Green Energy and Fuel Research

Review

Chemical Looping Gasification of Plastics toward Hydrogen-Rich Syngas: A Review of Oxygen Carrier Design and Process Optimization

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Abstract: In the dual pursuit of achieving the global goal of carbon neutrality and curbing plastic pollution, chemical looping gasification (CLG) technology has emerged a key approach to integrate plastic waste recycling with H_2-rich syngas production. This review systematically examines and compares the design strategy (support properties, active metal modification and bimetallic synergy) of oxygen carrier (OC) and the optimization progress of key process parameters (temperature, reactor staging and co-gasification of biomass) in the production of H_2-rich syngas from plastic CLG. The findings demonstrate that support optimization and metal synergy can significantly regulate the syngas yield and its H_2/CO ratio, while improving the cycle stability of OCs. Temperature and segmented CLG significantly improves the conversion rate and achieve flexible control of the H_2/CO ratio. CLG of biomass and plastics can improve the yield and quality of H_2-rich synthesis due to the complementary hydrocarbon and the coordinated conversion of tar. However, most studies at present use model plastics as fuel, which highlights the key shortcomings in understanding actual plastic waste, impurities, long-term CLG stability, and systematic economic evaluation. This review aims to provide guidance for the development of stable and selective OC systems and the optimization of CLG systems to achieve sustainable energy conversion of plastic waste.

Keywords: chemical looping gasification; plastic; oxygen carrier; process parameter; hydrogen-rich syngas

1. Introduction

Plastic is an indispensable material that is commonly used in modern industry and human daily life. Since the middle of the 20th century, global plastic production has grown rapidly due to its light weight, high strength, low cost, and easy processing of plastics [1]. In 2018, the total number of global plastic waste reached approximately 359 million tons, with China contributing nearly 30% of this volume. Alarmingly, about 12 billion tons of plastic waste are projected be landfilled by 2050 [2,3]. Traditional treatment methods such as landfill and incineration have many disadvantages, including land occupation, microplastic pollution, and the release of toxic substances-for example, dioxins produced by PVC incineration. These methods create a long-term threat to ecosystems and human health [4,5]. Therefore, developing efficient and green waste plastic conversion technology for transforming plastic into high-value products is of great significance to solve environmental pollution and realize resource recycling and sustainable development.

Among various waste treatment technologies, thermochemical conversion, particularly gasification, is considered an effective method for achieving the high-value utilization of waste plastics due to its broad adaptability to different raw materials and high product value [6–9]. As illustrated in Figure 1a, in the presence of of gasifiers (such as air, water vapor, or oxygen), waste plastic is converted into syngas at high temperature, which is mainly composed of hydrogen and carbon monoxide [10,11]. As a clean energy source, the syngas can play a key role in energy decarbonization and reducing industrial emissions [12]. It is especially suitable for green energy fields such as transportation, energy storage, and high-end manufacturing [13,14]. Therefore, utilizing waste plastics to produce hydrogen-rich syngas can be an effective solution to address the problems of waste management and green energy production at the same time [15].

Nevertheless, conventional gasification faces several major challenges. The process usually requires high temperatures (above 700 °C, sometimes even up to 1200–1500 °C), resulting in high energy consumption [16].

Moreover, when air is used as a gasifier, the introduced nitrogen dilutes the syngas, and may lead to the generation of pollutants such as nitrogen oxides (NO_x) [17]. Although the use of pure oxygen can improve the quality of syngas, it will greatly increase the operating cost.

As an innovative approach to fuel conversion, chemical looping gasification technology (CLG) provides a promising solution to meet these challenges [18]. This technology uses a recyclable OC to provide oxygen and divide the traditional single-step gasification process into two independent reaction stages: the fuel stage and the air stage (Figure 1b). In the fuel reactor, the OC provides lattice oxygen for partial oxidation of feedstocks, producing syngas rich in H_2 and CO. Meanwhile, the OC itself is reduced. The reduced OC is then regenerated by introducing air into the air reactor. Because the feedstocks in the CLG process do not directly contact the air, the resulting syngas will not be diluted by nitrogen, leading to higher purity and simplifying downstream utilization. Furthermore, the designed metal-based catalyst can also catalyze tar cracking, adsorb CO_2, or capture pollutants such as chlorine, achieving in-situ purification during gasification. Therefore, the CLG technology shows great potential to improve energy efficiency, reduce the generation of pollutants, and improve syngas quality [19]. For high-hydrogen plastics, CLG technology can effectively promote the conversion of plastic to produce H_2-rich syngas with various yield and composition through controlling key process parameters (such as temperature) and selecting different OCs (Table 1). However, compared to the extensive biomass CLG investigation, a systematic summary and perspective focusing on plastic is still lacking.

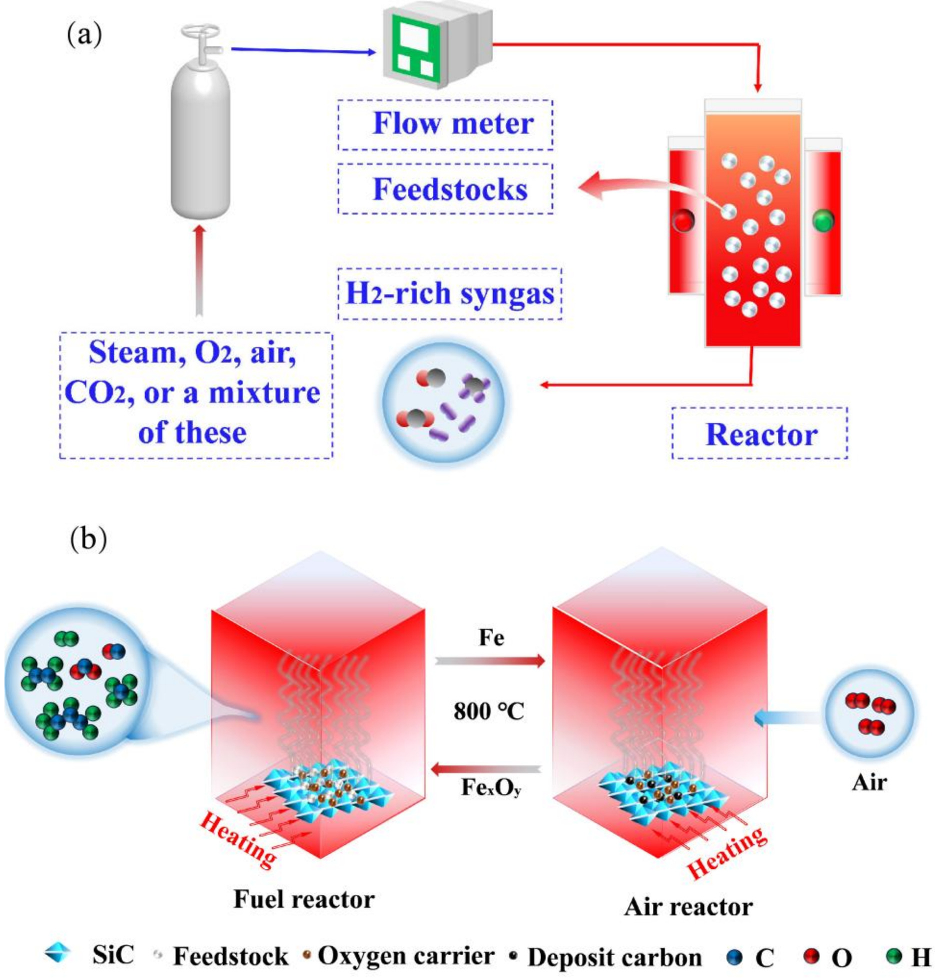


Figure 1. (**a**): The diagram for traditional gasification (**b**) and chemical looping gasification [20]. "Reprinted with permission from Ref. [20]. 2025, Zhou, J.M".

