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Review

Completion of Waste Heat Recovery and CO_2 Conversion Simultaneously Based on the Flue Gas Chemical Recuperative Cycle: A Review

Mengze He, Ping Zhou *, Xiqiang Zhao * and Tao Wang *

National Engineering Laboratory for Reducing Emissions from Coal Combustion, Engineering Research Center of Environmental Thermal Technology of Ministry of Education, Shandong Key Laboratory of Energy Carbon Reduction and Resource Utilization, School of Nuclear Science, Energy and Power Engineering, Shandong University, Jinan 250061, China; 19510166726@163.com

* Correspondence: zhousuiyan@sdu.edu.cn (P.Z.); zxq@sdu.edu.cn (X.Z.); wtao@sdu.edu.cn (T.W.)

Abstract: Energy shortage and greenhouse gas emission have become bottlenecks in current society development. Improving the efficiency of energy conversion and utilization systems through waste heat recovery and reduction of greenhouse gas through CO_2 capture/conversion are important solutions. Both can be achieved simultaneously by utilizing high-temperature flue gas or CO_2 in flue gas for organic matter gasification, which is called the flue gas chemical recuperative cycle. This paper provides a meaningful review of the latest advancements in the flue gas chemical recuperative cycle system, focusing on its application in diverse gasification systems for organic matters such as methane, sludge, etc. Additionally, this paper reviews methods for the integration of flue gas gasification into energy conversion and utilization systems under the application scenarios of gas turbine flue gas, air combustion flue gas, and oxy-fuel combustion flue gas. Subsequently, in order to improve the conversion efficiency of the chemical recuperative cycle, the applications of emerging gasification technologies in the field of the flue gas recuperative cycle, such as microwave gasification, plasma gasification, etc., are briefly summarized, offering an in-depth analysis of the mechanisms by which new methods enhance the process. Finally, the prospects and challenges of the field are discussed, and a comprehensive outlook is provided to guide future research.

Keywords: flue gas recuperative cycle; CO_2 gasification; syngas; methane reforming; biomass gasification

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1. Introduction

Energy is the cornerstone of human civilization and plays a crucial role in human survival and social development. However, large quantities of greenhouse gases are generated during the process of energy utilization, leading to drastic climate changes [1,2]. In addition, the over-exploitation of fossil energy and the resulting energy shortage are also serious constraints on the development of society [3]. Therefore, how to improve the efficiency of the energy conversion/utilization system and reduce greenhouse gas emissions are two daunting tasks before mankind today [4].

Traditional fossil energy utilization systems such as boilers and gas turbines often emit large amounts of exhaust gases, along with a large amount of waste heat emitted simultaneously. Although some of the heat can be recovered using waste heat recovery technology, gases such as CO_2 and H_2O are not collected and need to be captured and

further immobilized or converted in a series of trapping systems, resulting in a complicated process and high energy consumption [5]. The flue gas recuperative cycle as an advanced cycle aims to optimize the system performance by recovering the waste heat and converting CO_2 and H_2O directly into combustible gases for utilization [6]. This process not only reduces heat loss, but also reduces greenhouse gas emissions [7]. Therefore, the flue gas recuperative cycle technology has been widely noticed and applied, becoming an important research hotspot in the energy field. With the development of gasification technologies, the superiority of the flue gas recuperative cycle will be more prominent.

This article analyzes how to efficiently utilize the waste heat in flue gas and the chemical composition of flue gas, filling the gap found in a review of this field. Based on this, this paper reviews the flue gas recuperative cycle along the lines shown in Figure 1, looks into its development prospects, and gives recommendations.

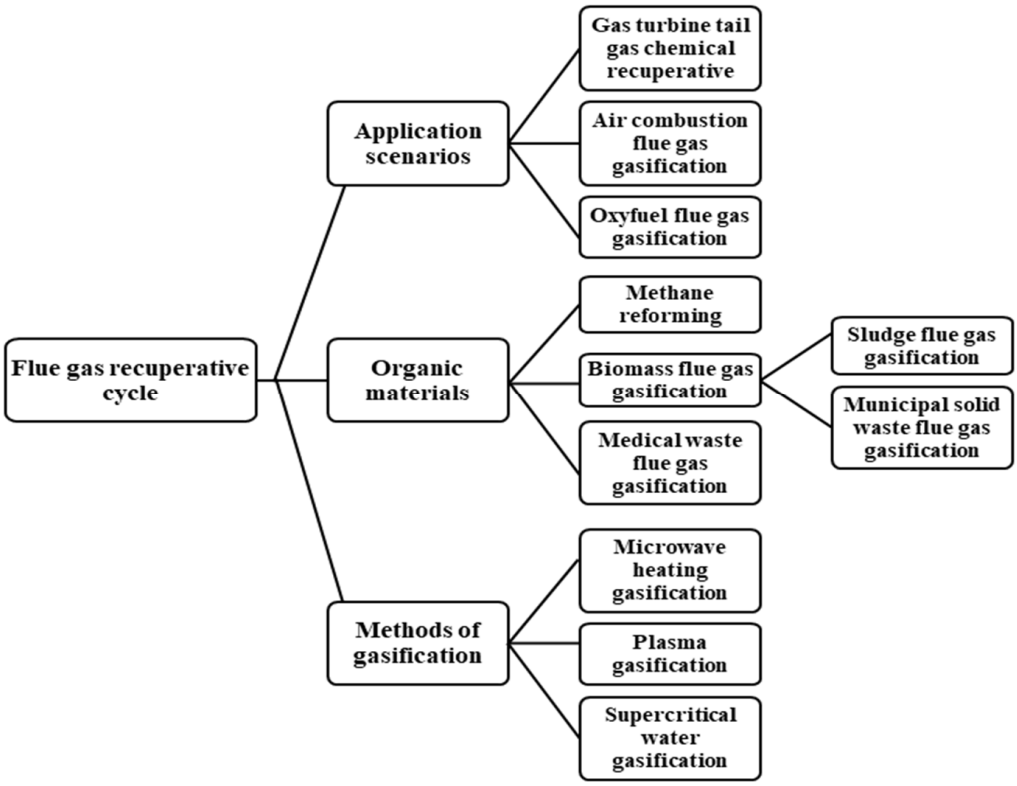


Figure 1. Different research areas of the flue gas recuperative cycle.

2. Concept and Development History of the Flue Gas Recuperative Cycle

Flue gas recuperative cycle technology was firstly applied in waste heat utilization of gas turbine tail gas [8]. Later, with the deepening of the research, the flue gas recuperative cycle was extended to broad flue gas recuperative technology, such as direct gasification with flue gas [9] or the main components of the flue gas to carry out CO_2 gasification [10,11], steam gasification [6,12,13], biomass gasification [14], and so on. At present, the flue gas recuperative cycle mainly refers to the use of flue gas as a gasification agent for the (catalytic) gasification or reforming of organic matter, and the waste heat in the flue gas provides energy/partial energy for the reaction to generate syngas, CO, and other renewable gases, which can be utilized again as fuels or products, e.g., to provide energy for the energy power units.

We searched Web of Science with the theme of "flue gas gasification" and analyzed the search results in terms of keyword statistics, the number of publications, citation frequency and distribution of publications, the major research countries, and the cooperation between the major research countries. VOS viewer(1.6.18) software, which is an important visualization tool in the field of academic research and information analysis, is used to process the data obtained from the search, which can provide valuable references for research in related fields.

2.1. Statistics on Keyword Search Results

According to the keyword statistics, 3778 keywords were retrieved and 29 keywords appeared more than 40 times. Among them, the most common keyword besides "gasification" was "combustion", which appeared 185 times, followed by "biomass", appearing 156 times, and then by "pyrolysis", appearing 122 times. The search results show that the research in the field of flue gas gasification mainly focuses on studies of biomass gasification and pyrolysis. The keywords were clustered using VOS viewer software to generate the clustering view of keyword co-occurrence, as shown in Figure 2, to obtain the research hotspots of flue gas gasification in recent years.

