)]
26. Lu, D.; Yoshikawa, K.; Ismail, T.M.; El-Salam, M.A. Assessment of the carbonized woody briquette gasification in an updraft fixed bed gasifier using the Euler-Euler model. *Appl. Energy* **2018**, *220*, 70–86. [[CrossRef](#)]
27. Lu, D.; Yoshikawa, K.; Ismail, T.M.; El-Salam, M.A. Computational fluid dynamics model on updraft gasifier using carbonized woody briquette as fuel. *Energy Procedia* **2017**, *142*, 166–171. [[CrossRef](#)]
28. James R, A.M.; Yuan, W.; Wang, D.; Wang, D.; Kumar, A. The Effect of Gasification Conditions on the Surface Properties of Biochar Produced in a Top-Lit Updraft Gasifier. *Appl. Sci.* **2020**, *10*, 688. [[CrossRef](#)]
29. Lei, M.; Hai, J.; Cheng, J.; Lu, J.; Zhang, J.; You, T. Variation of toxic pollutants emission during a feeding cycle from an updraft fixed bed gasifier for disposing rural solid waste. *Chin. J. Chem. Eng.* **2018**, *26*, 608–613. [[CrossRef](#)]
30. Font Palma, C. Modelling of tar formation and evolution for biomass gasification: A review. *Appl. Energy* **2013**, *111*, 129–141. [[CrossRef](#)]
31. Devi, J.; Ptasiński, K.J.; Janssen, F.J.J.G. A review of the primary measures for tar elimination in biomass gasification processes. *Biomass Bioenergy* **2003**, *24*, 125–140. [[CrossRef](#)]
32. Mahjoub, B.; Jayr, E.; Bayard, R.; Gourdon, R. Phase partition of organic pollutants between coal tar and water under variable experimental conditions. *Water Res.* **2000**, *34*, 3551–3560. [[CrossRef](#)]
33. Ruiz, J.; Juárez, M.; Morales, M.; Muñoz, P.; Mendivil, M. Biomass gasification for electricity generation: Review of current technology barriers. *Renew. Sustain. Energy Rev.* **2013**, *18*, 174–183. [[CrossRef](#)]
34. Park, H.I.; Wu, C.J.; Lin, L.S. Coal tar wastewater treatment and electricity production using a membrane-less tubular microbial fuel cell. *Biotechnol. Bioprocess Eng.* **2012**, *17*, 654–660. [[CrossRef](#)]
35. Luthy, R.G.; Dzombak, D.A.; Peters, C.A.; Roy, S.B.; Ramaswami, A.; Nakles, D.V.; Nott, B.R. Remediating tar-contaminated soils at manufactured gas plant sites—technological challenges. *Environ. Sci. Technol.* **1994**, *28*, 266–276. [[CrossRef](#)]
36. Milne, T.A.; Evans, R.J.; Abatzoglou, N. *Biomass Gasifier “Tars”: Their Nature, Formation, and Conversion*; Report NREL/TP-570-25357; U.S. National Renewable Energy Laboratory: Golden, CO, USA, 1998. Available online: <https://www.nrel.gov/docs/fy99osti/25357.pdf> (accessed on 1 October 2019).
37. Han, J.; Kim, H. The reduction and control technology of tar during biomass gasification/pyrolysis: An overview. *Renew. Sustain. Energy Rev.* **2008**, *12*, 397–416. [[CrossRef](#)]
38. Nyazi, K.; Baçaoui, A.; Yaacoubi, A.; Darmstadt, H.; Adnot, A.; Roy, C. Influence of carbon black surface chemistry on the adsorption of model herbicides from aqueous solution. *Carbon* **2005**, *43*, 2218–2221. [[CrossRef](#)]

39. Calí, G.; Deiana, P.; Bassano, C.; Maggio, E. Experimental activities on Sotacarbo 5 MWth gasification demonstration plant. *Fuel* **2017**, *207*, 671–679. [CrossRef]
40. Ghani, M.U.; Radulovic, P.T.; Smoot, L.D. An improved model for fixed-bed coal combustion and gasification: Sensitivity analysis and applications. *Fuel* **1996**, *75*, 1213–1226. [CrossRef]
41. Thimsen, D.P.; Maurer, R.E.; Pooler, A.R.; Pui, D.Y.H.; Liu, B.Y.H.; Kittelson, D.B. Fixed-Bed Gasification Research Using US Coals. Volume 19—Executive Summary. US Bureau of Mines Contract H0222001. 1985. Available online: <https://www.osti.gov/servlets/purl/5704367> (accessed on 15 May 2019). [CrossRef]
42. De Filippis, P.; Scarsella, M.; de Caprariis, B.; Uccellari, R. Biomass gasification plant and syngas clean-up system. *Energy Procedia* **2015**, *75*, 240–245. [CrossRef]
43. Calí, G.; Deiana, P.; Maggio, E.; Marotto, D.; Mascia, M.; Vacca, A. Management and treatment of the clean-up water from the scrubber of a coal and biomass gasification plant: An industrial case study. *Chem. Eng. Trans.* **2019**, *74*, 337–342.
44. Conte, L.; Falletti, L.; Zaggia, A.; Milan, M. Polyfluorinated organic micropollutants removal from water by ion exchange and adsorption. *Chem. Eng. Trans.* **2015**, *43*, 2257–2262.
45. Liu, Z.Q.; You, L.; Xiong, X.; Wang, Q.; Yan, Y.; Tu, J.; Cui, Y.H.; Li, X.Y.; Wen, G.; Wu, X. Potential of the integration of coagulation and ozonation as a pretreatment of reverse osmosis concentrate from coal gasification wastewater reclamation. *Chemosphere* **2019**, *222*, 696–704. [CrossRef] [PubMed]



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