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Editorial

An Overview of Waste Gasification and Syngas Upgrading Processes

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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 or chemical production.

The valorisation and disposal of municipal and industrial wastes are challenges of the next generation and must have key roles in energy transitions in order to reduce the carbon footprint of human activities. Landfilling is the most traditional and easiest method for waste disposal, but it is also characterized by significant impacts from both environmental and socio-economic viewpoints; hence, landfilling should be avoided and replaced with waste-reutilization techniques.

The use of waste or residual material as raw material for chemical production can be considered a smart solution for waste disposal, which can provide new life to a carbon source. Thermochemical treatments are seen as emerging technologies that can help reduce the volume of waste by recovering energy and/or matter by obtaining products with high added value during the final treatment of municipal solid wastes (MSWs). The thermochemical conversion of waste aimed at the synthesis of new chemical products represents a valid solution and is one of the pillars of the philosophy of circular economies.

Gasification is considered a clean and effective method for converting low quality biomass to higher value gas and for solving various waste utilization problems as well.

Gasification transforms low-value materials, converting them into gaseous products with usable calorific value and/or marketable products. Syngas, also called synthesis gas, is mainly composed of carbon monoxide (CO) and hydrogen (H_2) besides CO_2 . After proper cleaning and upgrading processes, syngas can be used as an alternative fuel in turbogas to generate heat and electricity or as a raw material for a large number of products in the petrochemical industry and in refinery, such as methanol, plastic monomers, oxo alcohols, ammonia, synthetic gasolines, etc.

This high gasification flexibility, both in terms of raw material type and power generation or chemicals production options, is what drives the expansion of research and implementation opportunities for biomass and waste gasification.

Although these technologies are commercially available, research studies on optimizing and exploring its further potential are still ongoing. In particular, in recent years, new pyrolysis and gasification technologies emerged with the aim of increasing possible feedstocks that improve energy recovery and reduce environmental impacts.

A number of theoretical studies on these kinds of processes are currently available in the scientific literature, and these are mainly focused on the development of thermodynamic models or on computational fluid dynamics (CFD) process simulation, in some cases with the experimental model validation in pilot units. Several studies are available on pilot-scale experimental development of the process for waste gasification.

In this editorial, an essential scenario of several aspects regarding waste gasification and syngas upgrades will be reported with reference to selected papers published in the



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Energies **2022**, 15, 6391 2 of 7

journal *Energies* during the past two years, thus providing an updated vision of what is being currently studied in the field.

The topics covered are numerous, and although the contributions include leading innovative elements in the field of waste gasification, the most important aspects have been reported following a common guideline among the studies analyzed. The selected papers are strongly interrelated among each other at several points, as quite different problems converge in the area of advanced waste to syngas plant challenges. Therefore, selecting the proper keywords that, according to the authors, characterize the research, allows building the correlation matrix reported in Table 1, which helps the reader to have a global view of the arguments of the considered advanced research papers. Moreover, a short summary of all papers is reported. The order of the discussed papers has been accurately chosen as follows.

[1]	[2]	[3]	[4]	[5]	[6]	[7]	[8]	[9]
Χ	Х	Х	Х	Х	Х	Х	Х	Х
X	X	X	X	X	X	X	X	X
X		Χ			X	X	X	X
X					X	X	X	X
X	X	X	X	X	X	X	X	Χ
X	X	X	X	X	X	X	X	Χ
			X			X		Χ
Χ				X	X			Χ
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Table 1. Correlation matrix of the selected keywords.

A performance aspect in which gasification technologies are unique compared to other conventional waste thermal-treatment methods is that the product of the treatment can be both energy and a chemical or fuel: consequently, treatments can be classified as either waste-to-energy or waste-to-chemical treatments.

This has not been considered very important in the past when reducing the volume of waste to avoid landfills was the main driver of heat treatments. However, as there is an increasing push towards the decarbonisation of transport systems and for biomaterials, this is an area that can be expected to become increasingly interesting in the future.

Waste-to-energy (WtE) or energy-from-waste (EfW) treatment is the process of generating energy in the form of electricity and/or heat from the primary treatment of waste or the processing of waste into a fuel source. WtE is a form of energy recovery. WtE gasification processes produce a combustible fuel commodity, such as syngas, to generate electricity and/or heat.