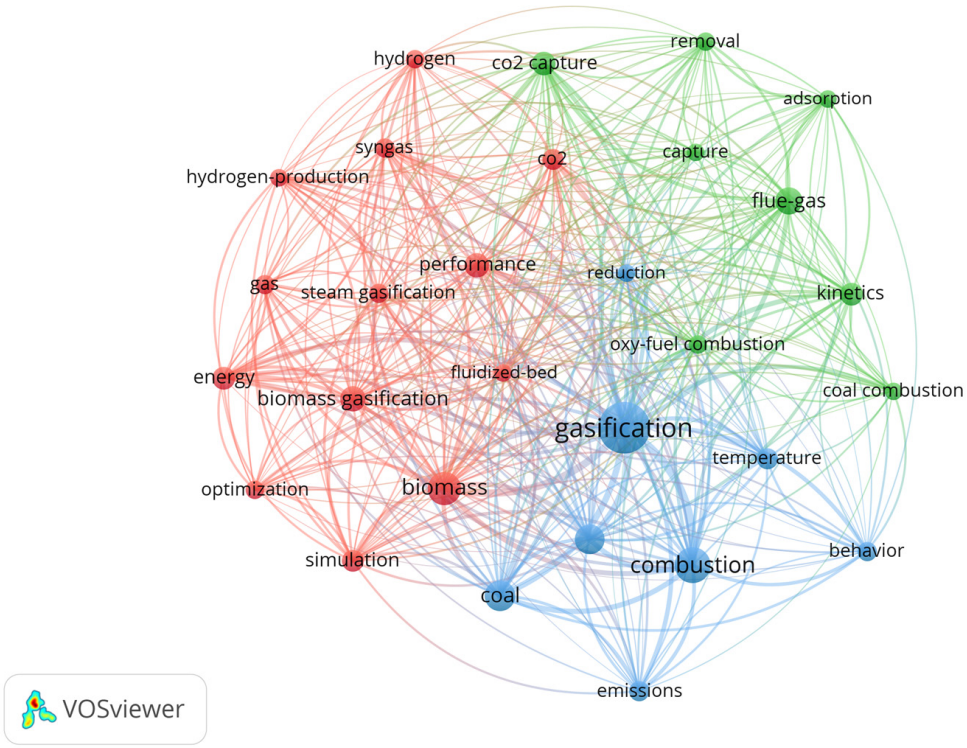


Figure 2. Clustered view of flue gas gasification keywords.

2.2. Statistics on the Number of Publications in Journals

The number of items of related literature published in each publication was counted. Relevant articles were published in 450 publications, of which the top 10 were selected. The results are presented in the form of a dendrogram, as shown in Figure 3. The results show that *Fuel* has the highest number of related literature published, with 151 articles, followed by *Energy*, with 83 articles, indicating that *Fuel* is more authoritative in this field.



Figure 3. Number of publications of related literature in various journals.

2.3. Statistics on Citation Frequency and Distribution of Publications

Since the number of publications and the number of citations can help scholars reveal whether the field is a research hotspot and a development trend, the citation frequency and the distribution of publications in this field are plotted by year (1996–2024), as shown in Figure 4. It can be seen from the figure that the number of publications in this field has increased significantly in 2011 and after, and the citation frequency of the literature has also increased significantly after 2013, which fully demonstrates that the field of flue gas gasification has a great potential for development.

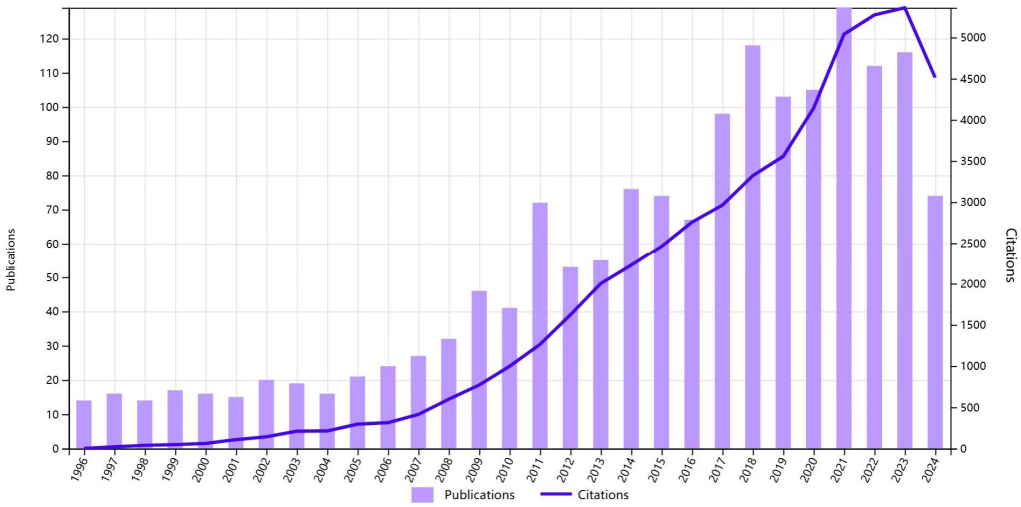


Figure 4. Distribution of citations and publications from 1996 to 2024.

2.4. Statistics on Major Research Countries and Cooperation Between Major Research Countries

In order to identify the main countries studying the related technologies and the cooperation between the main research countries, the national and regional distribution of the authors of the literature are analyzed. There are a total of 81 countries and regions with literature published in this field; the top 20 countries and regions with the highest amount of literature have been selected for statistics, and the results are shown in Figure 5. The results show that China has the largest amount of literature in this field, and it is significantly higher than in other countries, which indicates that China's research in this field is more extensive and in-depth, followed by the United States, Spain, and other countries. In terms of cooperation, as shown in Figure 6, China has exchanges and cooperation with

several major research countries, especially the United States, Sweden, France, Japan and the United Kingdom. Among them, China has the highest amount of cooperation with the United States.

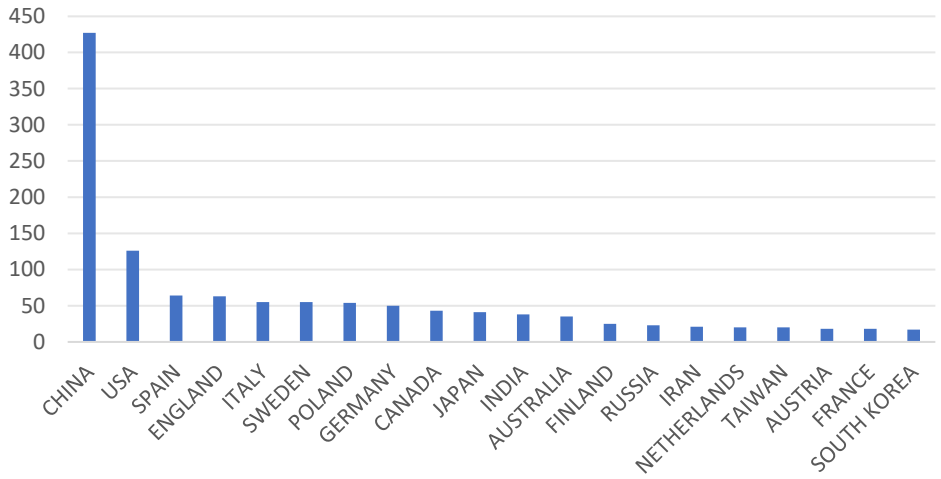


Figure 5. Top 20 countries or regions in terms of documentation.



Figure 6. Cooperation between China and major research countries.

3. Research Progress on Flue Gas Recuperative Cycle

3.1. Different Application Scenarios of the Flue Gas Recuperative Cycle

3.1.1. Chemical Recuperative Cycle with Gas Turbine High-Temperature Tail Gas

Gas turbines have the advantages of high stand-alone power, small size, and fast start-up [15], but low thermal efficiency and high pollutant emissions at low operating conditions. In order to compensate for these shortcomings, Olmsted and Grimes [8] in 1972 for the first time proposed the waste heat recovery of high-temperature gas from a simple cycle combustion turbine by means of a chemical reaction, i.e., the chemical recuperative cycle. Numerous subsequent studies have shown that compared with other cycles, it not only significantly improves the power and cycle efficiency of the device [16–19], but also significantly reduces the emission of pollutants such as NO_x [20], while attenuating the greenhouse effect, which is very promising for research.

The chemical recuperative gas turbine (CRGT) proposed based on the chemical recuperative cycle is an extension of the steam injection gas turbine (SIGT), which operates on the principle of using the exhaust heat from the tail gas to generate steam and heat the

steam reforming reaction of methane, where the steam is used in the methane reforming and the syngas produced by the reforming reaction is injected into the combustion chamber of the gas turbine for cycling. Figure 7 illustrates the CRGT concept in detail. Exhaust heat is first recovered in the reforming reactor to provide heat for the steam reforming reaction of methane, and then the exhaust gas is passed to a steam generator (SG) to recover the remaining heat and generate steam. The natural gas is mixed with the generated steam and fed into the reforming reactor, where the methane and steam undergo a reforming reaction and produce syngas, which is then injected into the combustion chamber to form a cycle. Studies have shown that the complete conversion of methane in this way can increase the effective fuel calorific value by about 30% [6].