Table 1. Plastic CLG under different conditions.

Fuel	Temperature (℃)	OC	Syngas Yield	H_2 Yield	Ref.
HDPE	800	Ni-Fe-Al	75.32 [a]	47.09 [a]	[20]
Disposable masks	Stage 1: 600 Stage 2: 800 Stage 3: 800	Fe1/Al2	129.89 [a]	95.20 [a]	[21]

Table 1. *Cont.*

Fuel	Temperature (°C)	OC	Syngas Yield	H_2 Yield	Ref.
PP	Stage 1: 500 Stage 2: 800	NiO/Al_2O_3	143 [a]	/	[22]
PP	900	$NiFe_{20}O_x$	81.3 [a]	46.0 [a]	[23]
HDPE	850	$(FeNiCOCrMn)_3O_4$	69.36 [a]	52.44 [a]	[24]
PP	Stage 1: 600 Stage 2: 200 Stage 3: 850	Ni-$LaFeO_3$	168.4 [a]	98.2 [a]	[25]
PE, PP, PS	600–950	Bauxite+waste tire	188.66 [a]	63.33 [a]	[26]
PE + corn stalk	800	$CuO/NiFe_2O_4$	62.9 [a]	28.1 [a]	[27]
LDPE + pine	600	Ni-Fe-Ca/H-Al	/	53.2 [a]	[28]
PE + pine	850	CaO/Fe_2O_3	/	71.0 [a]	[29]
PE + Alfalfa	950	Iron Ore	57.09 [a]	/	[30]
Disposable face mask	Stage 1: 600 Stage 2: 600 Stage 3: 850	Fe-Ca-Al	177.89 [a]	127.93 [a]	[31]

Note: Stage 1—pyrolysis stage; Stage 2—steam stage; Stage 3—air stage; [a] mmol/g. In order to facilitate the quantitative comparison of various studies, all gas yield data in this table have been normalized and expressed in "mmol/g". Conversion of the molar volume of 22.4 L/mol of the ideal gas under standard conditions: (1) For the data initially expressed as Nm^3/kg, the conversion coefficient of 1 $Nm^3/kg \approx 44.6$ mmol/g is used; (2) For the data originally expressed in mL/g, the conversion coefficient of 100 mL/g ≈ 4.46 mmol/g is adopted.

The OC is the core of CLG technology, directly determining plastic conversion efficiency and syngas quality [32,33]. An ideal OC should have the following properties: high reactive activity, excellent redox cycle stability, and resistance to carbon deposition. To date, various OC types have been proposed, including iron, nickel, copper, cobalt and natural ores-based OC [34–36]. Performance improvement is mainly achieved through strategies such as support optimization and metal doping.

Currently, research on CLG has primarily focused on biomass, with numerous reviews and detailed reports on the development of biomass-based CLG [37,38]. However, there is still a lack of systematic integration and critical evaluation of the growing research results in the field of plastic CLG, which primarily utilizes model plastics (such as polyethylene and polypropylene) as raw materials. It is crucial to fully integrate these studies, which helps to provide suggestions for OC developments and react system optimization toward H_2-rich syngas production. The review, therefore, illustrates the design strategy of OC, including selection of inert support, active metal doping, and composite structure design. Meanwhile, the influence of key operating parameters, including temperature, reactor configuration, and co-feedstock on the conversion process is also be analyzed (Figure 2). Through examining these studies' progress, this review also highlights the critical research gaps and practical challenges that must be addressed to drive the industrial application of plastic CLG. In addition, this review also pays attention to the key issues of data comparability between studies (e.g., through standardized presentation in Table 1). Finally, we summarize the current research limitations and propose future directions for developing high-performance, low-cost, and deactivation-resistant OCs. This review aims to provide a theoretical reference and design guidance for combining waste plastic resource recovery with clean H_2-rich syngas production.

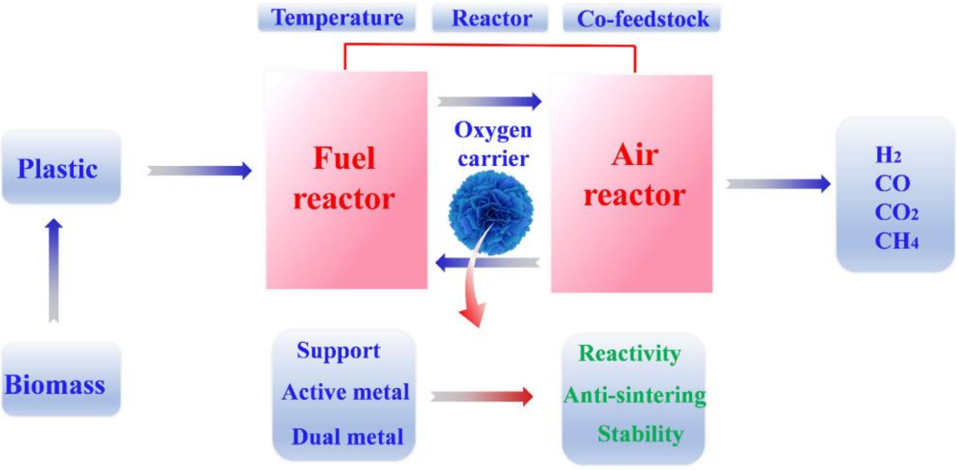


Figure 2. Schematic overview of chemical looping gasification of waste plastics for H_2 -rich syngas production.

2. Oxygen Carrier Design

2.1. Support of OC

In plastic CLG, the support of an OC plays a critical role for stabilizing the active metal. In addition, it also directly affects the syngas yield, hydrogen selectivity, and reaction stability by modifying its physicochemical properties [39]. Therefore, a systematic comparison of how different support materials affect OC performance is highly important for designing and optimizing OC aimed at H_2-rich syngas production.