Combined heat and power (CHP) production compared with single generation of power has two advantages: it helps improve the utilisation of energy production and helps reduce pollution.

Gasification technologies for small- and medium-scale combined heat and power (CHP) generation from waste biomass have been significantly developed in the last decades. Most of the attention is focused on fixed-bed downdraft processes, but a few studies also consider bubbling or circulating fluidised-bed gasification processes, sometimes promoted with specific catalysts or integrated with hydrothermal carbonisation to treat high-moisture biomass or waste. With respect to these technologies, fixed-bed updraft gasification introduces improved conversion efficiencies (thanks to the countercurrent heat exchanges) and it is typically not only 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%) but also involves a relatively high production of pyrolysis liquids (i.e., oils and tar).

Cali G. et al. [1] present an experimental development at the demonstration scale of an integrated gasification system fed with wood chips. This paper aims to establish the baseline performance of a 5 MWth demonstration-scale updraft gasifier, operating since 2014 at the Sotacarbo Research Centre in Sardinia, Italy, and tested for some 1500 h with

Energies **2022**, 15, 6391 3 of 7

different types of coal and biomass. In particular, the experimental results reported here are focused on the gasification of two forms of local waste biomass, with the aim of assessing operating conditions, syngas composition and properties and whole-plant performance. In addition, syngas cleaning and wastewater treatment process performances were 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 entire project).

The gasification unit is characterized by high cold-gas efficiencies (about 79–80%) and operation stabilities during each test. Particular attention has been directed to the optimization of an integrated double-stage wastewater management system—which includes an oil skimmer and an activated carbon adsorption filter—designed to minimize both liquid residues and water make-up. Syngas has a very stable composition during the entire run, 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). Moreover, the optimal process parameters for the operation of the syngas cleaning section have been identified and the configuration has been modified, with a result of a 60% reduction in wastewater disposal.

To meet the global demand for energy and to reduce the use of fossil fuels, alternative systems such as the production of syngas from renewable biomass gasification have been investigated by Muhammad et al. [2]. To design such gasification systems, model equations can be formulated and solved to predict the quantity and quality of the syngas produced with different operating conditions (temperature, the flow rate of gasifying agent, etc.) and with different types of biomasses (wood, grass, seeds, food waste, etc.).

The modelling of biomass gasification generally involves the formulation and solution of sets of equations, including mass and energy balances, in addition to either rate-based or equilibrium-based expressions to determine the effect of the occurring reactions. To simplify this approach, various assumptions can be made, and correlations can be utilized relating to experimentally measured properties (e.g., feed composition and operating conditions). The more complex approaches include the use of computational fluid dynamics (CFD) models and kinetic rate expression models, which require knowledge of the reaction and diffusion parameters and potentially require relatively longer computational times than the simpler modelling methods available. Several models can be obtained by using different correction factors, model parameters, and assumptions, and these models are compared and validated against experimental data and modelling studies from the literature.

In this study, several different stoichiometric thermodynamic models are compared to determine which are the most suitable and reliable. The analysis has been carried out for the prediction performance to predict syngas compositions. To correct some of the errors associated with thermodynamic models, correction factors are utilized to modify the equilibrium constants of methanation and water gas-shift reactions, using the data from 27 experimental values published in the literature, which allows them to better predict the real output composition of the gasification reactors. The models are formulated and optimized in MATLAB. These models also have the ability to estimate the performance of biomass gasification for different operating conditions, such as temperature and moisture contents. In this study, we show that similar results and model accuracies can be achieved with a simpler model in which correction factors are fixed parameters.

In future, more complex models can also be proposed, involving more fitted parameters in order to improve the prediction accuracy of the models. For example, this could be performed by including the dependence on different operating conditions or switching to a non-stoichiometric equilibrium model that could potentially have large numbers of fitted parameters. In addition, the minor gas products and trace products can also be included to provide more detailed outputs. This could also consider the production of toxic emissions, including dioxins and furans.

In order to overcome some of the technological barriers, a deep understanding of the different phenomena and operational parameters involved in gasification process is Energies **2022**, 15, 6391 4 of 7

required. To this aim, mathematical modeling is a useful tool for understanding physical and chemical mechanisms that occur inside the gasifier. A numerical model allows for the evaluation of process performance when varying biomass proprieties and operating parameters, providing a set of optimum operating conditions and designating the risks and limits of the system. Additionally, a numerical model helps considerably reduce costs associated with the research and development of new and innovative devices.