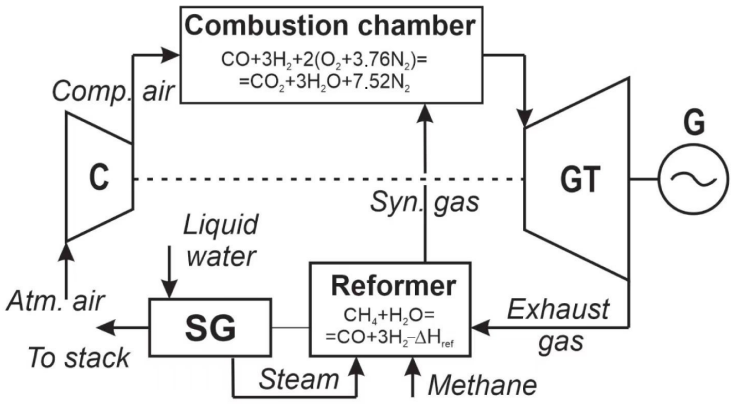


Figure 7. Chemically recuperated gas turbine concept [6].

More studies have been conducted in view of the superiority of the chemical recuperative cycle. Pashchenko et al. [6] found that the chemical recuperative cycle can replace the STIG. When the steam-to-methane ratio is the same, the efficiency of the CRGT is higher than that of the STIG. The efficiency of CRGT increases with the increase of reaction temperature. At a steam-to-methane ratio of 3 and a temperature of 1300 °C, the efficiencies of STIG and CRGT were 37.4% and 47.3%, respectively [6]. Pashchenko [12] studied a combined cycle power plant with a chemical recuperative gas turbine and found that the chemical recuperative cycle significantly increased the cycle efficiency of a high-temperature gas turbine [12]. In search of higher thermal efficiency, Pashchenko et al. [13] determined the effect of operating parameters (temperature, pressure, and steam-to-methane ratio) on the efficiency of the CRGT and found that up to 63% of the waste heat could be recovered [13]. In syngas production, Flórez-Orrego et al. [7] compared a conventional syngas production process with four CRGT-based processes and found that the new process could achieve higher temperatures and higher conversion rates than the conventional process with lower drive power, and could also provide the power needed for air compression and other auxiliary systems such as an air separation unit, a carbon capture system, a boiler feed pump, and recompressors. The average specific energy destruction and fuel consumption of the new process were reduced by 12.0% and 2.7%, respectively, and CO_2 emissions were reduced by 25% [20].

In order to expand the scope of application of the chemical recuperative cycle, many scholars have focused on the fuels of chemical recuperative gas turbines, including methane, dimethyl ether (DME), biomass ethanol, methanol, and other fuels, while in recent years many scholars have believed that ammonia, which has the characteristics of high energy density and ease of transportation and storage, is a major direction for the future low-carbon energy industry [21]. Therefore, many studies have been carried out on the use of ammonia as a fuel. Table 1 shows the fuel types and research results of some scholars.

Table 1. First author, fuel types, and findings of selected studies.

First Author	Fuel Types	Research Results
Nakagaki Takao [22,23]	DME	Compared to the STIC cycle, the CRGT cycle based on DME-steam reforming reaction has 10% less fuel consumption and less than 10 ppm NO_x emissions [22,23].
Cocco Daniele [24]	Methane, DME	The only difference between using DME as fuel and using natural gas as fuel is the increase in CO_2 emissions [24].
H Pak [25]	Biomass ethanol	The thermal efficiency of a CRGT cycle fueled by biomass ethanol can be increased by 16% compared to a simple cycle [25].
Abdallah H [26,27]	Methane, methanol	Methanol can reach 56.37% thermal efficiency when used as a fuel [26,27].
Christophe Duwig [28]	Methanol	Methanol-fueled micro-CRGTs can increase thermal efficiency by 5% at design operating conditions [28].
Vittorio Tola [29]	Renewable methanol	Combining an electric methanol storage system with a CRGT plant, the system can produce about 350 kg/h of methanol and generate about 600–750 kW of electricity [29].
Su Bosheng [30]	Ammonia	Ammonia-fueled CRGTs, which use liquid ammonia gasification as well as ammonia cracking to absorb waste heat from the flue gas, can achieve thermal efficiencies of 35.48–36.09% [30].
Yang Ren [31]	Ammonia	Ammonia-fueled CRGTs are capable of achieving thermal efficiencies of 40.4% to 50.5% in the chemical equilibrium reaction state in the range of 12 to 26 MW output power, and 37.8% to 46.6% in the presence of a Ru1-based catalyst [31].
Zhang Shijie [32]	Ammonia	The power generation efficiency of ammonia-fueled micro-CRGT is about 28.8% to 41.7% within the range of studied parameters, which is about 1.87% to 5.61% higher than the micro-CRGT cycle efficiency, with higher temperature ratios and more pronounced enhancements [32].
Gou Chenhua [33]	Methanol	Methanol-fueled CRGTs, which use methanol cracking to absorb the low-temperature heat of the pressurizer exit air, can achieve thermal efficiencies of up to 57.9% [33].

In addition, some scholars have also attempted to recover the high-temperature waste heat in the flue gas of gas turbines with different heat absorption reactions. Pan et al. [34] utilized the diesel-steam reforming reaction to recover the waste heat, and Figure 8 demonstrates the main flow of this configuration (MR-CRGT) [35]. Pashchenko et al. [35] analyzed the recovery of high-temperature waste heat from flue gas by ammonia decomposition through Aspen HYSYS, and the results showed a waste heat recovery of up to 43% [35].

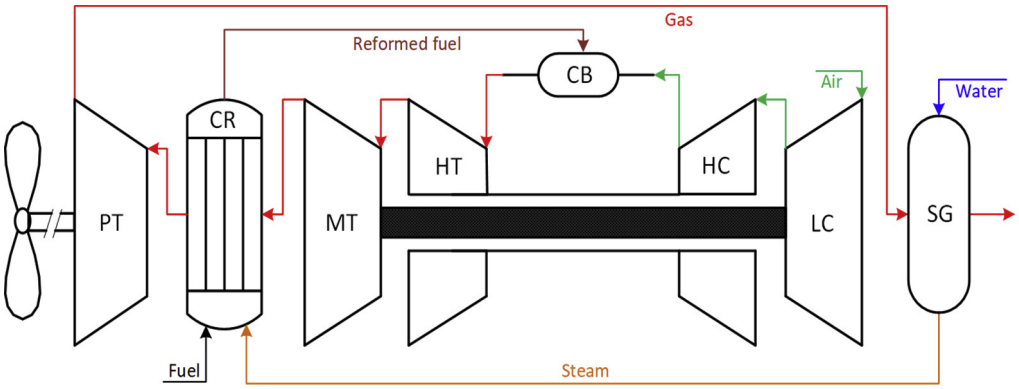
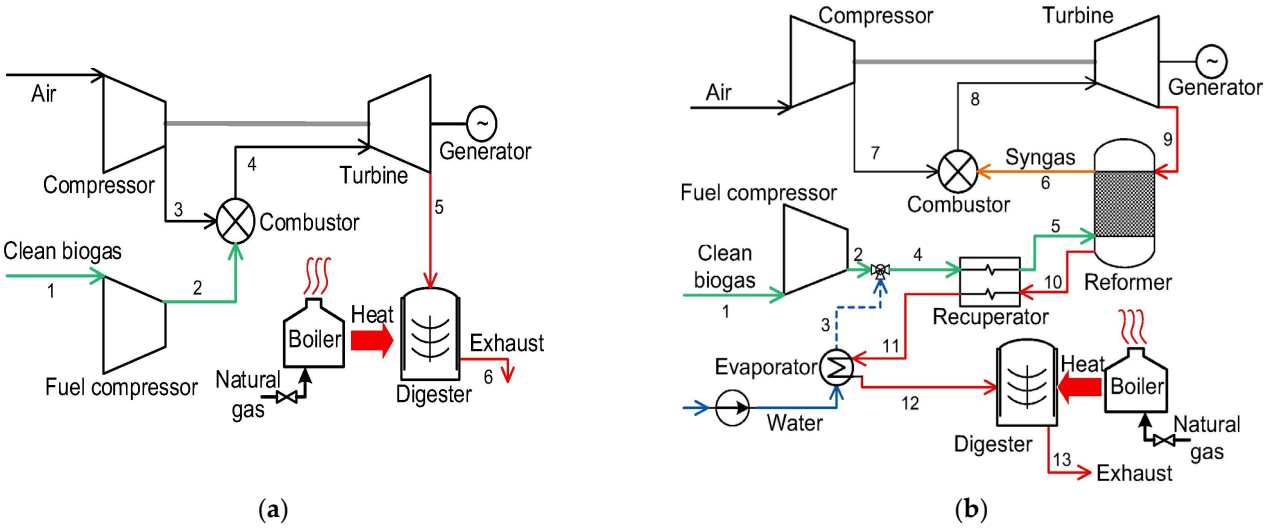


Figure 8. Schematic diagram of the MR-CRGT [34].