Al_2O_3 is a typical inert support that is frequently used to load active metals like Fe and Ni [40]. There are a lot of studies proving the excellent performance of this carrier in biomass CLG. For example, Hu et al. [41] prepared Fe-based OC with different supports (Al_2O_3, TiO_2, SiO_2, ZrO_2) and used wheat straw as feedstock. The results demonstrated that the Fe_2O_3/Al_2O_3 OC performed best, achieving the highest gas yield of 36.6 mmol/g, H_2/CO ratio of 0.9, and carbon conversion rate of 64.9% among four tested OCs with different supports. The difference in performance is related to the structural properties of the OC. Al_2O_3 has a higher specific surface area (the specific surface area of Fe_2O_3/Al_2O_3 of 60% metal load is 4.78 m^2/g, and Fe_2O_3/SiO_2 under the same conditions is 0.677 m^2/g) and a suitable pore structure, which is conducive to the division of active components. This property is beneficial for the reforming and water-gas shift reactions, thus improving the syngas production. The basic principle outlined–that is, a carrier with a large surface area and appropriate porosity can promote the distribution of active sites and improve the critical gasification reaction–represents a general material design strategy. These strategies provide valuable guidance for the initial selection of OC in plastic CLG. However, due to the inherent differences between raw materials, it is necessary to be careful to extrapolate the quantitative performance data of biomass directly to plastics. Compared with biomass, plastics usually exhibit different pyrolysis behaviors (for example, melting properties, different volatile material release curves), potential impurities (e.g., chlorine), and different tar formation tendencies. These factors will significantly change the optimal metal-carrier ratio and reaction conditions, and ultimately affect the specific gas yield and conversion efficiency. Therefore, although the excellent performance of Al_2O_3 as an OC demonstrated in biomass CLG provide an important reference base, its performance must be specially verified and advanced in the plastic CLG.

On the above basis, Cai et al. [42] systematically compared the effects of four typical supports (Al_2O_3, SiO_2, TiO_2, and ZSM-5) on syngas production from polypropylene. Their fundings align with the previous trend, demonstrating that Fe/Al_2O_3 achieved the highest hydrogen yield (58.7 vol%). Recently, Ren et al. [21] further revealed the structural mechanism of Al_2O_3 in plastic CLG. They found that within a Fe-Al OC, Al_2O_3 does not only provide physical support but also participate in forming a $FeAl_2O_4$ spinel structure, which is beneficial for providing lattice oxygen.

Some studies have also shown that the surface properties, pore structure, and mechanical strength of OCs can be regulated by different precursors and synthetic routes used [43]. Different precursors will release different decomposition products and gases during heat treatment, which directly impact the pore formation (Figure 3), surface acidity and alkalinity of the OC [44]. All these factors are closely related to the reactive activity and durability of the OC, resulting in different H_2 yield (Figures 4 and 5). In addition, the method used to load active metals onto the support (such as wet impregnation, co-precipitation, sol-gel method) will also affect the performance of the OC. The fundamental insights from these synthetic optimizations-aimed at enhancing surface area, pore structure, and metal-support interaction-are universally relevant to OC design. Nonetheless, their effectiveness in mitigating plastic-specific challenges (e.g., coke deposition from polyolefins or chlorine poisoning) requires direct experimental confirmation within plastic CLG systems. The researches of waste plastic pyrolysis-catalytic process provide important data resources for the design of OCs for CLG. Although pyrolysis catalysis does not emphasize material recycling, its core is highly relevant to CLG, as the catalyst is used for steam or dry reforming. Yao et al. [45] found that the Ni/Al_2O_3 and Fe/Al_2O_3 catalysts prepared by sol-gel method have higher specific surface area (Figure 3), pore volume and metal dispersion than similar materials prepared by the impregnation method. These optimized structural properties come from the fact that the sol-gel method can mix precursors at the molecular level, thus promoting the high dispersion of active metals and the formation of strong metal-carrier interactions. The conclusion of the study–excellent texture and strong metal-carrier interaction is the key to obtaining high catalytic activity and selectivity–provides direct guidance for the design of OC.

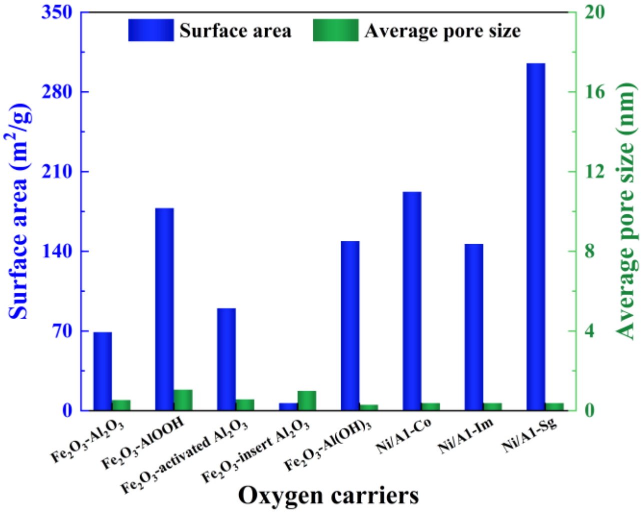


Figure 3. BET results of OCs with different support (Co: co-precipitation; Im: impregnation; Sg: sol-gel method) [44,45].

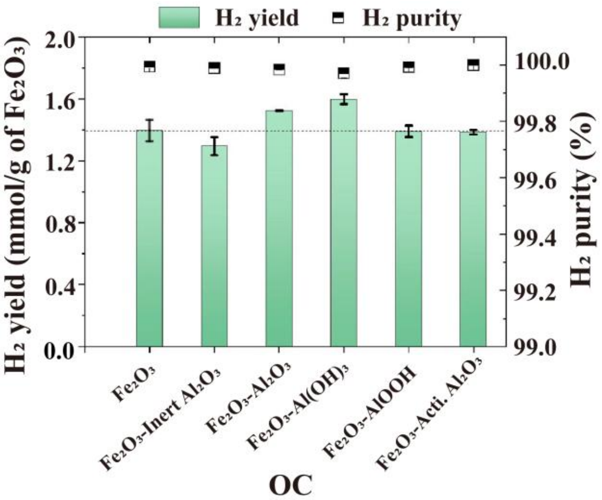


Figure 4. The H_2 yield results of OCs with different support precursor (Acti.: activated) [44]. “Reprinted with permission from Ref. [44]. 2024, Gao, Z.X”.