In Ref. [3], Moretti and co-authors present an overview of biomass gasification modeling approaches in order to evaluate their effectiveness as a tool for the design and optimization of poly-generation plants based on biomass gasification. The authors developed two equilibrium models using both a commercial software and a simulation tool implemented in a non-commercial script. In this paper, the reliability of equilibrium gasification models currently available in the scientific literature was investigated to establish whether they can be employed to build new control and optimization schemes and operating maps of biomass gasification systems integrated into polygeneration plants coupled with energy networks. The developed thermodynamic equilibrium models were applied to different case studies on downdraft gasifiers available in the literature in order to evaluate the accuracy of the simulation results upon varying the biomass type and composition and the operating conditions of the gasification process, evidencing the advantages and disadvantages of both models.

The obtained results highlighted strengths and limitations of using equilibrium models as a function of biomass type and composition. They showed that the analytical model predicted syngas composition with better accuracy for biomass types characterized by a low ash content, whereas the Aspen model appeared to fairly predict the syngas composition; however, its accuracy might be reduced if the properties of the treated biomass changed. The proposed overview of thermodynamic equilibrium models has significant industrial implications, allowing for the selection of the most suitable and reliable model as a function of biomass properties and industrial operating conditions. Therefore, industries will benefit from this study by having a clearer view of the most reliable models for the gasification process.

In the last years, many advanced gasification technologies (AGTs) using waste or biomass have been developed. AGTs are considered to be potential low-carbon routes for sectors that are known to be difficult or expensive to decarbonise.

Globally, several AGTs for the production of syngas are in various stages of development for operation mainly on biomass or RDF. Most projects in development are focussed more on waste streams than biomass for economic reasons.

The aspects most studied are the characteristic of reactors, the gasifying agents, the operative parameters, and the upgrading processes. Syngas clean-up systems for the removal of alkali metals, particulates, tars, sulphur compounds, and several other contaminants must be coupled with a gasification system to ensure that the syngas delivered to the upgrading system meets the technical specification for commercial operation.

Stasiek J. et al. [4] present an advanced thermal conversion system involving the high-temperature gasification of biomass and municipal waste into biofuel, syngas, or hydrogen-rich gas. The decomposition of solid biomass and waste by gasification was experimented with a modern and innovative updraft continuous regenerator and gasifier. A ceramic high-cycle regenerator provided extra energy for the thermal conversion of biomass or any other solid waste. Highly preheated air and steam gas (heated up to 1600 °C) were used as gasifying agents (feed gas). The preheated feed gas also improved the thermal decomposition of solid feedstock for fuel gas. However, the main objective of this study was to promote new and advanced technologies for the thermochemical conversion of biomass for the production of alternative energy. Finally, this novel and unique technology can ensure significant energy savings of more than 30%, the downsizing of equipment, very low NO_x emissions (around 50%), and a reduction in harmful pollutants (about 25%). This technology has tremendous potential for many applications, primarily for energy saving, and fulfills all new regulations on waste incineration proposed by the European Com-

Energies **2022**, 15, 6391 5 of 7

mission. The presented results concerning HiTAG/HiTSG technology proved to be very useful for gas fuel, hydrogen, or syngas generation. The high temperatures of the process generated in the preheater eliminate harmful substances such as tars, dioxins, and furans, which can be produced in low-temperature conversion processes. Fuel gas can be used later to produce heat and electricity or even hydrogen by means of separation processes.

In order to reach much higher efficiencies, coupling systems of biomass gasification with advanced power generation have been developed: The gas turbine, internal combustion engine (ICE), and solid oxide fuel cell (SOFC) are the most common types of coupling. These combined heat and power (CHP) units on a small scale, based on gasification, can use an ICE or micro-gas and small gas turbine with electrical efficiencies up to 25% of the biomass' lower heating value (LHV). Biomass gasification coupled with SOFC produces electrical efficiencies up to 60%. The SOFC has become a very important energy technology due to its clean, greener, and efficient operation. Fuel cells allow the conversion of energy with higher efficiency; in fact, they convert the chemical energy contained in a fuel gas directly to electrical energy by electrochemical reactions.