With the expansion of biogas use, Su et al. [36] proposed a biogas CHP based on a chemically recovered gas turbine cycle, and Figure 9 demonstrates the process comparison between this process and the conventional biogas CHP. In the new process, the gas turbine flue gas is first used for steam reforming of biogas, then for heating the biogas-steam mixture, then for turning water into steam, and finally for the holding digester. Compared with the conventional technology, wherein the exhaust gas is directly used in the holding digester, this technology achieves an efficient use of chemical and physical energy [36]. Luo et al. [37] proposed a new co-generation system combining a CRGT with a multi-effect thermal vapor compression desalination system, and thus used for both power generation and desalination of seawater, which is capable of generating power and desalinating seawater at the same time [37].



(**a**) (**b**)

Figure 9. (**a**) Flow chart of conventional biogas CHP system; (**b**) flow chart of biogas CHP system based on chemical recovery gas turbine cycle [36].

In addition to the aforementioned, there are many other ways to use the waste heat of a gas turbine. Tian et al. [38] proposed a tri-generation application using gas turbine flue gas fueled by methane. The co-generation system, which contains a helium turbine power plant, a regenerative steam Rankine cycle, a domestic hot water plant, and a multi-effect desalination subsystem, is modeled in Figure 10. The results show that the optimal values of the electrical power and the thermal load are 507.34 kW and 261.94 kW, respectively, through the multi-objective optimization of power and thermal load. The freshwater capacity obtained was about 0.076 kg/s [38]. Xue et al. [39] proposed a supercritical water gasification combined cycle power generation system based on the chemical recuperative cycle with supercritical water gasification technology as the core, using gas turbine high-temperature exhaust to provide reaction heat for gasification, coupling the gasification reaction with the power cycle through a thermochemical process. The results showed that the net power generation efficiency and the efficiency of the new system reached 53.37% and 52.11%, respectively, which are 6.24% and 6.09% higher than those of the reference system [39].

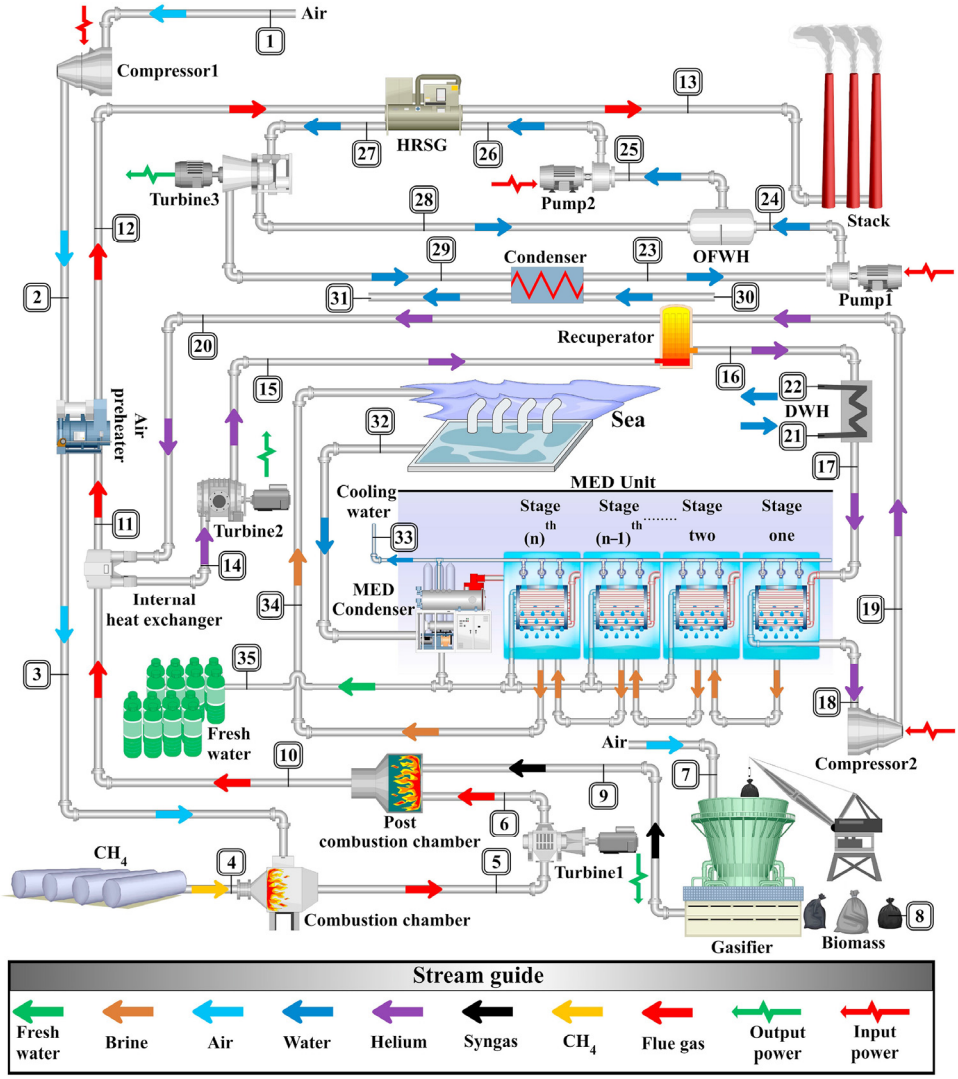


Figure 10. The proposed tri-generation model using methane fuel and biomass feedstock [38].

3.1.2. Gasification with Air Combustion Flue Gas

Air combustion flue gas is flue gas emitted from devices that utilize air for combustion. The chemical recuperative gas turbine is a typical application scenario of gasification using air combustion flue gas, which has been separately mentioned in the previous section due to its pioneering nature. The flue gas of a coal-fired power plant (CFPP) is also a typical representative of air combustion flue gas, and the use of the flue gas for organic matter gasification is one of the main focuses of research.

With the deterioration of climate, coal-fired power plants, one of the main sources of CO_2 emissions, need to be carbon-neutralized. At the same time, dry reforming of methane (DRM) and tri-reforming of methane both need a large amount of CO_2, so many scholars have considered combining the two. Chein et al. [40] used CFPP flue gas to produce syngas by tri-reforming and carbon gasification (CG) [40]. Minutillo et al. [41] used the syngas generated from the flue gas by tri-reforming for the synthesis of methanol [41]. Ouyang et al. [10,11] captured CO_2 from the flue gas, and the energy for the process was provided by the waste heat from the flue gas. The captured CO_2 was then used for DRM. The results showed that the innovative design could reduce 445,132.2 tons of CO_2 per year [10,11].

Qin et al. [9] proposed a new approach in which gasification of municipal solid waste (MSW) is carried out with cement kiln flue gas, and the main flow of this process is shown

in Figure 11. The results of the study show that the process reduces carbon emissions by 38.59% compared to conventional air gasification using flue gas and air as gasification agents. The total net CO_2 reduction was 172.11 kg equivalent CO_2/ton clinker [9].

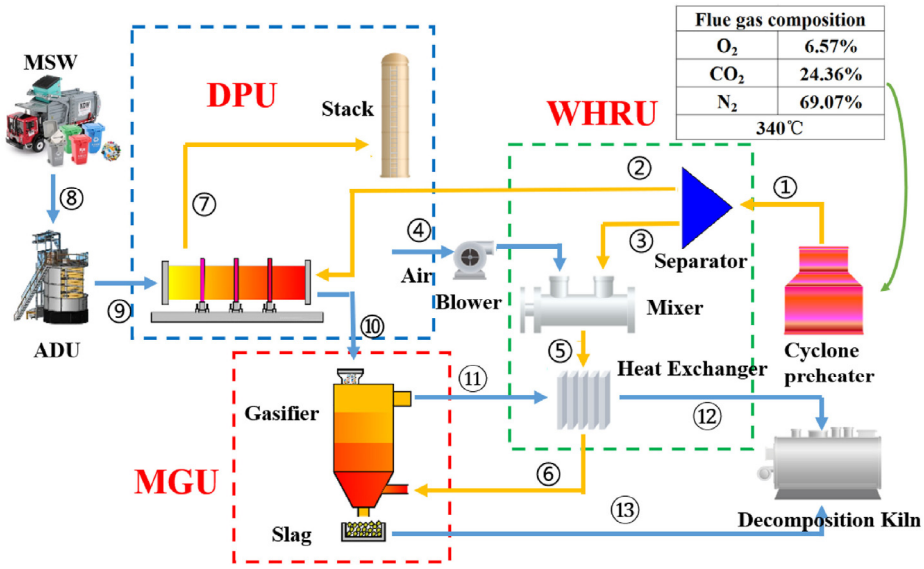


Figure 11. Schematic diagram of the stack gas–air MSW gasification and waste heat recovery system [9].