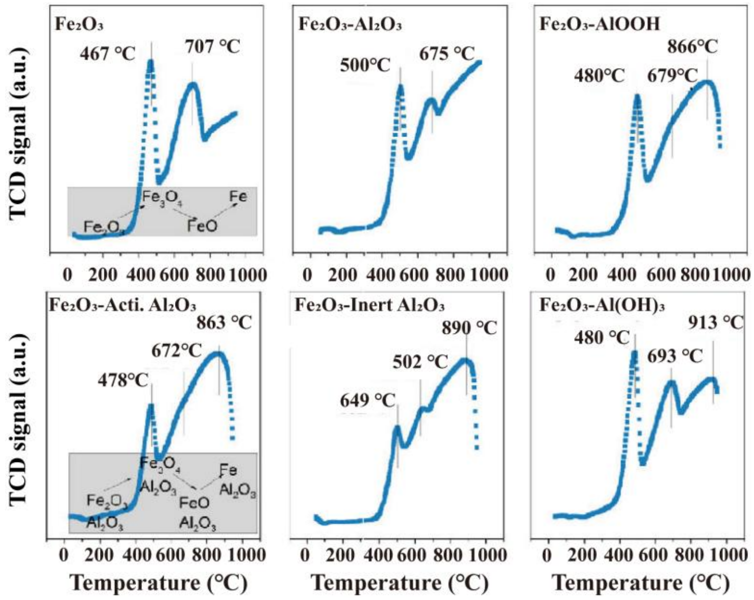


Figure 5. The H-TPO results of OCs with different support precursor [44]. “Reprinted with permission from Ref. [44]. 2024, Gao, Z.X”.

In short, the support affects metal dispersion and reaction transmission through its physical properties (such as specific surface area). The support can also significantly improve the cycle stability and reaction performance of the OC by forming a stable chemical structure (such as a spinel structure) with active metals. In addition to the inherent properties of the support material itself, the precursor form and the loading method of the active metal components will also affect their properties. For plastic CLG, the optimal support design must therefore not only adhere to these universal principles but also be specifically evaluated for its efficacy in handling the unique pyrolysis products, potential contaminants, and coking behaviors inherent to plastic feedstocks.

2.2. Metal Modification

In the process of CLG, the addition of active metals is one of the key methods for regulating the performance of OC and improving the yield and value of syngas. Among many metal types, Fe-based OCs have attracted wide attention from researchers owing to their advantages of low cost, environmental friendliness, and high activity. Furthermore, studies have shown that the proportion of Fe also has a significant impact on the OC properties and the resulting syngas composition.

Cai et al. [46] found that with the increase in the proportion of iron in Fe/Al_2O_3, the relative volume fraction of hydrogen in the syngas increased significantly. When the mass ratio of Fe to Al_2O_3 was enhance to 2:1, the volume fraction of H_2 reached the highest value (82%). At the same time, the decreasing trend of methane and ethylene was observed. These trends show that hydrocarbon cracking and water-gas transformation reaction were improved, effectively converting hydrocarbons into H_2 and CO.

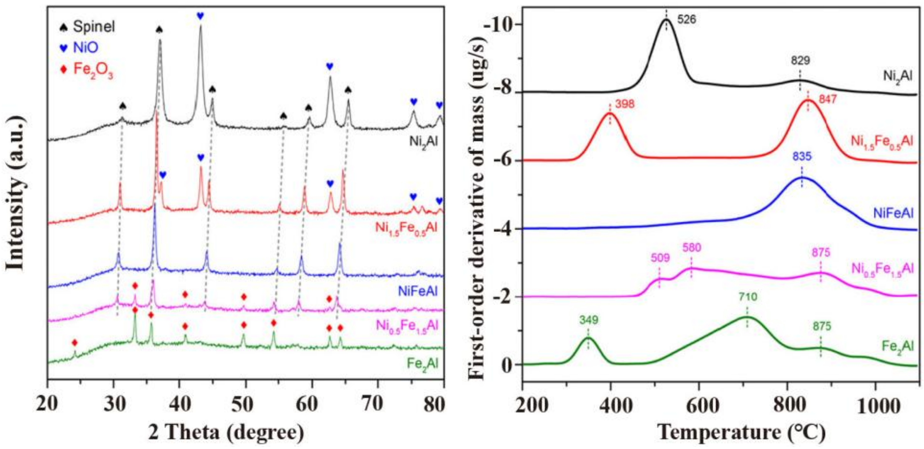
Ren et al. [21] studied the influence of different Fe/Al molar ratios on the properties of Fe-AlOCs, confirming the importance of the ratio of active metals to inert carriers for producing high-value-added syngas. They found that the Fe_1Al_2 (molar ratio of Fe/Al is 1:2) had the best performance, and the total syngas and H_2 yields reached 129.89 mmol/g plastic and 95.23 mmol/g plastic, respectively. This is because the formation of a stable Fe-Al spinel structure, which improves oxygen transfer and prevent sintering at high temperature.

Samprón et al. [47] conducted multiple cycle tests of OCs and found that high active metal loads (such as 25 wt% Fe_2O_3) will cause Fe cation migration and particle aggregation at high temperatures, thus accelerating the inactivation of OC. On the other hand, Fe_2O_3/Al_2O_3 with low iron load (such as 10 wt%) shows better structural durability and lasts longer in continuous tests. In addition, another study shows that the particle size of the OC is also a key factor in the CLG process. Liu et al. [48] prepared Fe/SiO_2 OCs with different particle sizes by the sol-gel method and applied them to polypropylene gasification. They found that OCs with large iron particles achieved the highest H_2 yield, reaching 25.60 mmol/g plastic. The relative content of H_2 in the syngas also increased to 50.30%. Nickel-based OCs are also often used in the study of CLG of plastics because of their high lattice oxygen activity and strong C-C and C-H bond fracture ability. Liu et al. [22] used NiO/Al_2O_3 as the dual-functional material of polypropylene CLG, achieving a syngas yield of 64.7 mmol/g-PP and a carbon conversion rate of 64.9% at 800°C, highlighting its efficiency and stability in plastic CLG. The applicability of other metal-based OCs (such as Ca, Mg, Cu, etc.) in plastic CLG remains to be systematically explored, despite their demonstrated efficiency in biomass CLG [49–51]. Therefore, future research should pay attention to expanding targeted OC evaluation for plastic CLG, with the focus on its high activity, selectivity, and strong resistance, promoting plastic CLG technology development.

2.3. Dual Metal Modification

Mono-metal OCs usually have certain limitations in activity and stability. Introducing a second metal to form a composite or doped structure can improve performance [52] (Figure 6). This approach can enhance OC's cyclic stability and selectivity of gas products.