In the last years, many studies were carried out to study the optimal operating conditions, the overall system performance, and the limitations and the potentials of a variety of SOFC power plants integrated with biomass gasification systems. However, the current commercial process simulation software packages do not include built-in models for SOFC. In the literature, the most common SOFC system modelling approach is to integrate process simulators with a stack model in a programming language.

Marcantonio et al. [5] coupled Solid-Oxide Fuel Cells (SOFCs) to gasification technology. A steady-state model of a biomass-SOFC was developed using a process simulation software, ASPEN Plus. The present paper showed an entire plant composed by a gasification part, a gas cleaning unit, and an SOFC system. This is one of the few studies available in the literature that includes all parts. The goal of this study was to develop a biomass-SOFC system capable of predicting performance in the function of operating conditions. The results show that there must be a trade-off between voltage, electrical efficiency, and power with respect to current density, and it is preferable to stay at a low steam-to-biomass ratio. The electrical efficiency achieved under the operating conditions is 57%, a high value, making these systems very attractive.

According to the study of Di Giuliano et al. [6], the chemical looping gasification of residual biomasses, operated in fluidized beds composed of oxygen carriers (OCs), may allow the production of biofuels from syngas. The chemical looping gasification for the sustainable production of biofuels (CLARA) research project, funded by the EU Horizon 2020 framework program, aims to contribute to this dimension. This project deals with the chemical looping gasification (CLG) of biogenic residues, with the obtained syngas used in Fischer–Tropsch syntheses to produce liquid fuels, as also obtained by the hydrocracking of waxes resulting from Fischer–Tropsch. The main goal of CLARA is the realization of a full biomass-to-fuel chain up to the 1 MWth scale in an industrially relevant environment (targets: cold gas efficiency of 82%, carbon conversion of 98%, tar in outlet syngas lower than 1 mg Sm⁻³).

Devolatilization is a key step of the gasification process and strongly influences both the quantity and quality of obtained syngas. At temperatures of gasification (typically up to 900 °C), vapours and tars—developed by primary devolatilization reactions—undergo secondary reactions, which contribute to both gaseous products (cracking and reforming) and solid products (polymerization).

This study aims to provide useful experimental data about the devolatilization of residual biomasses. The study investigates wheat straw pellets (WSP) and raw pine-forest residue (RPR) pellets as feedstocks for chemical looping gasification presenting experimental results from devolatilizations of WSP and RPR in bubbling beds made of three different OCs or sand (inert reference) at 700, 800, and 900 °C. Tests were performed at the laboratory scale by a quartz reactor using nitrogen as the fluidizing agent. For each test gas yield (η_{av}), carbon conversion (χ_{avC}), H₂/CO ratio (λ_{av}), and syngas composition

Energies **2022**, 15, 6391 6 of 7

were determined. Temperature is the dominating parameter: At 900 °C, the highest quality and quantity of syngas was obtained (WSP: $\eta_{av} = 0.035-0.042$ molgas·gbiomass⁻¹, $\chi_{avC} = 73-83\%$, $\lambda_{av} = 0.8-1.0$), RPR: $\eta_{av} = 0.036-0.041$ molgasgbiomass⁻¹, $\chi_{avC} = 67-71\%$, $\lambda_{av} = 0.9-1.0$), and oxygen carriers generally performed better than sand. The kinetic analysis suggested that the fastest conversion of C and H atoms into gases, at tested conditions, is ensured by the oxygen carrier, ilmenite.

Collected results have an important novelty value for both experimental and modelling studies since they deal with (i) residual biomasses with a great availability potential, which is currently unexploited; (ii) devolatilization, a single step of the more complex gasification process, tricky in being experimentally isolated especially at higher scales; (iii) the formulation of kinetic expressions of devolatilization/pyrolysis, a crucial point that is often lacking for a full-predictive modeling approach; and (iv) chemical looping gasification by means of OCs, a thermochemical process that has not been developed yet at industrial scales.

The next study [7] provides a comprehensive review of micro-scale thermal treatment technologies for non-sewered practices, which are up to date in commercial/pilot and small scales for various types of solid fuels. Furthermore, the challenges observed with the nominated (pyrolysis) technology are discussed in detail and addressed. This study suggests rapid energy recovery from byproducts primarily made up of the highest yield of syngas with a desirable calorific value. The optimum operating ranges are discussed to ensure a reliable thermal conversion of sludge materials considering the application's constraints and technological drawbacks due to the energy demand for drying. A pyrolysis temperature of 400–600 °C and the heating rates pursued at low ranks could result in a sustainable thermal conversion range for moist waste materials.