Qin et al. [42] later proposed a process for heat recovery of blast furnace gas (BFG) and gasification of MSW using the flue gas emitted from the blast furnace (BF), as shown in Figure 12. This technology, which is simple and easy to implement, can reduce CO emissions, and the CO_2 in BFG can be decarbonized and used as a fuel to be burned into BF again; it can also prevent the solid waste in MSW from entering into BF. However, the parameters and technical implications of this process are currently unknown [42].

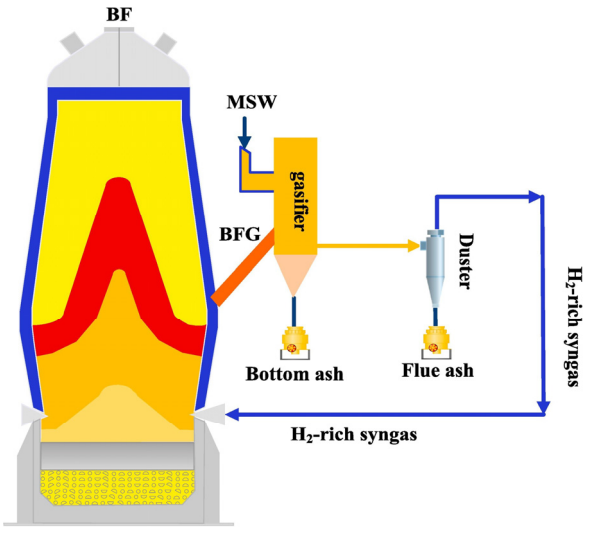


Figure 12. A process of MSW gasification by recycling hot BFG [42].

3.1.3. Gasification with Oxy-Fuel Combustion Flue Gas

Oxy-fuel combustion is often used in glass melting because of the high flame temperature, which improves combustion efficiency. The flue gas of oxy-fuel combustion is very useful for chemical recuperative cycles due to its simple flue gas composition. In recent years, Dong et al. [43] verified through simulation that the high-temperature flue

gas from oxy-fuel combustion of natural gas in a glass melting furnace can be utilized as waste heat in a multistage process [43], which significantly reduces the construction cost and is more energy-efficient and environmentally friendly than the traditional oxy-fuel combustion technology. Oxy-fuel combustion is an important means of carbon capture [44], and Tan [14] used the flue gas from oxy-fuel combustion as a gasification agent to gasify biomass to produce syngas. This technology combines carbon capture with the biomass integrated gasification combined cycle (BIGCC) and can achieve negative CO_2 emissions. Figure 13 describes the construction of the BIGCC.

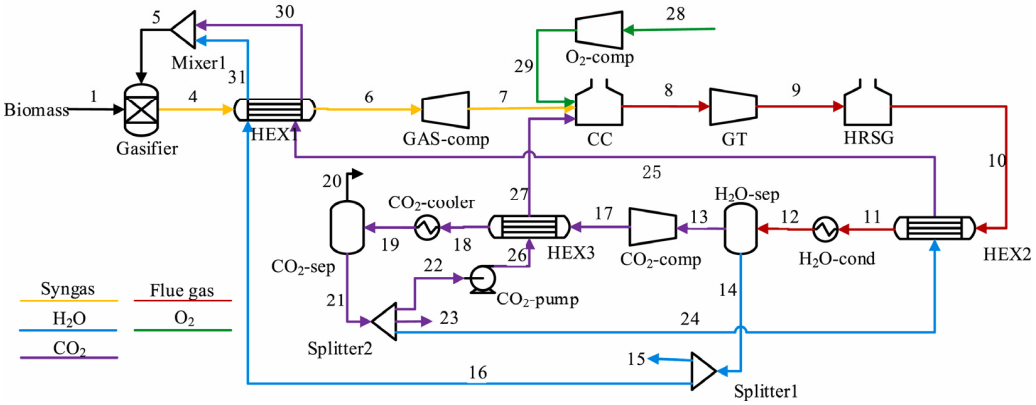


Figure 13. Configuration of complete BIGCC [14].

The results showed that the CO concentration in the syngas increased and then decreased when CO_2/C was increased from 0.2 to 1.0, and reached the maximum value at CO_2/C = 0.6. The H_2 concentration continued to decrease, and the CO_2 concentration continued to increase. The maximum efficiency of 29.1% was achieved at CO_2/C values, pressure ratios, and inlet temperatures of 1.0, 20, and 1150 °C, respectively [14].

3.2. Chemical Recuperative Cycle with Disposal of Different Organic Materials

3.2.1. Methane Reforming

Since methane reforming is a heat-absorbing reaction, many scholars have thought of utilizing the waste heat in the flue gas to supply heat for the reforming reactions. Methane reforming is capable of producing syngas on a large scale and is less harmful to the environment. Conventional reforming methods mainly include steam methane reforming (SMR) [45], partial oxidation methane reforming (POM) [46], dry reforming of methane [47], and bi- and tri-reforming of methane [48,49]. In addition to the traditional methods, the use of unconventional methods such as plasma reforming has also emerged and has achieved better results [50].

This section is presented in terms of the different types of reactions including DRM, bi-reforming, and tri-reforming of methane.

Dry Reforming of Methane

Dry reforming of methane has good prospects for application because of the advantages of wide source of feedstock, reduction of greenhouse effect, reduction of methane consumption, and suitability of the product as feedstock for Fischer–Tropsch synthesis [51].

Due to the effect of DRM emission reduction, Christoph Müller et al. [52] proposed a process combining DRM with CO_2 capture of flue gas that allows for the full conversion of the CO_2 captured and the selective production of a synthesis gas [52]. Ouyang et al. [10,11] captured CO_2 from the flue gas and the energy for the process was provided by the waste heat from the flue gas. The captured CO_2 was then used for DRM.

Bi-Reforming of Methane

Bi-reforming is the combination of SMR and DRM [53]. The main reaction is:

$$3CH_4 + 2H_2O + CO_2 = 4CO + 8H_2 \quad (1)$$

The advantage of the process is that it eliminates the need for separate units or reactors [48].

Bi-reforming is often used in the waste heat recovery of oxy-fuel flue gas, because the main components of oxy-fuel combustion are CO_2 and H_2O. Gaber et al. [54,55] investigated the situation of methane after bi-reforming using a fixed ratio of H_2O and CO_2 in pure oxygen combustion off-gas, in which a large amount of carbon deposition is formed, leading to a continuous decrease in methane conversion, and the addition of oxygen for tri-reforming significantly increased the methane conversion rate [54,55].

Tri-Reforming of Methane

Tri-reforming is the combination of the endothermic SMR and DRM with the exothermic POM [56]. The addition of oxygen not only reduces the total enthalpy of the reaction but also helps to prevent the formation of carbon deposits [57]. The main reactions are:

$$CH_4 + 1/2H_2O + 1/4CO_2 + 1/8O_2 = 5/4CO + 5/2H_2 \quad (2)$$

Tri-reforming is very promising, and some related studied have been introduced [40,41,55,56]. In addition, Zhang et al. [58] used tri-reforming to recover the waste heat in the flue gas. The syngas generated was then used for the synthesis of methanol. Dwivedi et al. [59] further improved and optimized the process. Zhang et al. [60] also developed an integrated process for the production of dimethyl ether (DME) using tri-reforming. The results showed that the process could make good use of CO_2 in flue gas [60]. Yang et al. [61] proposed a novel coal-to-ethylene glycol process incorporating methane tri-reforming technology, which was optimized for low carbon emission and high efficiency [61].

3.2.2. Flue Gas Gasification of Biomass

As mentioned at the beginning of the article, the flue gas recuperative cycle extends to the broader flue gas recuperative technology, such as direct gasification with flue gas or the main components of the flue gas to carry out CO_2 gasification, steam gasification, CO_2–CH_4 reforming, biomass gasification, and so on. This section mainly introduces the research on the gasification of biomass using flue gas or the main components of the flue gas such as carbon dioxide and steam.

Biomass gasification is the process of gasification and cracking of biomass feedstock after simple treatment to obtain combustible gas and purification to obtain product gas [62]. Compared with conventional fossil fuels, biomass has higher activity and is more suitable for gasification because it consists of cellulose, hemicellulose, lignin, etc., with high oxygen and volatile content and high activation of coke [63], and its gasification is more environmentally friendly due to its low sulfur and nitrogen content [64].