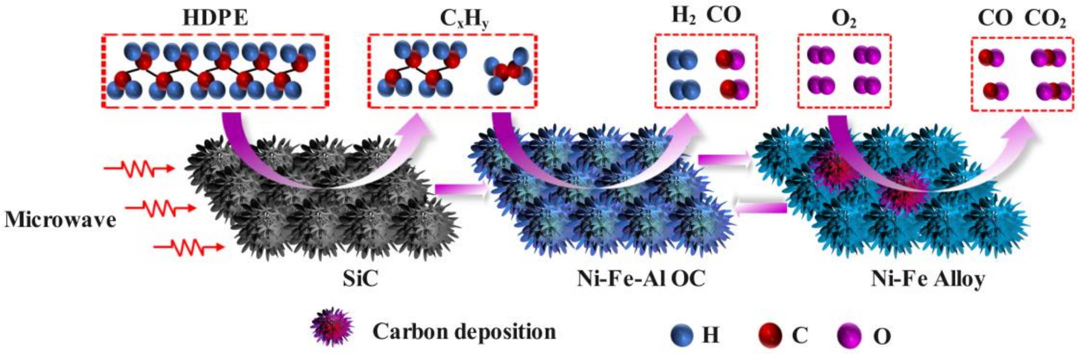
Ni-Fe OCs have demonstrated excellent ability in plastic CLG for producing H_2-rich syngas. Yao et al. [53] investigated the influence of Ni/Fe ratio on product selectivity. The results show that a Ni/Fe ratio of 1:3 yielded the highest hydrogen selectivity (73.93 vol%) and yield. During the reaction, Fe promotes deep dehydrogenation, while Ni promotes the reformation reactions. The combination of two active metals promotes the production of syngas with high H_2/CO ratio. In another study, Fu et al. [23] employed Ni-Fe OC to investigate the microwave-assisted CLG of PP. The experimental results prove that the addition of Ni not only improves the microwave absorption efficiency of OC but also create the synergy between Ni and Fe, which promotes the breakage of C-H bonds. Under optimized conditions (Ni/Fe = 1:20, microwave power 890 W), a high H_2 yield (46.0 mmol/g plastic) and concentration (56.5 vol%) were achieved. The above results not only prove the importance of the ratio between bimetals, but also reflect the great potential of combining energy supply mode.



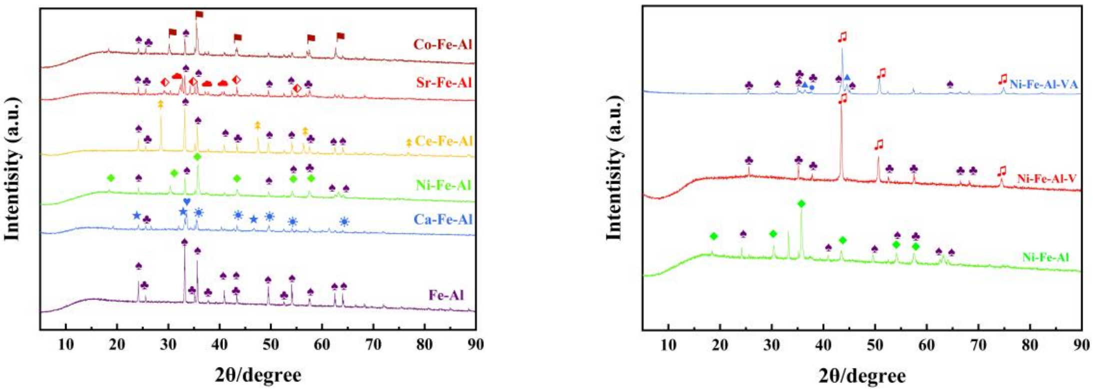
(**a**) XRD results (**b**) TPO results

Figure 6. XRD and TPO results of Ni-Fe redox catalysts [52]. "Reprinted with permission from Ref. [52]. 2018, Huang, J.J".

In addition to Fe/Ni OC, other bimetallic OC for CLG of plastics have been developed for plastic CLG. Zhou et al. [20] prepared a series of bimetallic OCs, comparing their performance in gasification process and proposed mechanisms (Figure 7a). In addition, their chemical structure differences were also analyzed in detail through XRD and other technologies. The results proved that the bimetallic-loaded OCs promote highly active crystal phases such as spinel phases, thus promoting the generation of syngas (Figure 7b,c). Falascino et al. [54] developed a Fe/Ti OC for the CLG process. This OC system tends to form $FeTiO_3$ phases with low oxidation, which effectively inhibits excessive oxidation and achieves high syngas purity (~95%). Xiao et al. [55] systematically compared four types of Fe-based OC-MFe_2O_4 (M = Co, Mn, Cu, Ni), providing systematic guidance for the OCs' design. They found that $CuFe_2O_4$ showed excellent CO_2 separation in the fuel reactor (>83%) while the $CoFe_2O_4$ and $MnFe_2O_4$ favored higher hydrogen in the steam reactor.



(**a**) Proposed mechanism for microwave-assisted chemical looping gasification of plastic using Ni-Fe-Al as OC.



(**b**) Fresh redox catalysts (**c**) Fresh and spent Ni-Fe-Al

Figure 7. (**a**) Proposed mechanism; (**b**,**c**) XRD results of OCs (♠: Fe_2O_3; ♣: Al_2O_3; ★: $Ca_2Fe_2O_5$; ♥: $CaFe_3O_5$; ☀: $CaAl_4FeO_{14}$; ◆: $NiFe_2O_4$; ‡: CeO; ◇: $SrAl_4O_7$; ☁: $SrFe_{10}O_{22}$; ⚑: $CoFe_2O_4$); ♫: Fe-Ni alloy; ●: $Fe_{0.98}O$; ▲:$FeAl_2O_4$) [20]. "Reprinted with permission from Ref. [20]. 2025, Zhou, J.M".

Chen et al. [56] prepared a Cu-Fe OC by doping CuO into Fe_2O_3-rich industrial waste red mud. The formed $CuFe_5O_8$ phase significantly enhanced the OC's reaction activity and oxygen transfer capacity. However, due to the decomposition of $CuFe_3O_8$ and the migration of Cu, the effect of the OC decayed after multiple cycles, thus reducing its efficiency. The above research highlights the challenges of maintaining the stability of the bimetallic OC. In order to address this problem, material design is developing in the direction of multi-component and high-entropy structures. For example, Wang et al. [24] introduced the concept of high entropy oxide to this field and synthesized a spinel-type five-component high entropy oxide $(FeNiCoCrMn)_3O_4$ for the CLG of high-density polyethylene. The high entropy induces lattice distortion and slow diffusion, stabilizing the material structure and promoting the formation of surface oxygen vacancy. After 20 cycles, the material maintained high hydrogen selectivity (75.6%) and hydrogen conversion efficiency (81.5%), opening up a new way for the design of next-generation OCs with high activity and excellent stability. It is important to note that real-world plastic waste presents additional challenges, such as chlorine content from PVC, which can release corrosive HCl and cause environmental concerns. Doping alkali/alkaline earth metals (e.g., K, Na, Ca) into the OC can achieve in situ chlorine deposition [57]. For the mixed plastic waste treatment technology, Yang et.al. [58] examined the chemical properties of the gasification process with iron nickel oxide ($NiFe_2O_4$) as OC in actual plastic and rubber compounds (PP, PE, SEBS). The results show that the carbon conversion rate, syngas yield and the H_2/CO ratio reached 96%, 142.8 mmol/g, and 2:1, respectively. The OC maintained high activity and structural stability in ten consecutive cycles, proving that $NiFe_2O_4$ has high efficiency and cyclic stability in the treatment of actual rubber and plastic waste. This provides important experimental support for promoting the bimetallic OC from material design to the energy application of actual waste.