One objective of this critical review is to discuss the solid waste management/stabilisation technologies and their operational challenges. The state-of-the-art applications of these thermal treatment technologies for on-site, non-sewered, sanitary applications at small scales are reviewed to clarify the specific operational challenges occurring through their implementation. The aim of this study overall is to outline challenges for developing a suitable technology for on-site sanitary applications and to address optimum ranges of operating conditions for reliable performances.

Various technologies have been employed to reduce, capture, and utilise CO_2 from power generation. One such technique includes integrated gasification combined cycle (IGCC) power generation with carbon capture and storage (CCS). CO_2 generated from gasification plants can be reused in the same cycle as a reactant, which helps cut costly oxygen-enriched air, oxygen, and steam. Furthermore, the recycling of CO_2 offers a wide range of CO/H_2 production, which is the precursor for a wide range of chemicals. Numerous studies on coal and biomass gasification have been conducted using CO_2 as a reactant.

Shahabuddin et al. [8] evaluated the entrained-flow co-gasification characteristics of coal and biomass using thermodynamic equilibrium modeling. The effects of the equivalence ratio, temperature, pressure, and biomass to coal ratios have been investigated using CO_2 reactant. It has been understood that the lower heating value (LHV) and cold gas efficiency (CGE) increase with increasing temperatures until the process reaches a steady-state temperature of 1100 °C. Pressure affects syngas composition only at lower temperatures (<1100 °C). The variation in syngas composition is minor up to the blending of 50% biomass (PB50). However, the PB50 shows a higher LHV and CGE than pure coal by 12% and 18%, respectively. Overall, the biomass blending of up to 50% favours gasification performance with an LHV of 12 MJ/kg and a CGE of 78%. Results showed that the combination of steam and oxygen performs better compared to other reactants. The steam–oxygen reactant showed the highest H_2/CO ratio of 0.74, while the ratio was about 0.32 using steam- CO_2 and pure oxygen. The concentration of pollutants decreased consistently with increasing biomass ratios. Thus, co-gasification using CO_2 reactants can potentially increase gasification efficiency in addition to reducing pollutant emissions.

Energies **2022**, 15, 6391 7 of 7

To achieve the goal of decreasing or even obtaining negative CO_2 emissions and producing clean energy, a proper model of biomass conversion needs to be developed. Because of the ultimate properties of biomass (low energy density and high moisture content), it is better to provide the thermochemical conversion of this material using processes such as pyrolysis or gasification than combustion.

The last analyzed study [9] focused on biomass thermochemical conversions with integrated CO_2 capture, investigating the impact of pyrolysis temperature (500, 600, and 700 °C) and CaO sorbent addition on the chemical and physical properties of obtained char and syngas. In this study, a novel approach for syngas production from biomass with parallel biochar production and negative carbon emission was proposed. The conducted research study allowed us to predict the most effective pyrolysis process conditions to obtain the highest yield of syngas and biochar.

The pyrolysis process temperature directly influences the syngas as well as biochar composition. The analysis of the studied temperature range allowed us to conclude that a temperature increase leads to the production of biochar with a higher carbon content and lower VM, as the gaseous phase of biomass was processed and converted into syngas. With the increase in process temperatures, higher hydrogen and methane concentrations were obtained. However, concentrations of higher hydrocarbons, such as C_2H_4 , C_2H_6 , C_3H_6 , and C_3H_8 , decrease.

The addition of a sorbent such as CaO for CO_2 capture improves the final gas composition. The presence of the sorbent allows the capture of carbon dioxide from the produced syngas. As the process temperature increases, the concentration of CO_2 in syngas decreases. The conducted tests and analysis prove the ability of capturing the CO_2 released during the pyrolysis process and can transform it into a carbonate phase by CaO. The processes of tar cracking are promoted by the presence of CaO with the parallel removal of CO_2 and lead to an increase in H_2 production. In particular, the concentration of CO_2 in syngas decreased with increased temperatures, and the highest decrease occurred in the presence of CaO from above 60% to below 30% at 600 °C.

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