Some scholars have studied the effects of the ratios between different components of flue gas and characteristics such as temperature on the gasification results. Liu et al. [65] investigated the effects of flue gas flow rate and temperature, $CO_2/H_2O/O_2$ ratio, and other factors on the gasification characteristics of biochar (a pyrolysis product of biomass feedstocks such as wood, rice straw, husk, etc.) at atmospheric pressure, and the results showed that (1) the CO_2 conversion rate could reach >90% when the temperature of flue gas was greater than or equal to 1200 °C, and (2) the 5% oxygen ratio in the mixture of CO_2/H_2O

promotes the gasification reaction to some extent [66]. Shang et al. [66] investigated the gasification characteristics of single-rod biomass in a hot $CO_2/H_2O/O_2$ atmosphere [66]. Mauerhofer et al. [67] conducted CO_2 gasification experiments using cork in a 100 kW dual-fluid steam gasification bed and showed that as the gasification steam/CO_2 ratio increased from 100/0 to 32/68 vol.%, the CO content of the resulting syngas increased, the H_2 content decreased, and the CH_4 decreased slightly [67].

Some scholars have also investigated the gasification characteristics of different types of biomass. Zhang et al. [68] investigated the CO_2 gasification reforming of biomass components (cellulose, xylan, lignin, and starch) using a two-stage fixed-bed reactor, which produces a CO-rich syngas, and the results showed that lignin had the lowest gas yield (5.7 mmol/g biomass), and cellulose had the highest gas yield (14.6 mmol/g biomass) [68]. The kinetic analysis of the whole CO_2 gasification process of corn stover was investigated by Wang et al. [69] through thermogravimetry and Fourier-transform infrared spectroscopy (TG-FTIR) [69].

Some studies on co-gasification of flue gas have also been carried out. Liu et al. [70] used sawdust, PE, and PVC for CO_2 co-gasification, and studied gasification properties by thermogravimetric experiments. The results showed that PVC improved the gasification reactivity of sawdust, while PE improved the quality of syngas [70]. Sinisalu et al. [71] investigated the co-gasification characteristics of biomass and oil shale under CO_2 atmosphere, and found that the co-gasification could increase the CO and CH_4 content in the products [71].

Furthermore, two types of biomass, sludge as well as municipal solid waste, have been selected to study the progress of research on flue gas gasification of biomass.

Flue Gas Gasification of Sludge

Sludge gasification technology has its unique advantages: the high-temperature environment of sludge gasification reaction can effectively destroy pathogens; organic components in sludge are converted into combustible gas, and the remaining part is converted into residue, with obvious capacity reduction effect; the heat generated by the gasification process can be recycled. Compared with incineration, the gas produced by sludge gasification can be directly used to generate electricity; the energy utilization efficiency of the sludge gasification process is higher without additional fuel; sulfur dioxide, nitrogen oxides, and other harmful gasification emissions are low; and the cost required to control emissions is less [72]. The disadvantage of sludge gasification technology is the high tar and dust content in the gas produced, which can adversely affect the pipelines and equipment behind [73].

Figure 14 shows the frequency of citations and publication distribution statistics of the literature on sewage sludge gasification from 1999 to 2024, and it can be seen that sludge gasification has held research popularity in recent years.

In the utilization of flue gas for sewage sludge gasification, Weng et al. [74] investigated the gasification characteristics of sewage sludge under six different flue gas atmospheres. The experiments were carried out in a fixed-bed downward suction gasifier at 950 °C with a gas flow rate of 300 mL/min. The combustible gas yield was in the range of 247.8–417.3 L/kg of sludge. The calorific value of the produced gas was in the range of 2881.8–6617.3 kJ/m^3. The gasification efficiency was in the range of 46.43–74.56% [74]. Ziółkowski et al. [75] simulated a system for gasification of sewage sludge using flue gas from a gas turbine and estimated the performance of a power plant fueled by syngas produced from the gasification of sewage sludge [75].

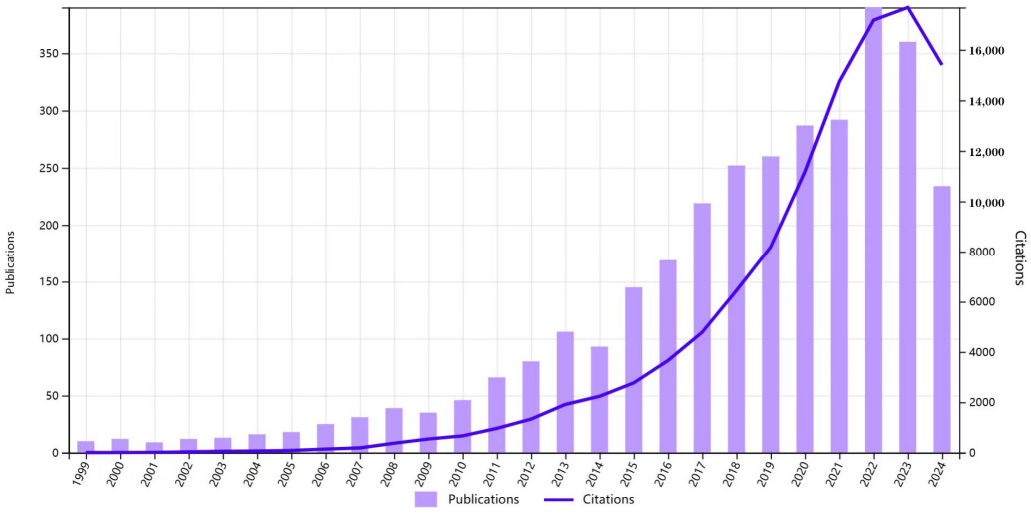


Figure 14. Distribution of citations and publications from 1999 to 2024.

Flue Gas Gasification of Municipal Solid Waste (MSW)

MSW refers to solid waste generated from the daily lives of urban residents or from activities that provide services for urban daily life. With the exponential rise in urban population, the need for effective and environmentally friendly disposal of MSW has increased dramatically [76].

In the utilization of flue gas for MSW gasification, Ge et al. [77] used high-temperature flue gas (HTFG) at 1200 °C as a heat source as well as a gasifier to gasify MSW, and investigated the effects of flow rate and composition of HTFG on the syngas yield and its energy share [77]. In a study by Zheng et al. [78], researchers investigated the gasification of MSW using a combination of steam and CO_2. They examined two crucial variables: the gasification temperature, which ranged from 1000 to 1100 °C, and the CO_2/steam ratio, which varied between 0.5 and 3. Their findings revealed that as the CO_2/steam ratio increased from 0.5 to 2.5, there was a corresponding rise in both H_2 and CO molar yields. Additionally, this increase in ratio led to improved CO_2 conversion efficiency. When the CO_2/steam ratio = 2.5 and the temperature was 1000 °C, the CO_2 conversion increased to 50.22%, at which time the maximum H_2 molar yield was 18.82 mol/kg MSW [78]. Vinoth Thangarasu et al. [79] investigated the gasification of MSW from a Brazilian city in air, CO_2, and air/CO_2 (70/30 vol%) atmosphere kinetics, and found that mixed air/CO_2 gasification improved combustion performance and reduced gasification temperature, at which time MSW exhibited better combustion performance, suggesting that an air/CO_2 atmosphere may be the best choice for MSW gasification [79]. Ge et al. [80] also investigated the effect of the contact mode between high-temperature CO_2 and MSW pyrolysis products, coke and volatiles, on the gasification results, in order to understand the relevant reaction behavior and provide guidelines for reactor design [80].

Flue Gas Gasification of Medical Waste (MW)

Due to its special characteristics of carrying viruses, MW is very likely to cause harm to people's health and the spread and transmission of diseases, which leads to a great increase in the difficulty of its treatment. Moreover, with the outbreak of COVID-19, the treatment of medical waste has become a focus of social development [81]. With the development of science and technology, high-temperature plasma gasification technology, microwave treatment technology, and other emerging technologies are able to effectively treat medical waste.

Regarding MW flue gasification, Li et al. [82] proposed a hybrid design solution combining hydrothermal treatment of sludge and plasma gasification of MW with waste heat

recovery in a cement plant to improve the waste-to-energy process [82]. Li et al. [83] also proposed another integrated system that combines plasma gasification for MW treatment, combined cycle power generation, and sludge pyrolysis. MW is first incinerated, and then the syngas from plasma gasification powers a gas turbine to generate electricity in this integrated system. The exhaust gas from the gas turbine heats steam and feedwater from the HRSG to achieve a combined gas–steam cycle. At the same time, the turbine exhaust gas powers the sludge pyrolysis, thus efficiently converting MW to electricity in an environmentally friendly manner. The results of the study show that these methods have excellent potential for development [83].