In general, bimetallic modification is an effective strategy to achieve OC target optimization and functional enhancement. Future research should focus on an in-depth understanding of the relationship between the structure of a specific metal combination and its properties. In addition, optimizing the energy supply mode and the design concept of advanced materials with high entropy are also important for developing economical, efficient, and stable OCs. Furthermore, future research should also explore the construction of multifunctional OC and extend its application to more complex actual mixed waste systems, so as to promote the large-scale application of CLG toward actual waste.

2.4. Cycle Stability of Oxygen Carrier

The long-term cycle stability of OCs remains a key challenge in transitioning this technology from laboratory applications to industrial applications. Althoug OC stability has been widely discussed, there remains a lack of system data covering the number of long cycles (>50–100 cycles), comparative studies across different OC systems, and in-depth explanations of the inactivation mechanism. This section aims to integrate existing researches and clarify the structural evolution mode and performance decay mechanism of OCs during circulation.

In the long-term study, Samprón et al. [47] conducted experiments on the Fe_2O_3/Al_2O_3 OC, with the number of cycles as high as 300 redox cycles (900 °C). The results clearly showed that there was a positive correlation between active metals loading and the degradation rate: high Fe load ($Fe_{25}Al$, 25 Wt%) exhibited obvious iron migration and surface aggregation after 50 cycles, while the degradation rate of low load ($Fe_{20}Al$, 20 wt%) was slower. However, particle breakage and severe aggregation still occurred after 300 cycles. These findings demonstrate high active substances loading may accelerate structural destabilization, with continuous iron cation migration and particle agglomeration emerging as priamary inactivation routes. In the study of medium and short cycles, Ren et al. [21] reported that the Fe_1Al_2 maintained a stable H_2 and CO yield over seven cycles. However, the SEM and BET results showed that as the number of cycles increased, the material gradually became dense and the specific surface area decreased, indicating that sintering has occurred. XRD analysis further shows that with the increase of the number of cycles, the intensity of the Fe_2O_3 diffraction peak decreased, indicating that the reoxidation is incomplete, and the spinel structure has not been fully restored. The study also emphasizes that the introduction of inert supports (such as Al_2O_3) can effectively inhibit the shedding and aggregation of active components. For bimetallic/polymetallic systems, the inactivation mechanism is more complex. Chen et al. [56] conducted 10 cycle tests on the red mud-based copper ferrite carrier, revealing a unique inactivation pattern: $CuFe_5O_8$ solid solution "decomposed" during circulation, resulting in the loss of active phase. At the same time, Cu migrated significantly to the particle surface, while Fe remains relatively stable. Huang et al. [52] studied the NiFeAl OC and found that the reaction temperature had a critical impact on the stability in 10 cycles: in the optimization range of 750–850 °C, the H_2 yield remained stable or even increased slightly; however, at 900 °C, structural degradation led to obvious inactivation. This is mainly due to sintering and phase separation at high temperatures. It is worth noting that the latest progress in the field of material design provides a new way to improve OC's cycle stability. Wang et al. [24] introduced the concept of high entropy oxide (HEO) to CLG and synthesized $(FeNiCoCrMn)_3O_4$, which maintained excellent stability after 20 cycles. The syngas yield, H_2 conversion rate and H_2/CO ratio have not been significantly impacted. The characterization results of OC confirm that the material still maintains a single-phase spinel structure after circulation, the distribution of elements is uniform, and there is no phase separation. This stability is attributed to the lattice distortion and diffusion slowdown caused by the high entropy effect, thus effectively inhibiting metal migration. This method points out the direction for the design of OCs with both high activity and long service life.

The comprehensive analysis of existing research reveals two major challenges in understanding the stability of the OC cycle: (1) the mechanism research is still on the surface, and most of the work is limited to performance characterization and post-cycle analysis. The understanding of micro-mechanisms (such as sintering dynamics, element migration drivers and phase change paths), especially the in-situ evolution under actual reaction conditions, is still insufficient; (2) the researches mainly rely on model plastics or pure components, and there is a serious lack of evaluation of the long-term toxic effects of impurities (chlorine, sulfur, additives, etc.) in real waste plastics. Therefore, future research urgently needs to make breakthroughs in the following areas: carry out long-cycle (>50 Figure 100 cycles) stability tests of the system to establish benchmark test conditions; combine in situ/operation characterization technology (for example, in situ XRD, TEM) with theoretical calculation depth to clarify the structural dynamics and inactivation mechanism of OCs during circulation; focus on evaluating the impact of complex actual raw materials, and explore advanced material design (such as high entropy, nuclear shell structure, interface engineering) to fundamentally improve the anti-sintering, anti-migration and anti-poisoning ability of OCs.

3. Key Operation Parameter and Optimization

3.1. Reaction Temperature

Temperature is one of the most critical parameters in the plastic CLG. It directly affects the reaction kinetics, gas product distribution, carbon conversion rate, and the stability of OCs. In the traditional catalytic pyrolysis gasification and CLG processes, temperature has always been the core factor for optimizing process and regulating product, with an optimal range for effective conversion. Researches show that the effect of temperature on the CLG process presents a characteristic "volcanic curve pattern" [59], revealing a clear optimal temperature range. At a lower temperature (usually below 700 °C), polymer macromolecules underwent incompletely pyrolysis. The fracture of C-H and C-C bonds is limited by dynamics, resulting in low volatile substances yield, excessive heavy tar production. As the temperature increases within the optimal temperature range (usually 750–850 °C), the reaction kinetics improve significantly. Complete pyrolysis of polymers is accompanied by enhanced oxygen release capacity from the OC, which effectively promotes the catalytic cracking, tar reorganization, and long-chain hydrocarbons. For example, Jagodzińska et al. [60] investigated the catalytic pyrolysis of landfill waste using FeNi/Al as catalyst, and investigated product distribution at three different reaction temperatures of 700, 800, and 900 °C. The corresponding results show that the H_2-rich syngas yield reached its maximum value at 800 °C. However, when the temperature further increased to 900 °C, the catalyst underwent sintering, resulting in a decrease in activity and a decrease in the total gas yield. Huang et al. [61] used the NiFeAl OC to explore the effect on the distribution of syngas from polyolefin CLG at a temperature range of 700–900 °C. The heighted yield was obtained at 800 °C. However, as the temperature reached 900 °C, the activity of the OC and the gas yield decreased. In addition, XRD results indicate the phase separation of Ni and Fe occurred at 900 °C, which is one of the important reasons for OC inactivation. Similarly, Yang et al. [58] explored the CLG of plastic and rubber using $NiFe_2O_4$ as the OC. They found that temperature exceeding 750 °C led to the sintering of OC and the limited release of lattice oxygen, resulting in gas yield decrease. The above results once again highlight the importance of maintaining an optimal temperature window in CLG operation. The fundamental goal of temperature control is to achieve the best balance between reaction kinetics, target product selectivity and structural stability of OC. Appropriately increasing the reaction temperature can promote C-H bond breakage and improve syngas yield. Excessive temperature will lead to sintering, phase separation or inactivation of OC. Therefore, determining and maintaining the optimal temperature range (usually between 750 °C and 850 °C) is crucial to obtain the required product distribution and ensure the long-term cycle stability of the OC. In addition to optimizing operating parameters, the development of next-generation OC materials with excellent sintering resistance and wide temperature range activity is also a key way to further break through the limitations of CLG technology.

3.2. Reactor Development

In the process of CLG, the contact between the feestocks and the OC is a key factor affecting the gasification path, product distribution and carbonization behavior. Compared with general CLG, the segmented chemical looping gasification (SCLG) process spatially separates pyrolysis and gasification into multiple stages, thereby effectively optimizing the reaction path and improving the control level of the whole process (Figure 8).

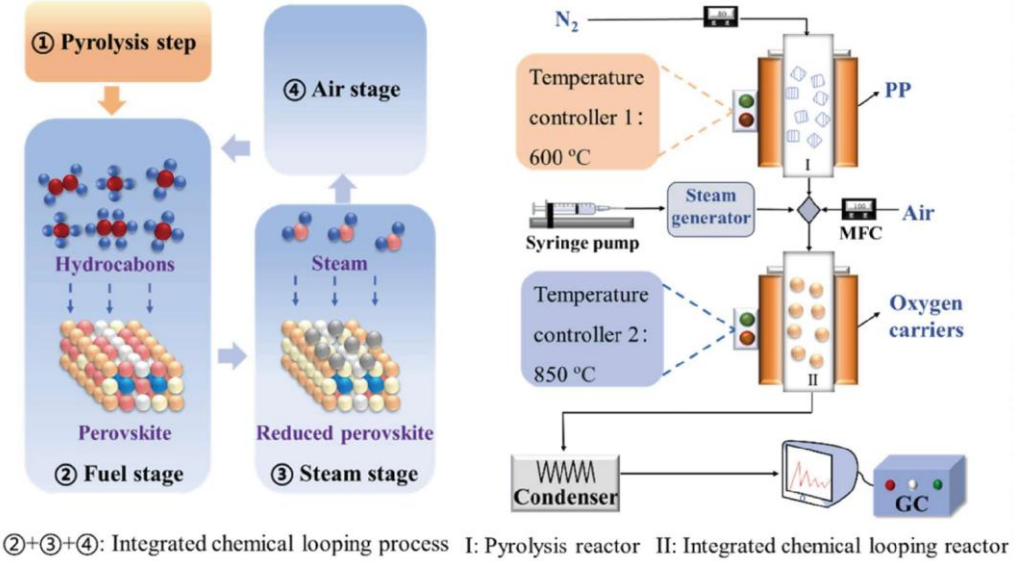


Figure 8. Schematic diagram of the SCLG process and experimental system [25]. "Reprinted with permission from Ref. [25]. 2026, Zhao, T.M".

Zhao et al. [25] have developed a graded process for plastic waste conversion. The core of this process lies in the grading design of the reactor: plastic raw materials are decomposed into volatiles at 600 °C in an independent cracking reactor. The generated volatiles were transported to the downstream reactor by the N_2 carrier gas and reacted with the OC at 850 °C (Table 2). This physical isolation design avoids direct contact between solid plastics and OC, fundamentally preventing serious carbon deposition and OC inactivation. Online gas monitoring clearly shows the function of each stage: in the fuel stage, the volatiles and the OC underwent partial oxidation and reforming reactions, mainly generating H_2 and CO; in the steam stage, the steam reacted with the reduced OC to produce additional H_2; in the air stage, the OC was regenerated for the next cycle.

Table 2. The reaction equations occurred during staged CLG process [25]. "Reprinted with permission from Ref. [25]. 2026, Zhao, T.M".

Reaction Type	**Reaction Equation**
PP pyrolysis	$PP \rightarrow CxHy$
Oxidation reforming	$CxHy+LaFe_{0.6}Ni_{0.4}O_3 \rightarrow CO + H_2 + LaO_3+Fe_3O_4+Fe\text{-}Ni$
Catalytic cracking	$CxHy \rightarrow C + H_2$
Carbon Oxidation	$C + H_2O \rightarrow CO + H_2$ $C + 2H_2O \rightarrow CO_2 + 2H_2$
Steam splitting	$La_2O_2 + Fe_3O_4+Fe\text{-}Ni+H_2O \rightarrow LaFe_{0.6}Ni_{0.4}O_3\text{-}\ \sigma + H_2$
Carbon combustion	$C +O_2 \rightarrow CO_2$
Regeneration	$LaFe_{0.6}Ni_{0.4}O_{3\text{-}\sigma}+O_2 \rightarrow LaFe_{0.6}Ni_{0.4}O_3$

Du et al. [26] developed a new plastic gasification process based on microwave-assisted heating and a staged reactor. They prepared a dual-purpose material FBRC using waste red mud (BR) and tire-derived carbon (TDC), serving as the role of OC and microwave absorber. The reactor was divided into two areas (D1 and D2): the upper region (D1) was loaded with a mixture of plastic and FBRC, while the lower region (D2) contained only FBRC. Under microwave radiation, OC can absorb microwave radiation due to the excellent electromagnetic loss characteristics of FBRC, thus providing a heat source for the plastics cracking. This method achieves partial mixing with spatial separation, allowing accurate control of reaction path. The process achieved a syngas yield of up to 188.66 mmol/g of plastic. It is crucial that by adjusting the FBRC feed in the D2 area, the H_2/CO ratio in the product (0.63–3.81) can be accurately controlled.

The transition from the traditional one-step CLG system to the segmented process means a fundamental change in the core mechanism and operation logic. In a traditional closed-loop single-stage CLCG system, all steps are usually completed in the same heating stage. The process essentially involves highly coupled concurrent reactions: plastic pyrolysis, oxidation, and catalytic cracking of volatiles, coke generation and initial gasification overlap and compete with each other. Therefore, it is difficult to control the reaction intensity of each stage independently, which severely limits the product distribution, especially the H_2/CO ratio and coke yield. In contrast, SCLG gasification first converts plastics into gaseous volatiles in a special pyrolysis stage. These volatiles then undergo reactions with the OC at the second stage, producing syngas and coke. In the subsequent special steam treatment stage, the coke produced in the previous stage and reduced OC react with the steam, efficiently generating additional syngas. Therefore, coke–a by-product that is difficult to control in a single-stage process–is converted into a precisely controlled secondary reaction raw material in a segmented process. Finally, the OC regenerates in the air stage. This operation of spatial separation provides the opportunity to optimize various processes (pyrolysis, reforming, gasification and regeneration) in an orderly manner. This paradigm shift from "parallel competition" to "sequential optimization" represents a key progress in the efficiency, flexibility and selectivity improvement of CLG technology.