4. New Gasification Methods for Chemical Recuperation

Self-heating gasification refers to the reaction by flue gas's own exothermic heat or heat that it carries. Since the purpose of flue gas gasification is to recover the residual heat in the flue gas by a heat absorption reaction, this is the main method of flue gas gasification. The flue gas gasification mentioned above basically uses this method, and the research by this method is also the most significant. However, this method has many disadvantages such as slow reaction, low product selectivity, etc. Therefore, microwave gasification, plasma gasification, supercritical water gasification, and other emerging gasification technologies were born. Table 2 lists some studies using microwave or plasma gasification.

Table 2. Different gasification methods, gasification agents, gasification raw materials, and their results.

First Author	Gasification Method	Gasifying Agent	Raw Materials	Gasification Results
PooyaLahijani [84]	Microwave heating gasification	CO_2	Biomass-derived coke	Up to 99% CO_2 conversion, with 13.7 times more CO produced in the microwave-driven reaction than in thermal gasification at 750 °C [84]
Nor Azlina Ahmad [85]		Steam, CO_2	Palm kernel shell	The optimal gasification was achieved at a steam/CO_2 ratio of 0.028 and a temperature of 850 °C, with CO_2 and oil palm shell conversions of 92% and 85%, respectively [85].
Peng Zhang [86]		CO_2	Plastics	More than 96% of hydrogen was converted to H_2 with a yield of 480 mmol/g plastic, while carbon and CO_2 conversion reached 70% and 53%, respectively [86].
Hu [87]		Steam, CO_2	Sewage sludge	At a steam/CO_2 volume ratio of 36, the maximum concentration of H_2 reaches 63.73 vol% (68.93 vol% for total H_2 and CO), and the maximum calorific value (HHV) of the syngas is about 10.51 MJ/Nm^3 [87].
Roni Mallick [88]	Plasma gasification	CO_2	RDF	Syngas with calorific values of 13.95 MJ/m^3 for H_2 (42.6 vol%) and CO (44.06 vol%) was obtained [88]
Jafar Fathi [89]		CO_2	Plastics	CO_2 conversion is 98.5%, and plastics (polypropylene) are almost completely converted [89]
Menglong Wang [90]		CO_2	Textile printing sludge	At a feed rate of 36 g/min and a CO_2 flow rate of 0.43 Nm^3/h, the carbon conversion gasification efficiency was 99.9%, the energy conversion efficiency was up to 71.8%, and the lower limit of the calorific value of the syngas-rich off-gas was 8.91 MJ/Nm^3 [90].

Table 2. *Cont.*

First Author	Gasification Method	Gasifying Agent	Raw Materials	Gasification Results
Roni Mallick [91]	Plasma gasification	CO_2	Electrical switching waste	At the highest flare power of 1.6 kW, the maximum syngas yield was 85% of H_2 (24.36 vol%) and CO (43.61 vol%), and the calorific value of the syngas was as high as 10.20 MJ/m^3 [91].
Jiarui Li [92]		Steam	MW	Process efficiency and process energy efficiency can reach up to 72.16% and 66.88%, respectively [92].
Dejan Cvetinović [93]		Steam	Sewage sludge	H_2 in the range of 65.5–66.2% and CO in the range of 7.80–10.60% [93]
Kexin Yin [94]		Steam	Sewage sludge, kitchen waste	The mole fractions of hydrogen and carbon monoxide in the syngas after co-gasification of municipal sludge and kitchen waste were 45.62% and 37.00%, respectively, at 2500 °C, with an SFR of 0.67 [94].

4.1. Microwave Gasification

Microwaves are electromagnetic waves with frequencies from 300 MHz to 3000 GHz [95]. Microwave heating is based on the principle that microwave irradiation induces dipolar rotation of polar molecules. The constantly fluctuating electric field generated by high-frequency microwaves induces successive alignment and realignment of the moving dipoles, which creates strong friction inside the material, and the resulting internal energy is dispersed in the form of heat, leading to heating of the material [96]. Thus, as shown in Figure 15, microwave heating occurs from the inside out. Compared to conventional heating methods, microwave heating is faster [97]. Secondly, it does not require contact, which minimizes processing time, reduces power consumption, and improves thermal diffusivity [98]. Microwave heating usually results in higher product yields [99]. Finally, microwave heating is more environmentally friendly [100,101]. Furthermore, the microwave-induced induced charge remains on the surface of the material (e.g., SiC, Fe), making it possible to increase the kinetic energy of some electrons, allowing them to jump out of the material. This is thought to be the formation of a spark or arc. However, at the microscopic level, these electrons are actually plasma with free radicals that can enhance chemical reactions [102,103]. Figure 16 reveals the reforming reaction of CH_4–CO_2 over Fe/SiC catalyst. As an excellent microwave-absorbing material with high thermal conductivity, SiC foam ceramics can realize the bulk heating of the catalyst and achieve a uniform temperature distribution, in addition to the plasma generation mentioned above, which enhances the reforming reaction, and the final conversion of CH_4 and CO_2 reaches 85%, whereas the conversion of the reactants is only about 20% in the conventional heating process [104].

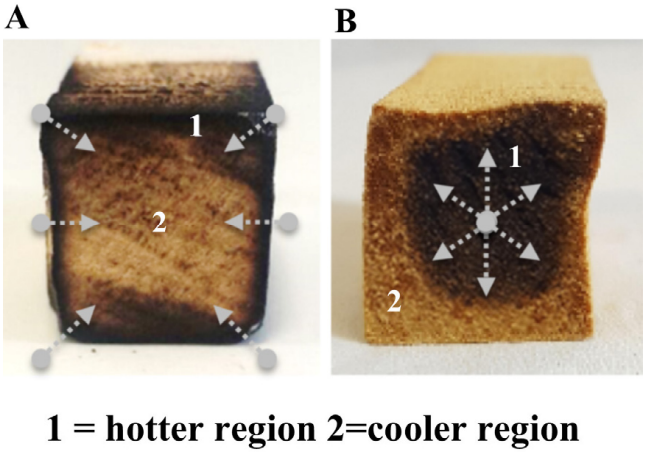


Figure 15. Difference in temperature distribution and direction of heat transfer between conventional heating and microwave heating. The arrows indicate the direction in which the heat is transferred. (**A**) Conventional heating: heat is transferred from the outside of the material inwards by conduction. Overheating can occur on the outside whist still maintaining a cooler inner region. (**B**) Microwave heating: MWs penetrate the material and heat volumetrically. Energy is then uniformly dissipated throughout the material [105].

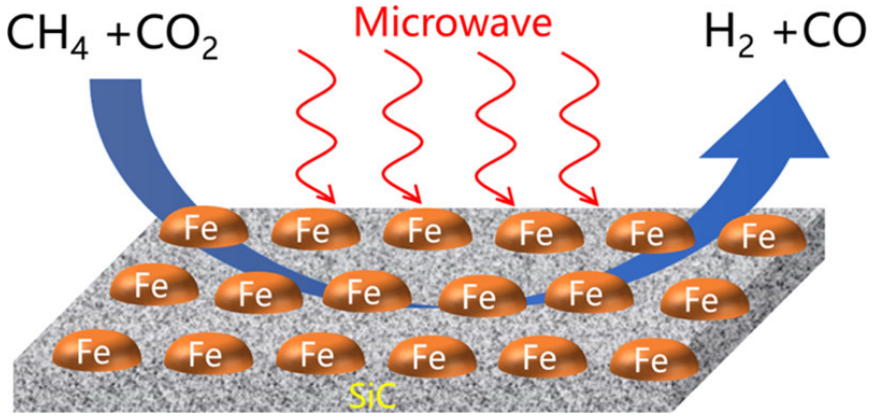


Figure 16. Schematic diagram of CH_4–CO_2 reforming reaction over Fe/SiC catalysts [104].

Table 2 shows some studies of microwave heating gasification.

Plastic, as a commonly used packaging material in modern society, is usually disposed of in landfills, but it often takes hundreds of years to degrade, which can be very harmful to the environment [106]. Zhang et al. [86] proposed a microwave-driven system for the gasification of plastic waste with CO_2. The plastic waste was decomposed into H_2 and solid carbon residues under microwave irradiation, which reacted with activated CO_2 gas to produce CO as the temperature increased. The results showed that the technique could produce H_2 and CO stably and continuously [86]. Hu et al. [87] investigated the microwave-assisted gasification of sewage sludge and the effect of gasification agents on the production of syngas in the microwave environment. The results showed that the mixed steam–CO_2 atmosphere helped to increase the syngas yield (to a maximum yield of 45.3 wt%) and improve the quality of the syngas, and Figure 17 demonstrates how H_2 was produced during the process [87].

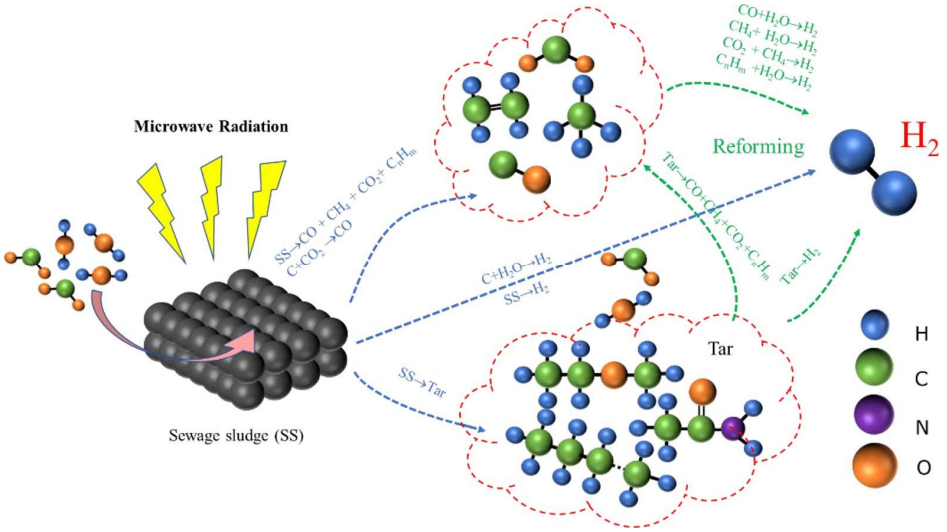


Figure 17. The process of H_2 generation in the steam–CO_2 mixed atmosphere [87].

4.2. Plasma Gasification

Plasma is an ionized gaseous substance consisting of positive and negative ions produced by the ionization of atoms and groups of atoms after some of them have been deprived of electrons. Plasma is currently used for dry reforming of methane [107], decomposition of carbon dioxide [108], and hydrogenation of carbon dioxide [109]. The basic principle of plasma gasification is to produce syngas by using a highly ionized gaseous plasma to thermally destroy waste. Compared to conventional gasification methods, plasma gasification technology has a high conversion efficiency, which contributes to syngas production and material recovery [110]. Currently, plasma gasification is commonly used to treat wastes, and it is capable of converting wastes to syngas with low environmental impact [111]. Figure 18 shows the advantages of plasma gasification.

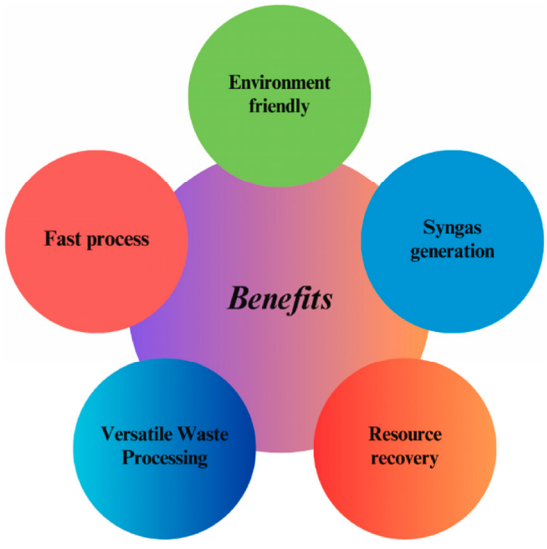


Figure 18. Advantages of plasma technologies [110].

Plasma gasification is very promising for the treatment of medical waste and sewage sludge. The studies on plasma gasification of medical waste conducted by Li et al. [82,83] were previously mentioned. In addition, Li et al. [92] proposed three methods for plasma gasification of medical waste to generate methanol or hydrogen using steam as a gasification agent [92]. In wastewater treatment, Cvetinović et al. [93] used plasma steam to gasify sewage sludge, determined the optimal parameters of the gasifier, and found that the

optimal gasification effect was achieved when the temperature was 1200K and the steam-to-fuel ratio was 0.1 [93]. Yin et al. [94] proposed a model for the plasma co-gasification of sludge and kitchen wastes using steam as a gasification agent, and used experimental data to validate the model [94].

4.3. Supercritical Water Gasification

In the biomass gasification process, the biomass must be dried first, leading to larger energy consumption, while supercritical water gasification (SCWG), as an emerging gasification technology, can directly gasify wet biomass without drying. The reaction efficiency, as well as the reaction rate, is higher compared to normal gasification [112]. One of the current research focuses of supercritical water gasification technology is the treatment of sewage sludge [113–116]. In addition, supercritical water gasification technology can also be used for gasification of coal; the coal can be efficiently converted into clean syngas, and the pollutants are discharged in the form of solid residues. As for the disadvantages of traditional gasification methods, such as low energy utilization efficiency, supercritical water coal gasification technology can solve these problems [117].

Many studies in recent years have combined supercritical water gasification with the flue gas recuperative cycle. Xue et al. [39] proposed a supercritical water–gas combined cycle power generation system based on the chemical recuperative cycle. Chen et al. [118] also proposed that water can be preheated to a supercritical state by using the heat in the flue gas from a gas turbine [118]. Wei et al. [119] proposed a near-zero-emission CO_2 power generation system that integrates the critical water gasification of coal with a solid oxide fuel cell (SOFC). In the gas turbine's combustion chamber, the SOFC anode's exhaust gas undergoes combustion with pure oxygen. This process generates a high-temperature flue gas composed of CO_2 and H_2O. The heat in the flue gas, as well as the heat generated by the SOFC, is used to preheat the feedwater and drive the gasification reaction [119].

In addition to supercritical water gasification, supercritical CO_2 gasification is also one of the hot spots in current research. This technology provides the possibility of consuming CO_2. Guan et al. [120] used supercritical water and supercritical CO_2 to gasify polyethylene terephthalate (PET) microplastics [120]. Wei et al. [121] used supercritical CO_2 to gasify diphenylmethane, and found that the carbon gasification efficiency (CE) and the hydrogen gasification efficiency (HE) at 700 °C for 40 min were 25.99% and 21.97%, respectively, with CO reaching a maximum value of 15.64 ± 0.16 mol/kg [121].

5. Prospects and Challenges

The concept of the flue gas recuperative cycle was first applied to the flue gas of gas turbines, which is essentially a conversion of energy, i.e., the use of a gasification reaction to convert the heat energy in the flue gas into the chemical energy in the reaction product (syngas). Later, the concept was gradually expanded; in addition to flue gas of natural gas turbines, flue gas of coal-fired power plants, cement kilns, and blast furnaces can be used for the flue gas recuperative cycle, and the recovery reaction is not limited to the SMR. Bi- and tri-reforming, biomass, as well as sewage sludge, municipal solid waste, and medical waste can be involved in the gasification reaction to recover heat. The flue gas chemical recuperative cycle can also be applied in combination with CO_2 capture. With the increasing demand for energy efficiency and greenhouse gas emission reduction, flue gas recuperative cycle technology will undoubtedly have a broader application space.

However, there are still some defects in the current research, such as a lack of overall research on the flue gas recuperative cycle, a lack of depth in that research, and a lack of theoretical analysis, especially the analysis of the impact of the introduction of the chemical recuperative cycle on the overall process energy efficiency and emission reduction of the

system, so it is necessary to carry out further research on these aspects. In addition, most of the flue gas gasification is now autothermal gasification, which has a slow reaction and low product selectivity, and there are fewer studies on gasification using emerging gasification methods such as microwave gasification, plasma gasification, supercritical water gasification, etc.; more of these studies will need to be carried out.

Beyond examining various technologies, it is crucial to evaluate their economic viability and performance thorough techno-economic studies. Nonetheless, the comprehensive utilization of flue gas chemical recuperative technology holds immense promise for enhancing the efficiency of energy conversion and utilization system. By delving deeper into the mechanisms underlying the promotion of the flue gas chemical recuperative cycle, further exploration of the feasibility and potential of this technology becomes feasible. This exploration could result in the development of cleaner and more efficient solutions for energy utilization and environmental protection, thereby steering the energy industry towards a more sustainable trajectory.

All in all, the flue gas chemical recuperative cycle is a very promising power cycle, which can greatly improve thermal efficiency and reduce CO_2 emissions, and is one of the key development directions for the low-carbon industry.

6. Conclusions

This paper reviews recent advancements in the flue gas recuperative cycle, including its systems and fundamental principles, with a focus on its application in energy utilization. Our study offers a thorough analysis of how the flue gas chemical recuperative cycle enhances system efficiency through a series of existing literature. We also discuss strategies for integrating this cycle into energy systems to explore more diverse applications. Key performance indicators, such as efficiency, stability, and cost, are identified. Future developments and challenges are examined, providing guidance for implementing stable and reliable production systems.

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