3.3. Co-Gasification with Biomass

Chemical looping co-gasification (CLCG) technology enable efficient resources recover from organic solid waste through the coordinated treatment of biomass and plastic waste, while significantly improving the quality of syngas and process economy [27,28]. The core of this technology lies in leveraging the complementary composition and reaction characteristics of biomass and plastics. Mechanisms such as hydrogen-carbon complementarity and tar synergistic conversion enhance the yield and selectivity of hydrogen. Biomass is rich in oxygen elements, which makes it easy to produce CO_2 and H_2O in the process of gasification. The main component of plastic is hydrocarbons, which produces volatile hydrocarbons and tar precursors after pyrolysis. In the process of co-gasification, the CO_2 produced by biomass participates in the dry reforming reaction of plastic tar and long-chain hydrocarbons, promoting their conversion into H_2 and CO. At the same time, the hydrogen-rich free radicals or intermediates produced from plastic pyrolysis can help stabilize the free radical generated from biomass and inhibit the polymerization and coke formation. In addition, the water vapor during the co-gasification system undergoes steam reforming reactions with hydrocarbons, which further improves the yield of hydrogen.

Liu et al. [29] systematically studied the CLCG of pine and polyethylene with CaO/Fe_2O_3 as the OC. They observed that the addition of polyethylene significantly improved the gas yield and product quality. The H_2 yield increased from 27.65 mmol/g to 70.94 mmol/g and the H_2/CO ratio rose from 1.54 to 1.88. Al-Qadri et al. [60] investigated the CLCG pf PE+alfalfa, confirming the positive synergy effect in the process of biomass and plastic co-gasification.

Wang et al. [62] have conducted in-depth research in CLCG field. During the CLCG of biomass and polyethylene (mass ratio 1:1), H_2 concentration of 58.05%, a total gas yield of 137.81 mmol/g and an H_2/CO ratio of 2.94 were achieved. From a system integration perspective, Tian et al. [63] proposed a bag-based multi-product co-gasification system for biomass and plastic waste. Utilizing $CuFe_2O_4$ as the OC, the system employed process simulation and machine learning for multi-objective optimization, reducing the energy consumption while improving the molar content of hydrogen, demonstrating the potential of intelligent algorithms in optimizing complex processes.

Based on the existing researches on the CLCG of model plastics (such as PE and PP) and biomass, it is particularly important to study the application of actual mixed plastic waste (including PVC, additives, metal impurities, etc.). The composition of actual plastic waste is usually very complex, containing chlorine (such as PVC), plasticizers, fillers, and heavy metals. These components will release harmful substances such as HCl and dioxin during the process, which may lead to catalyst poisoning, equipment corrosion or reduced syngas purity. Therefore, the study of the CLCG of actual mixed plastic waste is not only closer to practical application, but also a key step to transform this technology from the laboratory to industrial application. For example, Kudva et al. [64] conducted a CLCG experiment using corn cores mixed with actual mixed plastic waste (chlorine content of 1.13%) as fuel. They used iron-titanium-based OC to obtain syngas with a purity of about 95% at 950°C, proving that the system has good adaptability and high conversion efficiency for chlorinated plastics. The study systematically compares various operating parameters (oxygen flow, steam addition, and the mixing ratio of biomass and plastic waste raw materials). The results show high energy efficiency and low carbon dioxide emissions can be maintained. This shows that through reasonable OC design and process control, the adverse effects of chlorinated impurities can be reduced to a certain extent, thus maintaining high-quality syngas.

In summary, the CLCG of biomass and plastic waste has significant advantages in improving resource utilization efficiency and regulating the composition of syngas. Future research should focus on clarifying the structural evolution and synergy mechanism of OCs under actual complex co-gasification conditions, and strengthen the long-term stability assessment based on actual mixed waste logistics to promote the technology towards large-scale industrial applications. In addition, the influence of impurities in real waste on the structure and reaction path of OCs should also be systematically evaluated.

4. Conclusion and Prospects

CLG provides an efficient way to transform plastic waste into high-quality H_2-rich syngas, while solving the dual challenges of waste treatment and clean energy production. This paper systematically reviews the key factors affecting the performance of plastic CLG, with a focus on the OC design and optimization of process. Research indicates that the selection of OC materials, synthesis methods, and active metal components significantly affects both the quality and stability of syngas produced. Furthermore, process parameters such as gasification temperature, reactor structure (especially the transformation from single-stage reactor to multi-stage reactor), and biomass-plastic gasification strategy can effectively improve the syngas yield and allow precise control of the H_2/CO ratio. However, the current researches predominantly rely on model plastics or relatively clean waste. Critical bottlenecks that need to be overcome include: the impact of impurities (such as chlorine and heavy metals) in the actual mixed plastic waste, mechanical wear and chemical inactivation of OCs during long-term circulation, and the economic problems of reactor scaling and system integration.

In order to promote this technology development, future research should focus on the following possible ways:

(1) Co-optimization and technical preparation of OC performance: priority is given to the development of wear-resistant, non-toxic and cost-effective multifunctional OC material systems. For example, the structural stability may be improved through composite spinel phase. For plastic waste containing chlorine, sulfur, and other impurities, materials based on rare earth modification should be developed to have in situ adsorption or transformation ability. Combined with the recycling of industrial solid waste, the production cost of OC can be reduced.

(2) Study the process and reactor adaptability of plastic waste in practical applications: use actual mixed plastic waste (including polyvinyl chloride, additives, etc.) to conduct CLG experiments to systematically evaluate the impact of impurities on the reaction path and product distribution; develop integrated pretreatment methods using CLG technology (such as low-temperature de-chlorine). Design a fluidized-bed reactors capable of adapting to feedstock variability, and conduct a trial scale test to verify and optimize gas-solid contact and long-term operation stability.

(3) Integrate the entire process system and combine it with carbon dioxide neutral technology: promote the process coupling of CLG and downstream applications of syngas (for example, hydrogen, methanol and formaldehyde production), and improve energy efficiency through thermal integration and material recovery; explore joint solutions of CLG and carbon dioxide capture to reduce labor carbon dioxide emissions; based on life cycle assessment, use actual operating data for technical and economic analysis. Through these targeted research progress and a clear path to solve key bottlenecks, CLG technology is expected to become an important part of the plastic cycle economy and low-carbon energy system, thus supporting the goal of high-quality waste recycling and carbon neutrality.

(4) Establish a unified standard for performance evaluation and data reporting: it is recommended that “mmol/g” should be used as a reference yield unit in future research, and the carbon conversion rate, H_2/CO ratio and detailed reaction conditions should be clarified. This will help to establish a transparent and comparable data system, thus accelerating the selection of OCs and process improvement.

(5) Deepen the understanding of the reaction mechanism: combine advanced characterization technology with dynamic modeling. In order to complete the transformation from empirical optimization to rational design, future research urgently needs to improve the in-situ/operational characterization of OC dynamics and interface reactions in catalytic processes (for example, environmental transmission electron microscope, synchronous radiation X-ray absorption spectrum). At the same time, the first-principle calculation and microkinetic model should clarify the energy barrier and path of important element steps (such as lattice oxygen release, C-H bond activation, carbon deposition and elimination). This multi-scale mechanism research is crucial to exploring the design principles of high-performance and long-life OCs.

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